Norsk institutt for luftforskning
Norwegian Institute for Air Research

Air quality assessment of the surroundings of the Hydro Sunndal aluminium smelter

Measurements May – August 2019

Claudia Hak

NILU report 02/2021

NILU report 02/2021	ISBN: 978-82-425-3030-1 ISSN: 2464-3327	CLASSIFICATION: A – Unclassified (open report)
DATE 12.11.2021	SIGNATURE OF RESPONSIBLE PERSON Ole-Anders Braathen, Deputy director (sign.)	NUMBER OF PAGES 49

TITLE	PROJECT LEADER
Air quality assessment of the surroundings of the Hydro Sunndal aluminium smelter Measurements May – August 2019	Claudia Hak
	NILU PROJECT NO. O-119080
AUTHOR(S) Claudia Hak	QUALITY CONTROLLER Paul Hamer
REPORT PREPARED FOR Aluminiumindustriens Miljøsekretariat – AMS	CONTRACT REF. Leif Ongstad

ABSTRACT

On behalf of Aluminiumindustriens Miljøsekretariat (AMS), NILU – Norwegian Institute for Air Research has conducted a sampling campaign in the surroundings of the Hydro Sunndal aluminium smelter in order to update the knowledge on air quality around the smelter today. Samples were taken in summer 2019 and analysed for $PM_{2.5}$, PM_{10}, metals (Pb, Cd, Cr, Ni, As, Al, V, Ga, Sb, Bi), particle-bound PAHs, SO_2, particle-bound and gaseous fluorides. As a consequence of reduced emissions compared to earlier measurements, the ambient concentrations of PM_{10}, Cr, Pb, BaP (for PAHs), SO_2 and fluorides were strongly reduced. All measured compounds had concentrations below limit values and recommended guideline values.

NORWEGIAN TITLE

Vurdering av luftkvalitet i omgivelsene til Hydro Sunndal aluminiumsmelter

KEYWORDS

Air quality | Industrial pollution | Other chemical compounds

ABSTRACT (in Norwegian)

På oppdrag fra Aluminiumindustriens Miljøsekretariat (AMS), har NILU – Norsk institutt for luftforskning gjennomført en prøvetakingskampanje i omgivelsene til aluminiumssmelteverket Hydro Sunndal for å oppdatere kunnskapen om luftkvalitet rundt smelteverket i dag. Prøvene ble tatt sommeren 2019 og analysert for $PM_{2.5}$, PM_{10}, metaller (Pb, Cd, Cr, Ni, As, Al, V, Ga, Sb, Bi), partikkelbundne PAHer, SO_2, partikkelbundne og gassformige fluorider. Som en konsekvens av reduserte utslipp sammenlignet med tidligere målinger, ble konsentrasjonene av PM_{10}, Cr, Pb, BaP (for PAH), SO_2 og fluorider sterkt redusert. Alle målte forbindelser hadde konsentrasjoner under grenseverdier og anbefalte retningslinjer.

PUBLICATION TYPE: Digital document (pdf)	COVER PICTURE: Source: Hydro, press photo

© NILU – Norwegian Institute for Air Research

NILU's ISO Certifications: NS-EN ISO 9001 and NS-EN ISO 14001. NILU's Accreditation: NS-EN ISO/IEC 17025.

Contents

Summary

The effect of aluminium production on the environment surrounding aluminium smelters has been studied over several decades. NILU has studied their effects on air quality both in measurement and modelling studies since the early 1970s. The "Effect Study" in the beginning of the 1990s gave an overview over the effects of aluminium production on vegetation, water, farm and game animals and human health.

ESPIAL (=Ensuring the Environmental Sustainability of production of PrImary ALuminium) is a multidiscipline study initiated and sponsored by "Aluminiumindustriens Miljøsekretariat" (AMS) to update and supplement the Effect Study. This report is a contribution to this update, focusing on the effect on air quality today, by the example of measurements carried out around Hydro Sunndal.

In summer 2019 (23. May – 15. August 2019), weekly samples of SO_2, gaseous fluoride, particle-bound fluoride, 16 priority PAHs (particle-bound), $PM_{2.5}$, PM_{10} and metals in PM_{10} (lead, cadmium, chromium, nickel, arsenic, aluminium, vanadium, gallium, antimony and bismuth) were collected at two sites in the vicinity of the aluminium smelter Hydro Sunndal at the "Pensjonistsenter" and at Vennevold. Pensjonistsenteret is located close to the smelter and in the centre of Sunndalsøra whereas Vennevold is located 6 km up the valley Sunndalen. The measurements were carried out in summer due to the local meteorological and topographical conditions – the built-up area at Sunndalsøra is most affected by emissions from the smelter during summer.

Ambient concentrations of the measured compounds[1] have decreased since the beginning of the 1990s. The concentrations of all measured compounds in summer 2019 were below the respective annual limit values, target values and air quality criteria. As the highest concentrations in Sunndalen are expected in summer, compliance with limit values for summer averages suggests compliance for annual averages.

Particulate matter (PM) was measured as size fractions PM_{10} and $PM_{2.5}$. The PM_{10} average concentration over three months in summer 2019 was 50% of the PM_{10} average in summer 2006. Summer averages of PM_{10} and $PM_{2.5}$ were markedly lower than the respective annual limit values (for calendar year). There was no clear difference between levels at Pensjonistsenteret and levels at Vennevold.

Metals in PM_{10} were analysed, which comprise metals with a target value (Forurensningsforskriften), metals that Hydro Sunndal has an emission permit for, and other metals associated with aluminium production. Concentrations in summer 2019 were lower than in summer 2006 for chromium, lead, vanadium, aluminium. For nickel, arsenic and cadmium there was no clear decrease or even an increase compared to 2006. Summer averages of nickel, arsenic, cadmium and lead were below the respective annual target values and air quality criteria. Levels for chromium were above the air quality criterion, however, this is also the case at regional background stations.

Polycyclic aromatic hydrocarbons (PAHs) were analysed from filters (i.e., particle-bound compounds). Benzo(a)pyrene (BaP) which is almost exclusively particle-bound, has strongly decreased in concentration over recent decades. The summer average concentration of BaP close to Hydro Sunndal in summer 2019 (0.009 ng/m^3 at Pensjonistsenteret) was two orders of magnitude lower than the target value of 1 ng/m^3 (for calendar year). Also the air quality criterion for BaP in Norway (annual average 0.1 ng/m^3) is not expected to be exceeded. The level measured in Sunndalen in summer 2019 was slightly higher than the Norwegian regional background.

[1] Only PAH-measurements are available from the early 1990ies. Modelling results from the beginning of the 1990s exist for SO_2, fluorides and PM. Metals have not been measured before 2002.

Sulphur dioxide (SO_2) concentrations decreased about 63% between summer 2002 and summer 2019, as a consequence of reduced emissions. The summer average concentration of SO_2 in summer 2019 was well below the annual limit value (for the calendar year). The SO_2-levels measured in Sunndalen are elevated compared to the background level.

Fluorides were measured both in gaseous- and in particle-bound forms. Levels of gaseous- and particle-bound fluorides were lower in summer 2019 than in summer 2006, while there was no pronounced decrease in emissions. There are no limit values or target values valid today. Average fluoride concentrations in summer 2019 were below the WHO guideline value of 1 µg/m^3 as annual average.

Hydro Sunndal was selected for this study since the smelter is situated within a narrow valley and the plant comprises anode production. Air quality measurements in its surroundings may therefore represent a close to worst case[2] and cover a wide range of compounds – particulate matter, heavy metals, PAHs, sulphur dioxide and fluorides.

[2] Icelandic aluminium smelters do not have wet scrubbers to remove SO_2, so the term "worst case" is not applicable compared to them for this parameter. It is also likely that dispersion conditions in Årdal are even less favourable, so that ambient concentrations of some compounds may be higher there. Finally, Lista is using Söderberg technology with much higher emissions of PAH than Sunndal. A separate ESPIAL report covers this aspect.

Air quality assessment of the surroundings of the Hydro Sunndal aluminium smelter

Measurements May – August 2019

1 Introduction

A three-month sampling campaign focusing on air quality was carried out in the surroundings of the Hydro Sunndal aluminium smelter in summer 2019. Particulate matter ($PM_{2.5}$ and PM_{10}), sulphur dioxide (SO_2), heavy metals/metals (V, Cr, Ni, Ga, As, Cd, Sb, Pb, Bi, Al), particle-bound polycyclic aromatic hydrocarbons (PAHs, EPA 16 PAH) and fluorides (gaseous and particle bound) were sampled at two sites, one close to the smelter, the other one a few kilometres up the valley. At both sites, air quality measurements have been carried out in the past, so that measurement results can be compared to earlier levels.

1.1 Background and scope

The effect of aluminium production on the environment surrounding aluminium smelters has been studied over several decades. NILU has studied their effects on air quality both in measurement and modelling studies since the early 1970s. In the beginning of the 1990s, the "Effect Study" was commissioned by the Norwegian aluminium industry, focusing mainly on effects on vegetation, water, farm animals, game animals and human health. An update of the Effect Study, including effects on air quality, is being carried out now.

The main aim of this project is to advance the knowledge regarding the environmental consequences associated with emissions to air from the production of primary aluminium from the production technologies available today. The aim is achieved through the assessment of the effect of historical emissions on air quality in the past (report in preparation) and measurement of the most relevant air pollutants emitted during aluminium production (this study and measurement study in surroundings of Alcoa Lista, to be published later). The outcome from these activities will contribute to knowledge creation at the Al-industries and to secure the future sustainability of the aluminium industry in Norway/Northern Europe.

In order to establish up-to-date knowledge on the ambient air quality status in the surroundings of aluminium plants today, field campaigns are carried out at selected smelters. The 10 smelters participating in the ESPIAL[3] project are placed at very different locations, regarding dispersion conditions, population exposure, topography etc. This makes it difficult to conclude on the situation around the other smelters based on measurements at only one distinct location. Sunndal and Lista were indicated as suitable sites, one located in a topographically complex terrain, the other in a flat area at the coast. Two separate sampling campaigns were carried out. The present report covers measurements at Sunndalsøra in summer 2019 and comparison to the results from previous studies around Sunndalsøra, back to the early 1990s.

1.2 Hydro Sunndal

The Hydro Sunndal plant lies at Sunndalsøra, at the mouth of the Driva river into the Sunndalfjord. The Sunndal valley is a long, deep, U-shaped valley, flanked on both sides by 1500 – 1800 m high mountains

[3] ESPIAL - Ensuring the Environmental Sustainability of production of PrImary Aluminium

as illustrated in Figure 1. The prevailing wind direction driven by the sea breeze is up the valley in summer and during daytime, and down the valley in winter and at night-time. The topographic conditions around Sunndalsøra provide somewhat complicated dispersion conditions. Wind direction and speed can vary from one location to another within a short distance. To the south, there is a narrow valley, Litledalen. Sunndalen and Litledalen opening out into the Sunndalsfjord. Wind coming into the fjord from the north will transport emissions from Hydro Aluminium Sunndal into Sunndalen in a south-easterly direction, while drainage winds down Sunndalen will transport the emissions out into the fjord where the dispersion conditions also are influenced by wind coming down Litledalen.

Figure 1: Hydro Sunndal at Sunndalsøra. (Left): View from Sunndalsfjord towards southeast into the valley. (Right): Map over Sunndalsøra.

The municipality Sunndal has a population of about 7100, whereof nearly 60% are living in the village Sunndalsøra. The aluminium plant employs about 700 and is the largest industrial workplace in the municipality. There is extensive agriculture in the flat valley between Sunndalsøra and Gjøra, some 30 km southeast of Sunndalsøra. Grain and potatoes are the main products, but a substantial amount of livestock is also kept, for both meat and milk production. Due to risk of fluorosis, milk production was earlier restricted in the lower parts of the valley, but these restrictions were lifted since 2000[4].

The Sunndal plant started up in 1954 with one potline (SU1), and the next (SU2) commenced operations in 1958. Both these potlines were based on Söderberg technology. In 1968, SU3 was started, based on prebake technology. Another expansion came in 2002-2004, 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 2011-2015. Increased amperage over the years has also resulted in increased output, up to a total of 407 000 tonnes per year in 2017. An overview of major technological changes at Hydro Sunndal is given in Table 1.

The Sunndal plant also has a 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.

[4] Due to low fluorine emission in the end of the 1990s, low fluorine levels in animal feed and no further registrations of tooth damage in livestock, restrictions were lifted in 2000. Farmers in the lower valley were compensated for refraining from engaging in milk production. Increased fluorine emission due to underdesigned emission treatment plant between 2002 and 2008 (when the emission treatment plant should be expanded) led to further examination of the teeth of farm animals in 2008 and 2011 by the Norwegian Food Safety Authority and local veterinarians, respectively. These concluded that there is no animal welfare problem as a result of fluoride-emissions from the smelter and that there is no reason for restrictions on animal husbandry due to fluoride in the lower part of Sunndalen.

Table 1: Overview of major technological changes at Hydro Sunndal.

Year	Technological development
1954	Sunndal plant started up with one potline (SU1)
1958	The next (SU2) commenced operations. Both SU1 and SU2 were based on Søderberg technology
1968	SU3 was started, based on prebake technology
1969	Production capacity was doubled to 120 000 tonnes when the modern prebake plant (SU3) with 168 cells became operational
2002	Søderberg plant with 300 electrolytic cells was operational until 2002. The last Søderberg oven was shut down in the end of 2002
2002-04	Another expansion in 2002-04, modernization, when a new prebake line (SU4, 340 cells) gradually replaced SU1 and SU2[5]
2009	SU3 was temporarily closed following the financial crisis
2011-15	SU3 gradually restarted

Figure 2 shows the development of production and emissions of PM, metals, PAHs, SO_2 and total fluorides, respectively from 1992 to 2019.

[5] In connection with the modernisation and expansion of production at Hydro Aluminium Sunndal in the period 2001 – 2004, three treatment plants were built for the recovery of fluorine from the new electrolysis plant Su4. Furthermore, a new plant was built to replace the almost thirty-year-old treatment plants for the electrolysis plant Su3. Increased aluminium production also required expanded cleaning capacity. All four treatment plants have been expanded in 2008 to bring the emission of fluoride to air below the authorities' requirements of 10 kg/h on an annual basis and 13 kg/h on a monthly basis.

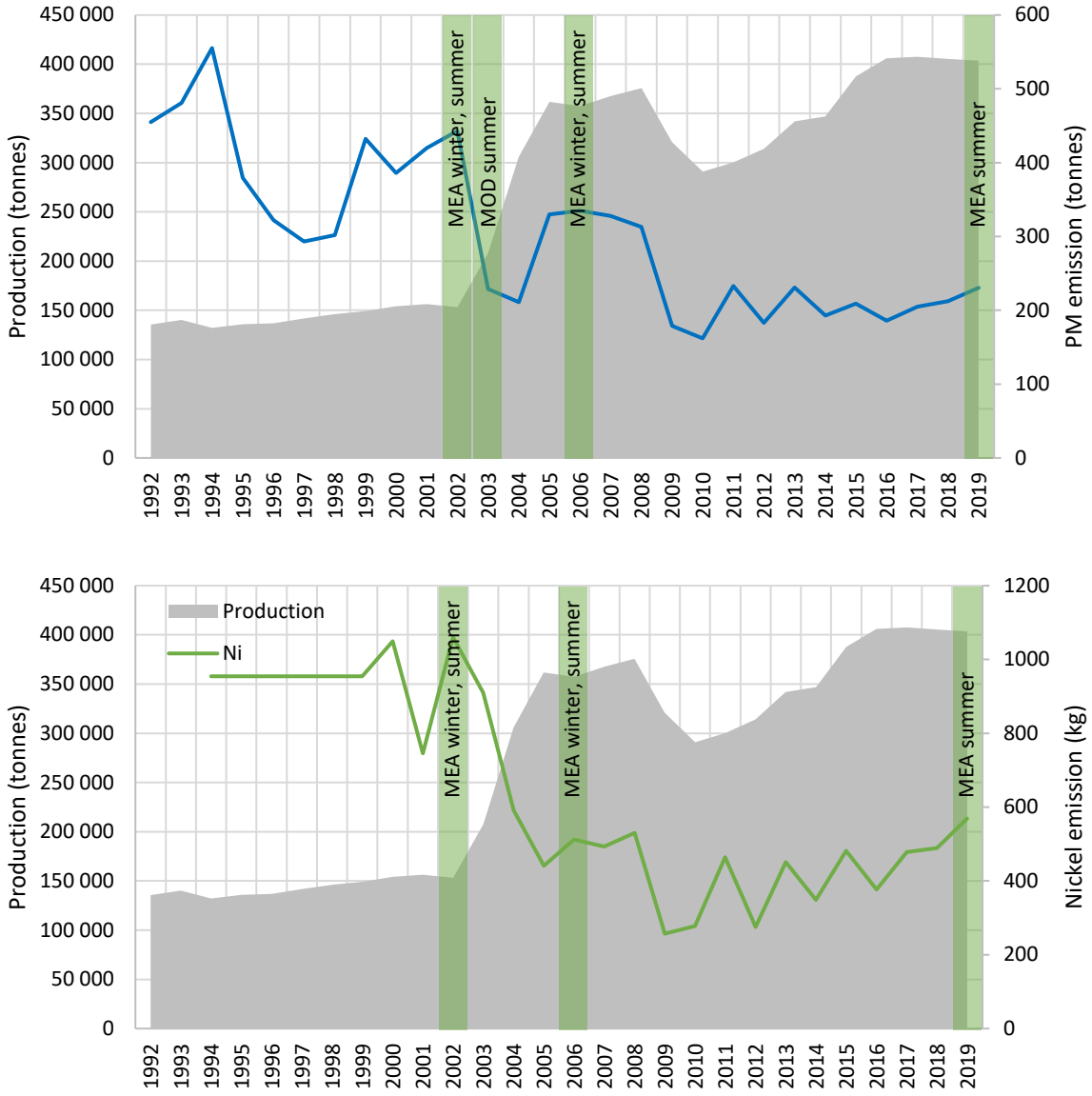


Figure 2: Development of production (grey background, unit: tonnes per year, left-hand y-axis) and emissions of PM, metals, PAHs, SO_2 and total fluorides, respectively, (unit: tonnes or kg per year) from 1992 to 2019. The right-hand y-axis indicates which compound is displayed. The years measurement (MEA) or modelling (MOD) studies for the individual compounds were carried out, are highlighted.

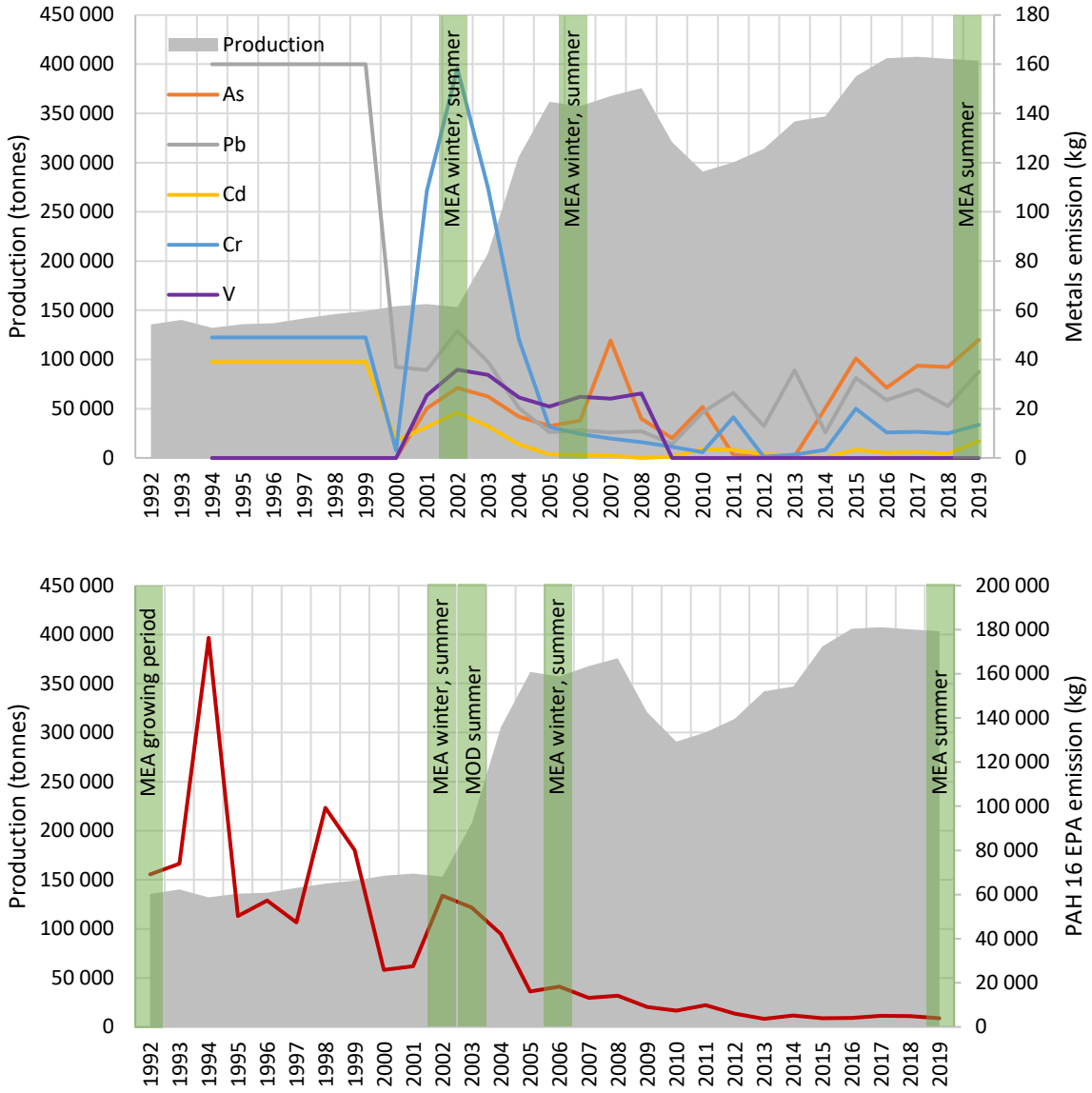


Figure 2 continued: Development of production (grey background, unit: tonnes per year, left-hand y-axis) and emissions of PM, metals, PAHs, SO_2 and total fluorides, respectively, (unit: tonnes or kg per year) from 1992 to 2019. The right-hand y-axis indicates which compound is displayed. The years measurement (MEA) or modelling (MOD) studies for the individual compounds were carried out, are highlighted.

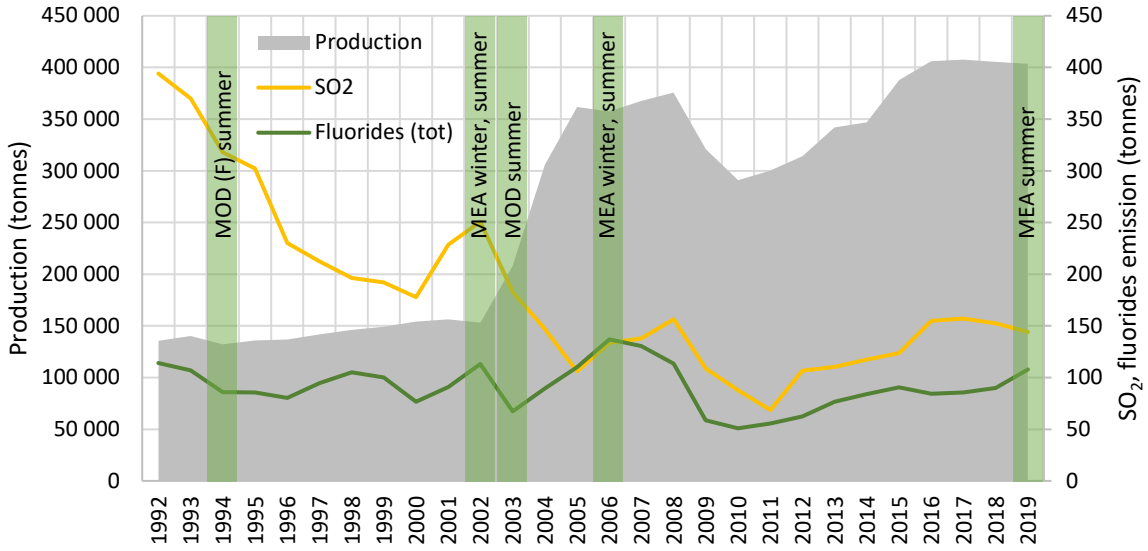


Figure 2 continued: Development of production (grey background, unit: tonnes per year, left-hand y-axis) and emissions of PM, metals, PAHs, SO_2 and total fluorides, respectively, (unit: tonnes or kg per year) from 1992 to 2019. The right-hand y-axis indicates which compound is displayed. The years measurement (MEA) or modelling (MOD) studies for the individual compounds were carried out, are highlighted.

- Production (tonnes per year) has increased almost threefold in the period 1992 – 2017, while total fluoride emissions (tonnes per year) have not changed markedly compared to the early 1990s, due to new fume-treatment plants removing fluorides.
- For SO_2 and PM (tonnes per year), emissions have been reduced about 60%. For comparability, the PM figures do not include fugitive emissions, since these have not been reported through the whole period, and the basis of reporting has been changed.
- For PAH to air (kg per year), the figures before 2015 are recalculated to the EPA standard. For the oldest figures, this recalculation entails quite big uncertainty. With this reservation regarding the accuracy of early measurements, the figure indicated a 90-95% reduction in this period (pers. comm. Leif Ongstad, 2019). Most of the reduction in PAH-emissions came as a result of the closure of the Söderberg potlines in 2003. The remaining emissions after 2004 are coming from the anode production.
- The emissions of metals were not reported in the beginning of the 1990s. Nickel emissions were almost halved between 1999 and 2019. No reduction of arsenic, lead, cadmium, chromium and vanadium emissions has been reported over the last 20 years. However, there are quite great uncertainties regarding the emission measurements of metals, and there have been some changes in methods, which makes historical trend comparisons difficult.

1.3 Earlier studies on air quality at Sunndal

Measurement and modelling studies

Overall, eight measurement studies and five modelling studies have been carried out around Hydro Sunndal before. Most earlier studies at Hydro Sunndal focussed on SO_2 and fluorides or PAHs in air (see overview in Table 2). Two measurement studies, in 2002 and 2006, also addressed particulate matter and heavy metals. Individual studies focused on fluorides and PAHs in crops and pasture land (OR 77/88 and TR 4/94).

Table 2: Overview of earlier measurement and modelling studies on air quality carried out in the surroundings of Hydro Sunndal. The present study covers measurements carried out in summer 2019 and comparison to results back to 1992 (shown with blue background). Studies before 1992 (grey background) are only mentioned to provide an overview.

SO_2	F_g / HF	F_p	PAH	PM	HM	Report
	Fluoride	Fluoride	PAH	PM (dust)		OR 1/83 (measurement, summer+ autumn 1981) PS
SO_2	Fluoride	Fluoride				OR 33/84 (measurement) PS
SO_2	Fluoride	Fluoride	(PAH)			OR 58/84 (modelling)
SO_2	Fluoride	Fluoride				OR 28/85 (measurement)
	Fluoride	Fluoride				OR 77/88 (measurement) pasture grass, 20-30 years
			33 PAHs			OR 42/91 (measurement)
SO_2	HF	Total fluorides	PAH	PM (dust)		OR 46/91 (modelling) summer 1990, summer 1997
			33 PAHs			OR 1/92 (measurement) air, salad summer 1991
			32 PAHs			TR 4/94 (measurements) crops, growth seasons 1989, 1990, 1991, 1992
	"Fluorides"	"Fluorides"				OR 15/94 uptake in

SO_2	F_g / HF	F_p	PAH	PM	HM	Report
						plants, wet deposition (modelling)
			33 PAHs*			OR 37/2002 (measurements) winter 2001-2002*
SO_2	F_g	F_p	33 PAHs	PM_{10}	Pb, Cd, Cu, Zn, Cr, Ni, Co, As, Al, V	OR 63/2003 (measurements) winter 2002, summer 2002
SO_2	Fluorides	Fluorides	PAH	PM_{10}		OR 88/2003 (modelling)
SO_2	F_g	F_p	33 PAHs	PM_{10}	Pb, Cd, Cu, Zn, Cr, Ni, Co, As, Al, V	OR 89/2006 (measurements) winter 2006, summer 2006
SO_2	F_g	F_p	16 PAHs particle bound	$PM_{2.5}$, PM_{10}	Pb, Cd, Cr, Ni, As, Al, V, Ga, Sb, Bi	This report 2/2021 (measurements) summer 2019

*Same samples as reported in OR 63/2003

Moss surveys

It was early discovered that terrestrial moss has the ability to be used for monitoring of atmospheric deposition of pollutants. In Norway, moss sampling is conducted by sampling the terrestrial moss *Hylocomium Splendens* which is shown to successfully sample deposition of trace metals. In Norway, nationwide atmospheric deposition surveys for heavy metals have been conducted at regular intervals since 1977. Sampling of moss is included in the state program for monitoring pollution, and is additionally part of an international survey where moss is sampled in several European countries. In addition, since 2000, moss surveys are carried out every 5 years around selected Norwegian industries (Steinnes et al., 2001; Steinnes et al., 2007; Steinnes et al., 2011; Steinnes and Uggerud, 2017). Hydro Sunndal participated in all four studies of heavy metals in moss around Norwegian industries, 2000, 2005, 2010 and 2015. The number of metals analysed and the number of sites varied between the studies. In the latest study, Al, Ti, V, Ni, Ga and Bi were moderately elevated compared to background values. The levels of these compounds have stayed relatively constant since 2000.

In summer 2015, a parallel moss survey focusing on PAHs was carried out around Norwegian industries, including Hydro Sunndal (Halse et al., 2017). Passive air samples and moss samples were collected around the industrial plants to be analysed for PAHs.

Passive air samplers were placed at 5 sites around Hydro Sunndal. The PAS were deployed along a transect going from each industrial site, for one month during summer 2015. The nearest background station is Kårvatn (20 km northeast of Sunndalsøra). The sum of 8 PAHs (fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benz(a)anthracene, chrysene, benzo(a)pyrene) was detected with higher amounts for all sites compared to the background station at Kårvatn.

1.4 Measurement sites

Two sampling sites in the surroundings of the aluminium smelter Hydro Sunndal, *Pensjonistsenteret* in the centre of Sunndalsøra and *Vennevold* 6 km up the valley Sunndalen, were selected in order to be able to compare measurement results with the results of the two most recent measurement projects, carried out in 2002 (Hagen, 2003) and 2006 (Hagen, 2006). Their locations were initially selected according to local topography, residents' exposure and local wind patterns. Figure 3 shows the locations of the stations and the aluminium smelter, and illustrates the topographic conditions in the area. Pictures of the measurement stations are given in Figure 4.

Figure 3: Map over the western part of Sunndalen, including Hydro Sunndal at the fjord, the measurement station "Pensjonistsenteret" (red marker) in the centre of Sunndalsøra, and the measurement station "Vennevold" (green marker) 6 km up the valley.

Figure 4: Sampling sites in the surroundings of the aluminium smelter Hydro Sunndal. Left: Pensjonistsenteret. The large intake above the roof is the intake for the PAH-sampler.

The PM-samplers are placed on the roof behind the PAH-intake. On the wall visible in the picture, the inlet for the SO_2- and fluoride-sampler can be seen. Right: Vennevold. The PAH-intake and the two PM-samplers on the roof are easily discernible. The inlet for the SO_2- and fluoride-sampler is on the remote wall of the station (not visible in the picture).

In studies earlier than 2002, measurements were carried out at several different sites within Sunndalsøra, making it more difficult to compare the levels measured.

1.5 Measurement program and methods

At both sites, weekly samples were taken of $PM_{2.5}$, PM_{10}, SO_2, fluorides and particle-bound PAHs in the three month period 23 May to 15 August 2019. Weekly average concentrations were analysed from the samples for $PM_{2.5}$, PM_{10}, 10 selected metals in PM_{10}, SO_2, gaseous fluoride and particle bound fluoride. The weekly PAH-samples were merged to monthly samples.

When measurements do not cover a whole year, it is recommended to carry out the measurements in the season which is characterised by the highest exposure to air pollution. For most locations, the situation is worst in the winter months due to unfavourable dispersion conditions. At Sunndalsøra, however, the distribution of wind direction frequencies leads to exposure of inhabited areas to emissions from the aluminium smelter mainly during summer. This is also the period when vegetation is active and will be most affected by the emissions. During the summer months, prevailing north-westerly wind, i.e., up the fjord, transports emissions from the aluminium plant over the built-up area at Sunndalsøra and into the valley Sunndalen, while during winter east-south-easterly wind carries emissions from the smelter mainly out towards the Sunndalsfjord and away from the settlement.

Particulate matter (PM)

Filter samplers (Kleinfiltergerät, Leckel Ingenieurbüro GmbH, Berlin) with $PM_{2.5}$- and PM_{10}-impactor, respectively, were used to sample $PM_{2.5}$ and PM_{10} in air. Particulate matter was sampled with an airflow of 2.3 m^3/h on filters (47 mm) which were gravimetrically analysed by NILU's laboratory in order to determine the average mass concentrations during the exposure time. The procedure used is according to the reference method for measurement of PM in ambient air (EN 12341:1999). The exposure time for each filter was 7 days.

Metals

Metals were analysed from the weekly PM_{10}-samples collected, as the existing limit/target values for heavy metals (As, Cd, Ni, Pb) apply to the size fraction of particles with diameters smaller than 10 µm (PM_{10}). The filters were digested by acid hydrolysis, applying a microwave-based decomposition technique, to extract (heavy) metals. Heavy metal concentrations are determined by inductively coupled plasma mass spectrometry (ICP-MS). The high temperature of the plasma ion source breaks apart the molecules present in the sample. The elemental ions in the plasma are quantified using a mass spectrometer. For heavy metals, there are specific requirements for cleanliness for preparation and treatment of the equipment to avoid contamination, i.e., acid-washed equipment is used for sampling and preparations. NILU has a clean room laboratory for handling and analysis of heavy metal samples. A selection of ten metal compounds was analysed using ICP-MS: lead (Pb), cadmium (Cd), arsenic (As), chromium (Cr), nickel (Ni), vanadium (V), gallium (Ga), antimony (Sb), bismuth (Bi) and aluminium (Al). The individual compounds analysed cover the metal compounds Hydro Sunndal has an

emission permit for (As, Pb, Cd, Cr, Ni[6]), compounds which show increased levels around aluminium smelters in moss studies (e.g. Ga, Bi, Sb) and thus cover most compounds analysed earlier, considering possible analytical constraints. Some heavy metals are among the emissions with potential for exceedance of regulations in the vicinity of aluminium smelters. Heavy metals in aluminium production occur both in raw materials and anode material. The content varies strongly.

Sulphur dioxide and fluorides

SO_2 and fluorides were sampled with NILU's "EK-sampler", which samples both aerosol and gaseous compounds in a filter package. The filter package contained two filters, the first one sampling aerosol (analysed for fluoride), the second one impregnated with KOH to sample gases (analysed for SO_2 and HF). The filter package is installed with an impactor and a pump, drawing 10 l air per minute through the filters. The EK samplers were placed at the two sites in the surroundings of the smelter to study spatial variations of particulate fluoride, gaseous fluoride (HF) and sulphur dioxide. The exposure time was 7 days.

The aerosol pre-filter is weighed under controlled conditions (20°C and 50% relative humidity) before and after field exposure to determine the mass of particles collected. In the further analysis, a water extraction is used to dissolve sulphate and fluoride. Ion chromatography is used for the analysis of particulate sulphate and fluoride. On the KOH-impregnated filter, SO_2 and HF (hydrogen fluoride) are absorbed. A water-H_2O_2 extract is used to dissolve what is absorbed. The water extract then contains sulphate and fluoride ions. Sulphate is analysed by ion chromatography. The amount of fluoride is determined by an ion selective electrode. Since HF is highly reactive, some of this gas can be absorbed on particles already deposited on the aerosol filter. It is therefore possible that some of what has been reported as particulate fluoride originally was HF. Hydrogen fluoride is a pollutant with a relatively low general background concentration in the atmosphere, which can be encountered in high concentrations close to aluminium smelters.

Polycyclic aromatic hydrocarbons (PAHs)

Particle-associated PAHs in air were quantitatively sampled on glass fibre filters at both sites using high-volume air samplers (Digitel Elektronik AG, Switzerland). The sampler consists of a pump that draws air through the samples with an average flow rate of 25 m^3/h. PAHs were sampled weekly as 7-day averages, covering the entire sampling period. In addition, field blank samples followed the sample batch in order to control potential contamination risks (as part of the extensive quality control procedure). All exposed samples were stored cold (2°C) prior to analysis and quantification. The filters were extracted with solvent. The weekly samples were merged to monthly samples, which were analysed for 16 priority[7] PAHs (EPA 16 PAH) by NILU's laboratory. For analysis, PAHs are extracted from the filters and the extracts are analysed using gas chromatography. Identification and quantification of the PAHs was carried out using a high-resolution gas chromatograph coupled to a low-resolution mass spectrometer as detector (GC/LRMS). The detection limit for PAH-compounds was 0.001 – 0.002 ng/m^3.

Benzo(a)pyrene (BaP) is the only PAH with an air quality target value. The target value is established for 1 year averages. BaP is mainly particle-bound. Sampling in summer, as decided for Sunndal, has the advantage that wood burning (residential heating), which is another major local source for PAHs, is not

[6] The selection of metals analysed does not include Hg (which Hydro Sunndal has an emission permit for), which according to moss studies does not appear to be a problem around the aluminium smelter.

[7] The EPA 16 PAHs are of environmental concern because of their potential toxicity in humans and other organisms and their prevalence and persistence in the environment. Several PAHs are probable or known carcinogens (IARC, 2010).

an interfering issue. The particles collected on the particle filter are mainly in the size range below 10 µm. Larger particles can occur, but not in significant amount.

The composition of the PAH samples, the so-called PAH-profile, tells us a lot about which sources contribute most to air concentrations. Some PAH compounds which are connected with emissions from the aluminium industry are phenanthrene, fluoranthene and pyrene. BaP is found in small quantities in the emissions from aluminium plants, as well as in emissions from car traffic and wood burning. Coronene is an indicator of traffic emissions (but is not among the 16 priority PAHs).

PAH-measurements at Årdal and in Oslo in winter 1991 (report OR 42/1991) show how PAHs split between gaseous and particle-bound state. In Figure 5, the gas-particle-split for the 16 priority PAHs is illustrated. The lightest compounds, naphthalene, acenaphthylene, acenaphthene and fluorene occur almost exclusively in the gas phase. Phenanthrene, anthracene, fluoranthene and pyrene are mainly in the gas phase (70-90%). Benz(a)anthracene and chrysene are mainly bound to particles (70-80%). Benzo(b,k)fluoranthenes, benzo(a)pyrene, inden(1,2,3-cd)pyrene, dibenzo(ac/ah)anthracenes and benzo(ghi)perylene occur almost exclusively in particulate phase. In summer, the split may be slightly shifted to the advantage of gaseous compounds, however, no PAH-data separated into gas phase and particle phase is available for summer.

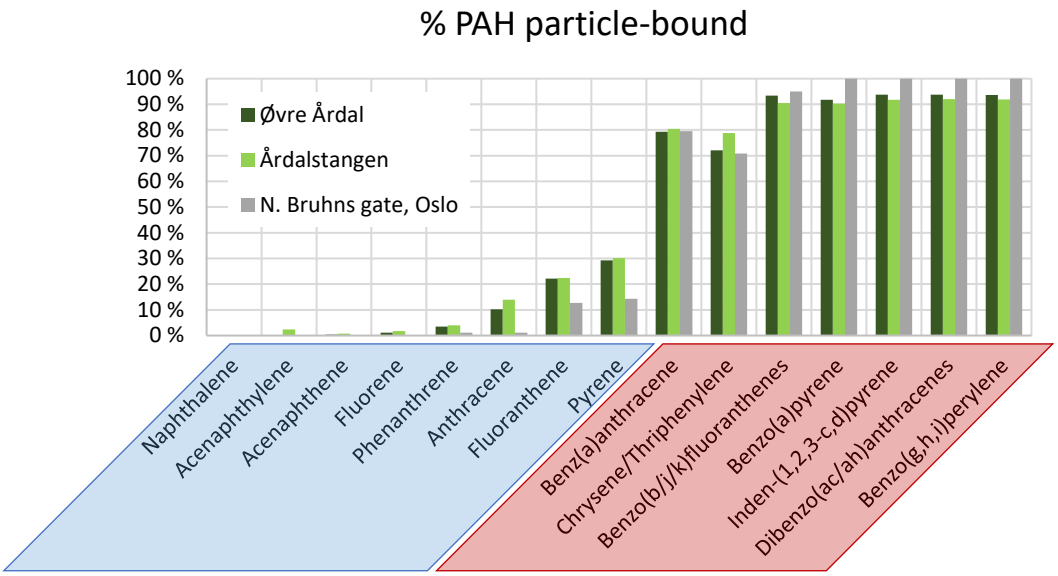


Figure 5: Split of 16 priority PAHs into mainly gaseous compounds (blue background) and mainly particle-bound compounds (red background). The height of the bars corresponds to the percentage of the compound concentration occurring in particle form. Data from measurement in Årdal and Oslo in winter 1991 (Hagen, 1991; OR 42/91).

2 Air quality guidelines and limit values

Humans can be adversely affected by exposure to air pollutants in ambient air. National and international limit and guideline values and air quality objectives have therefore been established for a number of pollutants present in the air. These limit, threshold and guideline values are summarised in Table 3. They apply over differing periods of time because the observed health impacts associated

with the various pollutants occur over different exposure times. Norwegian limit and target[8] values are equal to EU limit and target values (except for PM where Norway has stricter limit values). Limit values (Forurensningsforskriften, EU air quality directives) are legally binding. In case of exceedances, authorities must develop and implement air quality management plans which should aim to bring concentrations of air pollutants to levels below the limit and target values.

The WHO guideline values are set for the protection of health, and are generally stricter than the comparable politically agreed EU standards. Air quality criteria are adopted by the Norwegian Environment Agency and the Institute of Public Health. They are stricter than the limit and target values. The air quality criteria are based on existing knowledge about the potential health effects of exposure to air pollution. The criteria are set at a level that most people can be exposed to without experiencing harmful health effects. Guideline values and air quality criteria are recommendations.

Table 3: National limit values and air quality guidelines for the compounds measured. Note that Norwegian national limit values for particulate matter are stricter than EU limit values defined in the Air Quality Directive (AQD).

Compound	Averaging period	Limit value (FF[9])	WHO Air Quality Guidelines	Air quality criterion (LKK[10])
Particulate matter (PM)				
PM_{10}	Day (24 hours)	50 µg/m^3 (max. 30 exceedances) (AQD: max. 35 exceedances)	50 µg/m^3	30 µg/m^3
	Calendar year	25 µg/m^3 (AQD: 40 µg/m^3)	20 µg/m^3	20 µg/m^3
$PM_{2.5}$	Day (24 hours)	-	25 µg/m^3 (99th percentile)	15 µg/m^3
	Calendar year	15 µg/m^3 (AQD: 25 µg/m^3)	10 µg/m^3	8 µg/m^3
Sulphur dioxide (SO_2)				
SO_2	10 min		500 µg/m^3	
	15 min			300 µg/m^3
	1 hour	350 µg/m^3 (not to be exceeded more than 24 times per calendar year)		

[8] Target values are set out in the same way as limit values. They are to be attained where possible by taking all necessary measures not entailing disproportionate costs.

[9] FF: Forurensningsforskriften (Norwegian air quality regulation, embedding European air quality limit values). URL: https://lovdata.no/dokument/SF/forskrift/2004-06-01-931/KAPITTEL_3-1#KAPITTEL_3-1

[10] LKK: Luftkvalitetskriterier (Air quality criteria, set by Norwegian institute of public health and Norwegian environment agency).
URL: https://www.fhi.no/globalassets/dokumenterfiler/rapporter/2013/luftkvalitetskriterier---virkninger-av-luftforurensning-pa-helse-pdf.pdf

	Day (24 hours)	125 µg/m^3 (not to be exceeded more than 3 times per calendar year)	20 µg/m^3	20 µg/m^3
	Winter period 1.Oct – 31.March (ecosystem)	20 µg/m^3		
	Calendar year	20 µg/m^3		
Metals				
Arsenic (As) in PM_{10}	Calendar year	Target value 6 ng/m^3		2 ng/m^3
Lead (Pb) in PM_{10}	Calendar year	Limit value 0.5 µg/m^3		0.1 µg/m^3
Cadmium (Cd) in PM_{10}	Calendar year	Target value 5 ng/m^3		2.5 ng/m^3
Chromium (Cr) Cr VI	Calendar year			0.1 ng/m^3
Manganese (Mn)	Calendar year			0.15 µg/m^3
Nickel (Ni) in PM_{10}	Calendar year	Target value 20 ng/m^3		10 ng/m^3
Vanadium (V)	Day, 24 hours		1 µg/m^3	0.2 µg/m^3
Polycyclic aromatic hydrocarbons (PAHs)				
B(a)P in PM_{10}	Calendar year	Target value 1 ng/m^3		0.1 ng/m^3

There is no EU limit value for fluorides. According to WHO, a fluoride level (total fluoride) of 1 µg/m^3 as annual average, which is set to protect plants and crops will also be sufficient to protect human health (WHO, 2000). In the 1980s, two limit values for total fluoride (gaseous and particulate) taking into account human health were in force in Norway: 25 µg/m^3 for daily averages and 10 µg/m^3 for six month averages (SFT, 1982). SFT's recommended air quality criteria for gaseous fluoride earlier were set for protection of vegetation (SFT, 1992), 1.0 µg/m^3 as average over 24 hours, 0.4 µg/m^3 as average over 30 days and 0.3 µg/m^3 as average over 6 months. The criterion for animals was at 0.15 µg/m^3 averaged over 30 days.

Although there are some hints for toxic effects of aluminium, especially in the respiratory tract and nervous system, the knowledge of exposure conditions is so incomplete that FHI has chosen not to set air quality criteria for aluminium. Most studies, however, suggest that high concentrations of aluminium are needed before triggering health effects. This may indicate that the air quality criteria for particulate matter also will protect against health effects of aluminium. It is in accordance with international assessments not to set an air quality criterion for aluminium (FHI, 2013).

There is an air quality criterion for benzo[a]pyrene (B(a)P) as an indicator for carcinogenic effects of PAHs. PAHs are regulated in the Aarhus protocol on POPs (UN/ECE, 1998b) and the EU air quality directive (EU, 2004).

3 Measurement results

Results from measurements carried out in summer 2019 (23 May – 15 August) are shown and compared with the results from earlier measurements (see Table 2). Since the exposure time of individual samples and the sampling frequency differ for the individual campaigns, averages for the entire sampling periods are compared. Measurement results are also compared with regional background concentrations. Concentrations of selected environmental contaminants in air and precipitation at Norwegian background sites in 2019 are reported by Aas et al. (2020).

The meteorological station at Sunndalsøra (Sunndalsøra III[11]) was not operative during the measurement period. It is a long way to the next station. The complex topography in the area makes it difficult to say anything more about the wind conditions for Sunndalsøra during this period. Especially the lack of information on wind direction is disadvantageous for the interpretation of air pollutant results.

3.1 Particulate matter

PM_{10} and $PM_{2.5}$ were sampled at both sites, Pensjonistsenteret and Vennevold. In earlier measurement campaigns (winter 2002, summer 2002, winter 2006, summer 2006), only PM_{10} was measured. Particulate matter was measured as weekly average concentrations with equal exposure periods for both sites. In the earlier measurement campaigns (2002, 2006), two daily samples per week were taken.

In summer 2019 (23. May – 15. August 2019), rather low PM concentrations were measured, both at Pensjonistsenteret and at Vennevold (Figure 6). At Pensjonistsenteret, the third PM_{10} sample appeared not to have been exposed, so no PM_{10}-result exists for the period 6. June – 13. June 2019.

The weekly concentrations were at similar levels at both sites, both for PM_{10} and for $PM_{2.5}$ and the week-to-week variation over the 3-month period was similar at both sites (Figure 7). Weekly concentrations at Pensjonistsenteret varied between 3.6 µg/m^3 and 11.3 µg/m^3 for PM_{10} and between 2.0 µg/m^3 and 8.6 µg/m^3 for $PM_{2.5}$, at Vennevold weekly concentrations varied between 3.6 µg/m^3 and 12.2 µg/m^3 for PM_{10} and between 1.9 µg/m^3 and 9.7 µg/m^3 for $PM_{2.5}$. Average PM_{10} concentrations for the entire measurement period were 5.9 µg/m^3 at Pensjonistsenteret and 5.7 µg/m^3 at Vennevold. Average $PM_{2.5}$ concentrations were 3.6 µg/m^3 at Pensjonistsenteret and 3.9 µg/m^3 at Vennevold. An overview of these results is also given in Table 4.

At both sites, most PM_{10} was in the fine fraction, i.e., a particle diameter of less than 2.5 µm. Fine particulate matter, $PM_{2.5}$, represented between 46% and 76% (on average 59%) of PM_{10} at Pensjonistsenteret. Similarly, at Vennevold, between 52% and 80% (on average 66%) of PM_{10} were in the fine particle fraction. Roughly speaking, coarse particles, i.e., particles between 2.5 and 10 µm diameter are mainly mechanically produced, while fine particles, particles with diameter less than 2.5 µm are mainly produced by combustion processes and industrial processes.

Comparison to limit values

PM-concentrations measured around Sunndal in summer, which is the time of the year with highest air pollution in Sunndalen, were markedly lower during the 3-month measuring period than the annual limit value (PM_{10}: 25 µg/m^3 for calendar year, $PM_{2.5}$: 15 µg/m^3 for calendar year, see Table 3). This

[11] https://www.yr.no/nb/historikk/graf/563420/Norge/M%C3%B8re%20og%20Romsdal/Sunndal/Sunndals%C3%B8ra

indicates that international and national limit values and air quality guidelines were most likely not violated.

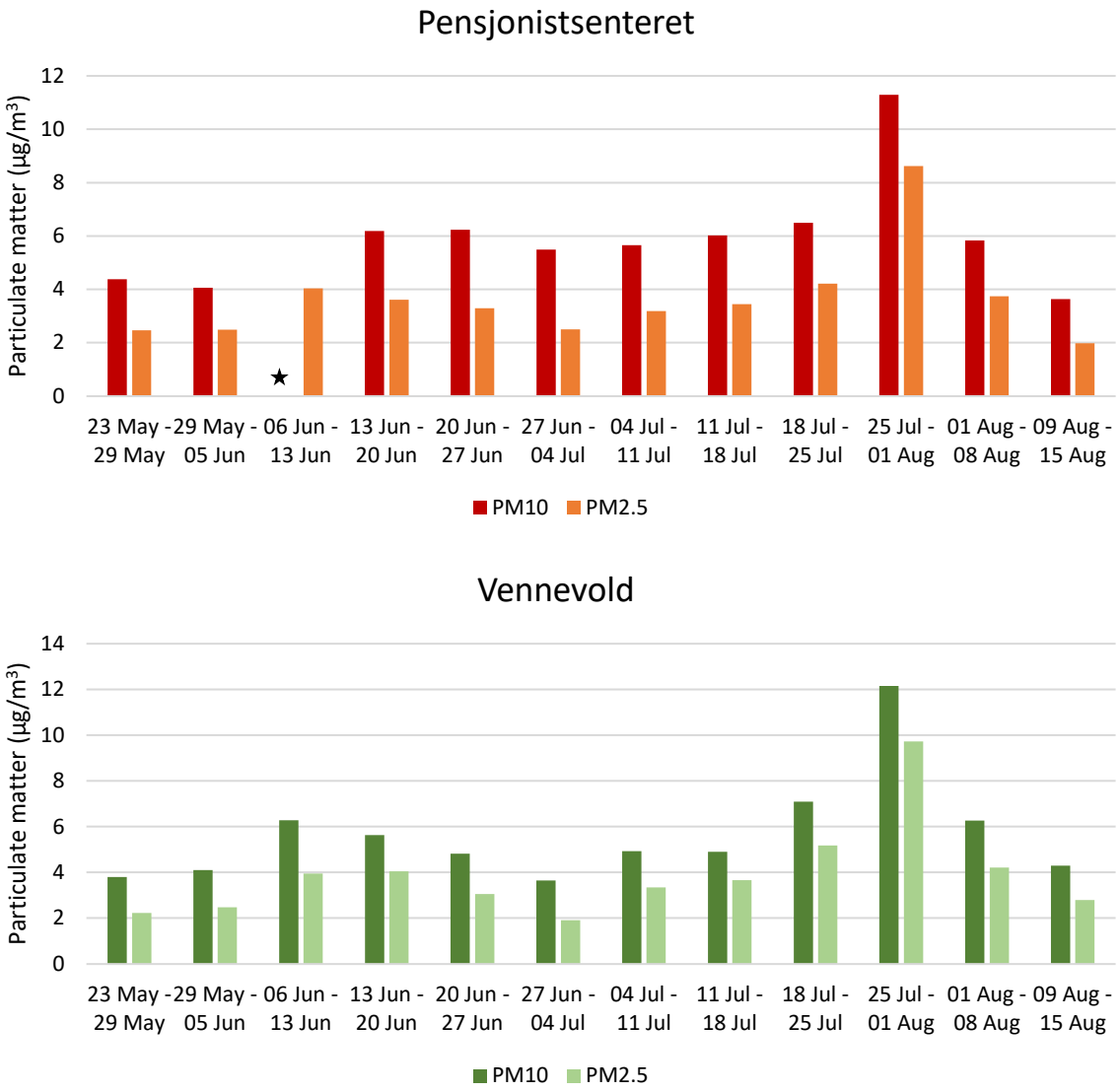


Figure 6: Weekly average concentrations of PM_{10} and $PM_{2.5}$ in summer 2019 (23. May – 15. August 2019 at Pensjonistsenteret (upper plot) and at Vennevold (lower plot). Asterisk marks a missing sample (see text). Limit values for annual average are 25 µg/m³ for PM_{10} and 15 µg/m³ for $PM_{2.5}$.

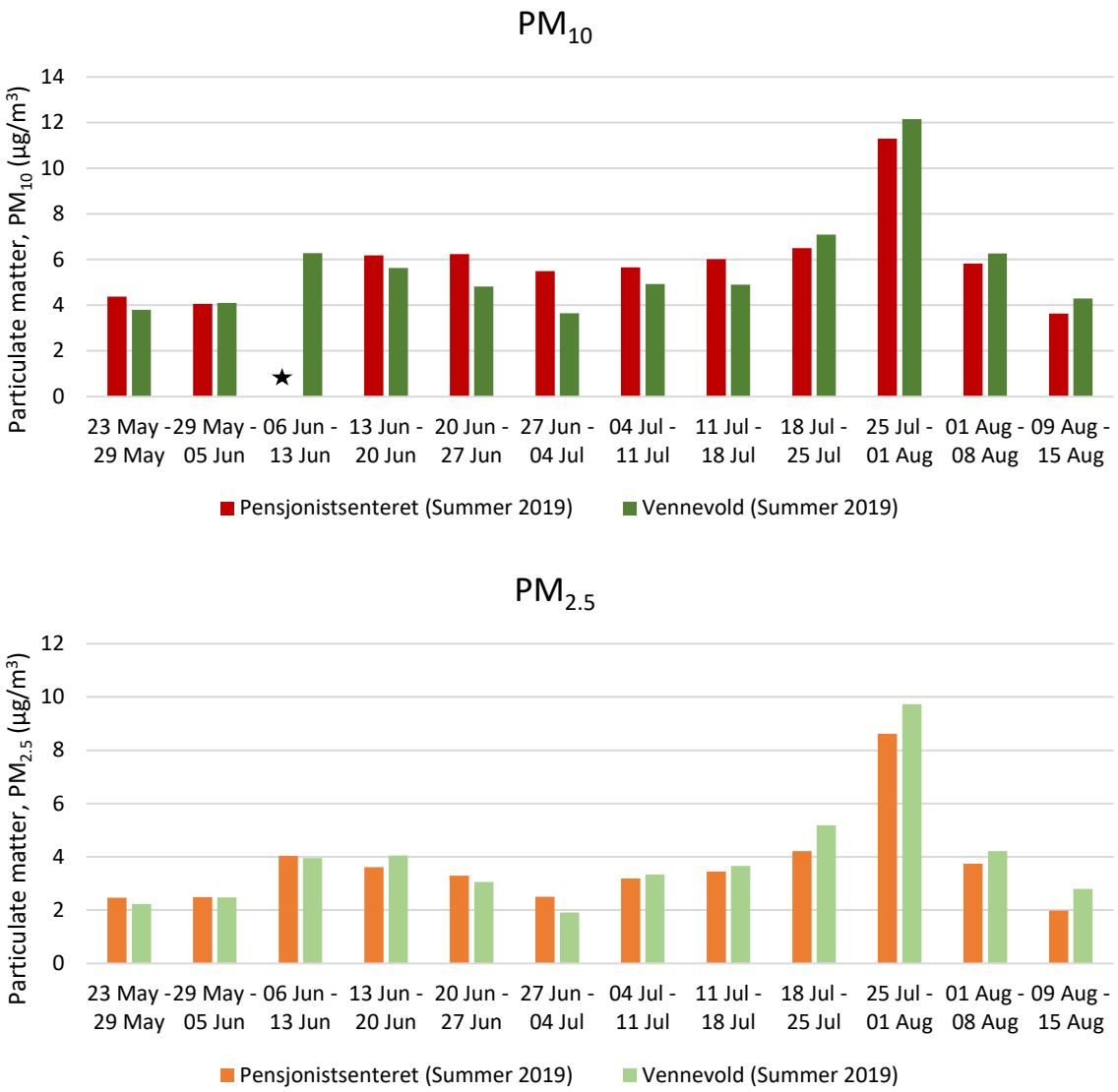


Figure 7: Weekly average concentrations of PM_{10} (upper plot) and $PM_{2.5}$ (lower plot) in summer 2019 (23. May – 15. August 2019). Comparison of concentrations at Pensjonistsenteret and at Vennevold. Asterisk marks a missing sample (see text).

Comparison to Norwegian background

Annual mean concentrations of PM_{10} and $PM_{2.5}$ observed at Norwegian rural background stations are amongst the lowest in Europe. Kårvatn (62°47' N, 8°53' E, 210 masl, Surnadal municipality, Møre og Romsdal county) is the background station closest to Sunndalsøra. It is indeed located very close, only 20 km northeast of Sunndalsøra, however in another valley. The annual average PM_{10} concentration at Kårvatn in 2019 was 2.9 µg/m^3, for $PM_{2.5}$ it was 1.9 µg/m^3 (Aas et al., 2020). There is a typical annual variation of PM at Kårvatn with maximum concentrations in summer. The average PM-level at Kårvatn in the period May – August 2019 was 3.8 µg/m^3 for PM_{10} and 2.7 µg/m^3 for $PM_{2.5}$ (see Table 4). At Kårvatn, the $PM_{2.5}/PM_{10}$-fraction was 66% in 2019, consistent with most previous years, and similar to the $PM_{2.5}/PM_{10}$-ratio observed in Sunndalen in the measurement period. The average PM_{10}-concentration in Sunndalen during the measurement period was ca. 50% higher than at Kårvatn in the period May – August 2019. For $PM_{2.5}$, the levels were ca. 30% higher.

Table 4: PM_{10}- and $PM_{2.5}$-concentrations ($\mu g/m^3$) at Pensjonistsenteret, Vennevold and the background station Kårvatn in the measurement period.

	Pensjonistsenteret	Vennevold	Kårvatn
PM_{10}			
Week, max	11.3	12.2	-
Week, min	3.6	3.6	-
Average	5.9	5.7	3.8
$PM_{2.5}$			
Week, max	8.6	9.7	-
Week, min	2.0	1.9	-
Average	3.6	3.9	2.7

The PM_{10}-level at Norwegian background sites showed a downward trend over the last 20 years. A trend analysis for Birkenes in Southern Norway reveals a statistically significant decrease of -33% from 2000 to 2019 (Aas et al., 2012). At Kårvatn, PM_{10} has been measured since 2010 and no trend analysis is available.

Comparison to previous campaigns

PM_{10} was included in measurement studies in 2002 and 2006 and in a modelling study in 2003 (see Table 2). In Figure 8, PM_{10}-levels from summer 2019 are compared to PM_{10}-levels in summer 2002 and summer 2006. Due to different meteorological patterns in winter, the results from the winter seasons in 2002 and 2006 are not included in the comparison. In summer 2019, PM_{10} at Pensjonistsenteret was about 50% lower than in summer 2006. PM was not measured at Kårvatn before 2010, thus no background levels are available for 2002 and 2006.

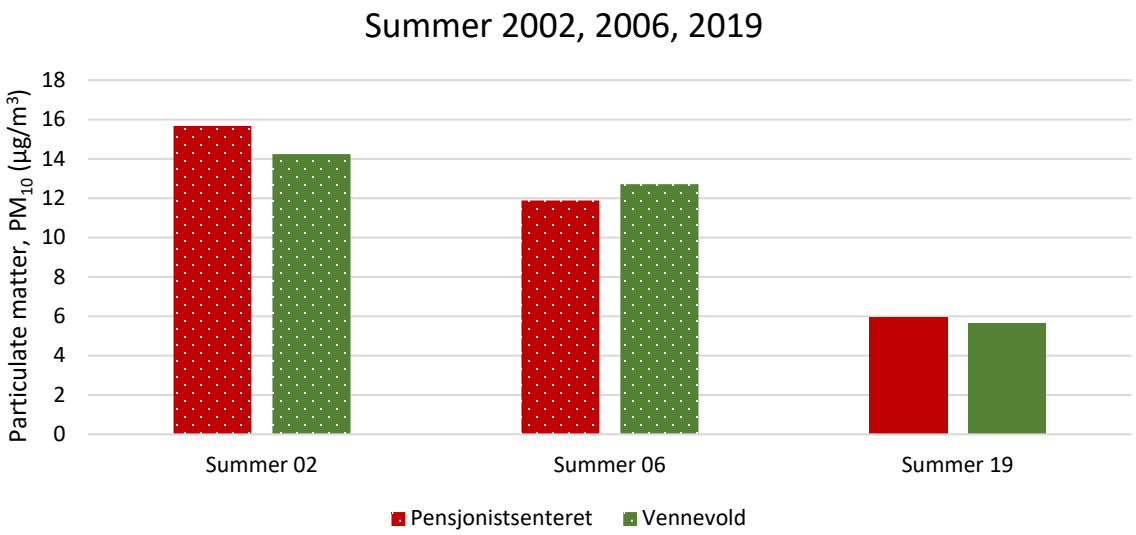


Figure 8: Average concentration of PM_{10} in summer 2002 (10 weeks), summer 2006 (9 weeks) and summer 2019 (12 weeks) at Pensjonistsenteret and at Vennevold. $PM_{2.5}$ was not measured at Sunndal in 2002 and 2006.

Emissions of particulate matter from Hydro Sunndal have decreased from 2002 to 2006 and further to 2019 (see Figure). The PM_{10}-levels measured during the three summer campaigns varied in a similar level as the PM emission. In 2002 and 2006, measurements were carried out on 20 days distributed over 3 months. These results are more sensitive to time variations and not directly comparable to long-term mean values over 3 months.

For comparison, an earlier modelling study (OR46/91, Bøhler and Larsen, 1991) calculated dispersion of pollution for the summers of 1990 and 1997 (i.e., after an expansion planned for 1997, consisting of building two new halls, SU4, at the same time as SU1/2 is closed). Dispersion calculations for summer 1990 (assuming particles below 10-15 µm in diameter) showed PM concentrations around 10-20 µg/m^3 in the centre of Sunndalsøra (half-year average over summer) and below 3 µg/m^3 several km up the valley. Closing SU1/2, which was the main PM-source, was assumed to result in a 50% reduction of dust emissions. The calculations show that summer averages in the range 5-10 µg/m^3 can appear in the centre of Sunndalsøra after the expansion.

3.2 Heavy metals

Metals were analysed from the PM_{10}-samples reported above (Section 3.1), i.e., weekly samples collected in the period 23. May – 15. August 2019 were analysed for 10 selected metals. Arsenic (As), lead (Pb), cadmium (Cd), chromium (Cr) and nickel (Ni) were analysed since these are compounds, Hydro Sunndal has an emission permit for. Vanadium (V), gallium (Ga), bismuth (Bi) and antimony (Sb) are compounds which showed increased levels in the surroundings of Hydro Sunndal and other aluminium smelters in previous moss surveys (see further down in this section). Aluminium (Al) is the product of the smelter, but is also an ubiquitous compound in the earth's crust, and has also been analysed in previous measurement campaigns. Metals were analysed both at Pensjonistsenteret and Vennevold, and measurement results from the same sites are available from previous campaigns in 2002 and 2006 (Hagen, 2003; Hagen, 2006).

Weekly average concentrations of the individual metals in PM_{10} are shown in Figure 9, comparing the levels at Pensjonistsenteret and Vennevold. For most compounds (Al, V, Ni, As, Cd, Sb, Pb, Bi), concentrations were higher at Pensjonistsenteret than at Vennevold. This may indicate that these compounds mainly originate from the aluminium smelter. In the week 25. July – 1. August 2019, some compounds (Al, Cr, Ga, Pb) had higher concentration at Vennevold. As no wind data are available, possible resuspension of dust cannot be verified.

Comparison to limit values

Target values (Forurensningsforskriften §7) exist for Ni, As, Cd and Pb. All weekly average concentrations of all compounds measured were distinctly lower than the respective target values for annual average values (Table 3, Table 5). Since the measurements were carried out in the time of the year with the highest expected pollution conditions in the valley, it is assumed that the annual average concentrations are below the respective target values as long as no incidents happen. In Table 5, average metal concentrations at the two sampling sites during the 3-month measuring period are compared to national target values and national air quality criteria (luftkvalitetskriterier), which are valid for annual average concentrations.

Table 5: Average metal concentrations (ng/m^3) at Pensjonistsenteret (PS) and Vennevold (VV) during the measurement period (23. May – 15. August 2019). National target values (FF) and air quality criteria (AQC) are defined for yearly average concentrations.

ng/m^3	Al	V	Cr	Ni	Ga	As	Cd	Sb	Pb	Bi
Average PS	468.7	0.96	0.24	3.45	0.52	1.18	0.10	0.88	1.10	0.25
Average VV	281.7	0.65	0.22	1.88	0.48	0.78	0.06	0.53	0.81	0.16
FF	-	-	-	20	-	6	5	-	500	-
AQC (LKK)	-	200 (day)	0.1*	10	-	2	2.5	-	100	-

*Air quality criterion for Cr is valid for hexavalent chromium (Cr VI)

Air quality criteria (LKK) are defined for, a.o. V, Cr (VI), Ni, As, Cd and Pb, based on research results on effects of the compounds on human health. The air quality criteria are so low that most people can be exposed for the pollutant levels without experiencing health effects. The weekly average concentrations of all compounds were well below the respective air quality criteria, defined for annual average values. The blue line in the panel showing chromium results in Figure 9 illustrates the air quality criterion for hexavalent chromium, which only represents a small fraction[12] of the total chromium measured. The analytical method does not distinguish between the valence states Cr III and Cr VI. The measurements were carried out during a 3-month period in summer, when highest concentrations are expected in Sunndalen. The annual limit values and air quality criteria for heavy metals were most likely not violated in 2019.

[12] There is only little information in the literature regarding the Cr VI fraction. The Norwegian Institute of Public Health (2013) indicates a fraction below 10%, measured in Mo i Rana.

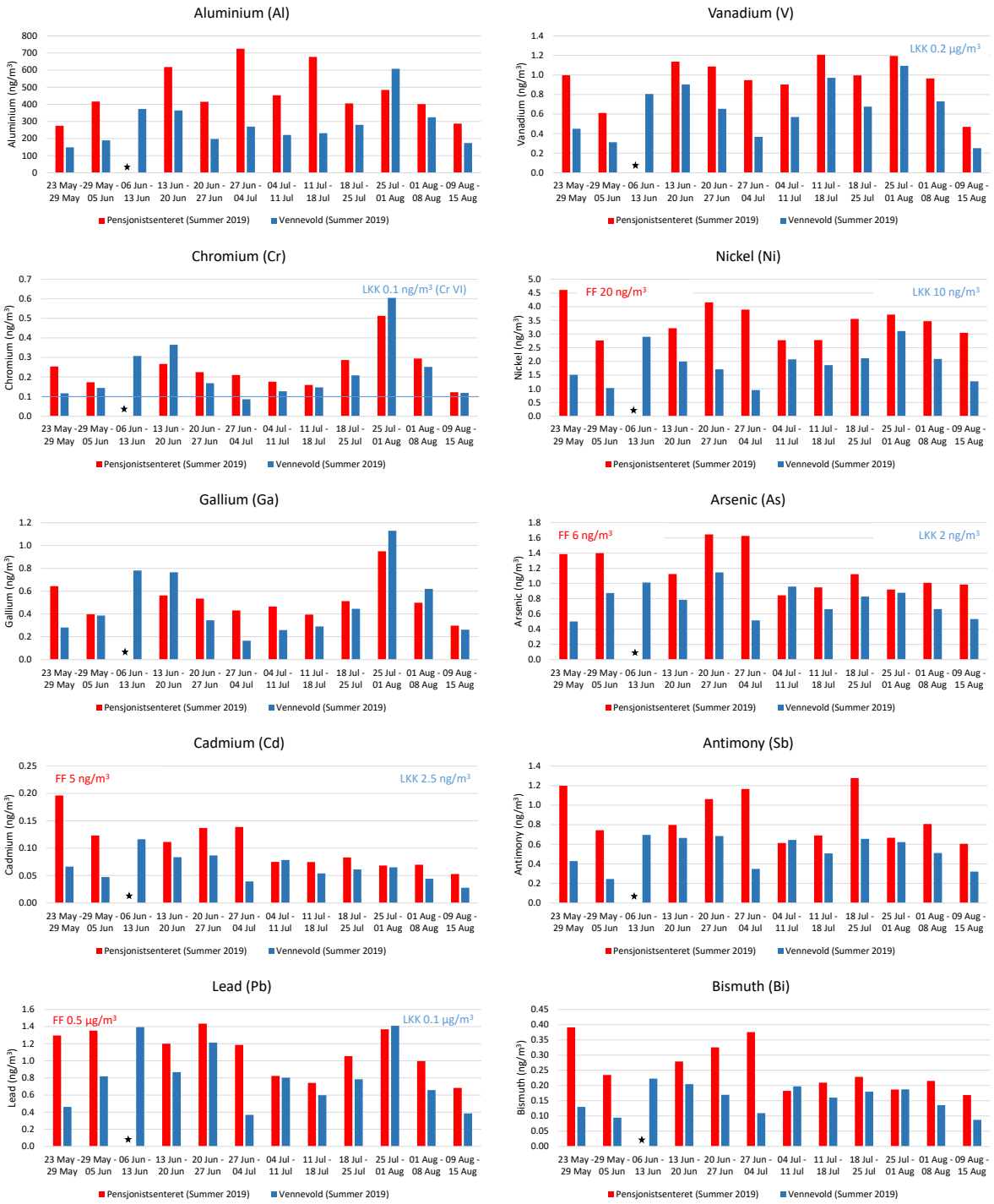


Figure 9: Weekly average concentrations of heavy metals (and aluminium) in summer 2019 (23. May – 15. August 2019) at Pensjonistsenteret and at Vennevold. The order of compounds is according to their atomic mass. The third PM_{10} sample at Pensjonistsenteret was not exposed (marked by asterisk).

Comparison to previous campaigns

Heavy metals in air/particles have been included in measurement studies in 2002 and 2006 (see Table 2). In Figure 10, levels of metals measured in summer 2019 are compared to results from summer 2002 and summer 2006. Gallium, antimony and bismuth were not analysed in previous

campaigns. Chromium and nickel were not analysed in 2002. For most compounds (Al, V, probably Cr, Pb), the levels went down since 2002. For nickel, the level is unchanged since 2006, at both sites. Arsenic levels were lower in 2006, compared to 2002, but are slightly higher in 2019 than in 2002 even though the PM_{10}-levels are reduced compared to earlier measurements.

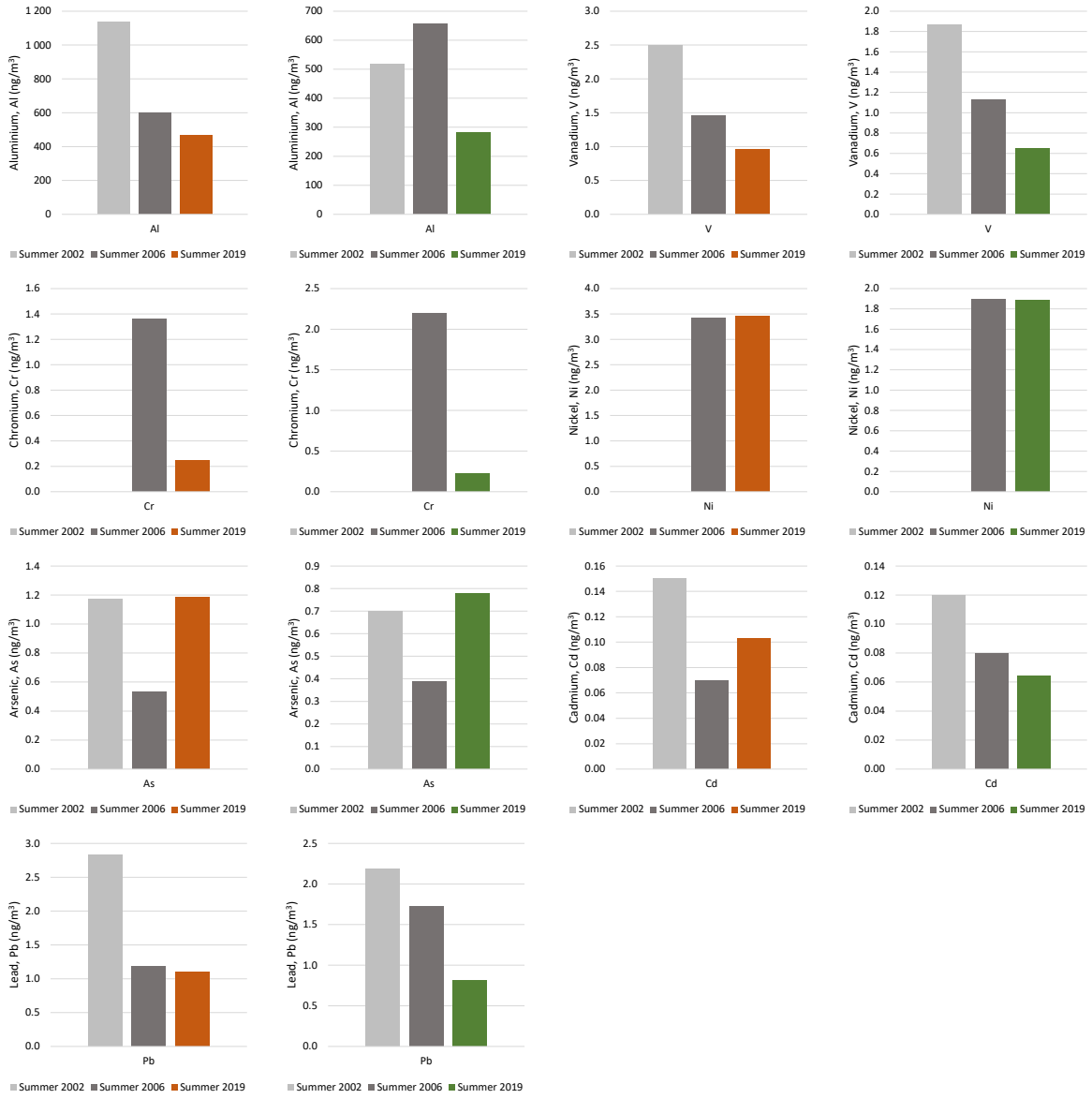


Figure 10: Average concentration of heavy metals in summer 2002 (10 weeks), summer 2006 (9 weeks) and summer 2019 (12 weeks) at Pensjonistsenteret (red) and at Vennevold (green).

In 2002 and 2006, measurements were carried out on 20 days distributed over 3 months. These results are more sensitive to time variations and not directly comparable to long-term mean values over 3 months.

Emissions of several metals are reported to the responsible authorities (see Figure). For nickel, the highest emissions were reported before the conversion of the smelter in 2003/2004. In 2002, when high emission was reported, nickel was not analysed. Today's (2019) nickel emission is at the same level as in 2006. Nickel concentrations were the same in 2019 as in 2006 at both sites. For chromium

high emission was reported for 2002. Emission in 2006 was strongly reduced and remained around the same level until 2019. However, chromium concentrations in 2019 were significantly lower than in 2006. For lead, higher emissions were reported 2002 than 2006, but also higher emissions were reported in 2019 compared to 2006. The lowest measured concentrations were observed in 2019. For vanadium, reported emission in 2002 was slightly higher than in 2006. Since 2009 no vanadium emissions are reported. Measured concentrations have decreased in the period 2002-2019. Arsenic emissions reported have been varying between 2000 and now and have increased recently. No decrease has been observed in measured concentrations. For cadmium, emissions reported in 2002 were higher than in 2006 and 2019. In 2019 higher Cd emissions than in recent years were reported. The highest ambient cadmium concentrations were observed in 2002. At Pensjonistsenteret, the Cd level in 2019 was higher than in 2006.

Comparison to Norwegian background

Heavy metals in air and aerosol are not measured at Kårvatn[13]. The closest Norwegian background site for which results for heavy metals in air and aerosol are reported, is Birkenes (58°23' N, 8°15' E, 219 masl) in Southern Norway, in addition to Andøya and Zeppelin in the Arctic. The same analytical method is used for samples from Birkenes and Sunndal.

In Table 6, average concentrations of heavy metals at Pensjonistsenteret and Vennevold during the measurement period are compared to concentrations of those compounds measured at Birkenes during the period May – August 2019 and to the annual averages for 2019 at Birkenes. Vanadium, nickel, arsenic, cadmium and lead show elevated concentrations compared to background values, while the chromium concentrations are higher at Birkenes compared to the measurement sites in Sunndalen. This indicates a long-range transport contribution of chromium, e.g. from the European continent, which is more pronounced at Birkenes compared to the study area. The remaining compounds are not analysed at Birkenes.

Table 6: Average metal concentrations (ng/m^3) at Pensjonistsenteret (PS) and Vennevold (VV) during the measurement period (23. May – 15. August 2019). Average concentrations of metals in air at Birkenes for the period May – August 2019 and annual average at Birkenes for 2019. Unit: ng/m^3.

ng/m^3	Al	V	Cr	Ni	Ga	As	Cd	Sb	Pb	Bi
Average PS	468.7	0.96	0.24	3.45	0.52	1.18	0.10	0.88	1.10	0.25
Average VV	281.7	0.65	0.22	1.88	0.48	0.78	0.06	0.53	0.81	0.16
Birkenes mai-aug	-	0.42	0.31	0.21	-	0.12	0.018	-	0.50	-
Birkenes ann.avg.	-	0.32	0.29	0.18	-	0.14	0.028	-	0.65	-

[13] At Kårvatn, heavy metals in precipitation are measured.

The air concentrations of metals at Birkenes and Andøya were slightly lower or equal in 2019 compared to 2018. At Lista[14]/Birkenes, there has been a significant reduction in air concentrations for all the measured elements for the period 1991 to 2019. The reductions were: lead 88%, cadmium 69%, arsenic 69%, chromium 82%, nickel 76%, and vanadium 93% (Bohlin-Nizzetto et al., 2020).

In general, the concentrations of most heavy metals in air at Birkenes in 2019 are 2-3 times higher than those observed at Andøya and Zeppelin. This is because Birkenes is closer to the emission sources at the European continent (Bohlin-Nizzetto et al., 2020), than the arctic stations. Sunndalsøra is located in between Birkenes and Andøya. The influence from the European continent is considered much lower, compared to local sources.

Moss surveys

Hydro Sunndal participated in the four moss surveys around Norwegian industries (2000, 2005, 2010 and 2015). In the most recent study (Steinnes and Uggerud, 2017), moderately elevated levels (i.e., factor 10 above background) were observed for Ti, V, Ni, Ga and Bi. These levels are reported to have stayed relatively constant since 2000 (Steinnes and Uggerud, 2017). Samples were collected at 5 sites (see Figure 11), where site 4 and site 7 are closest to the aluminium smelter and show the highest levels. Site 7 is close to Sunndalsøra and site 8 is some km up the valley, towards Vennevold. Similar to the present study, metal levels are lower at site 8 compared to site 7, which is one of the two most contaminated sites in the moss survey.

Segments of the moss plants corresponding to the last 3 years' growth are selected for chemical analysis, i.e., the spatial distribution of the metals is a 3-year-average dominated by the growth season. At the same time, the growth season is the season with highest expected pollution at Sunndal.

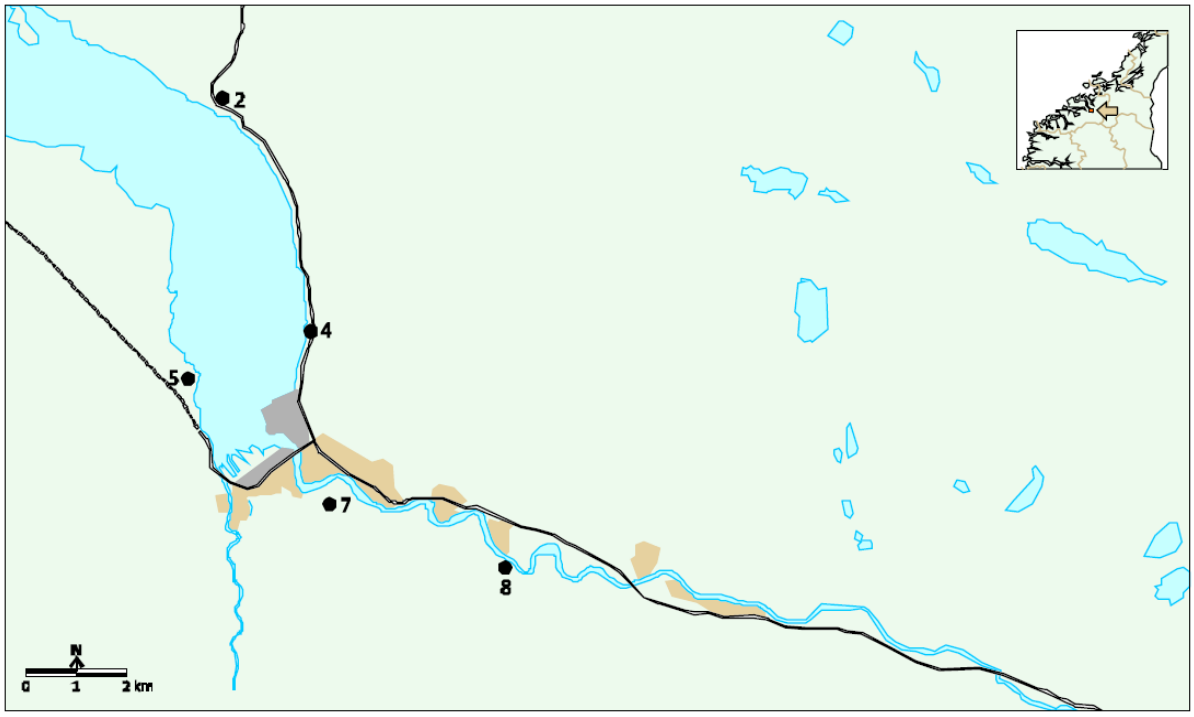


Figure 11: Moss sampling sites around Hydro Sunndal in 2015. Source: Steinnes and Uggerud (2017).

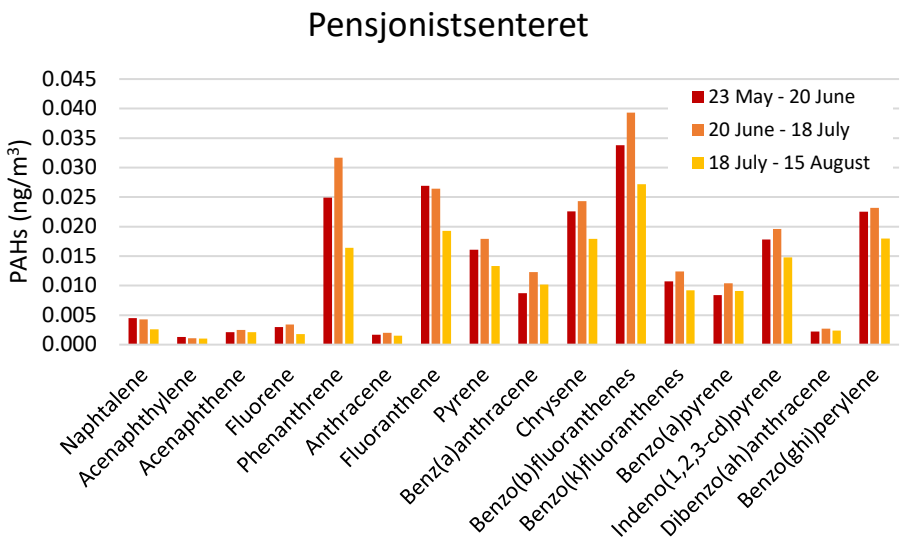
[14] The regional background station at Lista was closed in 2004, observations continued at Birkenes.

In the 2015 moss study, appreciable Ni deposition was observed around Hydro Sunndal. Al deposition is high around aluminium smelters. Out of the four aluminium smelters participating in the 2015-study, Sunndal had the highest Al deposition (followed by Årdal, Lista and Husnes).

3.3 Polycyclic aromatic hydrocarbons – PAHs

Weekly samples of particle-bound PAHs were collected at both sites, Pensjonistsenteret and Vennevold, and merged to monthly (i.e., 4 weeks) samples. The monthly samples were analysed for the 16 priority PAHs[15].

PAH profiles of the 16 priority PAHs in the measurement period are shown in Figure 12 for both sites. The profiles vary very little from month to month and are also very similar comparing the two stations. It needs to be underlined that exclusively particle-bound[16] PAHs were analysed in summer 2019. Compounds which mainly appear in gas form, like naphthalene, acenaphthylene, acenaphthene and fluorene were thus detected in very low concentrations. Benzo(a)pyrene is the only PAH with a target value. Benzo(a)pyrene is mainly particle-bound in all seasons. Phenanthrene, fluoranthene and pyrene are PAH-compounds which are connected with emissions from the aluminium industry and were prominent in the profiles observed. Also benzo(b)fluoranthene is seen as indicator for aluminium production as PAH source (Aubin and Farant, 2000).



[15] EPA 16 PAH: US EPA (1982) Determination of polynuclear aromatic hydrocarbons in industrial and municipal wastewaters. Cincinnati, U.S. Environmental Protection Agency, Environmental Monitoring and Support Laboratory (EPA-600/4-82-025).

[16] There were both analytical and economic reasons for analysing only particle-bound PAHs and not gaseous PAHs in the monthly samples.

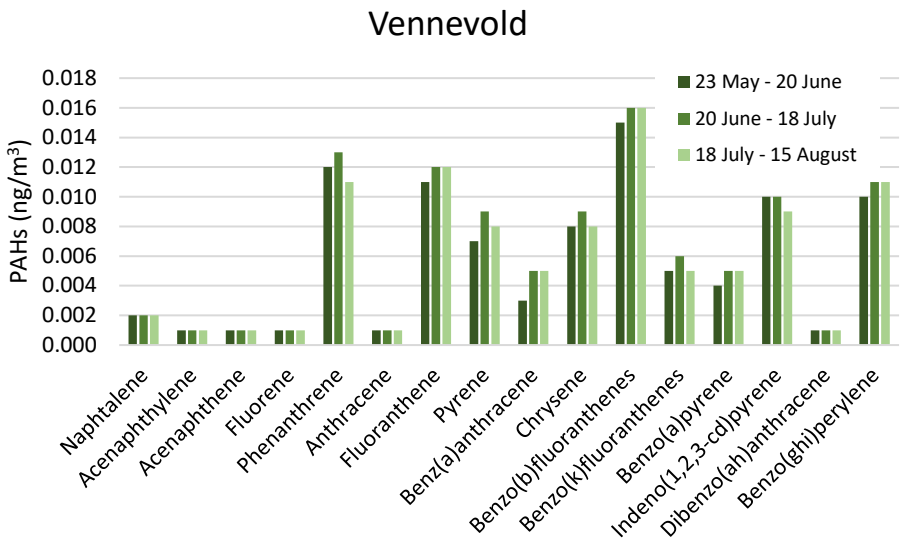


Figure 12: Monthly average profiles of particle-bound PAHs in summer 2019 at Pensjonistsenteret (upper plot) and Vennevold (lower plot).

The levels of all compounds were higher at Pensjonistsenteret than at Vennevold, which is several kilometres further away from the smelter (Figure 13). For most compounds, the concentration ratio was about a factor 2. The PAH profile at Vennevold was very similar to the PAH profile observed at Pensjonistsenteret, indicating that the same PAH sources influence both sites.

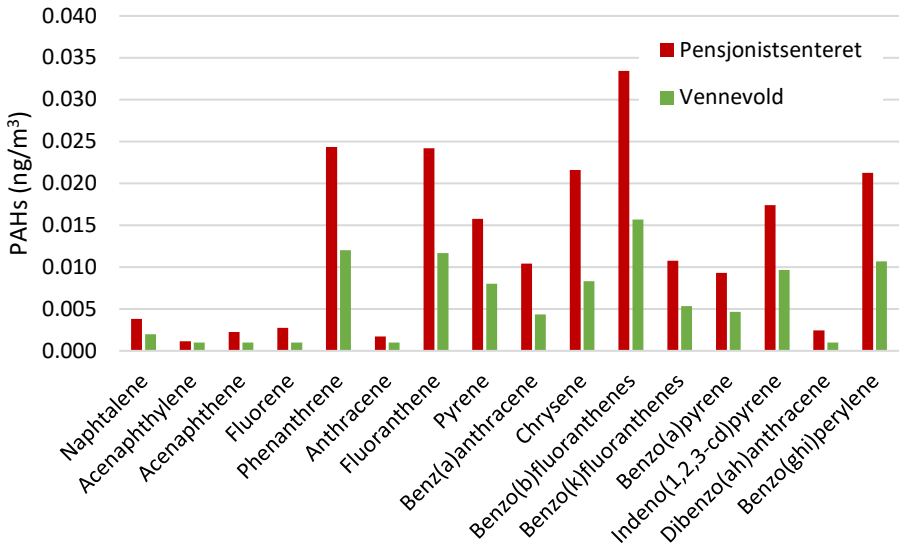


Figure 13: Average profiles (23. May – 15. August 2019) of particle-bound PAHs in summer 2019. Comparison of concentrations at Pensjonistsenteret and at Vennevold.

PAH-profiles from the present study cannot be compared to PAH-profiles from earlier measurements or PAH-profiles at background locations. Here, it needs to be taken into account that PAH-profiles from background sites and most earlier studies consist of both gaseous and particle-bound PAHs, while the

PAH-profiles from the present study consist of particle-bound PAHs. The concentrations of lighter PAHs (from naphthalene to pyrene) are therefore underestimated.

Benzo(a)pyrene occurs almost exclusively in particle phase. Quantitative comparison to limit values, background sites and previous measurements can thus be carried out.

Comparison to limit values

The target value for BaP is 1 ng/m^3 averaged over a calendar year. The monthly average values in summer 2019 were two orders of magnitude lower. Even if one expects higher ambient PAH levels in winter, due to e.g. residential heating, the target value is not expected to be exceeded as a consequence of emission from aluminium production at any of the two sites. Also, the air quality criterion for BaP in Norway (annual average 0.1 ng/m^3) is not expected to be exceeded.

Comparison to Norwegian background

The levels of environmental contaminants in air and precipitation, including PAHs, at Norwegian background sites is monitored continuously and reported yearly. For 2019, background concentrations at Birkenes[17] in Southern Norway and at Zeppelin in the Arctic are reported by Bohlin-Nizzetto et al. (2020). The monitoring program commissioned by the Norwegian environment agency covers 32 PAHs (including the 16 priority PAHs) in gas and particle phase. The most abundant PAHs at Birkenes were phenanthrene (0.7 ng/m^3, 37% of PAH_{16}), followed by dibenzofuran (0.5 ng/m^3, not part of PAH_{16}), fluorene (0.4 ng/m^3, 21% of PAH_{16}) and fluoranthene (0.2 ng/m^3, 9% of PAH_{16}). At Zeppelin, the volatile PAHs are more abundant. A strong seasonality is observed for all PAHs at Birkenes with a factor of 3 higher concentrations in wintertime (November – March) than in summertime. The monthly concentrations of sum PAH_{16} (gaseous and particle-bound) at Birkenes in the period May – August 2019 ranged between 1.0 and 1.9 ng/m^3. Monthly levels of particle-bound 16 EPA-PAHs at Pensjonistsenteret in the measurement period ranged between 0.17 and 0.23 ng/m^3. At Vennevold, the variation was between 0.09 and 0.10 ng/m^3. The annual mean concentration at Birkenes in 2019 (2.0 ng/m^3 for sum PAH_{16}) was lower than in 2018 and higher than in 2017, showing fluctuating concentrations over the last years (Bohlin-Nizzetto et al., 2020). PAH-observations at Birkenes started in 2009. The levels of benzo(a)pyrene at Birkenes are 2-3 orders of magnitude below the European air quality standard (1 ng/m^3).

As mentioned above, PAH-profiles in Sunndalen cannot be compared to background profiles. When comparing PAH profiles, it needs to be taken into account that PAH-profiles from the background site Birkenes consist of both gaseous and particle-bound PAHs, while the PAH-profiles from the present study consist of particle-bound PAHs. Due to the same reason, also sum PAH_{16} levels cannot be compared.

The levels of benzo(a)pyrene, which occurs almost exclusively in the particle phase, can be compared. The average concentration of B(a)P at Birkenes in the period May – August 2019 was 0.006 ng/m^3. At Pensjonistsenteret and Vennevold, the average B(a)P concentrations were 0.009 ng/m^3 and 0.005 ng/m^3, respectively.

[17] Birkenes in southern Norway is located ca. 480 km south of Sunndalsøra

Comparison to previous campaigns

For comparing the results from 2019 to earlier measurements, several differences in the measurements have to be noted:

- In 2019, weekly samples were merged to monthly samples. In 2002 and 2006, daily samples (24 hours) were analysed. This means that the air volume sampled was much lower than in the monthly samples, which has an effect on the detection limits, i.e., lower detection limits for weekly and monthly samples compared to daily samples. The daily samples for the earlier studies were collected twice a week and thus did not continuously cover the measurement periods. In addition, daily results are more sensitive to time variations and are not directly comparable to long-term mean values over 3 months.
- In 2019, particle-bound PAHs were analysed, while in 2002 and 2006, both particle-bound and gaseous PAHs were analysed. Some of the lighter PAHs are mainly gaseous (discussed above). The PAH-profiles from earlier studies will therefore differ from the PAH-profile from 2019.
- In 2019, PAH samples were analysed for 16 priority PAHs (EPA 16 PAHs). In 2002 and 2006, 33 PAHs were analysed. For comparison, the 16 priority PAHs were selected. Modelling results (OR46/91) for the situation in 1990 and 1997 show the distribution of PAHs without indicating which compounds are included in the calculations.

In previous campaigns, PAHs were only sampled at Pensjonistsenteret. Compared to previous campaigns at Sunndal, PAH-concentrations in summer 2019 were considerably lower (upper plot in Figure 15). The compounds with highest concentrations in 2002 (fluorene, phenanthrene, fluoranthene, pyrene and benzo(b,k)fluoranthenes) had decreased distinctly in 2006, with the exception of phenanthrene. It needs to be recalled that the PAH-profiles from 2002 and 2006 contain both gaseous and particle-bound PAHs. Since fluorene, phenanthrene, fluoranthene and pyrene occur mainly in gaseous form, it is natural that values in the 2019 profiles of particle-bound PAHs are lower than in previous campaigns.

Since the decline of concentrations between 2002 and 2019 is rather large, the percent contribution of each compound to PAH16 is compared, see lower plot in Figure 15. While in 2002, fluorene, phenanthrene, fluoranthene, pyrene and benzo(b,k)fluoranthenes (apart from benzo(b,k)-fluoranthenes these compounds occur mainly in gaseous form) were most prominent, the compounds standing out in the PAH-profile of 2019 are phenanthrene, fluoranthene, pyrene, chrysene, benzo(b)fluoranthene. The figure is only an illustration, but cannot be used for comparing, since the 2019 profile is biased by not including gaseous compounds.

Benzo(a)pyrene, which almost exclusively occurs in the particle phase, has decreased strongly over time. In summer 1991, the PAH-level at Sunndalsøra was reported to be 54% lower than in summer 1981 (Hagen, 1992). The benzo(a)pyrene concentration in the period June – August 1991 was 2.0 ng/m^3. As shown in Figure 14, the BaP concentration at Pensjonistsenteret was higher in summer 2002. After the technical conversion of the smelter in 2003/04, ambient PAH concentrations were strongly reduced. In summer 2006, a BaP-concentration of 0.16 ng/m^3 was measured at Pensjonistsenteret. In summer 2019, an even lower BaP concentration was measured, 0.009 ng/m^3.

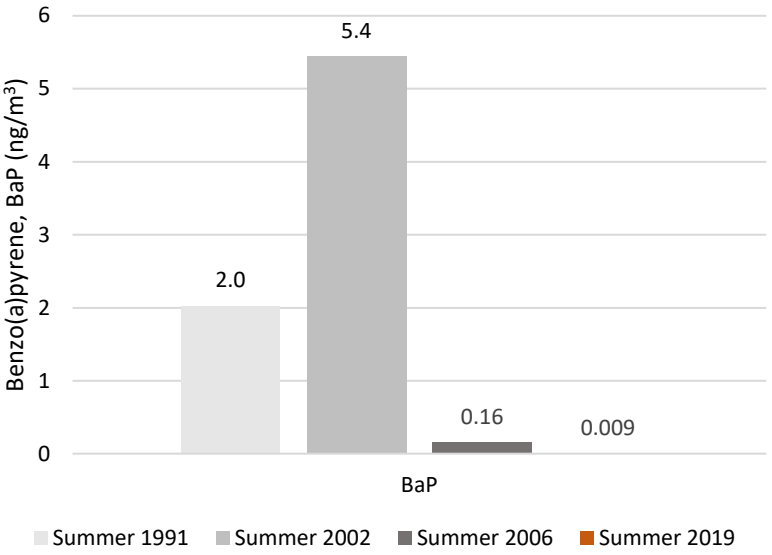


Figure 14: Average benzo(a)pyrene concentration in Sunndalsøra in summer 1991 (10 weeks), summer 2002 (10 weeks), summer 2006 (9 weeks) and summer 2019 (12 weeks).

Emissions of PAHs are reported to have decreased strongly, especially between 2002 and 2006, as a consequence of a change in production away from Söderberg technology, even though the production has more than doubled (see Figure). Söderberg technology was strongly associated with PAH emission due to the construction of the anode that consisted of coke and anthracite aggregates bound together with coal tar pitch. PAH-emissions decreased further between 2006 and 2019. The decrease of emissions seems to be reflected in the upper plot in Figure 15. However, for phenanthrene, the decrease between 2002 and 2006 is very low. For all 16 PAH compounds, the lowest values were observed in summer 2019.

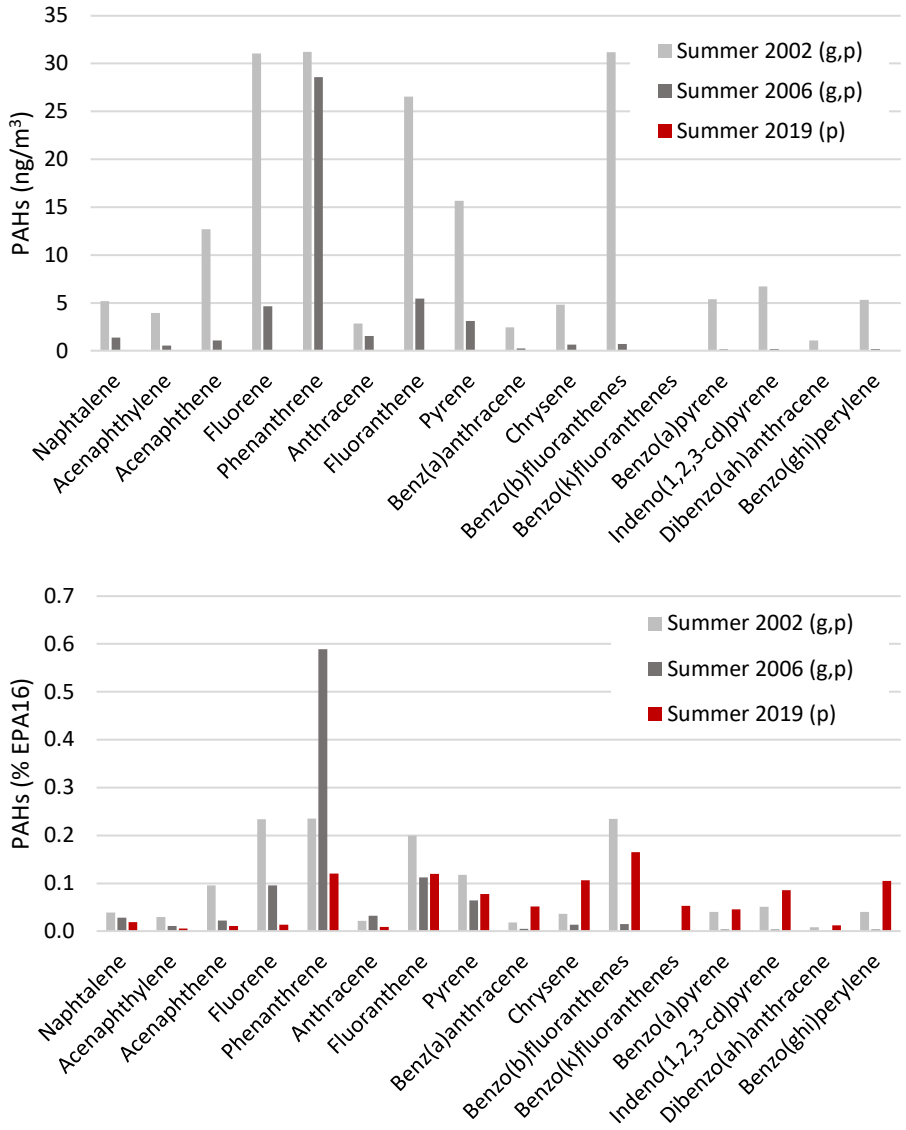


Figure 15: Average profiles of PAHs in summer 2002 (10 weeks), summer 2006 (9 weeks) and summer 2019 (12 weeks, particle-bound PAHs) at Pensjonistsenteret. In 2002 and 2006, no PAH measurements were performed at Vennevold. (Upper plot) PAH concentration profiles, (lower plot) profiles of the percentage of each compound to PAH16. Note that in the earlier studies, benzo(b)fluoranthene and benzo(k)fluoranthene were reported as group benzo(b,k)fluoranthenes.

For comparison, an earlier modelling study (OR46/91, Bøhler and Larsen, 1991) carried out dispersion calculations for the summers of 1990 and 1997 (i.e., after an expansion planned for 1997[18], consisting of building two new halls, SU4, at the same time as SU1/2 is closed). For 1990 emission conditions, PAH[19] summer averages of over 500 ng/m^3 700-800 m from the smelter were calculated, and several km up the valley the calculated concentration was below 100 ng/m^3. After the expansion, i.e., in 1997, PAH emissions were estimated to be 50% reduced, resulting in summer averages over 300 ng/m^3 up to 500 m from the smelter. PAH emissions in 1990 come from the cleaning facilities of the anode baking

[18] It appears that the expansion planned for 1997 was realised in 2003/04.
[19] It is uncertain, which PAH compounds are covered by the term "PAHs" in the modelling study.

plant and green anode plant and as roof emissions from SU1/2. After closing SU1/2 and launching the new halls in 1997, low PAH-emission was expected.

Moss survey

In the survey of PAHs in naturally growing moss collected around industrial enterprises in Norway in summer 2015 (Halse et al., 2017), atmospheric deposition of PAHs on moss was studied. In the same study also passive PAH air samplers (PAS) were placed around some of the enterprises, including Hydro Sunndal. The PAS at Sunndal were exposed for one month in summer/autumn 2015, from 28th July 2015 to 18th August 2015 at 5 locations upwind and downwind from the industrial site (see map in Figure 16). One pooled sample of moss from 50 x 50 m^2 was collected close to the industrial site in Sunndal. The location of the moss sampling site (see Figure 16) was selected in order to best characterise the local deposition pattern with respect to topography and dominant wind directions relative to the location of the industrial site. It was about 1 km from the smelter.

Figure 16: Distribution of passive air samplers (red circles) around Hydro Aluminium Sunndal and location for moss sampling (orange star). Source: Halse et al. (2017).

The lighter PAHs (2-3 benzene rings) appear solely in the gas phase, and are easily re-emitted from secondary sources as well as prone to long-range transport (LRT). Heavier PAHs (>5 benzene rings) are more attached to particles, and thereby less mobile and prone to be deposited close to source regions. Consequently, it appears likely that heavier PAHs are found in moss, while the lighter ones are collected by the air sampler (Halse et al., 2017). Moss and air samples together provide comprehensive information regarding the spatial distribution of PAHs around industrial sites.

Halse et al. (2017) present results from the passive air samplers as ng/sample for eight selected PAHs, PAH8 (fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benz(a)anthracene, chrysene and

benzo(a)pyrene). The highest PAH levels in passive air samples around the 6 selected industries were found at Sunndal (the other industries participating in the study were located at Kristiansand, Mosjøen, Orkanger, Ålvik and Hemne).

PAH air samples were collected at 5 locations around Hydro Sunndal. At all locations, the sum of PAH8 was detected with higher amounts compared to the background station at Kårvatn (Halse et al., 2017). The highest amount of the sum of PAH8 was found at Oppdøl (north-northwest of the industrial facility; 1687 ng/sample), followed by Nylykkjebekken (4 km east-southeast up the valley; 1216 ng/sample) and Blakåsbukta (at the fjord, 3 km northwest; 1040 ng/sample). The most abundant PAHs were the lighter PAHs with 2-4 rings, e.g. phenanthrene, fluorene, fluoranthene and acenaphthene. Phenanthrene contributed with 57% on the average loading of sum of 8 PAHs, while fluorene and fluoranthene contributed with 23% and 13%, correspondingly. Acenaphthene contributed with 8% to the sum of PAH8. Comparing the findings in the air samples with the background reveals that phenanthrene and fluorene were the most abundant components, but at a lower level. Heavier PAHs such as 5- to 7-ring PAHs are to a lesser extent detected in the sampler, since they are often associated to particle phase which is not effectively sampled by the PAS (Halse et al., 2017). Other sources than the industrial site may contribute to levels of PAHs collected by the air samplers.

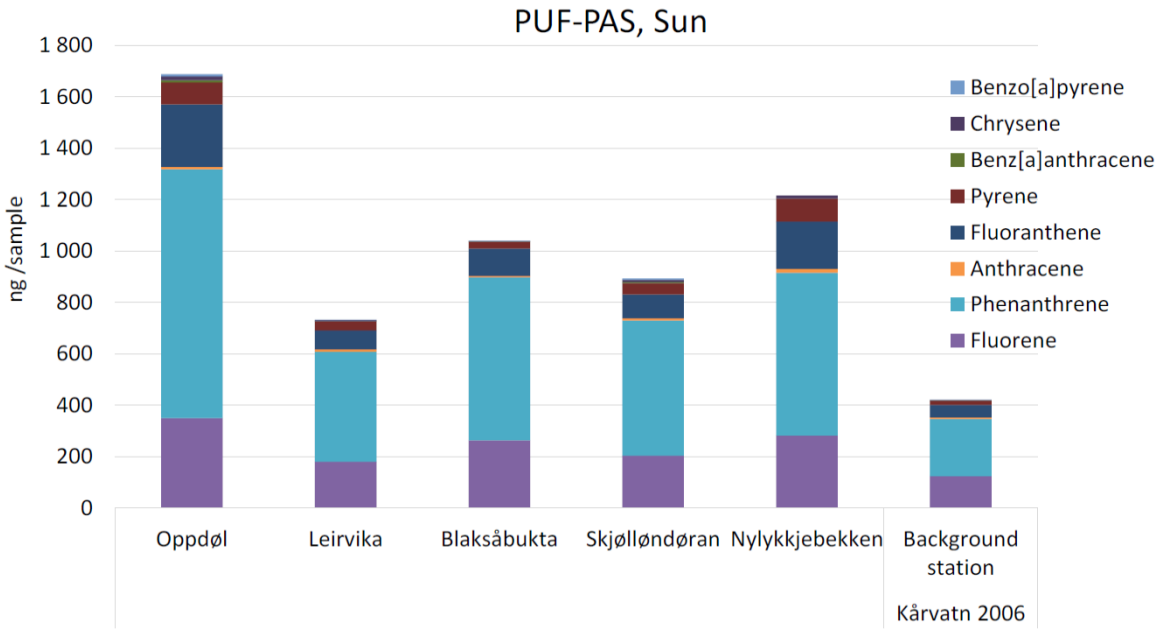


Figure 17: Distribution of selected PAHs (PAH8) in PUF-PAS deployed in the vicinity to Hydro Aluminium Sunndal. Source: Halse et al. (2017).

Generally, the PAH levels determined in moss collected around industrial sites in the moss survey were considerably higher than the PAH levels found in moss collected at background sites, especially for the heavier PAHs (phenanthrene, fluoranthene, pyrene, chrysene, benzo(b)fluoranthene and benzo(a)pyrene). The moss sample from Sunndal was detected with the second highest PAH levels when compared to the other industrial sites included in the moss survey. The sample was analysed for 16 priority PAHs.

The moss sample from Sunndal collected in summer 2015 showed high levels (1825 ng/g dw for sum PAH16) compared to background sites. Benzo(b)fluoranthene (13% of sum PAH16), fluoranthene (12% of sum PAH16), benzo(a)pyrene (12% of sum PAH16) and chrysene (11% of sum PAH16) were the most contributing PAH components. The lighter PAHs were detected at the lowest levels, with acenaphthylene, naphthalene and acenaphthene all contributing to < 1% to the sum PAH16. A comparison of the results from Sunndal with the findings for moss samples collected at the closest background site (Godøy) clearly shows elevated levels in the moss sample collected at Sunndal. The levels were considerably higher for, e.g., phenanthrene, fluoranthene, pyrene, benz(a)anthracene, chrysene, benzo(b)fluoranthene and benzo(a)pyrene (Halse et al., 2017).

The compounds found to be most abundant in the moss sample from Sunndal were the same compounds which were most abundant in the filter samples collected in summer 2019. Since the sampling techniques are different, it is difficult to compare the PAH-profiles.

3.4 Sulphur dioxide – SO_2

Weekly samples of SO_2 were collected at both sites in summer 2019. The sampling periods were identical at Pensjonistsenteret and Vennevold.

SO_2-concentrations were higher at Pensjonistsenteret, which is located very close to the aluminium smelter, than at Vennevold, which is located several kilometres up the valley (Figure 18). The ratio between concentrations at the two sites varied between the sampling periods, either due to changes in meteorology or variation in the emissions. Weekly concentrations at Pensjonistsenteret varied between 0.7 µg/m^3 and 1.8 µg/m^3, at Vennevold weekly concentrations varied between 0.3 µg/m^3 and 0.75 µg/m^3.

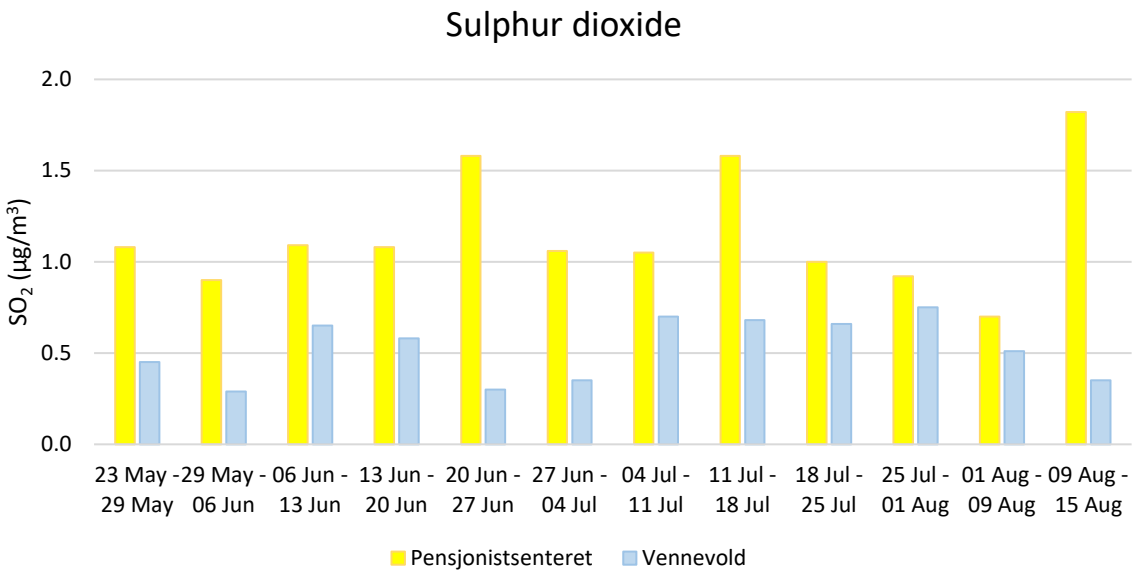


Figure 18: Weekly average concentrations of sulphur dioxide in summer 2019 (23. May – 15. August 2019). Comparison of concentrations at Pensjonistsenteret and at Vennevold.

Comparison to previous campaigns

The 3-month average concentration of SO_2 was 1.2 µg/m^3 at Pensjonistsenteret and 0.5 µg/m^3 at Vennevold. SO_2-levels have decreased between 2002 and 2006 and between 2006 and 2019 (see Figure 19). The emissions of SO_2 are reported to have decreased, especially between 2002 and 2006, even though the production has increased (see Figure above). The sampling method used was the same as in 2002 and 2006 and the same analysis principle was used. In 2002 and 2006, measurements were carried out on 20 days distributed over 3 months. These results are more sensitive to time variations than weekly samples.

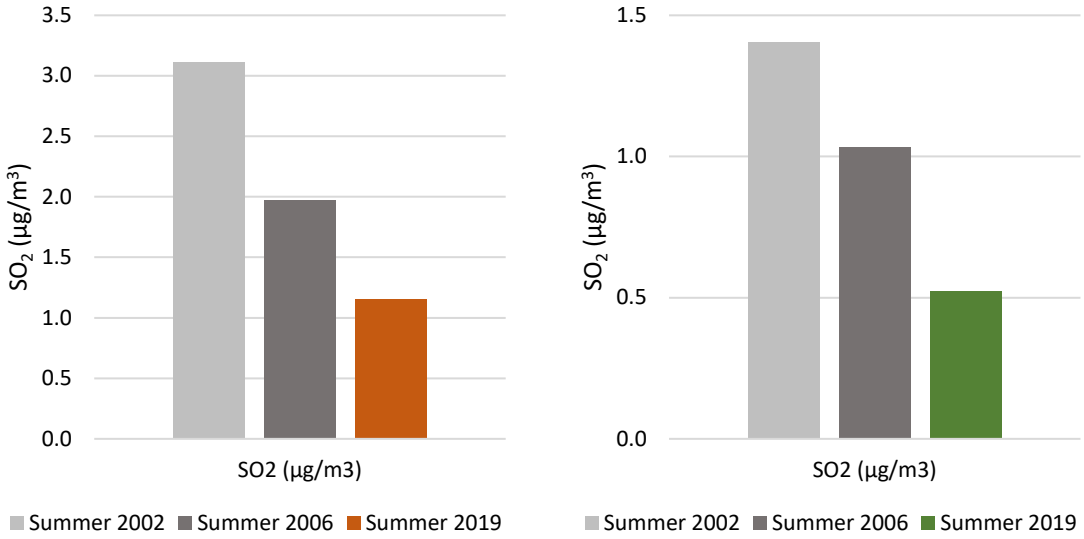


Figure 19: Average concentration of sulphur dioxide in summer 2002 (10 weeks), summer 2006 (9 weeks) and summer 2019 (12 weeks) at Pensjonistsenteret and at Vennevold.

For comparison, in an earlier modelling study (OR46/91, Bøhler and Larsen, 1991) results from dispersion calculations for summer 1990 and summer 1997 (after a planned expansion, closing down SU1 and SU2 and opening two new oven halls SU4) for the centre of Sunndalsøra showed around 10 µg/m^3 as a half-year average over summer 1990 and 5-10 µg/m^3 in summer 1997. The expansion of the smelter was estimated to lead to a little increase of SO_2-emissions and to a broader distribution of concentrations close to the smelter in 1997. In 1990, the largest SO_2 emissions were estimated to arise from the cleaning facilities for SU1/2 and SU3. The largest SO_2 emissions over the roof were estimated to come from SU1/2.

Comparison to limit values

SO_2-concentrations measured around Sunndal in the summer, which is the time of the year with the highest air pollution in Sunndalen, were lower during the 3-month measuring period than the annual limit value (20 µg/m^3 for calendar year, see Table 3). The measurement results indicate that there was little probability of violating international and national limit values. Recommended air quality criteria and WHO air quality guidelines exist for daily averages and 15 minute averages and cannot be evaluated from weekly samples.

Comparison to Norwegian background

Annual mean concentrations of SO_2 in air are quite low at the Norwegian background sites (Aas et al., 2020). At Kårvatn, close to Sunndalsøra, the annual mean of SO_2 in air is reported 0.03 µg S/m^3 in 2019. At Birkenes, levels were a bit higher, 0.07 µg S/m^3. These levels correspond[20] to SO_2 concentrations of 0.06 µg/m^3 and 0.14 µg/m^3 at Kårvatn and Birkenes, respectively. In the time period May – August 2019, average SO_2 concentration at Kårvatn was 0.035 µg S/m^3. At the two sites in Sunndalen, the SO_2 level is clearly above regional background levels.

Aas et al. (2020) carried out a trend analysis for annual levels of SO_2 at background sites. At Kårvatn, a -75% reduction of SO_2-levels was observed in the period 1990-2019. At Birkenes, the reduction in the same period was even -95%. Respective background values at Kårvatn for 2002 and 2006, the years earlier measurements were carried out in Sunndalen, were 0.07 µg S/m^3 and 0.06 µg S/m^3, which is factor 2 higher than in 2019.

3.5 Fluorides

Weekly samples of gaseous and particle bound fluorides were collected at both sites in summer 2019 using the same sampler as for SO_2 (see Section 1.5). The sampling periods were identical at Pensjonistsenteret and Vennevold.

At Pensjonistsenteret, fluorides were distributed rather equally between gaseous and particle-bound fluorides over the entire measurement period (see Figure 20, upper plot). At Vennevold, there were several weeks when gaseous fluorides had a distinctly larger concentration than particle-bound fluorides (see Figure 20, lower plot).

[20] Conversion factor for µg S/m^3, as reported in Aas et al. (2020), to µg/m^3 is: SO2-S (µg S/m^3) * 2 = SO2 (µg/m^3)

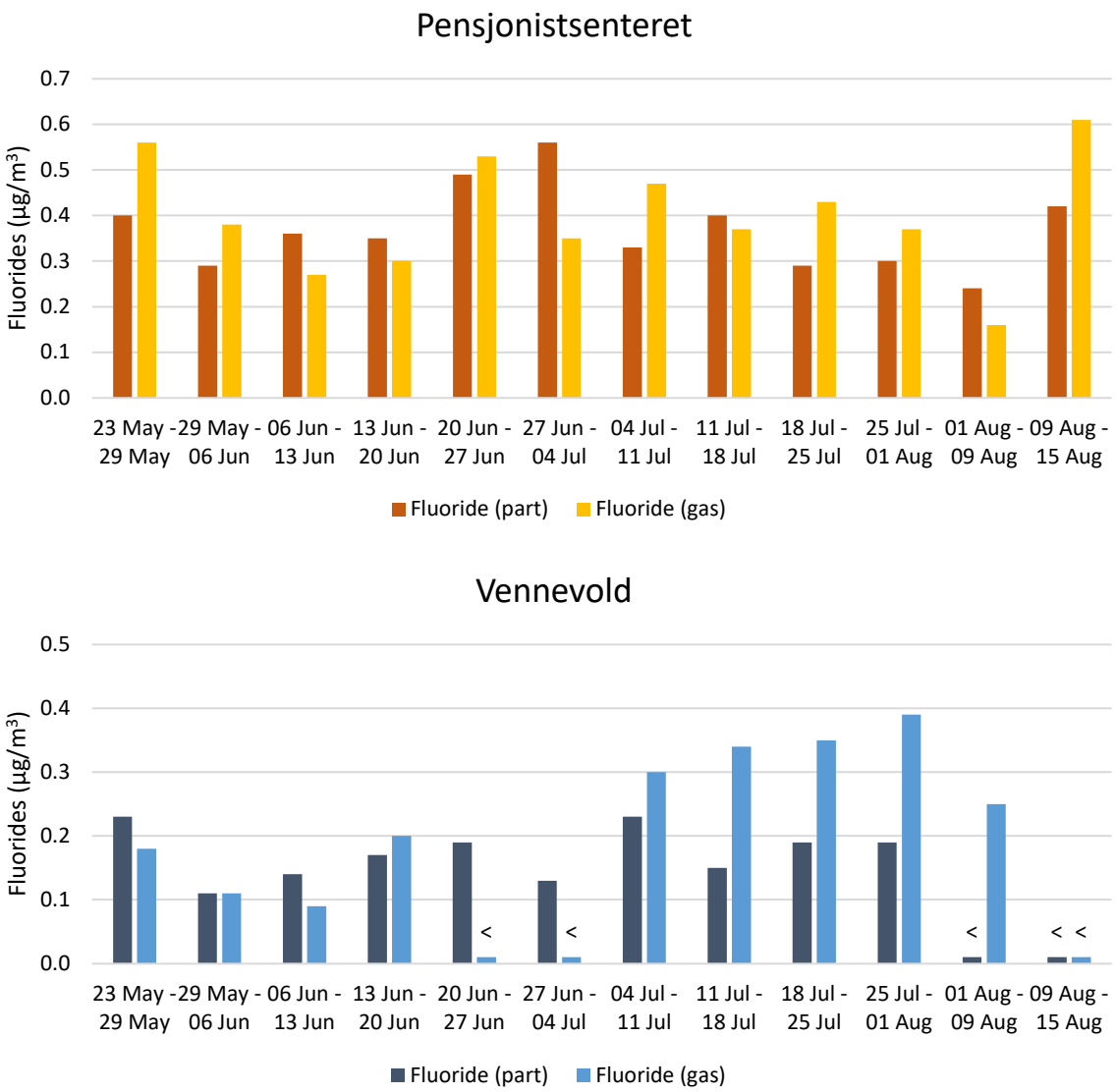


Figure 20: Weekly average concentrations of particle-bound and gaseous fluoride in summer 2019 (23. May – 15. August 2019) at Pensjonistsenteret (upper plot) and at Vennevold (lower plot). Samples with concentrations below detection limit are displayed with «<».

3.5.1 Particle-bound fluorides

Particle-bound fluorides had higher concentrations at Pensjonistsenteret, which is located very close to the aluminium smelter, than at Vennevold, which is located several kilometres up the valley (Figure 21). The ratio between concentrations at the two sites varied between the sampling periods in a similar way as for SO_2, probably due to changes in meteorology or variation in the emissions. Weekly concentrations at Pensjonistsenteret varied between 0.24 µg/m³ and 0.56 µg/m³, at Vennevold weekly concentrations varied between below 0.01 µg/m³ (detection limit) and 0.23 µg/m³. Particulate fluorides in the air around aluminium smelters occur in the size range 0.1 – 10 µm (WHO, 2000).

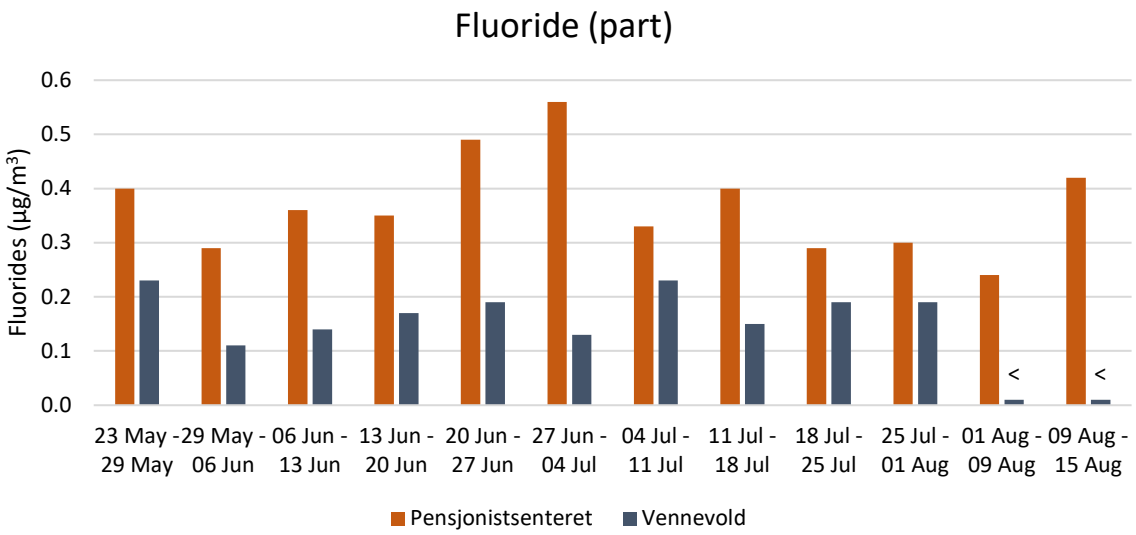


Figure 21: Weekly average concentrations of particle-bound fluoride in summer 2019 (23. May – 15. August 2019). Comparison of concentrations at Pensjonistsenteret and at Vennevold.

There is no limit value or guideline value for particle-bound fluorides.

There are no observations at Norwegian background stations for particle-bound fluorides.

Comparison to previous campaigns

The 3-month average concentration of particle-bound fluorides was 0.37 µg/m^3 at Pensjonistsenteret and 0.15 µg/m^3 at Vennevold. These levels are 24% and 42%, respectively, lower than levels of particle-bound fluorides reported for 2006 (see Figure 22). The emissions of total fluorides and the PM-emissions are reported to have decreased between 2006 and 2019, even though the production has increased (see Figure in Chapter 1.2). The sampling method used was the same as in 2002 and 2006 and the same analysis principle was used. In 2002 and 2006, measurements were carried out on 20 days distributed over 3 months. These results are more sensitive to time variations than weekly samples.

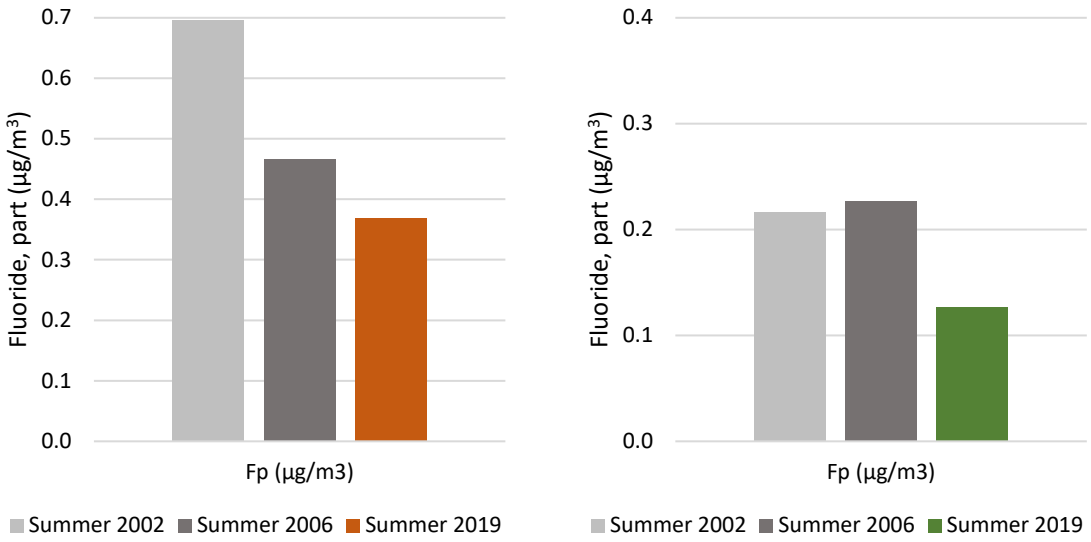


Figure 22: Average concentration of particle-bound fluoride in summer 2002 (10 weeks), summer 2006 (9 weeks) and summer 2019 (12 weeks) at Pensjonistsenteret and at Vennevold.

For comparison, results from dispersion calculations for summer 1990 and summer 1997 are presented in the modelling study by Bøhler and Larsen (1991), OR46/91. In the centre of Sunndalsøra, a half-year average over summer 1990 of around 3-5 µg/m^3 is shown for total fluoride concentrations. A summer average of ca. 0.4 µg/m^3 several km up the valley (area around Vennevold) is shown for summer 1990. After the expansion of the smelter, a reduction of the total fluoride emissions by 30% (compared to 1990) was estimated. Dispersion calculations for total fluoride for summer 1997 showed 1-3 µg/m^3 in the centre of Sunndalsøra and around 0.4 µg/m^3 several km up the valley. The expansion of SU4 and the closure of SU1/2 was estimated to lead to small changes in fluoride emissions. Emissions via the roof of SU1 and SU2 were estimated as the largest fluoride emissions at Hydro Sunndal in 1990. For the dispersion calculations it is assumed that the particles have a diameter less than 10-15 µm. If a substantial share of the particles have a diameter 30-50 µm, those will deposit close to the smelter and increase the gradient around the smelter.

3.5.2 Gaseous fluorides

The concentrations of gaseous fluorides were higher at Pensjonistsenteret, which is located very close to the aluminium smelter, than at Vennevold, which is located several kilometres up the valley (Figure 23). The ratio between concentrations at the two sites varied between the sampling periods in a similar way as for SO_2 and particle-bound fluorides, probably due to changes in meteorology or variation in the emissions. Weekly concentrations at Pensjonistsenteret varied between 0.16 µg/m^3 and 0.61 µg/m^3, at Vennevold weekly concentrations varied between below 0.08 µg/m^3 and 0.39 µg/m^3.

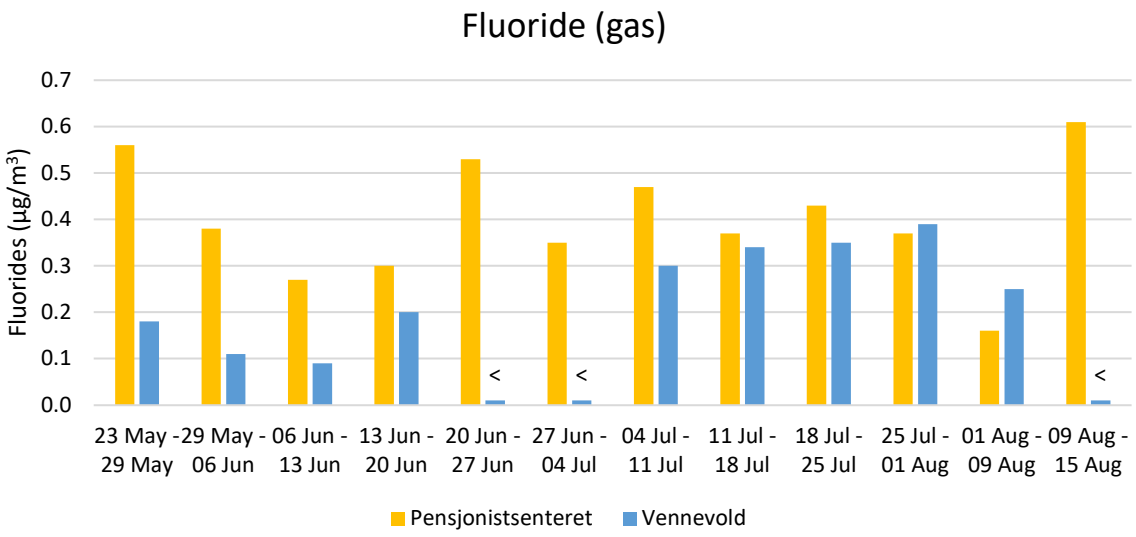


Figure 23: Weekly average concentrations of gaseous fluoride in summer 2019 (23. May – 15. August 2019). Comparison of concentrations at Pensjonistsenteret and at Vennevold.

There is no limit value or guideline value for gaseous fluorides.

There are no observations at Norwegian background stations for gaseous fluorides.

Comparison to previous campaigns

The 3-month average concentration of gaseous fluorides was 0.40 µg/m^3 at Pensjonistsenteret and 0.19 µg/m^3 at Vennevold. These levels are markedly lower than levels of gaseous fluorides reported for 2006 (see Figure 24) and little lower than in 2002. The emissions of total fluorides are reported to have decreased since 2006, even though the production has increased (see Figure in Chapter 1.2). The sampling method used was the same as in 2002 and 2006 and the same analysis principle was used. In 2002 and 2006, measurements were carried out on 20 days distributed over 3 months. These results are more sensitive to time variations than weekly samples.

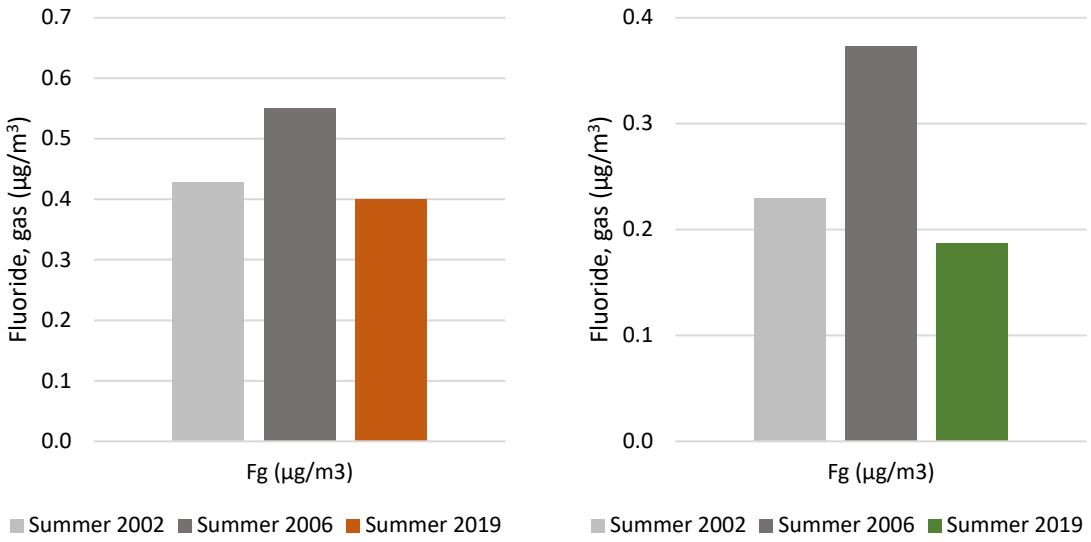


Figure 24: Average concentration of gaseous fluoride in summer 2002 (10 weeks), summer 2006 (9 weeks) and summer 2019 (12 weeks) at Pensjonistsenteret and at Vennevold.

Dispersion calculations for fluorides in Sunndalen were carried out by Bøhler et al. (1994) in order to calculate the uptake of fluorides in plants as well as the wet deposition of fluorides in Sunndalen (using the same emission data as Bøhler and Larsen (1991)). The calculated uptake is largest in July (32 mg/m^2 month at Sunndalsøra) compared to the entire year and varies depending on the vegetation type. Uptake calculated for areas around Vennevold was about half of that. Monthly average concentrations of fluoride in air calculated for the receptor Sunndalsøra for 1991 were 2.4 µg/m^3 in May, 1.94 µg/m^3 in June, 2.33 µg/m^3 in July and 2.07 µg/m^3 in August. Concentrations up towards Sunndalen calculated for July were 0.93 µg/m^3 at Furu and 0.54 µg/m^3 at Hoås. The receptors Furu (3.5 km from the smelter) and Hoås (10 km from the smelter) further up Sunndalen may represent the situation at Vennevold.

For comparison, results from dispersion calculations for summer 1990 and summer 1997 are presented in the modelling study by Bøhler and Larsen (1991), OR46/91[21]. In the centre of Sunndalsøra, a half-year average over summer 1990 around 2-3 µg/m^3 is shown for fluoride concentrations. The expansion in 1997 was estimated to lead to a 30% reduction of the HF-emission. However, the hall emissions from SU4 were assumed to give less updraught in still air than SU1/2, so that the reduction of pollution in the surrounding area was assumed to be less than 30%. Gaseous fluoride can have effects on vegetation. Dispersion calculations for gaseous fluoride for 1990 showed that 24 h averages over the guideline value for vegetation (valid at that time) may occur up to ca. 6 km from the smelter into Sunndalen. After reduction of the emissions, especially from the cleaning facility, the distance was estimated to be reduced to 4 km.

4 Discussion and conclusions

Comparison to the results from earlier studies indicate that although the production of aluminium has increased over the past 20-30 years, the emissions and the impact on the surroundings has decreased. This is valid for particulate matter, some metals (vanadium, chromium, lead), PAHs (using BaP as

[21] The study was part of an EIA for a planned expansion, which did not happen in 1997, but in 2004, and different from the original plan.

indicator), sulphur dioxide and fluorides. For some metals (nickel, arsenic, cadmium), ambient levels remained constant or increased slightly between 2006 and 2019. This development may be explained by year-to-year variation of the emission of these compounds.

The aluminium industry is the only source of fluoride to air at Sunndalsøra. The emission of fluorides reported did not decrease considerably over the past 30 years. The decrease of fluoride-concentrations in ambient air is mainly explained by the decrease of particle-bound fluoride, probably as a consequence of decreased PM-emission. Some studies in the 1990s addressed the uptake of fluorides in crops (e.g., salad, apples). These results are not discussed here.

The aluminium industry has been known to emit amounts of PAHs. By phasing out Söderberg technology at Hydro Sunndal in 2002, the emission of PAHs was strongly reduced. Remaining PAH-emissions at Hydro Sunndal are due to the production of pre-baked anodes.

The results from the present study (summer 2019) are compared to results from earlier measurements, mainly those carried out in summer 2002 and summer 2006. The sampling strategy applied in 2019 was different from the strategy in 2002 and 2006. While measurements were carried out on 20 days distributed over 3 months in 2002 and 2006, weekly samples were taken in 2019, evenly covering the entire 3-month period. The results from 24-hour samples are more sensitive to time variations compared to the measured concentrations for 7-day samples. However, 3-month averages are considered to be more comparable.

Previous measurements at Sunndalsøra (in 2002 and 2006) have shown that there was a distinct covariation between fluorides, SO_2 and PAHs, as well as metals like cobalt (Co), aluminium (Al) and nickel (Ni). Lead (Pb) and vanadium (V) showed to be partly correlated with fluorides, while copper (Cu) and chromium (Cr) were not correlated with the other compounds. No pronounced correlation was observed for these compounds in 2019. However, this may be explained by the lower variability between weekly averages (2019 study) compared to daily averages (studies from 2002 and 2006).

Hydro Sunndal appears to be the main source for aluminium, vanadium, nickel, arsenic, antimony, bismuth, PAHs, SO_2 and fluorides in the surroundings, as concentrations of these compounds were higher at Pensjonistsenteret than at Vennevold throughout the measuring period. For particulate matter, additional sources are of importance, leading to similar or slightly higher PM_{10}-and $PM_{2.5}$-concentrations at Vennevold, compared to Pensjonistsenteret.

5 References

Aubin, S., Farant, J.P. (2000) Benzo[b]fluoranthene, a potential alternative to benzo[a]pyrene as an indicator of exposure to airborne PAHs in the vicinity of Söderberg aluminum smelters. Journal of the Air & Waste Management Association, 50(12). https://doi.org/10.1080/10473289.2000.10464236

IARC, International Agency for Research on Cancer (2010) Some non-heterocyclic polycyclic aromatic hydrocarbons and some related exposures. IARC Monograph, 92. Lyon, France. http://monographs.iarc.fr/ENG/Meetings/92-pahs.pdf.

Norwegian Institute of Public Health (2013) Luftkvalitetskriterier – Virkninger av luftforurensning på helse ('Air quality criteria – Health effects of air pollution') (Report 2013:9). Oslo: Norwegian Institute of Public Health.

SFT, Statens Forurensningstilsyn (1982) Luftforurensning – virkninger på helse og miljø (SFT-rapport nr. 38).

SFT, Statens Forurensningstilsyn (1992) Virkninger av luftforurensning på helse og miljø (SFT-rapport 92:16).

WHO, World Health Organization (2000) Air quality guidelines for Europe. Second edition. WHO Regional publications, European series, No. 91

WHO, World Health Organization (2006) Air quality guidelines: Global update 2005. Particulate matter, ozone, nitrogen dioxide and sulfur dioxide. WHO Regional Office for Europe, Copenhagen.

Studies including Sunndal (period 1992 – today)

Bøhler, T., Larsen, M. (1993) Beregning av opptak og tørravsetning av fluorider i Sunndalen. In *Effektstudien. Prosjekt for effektstudier av industriutslipp fra primæraluminiumverk i Norge.* (pp. 27-30). Stabekk: Hydro Aluminium.

Bøhler, T., Larsen, M., Semb, A. (1994) Spredningsberegninger av fluorider i Sunndalen. Opptak i planter og våtavsetning (NILU OR 15/94). Kjeller: NILU.

Gjerstad, K.I. (2003) Luftkvalitet ved Hydro Aluminium Sunndal etter Su4-utbygging (NILU OR 88/2003). Kjeller: NILU.

Hagen, L.O. (1992) Kontrollmålinger av PAH i luft og nyttevekster ved aluminiumsverk sommeren 1991 (NILU OR 1/92). Kjeller: NILU.

Hagen, L.O. (2002) Kontrollmålinger av PAH i luft ved aluminiumsverk vinteren 2001/2002 (NILU OR 37/2002). Kjeller: NILU.

Hagen, L.O. (2003) Målinger av luftkvalitet ved Hydro Aluminium Sunndal i 2002 (NILU OR 63/2003). Kjeller: NILU.

Hagen, L.O. (2006) Målinger av luftkvalitet i Sunndal i 2006 (NILU OR 89/2006). Kjeller: NILU.

Halse, A.K., Uggerud, H., Schlabach, M., Steinnes, E. (2017) PAH measurements in air and moss around selected industrial sites in Norway 2015 (NILU report 23/2017). Kjeller: NILU.

Mikalsen, A., Hagen, L.O., Braathen, O.A. (1994) PAH i nyttevekster (NILU TR 4/94). Kjeller: NILU.

Schlabach, M., Steinnes, E., Uggerud, H.T. (2016) Atmospheric deposition of organic contaminants in Norway: National moss survey 2015 (Miljødirektoratet rapport, M-604/2016) (NILU report 30/2016). Kjeller, NILU).

Steinnes, E., Berg, T., Sjøbakk, T.E., Vadset, M. (2001) Nedfall av tungmetaller rundt utvalgte norske industrier. Studert ved analyse av mose (Statlig program for forurensningsovervåking, rapport 831/01) (TA-1819/2001).

Steinnes, E., Berg, T., Uggerud, H., Vadset, M. (2007) Nedfall av tungmetaller rundt norske industrier studert ved analyse av mose: Undersøkelse i 2005. Oslo, Statens forurensningstilsyn (Statlig program for forurensningsovervåking (Rapport 979/07) (TA-2240/2007). Trondheim: NTNU.

Steinnes, E., Uggerud, H.T., Pfaffhuber, K.A. (2011) Nedfall av tungmetaller rundt norske industrier studert ved analyse av mose: Undersøkelse i 2010 (Klif TA 2860/2011) (NILU OR 65/2011).

Steinnes, E., Uggerud, H. (2017) Metal pollution around Norwegian industries studied by analysis of naturally growing moss samples. 2015 survey (NILU report 1/2017). Kjeller: NILU.

Tønnesen, D. (2010) Tungmetallutslipp til luft fra aluminiumsindustrien i Norge (NILU OR 46/2010). Kjeller: NILU.

Earlier studies at Sunndal (before 1992)

Austrheim, K. (1971) Framstilling av aluminium og utslipp av fluor fra norsk aluminiumindustri (NILU OR 18/71). Lillestrøm: NILU.

Austrheim, K. (1971) Estimat for fluorutslipp som følge av aluminiumindustriens ekspansjonsplaner (NILU TR 4/71). Lillestrøm: NILU.

Bøhler, T., Larsen, M. (1991) Spredningsberegninger for utslipp til luft fra aluminiumsverk på Sunndalsøra (NILU OR 46/91). Lillestrøm: NILU.

Hagen, L.O. (1991) Kontrollmålinger av PAH i luft ved aluminiumsverk vinteren 1991 (NILU OR 42/91). Lillestrøm: NILU.

Thrane, K.E. (1983) Luftkvalitet i et boligområde på Sunndalsøra (NILU OR 1/83). Lillestrøm: NILU.

Thrane, K.E. (1984) Luftkvalitetsmålinger av svoveldioksid og fluorid på Sunndalsøra. Statusrapport (NILU OR 33/84). Lillestrøm: NILU.

Thrane, K.E. (1984) Estimat av aluminiumverkets bidrag til luftforurensning på Sunndalsøra (NILU OR 58/84). Lillestrøm: NILU.

Thrane, K.E. (1985) Luftkvalitetsmålinger av svoveldioksid og fluorid på Sunndalsøra (NILU OR 28/85). Lillestrøm: NILU.

Thrane, K.E., Aldrin, M. (1988) Fluorid i omgivelsene omkring aluminiumverket på Sunndalsøra (NILU OR 77/88). Lillestrøm: NILU.

Appendix A

Selection of metals analysed

Overview of metals included in emission reporting for Hydro Sunndal in www.norskeutslipp.no, metals Hydro Sunndal has an emission permit for, metals which are elevated around aluminium industry according to moss studies, metals which were analysed in earlier studies at Hydro Sunndal, and metals selected to be analysed in the present study.

Norskeutslipp	Emission permit	Moss studies (around industry)	Measurements 2002	Measurements 2006	This study (summer 2019)
		Ti V Ga Bi (around Sunndal)			V Ga Bi
As	As		As	As	As
Pb	Pb		Pb	Pb	Pb
Cd	Cd		Cd	Cd	Cd
Cu			Cu	Cu	
Cr	Cr		Cr	Cr	Cr
Hg	Hg				
Mo					
Ni	Ni	Ni	Ni	Ni	Ni
Zn			Zn	Zn	
V			V	V	
			Co	Co	
			Al	Al	Al
		Te Sb Be (around other Al-smelters)			Sb

NILU – Norwegian Institute for Air Research

NILU – Norwegian Institute for Air Research is an independent, nonprofit institution established in 1969. Through its research NILU increases the understanding of climate change, of the composition of the atmosphere, of air quality and of hazardous substances. Based on its research, NILU markets integrated services and products within analysing, monitoring and consulting. NILU is concerned with increasing public awareness about climate change and environmental pollution.

NILU's values: *Integrity - Competence - Benefit to society*
NILU's vision: *Research for a clean atmosphere*

NILU – Norwegian Institute for Air Research
P.O. Box 100, NO-2027 KJELLER, Norway

E-mail: nilu@nilu.no
http://www.nilu.no

ISBN: 978-82-425-3030-1
ISSN: 2464-3327

Norsk institutt for luftforskning
Norwegian Institute for Air Research