Vegetation survey at Årdal, Sunndal and Mosjøen aluminium smelters in 2019-2020

Impact of fluoride emissions on local vegetation

NIBIO REPORT | VOL. 7 | NO. 144 | 2021

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DATO/DATE:	RAPPORT NR./ REPORT NO.:	TILGJENGELIGHET/AVAILABILITY:	PROSJEKTNR./PROJECT NO.:	SAKSNR./ARCHIVE NO.:
01.02.2023	7/144/2021	Open	11341	18/01397

ISBN:	ISSN:	ANTALL SIDER/ NO. OF PAGES:	ANTALL VEDLEGG/ NO. OF APPENDICES:
978-82-17-02904-5	2464-1162	64	3

OPPDRAGSGIVER/EMPLOYER:	KONTAKTPERSON/CONTACT PERSON:
Aluminiumindustriens Miljøsekretariat (AMS)	Leif Ongstad

I

STIKKORD/KEYWORDS:	FAGOMRÅDE/FIELD OF WORK:
Stikkord norske: Fluoridskader på vegetasjon Keywords: Fluoride damage on vegetation	Forest pathology, Fluoride damage on vegetation

SAMMENDRAG/SUMMARY:

Målet med denne studien var å gi en oppdatering om effekter av fluor (F)-utslipp på vegetasjon rundt tre aluminiumssmelteverk. Vi besøkte Årdal og Sunndal smelteverk i 2019-2020 og Mosjøen i 2020, vurderte og dokumenterte de visuelle symptomene på F-skader på vegetasjon og relaterte disse til påviste verdier av F i plantevev.

Tre plantearter viste kvaliteter som nyttige indikatorer: Rogn, furu og perikum. Fordi ormetelg akkumulerte ekstreme F-verdier og viste tydelige beiteskader, kan overvåking av denne arten være berettiget på grunn av den mulige helsefaren for beitedyrene.

I Årdal og Sunndal har vi i løpet av 2019 og 2020 påvist de høyeste F-verdiene hos ormetelg, fra 94 til 925 mg F/kg. I rogn ble den høyeste F-konsentrasjonen påvist i trær som vokste i arealet til Årdal smelteverk (1161 mg F/kg), men på alle andre lokaliteter varierte F-konsentrasjonene i rogn fra 4 til

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327 mg F/kg. Hos furu varierte F-konsentrasjonene fra 6-351 mg F/kg for alle nålealdre, men eldre nåler samlet alltid mer F enn yngre. I perikum var de akkumulerte F-verdiene fra 10-84 mg F/kg. Ved alle smelteverk var det en gradient av avtagende F-konsentrasjon i vegetasjon med økende avstand fra smelteverket.

F-utslipp i Årdal (henholdsvis 12 og 11 kg F/time i 2019 og 2020) og i Sunndal i løpet av 2019 (12 kg F/time) var bare litt høyere enn anbefalte grenser (10 kg F/time) for skader på vegetasjon, mens i Mosjøen var F-utslippene 7 kg F/time i 2020. F-skader på vegetasjon stemte overens med rapporterte utslippsnivåer. På bakgrunn av denne vurderingen er utslippsreduksjoner fortsatt tilrådelig i Årdal og Sunndal, mens situasjonen er akseptabel i Mosjøen.

Summary

The scope of this study was to provide an update on fluoride (F) emission effects on vegetation around three aluminium smelters. We visited Årdal and Sunndal smelters in 2019-2020 and Mosjøen in 2020, assessed and documented the visual symptoms of F-damage on vegetation and related these to detected values of F in plant tissue.

Three plant species showed qualities as useful indicators: Rowan, pine and St. John's wort. Because male-fern accumulated extreme F-values and showed clear grazing damage, the monitoring of this species may be warranted because of the potential health hazard for the grazing animals.

In Årdal and Sunndal, during 2019 and 2020, we detected the highest F-values in male-fern, ranging from 94 to 925 mg F/kg. In rowan, the highest F-concentration was detected in trees growing within the Årdal smelter (1161 mg F/kg) but on all other locations the F-concentrations in rowan ranged from 4 to 327 mg F/kg. In pine, the F-concentrations ranged from 6-351 mg F/kg for all needle ages, but older needles always accumulated more F than younger ones. In St. John's wort the accumulated F-values ranged from 10-84 mg F/kg. At all smelters there was a gradient of decreasing F-concentration in vegetation with increasing distance from the smelter.

F-emissions in Årdal (12 and 11 kg F/hour in 2019 and 2020, respectively) and in Sunndal during 2019 (12 kg F/hour) were only slightly higher than the recommended limits (10 kg F/hour) for damage on vegetation, while in Mosjøen the F-emissions were 7 kg F/hour in 2020. The presence of F-damage on vegetation was consistent with the reported emission-levels. On basis of this evaluation, reductions in emissions are still advisable in Årdal and Sunndal, while the situation is acceptable in Mosjøen.

LAND/COUNTRY: Norway

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Preface

Results of an extensive Effect Study, sponsored by AMS Aluminium (Aluminiumindustriens Miljøsekretariat), were published in 1994 (Norsk aluminiumindustri og miljø, 1994). The study summarised effects of aluminium production on vegetation, soil, water, farm and game animals and human health.

This report is commissioned by AMS Aluminium and is a part of a multidisciplinary project, “Ensuring the Environmental Sustainability of Production of Primary Aluminium (ESPIAL)”, an update to the original Effect Study. This particular report focuses on emission effects on vegetation around three aluminium smelters: Årdal, Sunndal and Mosjøen.

The objectives of this report were to investigate the fluoride pollution gradients around the aluminium smelters in relation to visible symptoms on vegetation and compare the current situation with results from the Effect Study.

All photographs are taken by Isabella Børja, unless otherwise stated in the figure captions.

Ås, 01.02.23

Isabella Børja, Ari M. Hietala, Nina E. Nagy, Halvor Solheim, Volkmar Timmermann

Contens

1 Introduction

1.1 Background

In the early 1990'ies the Norwegian aluminium industry commissioned an extensive study to evaluate the local environmental impacts of its activities (the Effect Study, Norsk aluminiumindustri og miljø 1994). Since then, the production technology and biological assessment methods have improved significantly. The current project, Ensuring the Environmental Sustainability of Production of Primary Aluminium (ESPIAL), is a multidisciplinary project aiming to update and advance the knowledge regarding the environmental consequences of the production of primary aluminium and minimise the negative environmental impact of the production process. To reach this goal, the industry needs tools to detect, identify and monitor various emissions and an expertise to interpret whether these emissions might be harmful to the environment. The institutes involved in the original Effect Study, Norwegian Institute for Water Research (NIVA), Norwegian Institute for Air Research (NILU), Norwegian Institute of Bioeconomy Research (NIBIO) and Veterinary institute are also involved in the current ESPIAL project.

The emission effects on vegetation, were investigated by Horntvedt & Øyen (1994) as part of the Effect Study. Since the Effect Study, the individual smelters had local vegetation sampling campaigns, however these varied among the smelters and different plant species were sampled. Therefore, no comparable update of vegetation sampling and evaluation since the Effect Study is available. In ESPIAL we sampled and evaluated vegetation at three out of the seven smelters assessed in the Effect Study. Our study is based on one-day field campaigns around Årdal and Sunndal smelters in 2019 and 2020, and one-day field campaign around Mosjøen smelter in 2020.

1.1.1 AMS Aluminium (Aluminiumindustriens Miljøsekretariat)

ESPIAL study was commissioned by AMS Aluminium - a joint force for the primary aluminium plants in Norway, Sweden and Iceland on Environmental, Health and Safety projects.

AMS Aluminium was established in 1976. Four companies (Alcoa, Hydro, Kubal and Rio Tinto), with 10 primary aluminium smelters in Iceland, Sweden and Norway and one aluminium refinery in Norway, are currently members of AMS Aluminium.

AMS Aluminium is a project-oriented organization, pursuing its goals/mission for the members by initiating projects in the areas of work environment, emissions and effects on humans and ecosystems.

1.1.2 Smelters selected for vegetation sampling

Hydro Årdal and Hydro Sunndal were selected for sampling in 2019 and 2020, complemented with Alcoa Mosjøen in 2020. Some key production and meteorological data for the three smelters are given in Table 1.1. The technological development during the last three decades indicates that although the production of aluminium has steadily increased the fluoride emissions remained similar for the last 30 years (Figure 1.4).

Table 1.1. The aluminium smelters participating in the survey: production and emission in tonnes per year (t/yr), annual precipitation and length of growing season in 2019-2020.

Locality	Primary aluminium production[1] (t/yr) 2019/2020	Fluoride emission[1] (t/yr) 2019/2020	Growing season[2] (days, 2019/2020)	Annual precipitation[3] 2019 (mm)	Annual precipitation[3] 2020 (mm)
Årdal	209.761/211.473	104/95	207/232	744.1	1035.2
Sunndal	403.380/409.000	108/83	216/256	-	1319.7
Mosjøen	197.504/200.179	61/58	158/158	1345.7	1702.4

[1] **Source: Smelters (2020)**
[2] **Days >5°C, Source: seklima.met.no**
[3] **Source: Norsk Klimaservicesenter (no data available for Sunndal 2019)**

Hydro Årdal

The Hydro Årdal facilities comprise a primary metal smelter at Øvre Årdal (Figure 1.1), 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 11 km. The plants are located in a narrow valley surrounded by high mountains.

At Øvre Årdal, the valley branches into the Moadalen, Utladalen, Fardalen and Nundalen valleys. Winds blowing either up or down the valley occur with roughly the same frequency. There is little difference between winter and summer in this respect.

The Årdal plant started up in 1948 using Søderberg technology. Today it consists of two regular prebake reduction lines and a test centre where Hydro's technology department is testing improved reduction technology. The total production capacity is about 210.000 tonnes per year (t/yr, Table 1.1). Årdal Karbon at Årdalstangen manufactures anodes for prebake plants. A little more than half of the anode production of 190.000 tonnes per year goes to the metal plant in Øvre Årdal, the rest goes to Høyanger and to other Hydro plants.

Figure 1.1. Hydro Årdal

Hydro Sunndal

The Sunndal plant lies at Sunndalsøra (Figure 1.2), at the mouth of the Driva river. The Sunndal valley is a long, deep, U-shaped valley flanked on both sides by mountains 1000 - 1800 meters high.

The predominant wind directions are up the valley in summer and during the day, and down the valley in winter and at night.

The Sunndal plant started up in 1954 with the first potline (SU1), and the second (SU2) commenced operations in 1958. Both of these potlines 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. Increased amperage over the years has also resulted in increased output, up to a total of 409. 000 tonnes per year in 2020 (Table 1.1).

The Sunndal plant also has production of about 80.000 tonnes of anodes per year, covering about 40% of the anode consumption in the potlines. The remaining amounts of anodes are imported.

Figure 1.2. Hydro Sunndal. Photo: Ole Erik Loe

Alcoa Mosjøen

The Alcoa Mosjøen (Figure 1.3) aluminium plant lies directly north of Mosjøen centre, at the mouth of the Vefsna and Skjerva rivers. Vefsndalen valley is a long, relatively open valley flanked on both sides by mountains about 800-1000 meters high. Further down the Vefsnfjord, there are mountains 6-800 meters high on the west and north, while the hills to the east, facing Fustvatnet, are lower. There are conifer forests along the Vefsndalen valley, while vegetation is sparse further down the fjord and on the steepest mountain sides.

The prevailing wind is up the valley in the summer and during the day, while there is often a wind from the land in winter and at night. The aluminium plant started in 1958 with an annual production of 25.000 tonnes. Today the plant consists of four potrooms, all prebake, with a total capacity of nearly 200.000 tonnes per year (Table 1.1). Two of the prebake lines were converted from Søderberg in 1987-89, and the latest conversion took place in 2004-05. A new, large anode plant was completed in 2008, with a capacity of 300.000 tonnes of baked anodes per year and supplies anodes to Alcoa Mosjøen smelter and Alcoa Fjardaal (Iceland).

Figure 1.3. Alcoa Mosjøen

1.1.3 Emission of fluorides

During the production of aluminium, several phytotoxic air pollutants, such as fluorides (F) and sulphur dioxide (SO2), are released to the environment. Concentrations of these pollutants in the air vary according to industrial emissions, weather conditions and topography. Of the commonly occurring pollutants, F is considered as the most toxic and damage on vegetation around aluminium smelters is mostly attributable to F. While usually low in the atmosphere, the F-concentration close to emission sources can vary considerably between episodic peaks and values near to zero.

The most radical improvements in the emission levels took place in the 1980'ies and early 1990'ies. Since then, the replacement of Søderberg technology with prebake technology gave much lower emissions per tonne aluminium produced, but due to concurrent production increase, the fluoride emission levels are now close to the same level as around 1994, with about 100 tonnes F/year for Hydro Årdal and Sunndal, and 60 tonnes F/year for Alcoa Mosjøen (Table 1.1, Figure 1.4).

Although the gas treatment centres have a high cleaning efficiency, some fugitive emissions from the pots are released during the pot operations (e.g., anode changing). The major part of the fluoride emissions is therefore released through the roof ventilators of the potrooms.

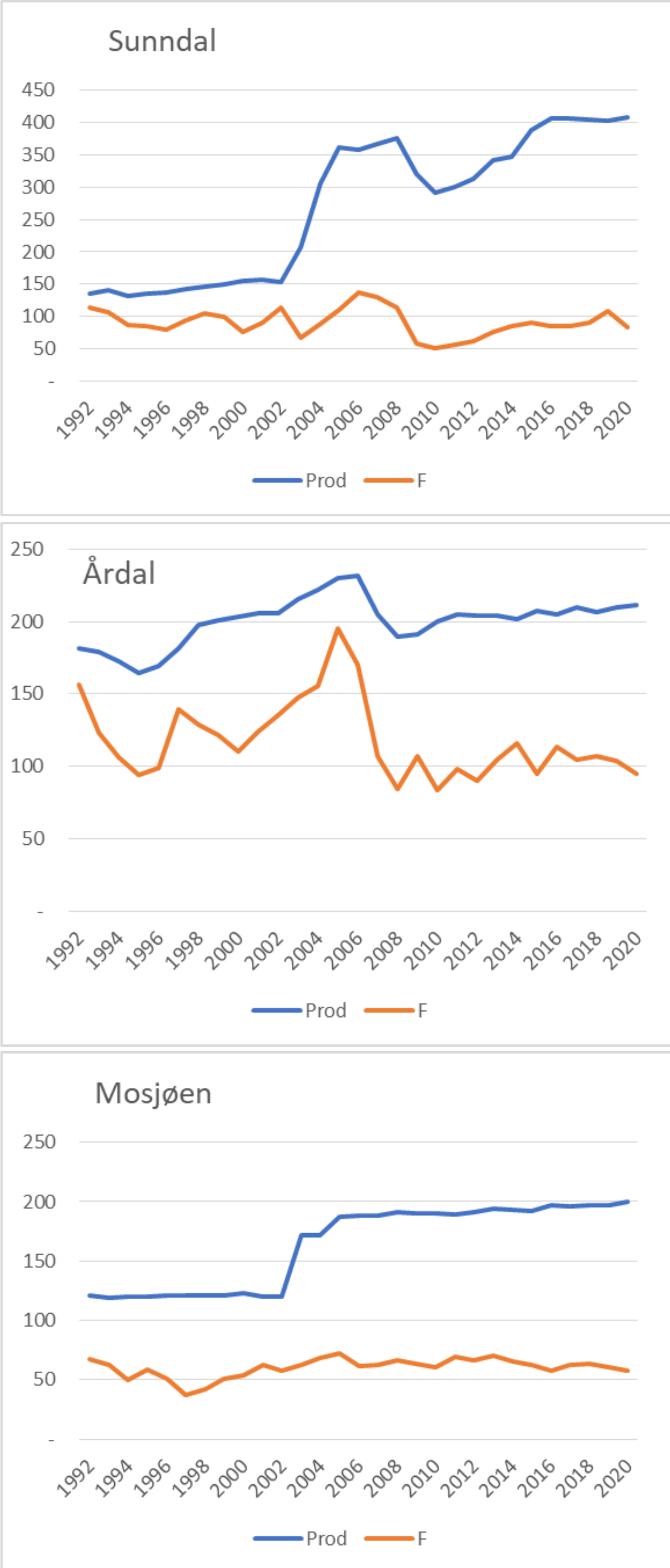


Figure 1.4. Production (Prod, t/yr) and fluoride emissions to the air in period 1992-2020, both particulate and gaseous, (F, t/yr) from Hydro Årdal, Hydro Sunndal and Alcoa Mosjøen (data obtained from: Årdal, Sunndal and Mosjøen smelters). The data for fluoride emissions to the air only include emissions from potlines (gas treatment + potroom ventilation) and anode production. In recent years, some minor emissions from the cast houses have also been reported in Norskeutslipp.no. Here we chose to exclude the cast house emissions, since these measurements are available only for few years. These emissions may amount to 10 - 15% of the potline emissions.

1.1.4 Vegetation monitoring and foliar analysis

The emissions of fluorides to the air are monitored at the sources, providing accurate measurements. When emissions disperse, the vegetation canopy is the very first surface available to these fugitive atmospheric pollutants. Vascular plants absorb the fluoride emissions through the stomata in their leaves and consequently accumulate fluoride in the tissue. The uptake depends on the concentration and composition of fluoride in ambient air and the duration of exposure, but it also varies with vegetation structure, plant species and growth conditions (Horntvedt & Øyen 1994). This uptake capacity makes vascular plants convenient organisms for monitoring airborne F-pollutants. Therefore, establishing a network of biomonitoring plants around large emitters is a feasible way to constantly follow the emission levels and to evaluate the efficiency of air control measures. Although the weather variations and topography may largely influence and overshadow any variations in the emissions, damage assessments of vegetation may still provide a valuable indication of emission levels. In addition, vegetation monitoring can provide a relatively cost-effective tool to assess the dispersion pattern in a qualitative way.

Fluoride is especially suitable for monitoring by biological methods as the visible symptoms in sensitive plants are rather distinctive and quantification of its accumulation in plants is feasible. Because fluoride, unlike sulphur or nitrogen, is not an essential element for plants and its uptake from the soil is very small, the background concentration in plant tissue is quite low, usually between 0 and 10 mg F/kg dry matter (Horntvedt & Øyen 1994). Although other sources mention that in unpolluted areas F-concentrations in plant tissue are in the range from 2-20 mg F/kg (Committee on Biological Effects of Atmospheric Pollutants, 1971), in this report we use the background threshold of 10 mg F/kg used by Horntvedt & Øyen (1994), as we surveyed vegetation at the same smelters. Therefore, any accumulation of F over about 10 mg/kg in foliage generally indicates the presence of atmospheric fluoride contamination.

The dispersion of fluoride emissions can be assessed through a) modelling, b) direct measurements in the air or c) by monitoring of selected plants. Modelling underestimated the range of dispersion, especially in complex terrains (Norsk aluminiumindustri og miljø, 1994). Direct air measurements are expensive and can only be performed at specific points and time periods. Biomonitoring is relatively cost-effective and feasible, can cover a larger area and is suitable for documenting the emission dispersion pattern and effects on vegetation. These advantages compensate for the inaccuracies in accumulation factors in different species.

There is a guideline value for total fluoride of 1 µg/m^3 ambient air as annual average to prevent harmful effects on plants, livestock and humans (WHO 2000). However, there is currently no official EU threshold limit for fluoride emissions (mostly in gaseous form as hydrogen fluoride, HF) causing damage to vegetation, only recommendations. Values, inducing appearance of foliar necroses, were recommended by the Norwegian Pollution Control Authority (SFT, 1992) and these were also used in the Effect Study by Horntvedt & Øyen (1994). Garrec (2021) published more recent damage limits by HF, which are less strict than those suggested by SFT (Table 1.2).

Table 1.2. Threshold concentrations of ambient HF causing damage on vegetation, as a function of exposure time.

Averaging period	HF (µg /m3)	
	SFT (1992)	Garrec (2021)
24 hours	1.0	1.5
1 month	0.4	0.9
Growing season/6 months	0.3	0.5

Based on their extensive data, Horntvedt & Øyen (1994) recognised 5 classes of F-damage in plants, related to F-concentrations detected in the rowan tissue (Table 1.3). With some reservations this framework can be used to relate the detected rowan F-concentrations to ambient F-concentrations and observed damage on plants. The "damage scale" presented by Horntvedt & Øyen (Table 1.3, 1994) indicates that F-related damage on sensitive plants occurs at lower F-concentrations (0.2-0.6 µg /m³) than those specified in the recommended air quality criteria by SFT (0.3 µg /m³, Table 1.2).

Table 1.3. Damage in plants exposed to increasing F-concentrations. Effects of different fluoride levels, measured as fluoride in rowan (F_{rowan}) following 100 days of exposure (5 May - 15 August). The corresponding concentration in the air (F_{air}) is calculated using the formula $F_{rowan} = K \bullet F_{air} \bullet T$. The K-value for rowan is 1.7. The table is adopted from Horntvedt & Øyen (1994).

Class	F_{rowan} (mg/kg)	F_{air} (µg /m³)	Damage in plants
1	0-10	<0.02	None. Background level
2	10-30	0.02-0.2	None
3	30-100	0.2-0.6	Damage on sensitive species
4	100-200	0.6-1.2	Damage on more species
5	>200	>1.2	Damage on many species, increasing risk for forest damage

1.2 Identification of fluoride damage symptoms

Fluorides in emissions from aluminium smelters occur in gaseous form (mostly as hydrogen fluoride, HF), but also as particle-bound fluorides. Gaseous F is absorbed from the air through stomata in leaves and passes into the transpiration stream where it is carried to its final site of accumulation in the tips and margins of leaves (Figure 3.1). The uptake is greatest when plants are most active (when stomata are open), during the day throughout the growing season. Particulate F is deposited on the plant surfaces and during rain it may to a certain degree be absorbed through the cuticle.

If the accumulation of F exceeds a tolerance threshold for the particular species, yellowing at the leaf edges and between the leaf veins (marginal and interveinal chloroses) are usually the first symptoms observed (see Appendix 1 for details). Depending on the species, reddish discoloration, dead tissue (anthocyanosis, necroses) and growth distortion (spoon-like distortion of leaves or missing leaf tips) may also occur (Appendix 1). Typical symptoms of F-damage on plant leaves (see also Chapter 3.1, Figure 3.1) are brown (dead) leaf margins and tips, sharply divided from the living tissue by a distinct dark line. Often a thin yellow zone is in front of the dark, dividing line, indicationg the chlorophyll degradation (Figure 3.1). Only little F moves downward in plants to roots, from leaf to leaf or from leaves to fruits. Young leaves and needles are the most sensitive to F and the degree of sensitivity decreases with age, until late in the growing season when they become relatively tolerant. This is most likely related to the normal metabolic shifts as tissues mature. However, leaves/needles of non-deciduous plants are known to accumulate F with time, the concentrations increasing in the tissues with years of exposure.

Although the F-damage symptoms on a plant may be typical, they may easily be confounded by other stress factors, such as drought stress or salt toxicity, that display similar symptoms. Therefore, to assess the cause of symptoms at each location, all other factors that may contribute to similar symptom development must be taken into consideration.

Higher plants show a vast range in F tolerance. Species that are sensitive in one region may be less sensitive in another because of differences in climatic factors, soil properties, as well as genetic differences. Therefore, setting up of a monitoring programme requires an individual approach designed specifically for each area.

1.3 Vascular plants in environmental monitoring: Bioindicators and biomonitors

Because plant foliage is effectively accumulating atmospheric pollutants it makes plants suitable and cost-effective indicators of the environmental status. In addition, use of plants as indicators may be regarded as an effective tool for detecting potential health risks to certain animals (mainly ruminants) feeding on the plants near sources of fluoride emissions.

Biomonitoring of air pollutants can be passive or active. Passive methods observe plants growing naturally within the area of interest, as in the case of ESPIAL. Active methods, in turn, detect the presence of air pollutants by placing test plants of known response into the study area.

Depending on the sensitivity and uptake of pollutants plants may serve as bioindicators or biomonitors. A bioindicator is any species whose function can reveal the **qualitative** status of the environment. The best bioindicator for airborne F is usually the most susceptible species with clear signature symptoms in the foliage. The presence of a good bioindicator enables conclusions based on the single indicating species instead of monitoring the whole plant community.

Within the ESPIAL study the sensitive species such as St. John's wort (*Hypericum* spp.), rowan (*Sorbus aucuparia*) and Scots pine (*Pinus sylvestris*) are examples of good bio indicators, all showing clear and early F-damage symptoms on their foliage.

A biological monitor or biomonitor is a species that may provide **quantitative** information on the pollution status of the surrounding environment. Because the leaves of biomonitors have the capacity to retain a proportion of the absorbed F after the exposure, their leaf analysis can provide an estimate of the intensity of exposure and a rough approximation of the concentration of F, provided the dynamics of F uptake and loss are known (Davison 1983). The formula often used to calculate the F-uptake is

$$F_{plant} = K * F_{air} * T$$

where F_{plant} is the F-concentration in plant tissue after exposure to F in the air, F_{air} is the concentration of gaseous and particulate fluoride in the air and T is the duration of the exposure. K is the accumulation coefficient, that depends on the plant species in question (Committee on Biological Effects of Atmospheric Pollutants, 1971; Horntvedt & Øyen 1994).

In the Effect Study, Horntvedt & Øyen (1994) found in rowan a clear linear relationship between the accumulation of F in the leaves and the duration of exposure to fluoride in ambient air (Figure 3.9). As this linear F-accumulation in rowan was consistent also across different sites (Figure 3.9), they singled out rowan as a good biomonitor that may also be used for estimating the retrospective F-concentrations in the air during the exposure period. Thus, rowan is a species suited both as bioindicator as well as biomonitor.

In the ESPIAL study, in addition to rowan, also pine may be considered as a biomonitor, as the presence of several annual needle age groups would enable estimation of the intensity of F-exposure over several years.

1.4 Objective and scope of the study

The objective of the present study was to assess the accumulation and effects of fluoride emissions on vegetation around Hydro Årdal, Hydro Sunndal and Alcoa Mosjøen smelters. This work included:

- Site visits during two growing seasons (2019 and 2020) to assess the health status of selected plant species including morphological changes in leaves, and accumulation of fluorides in plant tissue.

- Evaluation of the new vegetation data, in light of results obtained by the Effect Study from 1994.

- Evaluation of sampling procedure and historical vegetation data and give recommendations for standardization.

- Collect and systemise the existing data from chemical analyses related to vegetation monitoring at all smelters in a common database.

2 Material and Methods

2.1 Studied sites

For the vegetation monitoring program, we sampled plants at three locations around Årdal smelter (2019 and 2020, Figure 2.1), four and six locations around Sunndal smelter in 2019 and 2020, respectively (Figure 2.2), and six locations around Mosjøen smelter in 2020 (Figure 2.3). The sampling locations represented different distances from the respective smelters. In addition, in Årdal we also sampled vegetation at different elevation levels at the Timrebakkane hill, 100 - 760 m.a.s.l. close to the smelter.

2.1.1 Hydro Årdal

On the 22nd of August 2019 we collected leaf samples from three standard locations up to 4 km away from the smelter: Moa 0.72 km, Melheim 2 km and Hjelle 4 km (Figure 2.1). These locations were the same as in the Effect Study. In addition, we also collected rowan leaves from an "ad hoc" location inside the Årdal smelter area. These rowan trees were growing in front of the potline-vents and had clear F-damage symptoms and general signs of decline.

In 2020 we had no access to Årdal smelter area (due to the corona-situation) and we sampled on 17th August 2020 the same three locations outside the smelter. We sampled "ad hoc" also rowan and maple leaves with symptoms from Årdalstangen, 11 km southwest from the smelter (Figure 2.1), against the main wind direction.

To assess the effect of elevation, we sampled plant material at the Timrebakkane hill, right above the Hydro smelter at 100 - 760 m.a.s.l. during both the 2019 and 2020 surveys (Figure 2.1, Figure 3.3).

Figure 2.1. Hydro Årdal smelter and vegetation sampling sites at different distances from the smelter. At Timrebakkane the vegetation was sampled at different elevations, from 100 - 760 m.a.s.l. (Photo: modified from www.google.com/maps).

2.1.2 Hydro Sunndal

Leaf samples were collected during the 5th of September 2019 at 4 locations at different distances from the smelter: Einangen (4.5 km), Grøa (9 km), Hoås (11 km) and Gravem (30 km). All locations in 2019 were the same as those in the Effect Study. In 2020 the samples were collected during 1-2nd September and two additional locations were included: Skjølland (1.5 km) and Lensmannsøra (3 km) (Figure 2.2).

Figure 2.2. **Hydro Sunndal smelter and vegetation sampling sites at different distances from the smelter. Locality Ålvundeidet (11 km from the Sunndal smelter) was used as a reference site in 2020. (Photo: modified from www.google.com/maps).**

2.1.3 Alcoa Mosjøen

Leaf samples were collected during the 28th of August 2020 at 6 locations up to 10 km away from the smelter: Marsøraveien (1 km), Kjerringlia (1 km), Haug (2 km), Hestremveien (4 km), Finnmyro (4 km) and Holandstunnellen (10 km) (Figure 2.3). These locations were different than those used in the Effect Study and were selected based on the following criteria: Presence of the main four sampled species (rowan, pine, male-fern and St. John's wort) and a similar distance gradient as at the other two smelter sites.

Figure 2.3. **Alcoa Mosjøen smelter and vegetation sampling sites at different distances from the smelter. (Photo: modified from www.google.com/maps).**

2.2 Sampled plant species

At each site we assessed trees and herbs known to be sensitive to accumulation of F, but also the remaining vegetation, for visual symptoms of F-damage. In Årdal (2019 and 2020) we sampled ten different species/genera and analysed 120 samples in total. In Sunndal (2019 and 2020) we sampled eleven different species/genera and analysed 104 samples in total. In Mosjøen (2020) we sampled seven different species/genera and analysed 54 samples in total (Table 2.1).

In 2020 we adjusted the sampling around all smelters based on the F-analyses results from 2019 and sampled mostly plant species that seemed to be the best indicators of F-accumulation in plant tissues: rowan, St. John's wort, Scots pine (in the further text called "pine") and male-fern. A few samples from other species were taken as reference (Table 2.1).

Table 2.1. Overview over all sampled species at different locations during 2019 and 2020. n= total number of analysed samples.

Location	Species/Genus		n
Årdal 2019,2020	Rowan	*Sorbus aucuparia*	
	Male-fern	*Dryopteris filix-mas*	
	Scots pine	*Pinus sylvestris*	
	Birch	*Betula spp.*	
	Grass	*not identified*	
	Goat willow	*Salix caprea*	
	Lady-fern	*Athyrium filix-femina*	
	St. John's wort	*Hypericum spp.*	
	Bilberry	*Vaccinium myrtillus*	
	Maple	*Acer sp.*	
			120
Sunndal 2019, 2020	Rowan	*Sorbus aucuparia*	
	Male-fern	*Dryopteris filix-mas*	
	St. John's wort	*Hypericum spp.*	
	Scots pine	*Pinus sylvestris*	
	Lady-fern	*Athyrium filix-femina*	
	Elm	*Ulmus sp.*	
	Aspen	*Populus tremula*	
	Grey alder	*Alnus incana*	
	Bilberry	*Vaccinium myrtillus*	
	Eagle fern	*Pteridium aquilinum*	
	Maple	*Acer sp.*	
			104
Mosjøen 2020	Rowan	*Sorbus aucuparia*	
	Male-fern	*Dryopteris filix-mas*	
	Scots pine	*Pinus sylvestris*	
	Spruce	*Picea abies*	
	Lady-fern	*Athyrium filix-femina*	
	Small ferns	*Gymnocarpium dryopteris, Phegopteris connectilis*	
	Fireweed	*Chamaenerion angustifolium*	
			54

2.3 Sampling procedure

In 2019 we sampled leaves of plants showing characteristic F-damage symptoms, but also some plants without symptoms for reference. In 2020, however, we sampled systematically both plants with typical symptoms and nearby plants without symptoms, to be analysed separately. All samples were taken from adult plants. Whenever possible, samples of different individuals of the same plant species were pooled prior to the lab analysis. At each sampling site, plant samples were placed in labelled plastic bags and later photographed individually to document the symptoms. This enabled us to later relate the visual symptoms to F-content in the same tissue. Samples of pine and spruce needles were separated according to age into current-year needles, 1-year-old needles, etc. All leaf samples of deciduous trees included petioles. Samples of other plants included all the above-ground parts except the lower, leafless stalk. After the photographic documentation, the samples were stored in paper bags, for further chemical analysis of F-concentration in the tissue.

2.4 Chemical analysis

In 2019 the analyses of F-concentrations in leaf tissues from Årdal and Sunndal smelters were performed by the chemical laboratory facilities in Årdal smelter. In 2020 all chemical analyses were performed at laboratories at each participating smelter but were following the same protocol.

Briefly summarized, the analytic procedure for fluoride analysis was performed as follows: Unwashed leaves were dried at 80°C for 24 hours and ground in a mixer. Three parallel replicates of each sample were analysed (with about 2 g sample in the two first replicates and 1 g in the third).

Fluoride was then extracted with 0.1 M perchloric acid and the F-concentration was measured by Fluoride ion selective electrode. The results of F-concentrations in individual samples were entered in an Excel spreadsheet.

2.5 Statistical analysis

Mean F-concentration in vegetation samples was calculated for each plant species at each site. The standard error of the means was calculated whenever a sufficient number of analysed samples was available. However, due to the numerous specifics in the material (different species, distances, elevations etc.), in most cases the number of analysed samples was too small for further statistical analysis.

When calculating mean F-concentration in rowan, we excluded the extreme value detected in declining rowan trees at the Hydro Årdal site right outside the potroom vents (1161 mg F/kg).

In the tables in the Result chapter (Table 3.4, Table 3.5, Table 3.6, Table 3.7,Table 3.8), we set the mean F-accumulation in rowan to 100% at each location and established a rough index showing the uptake capacity of other sampled plants relative to rowan as described in Horntvedt & Øyen (1994). We chose rowan as this was the tree species found to have consistently the highest F-values of all tree species, present at most locations and showing clear symptoms of F-damage.

To compare our results with those of Horntvedt & Øyen (1994) in the Effect Study we used the available median values for all detected F-concentrations at the same locations in ESPIAL (Årdal 2019-2020 and Sunndal 2019).

3 Results

In general, during the vegetation assessments around Årdal, Sunndal and Mosjøen smelters, we did not see any immediately eye-catching F-related damage on vegetation. However, at a closer examination, especially in sensitive species, we found typical signature signs of F damage. The symptoms were most pronounced in Årdal, followed by Sunndal, while in Mosjøen the F-damage symptoms were sparse and found mostly on rowan.

During the sampling campaigns in 2019 and 2020, four species were singled out as useful indicators for various reasons. First, rowan, as a useful biomonitor as it consistently accumulated F in proportion to the ambient F-concentration and duration of the exposure. Rowan has also good bioindicator qualities as it displayed clear, typical symptoms of F-damage (Figure 3.1), and it grew at most of the locations we studied. Second, pine, as a bioindicator, showing clear typical symptoms of F-damage (Figure 3.1). As pine was the only non-deciduous tree, accumulating consistently increasing amounts of F in older needles, it may be used also as a biomonitor. Third, St. John's wort, as it showed early and clear signs of F-damage and could thus be useful as an "early warning"-bioindicator. Because St. John's wort accumulated F in tissue only moderately and inconsistently, this species is not suitable as a biomonitor. Finally, male-fern, as it accumulated extremely high amounts of F, up to 10 times more than rowan. Although not suited as a biomonitor or bioindicator, because we observed frequent grazing damage on male-fern at all sites, this species should be monitored due to the potential hazard for animals that have male-fern as substantial part of their diet.

3.1 Typical F-damage symptoms

In rowan we observed both the typical and non-typical F-damage symptoms. The typical, here called "edge"-symptom, was necrotic scorched tissue along the edges of the leaflets (Figure 3.1, A). The non-typical, less known symptoms, here called "tip"-symptom, were dead tips of the leaflets (Figure 3.1, B). We detected comparably high F-concentrations in rowan leaves with both types of symptoms.

Typical symptoms of F-damage on ferns were brown/dead tissue along the edges of the pinnate leaflets, often with dark line separating the live and dead tissue (Figure 3.1, C).

St. John's wort showed consistently visual symptoms of F-damage at all sites where it was found: scorched tips of the leaves, sharply delineated from the living tissue by a dark line (Figure 3.1, D).

On pine the distinct F-symptoms were dark brown/burgundy needle tips with a distinct dark line between the dead tissue in the needle tip and the living, green basal part of the needle (Figure 3.1, E).

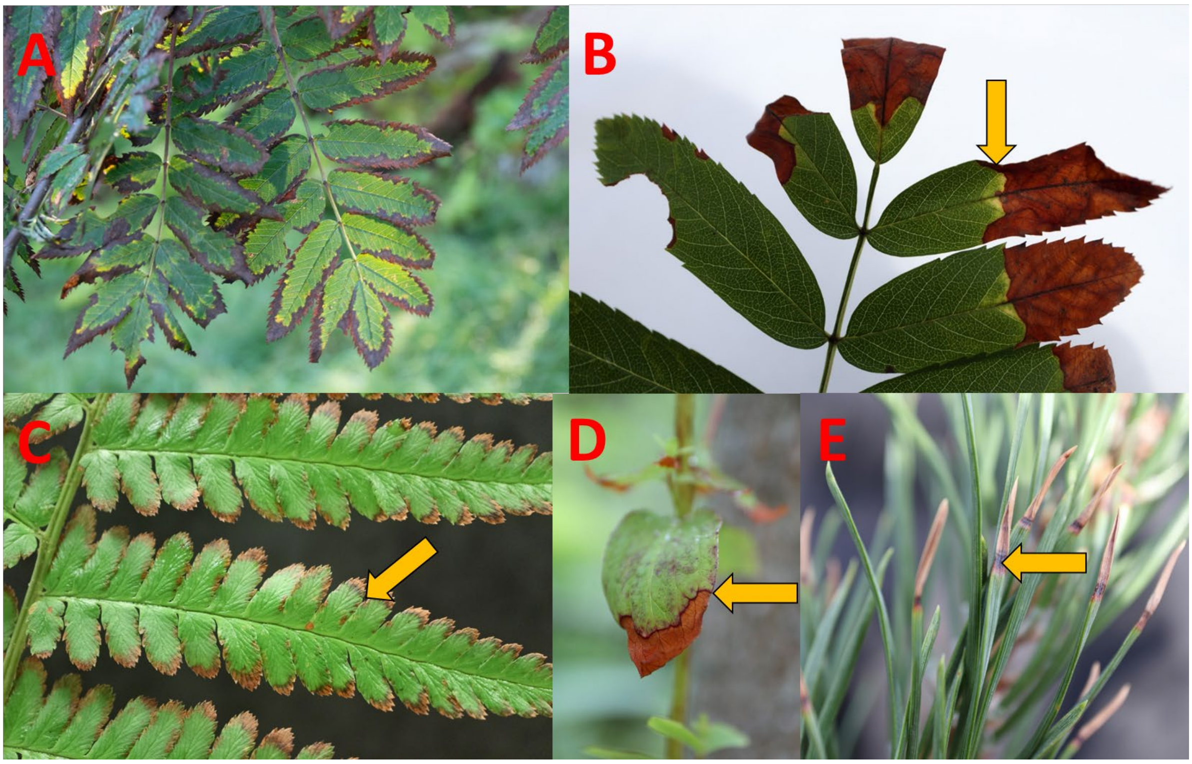


Figure 3.1. Typical F-damage symptoms in rowan, male-fern, St. John's wort and pine. A: Typical rowan "edge"-symptom with dead tissue along the edges of the leaflets. B: Non-typical "tip"-symptom on rowan with scorched, dead leaflet tips. C: Typical F-damage on male-fern with dead edges of the pinnate leaflets. D: Typical F-damage on St. John's wort, with scorched, leaf-tips. E: Typical F-damage on pine needles with dead, brown needle tips. Notice in all cases the characteristic, sharp, dark line separating the dead tissue from the living one (marked with yellow arrows).

3.2 F-concentrations in sampled plants

Considering all sampled species in ESPIAL dataset we found that in Årdal and Sunndal, 70% and 49%, respectively, of the analysed samples had F-concentrations higher than 30 mg F/kg (damage class 3-5 by Horntvedt & Øyen, 1994) while in Mosjøen the F-concentration never exceeded 30 mg F/kg (Table 3.1). Most of the samples with F-concentrations exceeding 200 mg F/kg were found in Årdal and Sunndal, 18 and 8% of all samples, respectively (Table 3.1).

Table 3.1. Distribution of all collected vegetation samples in different damage classes according to detected F-concentrations at Årdal and Sunndal (2019 and 2020) and at Mosjøen 2020.

Class	F plants (mg/kg)	Årdal n samples	Sunndal n samples	Mosjøen n samples
1	0-10	6	12	30
2	10-30	30	41	24
3	30-100	48	31	0
4	100-200	15	11	0
5	>200	21	8	0
Total n samples		120	103	54

We detected the highest median F-concentrations in Årdal and the median was similar for 2019 and 2020 (Table 3.2). In Sunndal the median F-concentrations were slightly lower than in Årdal in 2019 but in 2020 it decreased to about half of that detected in 2019 (Table 3.2). This anomaly was possibly caused by low amount of material used (or available) for analysis or the local weather conditions. In Mosjøen the median was the lowest of all three smelters (Table 3.2).

Table 3.2 **Median values for F-concentrations in all leaf samples at all locations in Årdal (2019 and 2020), Sunndal (2019 and 2020) and Mosjøen (2020).**

Location	Median ESPIAL
Årdal 2019	51
Årdal 2020	57
Sunndal 2019	42
Sunndal 2020	23*
Mosjøen 2020	8

*The majority of the analysed samples was marked to have “low amount of material” by the lab, possibly influencing the F-concentration results.

When we compared median values of F-concentrations in vegetation sampled during ESPIAL with those sampled during the Effect Study (Horntvedt & Øyen, 1994) we found that medians for Årdal (2019 and 2020) were only slightly higher in ESPIAL than in the Effect Study (Table 3.3). In Sunndal we compared only medians from 2019, as we sampled two additional localities in 2020 that were not part of the Effect Study. ESPIAL median values in Sunndal were almost twice as high as those detected in the Effect Study (Table 3.3). We could not compare medians in Mosjøen as we sampled different locations than those in the Effect Study.

Table 3.3. **Median values for F-concentrations in all leaf samples at all locations in Årdal and Sunndal (2019-2020) compared to those in all samples at all locations in Årdal and Sunndal 1990-1993 (Horntvedt & Øyen, 1994).**

Location	Median ESPIAL 2019-2020	Median Effect Study 1990-1993
Årdal	56	52
Sunndal 2019	42	20

3.3 Hydro Årdal vegetation survey

3.3.1 F-concentrations in sampled plant species

During 2019 and 2020 we collected and analysed 120 samples. In both 2019 and 2020, all the detected means for F-concentrations (within the same species at the same location) exceeded the background concentration of 10 F mg/kg, except for grass at one location (Table 3.4, Table 3.5).

Sampling campaign in 2019:

All sampled plants (except grass) showed clear visual symptoms of F-damage typical for the different species.

The mean F-concentrations were the highest in male-fern leaves, ranging from 374 to 416 mg F/kg (Table 3.4).

In rowan we detected mean concentrations ranging from 98 mg F/kg to 245 mg F/kg (Table 3.4). By far the highest F-concentration of all analysed plant materials was detected in a group of rowan trees (1161 mg/kg), growing right in front of the potroom vents at the Årdal smelter site (Appendix 1). This was an ad hoc location, selected because of the severely declining rowan trees, and was sampled only once. Because of the extreme F-values detected in leaves of the trees, we excluded these values when calculating the mean F-values for rowan.

Although St. John's wort consistently displayed typical F-damage symptoms (scorched leaf tips, Figure 3.1), the mean concentrations from all samples showed rather low F-levels ranging from 31 to 41 mg F/kg (Table 3.4).

In pine trees with typical F-symptoms (Figure 3.1) we detected mean F-concentrations from 23 to 145 mg F/kg for all needle ages (Table 3.4)

In 2019 we also sampled birch leaves showing slight yellowing between the leaf veins (Appendix 1) and detected high levels of mean F-concentration, 142 mg F/kg (Table 3.4). The goat willow leaves had similar symptoms as birch leaves: yellowing of tissue between the leaf veins but the mean F-concentrations ranged from 20 to 340 mg F/kg (Table 3.4).

Grass had mean concentrations ranging from 8 - 62 mg F/kg (Table 3.4) but showed no visual symptoms of F-damage.

When accumulation of F in rowan was set as 100% at each locality and compared to other species, male-fern was the only sampled species that repeatedly accumulated more F than rowan, 1.5-2 times more (Table 3.4).

Table 3.4. Årdal 2019: Mean F-concentrations detected in analysed species. Relative F-concentrations in leaves of analysed plants compared with rowan (100%). Sampling locations (distance to smelter), sampled species and F-concentration in plant tissue per species (mean values if more than one sample per species, n=number of samples). Total number of samples: 47.

Location	Species/Genus	n	Mean F (mg/kg)	% of rowan
Moa (0.72 km)	Rowan	3	212.8	100
	Male-fern	1	416.5	196
	Pine	3	144.5	68
	Birch	1	141.7	67
	Grass	3	62.0	29
	Goat willow	1	58.6	28
Timrebakkane (1 km)	Rowan	4	245.3	100
	Male-fern	2	374.3	153
	Goat willow	1	340.5	139
	Pine	3	142.3	58
	Lady-fern	2	53.2	22
Timrebakkane (2 km)	Rowan	2	33.4	100
	St. John's wort	1	31.0	93
	Lady-fern	1	16.2	49
Melheim (2.5 km)	Pine	6	48.1	
	St. John's wort	1	35.1	
	Grass	1	7.5	
Hjelle (4 km)	Rowan	1	98.3	100
	St. John's wort	1	41.4	42
	Lady-fern	1	25.7	26
	Pine	5	22.6	23
	Goat willow	1	20.3	21
	Grass	1	11.4	12

Sampling campaign in 2020:

All sampled plants had typical F-damage symptoms (Figure 3.1).

Also, in 2020, male-fern had the highest mean F-concentrations of all sampled plants at all Årdal locations, ranging from 221 to 732 mg F/kg. Rowan had lower mean F-concentrations than male-fern, ranging from 36 to 150 mg F/kg. In pine the concentrations ranged from 14 to 194 mg F/kg for all needle ages and in St. John's wort from 29-57 mg F/kg (Table 3.5).

When accumulation of F in rowan was set as 100% at each locality and compared to other species, male-fern showed 3.4-to-4.9-fold higher F-concentration than rowan. At the location closest to the smelter, also pine and goat willow accumulated slightly more F than rowan, 1.3 and 1.2 times, respectively. All the remaining species had lower F-accumulation than rowan (Table 3.5).

Table 3.5. Årdal 2020: Mean F-concentrations detected in the analysed species. Relative F-concentrations in leaves of analysed plants compared with rowan (100%). Sampling locations (distance to smelter), sampled species and F-concentration in plant tissue per species (mean values if more than one sample per species, n=number of samples). Total number of samples: 73.

Location	Species/Genus	n	Mean F (mg/kg)	% of rowan
Moa (0.72 km)	Rowan	5	149.6	100
	Male-fern	4	732.1	489
	Pine	3	194.4	130
	Goat willow	2	171.4	115
	Birch	2	74.7	50
Timrebakkane (1 km)	Rowan	3	118.3	100
	Pine	6	78.9	67
	Lady-fern	3	62.4	53
	Bilberry	2	44.7	38
Timrebakkane (2 km)	Rowan	1	36.4	100
	Lady-fern	2	15.8	43
	Pine	6	13.5	37
	Bilberry	1	10.8	30
Melheim (2.5 km)	Rowan	2	64.1	100
	Male-fern	2	221.8	346
	St. John's wort	2	57.2	89
	Lady-fern	1	47.6	74
	Pine	7	31.8	50
	Birch	1	29.7	46
Hjelle (4 km)	Rowan	3	66.6	100
	Male-fern	2	228.4	343
	Small ferns	1	55.7	84
	Bilberry	2	52.6	79
	St. John's wort	3	29.1	44
	Pine	5	24.4	37
Årdalstangen (10 km)	Rowan	1	45.2	100
	Maple	1	21.5	48

3.3.2 Effect of distance on F-concentration in vegetation

The concentration of F in vegetation tissue declined with increasing distance to the smelter in both 2019 and 2020 (Table 3.4, Table 3.5, Figure 3.2).

During both years we detected the highest F-concentrations for plants at the closest locations, Moa and Timrebakkane hill, ranging from the lowest in bilberry (45 mg F/kg) to the highest in male-fern (732 mg F/kg, Table 3.4, Table 3.5). The F-concentration in vegetation declined considerably at Melheim, 2.5 km from the smelter, ranging from 7.5 mg F/kg in grass to 222 mg F/kg in male-fern.

Similar mean F-concentrations were detected also at the most distant sampling point at Hjelle, 4 km from smelter, with F-concentration ranging from 11 mg F/kg in grass to 228 mg F/kg in male-fern (Table 3.4, Table 3.5).

When considering only the four main indicator species: male-fern, rowan, pine and St. John's wort, the tendency was the same, with highest concentrations closest to the smelter and declining with increasing distance to the smelter (Figure 3.2).

We sampled also symptomatic rowan and maple in Årdalstangen, an ad hoc location 11 km from the smelter, against the main wind direction. At this site rowan and maple showed elevated F-concentrations (45 and 22 mg/kg, respectively, Table 3.5).

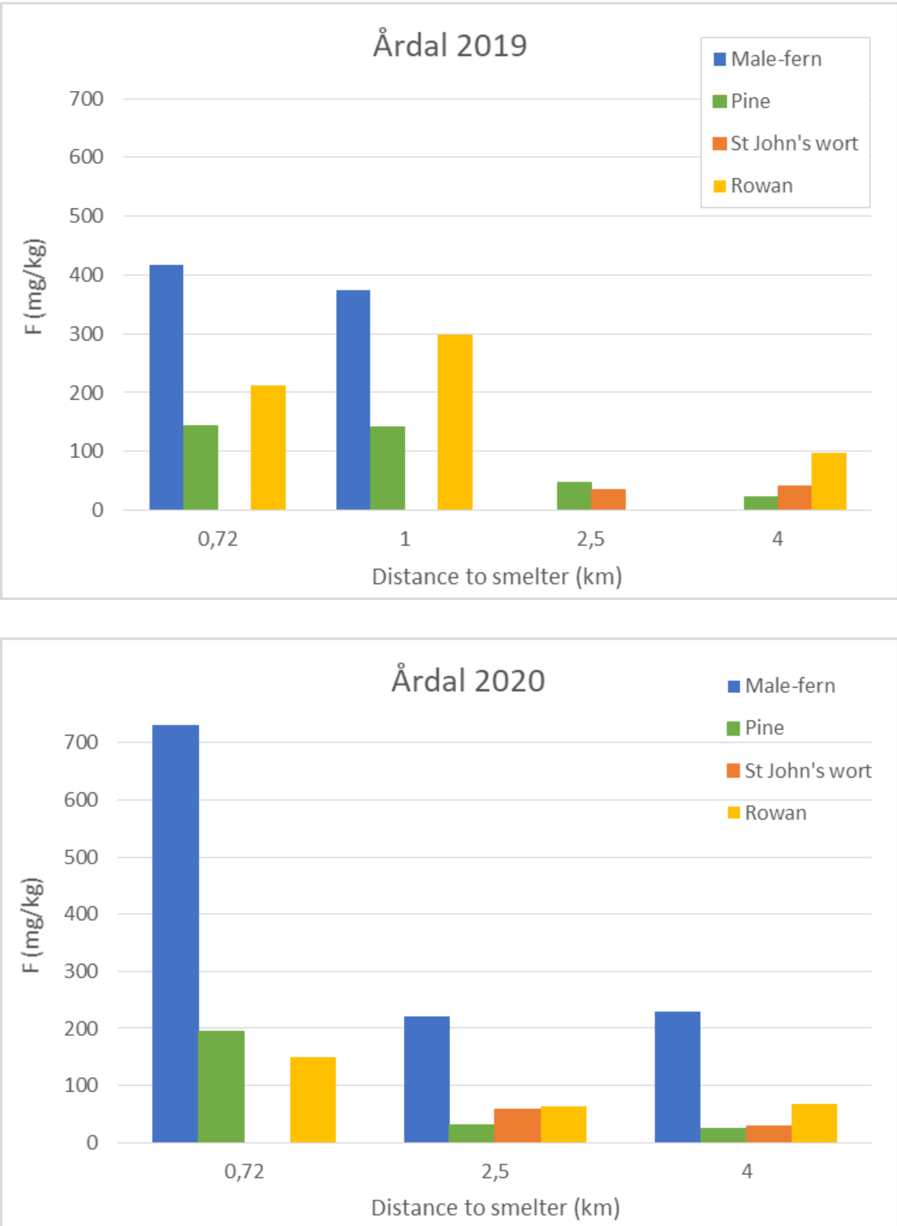


Figure 3.2. Mean F-concentrations in four plant species (male-fern, pine, St. John's wort and rowan) sampled at sites around Hydro Årdal smelter in 2019 (upper panel) and 2020 (lower panel). Locations and distances to smelter were: Moa (0.72km), Timrebakkane (1 km), Melheim (2.5 km) and Hjelle (4 km).

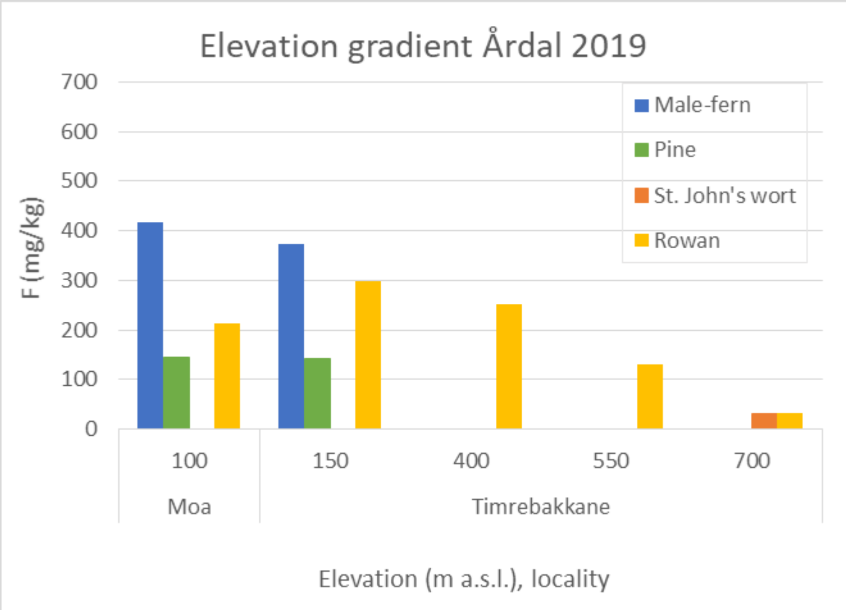
3.3.3 Effect of elevation gradient on F-concentration in vegetation

In general, mean F-concentrations in sampled vegetation (male-fern, rowan, St. John's wort and pine) decreased with increasing elevation (Figure 3.3).

In 2019 the mean F-concentrations in sampled vegetation were high at elevations from 100 to 400 m.a.s.l., ranging from the lowest in pine 142 mg F/kg to the highest in male-fern 417 mg F/kg. The mean F-concentrations declined gradually in vegetation sampled at 550 and 700 m.a.s.l., with the lowest in St. John's wort, 31 mg F/kg, and the highest in rowan (131 mg F/kg, Figure 3.3, upper panel).

In 2020 the male-fern had the highest mean F-concentration at Moa (732 mg F /kg), about 100 m.a.s.l. and closest to the Årdal smelter, while pine and rowan showed much lower concentrations at the same elevation, 194 and 150 mg F/kg, respectively. At elevations from 490-760 m.a.s.l., the mean F-concentrations in sampled plants declined, ranging from the lowest in pine (14 mg F/kg) to the highest in rowan (181 mg F/kg, Figure 3.3, lower panel).

Our results confim the findings of Horntvedt & Øyen (1994) from the same location 30 years earlier, who detected slightly increasing F-concentrations in rowan up to 400 m.a.s.l., then sharply declining with the increasing elevation.



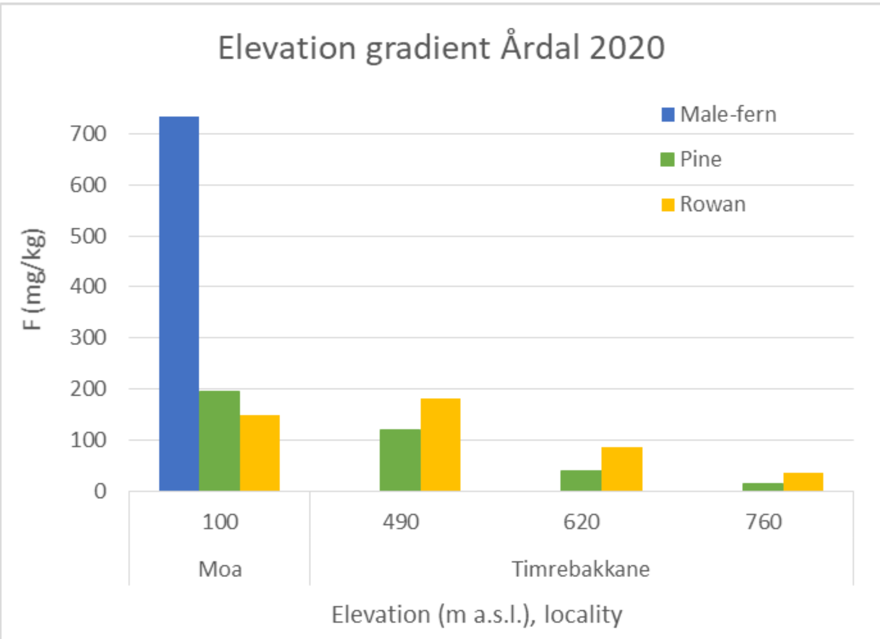


Figure 3.3. **Elevation gradient at Hydro Årdal. Mean F-concentrations in plant species (male-fern, pine, rowan and St. John's wort) sampled in 2019 (upper panel) and 2020 (lower panel) at different elevations above the Årdal smelter (0 m.a.s.l.). Missing data points indicate locations where the targeted plant species were not present.**

3.4 Hydro Sunndal vegetation survey

During 2019 and 2020 we collected and analysed 104 samples. In both 2019 and 2020, all the detected means for F-concentrations exceeded the background concentration of 10 F mg/kg, except for pine at Gravem, the most distant location from smelter (30 km, Table 3.6, Table 3.7) and rowan and lady-fern at Ålvundeidet, the "ad hoc" reference site.

3.4.1 F-concentrations in sampled plant species

Sampling campaign in 2019:

In general, at all sites there were only few typical F-symptoms visible on the surveyed vegetation. We noticed typical symptoms on rowan and St. John's wort. At Hoås we noticed some F-symptoms on pine, but mostly on current-year needles.

The highest mean concentrations in Sunndal area were detected in male-fern, ranging from 137 to 358 mg F/kg, and rowan, ranging from 72 to 146 mg F/kg (Table 3.6). The mean F-concentration in St. John's wort ranged from 52-84 mg/kg F (Table 3.6). The remaining vegetation showed mean F-concentrations ranging from the lowest in goat willow (18 mg F/kg) to the highest in bilberry (62 mg F/kg, Table 3.6).

When accumulation of F in rowan was set as 100% at each locality and compared to other species, only male-fern had higher concentrations than rowan ranging from 1.6 to almost three-fold higher than those of rowan (Table 3.6).

Table 3.6. **Sunndal 2019: Mean F-concentrations detected in analysed species. Relative F-concentrations in leaves of analysed plants compared with rowan (100%). Sampling locations (distance to smelter), sampled species and F-concentration in plant tissue per species (mean values if more than one sample per species, n=number of samples). Total number of samples: 26.**

Location	Species/Genus	n	Mean F (mg/kg)	% of rowan
Einangen (4.5 km)	Rowan	1	131.6	100
	Male-fern	3	215.5	164
	Elm	1	41.3	31
	Aspen	1	39.1	30
	Goat willow	1	32.2	24
Grøa (9 km)	Rowan	1	131.3	100
	Male-fern	1	358.2	273
	St. John's wort	1	51.7	39
	Grey alder	1	41.9	32
	Birch	1	28.4	22
Hoås (11 km)	Rowan	1	145.5	100
	St. John's wort	1	83.8	58
	Bilberry	1	61.7	42
	Lady-fern	1	40.1	28
	Eagle-fern	1	38.3	26
	Maple	1	29.6	20
	Pine	3	27.0	19
Gravem (30 km)	Rowan	1	72.1	100
	Male-fern	1	137.0	190
	St. John's wort	1	53.2	74
	Bilberry	1	23.6	33
	Goat willow	1	18.2	25

Sampling campaign in 2020:

In 2020 we found typical F-symptoms on ferns, rowan, St. John's wort on all locations, while on pine the symptoms were less visible.

Male-fern had the highest mean F-concentrations ranging from 98-381 mg F /kg, followed by rowan ranging from 22-97 mg F /kg (Table 3.7). On the other hand, pine and St. John's wort had much lower F-concentrations, ranging from 8-28 mg F/kg (Table 3.7).

When accumulation of F in rowan was set as 100% at each locality and compared to other species, male-fern showed 3-to-10-fold higher F-concentrations than rowan at the same locality, Grøa, about 9 km from the smelter (Table 3.7). Also, grey alder and lady-fern had slightly higher F-accumulation, about 1.7 and 1.2 times higher than that detected in rowan, respectively. The other species had lower F-accumulation than rowan (Table 3.7).

We sampled also a reference site (Ålvundeidet, 11 km north of the smelter) where none of the plants had any F-damage symptoms and F-values in all samples were close or below the background level (10 mg F/kg, Table 3.7).

Table 3.7. **Sunndal 2020: Mean F-concentrations detected in analysed species. Relative F-concentrations in leaves of analysed plants compared with rowan (100%). Sampling locations (distance to smelter), sampled species and F concentration in plant tissue per species (mean values if more than one sample per species, n=number of samples). Total number of samples: 78.**

Location	Species/Genus	n	Mean F (mg/kg)	% of rowan
Skjølland (1.5 km)	Rowan	2	43.5	100
	Male-fern	2	183.5	422
	St. John's wort	2	14.8	34
	Pine	5	14.2	33
Lensmannsøra (3 km)	Rowan	2	96.9	100
	Male-fern	2	352.9	364
	Lady-fern	2	76.9	79
	St. John's wort	2	27.6	28
	Pine	6	20.7	21
Einangen (4.5 km)	Rowan	2	58.6	100
	Male-fern	2	159.9	273
	St. John's wort	2	21.3	36
Grøa (9 km)	Rowan	2	36.5	100
	Male-fern	2	381.3	1045
	Grey alder	2	61.1	167
	Lady-fern	2	37.5	103
	Pine	5	20.5	56
	St. John's wort	2	17.6	48
Hoås (11 km)	Rowan	2	55.9	100
	Lady-fern	2	68.2	122
	Bilberry	2	22.9	41
	St. John's wort	2	22.4	40
	Eagle-fern	2	18.3	33
	Pine	5	17.1	31
Ålvundeidet (11 km)	Rowan	1	4.2	100
	St. John's wort	1	10.1	240
	Lady-fern	1	5.3	126
Gravem (30 km)	Rowan	2	21.5	100
	Male-fern	2	98.3	457
	St. John's wort	2	12.6	59
	Pine	8	8	37

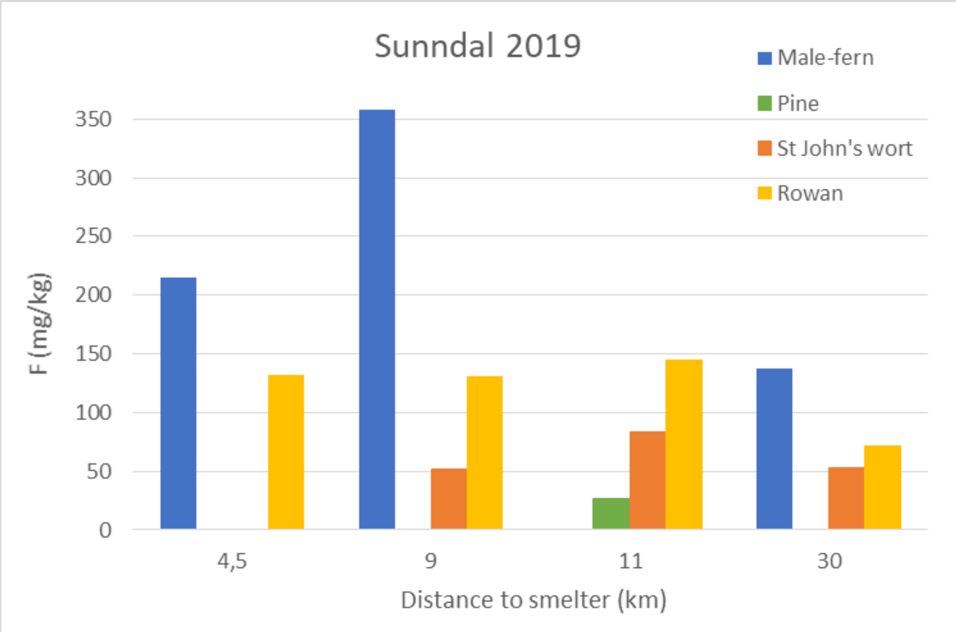
3.4.2 Effect of distance on F-concentration in vegetation

In 2019 and 2020 the F-concentration in sampled plants was rather even among the locations and decreased only in the most distant location Gravem, 30 km from the smelter (Table 3.6, Table 3.7).

In 2019, when considering only the four main indicator species (male-fern, rowan, pine and St. John's wort) male-fern had always the highest F-concentrations at the closest locations, 216 and 358 mg F/kg at Einangen and Grøa, respectively, and 137 mg F/kg at the furthest location Gravem. Rowan had similar F-concentrations (from 131 to 146 mg F/kg) at the three closest locations, but considerably lower (72 mg F/kg) at the most distant Gravem (Figure 3.4, upper panel).

Also, in 2020 the F-concentrations in the indicator species at the five closest locations were even, but all decreased somewhat at the most distant one, Gravem (Figure 3.4, lower panel). Male-fern had the highest concentrations at all four closest locations, ranging from 160 to 381 mg F/kg, but decreased at the most distant Gravem to 98 mg F/kg. Rowan, pine and St. John's wort had also the lowest concentrations at Gravem (Figure 3.4. lower panel).

Sunndal-smelter is located in a long, U-shaped valley which acts as a "chimney", where fumes have only limited escape possibilities. Therefore, the F-concentrations were elevated even at the at the most distant location, Gravem. Our findings are consistent with those of Horntvedt & Øyen (1994).



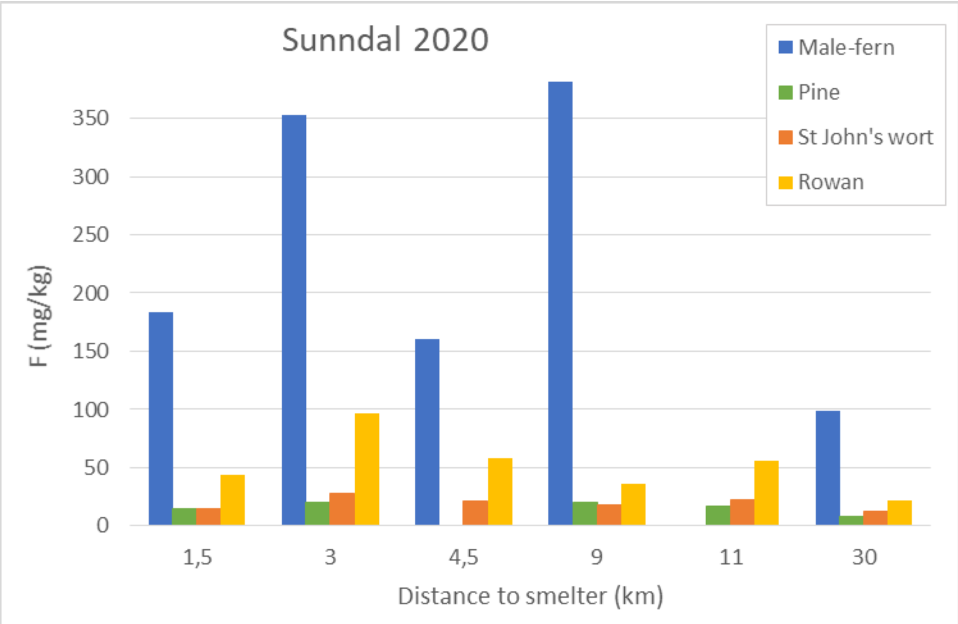


Figure 3.4. **Mean F-concentrations in different plant species (male-fern, pine, St. John's wort and rowan) sampled at sites around Hydro Sunndal smelter: Skjølland (1.5 km), Lensmannsøra (3 km), Einangen (4.5 km), Grøa (9 km), Hoås (11 km) and Gravem (30 km).**

3.5 Alcoa Mosjøen vegetation survey

3.5.1 F-concentrations in sampled plant species

During 2020 we collected and analysed 54 samples.

Rowan sampled at the location closest to the smelter showed typical F-damage symptoms, whereas the other sampled species did not show any typical F-damage symptoms at any of the sampled locations. Rowan was the only species present at all 6 locations, whereas we did not find St. John's wort at any location, and ferns and pine only at 4 and 5 locations, respectively.

All plants sampled around Mosjøen smelter had consistently mean F-concentrations below or slightly above the background level of 10 mg F/kg. Rowan had the highest F-concentrations at all locations, with the highest value (29 mg/kg F) at the closest location to the smelter (Table 3.8).

When accumulation of F in rowan was set as 100% at each locality and compared to other species, male-fern showed 57-77%, pine 27-62% and spruce 27-69% of the F-level detected in rowan (Table 3.8).

Table 3.8. Mosjøen 2020: Mean F-concentrations detected in analysed species. Relative F-concentrations in leaves of analysed plants compared with rowan (100%). Sampling locations (distance to smelter), sampled species and F-concentration in plant tissue per species (mean values if more than one sample per species, n=number of samples). Total number of samples 54.

Location	Species/Genus	n	Mean F (mg/kg)	% of rowan
Kjerringlia (0.75 km)	Rowan	1	28.7	100
	Spruce	3	19.9	69
	Pine	5	17.8	62
	Male-fern	2	16.5	57
Marsøra (0.85 km)	Rowan	2	22.2	100
	Lady-fern	2	15	68
	Male-fern	1	13.9	63
	Small ferns	2	13.8	62
	Spruce	3	7.7	35
	Pine	3	6	27
Haug (2 km)	Rowan	2	15	100
	Male-fern	2	11.6	77
	Small ferns	2	5.2	35
	Spruce	2	4.1	27
Finnmyra (3 km)	Rowan	2	5.4	100
	Small ferns	1	5.7	106
	Pine	2	3.1	57
	Spruce	2	2.3	43
Hestremvegen (4 km)	Rowan	2	13.2	100
	Pine	3	5.7	43
	Spruce	2	4.2	32
Holandstunnelen (10 km)	Rowan	1	10	100
	Fireweed	2	9.3	93
	Lady-fern	1	6.1	61
	Spruce	2	5.3	53
	Pine	2	3.4	34

3.5.2 Effect of distance on F-concentration in vegetation

In general, the F-concentrations in vegetation around Mosjøen were low, and decreasing with increasing distance to the smelter (Table 3.8 and Figure 3.5).

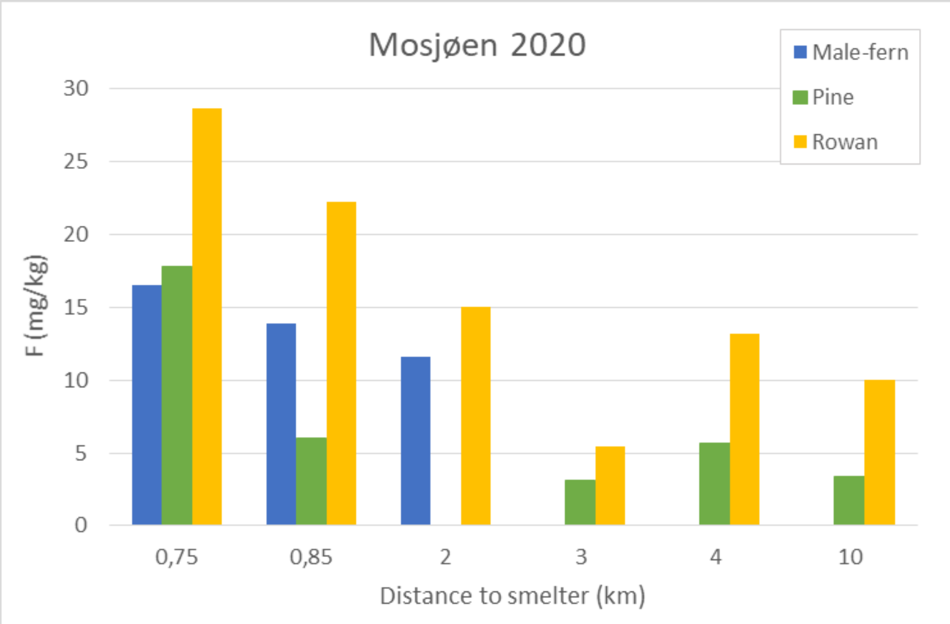


Figure 3.5. Mean F-concentrations in different plant species (male-fern, pine and rowan) sampled at sites around Alcoa Mosjøen smelter: Kjerringlia (0.75 km), Marsøra (0.85 km), Haug (2 km), Finnmyra (3 km), Hestremvegen (4 km) and Holandstunellen (10 km).

3.6 Accumulation of F in pine needles of different ages

In ESPIAL, pine was the only non-deciduous species systematically sampled around each smelter. We found usually three needle age groups; current-year needles (0), one-year-old needles (1) and two-year-old needles (2). The needle age groups differ in their exposure history to F-emissions. It is known that the oldest needles usually accumulate the highest F-concentrations (Horntvedt & Øyen, 1994). To distinguish the F-accumulation levels in pine needles of different ages, we analysed the needle age groups separately.

Around all three smelters, we found a consistent trend of lowest F-concentrations in the current-year needles, followed by one-year-old needles and finally in two-year-old needles (Figure 3.6), confirming the observation described by Horntvedt & Øyen (1994).

However, while at Årdal the mean F-concentrations in the 2-year-old needles were high, they were lower at Sunndal and mostly close to the background level (10 mg F/kg) at Mosjøen (Figure 3.6). At Årdal the pine needles sampled at Moa (0.72 km from the smelter) had the highest F-concentrations with 351 mg F/kg in 2-year-old needles, 177 mg F/kg in one-year-old needles and 55 mg F/kg in current-year needles (Figure 3.6).

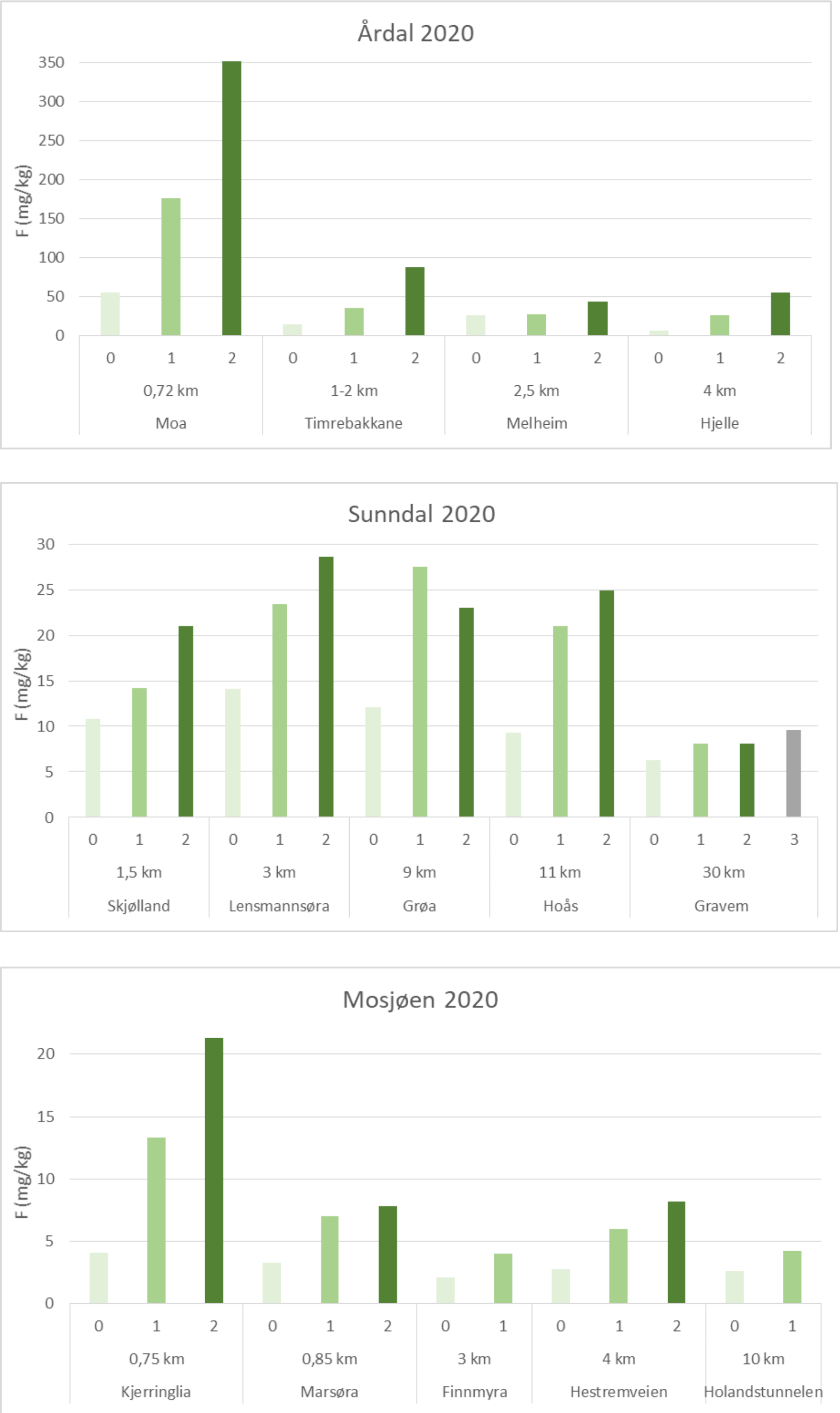


Figure 3.6. **Mean F-concentrations in pine needles of different ages at Hydro Årdal (upper panel), Hydro Sunndal (middle panel) and Alcoa Mosjøen (lower panel) smelters, sampled at different distances from smelters during 2020. Pine needles "0" are current-year needles, "1" are one-year-old needles and "2" are two-year-old needles.**

3.7 F-concentrations in plants with typical symptoms compared to plants without symptoms

In 2019 we sampled several plants with signature F-symptoms as well as plants without any symptoms, from the same location. Because we often found elevated F-concentrations in non-symptomatic plants, we decided to systematically sample both symptomatic and non-symptomatic plants in 2020 for all four indicator species (rowan, male-fern, pine and St. John's wort).

Vegetation at the Årdal, Sunndal and Mosjøen sites did not display eye-catching F-damage symptoms, but at closer examinations we recognised typical symptoms on several plant species. The most visible signature symptoms were on rowan and ferns, especially male-fern (Figure 3.1), which also had the highest F-concentrations in Årdal and Sunndal. In Mosjøen, on the other hand, the symptoms on rowan were much weaker and also the F-concentrations in all sampled plants were below 30 mg F/kg, and often even below the background level (10 mg F/kg, Table 3.8).

We found small, mostly non-significant differences in mean F-concentrations in plants with signature symptoms compared to those without symptoms (Figure 3.7). Male-fern and rowan had the highest mean F-concentrations in both symptomatic and non-symptomatic plants (Figure 3.7).

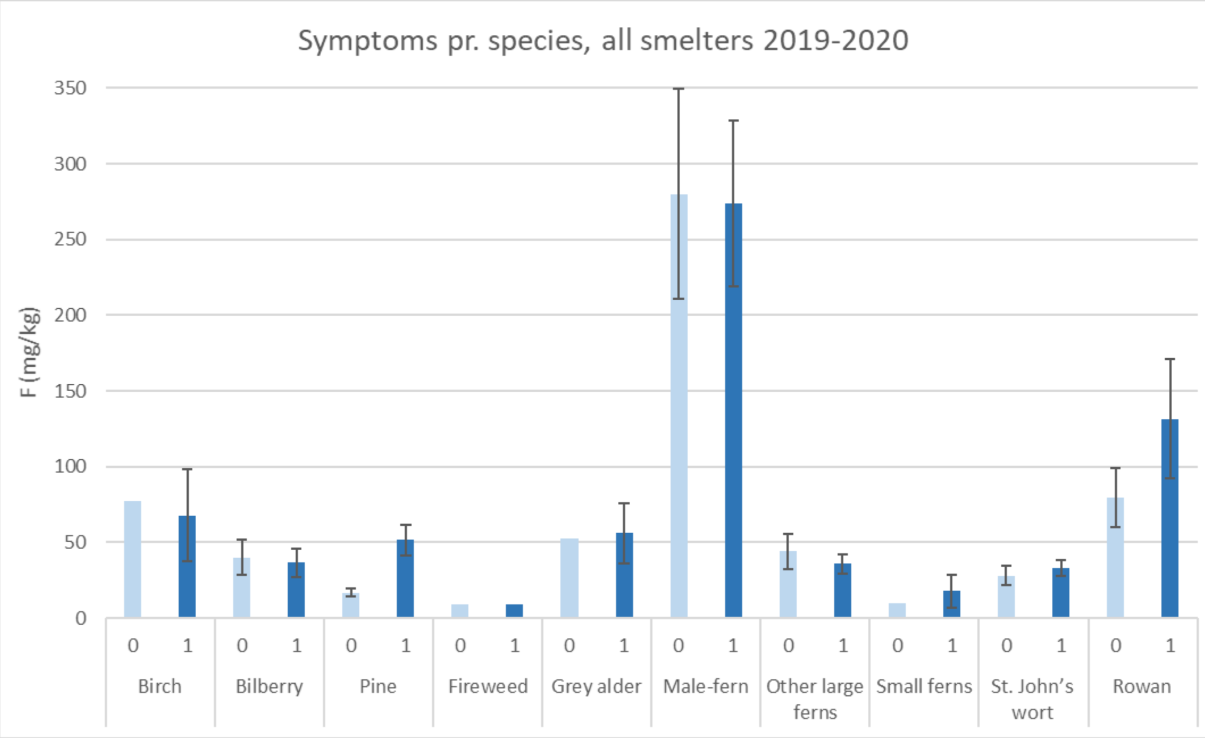


Figure 3.7. Mean F-concentrations in symptomatic vs. non-symptomatic plants sampled at all smelters (Årdal, Sunndal and Mosjøen) during 2019-2020. Symptomatic plants, marked as "1", indicate plants with typical visual symptoms of F-damage. Non-symptomatic plants, marked as "0", refer to plants without any symptoms. Whiskers indicate the standard error of the mean, in cases where it could be calculated.

3.8 Historical data sampled since 1995

At several smelters (Mosjøen, Lista, Høyanger, Karmøy, Sunndal and Årdal) vegetation has been sampled for analyses of F-concentrations since 1995. Each smelter used their own system concerning sampling frequency, plant species and data storage (Word, Excel or pdf-files). Thus, the quality of inputs was very variable. Also, some key information was missing as well as the description of the sampling procedures. To make the data useful for further analyses they needed unifying.

To compile the historical data in a format that is easily accessible and updatable we used the original Excel-format. We systemised and streamlined the data into a new Excel sheet, currently with 5.404 records. This will provide a blueprint and a template for future inputs. The unification of field names and variables will make it possible to use the historical, current and future data for analyses by using, for example, pivot tables or diagrams in Excel.

The historical data are based on sampling of mostly grass, pine, spruce, birch, rowan and salad. Of all input records (n=5.404) only about 3% exceeded concentrations of 100 mg F/kg and 0.7% exceed the F-concentration of 200 mg F/kg. The highest values of F-concentration in vegetation were detected mostly around Årdal and Sunndal smelters.

Of the four species that we mainly sampled during ESPIAL, pine was sampled regularly in Sunndal since 1967. However, in the current database the accessible data in Sunndal for rowan and pine start from 2012 and 2014, respectively (Figure 3.8). Our mean F-concentrations for pine and rowan sampled in Sunndal during ESPIAL study are in line with concentrations detected in previous years (Figure 3.8). In Årdal only grass and in Mosjøen only spruce was sampled regularly.

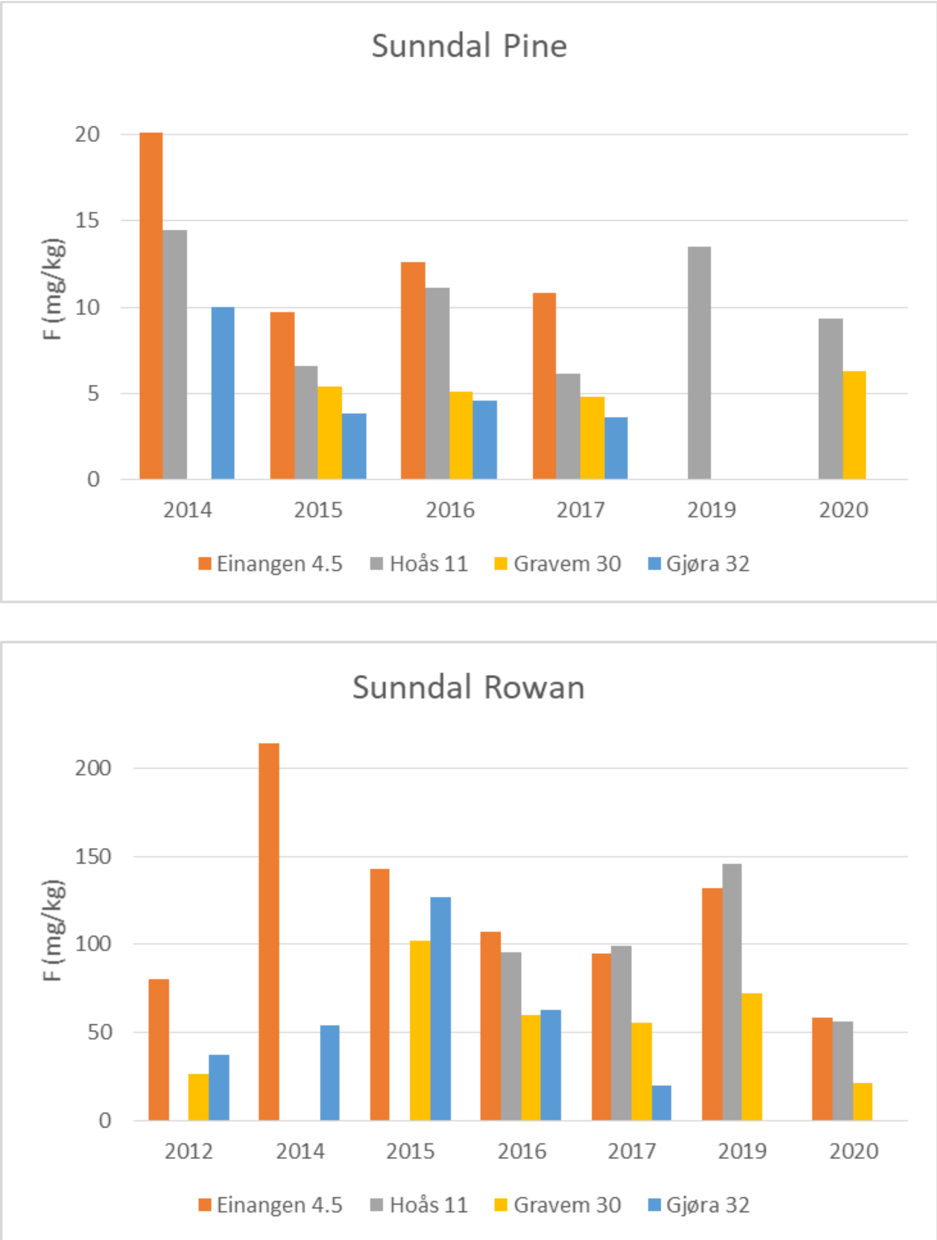


Figure 3.8. Available historical data compared to the ESPIAL sampling. Mean F-concentrations in pine (upper panel) and rowan (lower panel) sampled at sites around Hydro Sunndal smelter recorded since 2014 and 2012, respectively. Pine needles in 2019 and 2020 are current-year needles, for other years the needle age is not specified.

3.9 F-uptake in rowan; comparison between ESPIAL and the Effect Study

In the Effect Study, Horntvedt & Øyen (1994) showed a clear linear relationship between the F-concentrations in leaves and exposure to ambient fluoride over time (Figure 3.9).

With the assumption that plants are passive accumulators of fluoride, this relationship may be described by the equation $F_{plant} = K * F_{air} * T$, where F_{plant} is the detected fluoride content in plant tissue, K is the plant-species-specific accumulation coefficient, F_{air} is the concentration of fluoride in air, and T is the time of exposure (Committee on Biological Effects of Atmospheric Pollutants 1971, Horndtvedt & Øyen 1994).

Here we attempted to compare our data relevant for the comparison of the same data from the Effect Study. We superimposed our three directly comparable measurements for F-uptake in rowan (Table 3.9) from the same locations in Årdal (Moa and Hjelle) and Sunndal (Hoås) as used in the study of Horntvedt & Øyen (1994, Figure 3.9).

We found that our three points for F-uptake in rowan, detected almost 30 years later, at the same locations, aligned well with the original linear relationship between F-concentrations in rowan and exposure to ambient F over time, established by Horntvedt & Øyen (1994, Figure 3.9).

Table 3.9. **F-concentrations in rowan (F_{rowan}) detected at Årdal (Moa and Hjelle) and Sunndal (Hoås) during ESPIAL study. Time of exposure (T) is calculated as number of days from the leaf onset; 1 May in Årdal and 15 May in Sunndal.**

Location	Year	T (days)	n	F_{rowan} (mg/kg)
Moa	2020	78	5	150
Hjelle	2020	78	3	67
Hoås	2019	82	1	146

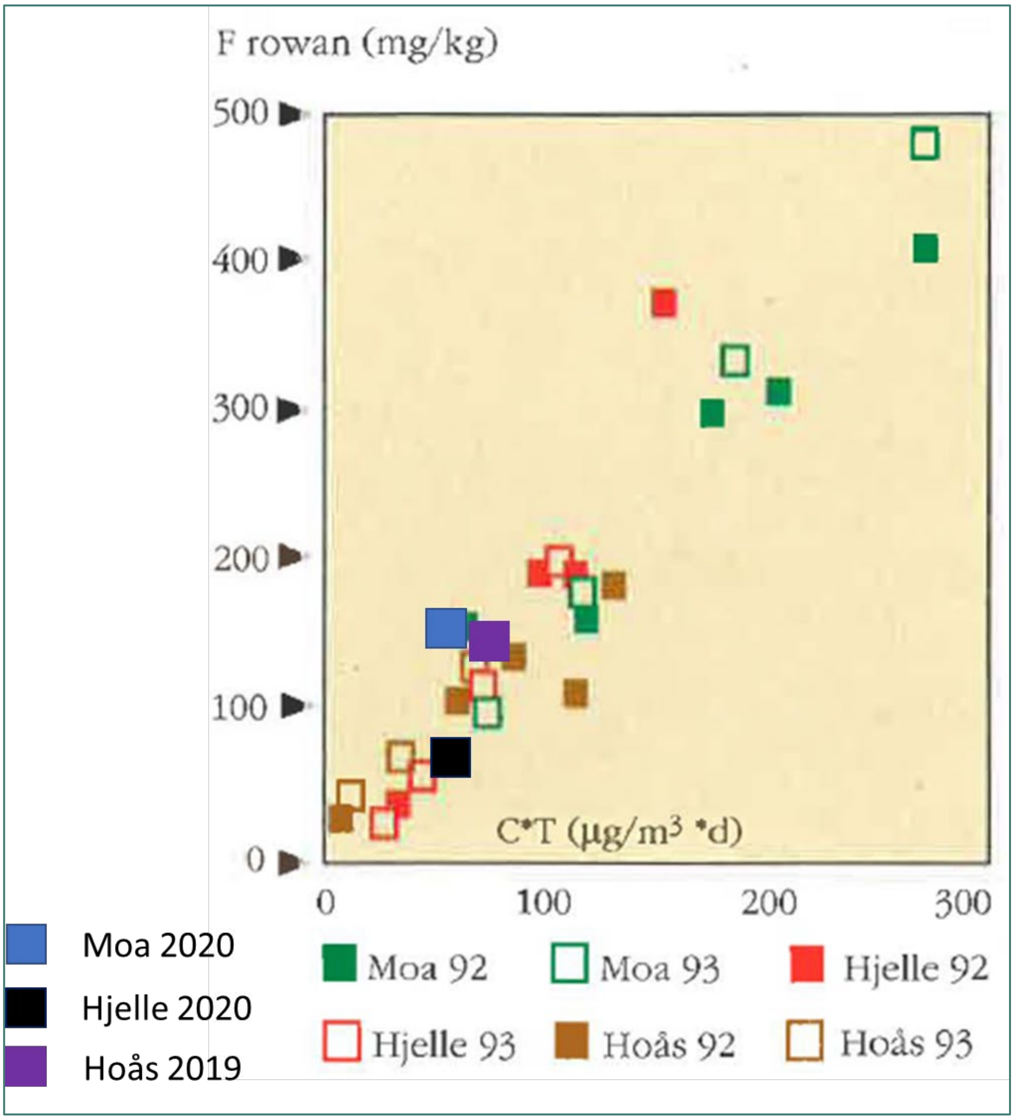


Figure 3.9. **Fluoride uptake in rowan leaves in relation to total fluoride exposure, figure adopted from Horntvedt & Øyen (1994). Exposure is expressed as the product of fluoride concentration in the atmosphere (C, µg/m³) and exposure time (T, days). Points (far left) from Moa and Hjelle (2020) and Hoås (2019) are added as results from ESPIAL study.**

4 Discussion

In the 1960's Robak (1969) reported on massive damage on vegetation over large areas around Norwegian aluminium smelters. About 30 years later, during the Effect Study, Horntvedt & Øyen (1994) reported that the F-damage on vegetation around Norwegian aluminium smelters had improved significantly although they still recommended reductions in emissions in Årdal and Sunndal, while the situation was acceptable in Mosjøen. In our ESPIAL report, almost 30 years after the Effect study, F-damage symptoms on vegetation, although not immediately obvious, were still present especially around Årdal and Sunndal. In Mosjøen F-symptoms were sparse. The symptoms were particularly evident on the following four plant species: Rowan, pine, St. John's wort and male fern.

On rowan we found two types of symptoms, the well-known "edge"-symptom and the lesser known "tip"-symptom (Figure 3.1, Appendix 1). While "edge"-symptom is widely described in the literature as characteristic of F-damage on rowan leaves, the "tip"-symptom is not mentioned as typical of F-damage. To our knowledge, only Romøren (1973) described the "tip"-symptom in relation with F-damage on rowan. We found high concentrations of F in plants with both types of symptoms, thus confirming Romøren's (1973) observation.

By far the highest concentration of F in all our sampled material (1161 mg/kg F) was detected in a group of declining rowan trees sampled inside the Årdal Hydro smelter area (an "ad hoc" location). These trees were growing in the immediate vicinity of the potline vents, thus directly exposed to the fugitive F-emissions. All rowans at this place were severely declining, with defoliated, dead branches and the remaining leaves had consistent "tip"-symptoms (Appendix 1), this strengthening the association of this symptom to F-damage. We did not find rowan trees with similar signs of severe decline on any other site or locality during the ESPIAL study.

In our sampling campaigns rowan showed clear and consistent signs of F-damage and thus may be suitable as a bioindicator. In addition, rowan was selected in the Effect Study (Horntvedt & Øyen, 1994) as a good biomonitor of airborne fluoride emission as it accumulated F to a great extent, had smaller individual variation in F-content than other species and was widespread on all locations. They therefore used rowan as a biomonitor to identify the fluoride dispersion pattern and to establish a new damage scale (Table 1.3).

In pines around both smelters, we confirmed that F is accumulating with increasing age of the needles. One- or two-year-old pine needles accumulated always higher levels of F than current-year needles. This pattern was consistent around all three smelters and confirms that the longer the tissue is exposed to emissions, the more F it accumulates. This tendency is known also from other non-deciduous trees such as eucalyptus, neem-tree, palms etc., where F gradually accumulates and its concentrations increase in older tissues (Weinstein & Davison, 2003).

In addition, Horntvedt & Øyen (1994) described that F-concentrations in pine needles monitored from 1967 to 1992 showed a close connection between F-emissions and F-levels in needles at Sunndal. Also, they mention that the gaseous F is of greater importance for uptake in pine needles than particulate F. This may be due to the small and smooth surface of pine needles as compared to larger leaves with rough surfaces in other species. The qualities of pine; such as clear presence of F-symptoms and consistent F-accumulation, suggest that also pine is suitable as a good bioindicator as well as biomonitor.

St. John's wort is known to be a sensitive bioindicator (Horntvedt & Øyen 1994). In ESPIAL it showed consistent presence of early F-symptoms, but only slightly elevated concentrations of F in the tissue were detected. It may therefore be considered a sensitive bioindicator that reacts with appearance of F-damage symptoms quickly and consistently. However, as the F-uptake does not seem to increase proportionally with the exposure duration and F-concentration in ambient air, it does not have the quality of a good biomonitor.

Several species of large ferns (male-fern, lady-fern and eagle-fern) had typical F-damage symptoms but only male-fern had extremely high concentrations of F both in Årdal and Sunndal, up to 10 times more than those in rowan (Table 3.7). Also, Horntvedt & Øyen (1994) mentioned the unusually high F-concentrations in male-ferns from the same sites in Årdal and Sunndal, 30 years earlier. They found that male-fern accumulated up to 3-4 times the F-concentrations found in rowan and the F-accumulation was also high in reference areas, with no fluoride deposition. They concluded that this abnormally high F-accumulation in male-fern must originate from soil uptake. In our study male-fern sampled from Mosjøen locations, with mostly low F-concentrations (below 30 mgF/kg) in all sampled vegetation, had low F-concentrations (from 12-17 mg F/kg). We showed also that the F-concentration in male-fern decreased with the distance from the smelters in Årdal and Sunndal, probably reflecting lower F-content in soil with distance. Although our current findings confirm the anomaly in male-fern F-uptake described by Horntvedt & Øyen (1994), the source, cause and mechanisms of this extreme uptake remain unsolved.

In addition to the extreme F-concentrations in male-fern, we observed grazing damage on male-fern (but also on lady-fern, Appendix 3) and rowan at all three smelters (Appendix 3). Because male-fern and rowan had the highest F-concentrations at polluted areas, their consumption may pose a health risk to grazing animals. According to Turid Vikøren (pers. comm.) grazing on male-fern is quantitatively of low importance. It is mentioned that moose may be grazing on ferns. Although male-ferns may also occasionally be a part of a deer diet, it is not established how quantitatively important part of their diet male-fern actually is. During the spring/summer the deer mostly feed on grass, but also on birch, rowan and goat willow, which also showed high concentrations at Årdal and Sunndal, especially close to the smelters. None of these plants, however, reached the extreme levels detected in male-ferns. If proven that it is a significant part of grazing animals diet, male-fern may be considered worth monitoring because of its potential health hazard for animals.

The F-damage symptoms on ferns were very similar to fern autumn colouring. However, the autumnal discoloration appears later in season and may often be spotty and discontinuous as opposed to F-damage appearing early in the summer and with necroses evenly distributed along the edges (not spotty).

We detected higher F-concentrations in vegetation closer to smelters, decreasing with the distance in line with results of Horntvedt & Øyen (1994). This may partly be due to the presence of particulate F, which is most abundant near the smelters and declines with the distance as the particles are heavier and do not spread far. Because our samples were not washed before analysis, a substantial portion of detected F may have been of the particulate F origin, especially at locations close to the smelter. Also, the leaf morphology may be decisive regarding the capturing (and washing off) of particulate F; large, hairy and rough leaves (such as those of rowan and ferns) may capture more F than, for example, thin and smooth needles of pine.

We documented the variation of F-concentration with altitude in Årdal (Figure 3.3). Similar concentrations were found from 100 - 400 m.a.s.l., but the concentrations dropped at higher elevations (400 - 760 m.a.s.l.). A possible explanation, in addition to local meteorological conditions, may be that trees come into leaf later at higher altitudes and thus have a shorter accumulation time as suggested by Horntvedt & Øyen (1994).

Our results showed that non-symptomatic plants had similar concentrations of F as those with symptoms at all sites around the smelters (Figure 3.7). This may be due to delayed development of the symptoms in some plants, plant-specific differences in genetic traits, and local differences in habitat characteristics and air currents. Therefore, although the presence of typical visual symptoms is a good guideline for sampling and evaluation, the lack of symptoms does not necessarily indicate low levels of F in plant tissues.

Historical data gathered since 1995 showed mostly moderate F-concentrations in different sampled plant species with only few samples exceeding 100 mg F/kg. Most of the elevated F-concentrations were from Årdal and Sunndal. Because different plant species were sampled around the smelters, there are limited possibilities for comparison. Our current data could only be compared to data from Sunndal where pine and rowan were sampled since 2012 (Figure 3.8).

During our sampling campaign in Sunndal 2019 it was raining heavily and all plants that we sampled probably had a great part of the particulate F on their leaf surface washed off. Vike (2005) showed that up to 50% of the particulate F on leaf surface could be washed off by rain. However, our results from the 2019-campaign in Sunndal did not show a decrease in detected F-concentrations in rowan or pine when compared to historical data (Figure 3.8).

While the F-concentrations in vegetation around Årdal and Sunndal smelters often were high (especially in male-fern and rowan), we found only slightly elevated F-concentrations (always below 30 mg/kg F) around Mosjøen. These findings may be due to topographical differences; while Årdal and Sunndal are located in deep, narrow valleys with prevailing wind direction from the sea, Mosjøen is located in a relatively open valley with presumably more air circulation. Also, most of locations sampled during 2020 in Mosjøen were upwind from the smelter, in contrast to locations such as Skogåsen, 3-4 km south of the smelter, where higher F-levels were detected in 1992-1993, during the Effect Study. Furthermore, reported emissions of total F from Årdal and Sunndal were about 100 t/yr each year during 2019-2020, while in Mosjøen it was about 60 t/yr in 2020 (Figure 1.4), resulting in lower detected F-concentrations in vegetation here. In Sunndal we found elevated levels of F in plants growing as far as 30 km from the smelter (Gravem). In Årdal the highest levels seemed to be in the vicinity of the smelter (Moa, within 1 km), while at 4 km distance (Hjelle) the concentrations decreased considerably. It seems that the particular site topography in Sunndal and Årdal has the largest contribution to this emission distribution pattern. In Sunndal the long, U-shaped valley functions as a chimney, able to carry the emissions much further than in the case of Årdal, where the valley branches into four other valleys, thus dispersing the emissions.

Foliar F content is generally determined by the F-concentration in the air, exposure time and the absorption capacity of the species in question, expressed by formula $F_{plant}=K*F_{air}*T$ (Committee on Biological Effects of Atmospheric Pollutants, 1971). In rowan, Horntvedt & Øyen (1994) found a high degree of agreement with the formula at two localities in Årdal and one in Sunndal (Figure 3.9). Their good fit indicates that the environmental conditions were similar at these sites, or that the uptake by rowan is not particularly sensitive to variations in environmental conditions and has a steady accumulation pattern (Horntvedt & Øyen, 1994). Our samples from the same species and localities, aligned well with the data by Horntvedt & Øyen (1994) and followed the same linear tendency (Figure 3.9).

In the centre of Sunndalsøra, NILU detected ambient air F-concentrations of 0.8 µg F/m^3, while in the more distant Hoås (Vennevold, 11 km from the smelter) the F-concentration was less than a half, 0.3 µg F/m^3 (Hak 2020, measurements from May - August 2019). In Årdal there were installed passive F-samplers at Moa and Hjelle from November 2019 - June 2020. In Moa (about 0.72 km from the smelter) the ambient F-concentration was 0.7 µg F/m^3, while in Hjelle (4 km from the smelter) it decreased to 0.1 µg F/m^3, a value at the detection limit (Hak, pers. comm). The recommended threshold values causing damage on sensitive vegetation are 0.3-0.5 µg F/m^3 (according to SFT and Garrec, see Table 1.2) and 0.2-0.6 µg F/m^3 (according to Horntvedt & Øyen, 1994, see Table 1.3). Based on this, the F-concentrations in ambient air in Sunndalsøra centre and in Årdal (Moa) exceeded the recommended F-damage thresholds potentially causing F-damage on more species than only sensitive ones (Table 1.3). At Hoås the F-concentrations were within the SFT-limit, but exceeded slightly the limit set by Horntvedt & Øyen (1994) suggesting a potential F-damage on sensitive species. In Hjelle, the F-concentrations were below the recommended F-concentration limits. The detected ambient air F-concentrations agreed with the F-concentrations detected in vegetation and presence of F-damage symptoms at these locations (Hoås in Sunndal, Moa and Hjelle in Årdal) during the ESPIAL study.

The annual F-emissions in Årdal were 12 and 11 kg F/hour in 2019 and 2020, respectively, while during the Effect Study in 1992 they were considerably higher, 18 kg F/hour. In Sunndal the F-emissions were 12 and 9 kg F/hour in 2019 and 2020, respectively, while in 1992 they were only slightly higher, 13 kg F/hour. In Mosjøen the F-emissions were 7 kg F/hour in 2020, while in 1992 they were only slightly higher, 8 kg F/hour (data provided by individual smelters, Figure 1.4).

In the Effect Study, Horntvedt & Øyen (1994) conclude that with emissions less than 10 kg F/hour the risk of damage to coniferous forest would be very small (case of Mosjøen). Exceptions are areas in the immediate vicinity of the smelters and extremely unfavourable dispersion conditions. Emission levels of 10-20 kg F/hour would cause some damage, limited in open areas, but amplified in narrow valleys (cases of Årdal and Sunndal). No emissions exceeding 20 kg F/hour, capable of causing significant damage, were reported from any of the three smelters. On basis of this evaluation, reductions in emissions are still advisable in Årdal and Sunndal, while the situation is acceptable in Mosjøen.

As the measured ambient F-concentrations are exceeded in Årdal and Sunndal, particularly in the immediate vicinity of the smelters, some F-damage on vegetation is to be expected, as we demonstrated in the ESPIAL study. With the current emission level, the risk of extensive damage to coniferous forest as described by Robak (1969) would be low. However, the constant pollution may also pose a constant “background” stress that may not be directly expressed as symptoms, but may be predisposing the vegetation to other damaging factors such as frost, drought, insects or fungi.

5 Recommendations

Based on our results from samplings around Årdal, Sunndal (2019-2020) and Mosjøen (2020) smelters we recommend some adjustments to the current procedures. Our comments are mainly related to the choice of locations, selection of species, sampling procedure and data processing.

General recommendations for all smelters:

- Species: In addition to plant samples taken regularly at each smelter, we recommend to include samples of rowan and male-fern (in case male-fern is proven to be a significant part of animal diet). Both species accumulate high levels of F, are easily found at all locations, and are grazed by deer (Appendix 3), which may potentially be harmful for the animals. Rowan has qualities of a good bioindicator and biomonitor. If possible, we also recommend including sampling of pine and St. John's wort. Pine needles are particularly suitable for tracking F over several years since this tissue accumulates F-emissions usually up to 3 years after exposure. St. John's wort is suitable since it is a sensitive species and shows clear, early symptoms of elevated F-levels. All of these four plants show visible, signature symptoms of elevated F-concentrations, that may be readily recognized by the trained eye.

- Identification of symptoms: Use the photo guide in the Appendix (Appendix 1, Appendix 2) for identification of typical F-damage symptoms on the selected species.

- Sampling:

    - Use the same locations for sampling every year. Geo-referencing is important to calculate distance to the smelter, take precise GPS coordinates.

    - Sampling of foliage from the same trees is recommended. In this case, trees should be marked. When sampling, use the side of the crown that faces the emission source.

    - At each location, sampling of two to four species, preferably with visible F-damage signs is recommended. Sample the same species at all locations.

    - Sample material from several individuals (3-4 individuals) of the same species and pool the sample before chemical analysis.

    - Sample sufficient amounts of plant material for the chemical analysis. Usually about 20 g of dry matter is needed for the analysis, so about 200 g of fresh plant material should be sufficient.

    - Sample whole leaves in ferns and rowan, stems with leaves in St. John's wort and needles in pine. Sample leaves og the same age (avoid sampling the youngest leaves).

    - If no symptomatic plants are found, also plants of the same species without visible symptoms may be sampled since they usually have similar concentrations as the plants with symptoms. However, sampling of plants with visual symptoms is preferred.

    - In cases where it is of interest to monitor F-accumulations over years the different needle-ages can be separated by cutting the twigs into age-groups and gathering the sitting needles in separate bags for each needle age-group for further analysis.

- Sampling frequency: We recommend sampling once a year.

- Sampling timepoint: Sampling should be done at the end of the growing season (late summer), but before the onset of autumn signs. This timepoint may be variable among the years and sites, but sampling during autumn should be avoided in order to avoid confusion of the F-damage symptoms with the senescence signs.

- Chemical analysis procedure: We recommend the use of “round robin test” and use of the same protocol among the smelters to harmonise the analyses.

- Results: Import the data into relevant columns in the Excel sheet of the common “Database” to enable easy access and processing/analyses of the results. Missing information in the “Database”-Excel sheet, such as the geographical coordinates for each site may be included to enable more exact evaluations of trends based on both historical and current data. Use the “Database”-Excel sheet as a common deposit of all future data.

Hydro Årdal:

- Locations: No change necessary. The location selection is sufficient and covers sites with high F-concentrations nearby the smelter and low F-levels at only 4 km distance (Hjelle) from the smelter and includes an elevation gradient as well. Vegetation (grass) at these locations has been sampled since 2004, and the gathered data have a historical value, mirroring the emission development around the smelter. We recommend further use and follow-up of all current locations.

Hydro Sunndal:

- Locations: We recommend further use of the current locations and additions of one or two locations nearby the smelter to establish an emission gradient, preferably sites where the recommended plant species may be found, for example the two additional locations that we used in 2020. Otherwise, the location selection is sufficient - with Gravem, 30 km from the Sunndal smelter, as the most distant but still showing elevated F-concentrations.

Alcoa Mosjøen:

- Locations: Select locations at different distances from the smelter. The locations that we sampled in 2020 were selected with the priority on presence of the species sampled and on the pollution gradient. However, the pollution gradient was not as evident as in the Effect Study-locations. Therefore add/use locations following the prevailing wind direction from the smelter, so that an emission gradient may be established.

6 Conclusions

- At Årdal and Sunndal we consistently found higher F-values in vegetation than in Mosjøen. While in Årdal and Sunndal the F-concentrations in male-fern, rowan, St. John's wort and pine were often far above the background level (10 mg F/kg), in Mosjøen they were low and never exceeded 30 mg F/kg. These variances may be due to much lower production and emission levels in Mosjøen but also differences in topography and predominant wind directions. Whereas both Årdal and Sunndal are located in deep, narrow valleys, Mosjøen is located in far more open terrain.
- We found that in Årdal and Sunndal the most useful plant indicators of F-accumulation were rowan, pine and St. John's wort. Rowan and pine showed clear F-symptoms (bioindicator), accumulated F in a consistent way (biomonitor) and were thus suitable both as bioindicator and biomonitor species. St. John's wort was suitable as an early bioindicator as it showed F-damage at low F-emission levels.
- Pine was the only non-deciduous species we sampled in 2020 and we always detected the highest F-concentrations in the oldest needles with the longest exposure to F-emissions.
- Highest F-concentrations were detected in rowan and male-fern growing in the vicinity of Årdal and Sunndal smelters. However, male-fern showed consistently extremely high F-concentrations sometimes up to 10 times higher than those in rowan at the same location.
- We noticed considerable amount of deer grazing damage on rowan, male-fern and lady-fern around all three smelters. Because rowan and male-fern also contained the highest F-concentrations at Årdal and Sunndal, grazing may potentially have negative health consequences for grazing animals.
- At all smelters there was a clear gradient of decreasing F-concentration in vegetation with increasing distance from the smelter.
- At Årdal we found lower F-concentrations in vegetation with increasing elevations. At elevations of about 760 m.a.s.l. the F-concentrations were close to background levels.
- In 2020 we sampled both symptomatic and non-symptomatic plants from the same locations and found that these two groups had similar F-concentrations. The appearance of symptoms may have been delayed, although the F-emissions were already absorbed. The presence of symptoms is a good visual sampling guide and preferably symptomatic plants should be sampled, but lack of symptoms does not necessarily indicate low levels of F in plant tissues.
- The historical data recorded since 1995 show moderate F-concentrations in sampled vegetation and only about 3% of the 5404 analysed samples had F-concentrations exceeding 100 mg F/kg. F-concentrations in plant tissue were highest around Årdal and Sunndal smelters.
- Our results indicate that a targeted sampling of vegetation with symptoms may provide a good tool for monitoring of the emission load in the vicinity of smelters.
- Although the situation at smelters has somewhat improved regarding the fluoride emissions during the Effect Study (1994), the recommended air quality criteria were still slightly exceeded during the ESPIAL study in Årdal and Sunndal. In Mosjøen the situation was acceptable.

7 Acknowledgements

We thank Aluminiumindustriens Miljødirektoratet (AMS) for funding of the study. We are especially grateful to Leif Ongstad, Hanne Hoel Pedersen, Jan Olav Polden, My Nhung Tran, Asbjørn Kjønås, Bjørn Tore Bergsnev, Ingvill Eldegard, Magne Edgar Vee, Knut Fredrik Øi, Turid Vikøren, Claudia Hak and Frank N. Kidder for their invaluable help in planning the work and providing an excellent support, advice and feedback during the ESPIAL survey.

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Appendix

Appendix 1 – Fluoride damage

Fluoride damage	Detected F-concentration (mg F/kg) and symptom description
	Rowan at Årdal smelter area Detected F-concentration: 1161 mg F/kg Location: Årdal smelter area Symptoms: All four trees had declining crowns and leaves with "tip"-symptoms: Necrotic, red-brown leaf tips with sharp, burgundy-coloured line separating the dead and living tissue.
	Rowan from Årdal smelter area Detected F-concentration: 1161 mg F/kg Location: Årdal smelter area Symptoms: "Tip"-symptom on rowan: Necrotic, red-brown leaf tips with sharp, burgundy-coloured line separating the dead and living tissue.

	Rowan – tip symptom Detected F-concentration: 170 mg F/kg Location: Årdal, Moa Symptoms: “Tip”-symptom in rowan: Necrotic, red-brown leaf tips with sharp, burgundy-coloured line separating the dead and living tissue.
	Rowan – tip symptom Detected F-concentration: 98 mg F/kg Location: Årdal, Hjelle Symptoms: Example of “tip”-symptoms in rowan: Necrotic, red-brown leaf tips with sharp, burgundy-coloured line separating the dead and living tissue. Notice the yellow region in front of the dead tissue, where chlorophyll becomes degraded.
	Rowan – no symptoms Detected F-concentration: 277 mg F/kg Location: Årdal, Moa Symptoms: Example of rowan leaves without visible symptoms, but high F-concentration

Rowan – edge symptom
Detected F-concentration: 146 F/kg
Location: Årdal, Moa
Symptoms: Typical “edge”-symptom in rowan: Necrotic, brown leaflet edges with sharp, dark-coloured line separating the dead and living tissue.

Rowan – edge symptom
Detected F-concentration: 190 mg F/kg
Location: Årdal, Moa
Symptoms: Typical “edge” -symptom at rowan: Necrotic, red-brown leaflet edges with sharp, dark-coloured line separating the dead and living tissue.

	Male-fern Detected F-concentration: 416 mg F/kg Location: Årdal, Moa Symptoms: Necrotic, red-brown leaflet edges with sharp, dark-coloured line separating the dead and living tissue.
	Male-fern Location: Årdal, Moa Symptoms: Symptoms on male fern are easily visible from a distance: Necrotic, red-brown leaflet edges with sharp, dark-coloured line separating the dead and living tissue.

Male-fern, detail
Detected F-concentration: 925 mg F/kg
Location: Årdal, Moa
Symptoms: Necrotic, red-brown leaflet edges with sharp, dark-coloured line separating the dead and living tissue.

St. John's wort
Detected F-concentration: 41 mg F/kg
Location: Årdal, Hjelle
Symptoms: Necrotic, red-brown leaf tip with sharp, burgundy-coloured line separating the dead and living tissue.

St. John's wort
Detected F-concentration: 49 mg F/kg
Location: Årdal, Melheim
Symptoms: Necrotic, red-brown leaf tips with sharp, burgundy-coloured line separating the dead and living tissue.

Pine
Detected F-concentration:
Current-year needles: 15 mg F/kg
1-yr old needles: 26 mg F/kg
2-yr old needles: 34 mg F/kg
Location: Årdal, Hjelle
Symptoms: Brown, dead needle tips with sharp, dark lines separating the dead tissue from the living.

Pine
Detected F-concentration:
Current-year needles: 42 mg F/kg
1-yr old needles: 147 mg F/kg
2-yr old needles: 243 mg F/kg
Location: Årdal, Moa
Symptoms: Light brown, dead needle tips with sharp, dark line separating the dead tissue from the living. Notice the burgundy and yellow area in front of the sharp line where the chlorophyll becomes degraded and tissue dies.

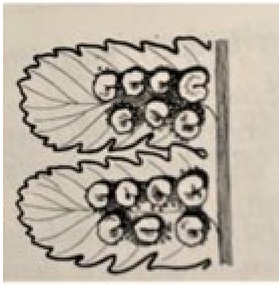
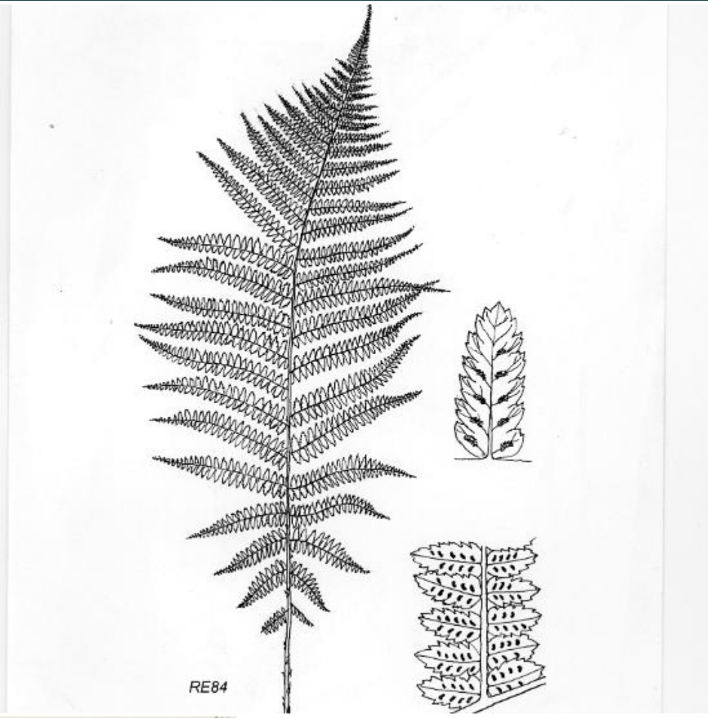
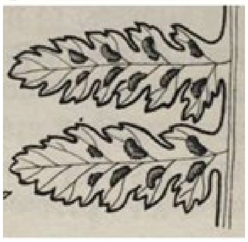
Goat willow
Detected F-concentration: 20 mg F/kg
Location: Årdal, Hjelle
Symptoms: Yellowing of tissue between the leaf veins (interveinal chloroses) and yellow leaf edges.

Birch
Detected F-concentration: 142 mg F/kg
Location: Årdal, Moa
Symptoms: Yellowing among the leaf veins (interveinal chlorosis).

	Goat willow Detected F-concentration: 58 mg F/kg Location: Årdal, Moa Symptoms: Yellowing among the leaf veins (interveinal chlorosis), yellow leaf edges and sometimes dead leaf tips.
	Elm Detected F-concentration: 41 mg F/kg Location: Sunndal, Einangen Symptoms: Necrotic, red-brown leaflet edges with sharp, dark-coloured line separating the dead and living tissue. Notice the spoon-like curling of leaf edges.
	Bilberry Detected F-concentration: 61 mg F/kg Location: Sunndal, Hoås Symptoms: Necrotic, red-brown leaflet edges sometimes with sharp, dark-coloured line separating the dead and living tissue.

	Aspen Detected F-concentration: 39 mg F/kg Location: Sunndal, Einangen Symptoms: Necrotic, brown leaflet edges with sharp, dark-coloured line separating the dead and living tissue.
	Grey alder Detected F-concentration: 41 mg F/kg Location: Sunndal, Grøa Symptoms: Necrotic, red-brown leaflet edges with sharp, dark-coloured line separating the dead and living tissue. Notice the inward, spoon-like curling of the leaves.
	Rose Not analysed Location: Øvre Årdal, Parking lot in front of the Hydro Årdal smelter entrance Symptoms: Necrotic, red-brown leaf tips with sharp, dark-coloured line separating the dead and living tissue.

Appendix 2 – Male- and lady-fern differences

Male- and lady-fern differences	
 	Male-fern Distinguishing signs: Rounded pinnate leaflets with smooth edge. Rounded spore-houses on the lower side of the leaves. (Detail illustration from Lid & Lid, Norsk flora)
 	Lady-fern Distinguishing signs: Pointed pinnate leaflets with incisions in leaflet edges that give a “lacy” and narrower look as compared to male-fern. Comma-like spore-houses on the lower side of the leaves. (Detail illustration from Lid & Lid, Norsk flora)

	Male-fern Distinguishing signs: Rounded pinnate leaflets with smooth edge.
	Male-fern Distinguishing signs: Rounded pinnate leaflets with smooth edge.
	Male-fern Distinguishing signs: Rounded spore-houses on the underside of the leaflets as opposed to comma-like spore-houses in lady-fern. Rounded pinnate leaflets with smooth edge.

Lady-fern
Pointed pinnate leaflets with incisions in leaflet edges that give a “lacy” and narrower look as compared to male-fern.

Lady-fern
Pointed pinnate leaflets with “cuts” in leaflet edges that give a “lacy” and narrower look as compared to male-fern.

Appendix 3 – Examples of grazing damage

Examples of grazing damage	
	Grazing damage on rowan in Sunndal
	Grazing damage on male-fern in Sunndal
	Grazing damage on lady-fern in Årdal

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