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ESPIAL Fauna - Current state for fluoride exposure of animals in the vicinity of aluminium smelters

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Preface

ESPIAL (Ensuring Sustainable Production of Primary Aluminium) is a project directed and funded by the Aluminiumindustriens Miljøsekretariat (AMS). Four companies with ten primary aluminium smelters in Iceland, Sweden and Norway and one aluminium refinery in Norway are part of AMS today. ESPIAL is an update to the comprehensive and multidisciplinary “Effect Study” describing the effects of industrial emissions from primary aluminium plants on the local environment in the early 1990s (Norsk aluminiumindustri og miljø 1994).

ESPIAL is focusing on the effects of emissions from primary aluminium smelters on the external environment and comprises four subprojects: marine environment, air, vegetation and animals. The latter subproject, named ESPIAL Fauna, is conducted by the Norwegian Veterinary Institute (NVI) in collaboration with the East Iceland Nature Research Centre (Náttúrustofa Austurlands - NA).

ESPIAL Fauna includes two work packages (WP). In WP1, review, validation and summary of the monitoring of fluoride exposure in domestic and wild animals around the ten primary aluminium smelters in the period 1994-2018 is given. For each smelter, a “present status” and recommendations for new studies are described. In addition, WP1 includes development of a novel database named the ESPIAL Fauna database. The results of WP1 were presented in a separate report: “Oppsummering av studiar av fluorutslepp frå aluminiumsverk sine effektar på dyr 1994-2018 og skildring av ESPIAL Fauna databasen” (Vikøren 2019).

WP2 includes the new studies of fluoride exposure in wild animals recommended in WP1, performed in the period 2019-20 around selected smelters. In Årdal, also material from 2017-18 was included.

This final report commissioned by AMS presents the results of WP2. In addition, the description of the ESPIAL Fauna database is included as an appendix and the database is updated with the results obtained in WP2. The East Iceland Nature Research Centre presents the results of new studies around Alcoa Fjarðaál, Iceland, in a separate report.

Summary

This study is an assessment of the current state for effects of fluoride (F) emissions on animals around five Norwegian aluminium smelters. Wild cervids like red deer (*Cervus elaphus*) and roe deer (*Capreolus capreolus*) were used as the main indicators due to high sensitivity to F-effects. The F-exposure was documented by assessing the F-accumulation in the lower jawbone and the detrimental F-effects like dental fluorosis and osteofluorosis. The material was mainly collected by hunters during ordinary autumn hunting season around Hydro Årdal, Hydro Sunndal, Hydro Husnes, Hydro Karmøy and Alcoa Lista.

The F-exposure of wild animals differed between the five smelters and seemed to be influenced by local topography, meteorological conditions and level of F-emissions. We found F-accumulation and detrimental F-effects (fluorosis) in animals at all smelter sites, however at very variable degree and severity.

Highest F-exposure was found in deer from Årdal within a distance of approximately 13 km from the smelter, and within the fenced area around Hydro Karmøy. The F-exposure further away than 1 km from Hydro Karmøy was found to be low. The F-exposure was moderate in deer at Sunndal (lower half of Sunndal valley) and Husnes (within 3 km) and generally low at Alcoa Lista, with a few exceptions.

The survey confirmed that 1.5-year-old (yearlings) red deer is a very useful animal indicator of recent F-exposure due to very high F-sensitivity. Young individuals are particularly prone to F-toxicosis due to skeletal growth and tooth development. The consequences of F-exposure in early life can be aggravated as time goes on.
Comparing Årdal, Sunndal and Husnes where 1.5-year-old red deer was the main indicator, we found that the F-exposure was highest in Årdal. Sunndal and Husnes showed similar F-exposure causing moderate dental fluorosis; however, the area affected was much larger in Sunndal. Difference in the level of F-emissions in the period of exposure (2018-20) and more unfavourable topography for dispersion of fumes particularly in Årdal, is probably the main explanation for different degree of F-exposure between the three sites. In addition, the habitat of the examined red deer might have been closer to the plant in Årdal than at Sunndal.

At Hydro Karmøy, roe deer within the fenced area around the smelter were examined due to their close proximity to the F-emission source. These animals were indeed sedentary living there for several years and thus, an optimal indicator. The F-exposure found was extremely high showing F-accumulation at levels never documented before in deer in Norway. Many of these animals had severe dental fluorosis and some had gross signs of osteofluorosis. Similar findings were also found in sheep grazing just south of the fence. The flat landscape and the windy conditions at Karmøy is considered favourable causing rapid dispersal and dilution of F-emissions. Despite this, our findings in roe deer are a reminder that the vegetation in close proximity of the smelter is heavily F-polluted. Therefore, of animal welfare reasons, this area is not suited for sensitive ruminants like deer, sheep and cattle with low F-tolerance threshold. It is reason to believe that the conditions detected at Karmøy also applies to the other smelters. Individuals found close to the smelter at other sites also showed evidence of high F-exposure.

Compared with the early 1990s, the current F-emissions are at approximately the same level in Husnes, Karmøy and Sunndal, a bit lower and Årdal, and 50 % lower in Lista. In Årdal, indisputable F-exposure showed a decreasing trend from the 1990s; however, this trend was not seen in the percentage of deer with dental fluorosis. In the Sunndalen valley, the percentage of deer with DF had increased. Thus, more individuals seem to be at risk of F-exposure at a level causing dental fluorosis compared to the early 1990s. This is most probably due to increased red deer population and density that also includes the most F-polluted areas close to the plants. Changes in migration pattern may also be a factor of concern, for example is red deer in the Sunndalen valley more sedentary than earlier.

To reduce F-exposure of animals we recommend lowering the F-emissions and avoiding high episodic emissions, particularly during the growing season and at sites like Årdal and Sunndal where the topography is especially unfavourable for dispersal of fumes.

We recommend monitoring of F-exposure of deer on a regular basis at Hydro Årdal, Hydro Sunndal and Hydro Husnes. If F-emissions increases significantly at Hydro Karmøy and Alcoa Lista we advise to launch a monitoring program for animals there as well. Wild deer entering the fenced area around a smelter should be relocated or euthanized.

In areas close to the smelters, we advise to continue/initiate the monitoring of F-content in vegetation used as pasture or feed for F-sensitive livestock like sheep, goats and cattle. The tolerance levels must be used as an important guideline for acceptable F-levels in feed.

1 Introduction

1.1 Background information

Fluorine-containing compounds are used in the aluminium producing process and fluorides (F) are emitted with the fumes. In Norway, the first primary aluminium reduction plants began operating in 1908. Prior to 1960, these smelters were located by fjords to enable transport of raw materials and processed aluminium 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 plants are still operating i.e. in Høyanger, Årdal (Figure 1), Sunndal, and Mosjøen.

Figure 1. The Hydro Årdal aluminium smelter at Øvre Årdal 2020 photographed from Moa, ca. 700 m to the east.

Chronic fluoride toxicosis (fluorosis) in animals due to F-emissions from Norwegian aluminium smelters was first reported in livestock in the early 1930s (see ref. in Aas Hansen 1994). The aluminium industry expanded rapidly during the 1950s. This resulted in industrial fluorosis becoming a severe problem in livestock in emission-exposed areas. The extent of adverse effects of F on animals and vegetation is influenced not only by levels and composition of the emissions (gaseous/particulate), but also largely by local topographical and meteorological conditions. Advances in power-transmission technology eventually made it possible to locate smelters independently of the power source. The smelters established after 1960, i.e. at Husnes, Karmøy, and Lista, were located in more flat, open landscape, which favoured more rapid dispersal of fumes.

The Norwegian Smoke Control Council (Kontrollutvalget) initiated a monitoring program, running from 1967 to 1992, to summarize data concerning emissions from all Norwegian aluminium plants, and assess detrimental effects on vegetation and livestock (Aas Hansen 1994). The programme formed the basis for recommendations given to livestock farmers in order to prevent fluorosis. Wild animals were not included in this monitoring program. However, sporadic cases of fluorosis in cervids were seen. In 1970, a survey of red deer (*Cervus elaphus*) from Årdal documented F-exposure and fluorosis (Holt 1978).

On the initiative of the Norwegian aluminium manufacturers, a comprehensive study for monitoring the effects of fluoride emissions on the local environment was running from 1990 to 1993, named the Effect Study (Norsk aluminiumindustri og miljø 1994). As part of the study, a systematic survey of the effects of F-exposure on wild animals including cervids, hare and birds was carried out. Since the Effect Study, monitoring of industrial F-exposure in wild animals have been rather scarce. The monitoring in the period 1994-2018 is summarized in a separate report representing the WP1 of the ESPIAL Fauna study (Vikøren 2019). Only Hydro Aluminium Årdal has performed larger systematic studies of cervids, mainly red deer, in the periods 1995-98 and 2013-16.

The ESPIAL Fauna follows up the work from the Effect Study and includes new studies (WP2) of F-exposure (2019-20) to document the current state.

1.2 Fluorides – sources and absorption, retention and excretion in animals

Fluorides are ubiquitous in our environment and are present in most rocks, soil, water, plant and animal tissues in various amounts. In Norway, aluminium production is the only F-emitting industry of concern. F-containing compounds are utilized in the aluminium producing process, thus emissions from the electrolysis cells include both gaseous and particulate fluorides.
In the vicinity of aluminium plants, ingestion of feed (plants) contaminated with F is the main source of F in animals. Intake with drinking water or by inhalation is of little importance under Norwegian conditions (Vikøren et al. 1994).

1.2.1 Absorption, retention and excretion of F in animals

The absorption of fluorides takes place from the alimentary canal and depends on their solubility. Highly soluble compounds (HF, NaF) are almost completely absorbed, while F in cryolite dust and aluminium fluoride is absorbed to a lesser degree. F is rapidly absorbed into the blood. F has great affinity for bone as well as for developing and mineralizing teeth and is rapidly stored in these tissues. The major route for removal of F from the body is excretion in the urine.

In mineralized tissues, F combines to form a compound with low solubility, and thus F accumulates in bone as long as the intake continues. If the F-intake ceases (the animal moves to an unpolluted area), F is very slowly excreted from mineralized tissues. Particularly young individuals during growth are prone to F-accumulation due to the high metabolic rate of the developing mineralized tissues (bone and teeth).

Males of the cervid species (and the females of reindeer) develop a new set of antlers over a period of 3-5 months each year. F is incorporated in the developing antlers during the mineralization phase. The amount accumulated is dependent on the blood F-concentration in this period that is a product of the amount of F-absorbed and any redistributed F from the skeleton.

The low retention of F in soft tissues makes meat and milk from animals subject to heavy F-loads safe for human consumption. F in the blood crosses the placenta to the foetus; however, there seems to be a species variation dependent on differences in placental types.

Also animals that are not exposed to excessive amounts of F during life, will, because F is ubiquitous, show a natural increase in the skeletal F-content with increasing age. This "background" level is the reference used when assessing industrial F-exposure. Bone F-concentration exceeding the expected background level at a given age in the concerned animal species, is one of the most definitive indicators of excessive F-exposure. Skeletal F-retention is roughly proportional with the amount ingested and experimental studies indicate that bone F-concentration is a good indicator of the total F-intake.

1.3 Fluoride toxicity – fluorosis

Normally, humans and animals ingest some F without any adverse effects, and small amounts may be beneficial – referring to the protective effect against dental caries. However, the intake of excessive amounts is toxic. Acute F-toxicosis is rare and in humans often a result of accidental high intake of F-compounds.

Chronic fluorine toxicosis, also named fluorosis, is a condition caused by prolonged excessive intake of F and has a gradual, insidious development and onset (Shupe and Olson 1983). It is a debilitating disease, and the point at which F-ingestion becomes detrimental to an animal varies, being influenced by the following factors:

1) amount of F ingested (dose)
2) duration of F-exposure
3) solubility of ingested F
4) variation in F-exposure (intermittent)
5) animal species
6) age
7) state of nutrition
8) general state of health
9) stress factors
10) exposure to other toxic agents (synergism or alleviation of effects)
11) individual biological response

Particularly young individuals are prone to F-toxicosis. The detrimental effects of F on bone (osteofluorosis) and teeth (dental fluorosis) have been thoroughly described and documented in various species, including humans. Ruminants like cattle, sheep, and various cervid species (deer) are frequently used as indicators of industrially emitted F, due to high F-sensitivity.

1.3.1 Osteofluorosis

F may accumulate in increasing amounts in bone over a period without any detectable changes in bone structure and function. However, ingestion of toxic amounts of F for an extended period may result in the development of structural bone changes. Osteofluorosis (OF), also referred to as generalized fluorosis, is a more severe condition than dental fluorosis (DF) (see below). OF can be induced at any time during life, although the metabolic active bones of young animals are more susceptible to the effects of excessive F. The degree and development of OF may differ between species.

OF is located on the periostal surface of a bone and the most typical lesions are seen as a general thickening of the bones (periostal hyperostosis) and/or as exostoses (Figure 2). The lesions are usually bilateral. The affected bones appear chalky white with a rough periostal surface, and may have increased diameter and be heavier than normal. Mineralization of tendons at the point of attachment of the bone is also commonly observed. In severe cases, this may affect the function of the joints causing various degree of lameness.

The more metabolically active bones, used in locomotion, chewing, or breathing, are most affected. Thus, OF is first seen on bones of the lower part of the limbs (the metatarsal and metacarpal bones), the mandible (lower jaw), and the ribs.

Figure 2*: The lower jaw of a 8.5-year-old male red deer from Årdal shot in February 2020 (H83/19) with lesions in accordance with osteofluorosis. The surface of the bone is uneven and appear chalky white with a rough periostal surface with exostosis most visible in this picture on the lower margin of the jaw. Compare with Figure 3, showing a normal jawbone. The red deer had 10,887 mg F/kg in bone ash and dental fluorosis of category 5 (severe effect). The cheek teeth are worn down to the gums.*

1.3.2 Dental fluorosis

Excessive F-intake during tooth formation and mineralization results in teeth erupting with characteristic lesions, named dental fluorosis (DF). Thus, the most sensitive criterion of excessive F-ingestion by young animals is the appearance of typical fluorotic lesions on the developing permanent dentition. DF reflects F-exposure during the first years of life (in cervids up to 1.5 year or 2.5 year dependent on species).

The condition is characterized by mottling (opaque horizontal areas or striations of enamel), enamel discoloration, hypoplasia, pitting or erosion of enamel, and abnormal/excessive abrasion (Figures 3-4). The mildest gross sign of DF is small foci of mottling, most easily seen with translucent light. Changes usually are bilateral, and teeth that are formed and mineralized simultaneously have lesions of similar severity. Single or intermittent large F-doses may induce lesions seen as single or multiple horizontal rows of enamel pits. Excessive abrasion is associated with moderate to severe DF. Abrasion becomes more severe with use of the teeth as animals ages. Abraded cheek teeth may interfere with proper mastication, and abrasive feeds such as coarse, fibrous roughage commonly eaten by wild cervids, will increase the rate of dental wear. Excessive wear may lead to exposure of the tooth pulp, thus inducing pain especially when drinking cold water. Uneven and/or excessive wear often create spaces between the teeth where food accumulates and can cause painful inflammation in the gums, teeth and the jawbone.

A short-term exposure to high F-concentrations can cause severe DF. Therefore, also short-lasting high F-emissions can cause adverse consequences for grazing animals particularly young animals. Whether DF occurs in deciduous teeth ("milk teeth") of calves (fawns) is discussed, however lesions very suggestive of DF have been observed in both cattle and cervids. Thus, if the F-intake by the mother is sufficiently high, it seems possible that high enough amounts pass with the blood through the placenta and interfere with tooth development in the foetus.

***Figure 3**: Dental fluorosis in red deer. To the left, cheek teeth (premolars and molars) of an adult red deer showing severe dental fluorosis. The DF is particularly severe on the three teeth to the left showing abnormal abrasion. To the right, normal cheek teeth of an adult red deer. It is normal that the cheek teeth have brownish coating. This brownish colour is different from the brownish staining of the enamel seen in dental fluorosis.*

Figure 4*: To the left, the front teeth (incisors) of a 1.5-year-old red deer showing two large permanent teeth and three small deciduous teeth. The latter have a smooth, normal enamel, whereas the two permanent teeth show severe pitting/erosion of enamel affecting the whole tooth crown. This individual also had DF of the cheek teeth. To the right, normal permanent front teeth (incisors) of an adult red deer.*

1.4 Fluoride tolerance

F-tolerance has been estimated in different animal species based on published data from feeding experiments and field studies. These estimates must be seen as guiding and not absolute as they are associated with some uncertainty. When establishing F-tolerances, target effects must be clearly defined. In some studies, one effect level has been given for “performance” and another lower level for “pathology”. The first estimates the F-amount that can be ingested without clinical interference with normal performance, whereas the second indicates the F-level at which distinct pathological changes (OF/DF) are induced. Animals can have minor DF without impairment of performance.

Table 1 is based on tolerance levels from several studies including Kontrollutvalget. The tolerance levels vary within species partly due to some differences in the definition used. Generally, the highest levels given express the “performance tolerance threshold”.

Table 1: *F-tolerance levels in feed for animals.*

Species	F-concentration in feed (mg F/kg dry weight)
Dairy cow and heifer (mjølkeku og kvige)	≤30
Beef cow (kjøttfe)	40
Sheep and goat	30-50
Wild cervids (deer, moose)	< 30
Horses	40-60
Swine, growing	70
Dogs, growing	50-100
Poultry	150-400

Generally, birds seem less sensitive to F than most other animal. There is considerable variation between species in background bone F-levels and this may partly be due to differences in diet, F-disposition and life span. Some bird species have a high tolerance threshold for F probably due to marine adaptation. For example, Adélie penguins feeding on F-rich krill had extremely high bone F-level without any adverse effects.

1.5 Bioindicators/biomonitors

Wild cervids have proven very useful as indicators/monitors of industrially emitted F (see ref. in Vikøren et al. 1994 and Vikøren 1995). They fulfil the bioindicator definition by "responding sensitively and specifically to the pollutant under consideration, and show recognizable effects caused by the pollutant", as well as the biomonitor definition: "organisms that take up and sometimes accumulate environmental pollutants, thus indicating special and/or concentration trends".

In this study, cervids respond as bioindicators by developing DF and OF, and as biomonitors by accumulation of F in mineralized tissue like the skeleton. Fluorine that is absorbed from the gastro-intestinal tract is quickly distributed and incorporated into mineralized tissue. Since the degree of excretion of incorporated F is very low, the bone F-level of a cervid reflects the total amount absorbed. Due to this age-accumulation of F, it is important to know the age of the individual in question to evaluate the F-level.

Cervids are herbivourous and contaminated vegetation is the main F-source in the vicinity of aluminium smelters. In contrast to domestic ruminants like sheep and cattle, cervids graze/browse the whole year around. Cervids are relatively long-lived and good methods are available for their age determination. The latter is essential when evaluating the severity of the F-accumulation.

Particularly roe deer (*Capreolus capreolus*) fulfil the criteria required of a biomonitoring species, since they are more stationary than red deer, moose (*Alces alces*) and reindeer (*Rangifer tarandus*). Thus, stationary individuals that live close to aluminium smelters are exposed to higher levels of F than animals that migrate between locations with variable degree of F or that are stationery in an area with lower level of pollution.

Further details and references to the text in the chapters 3.1 – 3.5 can be found in the Effect Study report (Vikøren et al. 1994) and Vikøren (1995).

1.6 Objective and scope of the WP2 study

The objective was to assess the current state of F-exposure of wild animals around the aluminium smelters Hydro Årdal, Hydro Sunndal, Hydro Husnes, Hydro Karmøy and Alcoa Lista. The main indicator was wild cervids (deer) and we used the F-concentration in the lower jawbone and the prevalence and severity of fluorosis as a marker of F-exposure.

2 Materials and methods

2.1 Selection of the main indicators

The main objective was to assess recent F-exposure in animals and therefore, young wild cervids (yearlings of red deer, roe deer and moose) were chosen as the main bioindicator/biomonitor species (in the following called indicators). These reflect the F-exposure the last 1.5 year. In areas with small hunting bag or lower collection success of yearlings, other age groups of cervids and other species were included (Karmøy, Lista).

In addition, the mountain hare (*Lepus timidus*) was considered as a possible indicator, however due to low hunting activity in the locations in question (Karmøy, Lista); no hares were included in the study.

2.2 Selection of specimens for examination

As in the Effect Study, the lower jawbone (mandible) was chosen as the main specimen in cervids and livestock, since this makes age assessment, bone F-analysis, and examination of detrimental effects (DF and OF) possible. The mandible is among those bones in the skeleton with the highest F-level.

Karmøy: Some supplementary material was collected from deer from within the fenced area around Hydro Karmøy. These comprised the whole carcass of two roe deer fawns, and the head and internal organs from nine roe deer and one red deer. After detection of high F-exposure in roe deer within this area in 2019, the materials collected in 2020 were supplemented with bones of the lower part of the limbs (metatarsus and metacarpus) to better document OF if present.

The femur was collected in gulls from Karmøy to compare with results from the Effect Study and other studies.

Figure 5: *Lower jawbones (mandibles) of reindeer ready for examination.*

2.3 Survey sites and collection of materials

Based on the conclusions in WP1 (Vikøren 2019), the new studies in WP2 were performed at the following sites: Hydro Årdal, Hydro Sunndal, Hydro Husnes, Hydro Karmøy, and Alcoa Lista. Alcoa Fjarðaál was also included and the results are presented in a separate rapport from the East Iceland Nature Research Center.

To ease the reading we have chosen to present the following information in the Results and Discussion chapter: 1) background data for the study sites including description of the examined area at each smelter, 2) information about the local cervid populations, and 3) details regarding examined species for each study site.

Hunters collected the lower jawbone from wild cervids during the ordinary autumn hunting season 2019-20 (in addition 2017-18 in Årdal), running from August to December. The local wildlife management (LWM) or representative from the local Norwegian Hunters and Fishermen's Association coordinated the sampling with the hunters. Jaws from traffic killed animals or other fallen stock were also collected.

Additionally, in Karmøy the head from domestic sheep and the lower jawbone of a cow grazing close to the smelter were collected from the farmers, and in Lista the head of an Eurasian beaver (*Castor fiber*) found dead close to the smelter was included.

After the end of the ordinary hunting season, the materials were submitted to the Norwegian Veterinary Institute (NVI) for laboratory examinations and specimens collection for F-analysis.

2.4 Methods

Mandibular F-concentration was used as the parameter reflecting level of F-exposure in animals and to evaluate the extent of exposure on local wildlife populations. In addition, the material was subject to gross examination for fluorosis: dental fluorosis (teeth) and osteofluorosis (bone).

The methods used for examining collected lower jawbones (mandibles) were as recommended in the ESPIAL Fauna WP1 report:

The Norwegian Veterinary Institute recommendation for standardisation of future studies of dead animals for fluoride exposure:

- Bone for fluoride analysis: the lower jawbone (mandible).
- The "mandible standard" method for sampling, described in Vikøren et al. (1996).
    - In short, collect the bone sample from the ventral part of the mandible between the fourth premolar and the second molar, with some modifications for roe deer and calves (fawns) (Figure 6).
- Gross examinations of the teeth for dental fluorosis and the mandible for osteofluorosis.

***Figure 6**: The bone sample for F-analysis (left) in cervids and sheep was collected from the ventral part of the mandible between the third (P4) and fifth (M2) cheek tooth (right: the sample was the ventral part of the jaw between the two horizontal saw marks).*

2.4.1 Examination for detrimental effects of fluoride

If needed the teeth were cleaned with a tooth brush before gross examination. Both direct and translucent illumination of the incisors were employed to reveal small fluorotic changes. Each animal was given an overall dentition score based on the individual tooth scores.

The teeth were inspected for signs of dental fluorosis and classified according to the following categories (DF-category). Photos of the various categories are found in Vikøren et al. (1994) and Vikøren and Stuve (1996):

1. Normal tooth: smooth, translucent, glossy, white enamel; normal tooth shape/size.
2. Questionable effect: may have enamel flecks; unilateral or bilateral cavities; no mottling; unable to determine exact cause.
3. Slight to moderate effect: slight to definite mottling of enamel; may have various degrees of staining.
4. Marked effect: generalized mottling, pitting of enamel, staining and/or signs of wear.
5. Severe effect: cream coloured and pitted or eroded enamel with definite wear; staining may be present.
6. Excessive effect: as 5, with excessive wear.
8. Normal advanced old age related wear.

Mandibles, and for some individuals also the lower limbs (metacarpals/metatarsus), were examined grossly for osteofluorotic changes. The soft tissue was removed before inspection.

2.4.2 Evaluation of bone F-concentration in cervids

2.4.2.1 Upper normal level (UNL)

In the Effect Study, in order to distinguish between cervids exposed and unexposed to F-emissions a limit called the upper normal limit (UNL) was estimated (Vikøren and Stuve 1994). This limit was estimated for each species and age group using the sample 95th percentiles of mandibular F-concentrations in cervids collected at reference sites (located far from aluminium smelters, representing background F-levels in Norway).

Since the individual mandibular F-concentration in animals from reference areas varies considerably within age groups, it is difficult to interpret whether levels slightly above UNL represent exposure caused by F-emissions. Thus, a higher level represented by UNL multiplied with a factor of 3 (UNL x 3) have been set as a threshold for levels showing indisputable F-exposure (Table 2). The extent of F-exposure in each study site were described by the percentage of cervids having mandibular F-concentrations exceeding the UNL x 3.This concentration is usually below the level causing fluorosis (DF and OF).

The UNL x 3 show some deviation from the expected age accumulation of F, particularly between calves (0.5 year) and yearlings (1.5 year) (discussed in Vikøren and Stuve 1994). This may reflect that the bone metabolism is particularly high in the calf the first months of life, and therefore more of the ingested F is accumulated in bone than later in life. Interestingly, the background F-level show species variation, roe deer having the highest and moose the lowest levels.

Table 2*: The upper normal F-level (mg F/kg bone ash) multiplied with a factor of 3 (UNL x 3) in the lower jawbone (mandible) of various age groups of Norwegian cervid species.*

Species	0.5 year	1.5 year	2.5 year	3.5-5.5 year	6.5 year and older
Roe deer	1,600	930	1,500	1,600	2,600
Red deer	930	810	1,100	1,100	1,800
Moose	450	330	690	480	2,300

2.4.2.2 *Dose-effect relationship established for 1.5-year-old red deer*

In 1.5-year-old red deer, a fluoride dose – effect relationship (Table 3) was generated from the results of the Effect study (Vikøren and Stuve 1994). This was used to describe the current F-exposure at those study sites in which 1.5-year-old red deer was the main indicator (Årdal, Sunndal, Husnes).

Table 3*: Fluoride dose - effect relationship for 1.5-year-old red deer.*

Cumulative F dose (mg F/kg bone ash in mandible)	Effect (visible dental fluorosis)
Less than 1,000	None
1,000 – 2,000	Risk of developing dental fluorosis
Above 2,000	Causes dental fluorosis

2.4.3 Evaluation of bone F-concentration in sheep

The bone F-level in sheep and a cow (Karmøy) was evaluated according to Table 4 showing F-levels related to detrimental F-effects.

Table 4: *Categories of detrimental F-effects in domestic ruminants (sheep, cattle), based on bone F-concentration from slaughtered animals (livestock) (mg F/kg bone ash) (Aas Hansen 1994).*

Age	Detrimental effect		
	None	Moderate	Severe
Up to 2 years	< 1,999	2,000-4,499	> 4,500
2 - 5 years	< 3,499	3,500-5,999	> 6,000
Older than 5 years	< 4,499	4,500-7,999	> 8,000

2.4.4 F-analysis

All the bone samples were analysed for F at the laboratory of Årdal Metallverk, Hydro Aluminium, using standard methods (Vikøren et al. 1996). Bone samples were converted to ash at 700°C for a minimum of 8 hrs. The F-concentration in bone ash was determined using a solid-state fluoride-selective combination electrode after addition of a buffer. Two parallel ash samples from each specimen were analysed, and the mean recorded as the F-concentration. If the concentration in the highest parallel exceeded the lowest parallel by more than 10 %, the result was rejected and the analysis repeated. The results were given as mg/kg F in bone ash and registered in the ESPIAL Fauna database (attached).

To reveal any deviation between protocols used in the Effect Study and ESPIAL, old bone samples from the 1990s representing low, medium and high F-concentrations were analysed with the present protocol. The new results did not deviate significantly from the old results (data not shown, the results can be provided if desired).

2.4.5 Age determination of animals

Ages of young red deer (≤ 2.5 year), and moose and roe deer (≤ 1.5 year), were determined by tooth replacement. All other cervids were age determined at the Norwegian Institute for Nature Research (NINA) based on reading growth layers in incisor tooth cementum after tooth sectioning (see ref. in Vikøren et al. 1996).

2.4.6 The site description and F-emission data

The information given for the various smelter sites is based on the descriptions from AMS.

The fluoride emission to the air data include both particulate and gaseous F and are given as tonnes F per year. The data are obtained from the smelters and include emissions from pot lines (gas treatment + pot room ventilation) and anode production. The recent years the numbers may deviate from those reported in www.norskeutslipp.no (Norske utslipp, the Norwegian Environment Agency) since these also include minor F-emissions from the cast houses. Due to the short timeline of the latter, AMS have chosen to exclude them from the total amount of F-emissions per year (these emissions may amount to 10–15 % of the pot line emissions).
Gaseous F is more bioavailable than particulate fluorides. In F-emissions from prebake smelters, the proportion of gaseous F of the total F-emissions is higher than for Søderberg technology. During the decade 2000-2010, all the Søderberg lines at Sunndal, Årdal and Karmøy were closed, thus the proportion of gaseous F in the emissions is currently higher than in the 1990s (Effect Study), particularly at Sunndal and Karmøy (had high production with Søderberg technology before replacement with prebake).

3 Results and Discussion

The results of the new studies describing the current state are presented and discussed separately for each study site. A separate report from the East Iceland Nature Research Centre presents the results from Alcoa Fjarðaál.

3.1 Hydro Årdal

3.1.1 Site description

The Hydro Årdal facilities comprise a primary metal smelter at Øvre Årdal, at the inlet to the lake Årdalsvatnet, and anode production facilities at Årdalstangen at the head of the Årdalsfjord. The distance between the two plants is about 12 km. The plants lie in a narrow valley surrounded by high mountains. At Øvre Årdal, the valley branches into the Moadalen, Utladalen, Fardalen and Nundalen valleys. Winds blowing up and down the valley occur with roughly the same frequency. There is little difference between winter and summer in this respect. The climate is relatively dry, and similar to an inland climate, with warm summers and cold winters. There is some agriculture in the municipality, mainly free grazing sheep, goats and some cattle, in addition to some hay harvesting.

The Årdal plant started up in 1948 using Søderberg technology. Today it consists of two prebake lines and a test center for improved reduction technology. From 1995 to 2005, the metal production increased, mainly as a result of expansions of the prebake lines. The Søderberg line was closed in 2007. The total production capacity is about 210,000 tonnes p.a. Årdal Karbon at Årdalstangen manufactures anodes for prebake plants.

The F-emissions increased slightly during the production increase and fell when the Søderberg line was closed in 2007. Nearly all the F-emissions come from the metal plant. With prebake technology, the proportion of gaseous F of the total F-emissions is higher than with Søderberg. The F-emissions in 2020 was 95 tonnes per year, which were 25 – 35 % lower than in the early 1990s (Figure 7).

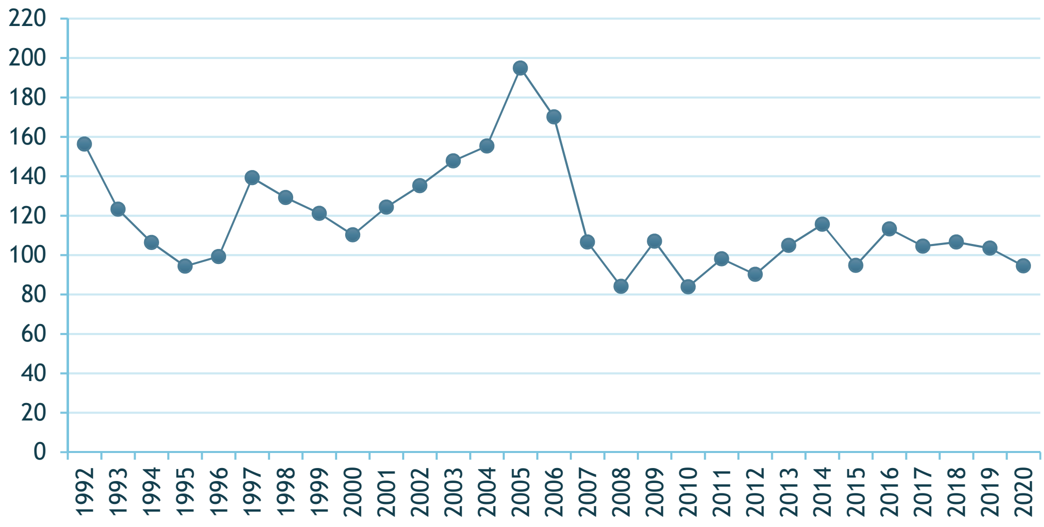

Figure 7: *Fluoride emissions (tonnes per year) from Hydro Årdal from 1992 to 2020. Source: AMS.*

3.1.2 The red deer population in Årdal

The red deer population in Årdal municipality has increased considerably from the early 1990s to 2020, reflected by a 5-fold increase in hunted animals (Figure 8). The particular high numbers harvested the last five years are a result of management measures to reduce the cervid population around the Nordfjella mountain area due to the occurrence of the infectious prion disease Chronic Wasting Disease (CWD) in wild reindeer.

The large increase in the red deer population and higher density most probably leads to more animals living closer to the plant in recent years. Hence, it will be higher competition regarding feed resources, particularly of the preferred plant species, and one could expect changes in habitat use.

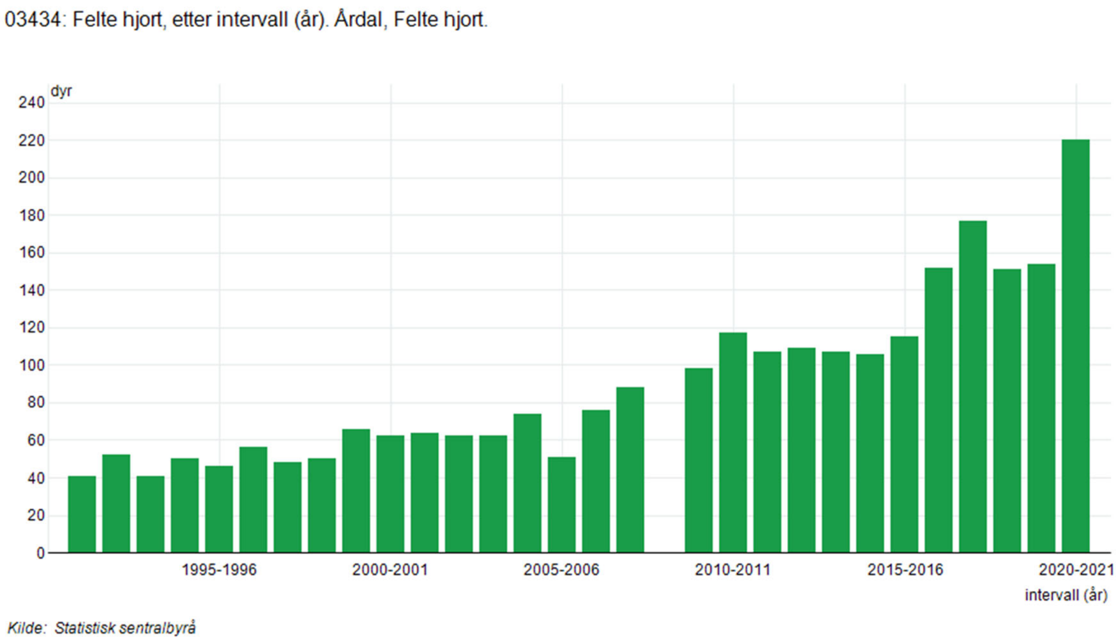

Figure 8: Number of hunted red deer in Årdal municipality from 1991 to 2020. Source: Statistics Norway (www.ssb.no).

3.1.3 Studies of fluoride exposure in wild cervids in Årdal, 2017-20

The main indicator in the studies 2017-2020 in Årdal was 1.5-year-old red deer shot during ordinary hunting and the study area included the whole municipality. The various hunting areas are shown in Figure 9. Other wild cervid species were also included if available.

The lower jawbone from 117 cervids (red deer, roe deer and moose) was collected 2017-20 in collaboration with the LWM and hunters and included hunted animals and fallen stock (traffic killed, other casualties) (Table 5).

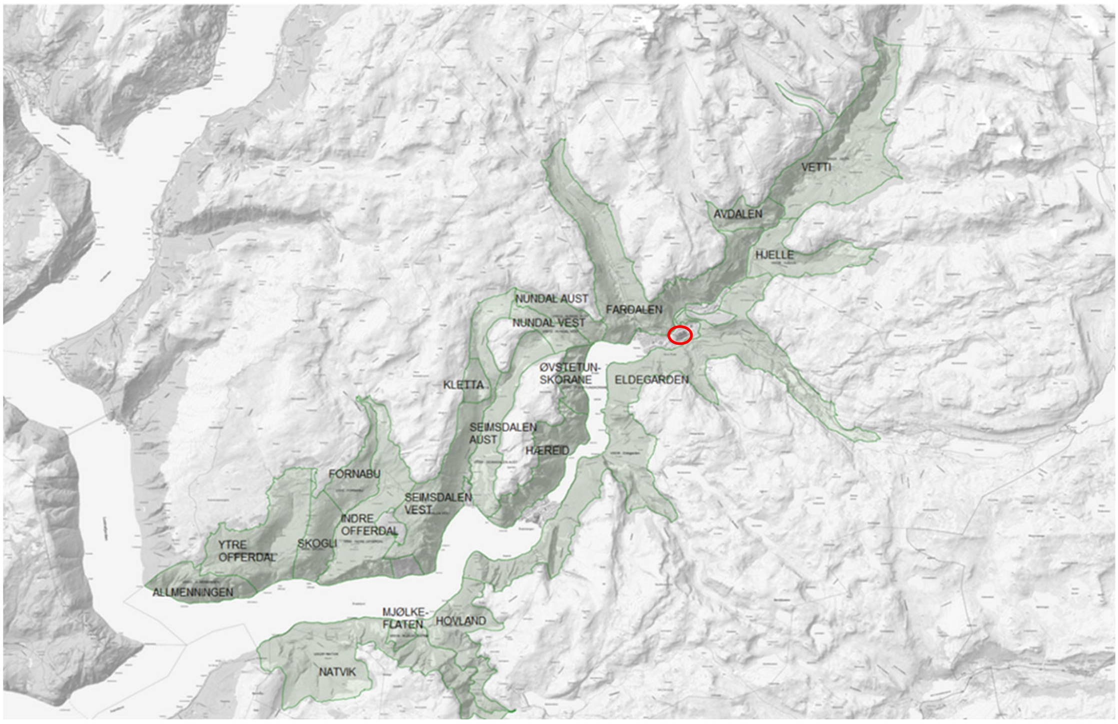

Figure 9: *Map showing the various hunting areas (vald) for red deer in Årdal municipality. The aluminium smelter is marked with a red circle. In the courtesy of Knut Fredrik Øi, local wildlife management officer.*

Table 5: *The number of wild cervids examined for F-exposure in Årdal 2017-20. The main F-indicator was 1.5-year-old red deer.*

Species	Year	Total number examined	Number of 1.5-year-old
Red deer	2017	30	28
	2018	19	19
	2019	16	15
	2020	41	36
	Total	106	98
Roe deer	2017-18	4	1
	2019	2	0
	2020	2	1
	Total	8	2
Moose	2017-18	3	0
	2019	0	
	2020	0	
	Total	3	0
All cervids	**Total**	**117**	**100 (85 %)**

3.1.3.1 Bone F-concentration in red deer

The results of the F-analyses are shown in Tables 6-7 and Figures 10-11. The individual F-level varied considerably. The data are registered in the ESPIAL Fauna database (updated version).

The main indicator in Årdal reflecting recent F-exposure was 1.5-year-old red deer and the individual F-levels are shown in Figure 10 for the various hunting areas. The F-level in other age groups of red deer is shown in Figure 11. All hunting areas in Årdal were represented in the materials except Nundal, Øvstetun Skorane, Kletta, and Mjølkeflaten. Four red deer were from an unknown area.

Among 1.5-year-old red deer, 32 (33 %) had F-level exceeding UNL x 3 (810 mg F/kg), thus showing indisputable F-exposure (Figure 10). The corresponding number for all ages of red deer was 34 (32 %). The majority of animals showing indisputable F-exposure was found in the areas closest to the plant (Figure 10 and Table 7). Eldegarden is a very large hunting area as shown in Figure 9, and the individuals shot at Moa (Figure 1, close to the smelter) had higher F-levels than those shot at Lægreid (further away from the smelter). All six red deer shot at Hæreid hunting area had indisputable F-exposure.

***Table 6**: The fluoride (F) concentration (mg F/kg bone ash) in the lower jawbone (mandible) of red deer from Årdal municipality given for the four monitoring periods 2017-20, 2013-16, 1995-98, and 1990-93 (the Effect Study). The F-level is given for each age group with mean, minimum, and maximum. The individual data are registered in the ESPIAL Fauna database (updated version).*

Sampling year		0.5 year	1.5 year	2.5 year	3.5-5.5 year	6.5 year and older
2017-20	Number	2	98	5	0	1
	Mean	953	1,036	347		
	Min	505	115	243		
	Max	1,401	6,210	471		11,679
2013-16*	Number	47	76	23	57	50
	Mean	450	1,102	1,122	2,007	3,026
	Min	53	108	266	377	361
	Max	4,053	7,365	2,946	12,462	14,905
1995-98	Number	13	14	16	20	11
	Mean	478	1,217	1,922	2,038	4,279
	Min	53	168	618	237	2,021
	Max	1,406	3,863	4,435	6,327	7,998
1990-93	Number	13	34	25	13	10
	Mean	657	1,283	1,374	2,512	4,641
	Min	76	252	524	522	914
	Max	4,165	5,436	2,859	6,820	12,707

** 2016: Tre adult animals with F-level of 11,419, 8,480 and 7,050 mg F/kg, respectively, were not included in the table because they were not aged.*

***Table 7**: Geographical distribution (hunting area, vald) and number and percentage of red deer with F-concentration above the upper normal level x 3 (> UNL x 3) in Årdal municipality, given for four different monitoring periods and the total (1990-2020).*

Hunting area - nr (vald)	2017-20*	2013-16	1995-98	1990-93	Total
Eldegarden V0038	8/13 (62 %)	26/36 (72 %)	13/18 (72 %)	12/13 (92 %)	59/80 (74 %)
Hjelle V0026	2/5 (40 %)	7/11 (64 %)	4/6 (67 %)	3/3 (100 %)	16/25 (64 %)
Vetti, Avdalen V0025, V0023	1/5 (20 %)	3/7 (43 %)	4/6 (67 %)	7/8 (88 %)	15/26 (58 %)
Fardalen V0043, V0017, V0016	8/12 (67 %)	11/19 (58%)	3/5 (60 %)	3/5 (60 %)	25/41 (61 %)
Nundal V0012, V0014	0/0	1/4 (25 %)	5/10 (50 %)	0/1 (0 %)	6/15 (40 %)
Hæreid, Seimsdal, Kletta V0040, V0008, V0009, V0011	7/10 (70 %)	25/47 (53 %)	7/10 (70 %)	5/7 (71 %)	44/74 (59 %)
Skogli, Fornabu, Indre Offerdal V0004,V0005, V0006	0/12 (0 %)	8/32 (25 %)	1/2 (50 %)	1/6 (17 %)	10/52 (19 %)
Allmenningen, Ytre Offerdal V0003, V0006	1/21 (5 %)	5/41 (12 %)	3/7 (43 %)	11/28 (39%)	20/97 (21 %)
Hovland V0032	2/11 (18 %)	6/28 (21 %)	3/7 (43 %)	2/8 (25 %)	13/54 (24 %)
Mjølkeflaten, Natvik V0019, V0020	2/13 (15 %)	4/25 (16 %)	1/3 (33 %)	3/8 (38 %)	10/49 (20 %)
Total**	**31/102 (30%)**	**96/250 (38 %)**	**44/74 (59 %)**	**47/87 (54 %)**	**218/513 (42 %)**

** The majority was 1.5-years-old. ** Total of those with known locality.*

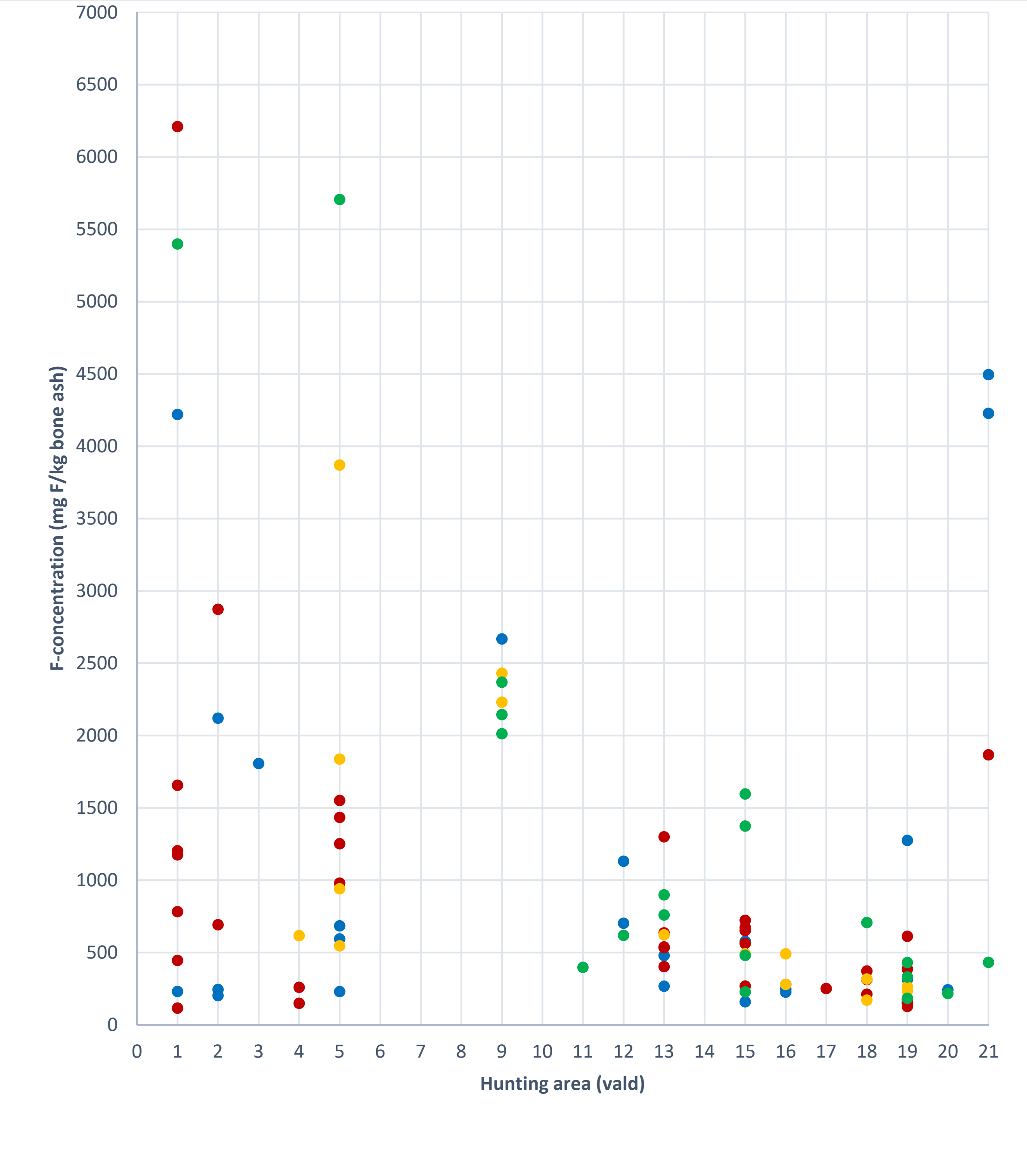

***Figure 10**: F-concentration (mg F/kg) in the lower jawbone (mandible) of 1.5-year-old red deer from Årdal municipality 2017-20, given for the various hunting areas (Figure 9): 1) Eldegarden, 2) Hjelle, 3) Avdalen, 4) Vetti, 5) Fardalen, 6) Nundal aust, 7) Nundal vest, 8) Øvstetun Skorane, 9) Hæreid, 10) Kletta, 11) Seimsdal aust, 12) Seimsdal vest, 13) Hovland, 14) Mjølkeflaten, 15) Natvik, 16) Indre Offerdal, 17) Fornabu, 18) Skogli, 19) Ytre Offerdal, 20) Almenningen and 21) unknown. The colour of the dots show the sampling year: blue = 2017, green = 2018, yellow = 2019, and red = 2020.*

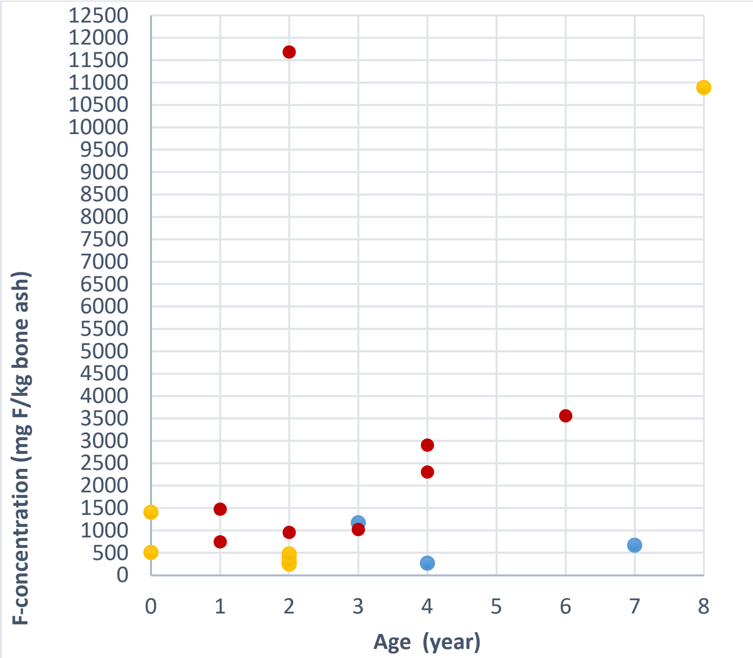

Figure 11: *Age and F-concentration (mg F/kg) in the lower jawbone (mandible) of red deer (yellow, other ages than 1.5-year-old), roe deer (red, all ages), and moose (blue, all ages) from Årdal, 2017-20. The 1.5-year-old red deer is shown separately in Figure 10 (above).*

3.1.3.2 Detrimental effects of F in red deer

Dental fluorosis: Totally 24 red deer (23 %) had DF, two in category 5 (severe), eight in category 4 (marked) and 14 in category 3 (slight to moderate) (Figure 12). All were 1.5-year-old except one 8.5-year-old (DF = 5). The animals with DF were from the hunting areas closest to the plant as shown in Figure 13 and Table 8 (Eldegarden, Hjelle, Avdalen, Fardalen, and Hæreid, Figure 9). All 1.5-year-old red deer with DF 4 and 5 had high F-levels ranging between 2,400 and 6,200 mg F/kg, whereas the range for those with DF 3 was 1,170 – 3,870 mg F/kg.

For three of the animals with DF, the location was unknown. The 8.5-year-old male showed pronounced F-exposure with severe DF and 10,887 mg F/kg in the jawbone and was shot in 2020 within the fenced area around the plant.

Figure 12: *The front teeth of 1.5-year-old red deer with dental fluorosis from Årdal. To the left, severe DF (category 5) on two teeth showing large enamel damage. The animal was from Moa, Eldegarden vald, and had a F-level of 6,210 mg F/kg. To the right, marked DF (category 4) showing a horizontal row of enamel pitting on two teeth. This animal came from Hjelledalen and had 2,872 mg F/kg bone ash.*

According to the fluoride dose – effect relationship for 1.5-year-old red deer (Table 3), 14 animals (14 %) had F-levels between 1,000 – 2,000 mg F/kg (risk of developing DF) and 15 (15 %) had F-level above 2,000 mg F/kg (causes DF) (Figure 13). This is in agreement with the findings of DF presented above. Of the 14 animals in the “risk of developing DF” group, DF was found in eight and four had questionable effect. Thus, a F-exposure that gives an accumulated dose of 1,000 to 2,000 mg F/kg in a 1.5-year-old red deer very often causes DF.

Osteofluorosis: Two of the red deer (2 %) had small gross signs of OF in the mandible. One was the 8.5-year-old male shot within the fenced area of the plant, presented above. The other was a 1.5-year-old animal collected in 2017 from an unknown locality having 4,495 mg F/kg and DF 4.

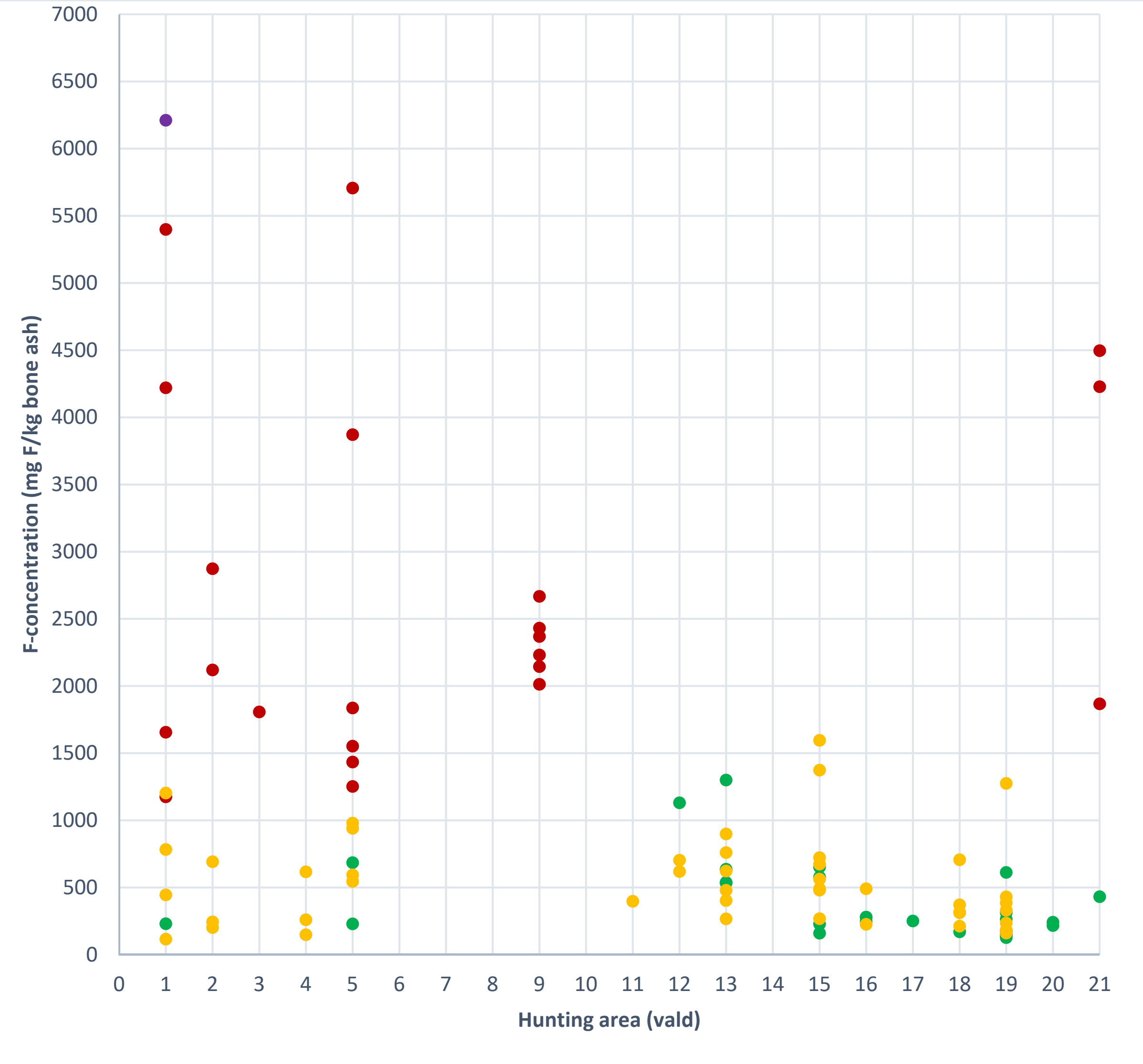

***Figure 13**: F-concentration (mg F/kg) in the lower jawbone (mandible) and dental fluorosis (DF) of 1.5-year-old red deer from Årdal municipality 2017-20. The colour of the dots show the dental category: green = DF 1 (normal), yellow = DF 2 (questionable), red = DF 3 and 4 (slight to marked DF), and purple = DF 5 (severe DF). The results are given for the various hunting areas (map in Figure 3): 1) Eldegarden, 2) Hjelle, 3) Avdalen, 4) Vetti, 5) Fardalen, 6) Nundal aust, 7) Nundal vest, 8) Øvstetun Skorane, 9) Hæreid, 10) Kletta, 11) Seimsdal aust, 12) Seimsdal vest, 13) Hovland, 14) Mjølkeflaten, 15) Natvik, 16) Indre Offerdal, 17) Fornabu, 18) Skogli, 19) Ytre Offerdal, 20) Almenningen, and 21) unknown.*

***Table 8**: Geographical distribution (hunting areas, see Figure 9) of number and percentage of red deer with dental fluorosis (DF) in Årdal municipality, given for four different monitoring periods and the total (1990-2020).*

	2017-20*	2013-16	1995-98	1990-93	Total all periods
Eldegarden V0038	6/13 (46 %)	22/36 (61 %)	4/18 (22 %)	4/13 (31 %)	36/80 (45 %)
Hjelle V0026	2/5 (40 %)	4/11 (36 %)	1/6 (17 %)	1/3 (33 %)	8/25 (32 %)
Vetti, Avdalen V0025, V0023	1/5 (20 %)	2/7 (29 %)	0/6 (0 %)	2/8 (25 %)	5/26 (19 %)
Fardalen V0043, V0017, V0016	6/12 (50 %)	10/19 (53 %)	1/5 (20 %)	2/5 (40 %)	19/41 (46 %)
Nundal V0012, V0014	0/0	0/4 (0 %)	2/10 (20 %)	0/1 (0 %)	2/15 (13 %)
Hæreid, Seimsdal, Kletta V0040, V0008, V0009, V0011	6/10 (60 %)	10/47 (21 %)	3/10 (30 %)	4/7 (57 %)	23/74 (31 %)
Skogli, Fornabu, Indre Offerdal V0004,V0005, V0006	0/12 (0 %)	5/32 (16 %)	0/2 (0 %)	0/6 (0 %)	5/52 (10 %)
Allmenningen, Ytre Offerdal V0003, V0006	0/21 (0 %)	4/41 (10 %)	0/7 (0 %)	3/28 (11 %)	7/97 (7 %)
Hovland V0032	0/11 (0 %)	3/28 (11 %)	2/7 (29 %)	0/8 (0 %)	5/54 (9 %)
Mjølkeflaten, Natvik V0019, V0020	0/13 (0 %)	1/25 (4 %)	1/3 (33 %)	2/8 (25 %)	4/49 (8 %)
Total**	**21/102 (21 %)**	**61/250 (24 %)**	**14/74 (19 %)**	**18/87 (19 %)**	**114/513 (22 %)**

** The majority was 1.5-years-old. ** Total of those with known locality.*

3.1.3.3 Bone F-concentration and detrimental effects in roe deer and moose

The results of the F-analyses of eight roe deer and three moose are shown in Figure 11. Five roe deer (63 %) and one moose (33 %) had F-level exceeding UNL x 3, thus showing indisputable F-exposure. The roe deer came from locations rather close to the smelter: within the smelter area, Eldegarden, Fardalen, and Seimsdalen (Figure 9). The moose came from Hjelle and Seimsdalen.

Three roe deer (38 %) had DF, one each in the categories DF 6 (excessive), DF 5 (severe), and DF 3 (slight to moderate). The first was a 2.5-year-old animal hit by a car within the fenced area of the smelter in 2017. It had an extreme high F-level of 11,679 mg F/kg. The other two were from Eldegarden and Fardalen.

One 3.5-year-old male moose from Seimsdalen had DF (33 %) of category 5. The F-level was surprisingly low (1,165 mg F/kg). It may be due to DF caused by higher F-exposure early in life, and less exposure the last two years (even causing decreasing F-level in bone due to F-excretion).

3.1.4 Local differences in F-exposure and relation to vegetation data

In the main indicator, 1.5-year-old red deer, indisputable F-exposure (>UNL x 3) was documented in the highest percentage of animals from Eldegarden, Avdalen, Fardalen and Hæreid, followed by a medium percentage in individuals from Hjelle (Figure 13). Individuals with DF also were restricted to these areas including the smelter area (not a hunting area).

Roe deer, usually a more sedentary species than red deer, showed the same pattern. Animals from Seimsdalen and particularly areas further west on both sides of the fjord, showed to a lesser degree F-exposure. A few individuals from these areas may have F-level above 1000 mg F/kg (being in risk of developing DF), but this is most probably due to migration from more polluted areas closer to the plant. No animals from Nundalen were examined, so the current F-exposure in this area is not known. Some caution in interpretation must be taken in those hunting areas were few animals were examined.

The ESPIAL vegetation survey in 2019-20 (Børja et al. 2021) included sampling at five locations in Øvre Årdal at distances of 0.7 km (Moa), 1 km and 2 km (Timrebakkane), 2.5 km (Melheim) and 4 km (Hjelle) from the smelter. The first three locations are a part of Eldegarden hunting area, Melheim is a part of Fardalen and Hjelle is within Hjelle hunting area. In 2020, Årdalstangen (10 km) was also included as a sampling site, being representative for the hunting areas Hæreid and western part of Eldegarden. The main conclusion was that the highest F-concentrations were found in vegetation closest to the smelter and declining with the distance from the smelter. Ferns had very high F-content. It is stated that deer may eat ferns, but there is no Norwegian studies and it is assumed that these plants are of little quantitative value in the diet. Probably are ferns most attractive in the spring when they sprout out. However, if an animal has a penchant for eating ferns, it may be a significant source of F causing F-accumulation and, in the worst case, dental fluorosis. We have chosen to relate our results to the F-level in rowan, a preferred browsing plant for cervids, even though it quantitatively is of less importance. We have also included blueberry, normally eaten in larger amounts especially during the autumn.

The vegetation survey showed that the F-level in rowan was over the tolerance threshold for cervids (30 mg F/kg) at all sampling sites in 2019-20, the highest concentrations (> 200 mg F/kg) being found within a distance of 1 km from the smelter and declining at the sampling sites further away. Blueberry was only available for sampling in 2020 at Timrebakkane hill at 1 km and 2 km from the smelter and at Hjelle, and the F-level was exceeding the tolerance threshold at Timrebakkane 1 km and Hjelle.

The vegetation study was done at locations rather close to the smelter, whereas the survey of cervids comprises a larger area. The results of the vegetation study are in accordance with the main findings in deer. We have documented considerable F-exposure of deer in the areas closest to the plant: Eldegarden and Fardalen, but also in areas like Hæreid and Hjelle.

3.1.5 New results compared with historical data

In the current study in Årdal, 32 % red deer had F-level exceeding UNL x 3 and DF was diagnosed in 23 %. The corresponding numbers in the three previous survey periods are shown in Table 9. It must be stressed that the age distribution in the various periods differed (Table 6) and therefore comparisons must be done with some caution. Thus, the table also includes the UNL x 3 for 1.5-year-old animals separately in the four periods. In addition, there is some variation between the periods regarding the number of animals examined from the various hunting areas.

If we compare the percentage of red deer with F-level exceeding the UNL x 3, demonstrating indisputable F-exposure, it shows a clear decreasing trend from the 90s until 2020 (Table 9). On the other hand, the percentage of animals with DF is at the same level as in the 1990s. The explanation is most probably that due to the large increase in the red deer population and higher density, more animals lives closer to the plant than earlier. In the ESPIAL vegetation study (Børja et al. 2021), plants at various sampling locations close to the smelter had signs of browsing. The maximum F-level in 1.5-year-old red deer was clearly higher in the last two periods compared to those in the 1990s (Table 6), showing that some individuals now inhabit areas where the vegetation is very F-polluted. Higher population density in areas close to the smelter lead to more animals being exposed to higher F-exposure than earlier, despite some reduction in the F-emissions. Also, the proportion of the more bioavailable gaseous F of the total F-emissions is currently higher than in the 1990s.

***Table 9**: The number and percentage of red deer from Årdal with F-concentration exceeding the upper normal level x 3 (> UNL x 3) and dental fluorosis (DF) given for the monitoring periods 2017-20, 2013-16, 1995-98, and 1990-93 (the Effect Study).*

Sampling year	Number analysed for F	>UNLx3 n (%)	Number of 1.5-year-old examined for F	1.5-year-old > UNL x 3 n (%)	Number examined for DF*	DF n (%)
2017-20**	106	34 (32 %)	98	32 (31 %)	104	24 (23 %)
2013-16	256	102 (40 %)	76	30 (39 %)	209	61 (29 %)
1995-98	74	44 (59 %)	14	7 (50 %)	61	14 (23 %)
1990-93	95	50 (53 %)	34	16 (47 %)	82	18 (22 %)

** To be able to compare with earlier periods, calves were excluded in the examination for DF.*
*** The majority of the animals were 1.5-year-old.*

3.1.6 Current state Hydro Årdal

In the area at a distance of 12-13 km from the smelter, cervids like deer and moose are subject to F-exposure that causes clearly increased F-level in the skeleton and dental fluorosis in a significant amount of animals. Some individuals from the location very close to the smelter (Moa) show signs of severe F-exposure. Fardalen, Hæreid and Hjelle are other hunting areas with marked F-exposed cervids. The DF documented in young cervids was mainly category 3 and 4 (moderate to marked). Only a few animals showed gross signs of osteofluorosis. More animals seem to be at risk of F-exposure compared to the early 1990s due to increased red deer population and density.

3.2 Hydro Sunndal

3.2.1 Site description

The Sunndal plant lies at Sunndalsøra, at the mouth of the river Driva. The Sunndal valley is a long, deep, U-shaped valley flanked on both sides by mountains 1,000-1,800 meters high. The prevailing wind directions are up the valley in summer and during the day, and down the valley in winter and at night. There is extensive agriculture in the flat 30 km long valley between Sunndalsøra and Gjøra. Grain and potatoes are the main products, and there are some livestock farms for both meat and milk production.

The Sunndal plant started up in 1954 with one potline (SU1), and the next (SU2) commenced operations in 1958, both were based on Søderberg technology. In 1968, SU3 was started, based on prebake technology. Another expansion came in 2002-04, when a new prebake line (SU4) gradually replaced SU1 and SU2. Following the financial crisis, SU3 was temporarily closed in 2009 and gradually restarted in 2011-15. In 2017, 407 000 tonnes of aluminium was produced.

With the exception of 2019 (108 tonnes per year), the F-emissions the last 5 years have been around 85 tonnes per year which are down about 15% compared to the early 90s (Figure 14). However, Søderberg was replaced with prebake technology in the early 2000s, and the proportion of gaseous F (more bioavailable) of the total F-emissions therefore increased. The F-emissions from the cast house are not included in Figure 14. Taking those into account, the total F-emissions from Hydro Sunndal was 127 tonnes in 2019 (norskeutslipp.no, numbers from 2020 not given), thus at the level of the early 90s.

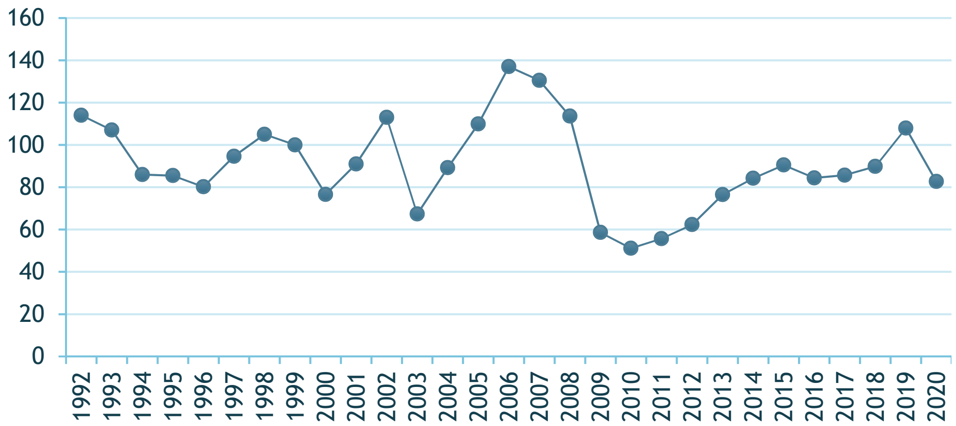

***Figure 14**: Fluoride emissions (tonnes per year) from Hydro Sunndal from 1992 to 2020. Source: AMS.*

3.2.2 The deer population in Sunndal municipality

The red deer population has shown an large increase from the early 1990s to 2020, reflected by a large increase in hunted animals, from 120 red deer in 1991 to 500 in 2020 (Figure 15). Migration pattern of red deer monitored by GPS in Sunndal municipality indicate that many animals are relative stationary within the various valleys, showing relatively short migration within the valley itself. The adult red deer in the Sunndal valley lives there the whole year and the population is so large that a reduction is recommended (Forvaltningsplan for hjortevilt i Sunndal municipality 2019-22 – høyringsutkast). According to LWM, there are more red deer in the lower parts of the Sunndal valley nowadays. Earlier, it was very uncommon to observe red deer further down of Furu (3.5 km from smelter).

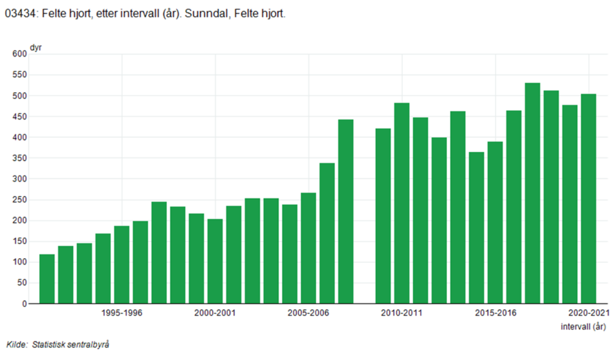

***Figure 15**: Number of hunted red deer in Sunndal municipality from 1991 to 2020. Source: Statistics Norway (www.ssb.no).*

The roe deer population in Sunndal is moderate due to predation and some winters with a lot of snow that causes increased deaths. In the period 2005 to 2018, the number of hunted roe deer in Sunndal municipitality was between 45 to 85, except in 2007 when 160 were shot (Forvaltningsplan for hjortevilt i Sunndal municipality 2019-22 – høyringsutkast). Nowadays it is less roe deer hunting than earlier probably due to extension of the hunting season for red deer, a more preferred game.

3.2.3 Studies of fluoride exposure in wild cervids, 2019-20

Based on the status for F-exposure of animals in Sunndal presented in WP1 (Vikøren 2019), the current study was limited to the main valley Sunndalen and the side valley Grødalen, as well as Litledalen/Trædal, in the following named "Sunndalen" if not otherwise described (Figure 16). The results are presented according to zones used by Hydro Sunndal in their monitoring of vegetation. The zones are:

1. Trædal – Litledalen
2. Øvre Sunndal
3. Fahle bru – Ottem bru
4. Grøa – Fahle bru
5. Sunndalsøra – Grøa
6. Gråura – Jenstad – Svisdal
7. Opdøl – Ålvundeid – Stomsvik
8. Viklandet – Melkild – Jordalsgrenda

This study comprises the zones 1-6.

Figure 16: *Map of Sunndalen showing the main area for the Espial Fauna study and the sampling points for grass in the vegetation monitoring performed by the Hydro Sunndal. In the courtesy of Kontaktutvalet v/Jan Olav Polden.*

Lower jawbones from 46 red deer and 12 roe deer were collected 2019-20 (Table 10) in collaboration with the LWM and hunters and included hunted animals and fallen stock (traffic killed, other casualties).

According to the LWM, very few roe deer are shot during ordinary hunting in Sunndalen. Thus, the main indicator was 1.5-year-old red deer (n = 34), with supplementary data from other age groups of red deer (n = 12) and from roe deer (n = 12).

***Table 10**: The number of wild cervids examined for fluoride exposure in Sunndalen 2019-20.*

Species	Year	Number examined	Comment
Red deer	2019	11	1.5-year-old: 9
	2020	35	1.5-year-old: 25
	Total	46	1.5-year-old: 34
Roe deer	2019	6	Varying age
	2020	6	Varying age
	Total	12	Varying age
All cervids	**Total**	**58**	

3.2.3.1 Bone F-concentration in red deer

The F-level showed individual variation (Table 11). In Figure 17 the age distribution of F-level is shown, whereas Figure 18 show the F-levels in the various areas. The highest F-concentration was found in the oldest red deer and mainly from the Sunndal valley zone 3-5.

***Table 11**: The fluoride (F) concentration (mg F/kg bone ash) in the lower jawbone (mandible) of red deer from Sunndalen given for the monitoring periods 2019-20, 2017-18, and 1990-93 (the Effect Study). The F-level is given for each age group with mean, minimum and maximum. The individual data are registered in the ESPIAL Fauna database (updated version).*

Sampling year		0.5 year	1.5 year	2.5 year	3.5-5.5 year	6.5 year and older
2019-20	Number	2	34	2	4	4
	Mean	597	717	856	2,738	2,566
	Min	356	208	600	2,046	1,118
	Max	838	1,577	1,112	3,677	4,645
2017-18*	Number	0	1	2	5	9
	Mean	-	1,314	1,858	1,618	2,884
	Min	-	-	1,202	786	1,347
	Max	-	-	2,514	3,505	4,295
1990-93	Number	24	28	23	26	6
	Mean	447	669	853	1,818	2,707
	Min	67	125	155	431	802
	Max	1,479	1,755	2,528	4,035	5,457

** Two adult red deer were not aged and are thus not included.*

In the present study, 20 (43 %) red deer had F-level exceeding UNL x 3 (Table 12), thus showing indisputable F-exposure. These were mainly from Sunndalen, zone 2-5 (16/27 = 59 %) while the other were from Grødalen and zone 6 (2/11 = 18 %) and from unknown location.

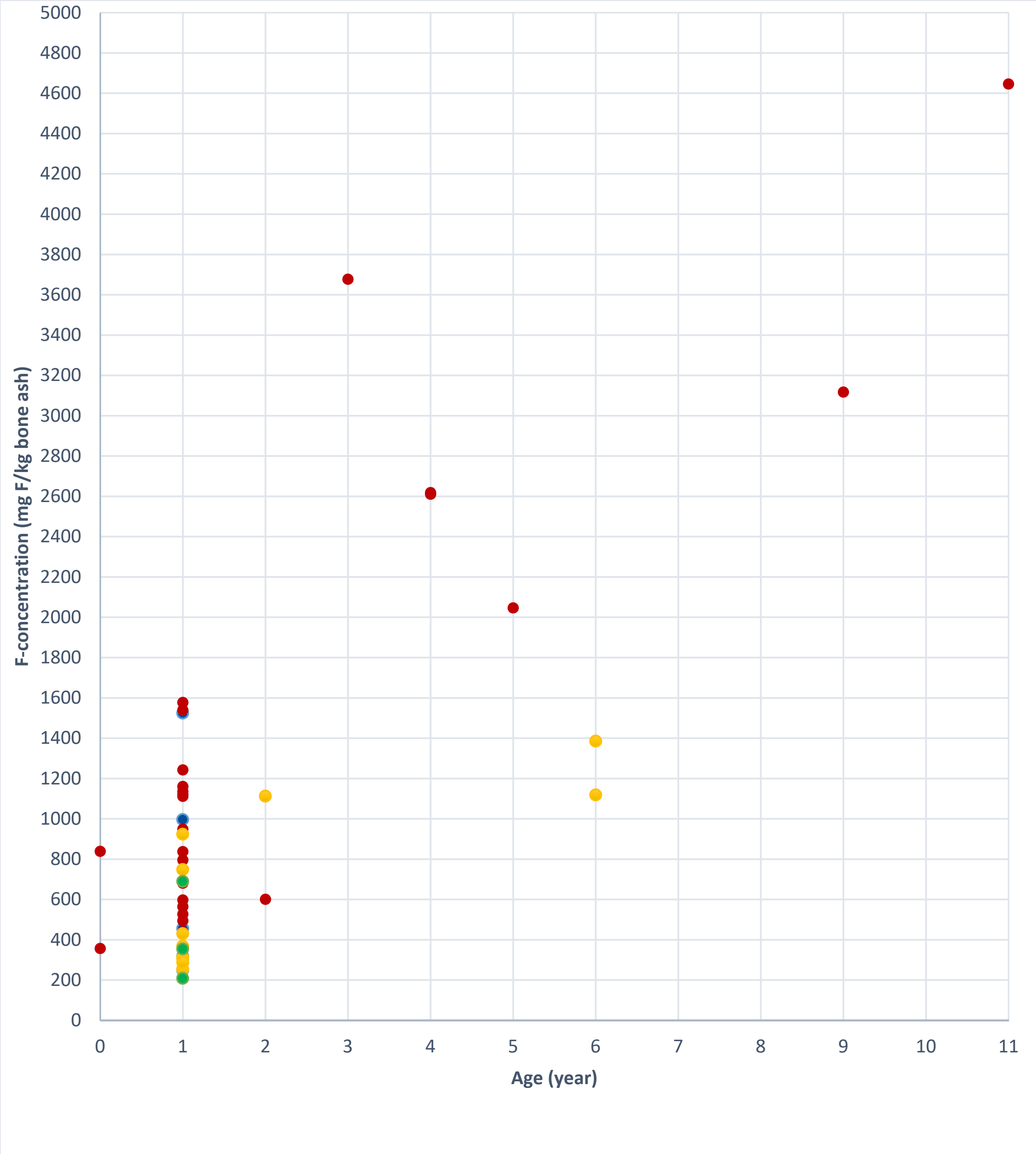

Figure 17*: Age and F-concentration (mg F/kg) in the lower jawbone (mandible) of red deer from Sunndalen 2019-20. The various areas is shown with different colours; red: Sunndalen (zone 2-5), n = 27; yellow: Grødalen and zone 6, n = 11; green: Litledalen/Trædal (zone 1), n = 3; and blue: unknown, n = 5. The 1.5-year-old group is shown in more detail in Figure 19 (below).*

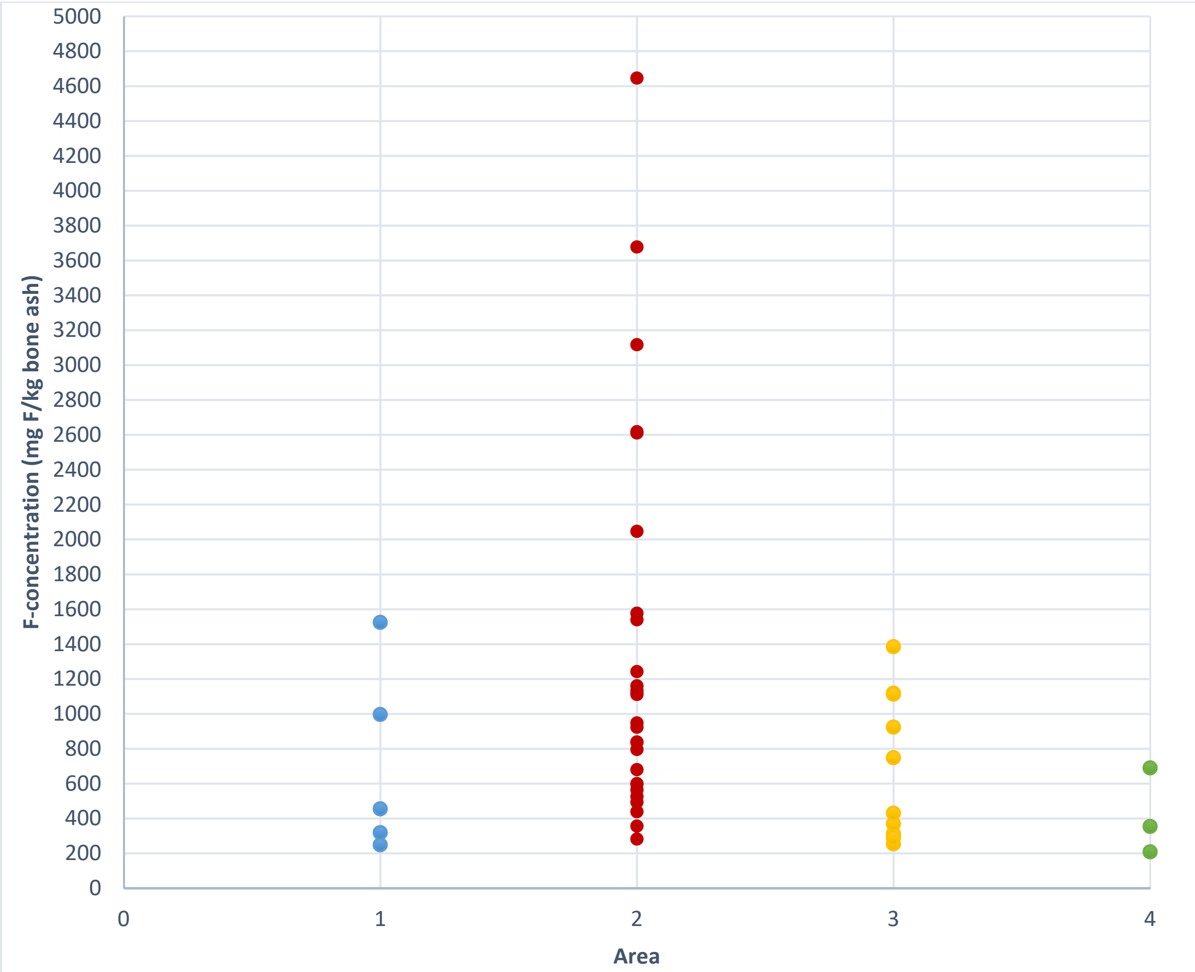

Figure 18: *F-concentration (mg F/kg) in the lower jawbone (mandible) of red deer (all age groups) from Sunndalen 2019-20, given for the various areas; 1) unknown, 2) Sunndalen (zone 2-5), 3) Grødalen and zone 6 og 4) Litledalen/Trædal (zone 1).*

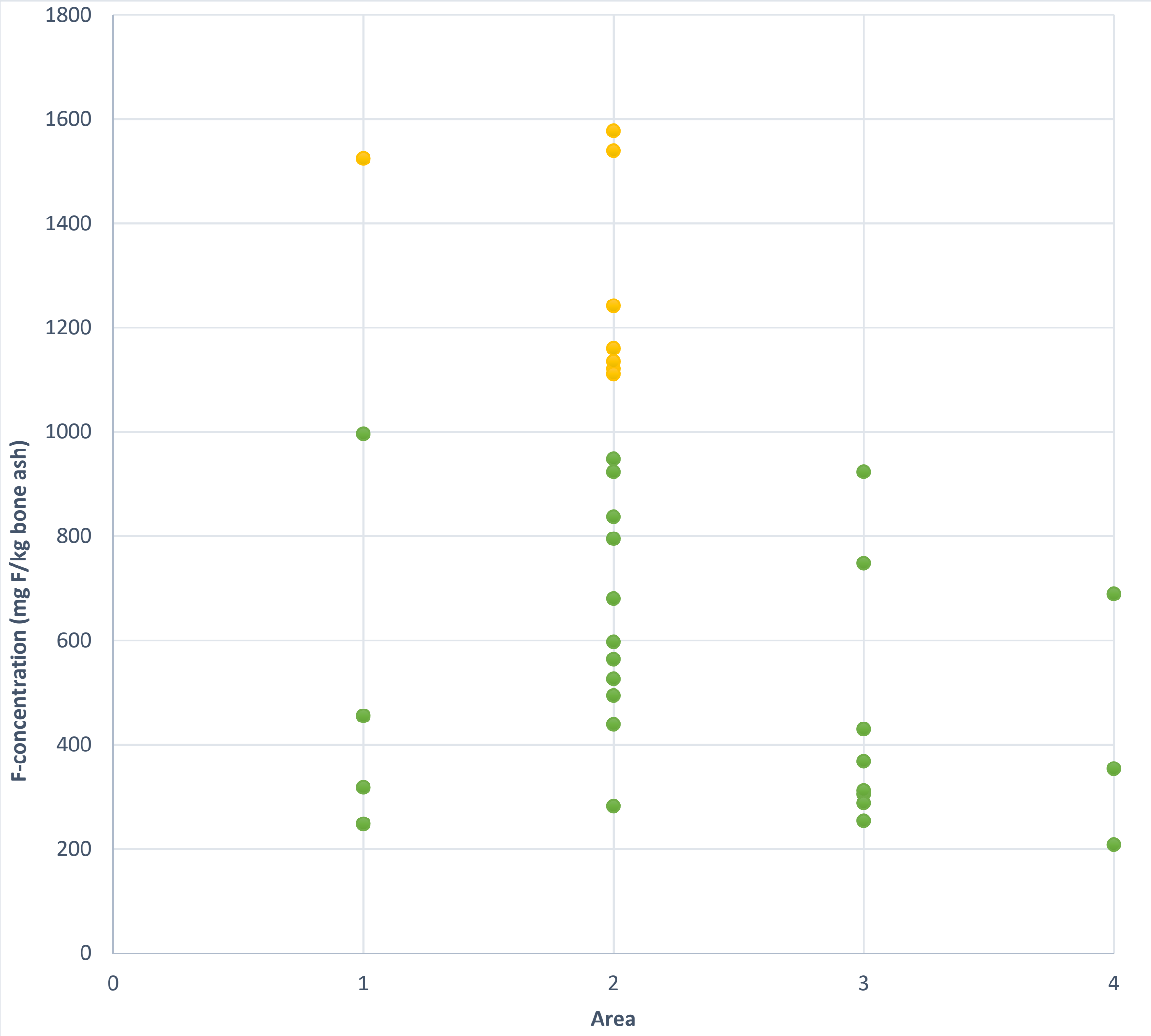

Figure 19: *F-concentration (mg F/kg) in the lower jawbone (mandible) of 1.5-year-old red deer from Sunndalen 2019-20, given for the various areas; 1) unknown, 2) Sunndalen (zone 2-5), 3) Grødalen and zone 6, og Litledalen/Trædal (zone 1). The individuals are marked with colour according to the fluoride dose - effect relationship for 1.5-year-old red deer: green < 1,000, yellow 1,000-2,000, and red > 2,000 mg F/kg bone ash (Table 3).*

According to the fluoride dose – effect relationship for 1.5-year-old red deer (Table 3), eight animals (24 %) had F-levels between 1,000 – 2,000 mg F/kg (yellow group = risk of developing DF) and none had F-level above 2,000 mg F/kg (red group = DF) (Figure 19). One in the risk group was from an unknown location whereas the remaining seven were from the Sunndal valley, zone 3-5 (lower to middle part of the valley; up to Fahle).

3.2.3.2 Detrimental effects of F in red deer

Dental fluorosis: The examination of the teeth showed that 13 red deer (28 %) had DF, one in category 4 (marked effect) and the rest in category 3 (slight to moderate effect). The animals with DF were from the Sunndal valley, zone 3-5 (lower to middle part of the valley; up to Fahle) except two (1.5-year-old) of unknown location. Ten red deer with DF were 1.5-years-old and the remaining three were three, four and 11 years old, respectively. The 1.5-year-old with DF includes all eight animals in the yellow group and two in the green group (< 1,000 mg F/kg) in Figure 19. The two latter had F-levels above 920 mg F/kg.

Osteofluorosis: None of the red deer had gross signs of OF in the mandible.

3.2.3.3 Bone F-concentration and detrimental effects of F in roe deer

The F-level showed large individual variation. In Figure 20, the age distribution of F-level is shown, whereas Figure 21 shows the F-levels in the various areas. The highest F-levels were found in the oldest roe deer mainly from the Sunndal valley zone 2-5, similar to the findings in red deer.

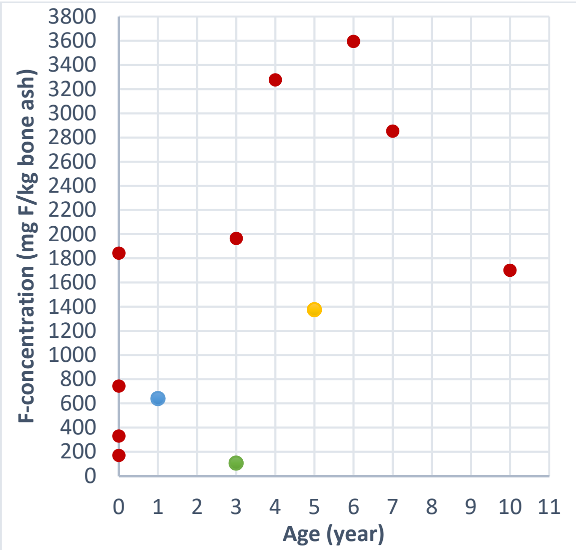

Figure 20*: Age and F-concentration (mg F/kg) in the lower jawbone (mandible) of roe deer from Sunndalen 2019-20. The various areas is shown with different colours; red = Sunndalen (zone 2-5), yellow = Grødalen and zone 6, green = Litledalen/Trædal (zone 1) and blue = unknown.*

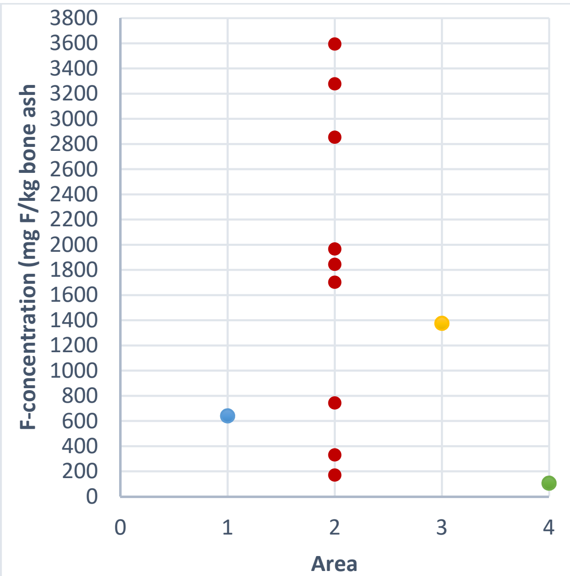

Figure 21*: F-concentration (mg F/kg) in the the lower jawbone (mandible) of roe deer from Sunndalen 2019-20, given for the various areas; 1) unknown, 2) Sunndalen (zone 2-5), 3) Grødalen and zone 6 og Litledalen/Trædal (zone 1).*

In the new study, five (42 %) roe deer had F-level exceeding UNL x 3, thus showing indisputable F exposure. All were from the Sunndal valley, zone 2-5.

Dental fluorosis: The examination of the teeth showed that four roe deer (33 %) had DF, all in category 3. The animals with DF were from Sunndalen, three from zone 5 (lower) and one from zone 4 (middle: Hoåslykkja, Figure 22). One roe deer with DF from zone 5 were a calf and the remaining three were four, six and seven years old, respectively.

Osteofluorosis: None of the roe deer had gross signs of OF in the mandible.

***Figure 22**: The six cheek teeth of the right lower jaw of a roe deer, 7.5-year-old female, from Hoåslykkja in Sunndalen (zone 4). Dental fluorosis of category 3; brown discoloration of enamel of several of the cheek teeth. The fourth teeth from the left (M1) has normal enamel.*

3.2.4 Local differences in F-exposure of cervids related to vegetation data

Indisputable F-exposure (>UNL x 3) was documented in red deer and roe deer from the Sunndal valley (zone 2-5), particularly the lower and middle part up to Fahle (zone 3-5). All red deer examined in zone 5 and 4 (Sunndalsøra up to Fahle bridge) had F-level exceeding UNL x 3. Dental fluorosis was found in cervids from the Sunndal valley zone 3-5, thus not in the upper part of the valley. The F-exposure seems to be low in cervids from Grødalen + zone 6 and Litledalen/Trædal (zone 1) and DF was not found in cervids from these areas. However, few animals were examined from zone 1, thus some caution in interpretation must be taken.

The ESPIAL vegetation survey (Børja et al. 2021) included sampling at four and six locations in the Sunndal valley (zone 2-5) in 2019 and 2020, respectively. As in Årdal, ferns had very high F-content and the possible significance as F-source for deer is discussed earlier (3.1.4). We have chosen to relate our results to the F-level in rowan, a preferred browsing plant for cervids, even though quantitatively of less importance. We have also included blueberry that normally is eaten in larger amounts especially during the autumn. The vegetation survey showed that the F-level in rowan was way over the tolerance threshold for cervids (30 mg F/kg) at all sampling sites up to Gravem (zone 2-5) in 2019. The same result was found in 2020, except at the most distant sampling point at Gravem (Figure 3.4, Tables 3.6 and 3.7, Børja et al. 2021). Blueberry was only available for sampling at Hoås (zone 4) and Gravem (zone 2) and the F-level was exceeding the tolerance threshold at Hoås in 2019.

Vegetation monitoring of rowan performed by Hydro Sunndal 2012-20 at four locations in the Sunndal valley up to Gjøra (32 km, zone 2) show F-levels exceeding the tolerance threshold for cervids in the majority of the samples (referred to in Figure 3.8, Børja et al. 2021).

We conclude that there is a correlation between the F-level in vegetation and cervids, showing considerable F-exposure in the lower half of the Sunndal valley, at a level that may cause DF. According to the LWM, the red deer is relative stationary showing migration within the valley itself.

3.2.5 New results compared with historical data

In the current study comprising Sunndalen, 43 % red deer and 42 % roe deer had F-level exceeding UNL x 3, and DF was diagnosed in 28 % and 33 %, respectively. The corresponding numbers in the 2017-18 study and the Effect Study (1990-93) are shown in Table 12. It must be stressed that the age distribution in the various study periods differed and therefore comparisons must be done with some caution. Thus, in the 2017-18 survey, the majority of red deer was in the two oldest age groups, thus representing individuals being F-exposed back to the 2000s. That might explain the high percentage of red deer with F-level > UNL x 3 and DF.

If we compare the 1.5-year-old red deer in the current study and the Effect Study, the number of animals examined, the bone F-levels, and the > UNL x 3 are relatively equal (Tables 11 and 12). On the other hand, the percentage of animals with DF is much higher in the 2019-20 study. The proportion of gaseous F of the total F-emissions is currently higher than in the 90s. Gaseous F is more bioavailable than particulate F. However, the main explanation is most probably that the current study includes more animals that have lived closer to the plant, thus the risk of being exposed to periods of high F-exposure causing DF is higher than if you live further from the plant and is exposed to a more moderate and even F-exposure. The red deer population has shown a 4-fold increase from the 1990s to 2020 and the density of animals has increased. According to LWM, there are more red deer in the lower parts of the Sunndal valley nowadays. In earlier times, it was very uncommon for red deer to use areas further down of Furu (3.5 km from smelter).

Table 12: *The number and percentage of red deer and roe deer from Sunndalen with F-concentration exceeding the upper normal level x 3 (> UNL x 3) and dental fluorosis (DF) given for the monitoring periods 2019-20, 2017-18, and 1990-93 (the Effect Study).*

Sampling year	Species	Number examined	>UNLx3 n (%)	DF n (%)
2019-20	Red deer	46	20 (43 %)	13 (28 %)
	Roe deer	12	5 (42 %)	4 (33 %)
2017-18*	Red deer	19	13 (76 %*)	7 (41 %)
1990-93	Red deer	107	42 (39 %)	3 (3 %)
	Roe deer	29	**	3 (10 %)

** Two red deer not aged were excluded. No roe deer from Sunndalen was examined. ** numbers not available for Sunndalen, only for the whole Sunndal municipality: 14% had F-level > UNL x 3.*

3.2.6 Current state Hydro Sunndal

In the lower and middle part of the Sunndal valley, roe deer and red deer are subject to F-exposure that causes increased F-level in the skeleton and dental fluorosis in a significant amount of animals. More animals seem to be at risk of F-exposure compared to the early 1990s due to increased red deer population and density. The DF currently documented in young cervids mainly was in category 3 (slight to moderate). Osteofluorosis was not found in the present study.

3.3 Hydro Husnes

3.3.1 Site description

Hydro Husnes is located in Kvinnherad municipality at an isthmus between the bay Husnesvågen and the lake Opsangervatnet, at the mouth of the Hardangerfjord. The area is relatively open, but directly to the east of the aluminium plant lies a mountainous area with peaks up to 1000 m high (part of Husnes hjortevald). North and west of the plant is the Hardangerfjord. The mountains affect the wind directions, which are mainly north/south. A northerly wind prevails in spring, and a southerly wind the rest of the year as a rule. The area has a mild coastal climate, with relatively high precipitation.

The plant started up in 1965 under the name Sør-Norge Aluminium (Søral) with Alusuisse as main owner. Hydro Aluminium later came in as a part owner and since 2014 as 100% owner. The plant originally had two potrooms, both prebake. Potroom B was temporarily shut down in 2009, but a decision to reconstruct it was taken in 2018, aiming at restart in 2020. Production capacity is 94,000 tonnes p.a., which will increase to 195,000 tonnes when potroom B is in full production.

The fluoride emissions (tonnes per year) in 2017 (34 tonnes) were approximately 25% lower than in the early 1990s (ca 50 tonnes). In 2018-20, the F-emissions increased to a level comparable to the early 90s due to increased production (Figure 23).

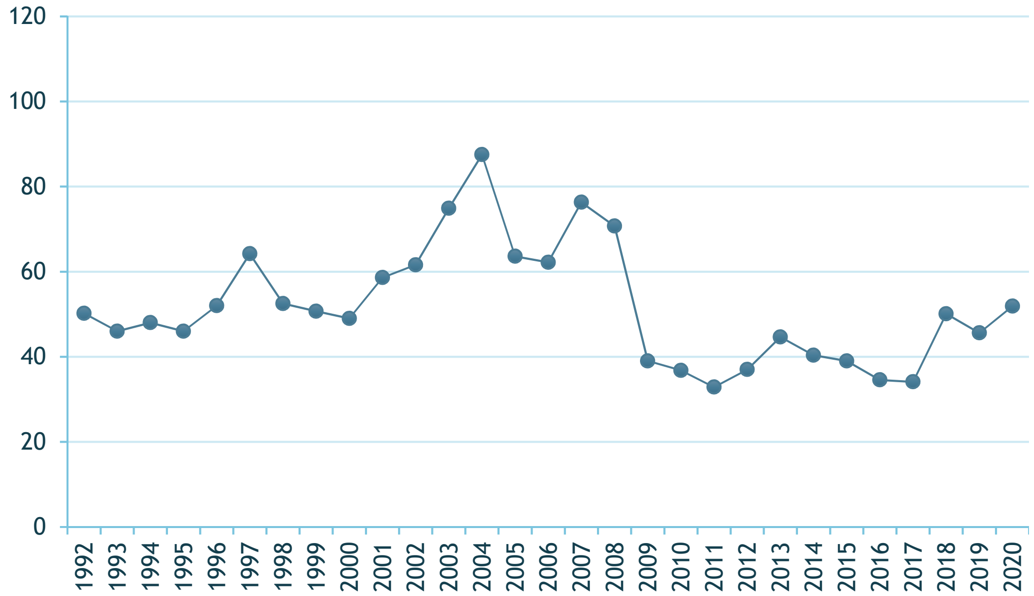

***Figure 23**: Fluoride emissions (tonnes per year) from Hydro Husnes from 1992 to 2020. Source: AMS.*

3.3.2 The red deer population in Kvinnherad

The red deer population in Kvinnherad is among the largest in the country based on hunting bag. In 1993-2020, the number of hunted animals increased more than a 3-fold from 436 to 1427 animals (Figure 24). This reflects a large increase and indicates a denser population in the area. It is reason to believe that more animals live closer to the plant than in the 1990s, and thus might be exposed to F-emissions.

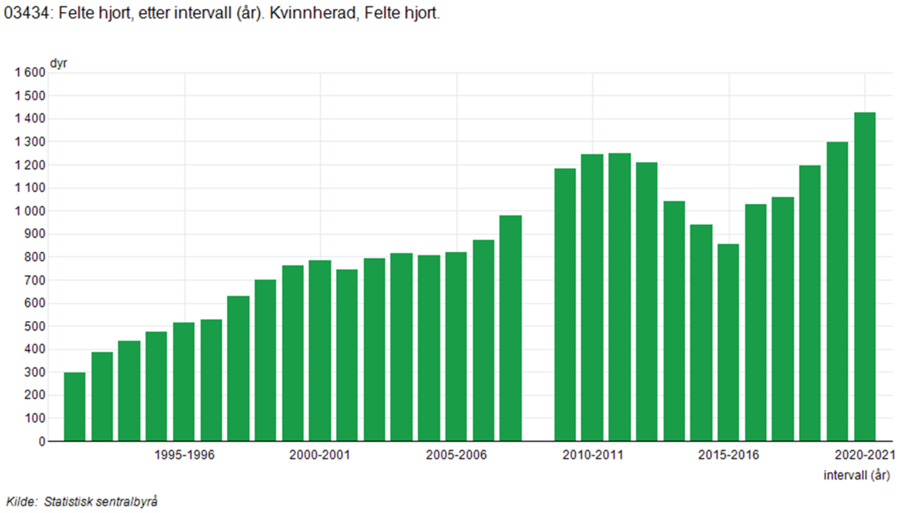

***Figure 24**: Number of hunted red deer in Kvinnherad municipality from 1991 to 2020. Source: Statistics Norway (www.ssb.no).*

3.3.3 Studies of fluoride exposure in red deer, 2019-20

Based on the status for F-exposure of animals in Husnes presented in WP1 (Vikøren 2019), the current study was limited to an area comprising a radius of approximately 10 km from the smelter. The indicator was 1.5-year-old red deer, monitoring the impact of F-emissions in the period 2018-20.

Hunters collected lower jawbones from red deer during the ordinary hunting seasons 2019-20. The collection was a cooperation between LWM, the Norwegian Institute for Nature Research (NINA) and NVI.

Totally 69 red deer, all 1.5-year-old, were examined: 47 sampled in 2019 and 22 in 2020. The 2019 material comprised the 10 km radius area, whereas the 2020 material was selected from the hunting areas (vald) in which animals exceeded 1,000 mg F/kg in 2019 (Husnes hjortevald, Kaldestadåsen, Øvre Feet/Døssland).

3.3.3.1 Bone F-concentration in red deer

The F-level showed large individual variation (Table 13, Figure 25). Sixteen (23 %) red deer yearlings had F-level exceeding UNL x 3 (810 mg F/kg), thus showing indisputable F-exposure. These were shot in the following hunting areas: Husnes hjortevald (10/26 examined = 38 %) Kaldestadåsen (2/6 = 33 %), Øvre Feet-Døssland in Uskedalen (2/6 = 33 %), Raudstein-Langgåt (1/1) and one unknown (Figure 25). The F-level in the other hunting areas was with a few exceptions low. The individual large variation in F-level in the hunting areas Husnes hjortevald, Kaldestadåsen and Øvre Feet-Døssland reflects that red deer to a variable degree is stationary.

According to the fluoride dose – effect relationship for 1.5-year-old red deer (Table 3), six animals had F-levels between 1,000 – 2,000 mg F/kg (risk of developing DF) and one had F-level above 2,000 mg F/kg (DF) (Figure 25).

***Table 13**: The F-concentration (mg F/kg bone ash) in the lower jawbone (mandible) of red deer from Kvinnherad (Hydro Husnes) given for the monitoring periods 2019-20 (10 km radius from smelter) and 1991-93 (the whole municipality, the Effect Study). The F-level is given for each age group with mean, minimum and maximum. The individual data is registered in the ESPIAL Fauna database (updated version).*

Sampling year		0.5 year	1.5 year	2.5 year	3.5-5.5 year	6.5 year and older
2019-20*	Number	-	69	-	-	-
	Mean	-	434	-	-	-
	Min	-	48	-	-	-
	Max	-	2,607	-	-	-
1991-93	Number	32	53	35	79	56
	Mean	161	258	401	716	907
	Min	31	51	119	154	138
	Max	767	1,158	1,762	8,433	5,761

** In 2019-20, the sampling area was restricted to a 10 km radius from the smelter.*

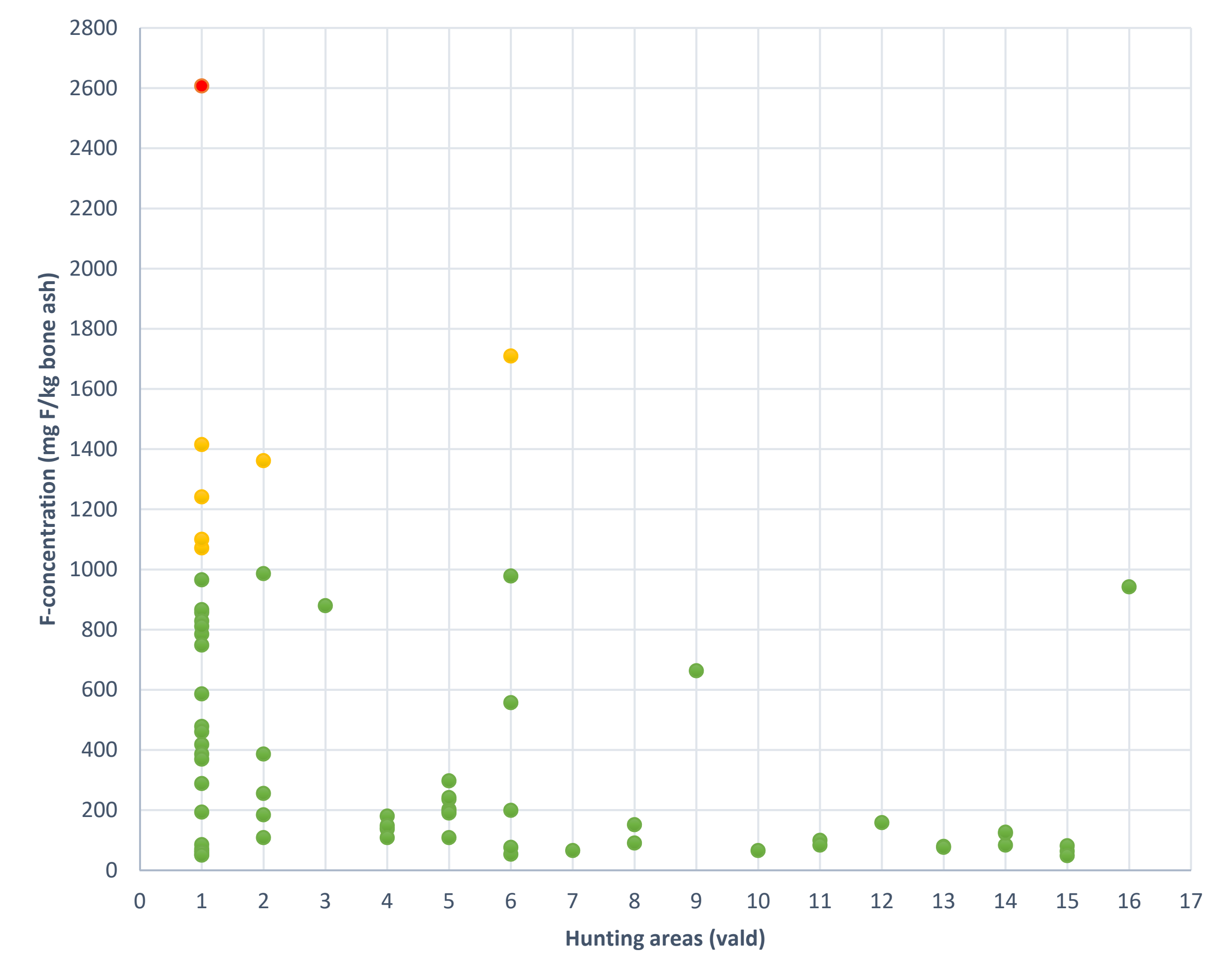

***Figure 25**: F-concentration (mg F/kg) in the lower jawbone (mandible) of 1.5-year-old red deer from Kvinnherad (Hydro Husnes) 2019-20, given for the various hunting areas: 1) Husnes, 2) Kaldestadåsen, 3) Raudstein - Langggåt, 4) Herøysundet, 5) Valen - Handeland, 6) Øvre Feet - Døssland, 7) Musland, 8) Kjærland - Myklebust - Ytre Rød, 9) Eikeland, 10) Høylandsbygd, 11) Halsnøy, 12) Jonsåsen, 13) Holmedal, 14) Giskafjell, 15) Indre Matre and 16) unknown. The individuals are marked with colour according to the fluoride dose - effect relationship for 1.5-year-old red deer: green <1,000, yellow 1,000-2,000, and red > 2,000 mg F/kg bone ash (Table 3).*

3.3.3.2 Detrimental effects of F in red deer

Dental fluorosis: Nine (13 %) red deer had DF, one in category 4 (marked) and eight in category 3 (slight to moderate). These includes all animals with F-level exceeding 1,000 mg F/kg (yellow and red dots in Figure 25) and two with F-level between 940 and 1,000 mg F/kg (green dots). The red deer with DF were shot in the two closest hunting areas to the smelter Husnes hjortevald (5/26 examined = 19 %) and Kaldestadåsen (2/6 examined = 33 %), except one from Døssland in Uskedalen and one from an unknown locality.

Osteofluorosis: None of the red deer had gross signs of OF in the mandible.

3.3.4 Local differences in F exposure

The examined animals shot in 2019-20 are an indication of the F-exposure from spring 2018 (birth year of those shot in 2019) until late autumn 2020 in an area comprising a radius of approximately 10 km from the smelter. Indisputable F-exposure was found in approximately 1/3 of the red deer from the hunting areas closest to the plant (Husnes hjortevald 38% and

Kaldestadåsen 33 %) and in a few animals from a further distance. The latter is most probably due to migration after the main F-exposure have taken place closer to the smelter. The F-exposure had caused dental fluorosis in 13 % of the animals, mainly close to the smelter. The F-level was generally very low in animals from the other hunting areas. Recent data of F-level in vegetation have not been available. The Effect Study showed that the F-level in rowan foliage exceeded the F-tolerance level of red deer (30 mg F/kg) in an area comprising approximately 3 km north and south of the smelter (Figure 22 in Horntvedt and Øyen 1994). In 2018-20, the F-emissions have been at the level of the Effect study period.

3.3.5 New results compared with historical data

In the Effect Study the main conclusion was that it was detected high F-levels and DF in some individual red deer, mainly close to the smelter. In Table 13 the results are shown for the two periods 2019-20 and 1991-93 (the Effect Study). It has been no monitoring of animals in the period 1994-2018. The results are not directly comparable since the 2019-20 material was restricted to a 10 km radius from the smelter, whereas the 1991-93 material comprised the whole municipality. Unfortunately, the details from the Effect Study are not available, thus it was not possible to use data from the 10 km radius zone only for comparison with the new results.
The proportion of animals with F-level exceeding UNL x 3 was 23 % (yearlings, 10 km radius) in 2019-20 and 6.7 % (all ages, the whole municipality) in 1991-93. A higher number of red deer had DF in 2018-20 than in 1991-93 (n = 3, category 4 and 5, >1.5-year old). As already mentioned, these two groups are not directly comparable. However, comparing 1.5-year-old animals in the two periods, a considerable higher maximum F-level was detected in 2019-20.

3.3.6 Current state Hydro Husnes

The F-emissions the last three years are comparable to the level in the 1990s. The current study was designed to better monitor the area close to the smelter. It shows that the F-exposure of red deer is restricted to an area of approximately 3 km radius of the smelter. Approximately 1/3 of the young red deer within this area showed indisputable F-exposure, particularly in south and southwesterly direction. The DF found was with one exception in category 3 (slight to moderate). Red deer having their main living areas further from the plant seem not to be affected by F-emissions. However, F-affected animals might also be found at a further distance due to migration.

In conclusion, the current F-exposure of red deer seem to be at the level found in the Effect Study, but due to the population increase more individuals seem now to be at risk of being exposed than in the early 1990s.
In view of the restart of potroom B and expected increase of F-emissions, further monitoring of the red deer population is recommended.

3.4 Hydro Karmøy

3.4.1 Site description

Hydro Karmøy is located at Karmsundet, the sound at the east side of Karmøy Island. The surrounding area consists of a flat or slightly undulating coastal landscape dominated by farmland, low hills and heather-covered slopes.

The climate is mild, windy and humid. The prevailing wind is from the north-west in spring and summer. In autumn and winter, the wind direction is more variable, but it tends to be easterly or southerly. The flat terrain and wind cause emissions to disperse rapidly, so that the area affected is relatively small.

Hydro Karmøy was started in 1967 based on Søderberg technology. Since then, a prebake line was built in three construction stages in 1982, 1987 and 1997. The total capacity reached 290,000 tonnes per year in 2008 before the Søderberg lines were shut down in 2009. In 2018 a new potline with a capacity of 75,000 tonnes per year came in production. This is a full scale pilot stage applying the latest technologies, and is prepared for further expansion up to 330,000 tonnes per year. This could bring total production up to 530,000 tonnes per year, the largest smelter in Northern Europe.

Dry and wet scrubbers are used to treat the off-gases from the pots. F-emissions in 2017 was 76 tonnes per year and due to the new potline, they increased to 89 tonnes in 2020, which is at the level of the early 1990s (Figure 26). The proportion of gaseous F (more bioavailable) of the total F-emissions increased from 2009 when Søderberg lines were closed.

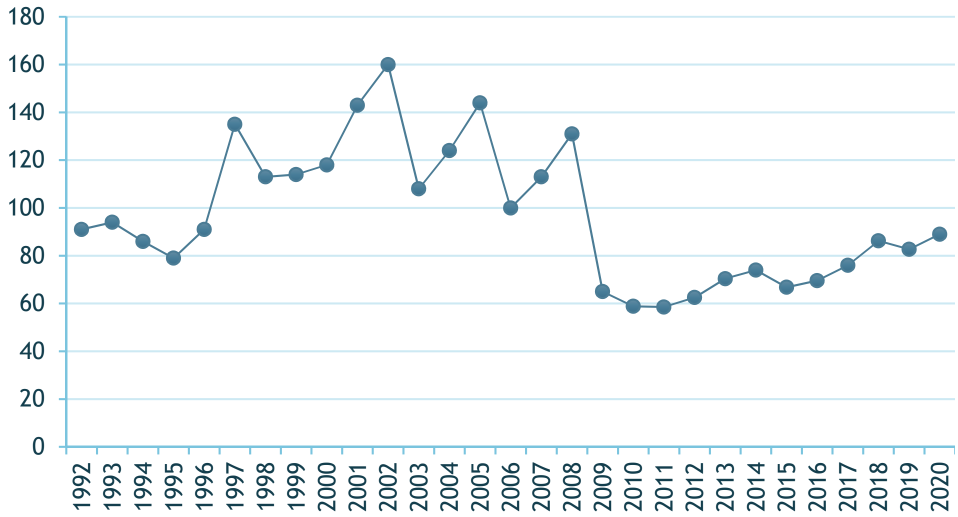

Figure 26*: Fluoride emissions (tonnes per year) from Hydro Karmøy from 1992 to 2020. Source: AMS.*

3.4.2 The deer population in Karmøy

The red deer population in Karmøy municipality is small and the number of hunted animals increased from 15 in 1991 to 32 in 2020. The roe deer population is larger and 160 animals were hunted in 2017 (Statistics Norway, www.ssb.no). Roe deer lives close to and within the fenced area around the smelter.

3.4.3 Studies of fluoride exposure in deer, birds and livestock, 2019-20 (21)

The collection of materials for the study was particularly challenging in Karmøy. In 2019, the collection was primarily based on hunter involvement during the ordinary hunting season, but it came out with no result.

There is no ordinary deer hunting within the area of Hydro Karmøy. Several groups of roe deer were living within this fenced area. The fence is so high that it is not possible for roe deer to leave the area. A special licence was given by the Karmøy municipality to hunt roe deer within the area around the smelter after an application from NVI in cooperation with Hydro. Three

roe deer were shot in November 2019. Additionally, a roe deer fawn found dead on the premises in October 19 was also included in the study.
Based on the results that showed excessive F-exposure in the four roe deer (see below), the NVI recommended euthanasia of all deer grazing within the fenced area around the smelter. Thus, a second application was sent to Karmøy municipality in 2020, and license given. The remaining six roe deer and a red deer were shot during the autumn, and additionally two roe deer fawns found dead were included in the study.

In 2020, a new strategy for collection of materials was implemented: Hydro Karmøy put a great effort in sampling material engaging employees and using their network of contacts to collect jaws from cervids. Both hunted and traffic killed animals were sampled. In addition, wild birds and sheep were included, as well as a cow slaughtered in 2021.

Roe deer and red deer: We examined 12 roe deer from within the fenced area around the smelter and 19 roe deer from other areas of Karmøy municipality (Table 14 and Figure 27). The material from the first group were more comprehensive for 11 of 12 roe deer including; the whole carcass of two fawns, and the head and some internal organs of nine roe deer including also the lower limbs in six of the latter.

Additionally, three red deer were examined: one shot within the smelter area (head, lower limbs, and internal organs) and two from other areas (one hunted and one traffic killed).

Livestock: Three sheep from a farm in Kopervik having their grazing area directly south of the plant were included in the study (Figure 31). These sheep are grazing the whole year (utegangarsau/"villsau") in this area. A cow that had its summer grazing area directly northwest of the plant was also included.

Wild birds: Seventeen gulls of various species and ages (8 juvenile and 9 adults) and one oystercatcher (*Haematopus ostralegus*) were collected in 2019 (n = 2) and 2020 within the area of the smelter.

Table 14: *The number of wild animals and sheep examined for fluoride exposure in Karmøy 2019-20 (21). The data is registered in the ESPIAL Fauna database (updated version).*

Species	Year	Number examined	Comment
Roe deer	2019-20	31	Various ages, 12 from smelter site
Red deer	2019-20	3	Various ages, one from smelter site
Sheep	2020	3	Various ages, directly south of smelter area
Cattle	2021	1	10-12 years, directly northwest of smelter area
Birds	2019-20	18	17 gulls and 1 oystercatcher

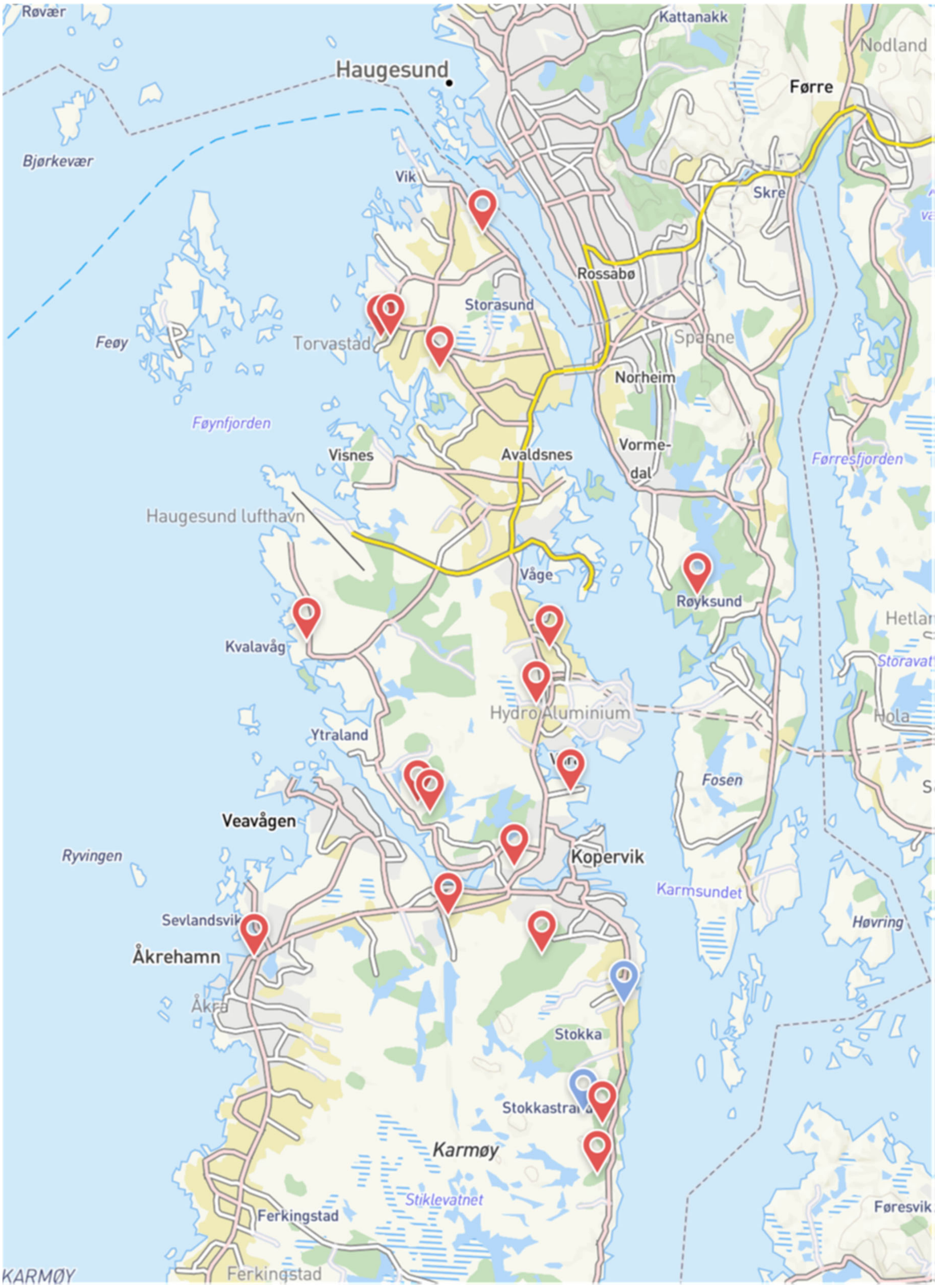

__Figure 27__: Map of Karmøy showing the location of roe deer (red) and red deer (blue) collected outside the premises of Hydro Aluminium Karmøy. For two roe deer the location data was unsufficient. © 2021 Norkart AS/Geovekst og kommunene/NASA, Meti © Mapbox © OpenStreetMap

3.4.3.1 Bone F-concentration in deer

The 19 roe deer and two red deer from areas outside the fenced area of the smelter (Figure 27), showed very moderate F- accumulation (maximum 1,200 mg F/kg) and all had F-level below UNL x 3. Taking the age into account, F-accumulation was highest in two roe deer closest to the smelter at Vorrå and Håvik.

Of the 12 roe deer from within the fenced area, there were three fawns (calves), one 1.5-year-old and eight adults (2.5 – 6.5 year). All except one fawn showed extreme F-accumulation (Figure 29: red and blue dots) and all 12 had F level ≥ UNL x 3. Eight roe deer had F-levels above the maximum previously registered in roe deer in Norway (11,680 mg F/kg).

The 2.5-year-old red deer shot within the fenced area also had high F-level (7,647 mg F/kg, Figure 29: yellow dot).

Antlers from two male roe deer shot in 2020, both 3.5-years-old, were F-analysed for comparison with mandibular F-level. One (R39/20, without OF and DF) had 311 and 4,265 mg F/kg and the other (R32/20, OF and DF cat. 6) had 10,533 and 14,639 mg F/kg in the antler and lower jawbone, respectively (Figure 29). F is incorporated in the developing antler and in roe deer this takes place from January to April each year. The large difference in F-level between antler and jawbone in one of the bucks indicates that the major F-exposure was not in the period of antler growth and mineralization. Thus, it is possible that this animal first entered the fenced area in late spring/early summer of 2020 and that the F-load in the jawbone was due to high F-exposure from May until it was shot in November. The other animal (R32/20) had very high F-levels in both tissues and had OF and excessive DF, which indicate that it had been within the fenced area for a longer time (years).

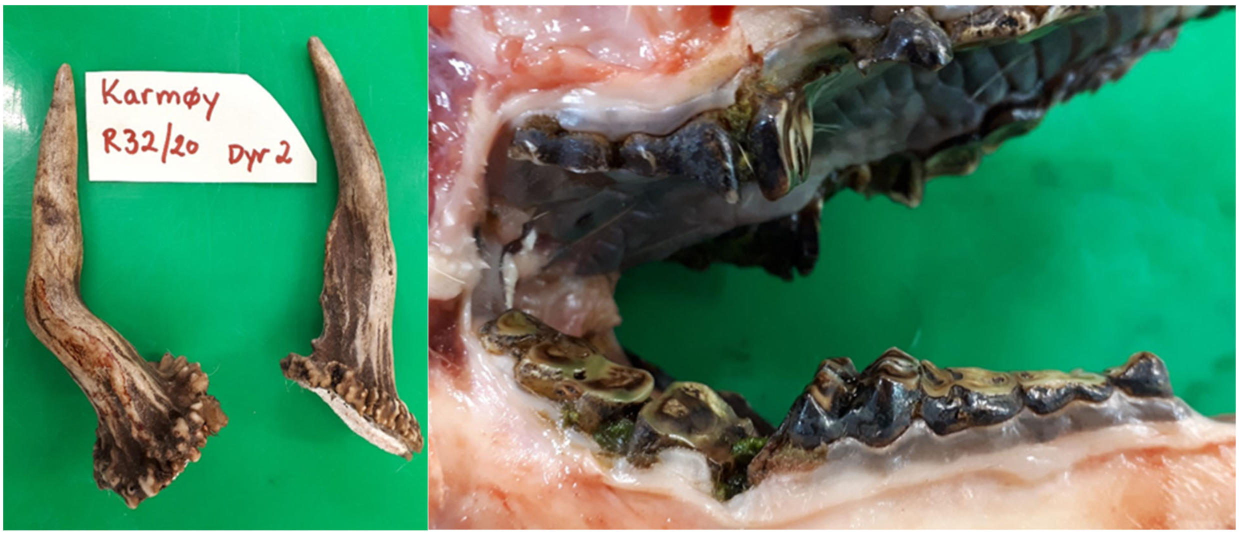

Figure 28*: The antlers (to the left) and the cheek teeth (to the right) of a 3.5-year-old roe deer buck (R32/20) shot within the fenced area of Hydro Karmøy, September 2020. The antlers were deformed and had a F-level of 10,533 mg F/kg. The animal had DF of category 6 (excessive effect) with excessive and abnormal wear, brownish discoloration and enamel lesions. OF were seen on the mandible.*

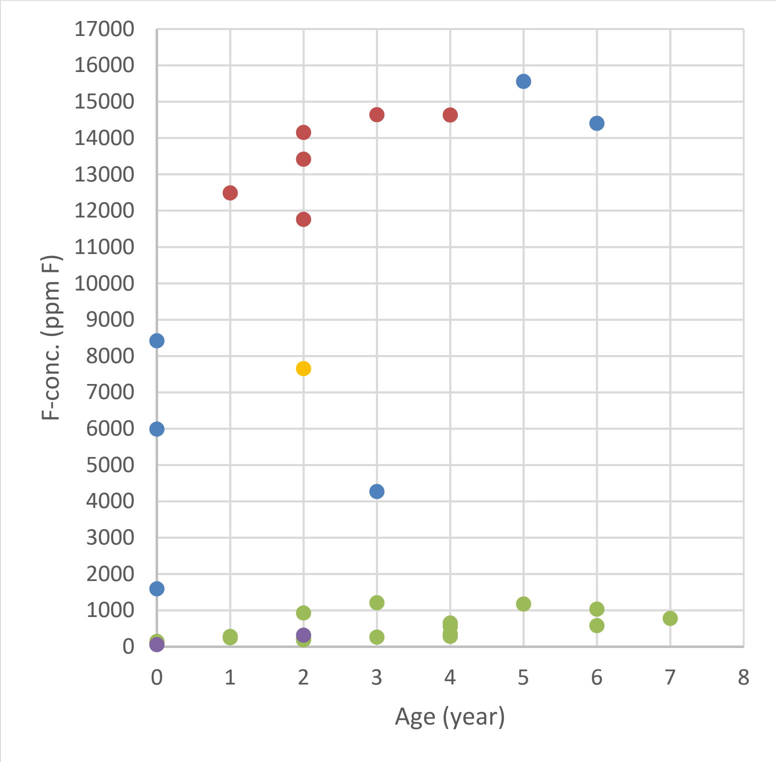

Figure 29: *F-concentration (mg F/kg bone ash) in the lower jawbone (mandible) of roe deer and red deer from Karmøy 2019-20. The red and blue dots are roe deer (n = 12) and the yellow dot are a red deer from within the smelter area; red represents roe deer with dental fluorosis. Green dots are roe deer (n = 18, one animal was not aged due to missing incisors) and purple dots are red deer (n = 2), from the outside the smelter area shown in Figure 27.*

3.4.3.2 Detrimental effects of F in deer

No signs of fluorosis (DF and OF) were found in the roe deer and red deer from outside the fenced area of the smelter, which was in accordance with their rather low F-level not exceeding UNL x 3 (Figure 29).

Of the 12 roe deer from within the fenced area, six had severe or excessive DF (category 5 and 6) (Figure 28 and 30). They were between 1.5 and 4.5-years-old age and their F-level were from 11,756 to 14,639 mg F/kg (Figure 29: red dots). DF was not found in three adult animals; two with excessive F-level (5.5 and 6.5-years-old), and one with high F-level (3.5-years old), respectively (Figure 29: blue dots). Most probably, these three roe deer entered the fenced area after their permanent dentition was erupted. In the three 0.5-year-old fawns, very few permanent teeth were fully erupted and available for examination.

OF was found in several animals within the fenced area. The three roe deer with the highest F-levels had osteofluorotic lesions on the lower jawbone. In addition, four had small signs in accordance with incipient/slight OF, all with F-levels exceeding 11,700 mg F/kg. Typical gross OF lesions were not found with certainty in the remaining five roe deer, including all the fawns.

The more comprehensive material from 11 of the 12 roe deer from within the fenced area, made supplementary examinations possible including evaluating the body condition. The necropsy of the two fawn carcasses showed that both animals were completely emaciated (no body fat) and had high load of internal and external parasites. Evaluating the body condition based on examination of the head (amount of fat) in nine roe deer indicated no emaciated animals, but a few had small fat deposits. Among the latter was the roe deer buck shown on Figure 28, thus it is reason to imply that this was due to the severe DF causing impairment in normal chewing.

The 2.5-year-old red deer shot within the fenced area had a high F-level (Figure 29, yellow dot) and dental lesions, not bilateral, was found. We could not conclude that the lesions were caused by F-exposure, but it is probable taking the young age and the high F-level into account. The lower jawbone and metatarsus/metacarpus were examined for OF, but typical gross lesions could not be found with certainty.

***Figure 30**: The front teeth of a 1.5-year-old roe deer female (R31/20) shot within the fenced area of Hydro Karmøy September 2020, showing excessive DF. There are multiple large enamel defects and cream coloured enamel (not glossy) with brown discoloration. It had a F-level of 12,483 mg F/kg and small signs of OF on the mandible.*

3.4.3.3 Bone F-concentration and detrimental effects of F in sheep and a cow

Three sheep (utegangarsau/"villsau") slaughtered in November 2020 were included in the study and the head submitted for examination. Their grazing area was just south of the Hydro Karmøy area (Figure 31). The oldest sheep (S1/20) was 7.5-years-old and according to the farmer; it had grazed in this area the last six years (2015-20). It had a bone F-level 796 mg F/kg and no signs of DF or OF (Figure 34). Due to the deviation of the F-level in this sheep compared to the two other, a new sample of the mandible was analysed and showed similar F-concentration (753 mg F/kg). The second sheep (S2/20) was 1.5-year-old and had grazed in the same area as S1/20 the whole life. It had an excessive F-load of 12,577 mg F/kg, severe DF (category 5-6) (Figures 32-33), and findings indicative of incipient OF in the mandible. The third sheep (S3/20) was a 0.5-year-old lamb that had a heavy F-load of 4,932 mg F/kg, and no signs of DF (no permanent teeth yet) or OF. The two young sheep fall into the group of severe detrimental fluoride effects according to Table 4.

The lower jawbone of a cow slaughtered in June 2021 due to high age (10-12 years) was also included in the study. The cow had been grazing during summer (May-October) in the area directly northwest of the smelter (Figure 31) for at least the last two summers, most probably longer. In addition, the farmer uses grass from this area as forage during winter. The examination revealed slight DF (DF 3) of some of the cheek teeth. The F-level was 8,611 mg

F/kg and there were signs of OF (exostosis like) on the lower jawbone. According to table 4, the cow was categorised as having severe F effects.

Figure 31*: The sheep grazing area south of the premises of Hydro Karmøy is marked with turquoise colour. The cow grazed in the area northwest of the smelter. In the courtesy of Tor-Erik Richardsen, Hydro Karmøy.*

Figure 32*: Front teeth of a 1.5-year-old sheep (S2/20) from Karmøy showing severe dental fluorosis of the two permanent teeth pairs (I1 and I2). The enamel has severe pitting with brown discoloration.*

Figure 33*: The cheek teeth of a 1.5-year-old sheep (S2/20) from Karmøy showing severe dental fluorosis with abnormal wear particularly of the 4th cheek tooth (M1), brown discoloration and enamel pits.*

***Figure 34**: The cheek teeth of a 7.5-year-old sheep (S1/20) from Karmøy showing normal wear.*

3.4.3.4 Bone F-concentration and detrimental effects of F in wild birds

Seventeen gulls of various species and ages (8 juvenile and 9 adults) and one oystercatcher were collected in 2019 (n = 2) and 2020 within the areas of the smelter. All had traumatic lesions of various severity. The bone F-level of the 17 gulls is shown in Table 15. The oystercatcher had 3,490 mg F/kg bone ash in the upper arm bone (humerus).

***Table 15**: F-concentration of femur (mg F/kg bone ash) in gulls from the area of Hydro Karmøy 2019-20 given for each species and age group (juvenile/adult).*

Species	total	adult	juvenile	mean F	median F	range F
Common gull (fiskemåke)	3	3	-			2,152 – 4,710
Lesser black-backed gull (sildemåke)	5	5				1,972 – 4,378
Herring gull (gråmåke)	1	1				3,391
All adults	9	9		3,179	3,007	1,972 – 4,710
Unknown (all juvenile)	8		8	1,310	1,154	966 – 2,140

3.4.4 Local differences in F-exposure of deer related to vegetation data

The new studies revealed severe F-exposure of deer and sheep grazing within the fenced areas or in the very close proximity south of the smelter. Further away (> 1km) the examined deer showed low F-exposure, among which the highest F-accumulation was found in the animals closest to the plant (Vorrå and Håvik).

Hydro Karmøy has monitored F-concentration in grass and spruce at many locations around the smelter dating back to the late 1960s. None is in the very close vicinity of the plant where the heavily F-exposed animals have been living. Thus, in autumn 2020, a new sample site for grass within the smelter area close to the sheep grazing land (Deponi 4) was included showing a level of 60 mg F/kg, Thus, the tolerance threshold was clearly exceeded in this sample (Table 1). The F-concentrations in vegetation samples vary considerably between years and are difficult to interpret. They have with few exceptions been highest at the sampling sites closest to the plant (Håvik, Vorrå), and the last five years rather few has exceeded the 30 mg F/kg threshold. An exception is the F-concentration in spruce showing high levels at Håvik.

3.4.5 New results compared with historical data

3.4.5.1 Deer and livestock

We have no historic data for the main indicators in the current study, roe deer, or for livestock. In the Effect Study (1992-93), red deer from Karmøy and neighbour municipality Bokn were studied; however, the material was rather scarce. The situation was quite different from today because deer was not living close to the smelter. The red deer were mainly located south on the island. Due to the major wind direction during summer, red deer from Vestre Bokn Island in southeasterly direction of the smelter was included. Indisputable F-exposure (> UNL x 3) was documented in 12 % of the animals from Vestre Bokn, mainly old animals, and none from Karmøy. No animal had DF or OF.

The current study has revealed severe F-exposure of ruminants living very close to the smelter, an area that was not examined in the Effect Study. The proportion of gaseous F of the total F-emissions is currently higher than in the 90s. Gaseous F is more bioavailable than particulate F.

3.4.5.2 Wild birds

Generally, birds seem to have high tolerance threshold for fluoride and many species have high background bone F-levels. In the Effect Study, F-levels were analysed in femur of adult herring gull collected in 1993 from a reference site not exposed to industrial F-emissions and from the area of Hydro Karmøy (Vikøren and Stuve, 1994). In 1993, a significant higher F-level was found in female herring gull compared to the reference group, but this difference was not found for males.

The maximum F-level in gulls from Karmøy was 6,185 mg F/kg in 1993 and 4,710 mg F/kg in 2019-20, respectively. In the current study, only four gulls from Karmøy had F-levels higher than the maximum F-level found in herring gulls from the reference area in 1993.

3.4.6 Current state Hydro Karmøy

Herbivorous animals like roe deer and livestock grazing within the fenced area or in the very close proximity of the Hydro Karmøy plant, show excessive F-exposure with extreme high bone F-levels and severe fluorosis. From an animal welfare point of view, no grazing animals should use these areas. Gulls nesting in the area do not show major F-accumulation.

Further away from the smelter (>1 km), the F-exposure is considerably lower and none of the examined deer showed indisputable F-exposure or fluorosis.

3.5 Alcoa Lista

3.5.1 Site description

Alcoa Lista is located in the Farsund municipality in the Agder County. The area is exposed to wind and weather with prevailing winds from northwest (NW) during much of the year. This will generally carry air borne pollutants seawards.

The climate is mild and humid, with a mean annual temperature of +7.6°C and annual precipitation of approximately 1,050 mm. The areas towards east are barren, low hills with sparse vegetation. Towards southeast (SE), there is a small forest between the plant and the Lomsesanden beach. The plant is close to the Husebysanden beach towards southwest (SW). Towards west (W) and NW there are mixed agricultural land with patches of forest and some built-up areas.

The aluminium plant was established in 1971 with Søderberg technology. The plant has three potrooms with a total capacity of about 94,000 tonnes p.a. This is an increase of about 17 % since 1992. Alcoa has upgraded the Søderberg technology to reduce emissions to the ventilation air.

F-emissions from Alcoa Lista in 2020 was 26.7 tonnes per year which is 40-50% lower than in the early 1990s (Figure 35). The prevailing winds from NW generally carry air borne pollutants seawards in a S/SE direction.

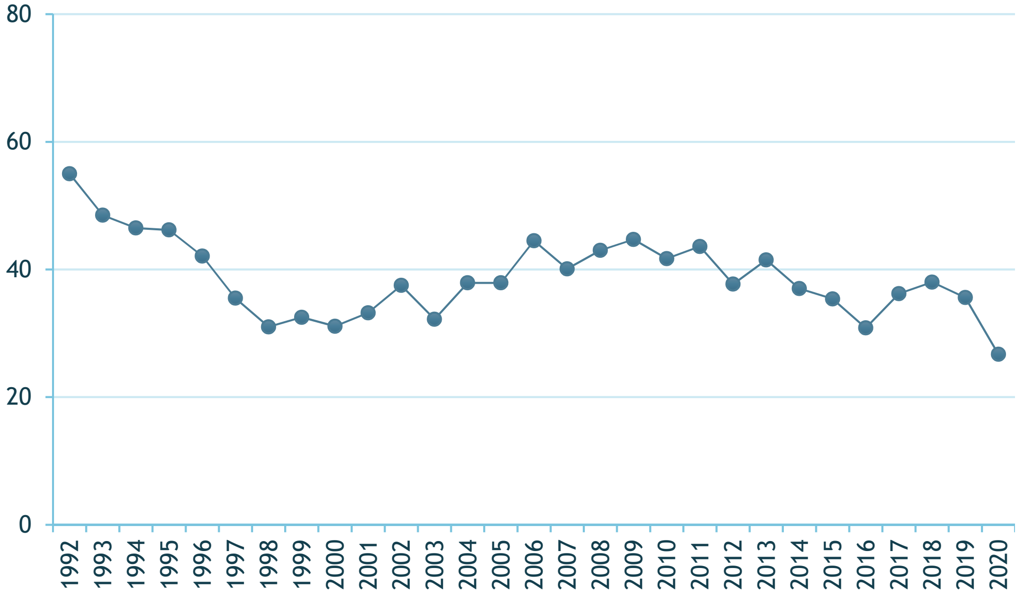

Figure 35: *Fluoride emissions (tonnes per year) from Alcoa Lista from 1992 to 2020. Source: AMS.*

3.5.2 The deer populations in Farsund

Roe deer is the most abundant cervid in Farsund municipality and it is harvested during the hunting season from August to December. There are several hunting areas for roe deer within the area of interest (Figure 37).
Red deer has populated Farsund during the last 30 years and in 2019 roundabout 80 were shot in the whole municipality, but there is no hunting in the area of interest.

3.5.3 Studies of fluoride exposure in wild animals, 2019-20

Based on the status for F-exposure of animals in Farsund presented in WP1, the current study was limited to an area comprising a radius of approximately 3.5 km from the Alcoa Lista plant (Figures 36-37). The main indicator was hunted and fallen stock (traffic killed and other casualties) of roe deer of all ages.

Totally, 19 roe deer of various ages were examined: nine sampled in 2019 and ten in 2020 (Figure 38). In addition, one beaver (*Castor fiber*) was included.

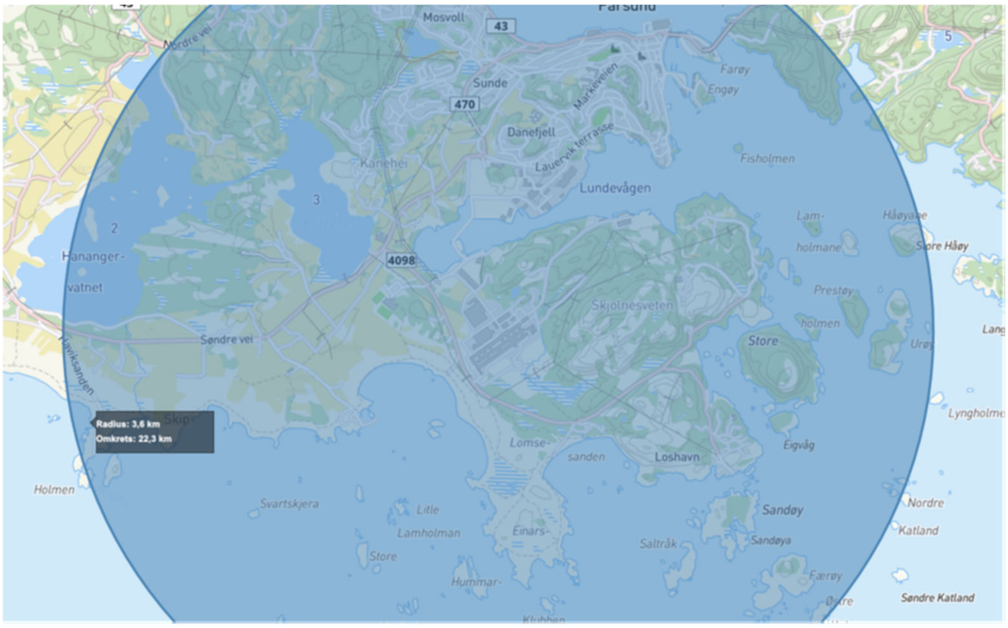

Figure 36: *The study area of F-exposure of roe deer from Farsund 2019-20 comprising a diameter of approximately 3.5 km from the Alcoa Lista smelter. © 2021 Norkart AS/Geovekst og kommunene/NASA, Meti © Mapbox © OpenStreetMap*

Figure 37: *The study area of F-exposure of roe deer in Farsund 2019-20 showing the hunting areas (vald) of interest. In the courtesy of Jan Fredrik Sundt, the agricultural department, Farsund municipality.*

3.5.3.1 Bone F-concentration and detrimental effects of F in roe deer

Figure 38 and Table 16 show the F-level, age and location of the examined roe deer. Only one roe deer (5.3 %) had F-level above UNL x 3, representing also the maximum level (4,450 mg F/kg) in Farsund. This individual was the only roe deer with dental fluorosis (category 4, marked effect). It was a 5.5-year-old animal from Krågenes, approximately 1.5 km in NW direction from the plant. Osteofluorosis was not found.

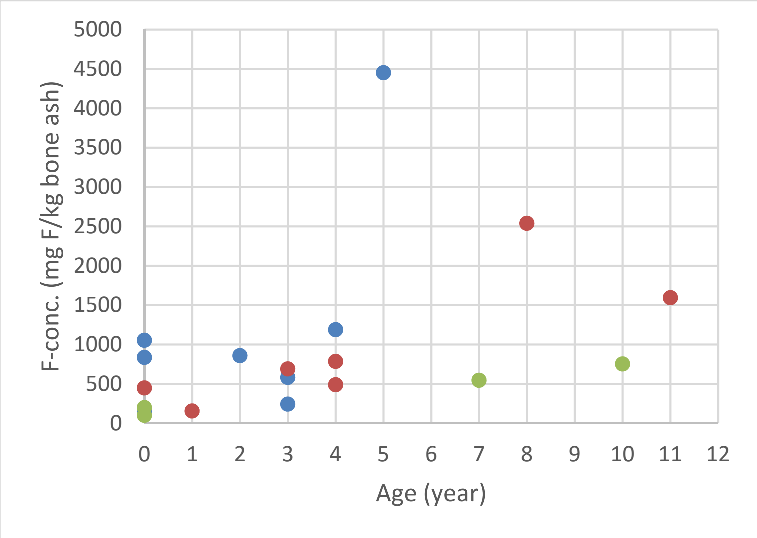

Figure 38: *F-concentration (mg F/kg bone ash) in the lower jawbone (mandible) of roe deer from Farsund 2019-20, sampled within 3.5 km from the Alcoa Lista smelter. The colour of the dots show the locality of the animals: blue is in the northwest (NW) direction (Lunde, Krågenes, Fjellestad), red is in the southeast/east (SE/E) direction (Lomsesanden, Loshavn, Eikenes, Skjoldnes) and green is in the west (W) (Radiosletta, Åsen) direction from the plant.*

Table 16: *The F-concentration (mg F/kg bone ash) in the lower jawbone (mandible) of roe deer from Farsund given for the monitoring periods 2019-20 (3.5 km radius from Alcoa Lista smelter) and 1991-93 (the whole municipality, the Effect Study, n=165). The F-level is given for each age group with mean, minimum and maximum. The individual data is registered in the ESPIAL Fauna database (updated version).*

Sampling year		0.5 year	1.5 year	2.5 year	3.5-5.5 year	6.5 year and older
2019-20*	Number	6	1	1	7	4
	Mean	463	154	857	1,203	1,357
	Min	99	-	-	242	545
	Max	1,052	-	-	4,450	2,538
1991-93**	Number	51	24	20	62	8
	Mean	304	563	935	833	1,004
	Min	38	125	141	135	251
	Max	2,759	2,034	7,430	6,830	4,644

** In 2019-20, the sampling area was restricted to a 3.5 km radius from the smelter.*
*** In 1991 and 93, the whole municipality was sampled, whereas in 1992 it was restricted to a 5 km radius from the smelter.*

3.5.3.2 Bone F-concentration and detrimental effects of F in a beaver
The beaver was euthanized in April 2020 at Lomsesanden (roughly 300 m SE from the smelter premises) due to traumatic lesions. It had 13,028 mg F/kg in the mandible, which is extremely high, compared to the maximum level found in roe deer. The age of the animal was unknown. Background F-level for beaver is not published and therefore we analysed the mandible of two beavers from our historic collection (1991): The first was a young male, presumed one year old from Eidsvoll, Viken County, and the other was an adult female (Figure 39) of unknown age from Kviteseid, Vestfold and Telemark County. The F-level in the mandible were 1,392 and 2,813 mg F/kg bone ash, respectively, thus higher than for wild cervids and hare.

In contrast to cervids, the teeth of the beaver grow the whole life due to constant wear, the front teeth at a higher speed than the cheek teeth. Thus, DF can develop during the whole life span. Another difference is that the front teeth of the beaver are only covered with enamel on the frontal part. The front teeth of the examined beaver had large abnormalities with pitted and eroded enamel with dark discoloration, particularly on the left tooth (Figure 39). In addition, the cheek teeth had dark brown-black discoloration. These lesions strongly indicate F-exposure. The expected bilateral findings were not so prominent, except at the top. In addition, we found lesions (exostosis) in the lower jawbone that was suggestive of OF.

Figure 39*: To the left: The lower front teeth of a beaver from Lomsesanden, Farsund municipality, showing large abnormalities in the enamel with dark brown-black discoloration, pitting and erosions, particularly on the left tooth (to the right in the picture). To the right: The normal lower and upper front teeth of an adult beaver showing the characteristic orange-brown colour and smooth enamel.*

The beaver has a long lifespan and can become up to 20-years-old. It feeds on various trees and water plants. The beaver prefers the foliage and the living bark of deciduous trees, particularly willow (*Salix* spp. = vier og selje) and aspen, but it eats birch, rowan and hazel as well. The summer feed consists essentially of herbs, particularly water plants.

3.5.4 Local differences in F-exposure of wild animals related to vegetation data
Of the 19 roe deer examined within 3.5 km from the plant, only one showed indisputable F-exposure. It was from Krågenes, approximately 1.5 km in NW direction of the plant.

There are available data from Alcoa Lista on F-level in vegetation (grass and conifer) at several locations up to 10 km from the smelter, from 1994 to 2020. None of the grass sample locations are in the southerly direction and the closest being Lunde, 1.5 km to the NW and Eikvåg, 1.8 km to the E. The last decade, only two grass samples collected in autumn (Vatne 5.5 km to the NV) had F-levels above 30 mg F/kg (dry weight) which is the tolerance level for roe deer. On the other hand, some of the conifer samples from Lomsesanden (S direction) have shown F-levels slightly above this threshold, also for samples collected in 2020.

Grass and conifer are not among the preferable diet for roe deer. However, based on these vegetation results and the fact that the roe deer in question had DF, it is reason to believe that the animal lived closer to the plant during the dental development the first 1.5 year of its life. In the Effect study, the F-concentration in foliage of rowan showed highest levels to the SE and E. In the area of Lomsesanden it was > 100 mg F/kg (Figure 23 in Horntvedt and Øyen 1994).

Regarding the F-exposure of the beaver, it is interesting to look at the part of the diet that is specific for this species; bark, branches and twigs. Analyses performed in the Effect Study showed that the F-content in bark consistently was low, and considerably lower than in the foliage of the same species at the same location and time. The F-content in branches and twigs was lower than in bark. However, the exception was birch, showing very high F-levels, probably because the bark of the birch becomes very old (see more details in Table 8 in Horntvedt and Øyen 1994).

The F-concentration of the two beavers representing background level was rather high compared with ruminants like deer. Thus, this probably reflect the diet of the beaver. The high F-load and the DF in the beaver from Lomsesanden indicate very high F-exposure, probably caused by ingestions of plants. Since the age was not known, it is difficult to interpret the findings in detail.

3.5.5 New results compared with historical data

The current study of roe deer show a lower F-exposure than in the Effect study (1991-93). One roe deer (5.3 %) had F-level above UNL x 3, a considerable reduction compared with the early 1990s (11 %, 18 roe deer: 16 within 2 km from the smelter). Only one roe deer had DF (category 4) and the corresponding number in the Effect study was four (category 3-5) shot at Fjellestad (N), Eikvåg (E), Skjoldnes (NE), and Alcoa/Huseby, respectively.

The Effect study also included hare and individuals shot within the premises of the smelter showed high F-load (mean and maximum of five individuals was 3,080 and 8,116 mg F/kg bone ash, respectively) and DF was detected in two animals. The hare cannot directly be compared with the beaver due to lower background F-level and different diet and lifespan. However, the beaver from 2020 also lived close to the plant and showed high F-level and DF.

3.5.6 Current state Alcoa Lista

The current study indicate that the F-exposure of roe deer in this area is lower than in the early 1990s. This is in accordance with lower F-emissions. One 5.5-year-old roe deer had moderate DF (due to F-emissions in 2014-15 during tooth development). Surprisingly, a heavily F-exposed beaver was detected close to the smelter. Therefore, we conclude that the F-emissions from Alcoa Lista might still cause high F-exposure of wild animals that lives in the very close vicinity of the smelter.

4 Conclusions

- The F-exposure (documented by F-accumulation and fluorosis) of wild animals differ between the five aluminium smelters and seems to be influenced by local topography, meteorological conditions, and level and composition of F-emissions.
- Detrimental F-effects (fluorosis) in wild animals were present at all smelter sites, however at a variable degree and severity.
- The highest F-exposure was found in deer from Årdal within a distance of approximately 12-13 km from the smelter, and within the fenced area around Hydro Karmøy. In Karmøy, the F-exposure of roe deer further away than approximately 1 km from the smelter was found to be low.
- The F-exposure was moderate in deer at Sunndal (lower half of Sunndal valley) and Husnes (within 3 km) and generally low at Alcoa Lista, with a few exceptions.
- Generally, some F-exposed deer might also be found at further distance from the smelters due to migration from locations closer to the smelter where the main F-intake have taken place.

- We have shown that 1.5-year-old (yearlings) red deer is a very useful animal indicator of recent F-exposure:
  - Young individuals are particularly prone to F-toxicosis due to skeletal growth and tooth development. The consequences of F-exposure in early life can be aggravated as time goes on, since dental fluorosis in the categories 3 and 4 imply that the teeth have reduced hardness and will wear faster. Thus, if the affected animals had lived longer, their DF could have ended in category 5 or 6.
  - The results for 1.5-year-old red deer in the present study confirm red deer's high F-sensitivity. In Sunndal and Husnes several individuals showed lesions we interpreted as very mild/small signs of dental fluorosis (category 3). We have found it necessary to adjust the dose-effect relationship due to findings of mild dental fluorosis in four animals having F-concentrations below 1,000 mg/kg (Table 17).

***Table 17:** Fluoride dose – effect relationship for 1.5-year-old red deer – adjusted 2021.*

Cumulative F dose (mg F/kg bone ash in mandible)	Effect (visible dental fluorosis)
Less than 900	None
900 – 2,000	Risk of developing dental fluorosis
Above 2,000	Causes dental fluorosis

  - The F-exposure of 1.5-year-old red deer was higher in Årdal compared with Sunndal and Husnes, and the most severe dental lesions were found in Årdal. Higher F-emissions in 2018-20 (except for 2019 when Sunndal had the highest) and more unfavourable topography is probably the main explanation. In addition, the habitat of the examined animals might have been closer to the plant in Årdal.
  - Red deer yearlings from Sunndal and Husnes showed similar F-exposure, however the area affected was much larger in Sunndal (up to approximately 15

km from the smelter in the Sunndal valley) than at Husnes (within 3 km). The F-emissions in 2018-20 were considerably higher in Sunndal than Husnes, and the topography is less favourable for dispersion of fumes.
    - In well-established red deer populations in western Norway, the hunting bag of yearlings is large which one can take advantage of for monitoring purposes. Roe deer is more sedentary than red deer and therefore considered a better indicator, but the low hunting bag limit the supply of material for monitoring.

- We consider the situation at Hydro Karmøy where roe deer inhabited the fenced area around the smelter, as extraordinary; these animals were indeed sedentary and thus fulfilled the requirements for an optimal indicator. Many of the animals had lived there for several years and we consider the F-exposure as extreme, mimicking a F-feeding experiment. The F-concentrations found were extremely high (the maximum exceeded 15,000 mg/kg bone ash) showing F-accumulation at levels never documented before in roe deer in Norway (Figure 29). Many of these animals had severe dental fluorosis and some had gross signs of osteofluorosis. Similar findings were also found in some sheep grazing just south of fence around Hydro Karmøy.
    - The topography (flat landscape) and the windy conditions at Karmøy are considered favourable causing fast dispersal and dilution of F-emissions. Despite this, the conditions for the roe deer within the fenced area around the smelter are a reminder that the close proximity of the smelter is heavily F-polluted. Therefore, of animal welfare reasons, this area is not suited for sensitive ruminants like deer, sheep and cattle with low F-tolerance threshold.
    - It is reason to believe that the conditions detected at Karmøy also applies to the other smelters. Individuals found close to the smelter at other sites also showed evidence of high F-exposure.

- Compared with the early 1990s, the current F-emission level is at approximately same level in Husnes, Karmøy and Sunndal, a bit lower in Årdal, and 50 % in Lista. In Årdal, indisputable F-exposure showed a decreasing trend from the 1990s; however, this trend was not seen in the percentage of deer with dental fluorosis. In addition, in the Sunndalen valley, the percentage of deer with DF had increased.
    - More animals seem to be at risk of F-exposure at a level causing dental fluorosis compared to the early 1990s due to increased red deer population and density that also includes the most F-polluted areas close to the plants. Changes in migration pattern may also be a factor of concern, for example is red deer in the Sunndalen valley more sedentary than earlier.
    - At several sites, prebake technology has replaced Søderberg technology, thus the proportion of the more bioavailable gaseous F of the total F emissions has increased. The significance of this is probably largest for Sunndal and Karmøy that had a relatively high production with Søderberg technology before it was replaced with prebake.

5 Recommendations

Recommendations for management and further monitoring of fluoride exposure of animals:

5.1 General recommendations for all smelter sites

- The F-emissions should be lowered to reduce the F-exposure of animals, particularly at sites like Årdal and Sunndal where the topography is especially unfavourable for dispersal of fumes.
    - Avoid episodic emissions, since short lasting (a few days or weeks) high F-emissions might cause dental fluorosis in young ruminants in the period of tooth formation/mineralization.
- Vegetation in areas close to aluminium smelters that is used (or planned used) as pasture or harvested as forage for F-sensitive domestic ruminants like sheep, goats and cattle, should be systematically monitored for F-content during the growing season. Generally, the tolerance levels in the feed (Table 1) should not be exceeded.
- Animals, particularly ruminants but also hare and beavers, found dead within the very close proximity of aluminium smelters, should be examined for F-exposure (F-level in bone and fluorosis).
- Wild deer entering the fenced area around a smelter should be relocated or euthanized.

5.2 Hydro Årdal

- We recommend monitoring of F-exposure in 1.5-year-old red deer on a regular basis of every 2nd year. The survey should include harvested animals in hunting areas at a distance of approximately 15 km from the smelter. Thus, the most western hunting areas on both sides of the Årdal fjord do not have to be included.
- If F-emissions increase significantly, we recommend expanding the monitoring area to include all hunting areas in the municipality and to survey on a yearly basis.

5.3 Hydro Sunndal

- We recommend monitoring of F-exposure in roe deer and 1.5-year-old red deer in the lower half of Sunndal valley (zone 3-5) on a regular basis of every 2nd or 3rd year. Focus should be on the areas closest to the smelter.
- If F-emissions increase significantly, we recommend to expand the monitoring area to the whole Sunndal valley and to survey every 2nd year.

5.4 Hydro Husnes

- We recommend monitoring of F-exposure in 1.5-year-old red deer in the hunting areas closest to the smelter (Husnes and Kaldestadåsen) on a regular basis of every 3rd year. In these areas, we found many individuals that were exposed to F at a “borderline” level (900-2,000 mg F/kg bone ash); a dose that gives risk for developing dental

fluorosis. It is important to follow this exposure in the years to come, especially if F-emissions are increasing.
- If F-emissions increase significantly, we recommend to expand the monitoring area to 5 km from the smelter and to survey every 2nd year.

5.5 Hydro Karmøy

- F-sensitive ruminants like deer, sheep and cattle should not graze within the fenced area of the smelter. Wild deer entering this area must either be 1) immobilised and moved to new living areas with background F-levels or 2) euthanized (shot).
- We stress that the general advice regarding F-monitoring of vegetation used as pasture/feed for livestock like sheep and cattle must be followed in areas close to the smelter.
    - If F-polluted feed (> 30 mg F/kg dry weight) constitute a significant amount of the diet for livestock, a possible preventive measure can be supplement of special feed with a high content of aluminium and calcium salts. These will bind to F in the alimentary canal forming heavy soluble compounds that are excreted in the faeces (Aas Hansen 1994).
- Further examination of F-exposure in livestock that grazes close to the smelter is recommended.
- If F-emissions increase significantly, we recommend monitoring of roe deer and livestock from an area of approximately 2 km from the smelter on a regular basis of every 2nd to 3rd year.

5.6 Alcoa Lista

- If F-emissions increase significantly, we recommend monitoring of roe deer (and red deer, hare and beaver) from an area of approximately 3 km from the smelter.

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8 Appendix

8.1 Historical data

Data from the period 1994-2018 received from the aluminium plants were registered in the ESPIAL Fauna database (information given by contacts at the plants, spring 2019). In addition, most of the data from wild animals collected around Norwegian aluminium plants in the period 1990-93 (Effect study) are included, as well as some historical data which have been received from various sources, among them the Monitoring Board ("Kontrollutvalget for aluminiumsindustrien"). The database is updated in 2021 with the data generated in ESPIAL Fauna WP 2 described previously in this report.

The purpose of the database is to be as complete as possible regarding information/data on fluoride exposure in animals around the aluminium plants being AMS members, both regarding historical as well as future data. The level of exposure (F-concentration in bone) and detrimental fluoride effects (dentalfluorosis/osteofluorosis) should be registered on an individual animal basis.

If there are historical data in various archives not included in the database yet, we advise to include these as well. The database will be an easy way to find standardised information in the future. The Norwegian Veterinary Institute can contribute to registration of data if requested.

The aim of the ESPIAL Fauna database is to comprise all monitoring data regarding fluoride exposure in animals - both wild and domestic - around the AMS member aluminium plants in Norway, Iceland and Sweden.

8.2 The ESPIAL Fauna database

The database is established as an EXCEL file. The results from ESPIAL Fauna – WP 2 have been included in a new updated version of the file in 2021.

Description of the ESPIAL Fauna database

Sheets: The EXCEL file includes a sheet giving instructions for how to register the data in a standardized way. There is one sheet for each aluminium plant, and a sheet for reference areas for Norway and Iceland, respectively.

Examinations of animals from municipalities bordering to aluminium plants is included in the sheet of the smelter, for example: Lærdal and Luster are included in the Årdal sheet.

Rows: One row per individual animal.

Individual data that should be included is below marked as:

- Mandatory – the data is of no value if this information is missing
- Very important
- Important

Regarding fluoride exposure, the fluoride concentration and the effects (dental fluorosis/osteofluorosis) are the parameters to include. If all these three parameters are missing from an animal, the registration is of no value.

Other missing data can be registered as "not given", "not analysed", "not examined" or "not applicable".

The database includes the following columns:

ID 1 and ID 2: individual identification: Two columns available for free-text registration.

Year dead: the year the animal died (shot/slaughtered) – **very important**

Species: name of the species in English (Norwegian) - **mandatory**

Location 1: name of municipality the animal was located – **mandatory**

Location 2: location more detailed – **very important.** For example:

- hunting area/«vald» for hunted cervids
- name of the farm/farmer

Location 3: other location data if applicable. For example:

- ID-number of the hunting area/"vald"
- the farmers production number (Norway)

Gender: **- important**

- m = male
- f = female

Age (year): **- very important**

- age in whole years if the age of the animal have been determined:
    - wild animals: based on teeth eruption pattern or counting annuli in incisor tooth cementum
    - farm animals: based on year of birth
- 0 = the first year of living (up to 1 year old)

Bone sampled: **- important**

- Name the skeletal bone used for fluoride analysis
- Preferably, the specific part of the bone sampled should also be registered due to some variation in fluoride concentration between different parts of the bone.
- "mandible standard" = referring to a standardised sampling of the lower jawbone (mandible) used by the Norwegian Veterinary Institute since the Effect Study. The method is described in Vikøren et al. 1996 and a short summary is given in Material and Methods in this report.

Fluoride concentration: **- mandatory**

There are two possible columns for registration of fluoride concentration:

- F conc ash (µg F/g): If the result of the fluoride analysis is given in bone ash – the concentration should be registered in this column as µg F/g.
- F conc dry matter (µg F/g): If the result of the fluoride analysis is given in dry matter – the concentration should be registered in this column as µg F/g.

DF-category: dental fluorosis (DF) category – **very important (mandatory if F conc. is missing)**

- The method used by the Norwegian Veterinary Institute for scoring teeth is described in Vikøren et al 1994, and Vikøren and Stuve 1996. Categories from 1 to 6 are used, and additionally 8 refers to normal advanced old age related wear. 1 = normal teeth, 2 = questionable effect, 3 = slight to moderate effect, 4 = marked effect, 5 = severe effect, 6 = excessive effect.
- NA has used the National Research Council (NRC 1974) method for samples from Fjarðaál.

DF- method: **- important**

State the method used for evaluation of DF. The alternatives are:

- the Effect Study method (Norwegian Veterinary Institute)(Vikøren et al 1994, Vikøren and Stuve 1996)
- National Research Council (NRC). 1974.
- other methods if applicable. Give a reference to the method used.

OF: osteofluorose (OF): **- very important**

- yes = gross lesions in accordance with osteofluorosis were found.
- no = examined and no gross lesions in accordance with OF were found.

F analysis lab: name of the laboratory that has performed the F analysis **- important**

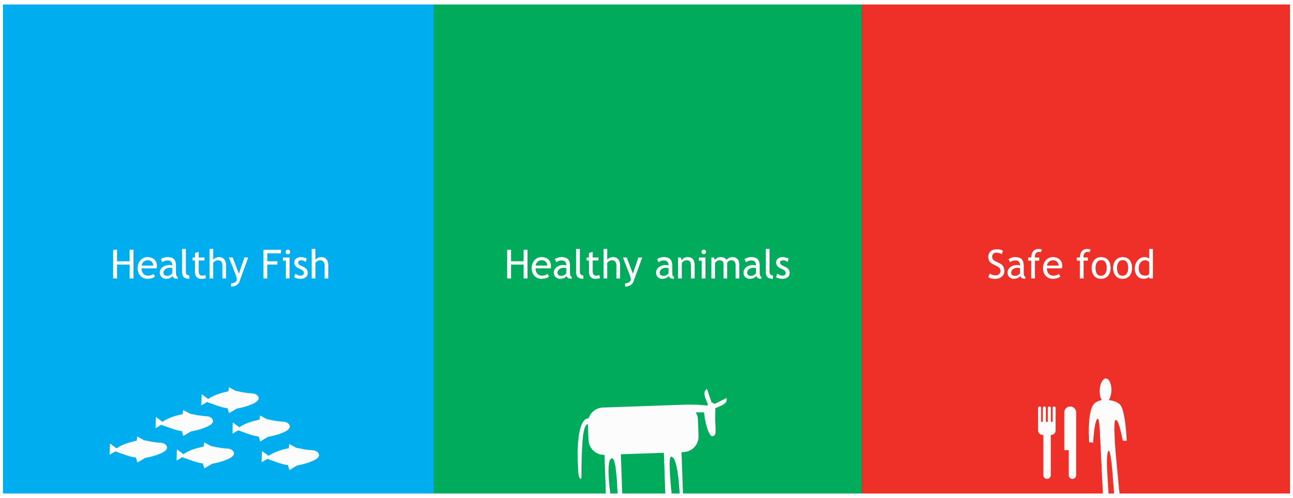
Comments: free text available for further information if applicable.

Recommendation for standardisation of future studies of dead animals for fluoride exposure:

- Bone for fluoride analysis: lower jawbone (mandible), use the “mandible standard” method (Vikøren et al. 1996).
- Gross examinations of the teeth for dental fluorosis and the mandible for osteofluorosis.

9 Attachment

The ESPIAL-Fauna database – updated version 2021 (Excel file)



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