Náttúrustofa Austurlands

Fluoride concentration in reindeer jawbones Pre- and Post-Alcoa Fjarðaál aluminium smelter in Reyðarfjörður - Iceland

-Effects of fluoride emissions on wild reindeer

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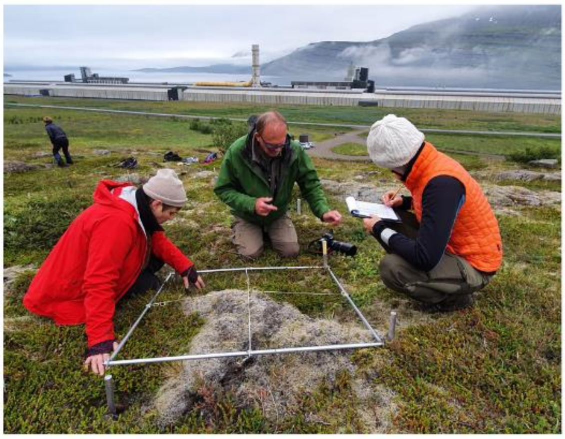

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Photographs on front page: Hilmar Sigurbjörnsson, Alcoa Fjarðaál and Kristín Ágústsdóttir

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Abstract: The Alcoa Fjarðaál aluminium smelter in Reyðarfjörður started production in 2007. After the startup, elevated levels of fluoride (F) concentration have been discovered in sheep jawbones and vegetation in the vicinity of the plant. Random analysis of jawbones of three adult reindeer bulls killed close to smelter in 2014 revealed elevated F-levels. As reindeer have been seen for extended periods around the smelter during the last decade it was considered important to get a better understanding of the possible F-exposure on these animals.

This study established background values of F-concentration in 67 lower jawbones from reindeer killed in the area prior to the smelter startup (2001 to 2007 – PRE-smelter animals). To examine possible F-effects due to the smelter, 61 jawbones from reindeer grazing in the area during full operation of the smelter (2014 to 2020 – POST-smelter animals) were examined. The study is a collaboration with the Norwegian Veterinary Institute (NVI) and is a part of the ESPIAL Project (Ensuring Sustainable Production of Primary Aluminium). Most samples were examined at NVI for detrimental effects and the F-concentration in bone was analysed Hydro Aluminium Metal Årdal in Norway. Lower jawbones are good specimen to assess effects of elevated F as they are among the bones with the highest levels of F-concentration. Additionally, age can be estimated, and detrimental effects of teeth (dental fluorosis) and bone (osteofluorosis) can be examined.

F-concentration in jawbones from POST-smelter animals was significantly higher than in PRE-smelter animals. F-concentrations in reindeer killed before the smelter increased with animal age but F-concentration decreased with increasing distance from the smelter site in animals killed after the smelter. The highest F-levels were found in animals shot close to the smelter after startup. None of the reindeer examined showed gross signs of osteofluorosis in the jawbone or dental fluorosis.

We conclude that a moderate increased fluoride load is documented in reindeer living in the vicinity of the Alcoa Fjarðaál smelter 2018-20, but no indisputable gross signs of fluorosis were found. Higher F-concentration in POST-smelter jawbones is concluded to be due to higher F-concentration in the vegetation that reindeer graze on in Reyðarfjörður area after smelter start up. Parts of the year reindeer graze close to the smelter where average values of F-concentration in vegetation is highest. Despite increased bone F-concentration, the F-accumulation in the bones of the reindeer did not reach limits known to cause detrimental effects in cervids. The fact that the animals migrate to areas further away from the smelter during large parts of the year and graze on none F-exposed vegetation could explain that. Thus, the nomadic way of life protects the reindeer from more severe F-exposure.

Despite increased F-concentration in the jawbones of reindeer, the values were very low compared to cervids grazing near some of the smelters in Norway. If F-concentration in vegetation in Reyðarfjörður increases significantly from what has been measured since the smelter startup and if F-concentration in bone ash from sheep jawbones reaches values above limits for possible detrimental effects, it will be important to monitor further the health of the reindeer grazing in the vicinity of the smelter.

Keywords: Alcoa-Fjarðaál, fluoride concentration, forage, jawbones, mandibles, reindeer, dental fluorosis, osteofluorosis, detrimental effects.	**ISSN nr:** 2547-7447 (electronic version)
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Íslensk samantekt (Icelandic summary)

Álver Alcoa Fjarðaáls í Reyðarfirði hóf starfsemi í apríl 2007. Styrkur flúors (F) í gróðri í Reyðarfirði og í kjálkabeinum kinda sem hafa gengið í Reyðarfirði hefur hækkað samanborið við grunngildi frá því áður en álverið tók til starfa. Flest ár, a.m.k. frá 2000, hafa hreindýr dvalið hluta úr ári á svæði sem nú er í næsta nágrenni álversins, þar sem styrkur F í gróðri hefur mælst hvað hæstur. Handahófskennd mæling á kjálkabeinum þriggja hreindýra sem drápust í árekstri við álverið árið 2014 leiddi í ljós að styrkur F í kjálkabeinum hreindýra hafði hækkað samanborið við grunngildi sem mæld voru í fimm hreindýrum sem drápust áður en álverið tók til starfa (2002 og 2003). Í ljósi þessa var talið mikilvægt að fá betra yfirlit á mögulegri útsetningu hreindýra fyrir flúor frá álverinu.

Þessi rannsókn miðaði að því að bera styrk F í kjálkabeinum 67 hreindýra, sem drepin voru eða drápust í Reyðarfirði á árunum 2001-2007, áður en álverið tók til starfa, saman við styrk í 61 dýri sem drápust á árunum 2014-2020 eftir að álverið tók til starfa. Rannsóknin er unnin samstarfi Náttúrustofu Austurlands og Norsku dýralæknastofnunarinnar (Veterinærinstituttet) og er hluti af stærra verkefni sem heitir ESPIAL (Ensuring Sustainable Production of Primary Aluminium) sem miðar að því að stuðla að sjálfbærari frumframleiðslu áls.

Neðri kjálkar eru taldir góðir til að fylgjast með styrk F í beinum dýra þar sem þeir eru meðal þeirra beina líkamans þar sem styrkur F mælist hæstur. Einnig eru neðri kjálkar ákjósanlegir til að ákvarða sjónrænt aldur dýra út frá tönnum, sem og mögulegar tannskemmdir (dental fluorosis) og uppsöfnun flúors í beinum, en slíkt getur valdið óeðlilegri beinmyndun og krónískum sjúkdóm (osteofluorosis) sem í verstu tilfellum veldur vanlíðan og vanþrifnaði hjá skepnum. Skemmdir á tönnum koma einkum fram ef dýr eru útsett fyrir F á meðan þau eru ung (<1,5 ára) og tennur eru að myndast, en afmyndun beina getur komið fram hvenær sem er á ævinni, þó yngri dýr séu viðkvæmari. Styrkur F eykst í beinum eftir því sem dýr eldast.

Samband milli styrks F í kjálkabeinum hreindýra á Íslandi og mögulegra skemmda á tönnum og beinum hefur ekki verið skilgreint, en í eins og hálfs árs gömlum krónhjörtum í Noregi hafa mörk verið skilgreind. Við styrk F upp á 900-2.000 ppm er talin hætta á að ungir krónhirtir geti þróað tannskemmdir (dental fluorosis) og styrkur yfir 2.000 ppm veldur tannskemmdum. Þá hafa verið skilgreind mörk styrks F í kjálkabeinum kinda í Noregi til að meta skaðleg áhrif flúors á þær. Ekki er gert ráð fyrir skaðlegum áhrifum ef styrkur F er undir 4.500 ppm í kindum sem eru eldri en fimm ára, 3.500 ppm fyrir 2-5 ára kindur og 2.000 ppm í yngri dýrum. Engin slík skilgreining er til fyrir hreindýr eða önnur hjartardýr.

Styrkur F í kjálkabeinum hreindýra sem drápust eftir að álverið tók til starfa var marktækt hærri (að meðaltali 610 ppm í dýrum 6 ára og eldri og að meðaltali 103 ppm í ungum dýrum) en í dýrum sem drápust áður en það tók til starfa (meðaltal 180 í dýrum 6 ára og eldri og að meðaltali 94 ppm í ungum dýrum). Hæsti styrkur F í dýrum áður en álverið tók til starfa mældist 242 ppm í 12 ára gamalli kú sem drapst 2003, en mest 1.608 ppm í 3 ára gömlum tarfi í hópi þeirra dýra sem drápust eftir að álver tók til starfa. Styrkur F í dýrum sem drápust eftir að álverið tók til starfa jókst marktækt með aldri og var hæstur í dýrum næst álverinu og lækkaði marktækt eftir því sem dýrin drápust lengra frá. Engu að síður var styrkur F í kjálkabeinum hreindýra sem dvöldu í nágrenni álversins í Reyðarfirði lágur, en í öllum sýnum nema 9 af 128 mældist styrkur F undir 1.000 ppm sem mun lægra en t.d. mælst hefur í krónhjörtum við álver í Noregi (allt að 11.000 ppm).

Við ályktum að hærri styrkur F sem mælist í kjálkabeinum hreindýra í Reyðarfirði sé vegna aukins styrks F í gróðri eftir að álverið tók til starfa. Þrátt fyrir hækkaðan F styrk í kjálkabeinum hreindýra í Reyðarfirði voru engar sýnilegar tannskemmdir eða óeðlilegar beinmyndanir í kjálkunum. Dýrin eru ekki staðbundin á Reyðarfirði, heldur ferðast þau á milli svæða á ólíkum árstímum og bíta því hluta ársins á svæðum þar sem gróður er ekki útsettur fyrir flúor frá álverinu og því er meðaltal styrks F í fóðri þeirra á ársgrundvelli lægra en ella.

Ef styrkur F í gróðri í Reyðarfirði eykst frá því sem verið hefur og styrkur F í kjálkabeinum kinda sem ganga þar hækkar er ástæða til að huga frekar að heilbrigði hreindýra sem dvelja í nágrenni álversins. Sér í lagi ef þar sjást kýr með kálfa eða vetrungar í meira mæli en verið hefur, en mjólkandi dýr eru talin þola mun minni styrk F í fóðri en önnur dýr.

Introduction

Concern for wildlife and animal welfare is growing and it is important to explore the effects of aluminium smelters on wildlife and the wellbeing of animals. Reindeer roam wild in East-Iceland and small groups graze in close vicinity of the Alcoa Fjarðaál aluminium smelter in Reyðarfjörður, especially during late winter and early summer (Figure 1) (East Iceland Nature Research Centre, unpublished data, and annual reindeer monitoring reports, see www.na.is). The smelter is the only known source of non-natural exposure of fluoride (F) to reindeer in East-Iceland. Animals grazing closer to the central highlands, where volcanos have been active, could occasionally be exposed to fluoride due to eruptions and dispersion of tephra (see effects on eruptions in Iceland on livestock in Weinstein & Davison 2004) Animals from such areas are not considered likely to migrate to Reyðarfjörður. Exposure to fluoride due to volcanic eruption in reindeer in recent decades has not been documented.

Figure 1. Reindeer bulls in early summer by Alcoa - Fjarðaál aluminium smelter (Photo: Hilmar Sigurbjörnsson, Alcoa - Fjarðaál).

Reindeer in Iceland have no predators and the population size is managed through hunting. The total population doubled in the period 2000–2008, from about 2,500 animals to 5,000 animals in winter, most likely due to underestimation of population size and recruitment rates. The estimated winter population in 2020–2021 is around 5,000 animals and hunting management aims keeping the population stable. East Iceland is divided into nine reindeer hunting areas for management purposes (Figure 5) (Þórisson, Þórarinsdóttir & Ágústsdóttir 2021). The animals grazing in Reyðarfjörður are considered to belong to hunting areas 4 and 5 which extends from Seyðisfjörður to the north and to Reyðarfjörður in the south. The estimated winter population in these two areas averaged 545 animals during the period 2000–2021 with the fewest animals in winter 2000–2001 (346) and the maximum number (700) during the two winters from 2008 to 2010. Estimated number in Reyðarfjörður only averaged 107 animals during winter from 2000–

2021, with the fewest animals in 2000–2001 (52) and no animals during the most intensive construction period of the smelter from 2004 to 2006. The maximum number of animals estimated was 250 during the winter 2001–2012 (East Iceland Nature Research Centre, unpublished data). Animals grazing in Reyðarfjörður parts of the winter and sometimes during early summer migrate to other areas further away during parts of the year, especially during calving (May), hunting (August-September), rutting (October) and in early winter.

It is not well documented what reindeer in Reyðarfjörður mainly eat, but studies in other areas in Iceland suggest that the animals adapt to their surroundings, grazing mainly in Carex moors and peatbogs (starmóar og - mýrar) where available forage is of poorer quality, and in Salix heathlands (víðimóar) where available forage is of richer quality (Þórisson, 2018). Reindeer seek different habitat types during different time of year, but heath- and mossland and wetlands are important throughout the year (Þórisson & Ágústsdóttir, 2014). Lichens are important forage during autumn and winter in areas where they are available. In areas where lichens are scarcer, the main autumn and winter forage are grasses, Carex species and willow shrubs (Egilsson, 1983).

External Environmental Monitoring of F-concentration in vegetation has been carried out in Reyðarfjörður since prior to start-up of Alcoa Fjarðaál. The survey shows that F-concentration in vegetation has increased considerably in all types of vegetation (lichen, moss, gras, blueberry leaves, rowan leaves) sampled since the smelter start-up, compared to baseline study, except for rhubarb stalks and berries. The F-concentration in the vegetation is highest just west of the smelter. Visual inspection and study of vegetation cover also shows that damage due to fluoride stress in plants is mostly visible just west of the smelter (Jóhannsdóttir et.al., 2019). The F-concentration in vegetation can change rapidly in parallel with changes in the weather and the amount of fluoride in the air (Weinstein & Davison, 2004; Jóhannsdóttir et.al. 2019).

Prolonged excessive fluoride intake by livestock and cervids causes accumulation of F in bones, which can lead to fluorosis in teeth (dental fluorosis) or in bones (osteoflurosis). Fluorosis is a chronic disease causing various detrimental effects on animals, depending on many factors, such as the dose of F ingested, the durability of exposure, species, age, health, and stress condition of the animal (see in more detail in Vikøren, 2021). Effects have been described for various species, including cervids such as deer, but not specifically for reindeer. Excessive fluoride intake during tooth formation is known to affect formation of the enamel and causes dental fluorosis in cervids and livestock if it exceeds certain limits in vegetation they feed on (Weinstein & Davison, 2004; Vikøren, 2021). Young animals are thus especially vulnerable to dental fluorosis which reflects F-exposure during the first years when the teeth are formed and mineralized. Osteofluorosis has more severe symptoms than dental fluorosis, as it can change the structure of the bones when fluoride accumulates as the animal ages. Osteofluorosis can cause lameness, including breathing and chewing problems and can be induced at any time during the lifespan of the animal, but young animals are more susceptible (see in more detail in Vikøren, 2021).

The defined fluorine limit values in feed varies for individual animal species, age, and condition (e.g., lactating) of the animals. In Iceland, limits for F in feed for cervids has not been defined. The defined maximum average limits for feed with 12% moisture on annual basis for livestock ruminants, i.e., cows, sheep, and goats are 50 ppm and 30 ppm for lactating animals. Limits for horses are not specifically defined, but horses are categorized as animals considered to tolerate 150 ppm averaged on annual basis (Regulation on feed control nr. 340/2001 and subsequent amendments). In Norway the limit for wild cervids (deer and moose) is set < 30 (ppm dry weight) (see in more detail in Vikøren, 2021).

A relationship between accumulated F-concentration in jawbones and possible dental fluorosis (dose – effect) has been established for 1.5-year-old red deer in Norway based on a study in the early 1990s (the Effect Study). Due to findings in the ESPIAL Fauna study (Vikøren, 2021) the dose – effect relationship has been adjusted and, limits for risk of developing dental fluorosis in 1.5-year-old red deer is 900-2,000 ppm and F-concentration above 2,000 ppm causes dental fluorosis. For evaluation of detrimental F-effects in livestock like sheep, the Kontrollutvalget in Norway made categories based on age and accumulated F in bone. No detrimental effects are expected with values below 4,500 ppm for sheep older than 5 years, 3,500 ppm for sheep 2–5 years old and 2,000 ppm for younger animals (see in more detail in Vikøren, 2021).

A dose-effect relationship in reindeer living in Iceland has not been established.

F-concentration in jawbones from sheep grazing in Reyðarfjörður has increased since the smelter startup, both in lamb and adult sheep, compared to background values in Reyðarfjörður and to F-concentration in jawbones from sheep from other areas in Iceland where there is no non-natural source of fluoride (Jóhannsdóttir et.al, 2019). Despite of increased F-concentration values in lambs and adult sheep, the animals are generally healthy and there is little or no sign of osteo- or dental fluorosis (Þórarinsdóttir, 2021; Arnardóttir, 2021).

A study on F-concentration in jawbones of horses living close to an aluminium smelter in West Iceland (Straumsvík, Hvalfjörður) showed a significant negative regression with increased distance from the smelter, indicating that the greatest source of fluoride in the environment originates from the aluminium industry. But there was no evidence of dental fluorosis. It was also noticed that fluoride accumulation was lower in the bones of horses compared to sheep (Valgeirsdóttir, 2020).

The monitoring of vegetation and livestock in Reyðarfjörður POST-smelter (2012–2018) has shown that annual mean average F- concentration in lamb jawbones grazing in Reyðarfjörður have significant correlation with the F- concentration in grass (Jóhannsdóttir, et.al., 2019).

Due to elevated F-levels in sheep jawbones and vegetation in Reyðarfjörður after the smelter startup, a random analysis of F-concentration in jawbones of three adult reindeer bulls killed close to the Alcoa Fjarðaál aluminium smelter in 2014 was carried out. The results revealed elevated F - levels compared to analysis of F-concentration in five animals killed in 2002 and 2003 prior to smelter start-up. As reindeer have been seen for extended periods around the smelter during the last decade it was considered important to get a better understanding of the possible F-exposure of the animals grazing in the vicinity of the smelter. This study was carried out shed light on this, where background values of fluoride in jawbones from reindeer killed in the area prior to the smelter start-up (2001–2004 and 2007) were established and compared to values of fluoride in jawbones in the area during full operation of the smelter (2014 to 2020). The study was carried out in collaboration with the Norwegian Veterinary Institute (NVI) and is a part of the ESPIAL (Ensuring Sustainable Production of Primary Aluminium) project directed and funded by the Aluminiumindustriens Miljøsekretariat (AMS).

Methods

Site description

Alcoa Fjarðaál smelter started operation in April 2007. The production capacity is up to 360 thousand tonnes of aluminium per year (Alcoa Fjarðaál, 2019). Emission of fluoride to the atmosphere at full production was highest in 2009 (116 F-total tons) and lowest in 2011 (74 F-total tons) and mainly follows the amount of aluminium produced (Figure 2).

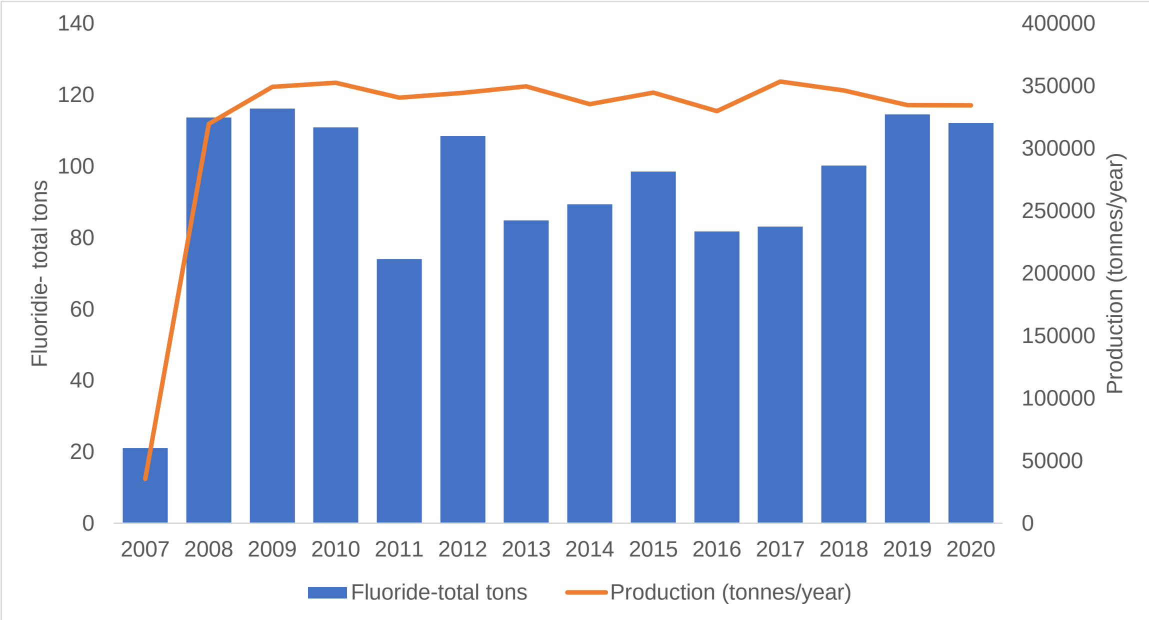

Figure 2. Total fluoride emission to the atmosphere and production of aluminium from 2007 (start-up) to 2020 at Fjarðaál (Data based on email from Sveinsson, G., environmental specialist at Alcoa Fjarðaál on the 15th of September 2021).

The smelter is located on the northern side of Reyðarfjörður in the East fjords of Iceland. The fjord is about 30 km long, from mouth in east to the bottom in west. It is about 6 km wide at the mouth (Þórarinsson et al., 1984) but narrower towards the bottom, and about 2 km wide at the smelter site. The land is rather steep on both sides of the fjord, with highest peaks about 1,000 m, which channel the wind to easterly and westerly directions. During summer, land – sea breeze effects further enhance the east – west directions. The climate is windy, humid, and cool with monthly averages around 10°C in summer and 1–2°C in winter.

Sample collection and processing

Lower jawbones are good specimen to assess effects of elevated fluoride concentration in animals, as the jawbones are among the bones in the skeleton with the highest levels of F-concentration. Additionally, age can be estimated from the status of teeth and detrimental effects of teeth (dental fluorosis) and bone (osteofluorosis) can be examined (Vikøren, 2021).

All samples were collected from the reindeer killed in hunting areas 4 and 5, which the animals grazing in Reyðarfjörður belong to. For PRE-smelter analysis, 67 lower jawbones were sampled from a large jawbone collection stored at the East Iceland Nature Research Centre. These PRE-smelter samples were from animals hunted in the Reyðarfjörður area during the hunting season (from August to September) in the period 2001–2004 and one animal dead from a roadkill in early 2007. The animals aged from 1 to 12 years old. For the POST-smelter analysis 61 lower jawbones were sampled from hunting and roadkill from 2014 to 2020, from animals aged from 0 to 7 years old (Table 1) located from 0 to 81 km from the smelter site (Figure 5). The data is shown in Appendix I.

The PRE-smelter jawbone samples were boiled in water and all meat rinsed of the bones and marked with a unique ID-number. The jawbones were then air dried and kept in a dry storage for future research. After being stored for up to two decades they were sent to NVI for analysis in early 2020. The POST-smelter jawbone samples were not boiled. Most of the meat was scraped off and the jawbones were air dried and marked with a unique ID-number before submitted to NVI in late 2020.

This study of totally 128 samples also included eight reindeer which had been F-analysed prior to this survey. Five samples in the PRE-smelter dataset were analysed for F-concentration in 2006 by the Center of Chemical Analysis at Keldnaholt, Iceland and inspected by veterinarian to determine age and for dental fluorosis, but not osteofluorosis. Three samples in the POST-smelter dataset were analysed for F- concentration in 2014 by the Innovation center Iceland, but not inspected for dental- and osteofluorosis nor age determined.

All other samples were age determined and examined at NVI for detrimental effects of fluoride on teeth and bones as described in Vikøren (2021). The teeth were classified into 6 categories based on the severity of damage, from category 1 where no effects were seen in teeth to category 6 where excessive effects were seen. Additionally, category 8 is for old age-related wear of teeth.

After inspection of detrimental effects, samples were extracted based on the “mandible standard” from the ventral part of the lower jawbones between third and fifth cheek tooth and sent to the laboratory of Hydro Aluminium Metal Årdal m in Norway for analysis of F- concentration. The analysis was carried out using standard methods. Further information about the examination for detrimental effects of fluoride and analysis of F-concentration is described in a report from NVI (Vikøren, 2021).

Statistical analyses

PRE- and POST-smelter difference in F-concentration in reindeer jawbones was assessed using descriptive statistics, and a *Welch t-test* on *log-transformed* data. Correlation between age and F-concentration in PRE-smelter data was inspected by using linear model (LM). For the POST-smelter data, age related and spatial variation in F-concentration in jawbones was studied using LM on log-transformed data for distance from smelter and untransformed data for age. In addition, a *generalized linear model* (GLM) with Gaussian distribution and a log-link function was used to study interaction of the independent variables; age of animals (years) and the distance from smelter where they were killed (km).

All analyses and data handling were conducted in *R v.4.1.0* (R Core Team, 2021). Graphs were produced using the packages *ggplot2 v.3.3.5* (Wickham, 2016) and *jtools v.2.1.3* (Long, 2020). Distance from the location of killed animals to the smelter site was measured with the pointDistance tool in the package *Raster* (Hijmans, 2020). The distance was calculated as the shortest direct line between the two points (Euclidian distance) and did not account for landscape.

Results

F-concentration in jawbones from POST-smelter animals was significantly higher than in PRE-smelter animals ($p < 0.001$) (Figure 3). A large difference in the variation of F-concentrations in jawbones was noticed between reindeer killed before and after the Alcoa Fjarðaál smelter startup (Figure 3 and Table 1). Most of the animals examined, both for PRE- and POST-smelter data, were in the age group 3–5 years (Table 1).

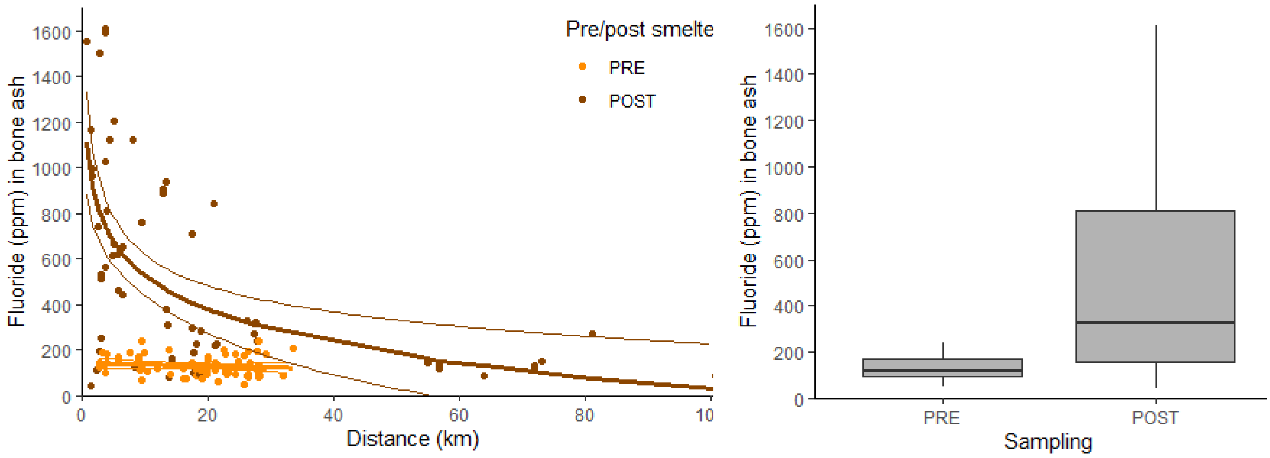

Figure 3. Left: F-concentration (ppm) in jawbones of reindeer killed PRE- and POST-Alcoa Fjarðaál aluminium smelter, showing each value along with the animal's distance from the smelter site and a regression line of the LM for both POST- and PRE smelter animals shown, with 95% confidence intervals. Right: boxplot with median (line), quartiles (box) and min/max values (lines) for each group (right).

All the animals from the PRE-smelter period had F-concentration below 250 ppm (Figure 3 and Table 1). The F-concentration increased significantly with increased animal age (R^2=0.488; $p<0.001$) with lowest value (52 ppm) measured in a one-year-old female and the highest (242 ppm) in a 12-year-old female (Figure 4). One calf in the ≤ 1-year-old group had surprisingly high F-concentration (238 ppm bone ash) (Table 1). However, that age group was still the only group within the whole dataset that had average F-concentration below 100 ppm.

The animals from the POST-smelter period had F-concentrations from 44 ppm in a 1-year-old female to 1,608 in a 3-year- old bull (Table 1 and Figure 4). All but nine animals had F-concentration below 1,000 ppm. The correlation was not as strong between F-concentration and age (R^2=0.122; $p<0.01$) for POST-smelter animals compared to PRE-smelter (Figure 4).

Table 1. The F-concentration (ppm bone ash) in the lower jawbone of reindeer grazing in Reyðarfjörður area for different age groups for the different data sets PRE-smelter (baseline, 2001–2007) and POST-smelter (2018–2020). Three samples from POST-smelter data from 2014 are not included as they were not age determined. Their F-concentrations was 614, 1,502 and 1,555 (ppm bone ash).

Sampling year		0.5*-1 year	2 year	3-5 year	6 year and older
PRE-smelter 2001-2007	Number	7	8	36	17
	Mean	94	101	123	180
	Min	52	76	82	111
	Max	238	150	187	242
POST-smelter 2018-2020	Number	4	12	34	7
	Mean	103	267	595	610
	Min	44	88	98	272
	Max	127	1,025	1,608	1,168

* only one sample

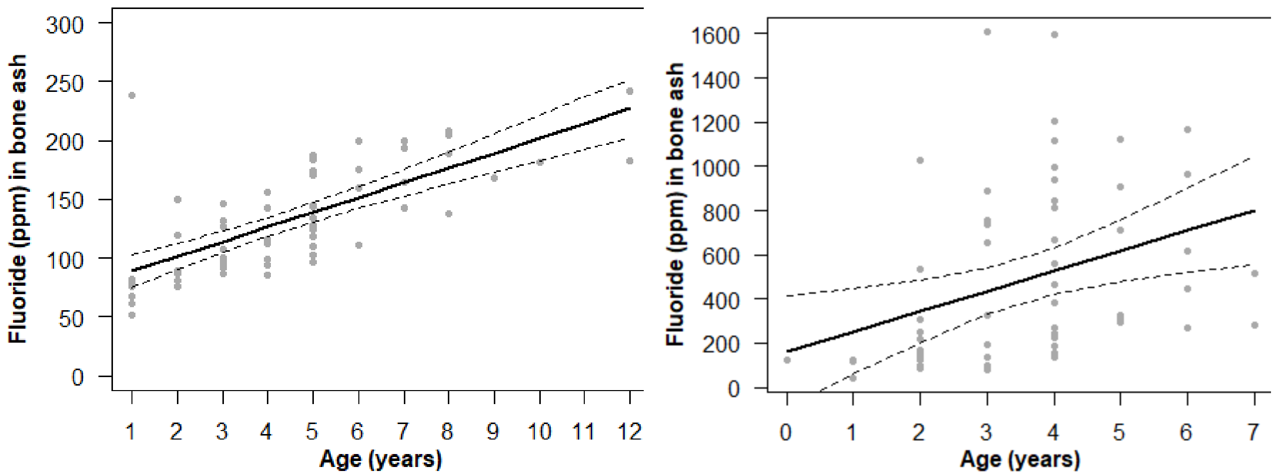

Figure 4. F-concentration (ppm) in jawbones vs. age in PRE-smelter reindeer (left) and POST-smelter reindeer (right). Regression lines of the LM are shown, with 95% confidence intervals (dotted lines). Notice different values on Y-axis.

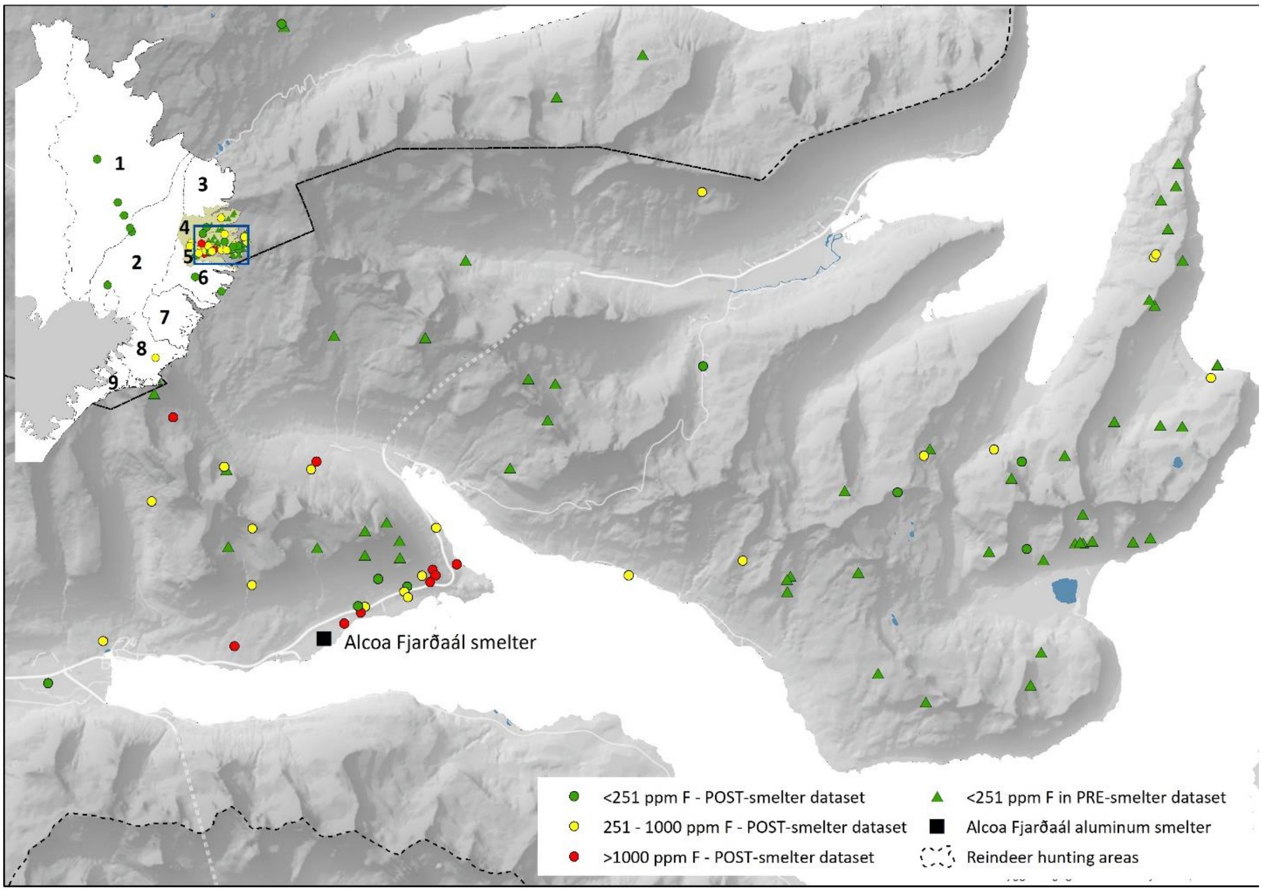

Figure 5. Location of the dead reindeer sampled for analysis and the F-concentration in their jawbone ash. The nine hunting areas in East Iceland are shown on the overview map embedded. F-concentration is classified into 3 classes: 1) less than 251 ppm F which defines the background value limits in Reyðarfjörður based on the PRE- smelter samples. 2) 251-1,000 ppm F and 3) above 1,000 ppm F.

The F-concentration varied more by distance in jawbones sampled from POST-smelter animals compared to F-concentration in jawbones from PRE-smelter animals (Figure 3). F-concentrations in reindeer killed after the smelter startup decreased with increasing distance from smelter

(R^2=0.352, $p < 0.001$) but this pattern was not obvious for PRE-smelter animals (R^2=0.0152, $p < 0.321$) (Figure 3 and Figure 5). POST- smelter jawbone samples from animals killed >35 km from the smelter had similar values and variation of F-concentrations as seen in the PRE-smelter data (Figure 3). The highest F-levels were found in animals shot close to the smelter. According to the GLM model the interaction between the two variables age and distance was not significant (p = 0.74).

None of the reindeer examined showed gross signs of osteofluorosis in the jawbone or dental fluorosis. The examination of the teeth from both PRE- and POST-smelter animals in Reyðarfjörður classified them as either category 1 (normal), 2 (questionable) or 8 (old age-related wear). The majority of reindeer were classified as having normal teeth, both in PRE-smelter animals (69%) and in POST-smelter animals (76%). About 1/5 of samples in both groups (19%) were classified as having questionable effects: the exact cause could not be determined. Many of these animals in category 2 (both PRE- and POST-smelter) had a slight brown enamel discoloration of one or several of the premolar and molars (cheek teeth), but we could not conclude that this was related to F-exposure. The data set from PRE-smelter had a higher proportion of older animals and thus 11% were classified as having old age-related dental wear compared to 5% in the POST- smelter animals (Table 2).

Table 2. Number and proportion (%) of reindeer in both POST- and PRE- smelter samples grouped in dental fluorosis categories.

		Post-smelter			Pre-smelter		
Effects	**Dental fluorosis-category**	**No. of animals**	**%**	**Age range**	**No. of animals**	**%**	**Age range**
Normal	DF 1	44	76%	0-7	43	69%	1-8
Questionable	DF 2	11	19%	3-6	12	19%	1-8
Old age-related	DF 8	3	5%	6	7	11%	7-12
Total*		58	100%		62	100%	

*5 animals from PRE- smelter data set and 3 animals from POST-smelter dataset were analyzed by Innovation Center Iceland were not examined for fluorosis.

Discussion and Conclusion

We conclude that a moderate increased fluoride load is documented in reindeer living in the vicinity of the Alcoa Fjarðaál smelter 2018-20, but no indisputable gross signs of fluorosis have been found. Higher F-concentration in jawbones of reindeer POST-smelter compared to PRE-smelter is concluded to be due to higher F-concentration in the vegetation that reindeer graze on in Reyðarfjörður area after smelter start-up. The F-concentration in vegetation POST-smelter (2008–2018) is significantly higher than in the baseline study (PRE-smelter 2004–2006) for all vegetation species (Jóhannsdóttir et.al., 2019), including lichens and moss which are known to be an important source of feed for reindeer especially during winter (Egilsson, 1983).

Part of the year the animals graze in closest vicinity to the smelter (0–2 km) where average values of F-concentration in vegetation is highest. In the period of 2008–2019, the F-concentration in this area in moss was 224 ppm and 67 ppm in lichens, which is above the tolerance threshold for wild cervids (<30 ppm) (see in Vikøren, 2021). The F-concentration in vegetation decreased significantly with increasing distance from the smelter as was seen in the bone F-concentration of reindeer

(Jóhannsdóttir et.al., 2019). All reindeer that were hunted in more than 12 km distance from the smelter site had F-concentration < 1,000 ppm in their jawbones.

Despite the increased bone F-concentration compared with the PRE-smelter F-level (baseline), the F-accumulation in the bones of the reindeer did not reach limits that is known to cause detrimental effects in cervids, and there was no sign of visible dental fluorosis or osteofluorosis. The fact that the animals migrate to areas further away from the smelter during large parts of the year and graze on none F-exposed vegetation could explain that. Lactating female reindeer with calves have not been observed in the area closest to the smelter, but yearlings and young bulls are often seen in groups with adult bulls. All the youngest reindeer (0.5–1.5-year-old) in the POST-smelter group had F-concentration below 1,000 (ppm bone ash). For 1.5-year-old red deer from Norway, a F dose – F effect (visible dental fluorosis) relationship have been made, and no effects are expected at F-levels < 1,000 ppm (in 2021 adjusted to < 900 based on the ESPIAL study) (Vikøren, 2021).

Even tough F-concentration in the jawbones of reindeer grazing near Alcoa Fjarðaál in Reyðar-fjörður have increased significantly from baseline, the values are very low compared to cervids grazing near smelters in Norway. As an example, eight of twelve roe deer living within the fenced area around Hydro Karmøy had F-concentration above 11,000 ppm (Vikøren, 2021). Some of the animals suffered from osteofluorosis and/or dental fluorosis. The highest F-value in jawbones in Reyðafjörður was 1,608 ppm in a 3-year-old reindeer bull and with no signs of dental- or osteofluorosis. The nomadic way of life of reindeer seems beneficial to avoid prolonged high F-exposure near the smelter.

If F-concentration in vegetation in Reyðarfjörður increases significantly from what has been measured since the smelter start-up and if F-concentration in bone ash from sheep jawbones reaches values above limits for possible detrimental effects, it will be important to monitor further the health of the reindeer grazing in the vicinity of the smelter. The same applies if lactating female reindeer with calves become common in the area. Young growing ruminants are particularly prone to F-toxicosis due to tooth development and skeletal growth and limits for F concentration in feed are lower for lactating animals.

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