

PAH measurements at Lista

January 2020 – December 2020

Claudia Hak



NILU report 11/2021

NILU report 11/2021	ISBN: 978-82-425-3042-4	CLASSIFICATION:			
	ISSN: 2464-3327	A – Unclassified (open report)			
DATE	SIGNATURE OF RESPONSIBLE PERSON	NUMBER OF PAGES			
15.06.2021	Ole-Anders Braathen, Viseadministrerende direktør (sign.)	33			
TITLE		PROJECT LEADER			
PAH measurements at Lista	Claudia Hak				
January 2020 – December 2020	NILU PROJECT NO.				
		O-119080			
AUTHOR(S)		QUALITY CONTROLLER			
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ABSTRACT					
On behalf of Aluminiumindustriens Miljøsekretariat (AMS) and Alcoa Lista, NILU – Norwegian institute for air research has conducted a sampling campaign in the surroundings of the Alcoa Lista aluminium smelter in order to update the knowledge on PAH-concentrations around the smelter today. Samples were taken in the period January – December 2020 and analysed for particle bound PAHs. As a consequence of reduced emissions compared to earlier measurements, the ambient concentrations of benzo(a)pyrene (BaP) were reduced. BaP had an annual average concentration below the target value at both sampling sites. At Huseby, the lower assessment threshold was exceeded. PAH-levels in the area were similar to those observed in Norwegian cities.					
NORWEGIAN TITLE					
PAH målinger ved Alcoa Lista					
KEYWORDS					
Air quality	Industrial pollution	Polycyclic aromatic hydrocarbons			
ABSTRACT (in Norwegian)					
NILU – Norsk institutt for luftforskning har, på oppdrag fra Aluminiumindustriens Miljøsekretariat (AMS) og Alcoa Lista, tatt PAH-prøver i omgivelsene til Alcoa Lista aluminiums-smelteverk for å oppdatere kunnskapen om PAH-konsentrasjoner rundt smelteverket i dag. Prøvene ble tatt i perioden januar – desember 2020 og analysert for partikkelbundne PAHer. Som en konsekvens av reduserte utslipp sammenlignet med tidligere målinger, er konsentrasjonen av benzo(a)pyren (BaP) redusert. Årsmiddelet av BaP-konsentrasjonen i 2020 var under målsettingsverdien på begge prøvetakingsstedene. På Huseby ble den nedre vurderingsterskelen overskredet. PAH-konsentrasjonen i området på samme nivå som i norske byer.					
PUBLICATION TYPE: Digital document (pdf) COVER PICTURE: T	neis Roksvåg Pedersen, Farsunds Avis			

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

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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 measurements 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 multidisciplinary study initiated and sponsored by "Aluminiumsindustriens Miljøsekretariat" (AMS) to update and supplement the Effect Study. The present report is a contribution to this update, focusing on the effect on air quality today, by the example of measurements carried out around Alcoa Lista.

In the period January 2020 – December 2020, weekly samples of 16 priority PAHs (particle-bound) were collected at two sites in the vicinity of the aluminium smelter Alcoa Lista. Huseby is located close to the smelter and Hanangermona is located 4 km west from the smelter in the direction towards Vanse. The measurements were carried out during the entire calendar year 2020. The annual variation of PAH-concentrations showed the highest levels in the winter months and lowest levels in the summer months.

Emissions to air and ambient concentrations of the measured compounds have decreased since the beginning of the 1990s as a result of improvement of the production technology.

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 average concentration of BaP close to Alcoa Lista in 2020 (0.48 ng/m³ at Huseby) was below the target value of 1 ng/m³ (for calendar year). At Hanangermona, the BaP average concentration in 2020 was 0.11 ng/m³. The BaP-concentration at Huseby was above the lower assessment threshold (0.4 ng/m³), but below the upper assessment threshold (0.6 ng/m³). At Hananger, the BaP-level was below the lower assessment threshold in 2020. The air quality criterion for BaP in Norway (annual average 0.1 ng/m³) was exceeded at Huseby. The level measured around Lista in 2020 was a factor 10-40 higher compared to the Norwegian regional background. The BaP-level at Lista is comparable to the level in Norwegian cities.

Alcoa Lista was selected for this study since the smelter is situated in a well-ventilated area (in contrast to the other smelter studied in the project, Hydro Sunndal, see Hak et al. (2021)) and the plant applies the Søderberg technology with much higher emissions of PAH than the widely used technology with prebaked anodes. Further, Alcoa Lista was required by the Norwegian Environment Agency to report annual PAH-levels in ambient air.

PAH measurements at Lista January 2020 – December 2020

1 Introduction

A 12-month sampling campaign focusing on the levels of PAHs in ambient air was carried out in the surroundings of the Alcoa Lista aluminium smelter in the period January – December 2020. Particlebound polycyclic aromatic hydrocarbons (PAHs, EPA 16 PAH) were sampled at two sites, one close to the smelter, the other one a few kilometres towards the small town Vanse. At one of the sites (Huseby), air quality measurements (in particular PAHs) 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 Hydro Sunndal, see Hak (2021)). The outcome from these activities will contribute to knowledge creation at the Al-industries and to secure 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 were carried out at selected smelters. The 10 smelters participating in the ESPIAL¹ project are placed at locations largely differing regarding dispersion conditions, population exposure, topography etc. This makes it difficult to conclude on the situation around other smelters based on measurements at only one distinct location. Lista and Sunndal were indicated as suitable sites, one located in a flat area at the coast, the other in a topographically complex terrain. Two separate sampling campaigns were carried out. The present report covers measurements at Lista in 2020 and comparison to the results from previous studies around Lista, back to the early 1990s.

1.2 Alcoa Lista

Alcoa Lista is located in the Farsund municipality in the Agder County. The surrounding area is characterised by flat terrain with a generally high wind exposure and good dispersion conditions so that emissions can be spread over larger areas. The prevailing wind direction is from north-west during much of the year. This will generally carry airborne pollutants seawards.

¹ ESPIAL – Ensuring the Environmental Sustainability of production of PrImary ALuminium



Figure 1: Alcoa Lista at Farsund. (Left): View towards southwest and the North Sea. (Right): Map over Farsund.

The climate at Lista is mild and humid, with a mean annual temperature of +7.6°C and annual precipitation of approximately 1050 mm.

The areas towards east are barren, low hills with sparse vegetation. Towards south-east, there is a small forest between the industrial site and the Lomsesanden Beach. The aluminium plant is close to the Husebysanden beach towards south-west. Towards west and north-west, there is mixed agricultural land with patches of forest and some built-up areas. A school and a pre-school are located in this area. To the north, there is a harbour area in Lundevågen.

The municipality has a population of nearly 9 800, whereof one-third are living in the town Farsund. The aluminium plant employs about 265 in addition to some regular maintenance contractors. It is the largest industrial workplace in the municipality. It delivers raw materials to Aludyne, an aluminium casting plant and previously an integrated part of Alcoa. About one-third of the workforce in the municipality work in the industry sector.

The aluminium plant was established in 1971 and was the last in Norway to be built with Søderberg technology. Specific to Søderberg is that the anode is baked in operation and PAHs evaporate from the anode top. The plant has three pot rooms with a total production capacity of about 94 000 tonnes p.a. This is an increase of about 17% since 1992. In addition, scrap aluminium and purchased metal are remelted and upgraded.

Alcoa Lista has upgraded the Søderberg technology to reduce environmental emissions while production results are comparable to what is achieved with prebake-technology. At traditional Søderberg, the anode top is open. New Søderberg technology involves closing the anode tops to reduce emission of process gases, point feeders for oxide and fluoride, modified paste recipe, etc. With regard to furnace operation, the addition of oxide was changed from large additions a few times a day, when the oven was opened, implicating emissions of process gases and dust, to a closed system with small frequent additions via point feeders. The potline has three separate gas treatment systems: One for the pot off-gas (dry scrubber + seawater scrubber), one for anode off-gas (dry scrubber) and one for pot room ventilation (seawater scrubbers). All cells have been upgraded with a closed point feeding system, which has increased the capacity and reduced greenhouse gas emissions. Point feeding and improvements at the gas treatment plants have resulted in a large reduction in emissions of PAHs, fluorides and dust. The development of New Søderberg was done in the 1990s. A large part of the transition was completed in 2000. Closing of anode tops and treatment plants were completed in 2006-2007.

Upgrade to New Søderberg:

- March 1995: Anode top covers with associated treatment plant in entire hall 3 (closer upper part of the ovens)
- December 1996: Point feeders in entire hall 3 (closing lower part of the ovens)
- 1999: Point feeders in hall 2
- 2000: Point feeders in hall 1
- 2006: Anode top covers in hall 2
- 2007: Anode top covers in hall 1

Figure 2 illustrates the development in production and PAH-emissions from 1992 to 2020, as reported by Alcoa Lista (<u>www.norskeutslipp.no</u>). For PAH to air (kg per year), the NS-9815 standard² was used until 2015, and this standard is used here in order to give a correct picture of the relative change. With some reservations regarding the accuracy of early measurements, the figure indicates a 75% reduction in this period.

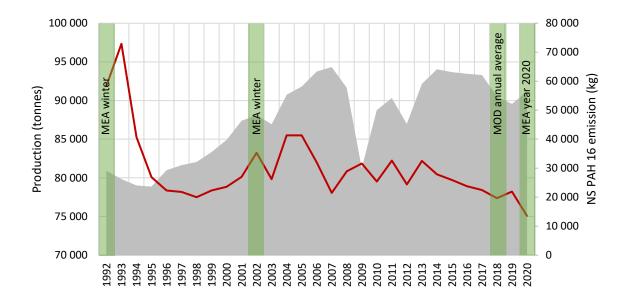


Figure 2: Development of production (grey background, unit: tonnes per year, left-hand y-axis) and emission of PAHs (unit: kg per year, right-hand y-axis) form 1992 to 2020. The years measurement (MEA) or modelling (MOD) studies for PAHs were carried out at Lista are highlighted.

1.3 Earlier studies on air quality at Lista

Measurement and modelling studies

Three measurement and modelling studies on PAHs have been carried out around Alcoa Lista before (see Table 1). Reports including air quality around the aluminium smelter at Lista from the 1970s cover

² Norwegian standard NS-9815 covered 16 PAH compounds to be included in the calculations of the total amount of PAHs. In 2015, the Norwegian standard has been replaced by two new international ISO standards. In the new reporting format, the sum of the 16 PAH compounds included in the US EPA standard must be reported. The transition to the new standard entails an apparently higher PAH emission. This is because the US EPA standard includes a number of volatile PAH compounds that were not included in the Norwegian standard.

fluorides and dispersion. Measurements of PAHs (sum of gaseous and particle-bound form) were performed in winter 1991 and winter 2002. In these two earlier studies, samples were collected at one site, Huseby, which is located ca. 1 km from the smelter. Since then, production conditions have changed and emissions are reduced. Samples for the present study were collected in 2020 at two sites (Huseby, as in earlier studies, and Hanangermona) and analysed for particle-bound PAHs.

Table 1:	Overview of earlier measurement and modelling studies on air quality carried out in the
surroundings of Alcoa Lista. The present study covers measurements carried out	
	and comparison to results back to 1991.

Report	PAHs	Other parameters	Study details
Report TR 4/71	-	Fluorides	Emission estimations
Report OR 18/71	-	Fluorides	Emission estimations
Report OR 92/74	-	Wind, temperature, stability	Measurement of meteorological parameters
Report OR 42/91	33 PAHs (gaseous and particle-bound)		Measurements winter 1991, 1 site (Huseby)
Report OR 37/2002	33 PAHs (gaseous and particle-bound)		Measurements winter 2001/02, 1 site (Huseby)
NILU report 27/2018	PAHs		Modelling (dispersion calculations)
NILU report 11/2021	16 PAHs (particle- bound)	-	Measurements, 2 sites (Huseby, Hanangermona) This study

The most recent study on PAHs in the surroundings of Alcoa Lista is a modelling study (NILU report 27/2017; Tønnesen, 2018). Dispersion calculations of PAHs, including BaP, in air were carried out based on emission data from 2017 given by Alcoa Lista (calculated from measurements of concentration and estimates for the various emission points) and wind measurements from Lista fyr from the period 2012-2017. Modelling results for the site Huseby, where measurements were carried out in 1991, 2001 and the present study, are given for the annual average concentrations of total PAHs and BaP.

Measurements of PAHs were carried out both in winter 2001/2002 (report OR 37/2002; Hagen, 2002) and in winter 1991 (report OR 42/1991; Hagen, 1991). In both studies, measurements of 33 PAH compounds (gaseous and particle-bound) were carried out at Huseby, which is also one of the two sites in the present study. The measurements in 1991 and 2001/02 were performed as daily averages from ca. 08:00 one day until ca. 08:00 the next day. In each of the studies, one sample per week was taken during a 10 week period. Equivalent measurements were carried out at most smelters in the period 1980-1982. No measurements of PAHs have been carried out at Lista before winter 1991. Parallel to sampling at Lista and other aluminium smelters, the measurements in winter 1991 also included sampling at two reference stations, Oslo and Lillestrøm.

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). Alcoa Lista³ participated in three of the four studies of heavy metals around Norwegian industries (2000, 2010, 2015). The number of metals analysed varied between the studies. Only gallium (Ga), a very likely contribution from an aluminium smelter, is deposited at distinctly higher levels near the smelter at Lista compared to background values. Ga is chemically related to aluminium (AI) and occurs in the deposition around all aluminium smelters.

In summer 2015, a parallel moss survey focusing on PAHs was carried out around Norwegian industries (Halse et al., 2017), however, no sampling was carried out around Alcoa Lista.

1.4 Measurement sites

Two sampling sites in the surroundings of the aluminium smelter Alcoa Lista, *Huseby* close to the smelter and *Hanangermona* ca. 4 km towards the small town of Vanse, were selected in order to be able to compare measurement results with the results from recent measurement and modelling projects and to assess the possible population exposure at Vanse. Their locations were selected according to local wind patterns and residents' exposure. At Huseby (Alcoa Miljøpark), measurements have been carried out earlier (see Section 1.3). 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.

³ Earlier Elkem Aluminium ANS Lista

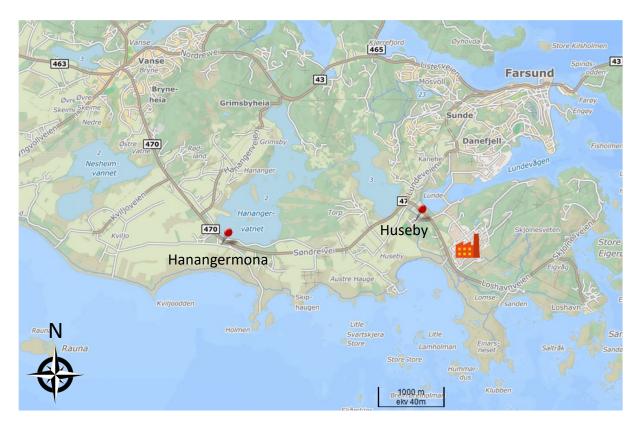


Figure 3: Map over area around Farsund and Vanse, including Alcoa Lista south of Farsund, the measurement station "Huseby" ca. 1 km northwest of the smelter and the measurement station "Hanangermona" 4 km towards west.



Figure 4: Sampling sites in the surroundings of the aluminium smelter Alcoa Lista. Left: Huseby. Right: Hanangermona. High volume samplers (Digitel DH-77) were used to sample PAHs.

1.5 Measurement program and method

At both sites, weekly samples of particle-bound PAHs were taken in the 12-month period 31 December 2019 to 6 January 2021. The weekly PAH-samples were merged to monthly samples.

The measurements covered the full calendar year 2020. Covering a full year is important to include different ambient concentrations of air pollutants due to varying wind directions, wind speeds and vertical dispersion conditions.

Polycyclic aromatic hydrocarbons (PAHs) are a group of organic compounds consisting of several hundred compounds with different properties. Some of the PAH compounds are toxic, mutagenic or carcinogenic and may therefore pose a health risk. PAHs are formed by all incomplete combustion of organic (carbonaceous) material. PAHs appear in polluted air in the gas phase and bound to particles. Traffic is an important source for PAH-exposure in larger cities. Stationary combustion, and especially wood combustion (residential heating), can contribute significantly to PAH-levels in the winter season. Other PAH sources are various types of industry (e.g. aluminium industry), especially electrolysis with Søderberg electrodes in aluminium production. PAHs evaporate from the electrode mass at high temperature. PAHs are also emitted during the production of electrode mass.

Benzo(a)pyrene, abbreviated BaP or B(a)P, is one of the compounds included in the group PAH. The chemical formula is $C_{20}H_{12}$. BaP is carcinogenic and has therefore a separate target value in air. BaP is often used as an indicator of the carcinogenic PAHs present in the environment.

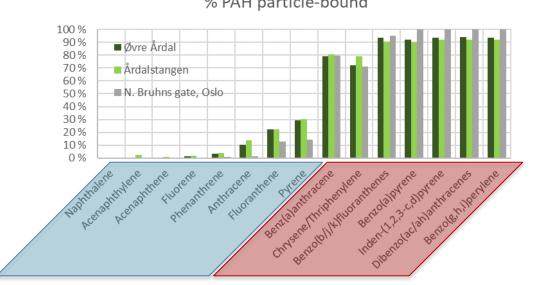
Particle-associated PAHs in air were quantitatively sampled on glass fibre filters at both sites using high-volume air samplers (Digitel Elektronik AG, Switzerland). The sampler consists of a pump that draws air through the samples with an average flow rate of 25 m³/h. PAHs were sampled weekly as 7-day 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 (cyclohexane). The weekly samples were merged to monthly samples, which were analysed for 16 priority⁴ 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/m³.

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. The particles collected on the particle filter are mainly in the size range below 10 μ m.

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 associated 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 residential 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, indeno(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.

⁴ 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).



% PAH particle-bound

Split of 16 priority PAHs into mainly gaseous compounds (blue background) and mainly Figure 5: 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).

In earlier measurement studies (1991 and 2001/02), NILU's PUR sampler has been used for sampling PAHs. Using this sampler, with polyurethane foam plugs after the particle filter, both particle-bound and gaseous PAH components are quantitatively collected. A total of 33 different PAH-compounds are reported in Hagen (1991) and Hagen (2002). In the analysis, the gas and particle phases were determined together. PAH are extracted from filters and plugs, and the extracts are analysed by gas chromatography. The detection limit for PAH was 0.01 - 0.02 ng/m³.

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. Norwegian limit and target⁵ values are equal to EU limit and target values (except for PM where Norway has stricter limit values). The target, threshold and guideline values for benzo(a)pyrene (as an indicator of carcinogenic PAHs) in ambient air are summarised in Table 2. The concentrations of benzo(a)pyrene shall be determined on the basis of the total content in the PM_{10} fraction, as an average over a calendar year. 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 air quality directive 2004/107/EC (and, analogously, Forurensningsforskriften FF) also defines assessment thresholds, which are pollution levels lower than the limit/target value, which specify requirements for measurements of air quality (FF Chapter 7 §7-8 and Appendix 3). Measurements shall

⁵ 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.

be carried out if the upper assessment threshold is exceeded (FF §7-9). When assessing the quality of the ambient air where the level in a representative period is between the upper and lower assessment threshold, the requirement for measurements is reduced, indicative⁶ measurements are sufficient (Directive 2004/107/EC). Below the lower assessment threshold (40% of the target value) there will be no need for measurements. The levels for BaP are specified in Table 2. Exceedances of upper and lower assessment thresholds are determined on the basis of concentrations during the previous five years where sufficient data are available. An assessment threshold is deemed to have been exceeded if it has been exceeded during at least three calendar years out of those previous five years.

The WHO guideline values are set for the protection of health, and are generally stricter than the comparable politically agreed EU standards. No specific guideline value is recommended by WHO for PAHs as such in air.

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.

Compound	Averaging period	Limit value (FF ⁷)	Air quality criterion (LKK ⁸)			
Polycyclic aromatic hydrocarbons (PAHs)						
B(a)P in PM ₁₀	Calendar year	Target value 1 ng/m³	0.1 ng/m ³			
		Upper assessment threshold 0.6 ng/m ³				
		Lower assessment threshold 0.4 ng/m ³				

 Table 2:
 National target value and air quality criterion for benzo(a)pyrene.

There is an air quality criterion for benzo(a)pyrene (B(a)P) as an indicator for carcinogenic effects of PAHs. The Norwegian air quality criterion is 0.1 ng/m³ as annual average concentration. PAHs are regulated in the Aarhus protocol on POPs (UN/ECE, 1998b) and the EU air quality directive (EU, 2004).

Within the premises of industries, the Limit values for pollutants in the work atmosphere (former Administrative norm) given by the Norwegian Labour Inspection Authority (Arbeidstilsynet) apply. The limit value for particulate PAHs collected on filters, based on the sum of 21 given⁹ PAH-compounds is 0.04 mg/m³ (40 μ g/m³).

⁶ Indicative measurements are measurements which are performed at reduced regularity but fulfil the other data quality objectives.

⁷ FF: Forurensningsforskriften (Norwegian air quality regulation, embedding European air quality limit values). URL: <u>https://lovdata.no/dokument/SF/forskrift/2004-06-01-931/KAPITTEL_3-1#KAPITTEL_3-1</u>

⁸ LKK: Luftkvalitetskriterier (Air quality criteria, set by Norwegian institute of public health and Norwegian environment agency). URL:

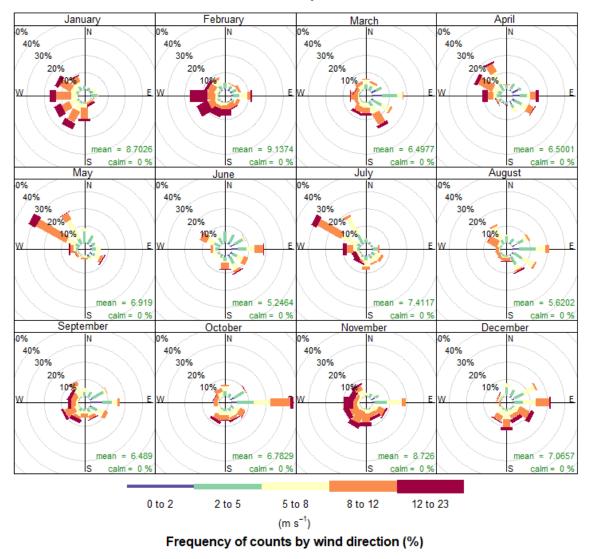
https://www.fhi.no/globalassets/dokumenterfiler/rapporter/2013/luftkvalitetskriterier---virkninger-avluftforurensning-pa-helse-pdf.pdf

⁹ Limit values for pollutants in the work atmosphere: <u>https://www.arbeidstilsynet.no/regelverk/forskrifter/forskrift-om-tiltaks--og-grenseverdier/8/1/</u>

3 Measurement results

Results from PAH-measurements carried out in 2020 (31 December 2019/1 January 2020 – 6 January 2021) are shown in the following subchapters and BaP-levels are compared with the results from earlier measurements (see Table 1). Since the exposure time of individual samples and the sampling frequency differ for the individual campaigns, seasonal averages 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 and previous years are reported by Aas et al. (2020). The report for 2020 will be available by summer 2021.

A meteorological station is operated by the Meteorological institute at Lista fyr, ca. 13 km westnorthwest from the smelter and is considered representative for the dispersion conditions in the area. Monthly wind roses from Lista fyr in 2020 are shown in Figure 6. Wind roses show the distribution of wind speed and wind direction at a site, i.e. the frequency of the occurrence of wind coming from different wind direction sectors. The main wind direction was from north-west in the summer months and from easterly directions in autumn and winter. The wind roses from 2020 agree quite well with the general wind distribution at Lista with west-northwest and east being the most frequent wind directions.



Lista Fyr

Figure 6: Monthly wind roses for the period January – December 2020, based on hourly values measured at Lista fyr.

The temperature and precipitation rate measured at Lista fyr in 2020 are shown in Figure 7. Due to its location at the coast, Lista has a temperate coastal climate with temperatures basically above 0°C. The daily average temperature in 2020 varied between 0.1°C (27 February 2020) and 21.5°C (26 June 2020). The ambient temperature may have an effect on the split of PAHs into gaseous and particle-bound compounds. Some lighter compounds will be less volatile at low temperatures, leading to a change of the PAH-profile in winter, compared to summer (see Section 1.5). There was no pronounced seasonal variation of the precipitation amount. The lowest precipitation rates occurred in April, May and August 2020.

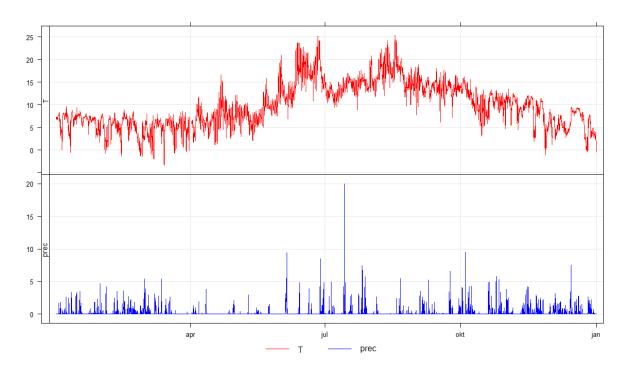


Figure 7: Temperature (°C) and precipitation rate (mm) measured at Lista fyr in 2020 (hourly values).

3.1 Polycyclic aromatic hydrocarbons – PAHs in 2020

Weekly samples of particle-bound PAHs were collected at both sites, Huseby and Hanangermona, and merged to monthly (ca. 4 weeks) samples. The monthly samples were analysed for the group EPA 16 PAH¹⁰. The sampling site at Huseby was the same as during earlier measurements in 1991 and 2001/02.

PAH-profiles of the 16 priority PAHs are shown in Figure 8 for both sites. The profiles show a seasonal variation and are very similar comparing the two stations. It needs to be underlined that exclusively particle-bound¹¹ PAHs were analysed. 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 (see Figure 5). Phenanthrene, fluoranthene and pyrene are PAH-compounds which are connected with emissions from the aluminium industry and were present in the profiles observed. Also benzo(b)fluoranthene is seen as indicator for aluminium production as PAH source (Aubin and Farant, 2000) and was very prominent in the profiles observed. The highest levels of most PAH compounds were observed in October, November and December. Biomass burning (residential heating) may have contributed to the increased levels. In January 2020, PAH-concentrations measured at both sites were substantially lower than in December 2020, despite similar ambient temperatures. The wind direction in January 2020, where winds predominantly came from westerly sectors (see Figure 6) was favourable for the sampling sites, compared to December 2020, where wind from east and southeast was measured, bringing emissions towards both Huseby and Hananger.

¹⁰ 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).

¹¹ There were both analytical and economic reasons for analysing only particle-bound PAHs and not gaseous PAHs in the monthly samples.

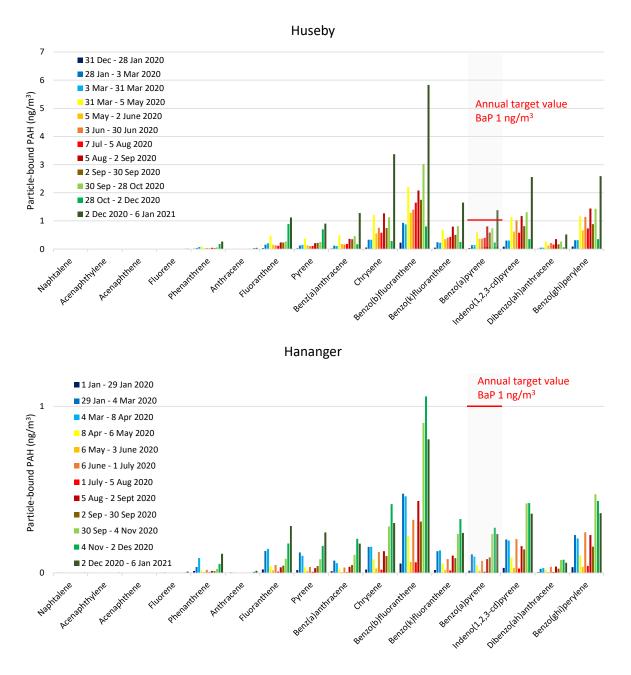
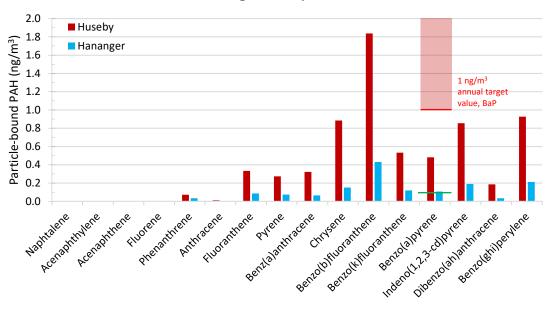


Figure 8: Monthly average profiles of particle-bound PAHs in 2020 at Huseby (upper plot) and Hananger (lower plot). Unit: ng/m³.

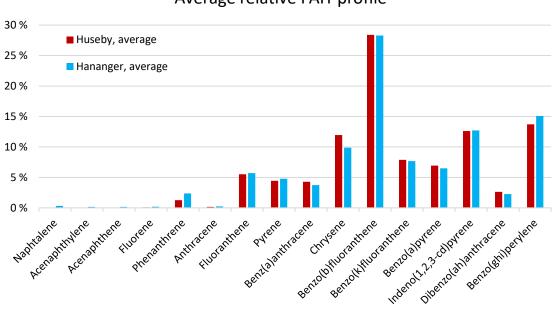
The levels of all compounds were higher at Huseby than at Hananger (Figure 9), which is a few kilometres further away from the smelter. For most compounds, the concentration ratio was about a factor 5.



Average PAH-profile

Figure 9: Average profiles (January – December 2020) of particle-bound PAHs in 2020. Comparison of concentrations at Huseby and Hananger. Unit: ng/m³. The red line shows the annual target value for BaP (1 ng/m³). The green line shows the Norwegian air quality criterion for BaP (0.1 ng/m³).

The PAH profile at Hananger was very similar to the PAH profile observed at Huseby (Figure 10), indicating that the same PAH sources influence both sites. The figure shows relative profiles, i.e. the percentage of each compound contributing to the sum of 16 priority PAHs.



Average relative PAH-profile

Figure 10: Average relative profiles (January – December 2020) of particle-bound PAHs at Huseby and Hananger in 2020. %-contribution of each compound to sum PAH₁₆.

The seasonal variation of relative PAH-profiles at Huseby and Hananger is shown in Figure 11. For the lightest and heaviest compounds, the temperature effect on the split between gaseous and particlebound form is observable. Heavier compounds like indeno(1,2,3-cd)pyrene, dibenzo(ah)anthracene and benzo(ghi)perylene have larger relative contributions to sum PAH₁₆ in summer months, Lighter compounds like phenanthrene, fluoranthene and pyrene have larger relative contributions to sum PAH₁₆ in the winter months, showing that the split between gaseous and particle phase is moved under cold conditions in favour of the particle phase.

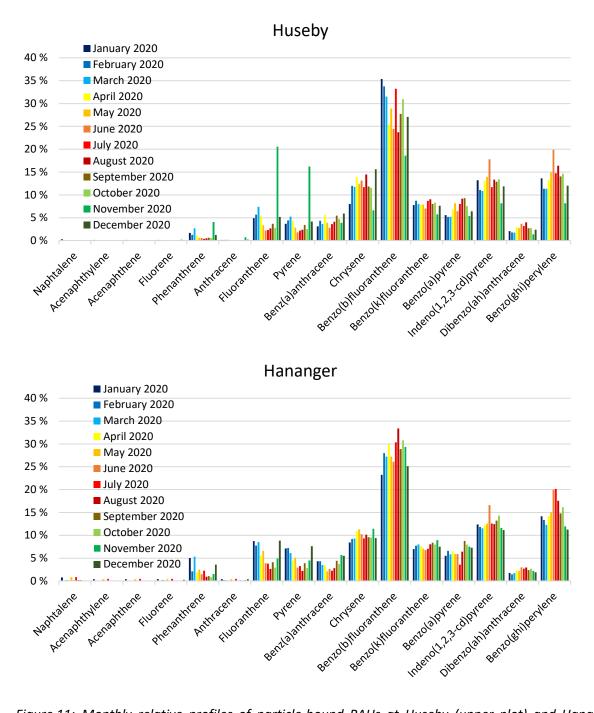


Figure 11: Monthly relative profiles of particle-bound PAHs at Huseby (upper plot) and Hananger (lower plot) in 2020. %-contribution of each compound to sum PAH₁₆.

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.

3.2 Comparison to limit values

The target value for BaP is 1 ng/m³ averaged over a calendar year. The annual average in the period January – December 2020 was 0.48 ng/m³ at Huseby and 0.11 ng/m³ at Hananger. The benzo(a)pyrene concentration at the sampling sites as a consequence of emission from aluminium production in addition to a possible background concentration did not exceed the target value.

The highest monthly average value in 2020 was 1.38 ng/m^3 at Huseby in December 2020.

The BaP-concentration at Huseby was higher than the lower assessment threshold (0.4 ng/m³), but lower than the upper assessment threshold (0.6 ng/m³). At Hananger, the BaP-level was below the lower assessment threshold in 2020.

The air quality criterion for BaP in Norway (annual average 0.1 ng/m^3) was clearly exceeded at Huseby (0.48 ng/m³), and marginally exceeded at Hananger (0.11 ng/m³).

3.3 Comparison to Norwegian background and cities

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 Birkenes¹² are reported by Bohlin-Nizzetto et al. (2020), also including previous years. The report for 2020 will not be published before summer 2021. 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 in 2019 were phenanthrene (0.7 ng/m³, 37% of PAH₁₆), followed by dibenzofuran (0.5 ng/m³, not part of PAH₁₆), fluorene (0.4 ng/m³, 21% of PAH₁₆) and fluoranthene (0.2 ng/m³, 9% of PAH₁₆). 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 annual mean concentration at Birkenes in 2019 (2.0 ng/m³ for sum PAH₁₆) 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/m³).

As mentioned above, PAH-profiles at Lista 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 PAH₁₆ 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 BaP at Birkenes in 2020 was 0.009 ng/m³ (0.011 ng/m³ in 2019). At Huseby and Hananger, the average BaP concentrations were an order of magnitude higher, 0.48 ng/m³ and 0.11 ng/m³, respectively. Benzo(a)pyrene concentrations at Birkenes have decreased since 2009.

¹² Birkenes is located ca. 93 km east-northeast of Lista. <u>https://www.nilu.no/anlegg/nilus-observatorier-og-malestasjoner/birkenesobservatoriet/</u>

Particle-bound PAHs are routinely measured in selected Norwegian cities (Oslo, Lillehammer, Trondheim). Filter samples are collected every third¹³ day and merged to monthly samples which are analysed for 7 PAH compounds¹⁴, which are classified as potentially carcinogenic within the 16 priority PAHs. Annual average values of BaP in city centres in Norway have mainly been between 0.3 ng/m³ and 0.6 ng/m³ in recent years. The annual BaP-concentration at Huseby, 1 km from the smelter, was comparable to levels in Norwegian cities. At Hananger, ca. 4 km from the smelter, the levels were lower. The relative contribution of BaP to the sum of the seven PAHs is lower at Lista compared to Norwegian cities. Benzo(b)fluoranthene has a considerably higher relative contribution to the sum of the seven PAHs than in Norwegian cities. This indicates that aluminium industry is a local source for benzo(b)fluoranthene at Lista.

3.4 Comparison to previous campaigns

For comparing the results from 2020 to earlier measurements, carried out in 1991 and 2001, several differences in the measurements have to be noted:

- In 2020, weekly samples were merged to monthly samples. In 1991 and 2001, 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 once 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 a month.
- In 2020, particle-bound PAHs were analysed, while in 1991 and 2001, both¹⁵ 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 2020.
- In 2020, PAH samples were analysed for 16 priority PAHs (EPA 16 PAHs). In 1991 and 2001, 33 PAHs were analysed. For comparison, the 16 priority PAHs were selected. Modelling results (NILU report 27/2018) using emissions from 2017 show the sum of NS 16 PAHs footnote ².

In previous measurement campaigns, PAHs were only sampled at Huseby. Compared to previous campaigns at Lista, PAH-concentrations in 2020 were lower (Figure 12). Note, however, that only particle-bound PAHs were analysed in 2020. The average concentration for sum PAH₁₆ from 24-hour samples in winter 2001 was 56 ng/m³. Compared with similar measurements in winter 1991 (sum PAH₁₆ 194 ng/m³), the average concentration of both total PAH and BaP in winter 2001 was significantly reduced at Lista. PAH-emissions at Lista have been reduced by 50-70% between winter 1991 and winter 2001/02. In the period 2001-2020, emissions were reduced by further 50%.

A general observation from measurements in the surroundings of aluminium plants in 1991 and 2001/02 was that the concentration of BaP was reduced more than the concentration of sum PAH. This may indicate that PAH emissions from car traffic were even more reduced than those from aluminium plants in the period 1991-2001 (Report OR 37/2002) or that reduction measures at the smelter have been more effective for the heavy PAHs than for the volatile compounds.

¹³ Exception: at Sofienbergparken in Oslo, samples are collected every sixth day

¹⁴ benz(a)anthracene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(j)fluoranthene, benzo(a)pyrene, indeno(1,2,3-cd)pyrene, dibenzo(ah)anthracene

¹⁵ The sum of particle-bound and gaseous PAHs were analysed in previous campaigns, not separately.

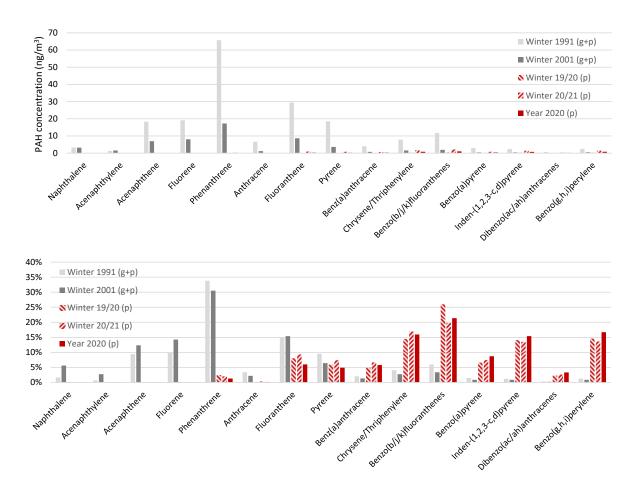


Figure 12: Average profiles of PAHs in winter 1991 (10 weeks), winter 2001 (10 weeks), winter 2019/20, winter (2020/21) and year 2020 at Huseby. In 1991 and 2001, no PAH measurements were performed at Hanangermona. (Upper plot) PAH concentration profiles, (lower plot) profiles of the percentage of each compound to PAH₁₆. Note that in the earlier studies, benzo(b)fluoranthene and benzo(k)fluoranthene were reported as group benzo(b,k)fluoranthenes.

The PAH-profile in winter 2001 was very similar to the profile from winter 1991 (Figure 12). Although PAH-levels have been significantly reduced since 1991, the relative ratios of the compounds, and thus the main sources, were about the same. The compounds with highest concentrations in 1991 (phenanthrene, fluoranthene, pyrene, fluorene) had decreased distinctly between 1991 and 2001, especially phenanthrene. It needs to be recalled that the PAH-profiles from 1991 and 2001 contain both gaseous and particle-bound PAHs. Since phenanthrene, fluoranthene, pyrene and fluorene occur mainly in gaseous form, it is natural that values in the 2020 profiles of particle-bound PAHs are lower than in previous campaigns.

Since the decline of concentrations between 1991 and 2020 is rather large, the percent contribution of each compound to PAH₁₆ is compared, see Figure 12. While in 1991, phenanthrene, fluoranthene, pyrene and fluorene (which occur mainly in gaseous form) were most prominent, the compounds standing out in the PAH-profile of 2020 are benzo(b)fluoranthene, chrysene, benzo(ghi)perylene and indeno(1,2,3-cd)pyrene. The figure is only an illustration, but cannot be used for comparing, since the 2020 profile is biased by not including gaseous compounds.

Benzo(a)pyrene, which almost exclusively occurs in the particle phase, has decreased strongly between 1991 and 2001. In winter 2001, the BaP-level at Lista was reported to be 83% lower than in winter

1991. The benzo(a)pyrene concentration at Huseby in the period 17. January – 22. March 1991 was 2.9 ng/m³. In winter 2001 (1. November 2001 – 3. January 2002), a BaP concentration of 0.5 ng/m³ was measured at Huseby (Figure 13). In winter 2019/2020 and winter 2020/2021, average BaP concentrations of 0.1 ng/m³ and 0.8 ng/m³ were measured at the same site. The average over both winter seasons (0.46 ng/m³) was comparable to the winter average in 2001, although PAH-emissions are reported to have decreased by 50% between 2001 and 2020. The annual BaP average for the year 2020 was 0.48 ng/m³, i.e. above the lower assessment threshold.

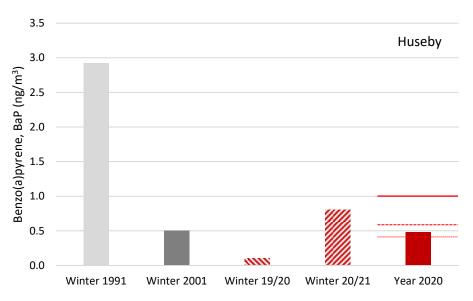


Figure 13: Average benzo(a)pyrene concentration (ng/m³) in Lista in winter 1991 (10 weeks, January – March), winter 2001 (10 weeks, November – January), winter 2019/2020 (January – March) and winter 2020/2021 (November – December) and annual average for 2020. The annual target value (1 ng/m³), upper assessment threshold (0.6 ng/m³) and lower assessment threshold (0.4 ng/m³) are also shown.

It needs to be pointed out again that 24 hour samples were collected in the previous campaigns (1991 and 2001), once a week, and that the concentrations are reported to have varied a lot from day to day. There is an uncertainty in the representativeness of the seasonal averages from winter 1991 and winter 2001 compared to the current measurements which evenly cover every month.

Emissions of PAHs are reported to have decreased, especially in the beginning of the 1990s. The production, however, has increased by ca. 13% since 1992 (see Figure 2). The aluminium plant at Lista is using Søderberg technology which is strongly associated with PAH emission due to the construction of the anode that consists of coke and anthracite aggregates bound together with coal tar pitch. At Lista, a new Søderberg technology has been developed, where alumina is fed continuously in closed systems (closed point feeding technology). An improved pot gas collection system collects gases such as PAHs and fluorine and dust from aluminium production with closed ovens/furnaces, so that they do not escape into the oven hall or into the atmosphere. Anode gases are collected and sent via treatment plants. This provides, a.o. a better working environment and reduced emissions and keeps high productivity. PAH-emissions decreased further since ca. 2005. The decrease of emissions is reflected in Figure 2.

For comparison, in a recent modelling study (NILU report 27/2018, Tønnesen (2018)), dispersion calculations for PAHs in air were carried out using meteorological data from Lista fyr from the period 2012-2017 and PAH emission data from 2017. An annual BaP-average of 1.3 ng/m³ was calculated at Huseby. For PAHs, the calculated annual average at Huseby was 76 ng/m³. The model calculations from

2018 showed good agreement with previous measurements for PAHs, but gave a somewhat higher BaP concentration level at Huseby than the measurements in winter 2001/02, where 0.5 ng/m³ were measured (Hagen, 2002). Both operating and emission conditions have changed in the period 2001-2017. As shown in Figure 2, the PAH emissions have decreased between 2001/02 and 2017. The calculated area with BaP concentrations exceeding the target value of 1 ng/m³ ranged 1 km north and south of the smelter and ca. 2 km west and southeast of the smelter. The highest concentrations of PAH and BaP occur inside the premises of the plant between the halls. Calculated maximum concentrations in this area were 32.5 ng/m³ BaP and 3.5 μ g/m³ PAH. Note that within the industry premises, the limit values for pollutants in the work atmosphere apply (see also Section 2). The model results from 2017/2018 gave considerably higher BaP concentration at Huseby than measured in 2020. This may be explained by a combination of several factors. First of all, the model is conservative, i.e. it interprets uncertainties in a way that calculated concentrations may overestimate the experienced levels "to be on the safe side". Production in 2020 was reported to be 2% lower than in 2017. The emission of sum PAH₁₆ reported for 2020 was 40% lower than in 2017, the emission of BaP was 20% lower in 2020 than in 2017. The named factors support higher concentrations to be estimated for 2017. Both years had a distribution of wind directions which corresponds to the long-term average at Lista.

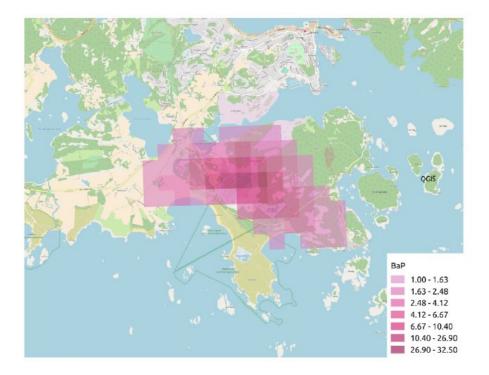
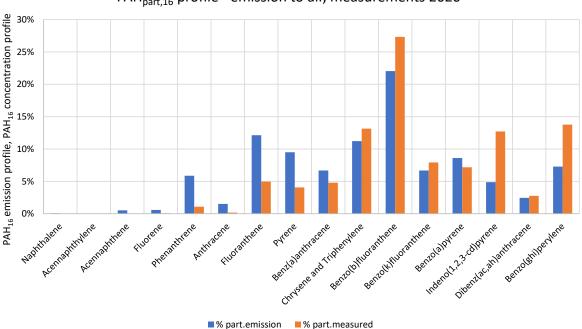


Figure 14: Calculated annual average concentration of BaP around Alcoa Lista. Unit ng/m³. Values above 1 ng/m³ (target value for BaP). Size of each square 250 m x 250 m.

3.5 Comparison to emission profile

PAH profiles measured in ambient air at Huseby and Hananger are compared to the emission profile of particle-bound PAHs obtained from Alcoa Lista (see Figure 15). The emission profile and the measured profile agree very well. Phenanthrene, fluoranthene and pyrene have a larger relative contribution to the emission profile than to the measured PAH profile. Indeno(1,2,3-cd)pyrene and benzo(ghi)perylene have a larger contribution to the measured profile. The emission profile of gaseous PAHs is dominated by light compounds like naphthalene, acenaphthene, phenanthrene and fluorene which stay gaseous and are not detected as particle-bound compounds.



PAH_{part.16} profile - emission to air, measurements 2020

Figure 15: Relative profile of particle-bound PAHs emitted by Alcoa Lista in 2020 (blue) and relative profile of particle-bound PAHs measured at Huseby in 2020 (orange).

4 Discussion and conclusions

Comparison to the results from earlier studies indicates that although the production of aluminium has increased over the past 30 years, the emissions of PAHs and the impact on the surroundings have decreased.

Alcoa Lista is the only Søderberg plant in Northern Europe today. Alcoa Lista has developed a new Søderberg technology in the 1990s in order to improve the environmental performance. Introducing a closed anode top strongly reduced the PAH-emissions. A better alumina feeding control technology was achieved by using point feeders reducing the frequency of anode effects. At the same time, the technology helped improving current efficiency, reducing energy consumption and achieving higher production. A large part of the conversion was completed in 2000.

PAHs show an annual variation with higher concentrations in the cold season. In winter, residential heating is another important PAH-source and more frequent stable conditions in winter may lead to an accumulation of pollutants. In summer, the aluminium industry is the main source of PAHs to air at Lista. The emission of PAHs/pollutants has been reduced, especially between 1990 and 2000, and the reduction is reflected in reduced ambient PAH-concentrations today.

The results from the present study (January – December 2020) are compared to results from earlier measurements carried out in 2001/02 and 1991 and to recent modelling results (2017). The sampling strategy applied in 2020 was different from the strategy in 2001/02 and 1991. While measurements were carried out on 10 days distributed over 3 months in 2001/02 and 1991, weekly samples were taken in 2020 and merged to monthly samples, evenly covering the entire 12-month period. The results from 24-hour samples are more sensitive to time variations compared to the measured concentrations for monthly samples. However, seasonal averages are considered to be more comparable.

In the present study, only particle-bound PAHs were analysed, while the sum of gaseous and particlebound PAHs was reported for previous studies. Analysis of only particle-bound PAHs was chosen assessing both economic and analytical reasons. Four weekly samples were decided to be merged to a monthly sample instead of collecting monthly samples, in order to decrease the risk of losing an entire month in case of sampling problems. Merging filter samples to monthly samples is a standard technique, also used for Norwegian cities. Gaseous PAHs are collected with polyurethane (PUR) foam blocks. Merging four weekly PUR blocks into one monthly sample was evaluated not viable. The alternative, analysing weekly PAH samples (both gaseous and particle-bound) would have exceeded the frame of the project.

Alcoa Lista appears to be the main source for PAHs in the surroundings, as concentrations of the individual compounds were higher at Huseby than at Hanangermona throughout the measuring period.

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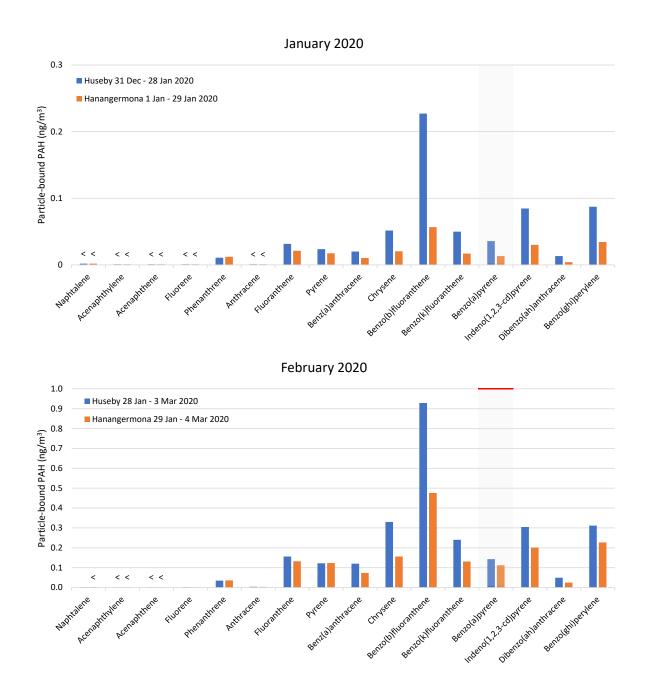
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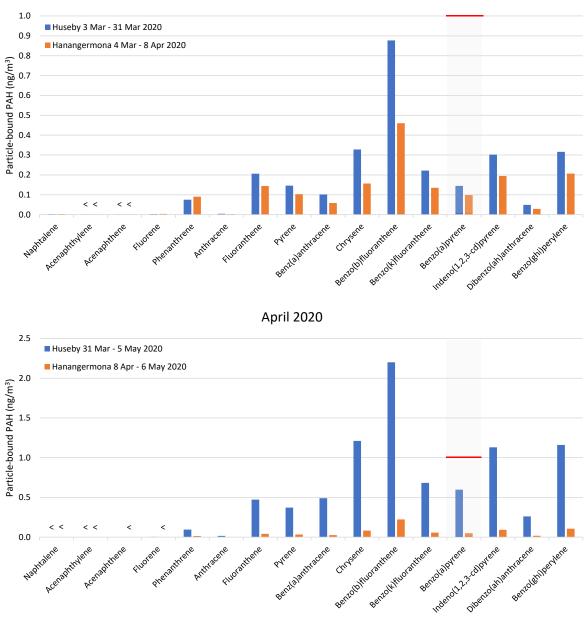
Appendix A

Monthly PAH profiles

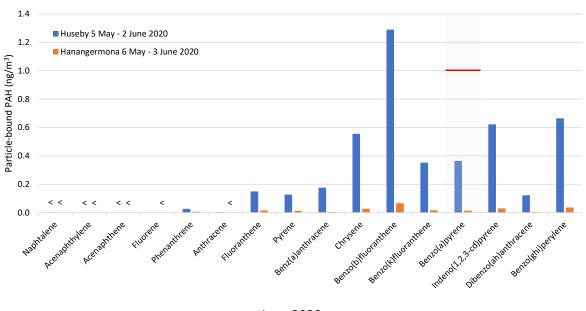


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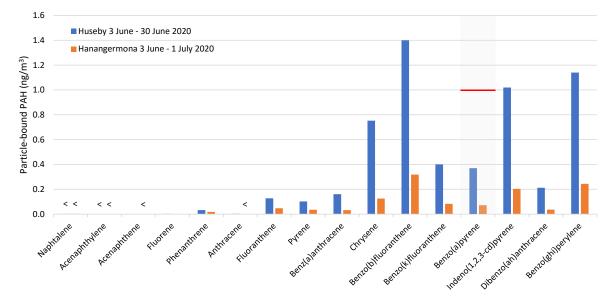




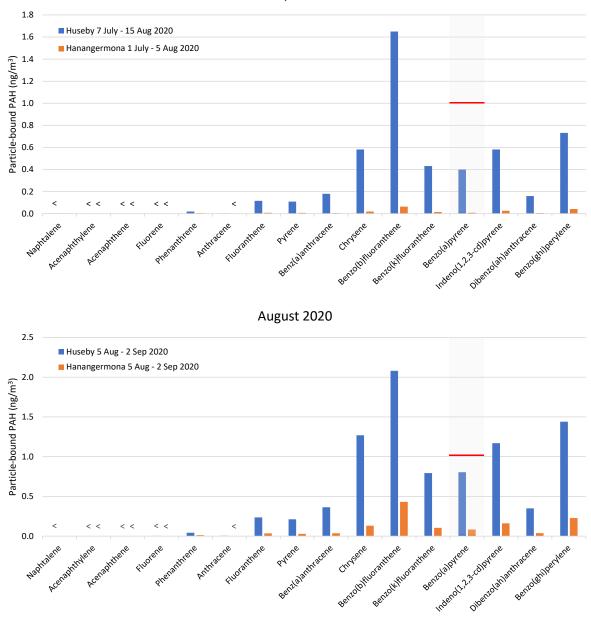


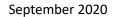


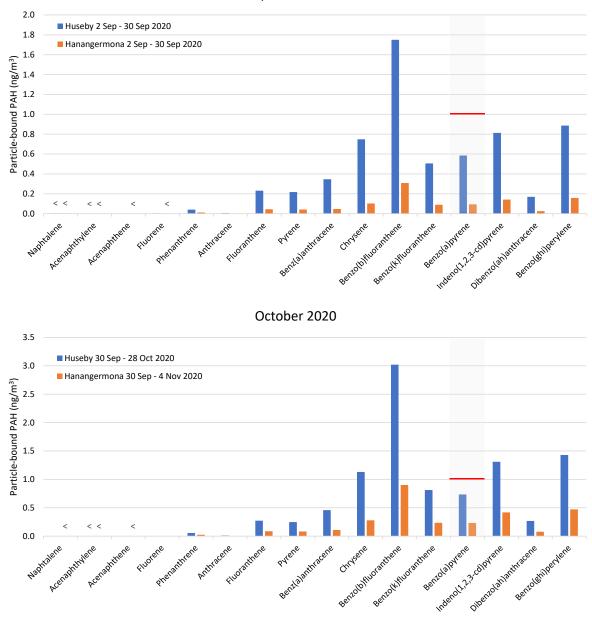
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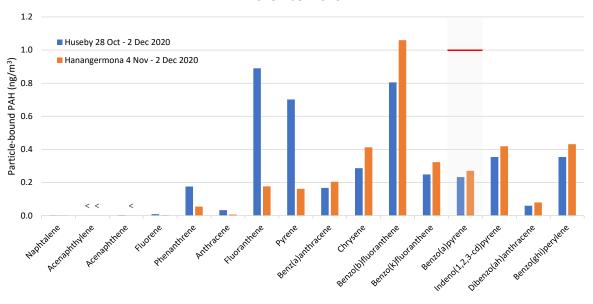




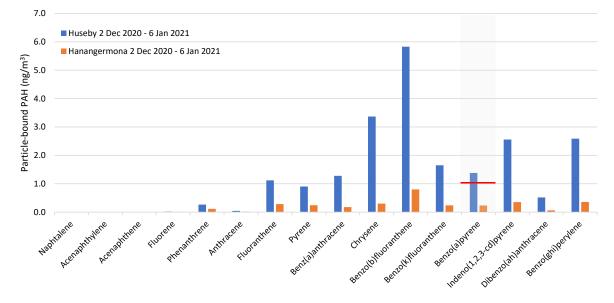




November 2020



December 2020



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ISBN: 978-82-425-3042-4 ISSN: 2464-3327

