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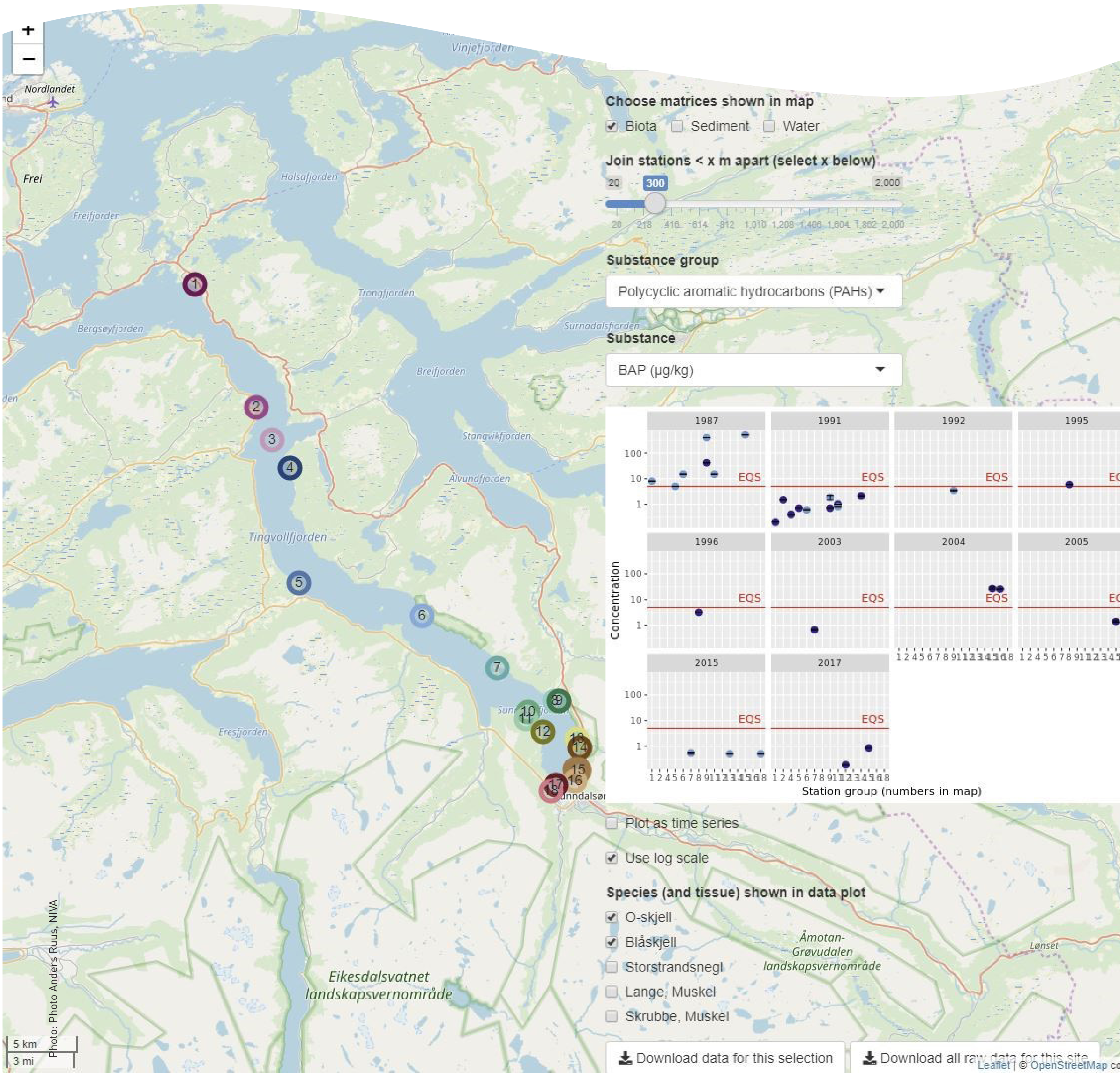
ESPIAL - Data review and evaluation of historical data (WP1)



Norwegian Institute for Water Research

REPORT

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Summary

The present report details the outcome of work package 1 of ESPIAL, with the aim of compiling the historical data from previous surveys at the aluminium sites in a common database and further to analyse the data. A graphical data displaying tool was developed as a custom interactive tool for data visualisations and was designed to be intuitive and user friendly. The main outcomes and findings of this work package can be summarized as follows: Polycyclic aromatic hydrocarbons (PAHs) constitute most data in the database; There is a decent amount of data on sediment samples in the data base, however these do not form time series; Most time series available in the data base are on blue mussel from Sunndal and northern horsemussel from Årdal; Most time series show a declining tendency; All statistically significant PAH time trends show declining concentrations; The database is likely not complete, and more data could be available; The graphical data displaying tool is a promising aid for instant insight in concentrations and time series of different analytes in environmental samples, historically collected in the vicinity of the smelters; The graphical data displaying tool makes it possible to view concentrations and trends in relation to the EQSs, giving insight in chemical status and sites of interest, in terms of possible need of remedial action or natural recovery.

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ESPIAL

Data review and evaluation of historical data

(WP1)

Preface

In the early 90s the aluminium industry commissioned an effects assessment study on the impact of it's production activities on the environment. The present project, ESPIAL (Ensuring Sustainable Production of Primary Aluminium) aims to revisit this investigation in a new more holistic way focusing on new technologies and tools in order to secure the future sustainability of the industry. The main objective is to provide the basis for developing existing expertise and the innovation of new technologies and tools for a more sustainable future for the aluminium industry, aspiring to gain a better understanding of the interactions between ecosystems (Marine, terrestrial, freshwater) and impacts of historical and future activities within the industry.

The present report details the outcome of work package 1 of ESPIAL, with the aim of compiling historical data from previous surveys at the aluminium sites in a common database and further to analyse the data. Jarle Håvardstun and Sigurd Øxnevad have been responsible for data gathering and Dag Hjermann has performed the statistical analyses and designed the graphical data displaying tool that has been developed in this work package. Anders Ruus has authored the report with assistance from all work package participants.

Oslo, Sept 2022

Ailbhe Macken

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Summary

The present report details the outcome of work package 1 of ESPIAL, with the aim of compiling the historical data from previous surveys at the aluminium sites in a common database and further to analyse the data. Data from relevant surveys (gathered from a literature review) were merged in a common database (available as a Microsoft Excel file), functioning as the data source for trend analyses and for the graphical data displaying tool developed within this work package. Statistical trend analyses were performed on time series defined as the data for a given station, matrix (e.g. sediment, or the combination of species and tissue) and analyte/contaminant. For each time series, linear regression was performed on the annual median values. The analyses were performed only for time series where the data available were sufficient, according to criteria adopted from OSPAR (the Oslo-Paris Convention for the protection of the marine environment of the North-East Atlantic).

The graphical data displaying tool was developed as a custom interactive tool for data visualisations, and was designed to be intuitive and user friendly, with roll-down menus, check boxes, graphical presentation of data (graphs) and a geographical interface (presentation on map).

The main outcomes and findings of this work package can be summarized as follows:

- Polycyclic aromatic hydrocarbons (PAHs) constitute most data in the database.
- There is a decent amount of data on sediment samples in the database, however these do not form a time series.
- Most time series available in the database are on blue mussel from Sunndal and northern horsemussel from Årdal.
- Most time series show a declining tendency.
- All statistically significant PAH time trends show declining concentrations.
- The database is likely not complete, and more data could be available.
- The graphical data displaying tool is a promising aid for instant insight in concentrations and time series of different analytes in environmental samples, historically collected in the vicinity of the smelters.
- The graphical data displaying tool makes it possible to view concentrations and trends in relation to the EQSs, giving insight into chemical status and sites of interest, in terms of possible need of remedial action or natural recovery.
- Extended monitoring continuing established stations and matrices will increase/improve the basis for time trend analysis and thus also predictions of environmental concentrations.

Sammendrag

Tittel: ESPIAL - Data review and evaluation of historical data (WP1)
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Denne rapporten presenterer resultatet av arbeidspakke 1 i ESPIAL-prosjektet, med mål om å samle de historiske dataene fra tidligere undersøkelser på lokaliteter hvor det har foregått aluminiums-produksjon i en felles database og videre analysere disse dataene. Data fra relevante undersøkelser (samlet gjennom en litteraturgjennomgang) ble slått sammen i en felles database (tilgjengelig som Microsoft Excel-fil), og fungerte som datakilde for trendanalyser og for det grafiske datavisnings-verktøyet som er utviklet i denne arbeidspakken. Statistiske trendanalyser ble utført på tidsserier definert som dataene for en gitt stasjon, matriks (f.eks. sediment, eller kombinasjonen av arter og vev) og analytt/miljøgift. For hver tidsserie ble lineær regresjon utført på de årlige medianverdiene. Analysene ble utført kun for tidsserier der tilgjengelige data var tilstrekkelige, i henhold til kriterier som anvendes av OSPAR (Oslo-Paris-konvensjonen).

Det grafiske datavisningsverktøyet ble utviklet som et tilpasset interaktivt verktøy for datavisualisering, og ble designet for å være intuitivt og brukervennlig, med rullegardinmenyer, avmerkingsbokser, grafisk presentasjon av data (grafer) og et geografisk grensesnitt (presentasjon på kart).

Hovedresultatene og funnene av denne arbeidspakken kan oppsummeres som følger:

- Polysykliske aromatiske hydrokarboner (PAH) utgjør de fleste data i databasen.
- Det er en anstendig mengde data fra sedimentprøver i databasen, men disse danner ikke tidsserier.
- De fleste tidsserier som er tilgjengelige i databasen er på blåskjell fra Sunndal og oskjell fra Årdal.
- De fleste tidsserier viser en nedadgående tendens.
- Alle statistisk signifikante PAH-tidstrender viser nedadgående konsentrasjoner.
- Databasen er sannsynligvis ikke komplett, og mer data kan være tilgjengelig.
- Det grafiske datavisningsverktøyet er et lovende hjelpemiddel for øyeblikkelig innsikt i konsentrasjoner og tidsserier for forskjellige analytter i miljøprøver, historisk samlet i nærheten av smelteverkene.
- Det grafiske datavisningsverktøyet gjør det mulig å se konsentrasjoner og trender i forhold til miljøkvalitetsstandarder (EQS), og gir innsikt i kjemisk status og interessante steder, med tanke på mulig behov for avbøtende tiltak eller naturlig forbedring.
- Fortsatt overvåking på etablerte stasjoner og med etablerte prøvematriser vill øke/bedre grunnlaget for tidstrendanalyser og således også prediksjoner om fremtidige konsentrasjoner i miljøet.

1 Introduction

The Work described in this report is part of the ESPIAL project. ESPIAL (Ensuring the Environmental Sustainability of production of PrImary ALuminium) is a multidisciplinary study on the local environmental effects around aluminium smelters in the Nordic countries. The study is initiated and sponsored by the "Aluminiumsindustriens Miljøsekretariat" (AMS), and is building on a similar, but more extensive study of the Norwegian smelters from 1991-94, called the "Effect Study". ESPIAL involves new field studies conducted in 2018-2020 on the marine environment, air quality, effects on vegetation and on wildlife, in addition to a review of studies conducted through the period after the Effect Study on these subjects. The report at hand is a contribution to ESPIAL. This report deals with work package 1 (WP1) of the Marine Environment part of ESPIAL.

An objective of the ESPIAL project is to look at today's status and to identify and describe changes since the original effect study that was conducted approximately 25 years ago, specifically focusing on the changes in environmental pollution and risks, changes in discharges and changes in environmental management. As such, the aim of this work package has been to compile the historical data from previous surveys at the aluminium sites in a common database and further to analyse the data to achieve a more profound understanding of the causal relationships between emissions and observed effects. The ultimate goal of this exercise is to be able to eventually answer the questions: Which sites still exceed environmental quality standards (EQSs)? Based on historical trends, which sites need action and which will improve if left as is? And if left as is, how long until good ecological and chemical status is achieved? In this work package, we aspire to help answer these questions also by providing data and information for use in work package 2 (WP2), a modelling exercise for understanding causal relationships and trends.

WP1 encompasses all smelter sites relevant for ESPIAL:

- Sunndal (Hydro)
- Høyanger (Hydro)
- Årdal (Hydro)
- Karmøy (Hydro)
- Husnes (Hydro)
- Lista (Alcoa)
- Mosjøen (Alcoa)
- KUBAL (Rusal)
- ISAL (Rio Tinto Alcan)
- Fjardaal (Alcoa)

The tasks of this work package have been:

- A literature review of relevant reports from recent years
- To gather relevant discharge data from smelters
- Construction of a database
- Statistical analysis and graphical presentation

The literature review (review of reports from environmental surveys in vicinity of the smelters) was focused towards polycyclic aromatic hydrocarbons (PAHs) and heavy metal data in matrices such as sediment, porewater, mussels, seawater, fish, benthic invertebrates and other organisms on which data would be available.

The deliverables of this work package are:

- A database containing relevant information from the 10 smelter sites on information gathered on PAHs and heavy metals in various matrices.
- A statistical analysis of trends.
- This report.

2 Methods

2.1 Literature review of relevant reports from recent years

NIVA has conducted several environmental surveys in vicinity of Norwegian aluminium smelters. The reports produced by NIVA are registered in NIVAs archive, and data from several of these surveys have been transferred to NIVAs central data base, "*Aquamonitor*".

Information was also gathered by contacting the smelters (in Norway, Sweden and Iceland), regarding data on environmental concentrations not included in the NIVA database.

Data from NIVAs central database and the additionally acquired data were merged in a common database (available as a Microsoft Excel file), functioning as the data source for trend analyses (chapter 2.2) and for the graphical data displaying tool developed within this work package (chapter 2.3).

The reports were also reviewed for a summary of the environmental status at the specific sites.

2.2 Statistical trend analysis

Statistical trend analyses were performed using R software version 3.6.1. A time series was defined as the data for a given station, matrix (e.g. sediment, or the combination of species and tissue) and analyte/contaminant. For each time series, we performed linear regression on the annual median values. The analyses were performed only for time series where the data available were sufficient. As such, criteria adopted from OSPAR (the Oslo-Paris Convention for the protection of the marine environment of the North-East Atlantic) were used: statistical trend analyses were run on time series where there are at least 5 years of data, counting only years when at least half of the concentrations are higher than the analytical limit of quantification (>LoQ).

Some very crude predictions, in terms of the development in concentrations (and when to expect concentrations ≤EQS), were made based on the slopes of linear temporal trends. These prospects are presented in Chapter 3.5.

2.3 Graphical data displaying tool

A graphical data displaying tool was developed within this work package, using R software, and the R package "Shiny", specifically, for visualisation. This software allows for relatively easy making custom interactive tools for data visualisations. The tool is meant to be intuitive and user friendly, with roll-down menus, check boxes, graphical presentation of data (graphs) and a geographical interface (presentation on map).

The above mentioned data source and the graphical data tool is preliminary located on a test site at NIVA for easy access https://espial.test.niva.no/. However, it will later be migrated to an appropriate web address at NIVA.

3 Results and discussion

3.1 Literature review of relevant reports from recent years

As mentioned, relevant data from NIVAs central database and additionally acquired data, from the literature review, were merged in a common database (available as a Microsoft Excel file). Acquiring data from reports was a time consuming and laborious task, as the data was available in different degrees and on different forms. As such, more data will most likely be entered into the data base, also after the statistical trend analyses have been performed. They will then instantly be available to view also in the Graphical data displaying tool.

An overview of reports reviewed and from which data were (and are) added to the data base is presented in **Table 1**. Data from the Swedish and Icelandic smelters (**Table 1**) are currently formatted for inclusion in the data base.

Table 1. Reports reviewed and from which data were added to the ESPIAL data base. The reports are sorted by different smelters.

Årdalsfjorden	**Report no.**
Overvåking av Årdalsfjorden 1983. En tiltaksorientert undersøkelse av forurensninger fra aluminiumsindustri og befolkning.	NIVA report: 1870-1985
Overvåking av polysykliske aromatiske hydrokarboner (PAH) i o-skjell fra Årdalsfjorden 1992	NIVA report: 2811-1992
Overvåkning av PAH i o-skjell Årdalsfjorden 1994, med orienterende analyser av dioksiner og non-orto PBC.	NIVA report: 3248-1995
O-skjell fra årdalsfjorden. Polysykliske aromatiske hydrokarboner (PAH) i o-skjell fra Årdalsfjorden 2006 og 2007	NIVA report: 5553-2008
Sammenstilling av tungmetallinnhold i resipientene til norske aluminiumsverk	NIVA report: 5907-2010
Overvåking av Årdalsfjorden i 2011	NIVA report: 6185-2011
Tiltaksrettet overvåking av Årdalsfjorden i henhold til vannforskriften. Overvåking for Hydro Aluminium Årdal Karbon, Hydro Aluminium Årdal Metallverk og Norsun	NIVA report: 6987-2016
Tiltaksrettet overvåking av Årdalsfjorden i 2017. Overvåking for Hydro Aluminium Årdal.	NIVA report: 7248-2018
Tiltaksorientert overvåking av Årdalsfjorden i 2018. Overvåking for Hydro Aluminium Årdal	NIVA report: 7344-2019
Karmsundet	**Report no.**
Tiltaksorientert overvåking av Karmsundet. Undersøkelse av sedimenter, bløtbunnsfauna og miljøgifter i organismer.	NIVA report: 2284-1989
Joint Assessment and Monitoring Programme (JAMP) Contaminant and effects data for sediments, shellfish and fish 1981-2006	NIVA report: 5562-2008
Forurensningssituasjonen i Karmsundet i 2008 med vekt på påvirkning fra Hydro Aluminium Karmøy. Metaller, PAH og klorerte forbindelser i vannmasser, blåskjell, torsk, krabbe og sedimenter	NIVA report: 5881-2009
Tiltaksrettet overvåking for Hydro Aluminium Karmøy AS i 2015, i henhold til vannforskriften	NIVA report: 7012-2016
Tiltaksrettet overvåking for Hydro Aluminium Karmøy i 2017	NIVA report: 7247-2018
Tiltaksorientert overvåking av karmsundet i 2019. overvåking for Hydro Aluminium Karmøy,	NIVA report: 7466-2020

Determining the effects of the discharge effluent from the Karmøy aluminium smelter using an integrated biological effects approach	NIVA report: 2022
Mosjøen	**Report no.**
Overvåking av PAH i sjøområdet utenfor Elkem Aluminium Mosjøen ANS i forbindelse med utvidelse og omlegging til Prebaketeknologi	NIVA report: 4906-2004
Vefsnfjorden som resipient for avfall fra Mosjøen aluminiumverk. Rapport 1. Undersøkelser 1978-1980	NIVA report: 1330-1981
Tiltaksrettet overvåking av Vefsnfjorden i henhold til vannforskriften. Overvåking for Alcoa Mosjøen	NIVA report: 6976-2016
Overvåking av Vefsnfjorden, Sunndalsfjorden og Årdalsfjorden 2000. PAH, klorerte forbindelser og metaller i organismer og sedimenter, sammensetning av bløtbunnsfauna.	NIVA report: 4440-2001
Tiltaksrettet overvåking av Vefsnfjorden i 2017. Overvåking for Alcoa Mosjøen	NIVA report: 7242-2018
Overvåking i Vefsnfjorden for Mosjøen aluminiumverk 1989. Delrapport 1. sedimenter.	NIVA report: 2521-1991
Overvåking av Vefsnfjorden 2006. PAH, metaller og klororganiske forbindelser i organismer og sedimenter, bunnfauna i sedimenter	NIVA report: 5329-2007
Overvåking i Vefsnfjorden for Mosjøen aluminiumverk 1984	NIVA report: 1876-1986
Sunndalsfjorden	**Report no.**
Tiltaksrettet overvåking av Sunndalsfjorden i henhold til vannforskriften. Overvåking for Hydro Aluminium Sunndal	NIVA report: 6980-2016
Tiltaksorientert overvåking av Sunndalsfjorden, Møre og Romsdal. Delrapport 1. Sedimenter og bløtbunnsfauna 1986.	NIVA report: 2093-1988
Tiltaksorientert overvåking av Sunndalsfjorden, Møre og Romsdal, 1986-88. Konklusjoner	NIVA report: 2425-1990
Overvåking av PAH i muslinger snegl og fisk fra Sunndalsfjorden 1991-1992 Overvåkingsrapport nr. 504/92	NIVA report: 2818-1992
Overvåking av Vefsnfjorden, Sunndalsfjorden og Årdalsfjorden 2000. PAH, klorerte forbindelser og metaller i organismer og sedimenter, sammensetning av bløtbunnsfauna.	NIVA report:4440-2001
Kartlegging av miljøtilstand i fjordområdet ved Rausand, Sunndalsfjorden i 2003. Undersøkelser utført for Aluvest AS.	NIVA report: 4727-2003
Kontroll av utslipp av tundgmetaller fra Hydro Aluminium Sunndal	NIVA report: 5935-2010
Tiltaksrettet overvaking av Sunndalsfjorden i 2017. Overvaking for Hydro Aluminium Sunndal.	NIVA report: 7246-2018
Tiltaksorientert overvåking av Sunndalsfjorden basert på overvåkingsdata fra 2019. Overvåking for Hydro Aluminium Sunndal	NIVA report: 7559-2020
Integrated biological effects assessment of the discharge water into the Sunndals fjord from an aluminium smelter.	NIVA report: XXXX-2022
Høyangsfjorden	**Report no.**
Undersøkelser i Høyangsfjorden 1997.Miljøgifter i sedimenter og o-skjell. Sammensetningen av bløtbunnsfauna.	NIVA report: 3807-1998
Tiltaksorientert overvåking av Høyangsfjorden i 2018.	NIVA report: 7345-2019
Tiltaksrettet overvåking av Høyangsfjorden i henhold til vannforskriften. Overvåking for Hydro Høyanger.	NIVA report: 6973-2016
Kubal	**Report no.**
Kubikenborg Aluminium AB Sundsvall. Assessment of Sediment and Mussel Monitoring Results from Bay.	Byrne Ref: 326X019. FBS: 326: 07.06.06 November 2003
Analyses of polyamoratic compounds (PACs) and metals in sediment and mussels near Kubal aluminium smelter	NIVA report: 7547-2020
ISAL	**Report no./Reference**
A survey of intertidal organisms around dumping pits for pot linings at Straumsvik. Institute of Biology, University of Iceland.	Ingólfsson, A (1990) Report No. 27b
Studies on the rocky subtidal communities in vicinity of a dumping pit for pot linings at Straumsvik, Southwestern Iceland. Institute of Biology, University of Iceland.	Svavarsson, J (1990) Report No. 28
The environmental impact of dumping pits for potlinings and filterdust from ISAL aluminium smelter at Straumsvik. A review of research carried out on the biotic diversity and accumulation of heavy metals and PAH in organisms. Institute of Biology, University of Iceland.	Gíslason, G.M (1998) Report No. 42b
The communities of the Hraunsvik coast east of Straumsvik. Institute of Biology, University of Iceland.	Ingofsson A., Steinarsdottir, M.B. (2002) Report No. 64.

The communities of the rocky subtidal in Hraunsvik east coast of Straumsvik. Institute of Biology, University of Iceland.	Svavarsson, J. (2002) Report No. 65
Examination of trace elements and polyaromatic hydrocarbons (PAHs) in blue mussels (*Mytilus edulis*) and the brown seaweed *Fucus distichus (arctic wrack)* at the aluminium smelter at Straumsvík.	Auðunsson, G.A., Árnadóttir, E., Halldórsdóttir, H (2005)
Könnun á ólífrænum snefilefnum og aromatískum fjölhringjasamböndum (PAH) í kræklingi og skúfþangi við álverið í Straumsvík, sýnataka 2008. / Survey of inorganic trace elements and aromatic polycyclic compounds (PAH) in mussels and shingles at the plant in Straumsvík, sampling 2008.	Auðunsson, G.A (2012) NMI 12-01
Könnun á ólífrænum snefilefnum og aromatískum fjölhringa- samböndum (PAH) í kræklingi við álverið í Straumsvík. Sýnataka 2013 / Survey of inorganic trace elements and aromatic polycyclic ring relationships (PAH) in mussels at the Straumsvík smelter. Sampling 2013.	Auðunsson, G.A (2014) NMI 14-06 (6EM13087)
Alcoa Fjardaál	**Report no./Reference**
Sjálfbærnivísir:Mengunarefni í dýralífi sjávar / Sustainability Indicators: Wildlife pollutants in the sea. Fjardaal, Alcoa.	HRV Engineering, (2010)
Alcoa Fjarðaál Umhverfisvöktun 2015 Skýrsla unnin af Náttúrustofu Austurlands og Nýsköpunarmiðstöð Íslands fyrir Alcoa Fjarðaál / Alcoa Fjardaál Environmental monitoring 2015. A report prepared by the East Icelandic Nature Agency and the Icelandic Innovation Center for Alcoa Fjardaál.	NA-160160 Neskaupstaður (2016)
Sør-al Husnes	**Report no.**
Resipientundersøking i Kvinnherad 1990	NIVA report: 2565-1991
Tiltaksrettet overvåking av Husnesfjorden i henhold til vannforskriften, 2015 Overvåking for Sør-Norge Aluminium AS	NIVA report: 6978-2016
Tiltaksorientert overvåking av Husnesfjorden i 2018. Overvåking for Hydro Aluminium Husnes.	NIVA report: 7346-2019
Alcoa Lista	**Report no.**
Tiltaksrettet industriovervåking iht. vannforskriften for Alcoa Lista. EUs prioriterte miljøgifter og vannregionspesifikke stoffer i sjøvann og organismer.	NIVA report: 6974-2016

The latest reports from the different sites (**Table 1**) were reviewed for an update on environmental status. This is summarized in chapter 3.4.

3.2 Statistical trend analysis

At the time of the performance of the statistical trend analyses, the database contained a total of 21880 datapoints shared among all areas/smelters/stations, sample types/matrices and analytes (**Table 2**). These datapoints comprised then 8351 potential time series (specific analyte in specific matrix at specific station; (**Table 3**). As mentioned, statistical trend analyses were run on compounds/matrices/stations where there are at least 5 datapoints (data from at least 5 years) and at least half of the data are concentrations higher than the analytical limit of quantification (>LoQ). 311 time series fulfilled these criteria (**Table 4**).

Table 2. Number of data points (all analytes and stations), specified for matrix/sample type and smelter.

	Biota	Sediment	Water	Total
Alcoa Lista	413	0	138	**551**
Alcoa Mosjøen	1544	1146	1085	**3775**
Hydro Høyanger	1081	743	0	**1824**
Hydro Karmøy	1357	861	0	**2218**
Hydro Sunndal	3520	2475	0	**5995**
Hydro Husnes	529	506	0	**1035**
Hydro Årdal	4459	2023	0	**6482**
Total	**12903**	**7754**	**1223**	**21880**

Table 3. Number of potential time series (for specific compounds in specific matrix at specific stations) represented by the data in the data base. Here specified for matrix and smelter.

	Water	Sediment	Blue mussel	Northern horsemussel	Snails	Fish	Seaweed	Total
Alcoa Lista	43	0	145	0	0	0	0	**188**
Alcoa Mosjøen	92	614	532	82	46	15	2	**1383**
Hydro Høyanger	0	283	325	0	0	0	0	**608**
Hydro Karmøy	0	367	312	0	69	0	0	**748**
Hydro Sunndal	0	1357	541	211	217	115	107	**2548**
Hydro Husnes	0	186	187	0	0	0	0	**373**
Hydro Årdal	0	725	525	1253	0	0	0	**2503**
Total	**135**	**3532**	**2567**	**1546**	**332**	**130**	**109**	**8351**

Table 4. The number of time series fulfilling the criteria of having at least 5 datapoints (data from at least 5 years) and that at least half of the data are concentrations higher than the analytic limit of quantification (>LoQ). Specified for matrix/sample type and smelter (**A.**) or analyte group (**B.**).

A.	**Water**	**Sediment**	**Blue mussel**	**Northern horsemussel**	**Snails**	**Fish**	**Seaweed**	**Total**
Alcoa Mosjøen	18	0	0	0	0	0	0	**18**
Hydro Karmøy	0	0	8	0	16	0	0	**24**
Hydro Sunndal	0	0	118	0	0	0	0	**118**
Hydro Årdal	0	0	0	151	0	0	0	**151**
Total	**18**	**0**	**126**	**151**	**16**	**0**	**0**	**311**

B.	**Water**	**Sediment**	**Blue mussel**	**Northern horsemussel**	**Snails**	**Fish**	**Seaweed**	**Total**
Biomarkers	0	0	0	0	1	0	0	**1**
Chlorobiphenyls (PCBs)	1	0	0	0	0	0	0	**1**
Dichloro-diphenyl-trichloroethane (DDTs)	0	0	4	0	8	0	0	**12**
Hexachlorocyclohexanes	1	0	0	0	0	0	0	**1**
Metals and metalloids	7	0	0	0	0	0	0	**7**
Organo-metallic compounds	0	0	5	0	5	0	0	**10**
Others	9	0	3	9	1	0	0	**22**
Polycyclic aromatic hydrocarbons (PAHs)	0	0	114	142	0	0	0	**256**
Total	**18**	**0**	**126**	**151**	**16**	**0**	**0**	**311**

As can be read from **Table 4**, data sufficient for time trend analyses are mostly relevant in water samples outside Alcoa Mosjøen (e.g. metals), in blue mussel (*Mytilus edulis*) samples outside Hydro Sunndal (particularly PAH compounds), and in horsemussel (*Modiolus modiolus* ;in Norwegian: "o-skjell") outside Hydro Årdal (also particularly PAH-compounds). Furthermore, there are time series to analyse for snails outside Hydro Karmøy. Among these are one time series on "Biomarkers". More specifically this is a measure of imposex (superimposition of male sexual characters onto females), measured as Vas Deferens Sequence Index (VDSI). Imposex is an irreversible pseudohermaphrodism disorder in females of certain sea snails caused by the effect of prevoiusly used organotin antifouling agents on ships, especially tributyltin (TBT) (Bryan et al. 1986; Schøyen et al. 2019).

The number of statistically significant time trends can be read from **Table 5**. The table shows that most statistically significant trends were found for PAH-compounds. **Figure 1** shows the number of statistically significant and non-significant time-trends for concentrations of different PAHs in different matrices/samples types, at different stations. All statistically significant trends are negative (downward), and the general picture is a reduction in PAH concentrations with time. **Figure 2** shows the same, however, for individual PAH compounds.

Figure 3 to **Figure 13** presents a selection of time series/analytes and the results of the statistical trend analyses. As indicated, some of these trends are statistically significant and some are not. It is a selection of samples of different matrices and at different localities. **Figure 3** and **Figure 4** show statistically significant downward trends of chromium (Cr) and lead (Pb), respectively, in water from a station a few km out the fjord from Alcoa Mosjøen, during the years 1992-2003. Note that the source of pollution is likely not the smelter, Drevjamoen shooting and training field is situated a few km north-east of the site. It is worth noting also that there are emissions of leachate from landfills along the Vefsnfjord (near Rynes; Øxnevad and Hjermann, 2020). **Figure 5** to **Figure 7** show the variation in concentrations of PAH-compounds (benzo[a]pyrene, fluoranthene and pyrene, respectively) in blue mussel at a station some km out the fjord from Hydro Sunndal, during the years 1995-2011 (fluoranthene and pyrene show statistically significant downward trends). **Figure 8** to **Figure 11** show the variation in concentrations of PAH-compounds (benzo[a]pyrene, fluoranthene, phenanthrene and pyrene, respectively) in Northern horsemussel at a station a few km out the fjord from Hydro Årdal, during the years 1983-2015 (only phenanthrene shows a statistically significant downward trends). **Figure 12** shows a statistically significant downward trend in VDSI in dogwhelk from a station in Karmsundet, approximately one km north of Hydro Karmøy, during the years 1997-2010. The downward trend in VDSI corresponds with a statistically significant downward trend in concentrations of TBT in blue mussel from a nearby (300 m) station (**Figure 13**; 2002-2010).

Table 5. Number of time series that show a significant change in concentrations with time. All matrices/sample types and stations are included. The single analytical parameters are assigned to different groups (PAHs, PCBs, DDTs, Metals and metalloids, organo-metallic compounds, biomarkers and hexachlorocyclohexanes). The results are given in intervals of percentage change per year. The numbers in each cell give the number of time series that show a statistical change with time, in the indicated change percentage interval for an analyte in the specified analyte group. The numbers in parentheses give the number of non-significant trends in the indicated percentage interval for analytes in the specified analyte group.

	Percentage change								
	(-100,-50]	**(-50,-20]**	**(-20,-10]**	**(-10,0]**	**(0,10]**	**(10,20]**	**(20,50]**	**(50,100]**	**NA**
Polycyclic aromatic hydrocarbons (PAHs)	1 (6)	11 (44)	2 (110)	0 (41)	0 (14)	0 (4)	0 (8)	0 (1)	0 (2)
Polychlorinated biphenyls (PCBs)		1 (0)							
Dichloro-diphenyl-trichloroethane (DDTs)		3 (4)	0 (4)		0 (1)				
Metals and metalloids		1 (0)	1 (1)	1 (2)				0 (1)	
Organo-metallic compounds		1 (1)	1 (6)						
Biomarkers				1 (0)	1 (0)				
Hexachlorocyclohexanes				0 (1)					

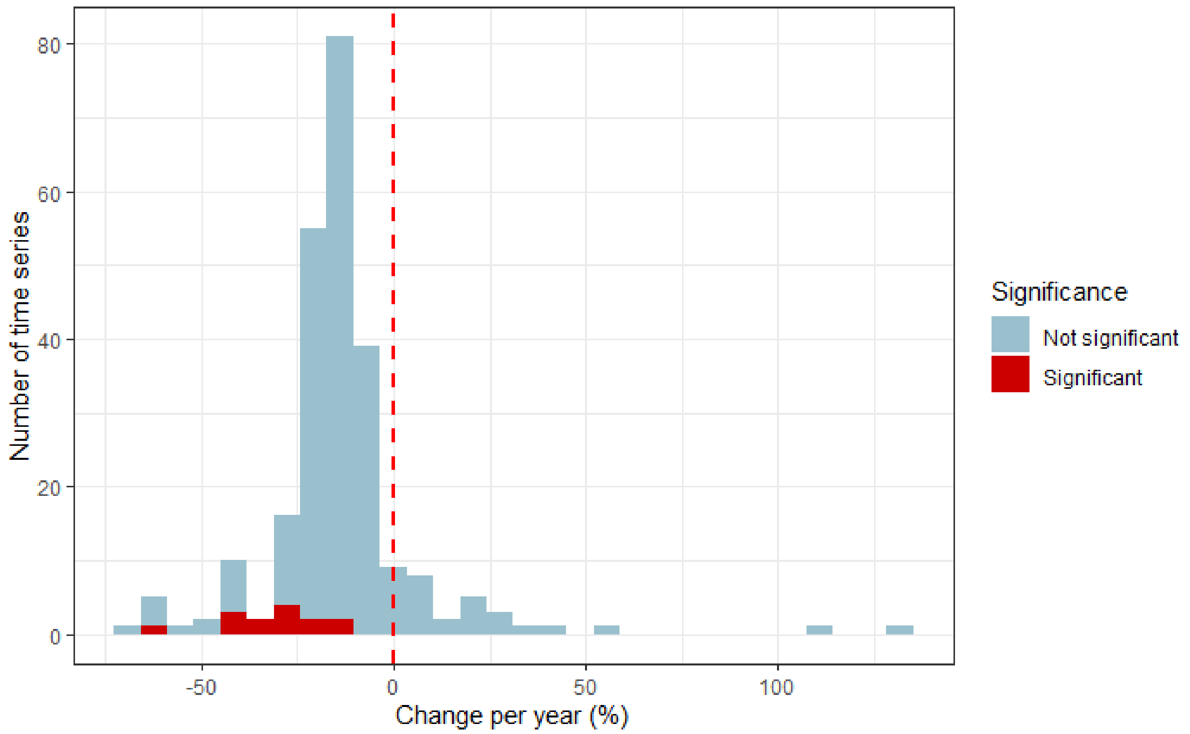


Figure 1. Number of statistically significant (red) and non-significant (blue) time-trends for concentrations of different polycyclic aromatic hydrocarbons (PAH-compounds) in different matrices/samples types, at different stations. Different intervals of percentage change per year are indicated.

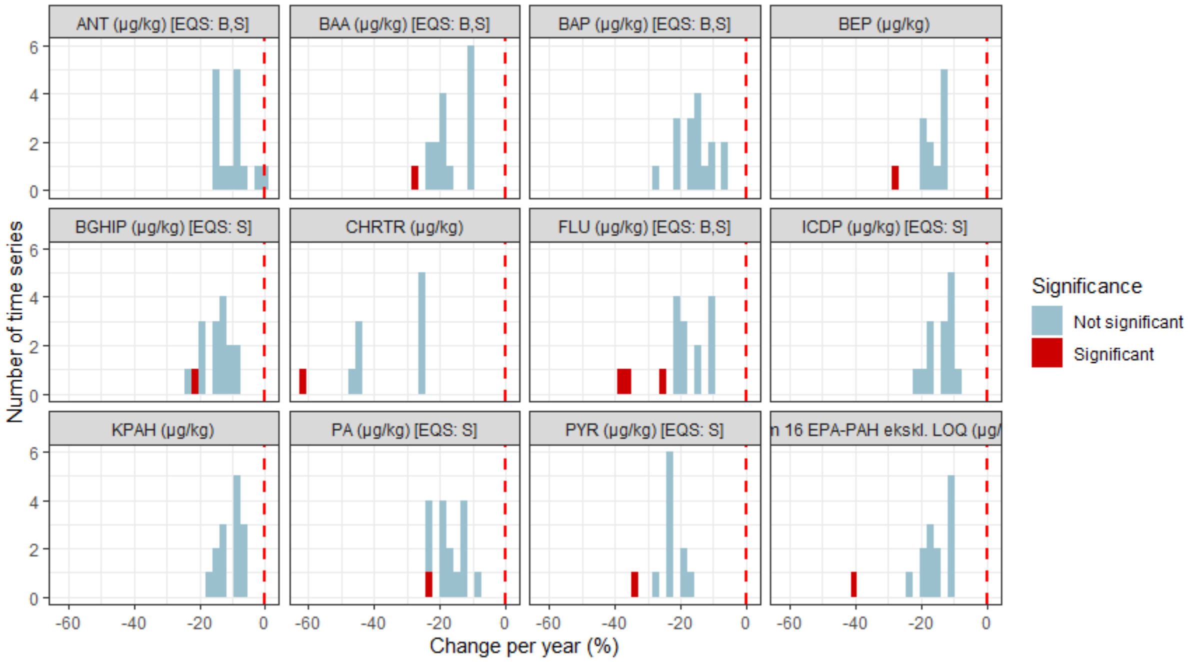


Figure 2. Number of statistically significant (red) and non-significant (blue) time-trends for concentrations of specified polycyclic aromatic hydrocarbons (PAH-compounds) in different matrices/samples types, at different stations. Different intervals of percentage change per year are indicated.

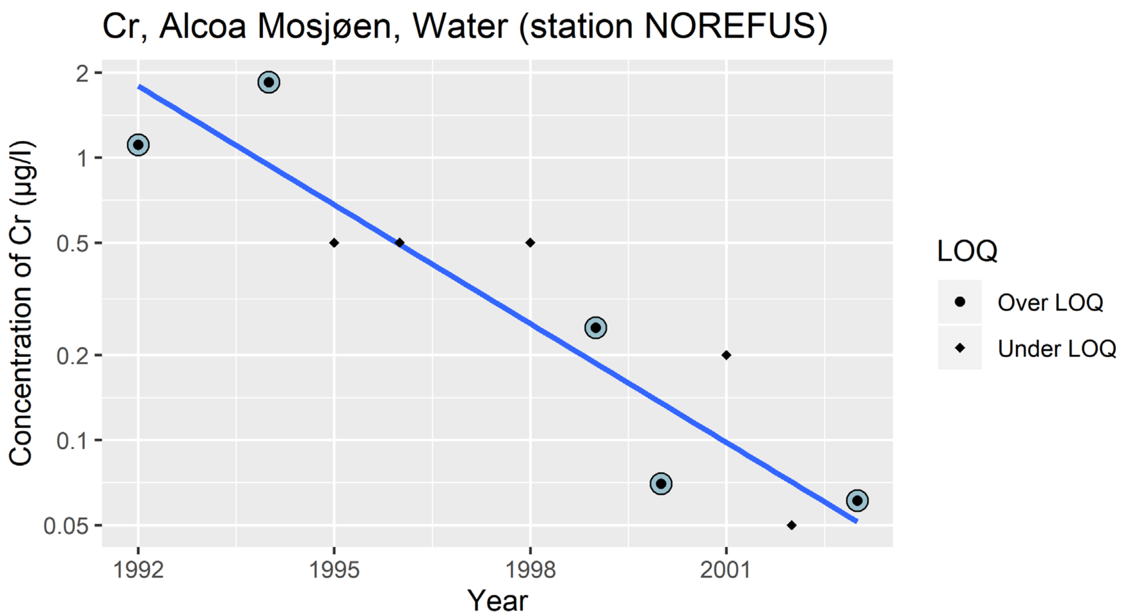


Figure 3. Chromium (Cr) in water from station NOREFUS outside Alcoa Mosjøen. There is a statistically significant downward trend; P=0.020109 (sign.); Change per year (%): -27.6; n=10; (Years: 1992 – 2003). Note that the source of pollution is likely not the smelter, Drevjamoen shooting and training field is situated a few km north-east of the site. Chromium is a river basin specific substance with an EQS of 3.4 µg/L in coastal water, i.e. above the scale presented here.

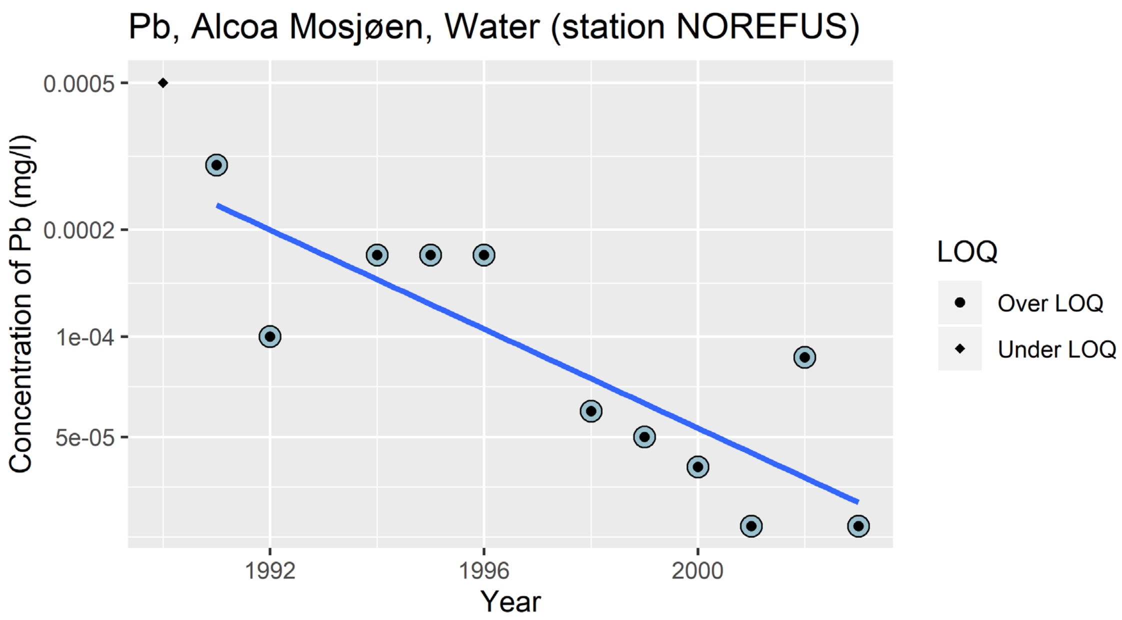


Figure 4. Lead (Pb) in water from station NOREFUS outside Alcoa Mosjøen. There is a statistically significant downward trend; P=0.001557 (sign.); Change per year (%): -13.9; n=12; (Years: 1990 – 2003). Note that the source of pollution is likely not the smelter, Drevjamoen shooting and training field is situated a few km north-east of the site. Lead is a priority substance with an EQS of 1.3 µg/L (0.0013 mg/L) in coastal water, i.e. above the scale presented here.

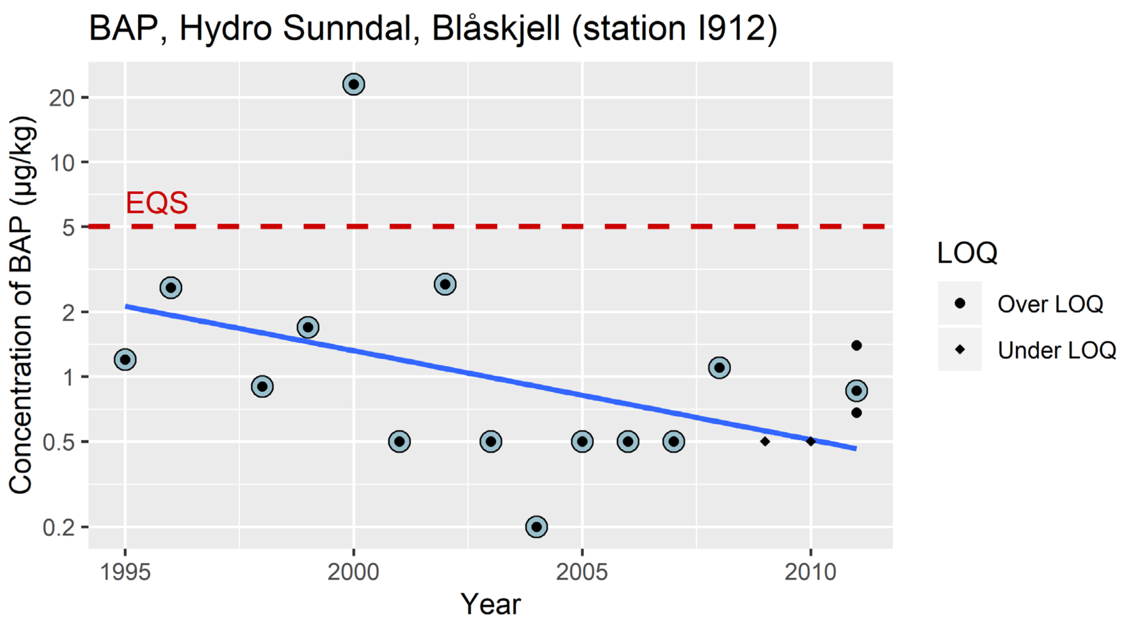


Figure 5. Benzo(a)pyrene in blue mussel from station I912 outside Hydro Sunndal. There is a downward tendency, however the trend is not statistically significant; P=0.169204 (not sign.); Change per year (%): -9.1; n=16; (Years: 1995 – 2011).

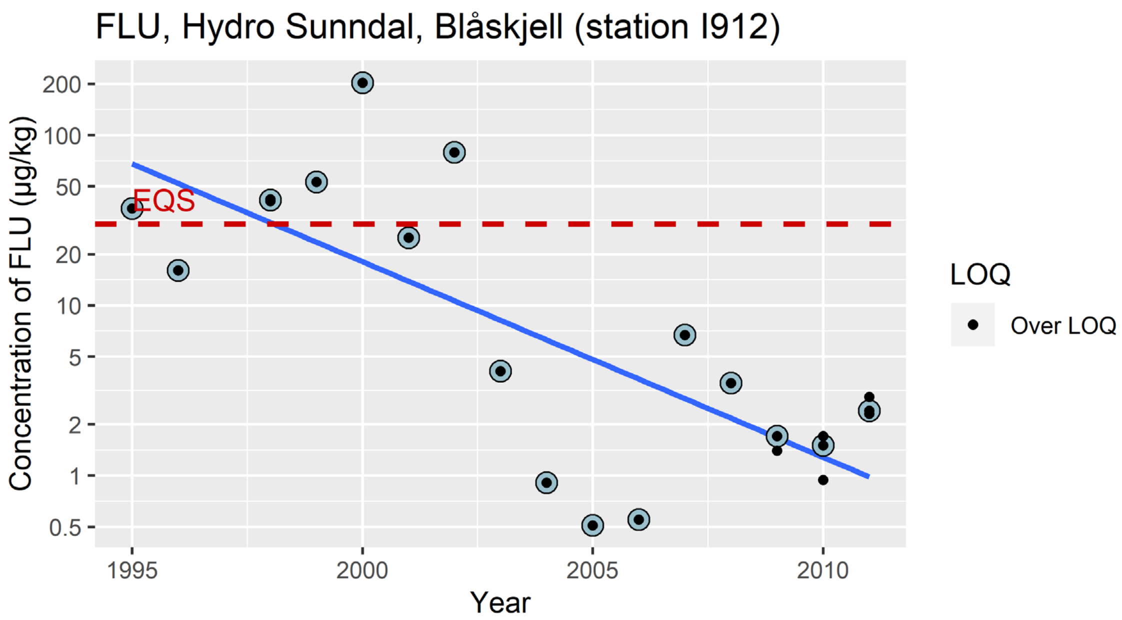


Figure 6. Fluoranthene in blue mussel from station I912 outside Hydro Sunndal. There is a statistically significant downward trend; P=0.002676 (sign.); Change per year (%): -23.3; n=16; (Years: 1995 – 2011).

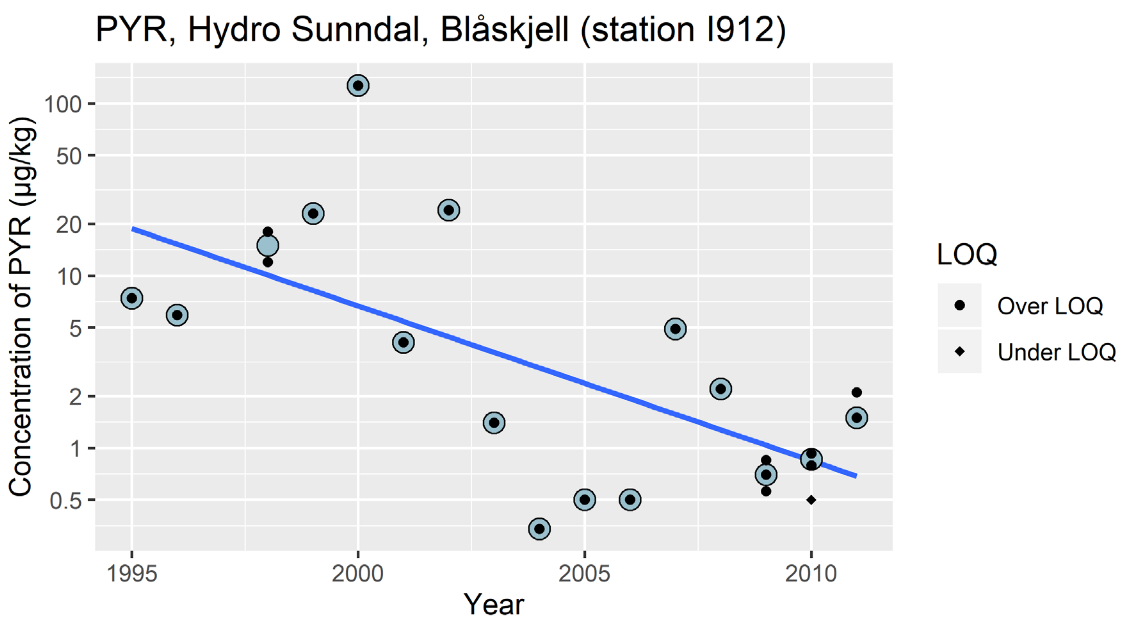


Figure 7. Pyrene in blue mussel from station I912 outside Hydro Sunndal. There is a statistically significant downward trend; P=0.012178 (sign.); Change per year (%): -18.7; n=16; (Years: 1995 – 2011).

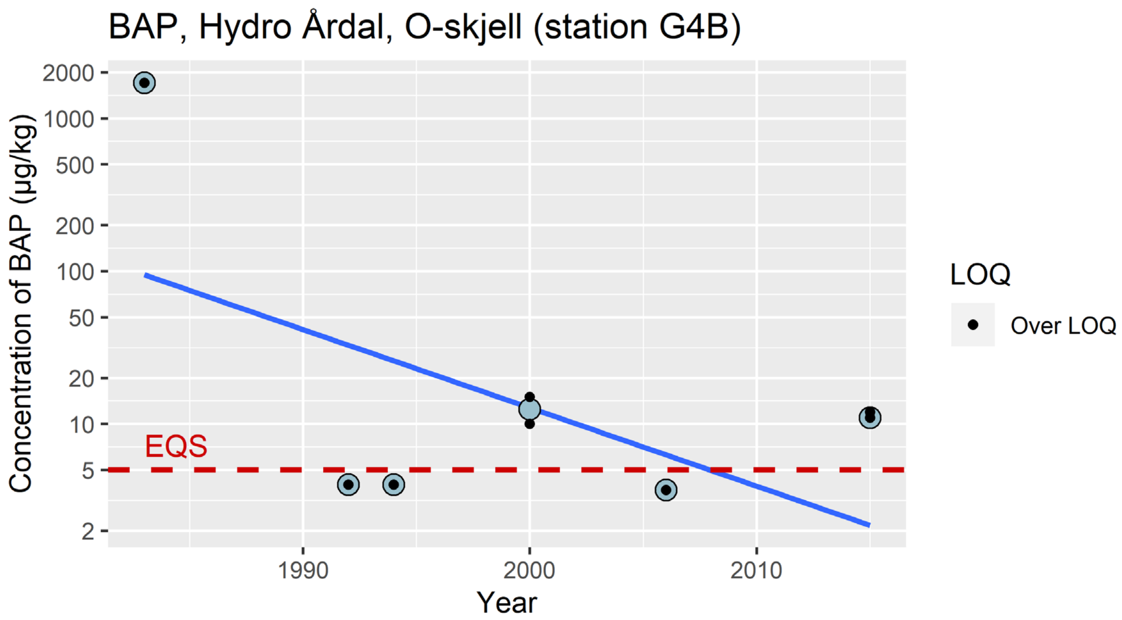


Figure 8. Benzo(a)pyrene in horsemussel from station G4B outside Hydro Årdal. There is a downward tendency, however the trend is not statistically significant; P=0.246697 (not sign.); Change per year (%): -11.1; n=6; (Years: 1983 – 2015).

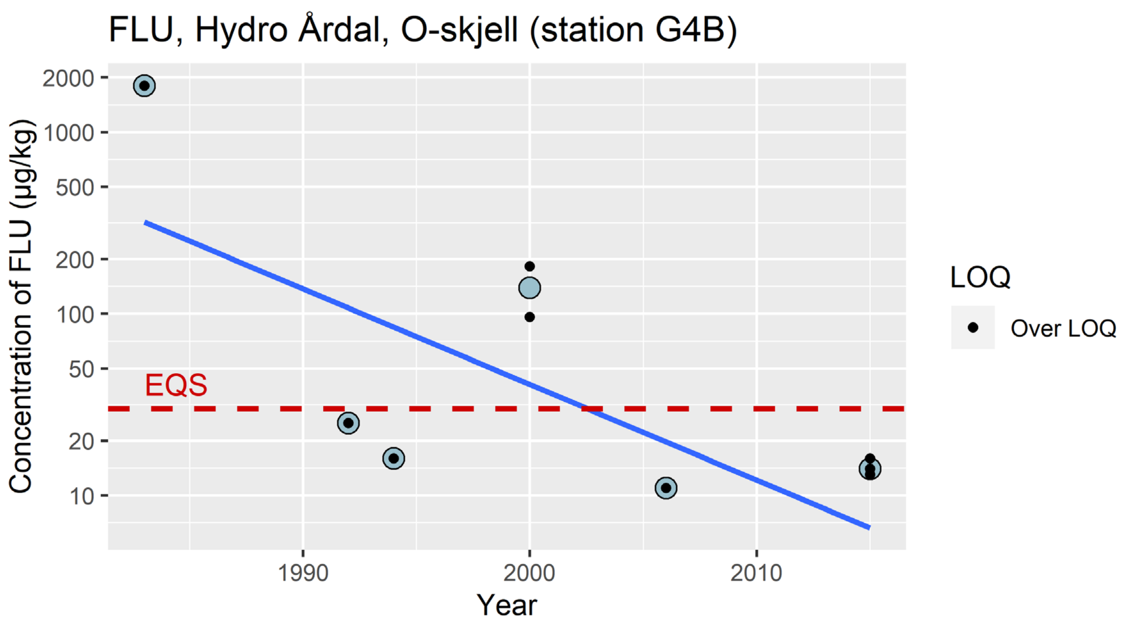


Figure 9. Fluoranthene in horsemussel from station G4B outside Hydro Årdal. There is a downward tendency, however the trend is not statistically significant; P=0.129923 (not sign.); Change per year (%): -11.4; n=6; (Years: 1983 – 2015).

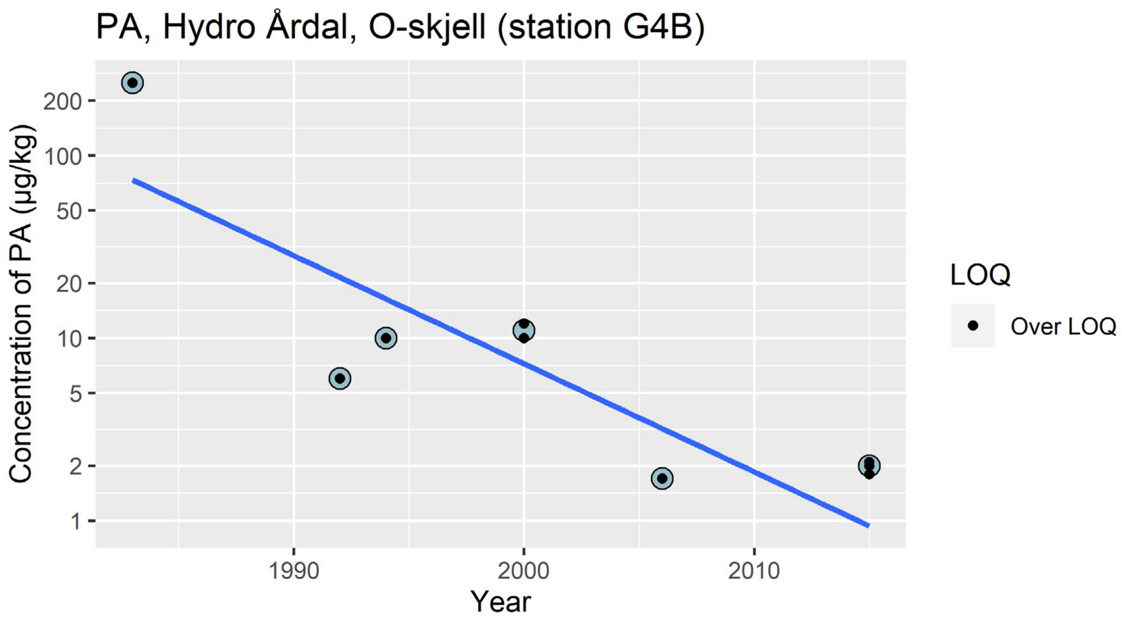


Figure 10. Phenanthrene in horsemussel from station G4B outside Hydro Årdal. There is a statistically significant downward trend; P=0.032092 (sign.); Change per year (%): -12.7; n=6; (Years: 1983 – 2015).

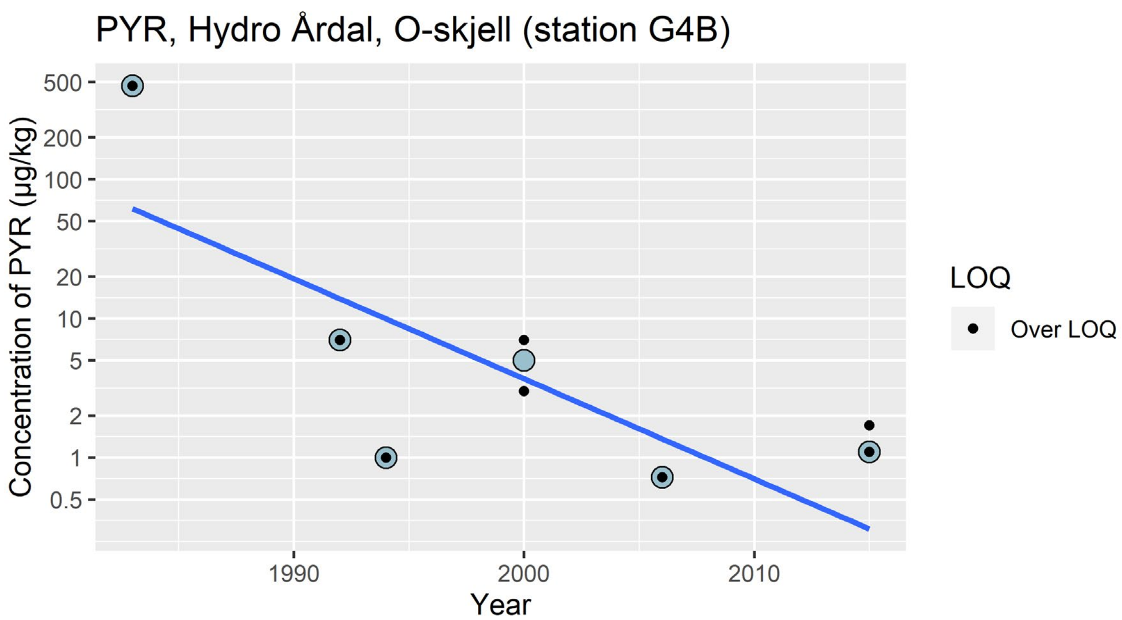


Figure 11. Pyrene in horsemussel from station G4B outside Hydro Årdal. There is a downward tendency, however the trend is not statistically significant; P=0.073871 (not sign.); Change per year (%): -15.3; n=6; (Years: 1983 – 2015).

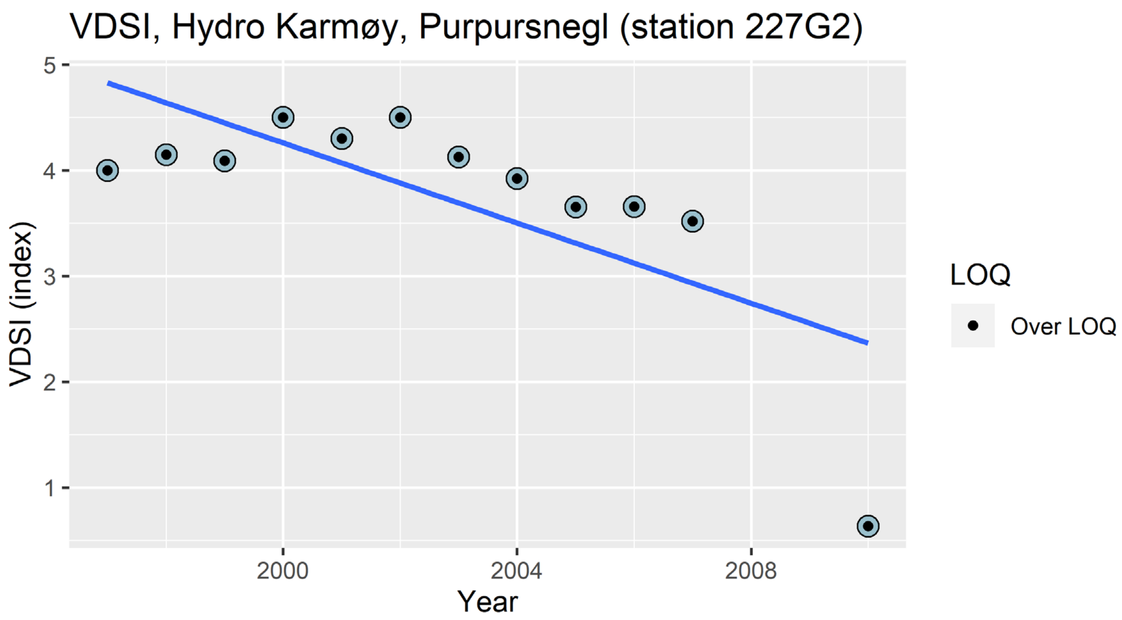


Figure 12. Vas Deferens Sequence Index (VDSI) in dogwhelk from station 227G2 outside Hydro Karmøy. There is a statistically significant downward trend; P=0.008348 (sign.); Change per year (%): -17.3; n=12; (Years: 1997 – 2010).

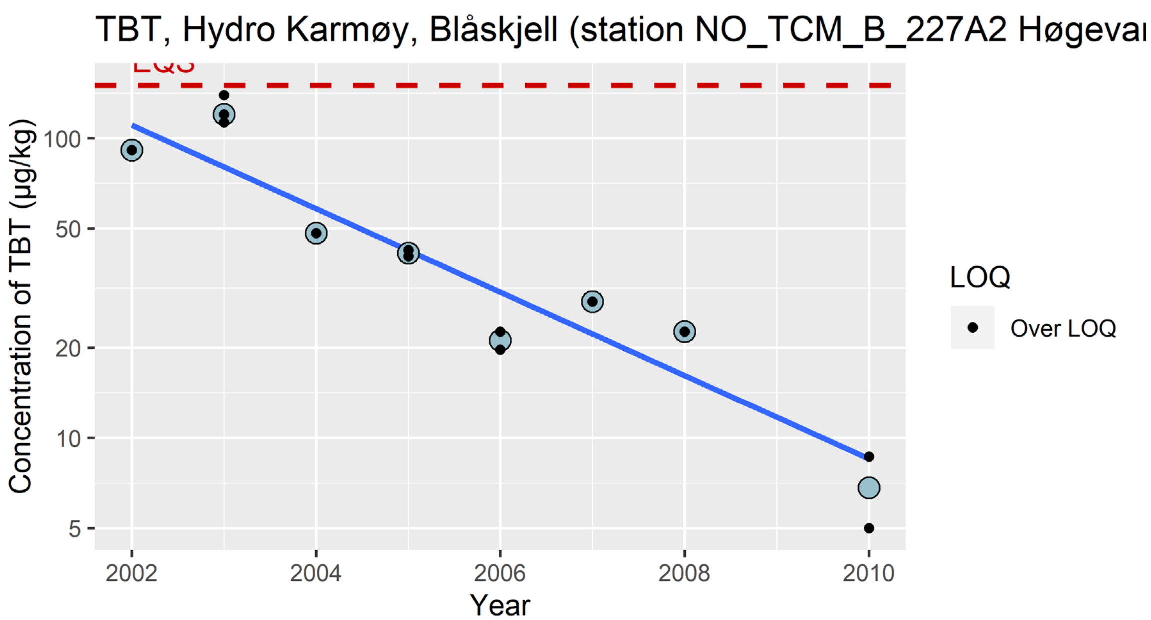


Figure 13. Tributyltin (TBT) in blue mussel from station 227A2 outside Hydro Karmøy. There is a statistically significant downward trend; P=0.00037 (sign.); Change per year (%): -27.4; n=8; (Years: 2002 – 2010).

3.3 Graphical data displaying tool

A Graphical data displaying tool was developed in this work package to make the data in the database accessible and to make it easy to display concentrations and trends of specific analytes in specific matrices at specific locations. As mentioned, we aspired to make the graphical data displaying tool intuitive and user friendly, with roll-down menus, check boxes, graphical presentation of data (graphs) and a geographical interface (presentation on map). The result is presented in **Figure 14** to **Figure 17**, where Hydro Sunndal is used as example. As shown, the interactivity (user interface) is maintained by use of roll down menus and check boxes, as well as a slider (to merge or separate stations at specific distances from each other).

The user may first choose the smelter of interest and will be presented with a map of the relevant area in the vicinity of this smelter. The stations from which there are data in the data base will be positioned in the map (stations are numbered and given a colour, to be recognised in the graphical output of concentration data). The user may then choose the matrix/matrices of interest (specifically biota, sediment or water). Regarding biota, there are data on several species available in the data base, and one may specify those that are of interest. The specific substance/analyte is chosen, first by "Substance group" and then by specific substance. The graphical output, in terms of concentration data, is a series of graphs (one for each year with data available), with the concentrations (as bars; linear scale) in the selected matrix/matrices (kept separate by different colours), at each of the available stations (**Figure 14**). One can choose a single matrix/species (**Figure 15**), and it is also possible to change the view to logarithmic scale on the concentration axis, e.g. when variability is high (**Figure 16**). The data will then be presented as points, instead of bars. For substances for which there are environmental quality standards (EQS), these are shown in the plots for comparative purposes (**Figure 15** to **Figure 17**).

The tool may also present time series of the selected data (**Figure 17**). In that case, data on the selected substance in the selected matrices are presented for each station combined in one graph with year as the abscissa and concentration as the ordinate.

Figure 18 and **Figure 19** illustrate how two of the time series analysed statistically (presented in **Figure 3** and **Figure 8**, respectively) are displayed in the graphical data displaying tool.

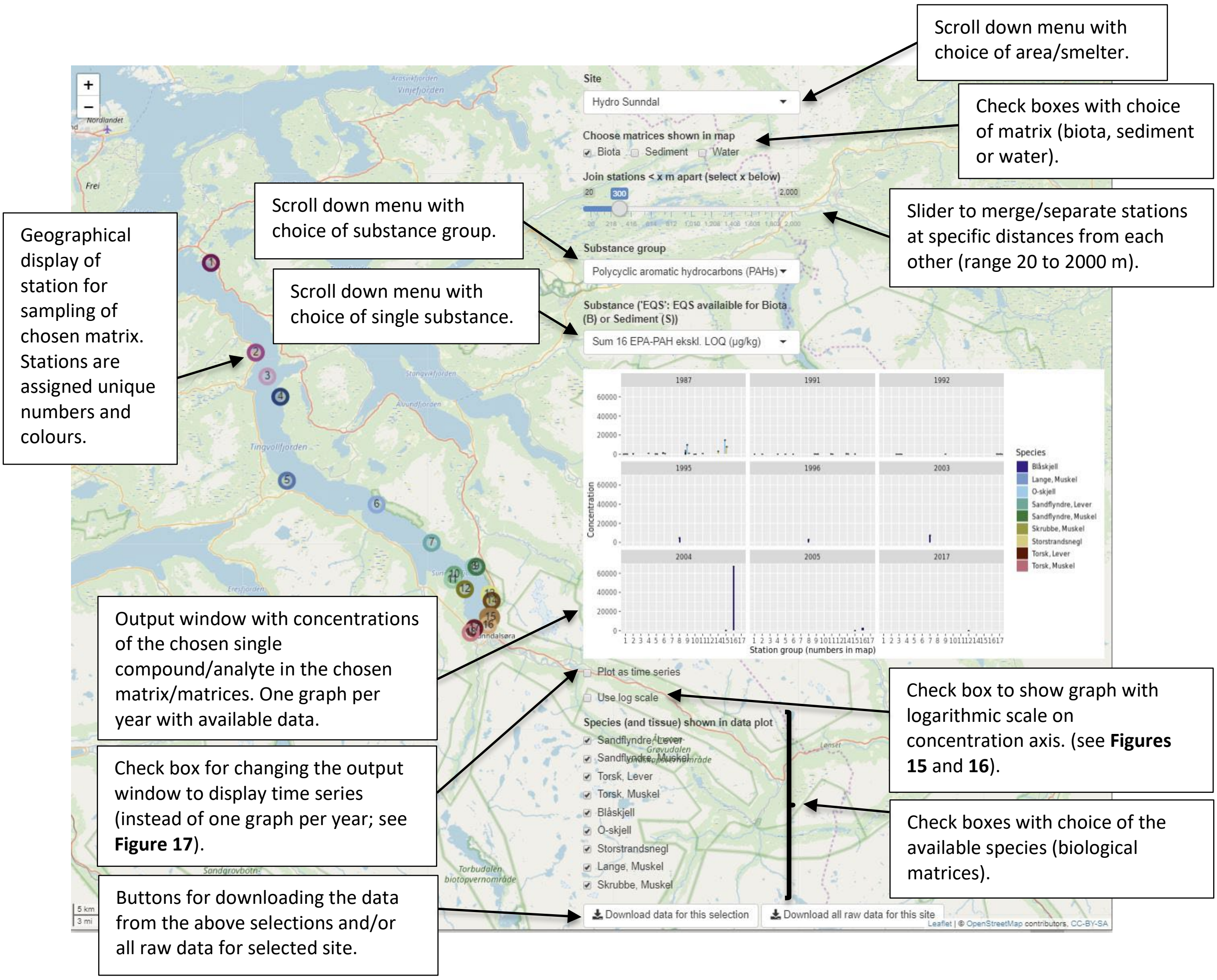


Figure 14. Screen dump of graphical data displaying tool with explanation of different tool elements. In this example, data on Sum 16PAH in different biological matrices at stations in the Sunndalsfjord is shown.

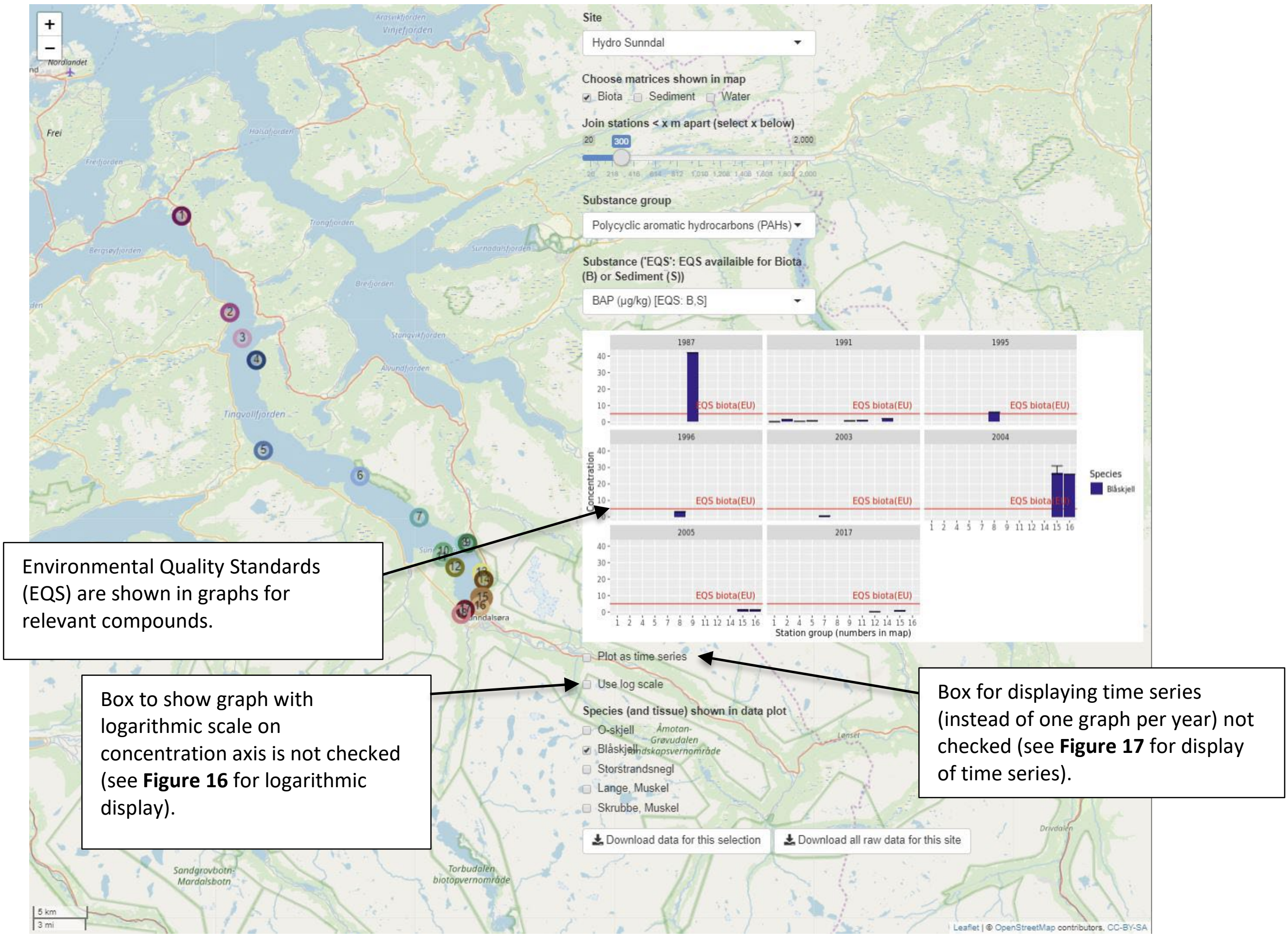


Figure 15. Screen dump of graphical data displaying tool. In this example, data on Benzo[a]pyrene in blue mussel at stations in the Sunndalsfjord are shown (data from 8 years at various station). Environmental Quality Standards (EQS) are shown to compare with the data (displayed as bars; linear scale). One graph for each individual year.

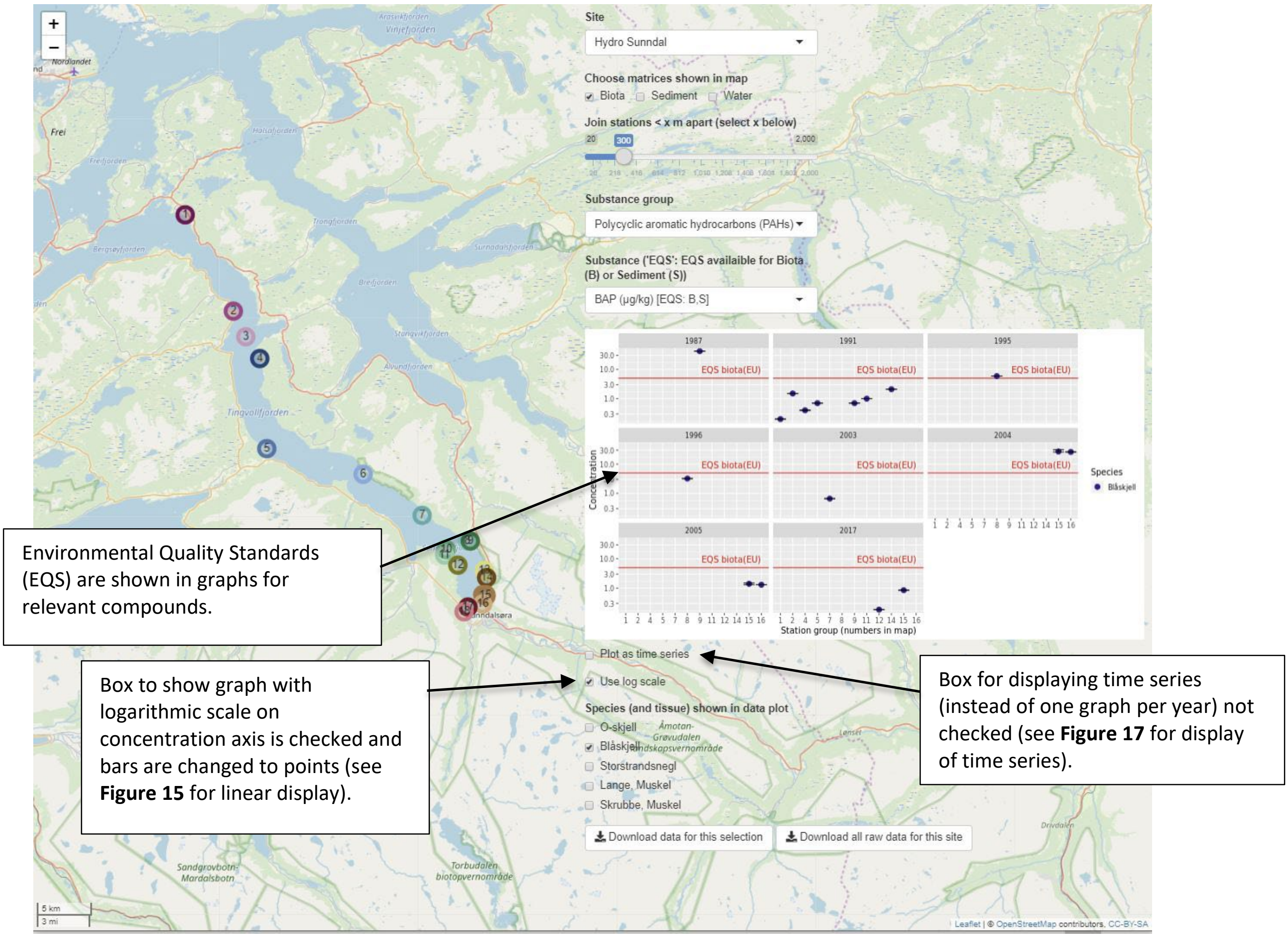


Figure 16. Screen dump of graphical data displaying tool. In this example, data on Benzo[a]pyrene in blue mussel at stations in the Sunndalsfjord is shown (data from 8 years at various station). Environmental Quality Standards (EQS) are shown to compare with the data (displayed as points; logarithmic scale). One graph for each individual year.

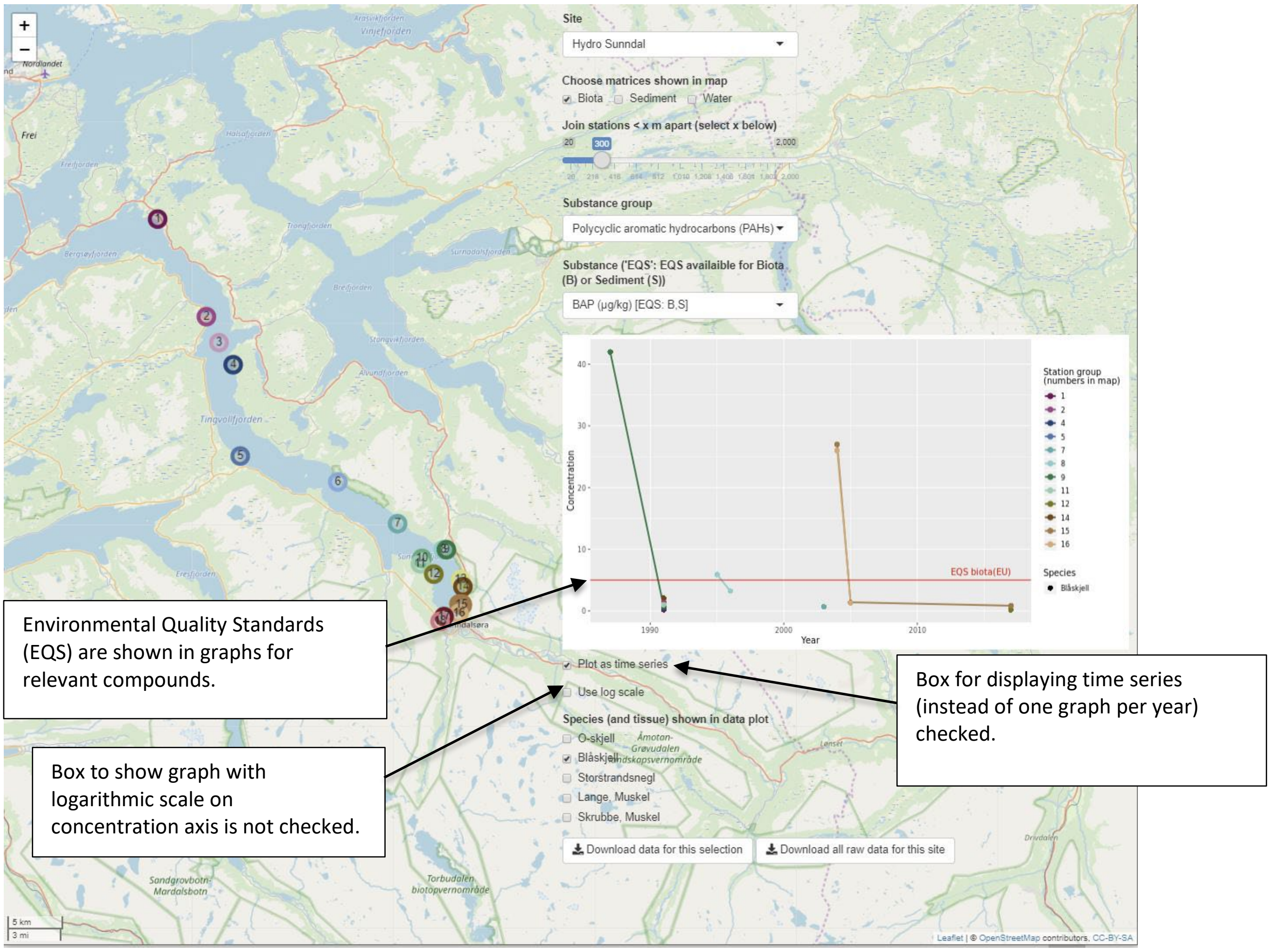


Figure 17. Screen dump of graphical data displaying tool. In this example, data on Benzo[a]pyrene in blue mussel at stations in the Sunndalsfjord is shown (data from 8 years at various station). Environmental Quality Standards (EQS) are shown to compare with the data. Time series of available data on each station is combined in one graph.

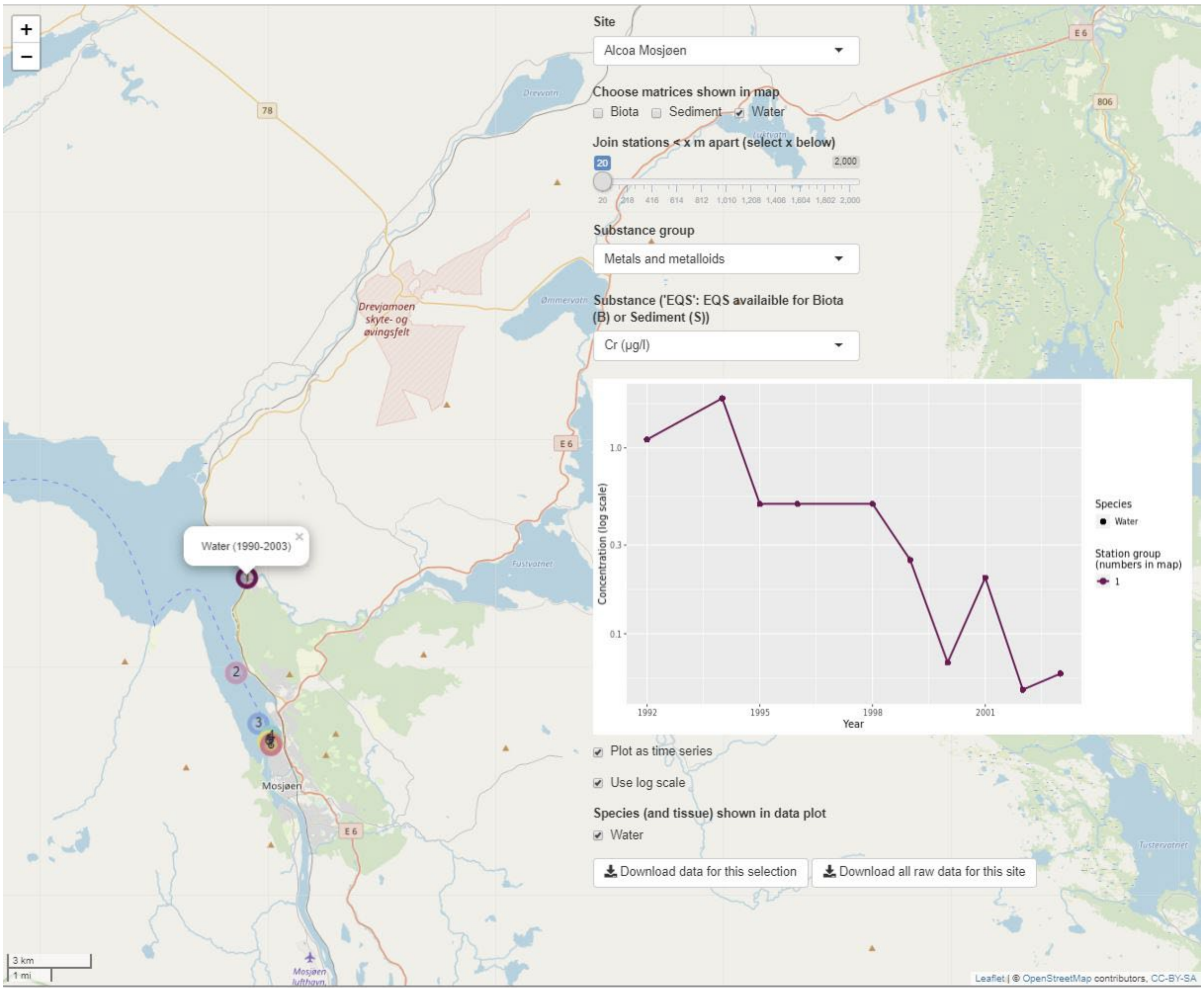


Figure 18. Chromium (Cr) in water at station NOREFUS outside Alcoa Mosjøen (1992-2003) as displayed in the graphical data displaying tool. By clicking on the station in the map, information regarding available data associated with the station is displayed. The selected time series corresponds with that displayed in **Figure 3** (in which results of the statistical time trend analysis is displayed).

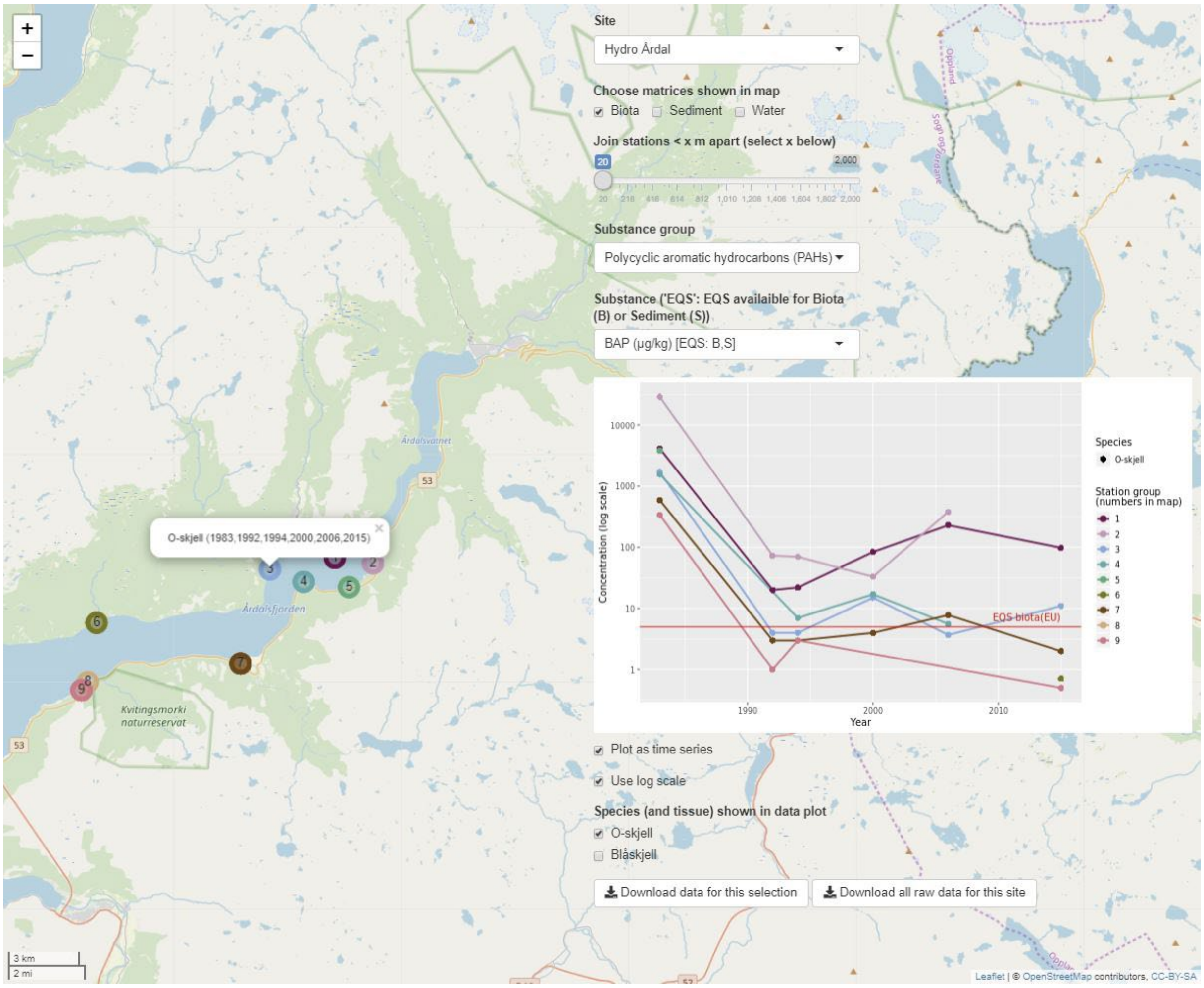


Figure 19. Benzo(a)pyrene in Northern horsemussel at station G4B outside Hydro Årdal (1983-2015) as displayed in the graphical data displaying tool. By clicking on the station in the map, information regarding available data associated with the station is displayed. Here, the station is assigned the number 3 and a light blue colour. The selected time series corresponds with that displayed in **Figure 8** (in which results of the statistical time trend analysis is displayed).

3.4 Status summary for each site

Below is a summary of results from the latest relevant reports from the smelter sites.

3.4.1 Sunndal

3.4.1.1 Biota

In 2019, NIVA monitored metals and PAHs in sediment and deployed mussels in Sunndalsfjord on behalf of the AMS. This data was reported as part of the ESPIAL project (Brooks et al., 2022a). The Norwegian Environment Agency approved the use of this ESPIAL data for reporting action-orientated monitoring of Sunndalsfjord for Hydro Aluminium Sunndal for 2019 (Øxnevad , 2021).
Mussels were deployed at corresponding sediment stations for a period of six weeks. After six weeks of exposure in Sunndalsfjord, there was no increase in the concentrations of PAH compounds. For

metals, one of the mussel stations had a concentration of mercury of 20 µg/kg wet weight, which is the limit value for mercury in biota according to the Water Framework Directive (WFD). The chemical status of that stations is therefore classified as “not good”. There were no exceedances of limit values at the other stations, and the chemical condition of the other four stations is therefore classified as “good”.

There were no exceedances of the quality standards for the priority substances at any of the blue mussel stations in 2017 (Øxnevad et al. 2018). Thus, Sunndalsfjorden was classified to "good chemical status". For three of the stations there were concentrations of zinc that was slightly above “high provisional background concentrations”. Low concentrations of PAH-compounds were observed in the mussels.

Trend analyses that have been performed on blue mussel samples showed two statistically significant downward trends; for benzo(a)pyrene and fluoranthene.

In 2015, analyses of PAH-compounds and metals in samples of horsemussels and common periwinkle (*Littorina littorea*) were done (Borgersen et al. 2016). Common periwinkle at the biota stations ST1 and I911 was classified in "good chemical status" and further showed no exceedances of the quality standards for river basin specific substances. Horsemussel at stations ST2, ST5 and I914 could not be classified in a " good chemical status" due to the exceedance of the quality standards for cadmium and fluoranthene. The concentrations of the river basin specific substances zinc and copper (copper only at I914) in horsemussels, also exceeded the quality standards and the aim of "good ecological condition" could consequently not be fulfilled.

3.4.1.2 Sediments

For the sediment analysed in 2019 (Brooks et al., 2022a) there were exceedances of limit values for nickel and eight PAH compounds that belong to the priority substance list in the WFD. Therefore, the chemical status of the five sediment stations was classified as “not good” (Øxnevad, 2021).

In 2015, analyses of PAH compounds and metals in samples of sediments were done. “Good chemical condition" could not be reached, due to exceedances of the quality standards for several priority PAH-compounds, including benzo(a)pyrene. Nickel exceeded the quality standard on two stations. Several of the river basin specific PAH compounds also exceeded the quality standards, and the aim of "good ecological status" was not reached.

The main conclusion was that the sediments in the examined area were little influenced by metals, but consistently heavily influenced by PAH-compounds. The concentration of PAHs in sediment was highest at the stations near the discharge and decreased with increasing distance from the discharge point. Over time, however, there has been a positive development in the fjord, regarding PAH-concentrations in sediment.

3.4.2 Høyanger

3.4.2.1 Biota

In Høyangsfjorden, levels of PAHs, fluoride and metals were analyzed in blue mussel from five stations in 2018 (Øxnevad et al. 2019). The quality standard for the priority substance mercury was exceeded at all stations. The chemical status could therefore not be classified as good. There has been a reduction in concentration of mercury in blue mussels from 2015 to 2018, at three of the stations. The reduction was most evident in blue mussels from the innermost station (Sandvika), where the concentration of mercury was reduced by one third. Blue mussels from Sæbøneset had lower concentration of mercury than the other stations in the Høyangsfjord. The concentration of mercury in blue mussels from Sæbøneset was at the same level in 2018 as in 2015. In 2015 the chemical status for blue mussels were classified as “good”, and this was mainly due to the different quality standards. In 2015, a quality standard of 0.5 mg Hg/kg dw was used, but in 2018 the quality standard was 0.02 mg/kg ww.

The concentrations of PAH-compounds were low, but there was a small increase in concentration of PAH_{16} from 2015 to 2018. The mussel concentrations exceeded the quality standard for arsenic, which is a river basin specific substance, thus the environmental objective of a “good ecological status” could not be achieved.

3.4.2.2 Sediments

At the sediment stations there were exceedances of the quality standards for eight of the priority substances; naphthalene, anthracene, fluroanthene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, indeno(1,2,3-cd)pyrene and benzo(g,h,i)perylene. Therefore, “good chemical condition” was not achieved. There were also exceedances of the quality standards for the river basin specific substances; acenaphthene, fluorene, phenanthrene, benzo(a)anthracene, chrysene and dibenzo(a,h)anthracene.

3.4.3 Årdal

3.4.3.1 Biota

Levels of PAHs and metals were analyzed in blue mussel from five stations in 2018. The innermost station is located near the smelter and the outlet of discharges to the fjord. The concentrations of metals and PAH-compounds in blue mussel were generally low, but at the innermost station in the fjord, the quality standards for two priority PAH-compounds were exceeded: benzo(a)pyrene and fluoranthene. The chemical status could therefore not be classified as "good". There were no exceedances of the quality standards at the other mussel stations. There were no exceedances of the quality standards for the river basin specific pollutants.

There has been a considerable reduction in concentration of PAH-compounds in blue mussel from the inner part of the Årdalsfjord. The concentration of PAH_{16} in blue mussel at the innermost station have been reduced by approximately 50 % from 2017 to 2018.

The composition of species and main groups of soft bottom fauna indicate that the situation was not satisfactory in 2011 (Øxnevad et al. 2001). However, compared to investigations in 1998 and 2001 the situation had improved at both stations. There had also been a decrease in concentrations of PAH_{16} and benzo(a)pyrene in horsemussels compared with previous years (2000, 2006, 2007 and 2011).

3.4.3.2 Sediments

Sediment investigations conducted in 2011 showed that the inner Årdalsfjord was heavily affected by PAHs, but sediments from the inner stations were significantly less polluted by PAHs, compared to 2001. The PAH-concentrations in sediment has over time shown a positive development at the two innermost stations AR4 and AR8 from the years 2001 and 2011, to 2015, with reduced concentrations of both benzo(a)pyrene and PAH_{16}. Lower discharges of PAHs might be the reason for this reduction. The sediment showed elevated concentrations of nickel and copper. Nine sediment stations were investigated by the use of sediment profile imaging camera (SPI; Beylich et al. 2019), and were according to benthic habitat quality (BHQ) index found to be in Class 3-4 (moderate to poor conditions).

3.4.4 Karmøy

3.4.4.1 Biota

In August 2020, NIVA implemented an integrated effects assessment at Karmøy under the auspices of the ESPIAL project (Brooks et al, 2022b). For this project mussels were deployed for 8 weeks at various locations around Karmøy. Mussels were deployed at three stations both north and south of the smelter discharge points, a reference site was also included. At each station, mussels were positioned at two depths: 1-2 m from the seafloor and approximately 5 m form the surface. Additionally, sediment samples were taken at 7 sites including a reference site, furthest away from the Smelter. Both mussels and sediment were analysed for a suite of metals and PAHs. Furthermore, a suite of biomarkers were assessed in the mussels at time 0 h and after the 8 weeks deployment.
After 8 weeks deployment, PAH contamination in mussels at the closest stations to the aluminium smelter reflected the PAH profiles of the sediment concentrations at each station (see section 3.4.4.2). This was not true for metals, which were relatively low in all mussels. The PAH concentration in mussels 2 m above the seafloor (bottom), were higher than mussels 5 m from the surface (top/surface) for three out of the four stations. Highest PAH concentrations in mussels were from station 3 bottom (South of the smelter) reflecting the higher PAH sediment concentrations at this station.
Despite the accumulation of PAH in the tissues of the mussels, the five biomarker responses measured were low and overall did not differentiate significantly between the mussel groups. Due to the low response of the biomarkers the IBR/n (Integrative Biological Response index) was equally low and based on the biomarkers measured, there appeared to be minimal impact on the overall health status of the mussels from all stations, including those closest to the aluminium smelter. The Principal Component Analysis (PCA) showed a clear differentiation among transplanted mussels, highlighting mussels from stations 1 top and bottom and stations 2 (Both North of the smelter) and 3 bottom as those with higher chemical body burden. No clear association was found between the impact of PAH's and metals with the biological responses in mussels, even in mussels placed closest to the aluminium shelter.

NIVA conducted operational monitoring in 2019 on behalf of Hydro Aluminium karmøy (Øxnevad et al., 2020). The monitoring programme was prepared in accordance with the water framework directive and approved by the Norwegian Environmental Agency. PAH compounds, arsenic, lead, cadmium, chromium, copper, mercury, nickel and zinc in mussels from three stations were measured. The highest concentration of PAH compounds was measured in mussels from Høgevarde, located in proximity to the northern sedimentation basin at Hydro Karmøy. Mussels from this station exceeded the limit value (EQS) of the priority substance benzo (a) pyrene. Therefore, chemical status of this station is classified as "not good". There were no exceedances of limit values for priority substance in mussels from Helgelandsvika and Bygnesvågen. Therefore, the chemical status of these two stations is "good". There was a lower concentration of PAH compounds in mussels from Høgevarde in 2019 compared to 2017. The concentration of PAH compounds in mussels from this station was approximately halved compared to 2017. There were elevated concentrations of several heavy metals in blue mussels from Høgevarde in 2019.

NIVA also conducted operational monitoring in the Karmsund on behalf of Hydro Aluminium Karmøy in 2017 (Øxnevad. et al 2017). Levels of PAHs, arsenic and lead were analyzed in samples of blue mussel. Blue mussel from station Høgevarde did not obtain "good chemical status" because of concentrations of the PAH-compounds fluoranthene and benzo(a)pyrene that exceeded the EQS for these substances. The two other stations obtained "good chemical status" and showed low concentrations of PAH compounds. Blue mussels from Høgevarde also had elevated concentrations of lead and arsenic. Furthermore, blue mussel from Bygnesvågen showed elevated concentrations of lead.

Results from this and other surveys show that the content of PAH-compounds in mussels has decreased since 2008. The company has significantly reduced their PAH emissions after 2009, when the Søderberg line for production was discontinued. This has given a good effect in the mussels. Biological quality elements were not investigated and the ecological status could therefore not be classified.

3.4.4.2 Sediments

The most recent sediment data from Karmøy was generated in 2020 throughout the ESPIAL project (Brooks et al., 2022b). In this study, measurements of PAHs and metals were taken at 7 stations (including a reference site). The highest PAH and metal contamination of the sediment was observed at station 4, a sheltered lagoon 1-1.5 km south of the smelter (Nordalsvågen), and also elevated at station 3, 500 m south of the smelter. Sediment at station 3 has been previously reported to contain historical PAH and metal concentrations, which had derived from the smelter when contaminant discharges were higher. It was unclear whether the PAH and metal contamination at station 4 originated from historical discharges from the smelter or if other industries were also responsible. Sediment concentrations of several PAHs correlated negatively with distance to the southern outlet of the smelter (i.e., lower concentrations observed further from the outlet). However, the number of stations investigated (n=7) were limited.

Based on the Norwegian classification scheme for the EPA-PAH16 concentrations in sediment, the values for the sum of PAH16 measured, the reference station was background (I) and station 2 (north of smelter) was good (II), whilst station 6 (furthest south of the smelter) was moderate (III). Stations 1 (furthest north), 3 and 5 (South of the smelter) were consider bad (IV), whilst station 4 was three times over the threshold of 20 000 µg/kg and categorised as extreme (V). When looking at the individual PAH, Benzo(g,h,i)perylene concentrations were considered extreme at stations 3 and 4, whilst anthracene, fluoranthene, chrysene, benzo(b,j)fluoranthene, benzo(k)fluoranthene and indeno(1,2,3-cd)pyrene were considered extreme at station 4 only. These PAHs were also classified as bad (IV) in sediment at station 1, 3, 5 and 6. Despite station 2, the closest to the northern discharge pond, having an overall PAH16 classification as good (II), PAH concentrations, indicating bad status (IV), were found for chrysene, indeno(1,2,3-cd)pyrene and benzo(g,h,i)perylene.
Despite some elevated levels of PAHs in sediment, the integrated effects assessment at Karmøy in 2020 showed no clear association between PAHs and metals and biological responses recorded in the mussels, even those located close to the smelter.

The most recent sediment data are from 2015 (Håvardstun 2015). Sediments were sampled from seven stations. The contamination of metals and PAH-compounds were determined. Sediments from the same stations had been included in an earlier sampling program in 2008 (Næs et al. 2009), and the results were compared. Concentrations in sediments from station K5, furthest from the smelter site, did not exceed the quality standards for any river basin specific substance analyzed. High concentrations of PAH_{16} were, however, found at the six other sediment stations. Station K6 additionally exceeded the quality standard for As, and station K10 exceeded the quality standard for Zn. All of the seven sediment stations exceeded the quality standard for one or more of the priority PAH-compounds, thus “good chemical status” was not obtained.

3.4.5 Husnes

3.4.5.1 Biota

Concentrations of PAHs and metals were analyzed in caged blue mussels at five stations in 2018. The quality standards were exceeded for the priority substance mercury at one station, thus "good chemical status" was not obtained. The Hg-concentration, however, was only slightly higher than the quality standard. The station was located in the nearby marina, and the concentration of mercury detected there may have been caused by pollution from the marina. There were no exceedances of the quality standards at any of the other four stations. There were no exceedances of quality standards for any of the PAH-compounds. Only low concentrations of fluoride were detected in the blue mussels.

3.4.5.2 Sediments

Sediments were analyzed in 2015, and exceedances of quality standards were observed only at the station closest to the emission point, for the following river basin specific substances; pyrene and benzo(a)anthracene, as well as for the priority substances anthracene, benzo(b)fluoranthene and indeno(1,2,3-cd)pyrene. The main conclusion of the survey was that elevated concentrations of pollutants were only recorded in the emission's near zone. These pollutants were all PAHs. None of these are longer present in the discharge, and probably leak from an old landfill near the emission point.

3.4.6 Lista

3.4.6.1 Biota

In 2018 NIVA has conducted operational monitoring of Husebybukta on behalf of Alcoa Lista. The programme was designed based on the company's discharges of contaminants to Husebybukta. Concentrations of polycyclic aromatic hydrocarbons (PAH), and metals were analyzed in blue mussels from one station and in common limpets (*Patella vulgata*) from four stations. In addition, monthly water samples were collected in Husebybukta and analyzed for lead and cadmium. Quality standards were exceeded for the priority substances benzo(a)pyrene and fluoranthene in samples of common limpets from Ytre Tjuvholmen and in blue mussels from Haugestranda, thus "good chemical status" was not obtained. There were no exceedances of quality standards for priority substances at the other stations. The quality standard for the river basin specific substance benzo(a)anthracene was exceeded in blue mussels from Haugestranda. Like in 2015, the stations closest to the discharge from Alcoa Lista showed the highest concentrations of priority substances. The concentration of PAH16 was lower in blue mussels from station Haugestranda in 2018 than in 2015.
Concentrations of fluoride appeared slightly elevated at four of the stations.

3.4.6.2 Water

In 2018, samples were collected every month from one station in Husebybukta (H1), placed near the discharge point. The annual average environmental quality standards (AA-EQS) for lead and cadmium in water was exceeded, thus "good chemical condition" was not obtained. The concentration of lead was lower in 2018 than in 2015, while the mean concentration of cadmium was unchanged since 2015.

3.4.7 Mosjøen

3.4.7.1 Biota

The monitoring programme in Vefsnfjorden is designed based on Alcoa Mosjøen's discharges of contaminants to the fjord. Levels of PAHs and metals were analyzed in samples of blue mussel in 2015. Two of the blue mussel stations in the Vefsnfjord (Høyneset and Korsneset) showed concentrations of mercury that exceeded the environmental quality standard for mercury in biota, thus «good chemical status» was not obtained. Blue mussels from station Alterneset, on the other hand, showed concentrations corresponding to «good chemical status». There were low concentrations of PAH-compounds in all blue mussel samples. Compared to 2009, PAH_{16} in horsemussels was significantly reduced.

The bottom fauna in the fjord was moderate to medium species-rich but was characterized by very high individual densities. The composition of species was dominated by some very numerous species that are considered tolerant or opportunistic. The condition of the bottom fauna in Vefsnfjord is unlikely to be due to the company's discharge of suspended substances, as the spill constitutes only 0.5% of the natural supply to the fjord via the Vefsna, Fusta and Drevjo rivers. Similarly, reduced species diversities have been detected in other deep fjords in Northern Norway, and is likely normal.

3.4.7.2 Sediments

Vefsnfjorden has in recent years showed a positive development with regard to concentrations of PAHs in sediments. Data from 2015, compared to 2009, shows that PAH_{16} in sediment is significantly reduced.

3.4.8 KUBAL

3.4.8.1 Sediments/Biota

In 2008, Söderberg furnaces were phased out at Kubal aluminum smelter, and production of aluminium continued with prebaked anodes. Analyses of polyaromatic compounds (PACs) and metals was conducted in 2019 and levels in both sediments (19 stations) and mussels (*Limecola balthica*, 5 stations) were investigated. More than 50 different PACs (including alkylated PACs) and 8 metals were analysed (this was conducted as part of the ESPIAL project (Grung et al., 2020). Many PACs exceeded the EQS near the outlet of Kubal, while the exceedances of metals were less often. The concentrations of most PACs correlated negatively with distance from Kubal, but this pattern was not observed for metals. PACs that did not correlate negatively with distance from Kubal probably had other sources in addition or instead of Kubal. The PAC pattern in sediments correlated well with the pattern observed in mussels, indicating an uptake of PAC from sediment. Concentrations of PACs and metals in sediment compared with historical data from 2002 revealed an overall reduction in concentrations for PACs. A mean of 62% reduction of PAC was observed, and a reduction was observed for 75% of the stations investigated both years (n=12). For metals, increasing concentrations were found for As, Mn and Hg, while Cu, Cr and Vn concentrations were decreased. The stations associated with the greatest increase in concentrations were not close to the discharge points of Kubal.

In previous studies, the PAH-monitoring results show decreasing concentrations with increasing distance from the Kubal-site. This indicates that Kubal contributes to the PAH levels in the bay, but also that this effect is largely limited to the immediate vicinity of the plant and diminishes rapidly at greater distances from the plant. The PAHs in sediment samples from locations close to Kubal generally correponds to Class 5 using the Naturvårdsverket system, indicating contamination, and in the Red Zone using OSPAR's environmental assessment criteria (EAC), indicating potential biological effects. These readings are also higher than the Background Concentrations established by OSPAR for uncontaminated sediments. However, for locations further away from Kubal the concentrations are generally lower than the limit of detection indicating that any impact from the plant is localized. In addition, the PAH concentrations in sediment samples have not increased when compared with the results of a previous survey, conducted in 1988, except at the monitoring point closest to the effluent outfalls. The PAHs in mussel samples are generally in the Green Zone using the EAC values.

3.4.9 ISAL

3.4.9.1 Biota

In 2013, a study was conducted, where PAHs and metals were analyzed in caged blue mussels (Guðjón Atli Auðunsson 2014). A similar proportional distribution of PAHs was found in all mussels sampled from 1997, 2003, 2008 and 2013, suggesting that the sources came from high-temperature treatment of coal, oil or coal salts. Concentrations of PAH_{16}, KPAH, PAH_4 and benzo(a)pyrene increased significantly in blue mussels after 1997. Environmental factors and growth conditions of mussels may affect the concentration of PAHs, i.e. the PAH-content can increase with decreasing growth rates. It was not possible to compare concentrations in molluscs from 2013, with concentrations previous years, due to high limits of detection for benzo(a)pyrene in 2013. The concentrations of PAHs in molluscs correspond to Class II according to Norwegian criteria.

Comprehensive comparisons of data in domestic and foreign databases were made with data from the 1997 and 2003 Straumsvík studies. On the whole, the mussels in Straumsvík in 2013, 2008 and 2003 are lower or similar to those found for human consumption off the coast of Europe and the USA, and lower than wild mussels from the Faroe Islands. As such, the level of pollution can be regarded as low, although the immediate area is obviously under the influence of the factory operations.

3.4.10 Fjardaal

3.4.10.1 Biota

Concentrations of metals (As, Cd, Cr, Cu, Hg, Ni, Pb and Zn) in blue mussels has only been measured twice (2010 and 2015) since the start-up of the smelter. In 2015, the concentrations of metals were lower than, or similar to, those observed in 2010. The maximum concentration of cadmium for consumption in Icelandic regulation is 1.0 mg/kg wet wt, 0.5 mg/kg for mercury and 1.5 mg/kg for lead (regulation 265/2010). No maximum concentration for consumption currently exists for arsenic, chromium, copper, nickel or zinc in the regulation. In all samples of blue mussels collected in 2015, the concentration of mercury, cadmium and lead were below maximum concentration values given according to Icelandic regulations. The concentration of mercury was 0.02–0.10 mg/kg wet wt, for cadmium it was 0.24–0.68 mg/kg wet wt and lead 0.01–0.03 mg/kg wet wt.

According to Icelandic regulation no. 265/2010, the maximum concentration for consumption of benzo(a)pyrene is 10 µg/kg wet wt. Observed concentrations were <0.33 µg/kg for blue mussel and therefore below maximum concentration, according to Icelandic regulations. Of the PAH-compounds analyzed in blue mussels and benthic worms, pyrene was the only one that was much higher in 2015, than in the baseline study (Guðfinsson, 2001), and a study from 2010 (HRV, 2010). However, no specific concentration distribution pattern could be found along the coast of Reyðarfjorður, thus concluding about the source of pollution was difficult.

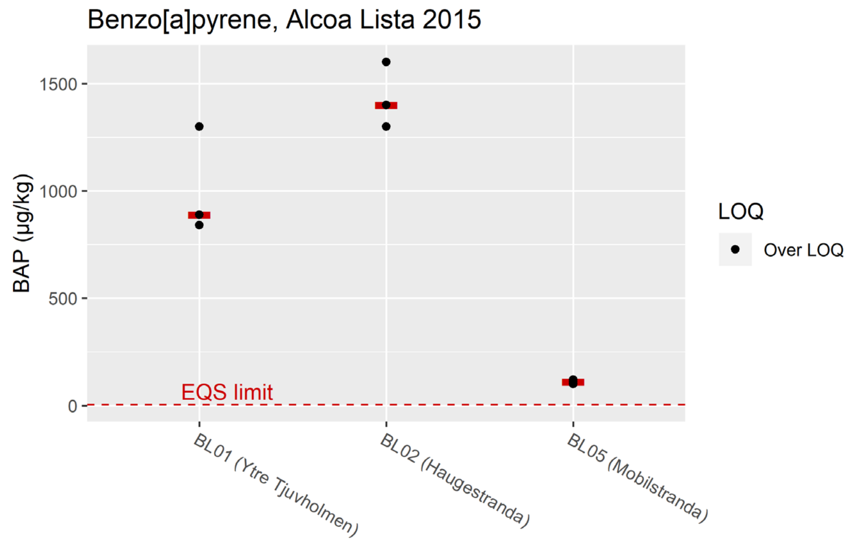
Figure 20 and **Figure 21** show the latest measured concentrations of benzo(a)pyrene and Hg, respectively in sediment and blue mussel at the Norwegian smelter sites. Furthermore, Environmental quality standards are depicted for indication of environmental status.

Figure 20. The latest measured concentrations of benzo(a)pyrene in sediment (left; a, c, e, g, i, k, m) and blue mussel * (right; b, d, f, h, j, l, n) at the Norwegian smelter sites. The year of sampling/analysis is given above each graph. Furthermore, Environmental quality standards are depicted for indication of environmental status. Note: Different scales on axes. Median (-) and replicate values (over/under limit of quantification, LOQ, is indicated) are given.

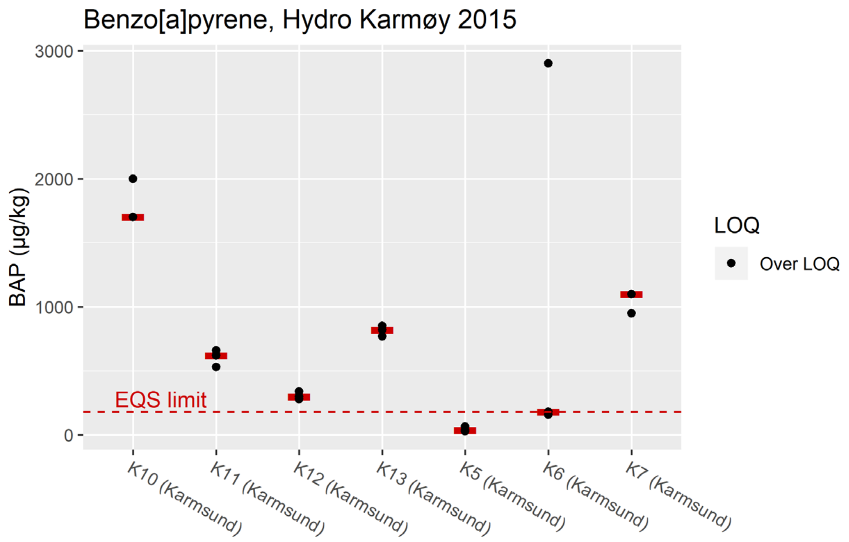
a. Lista (Sediment)

No data

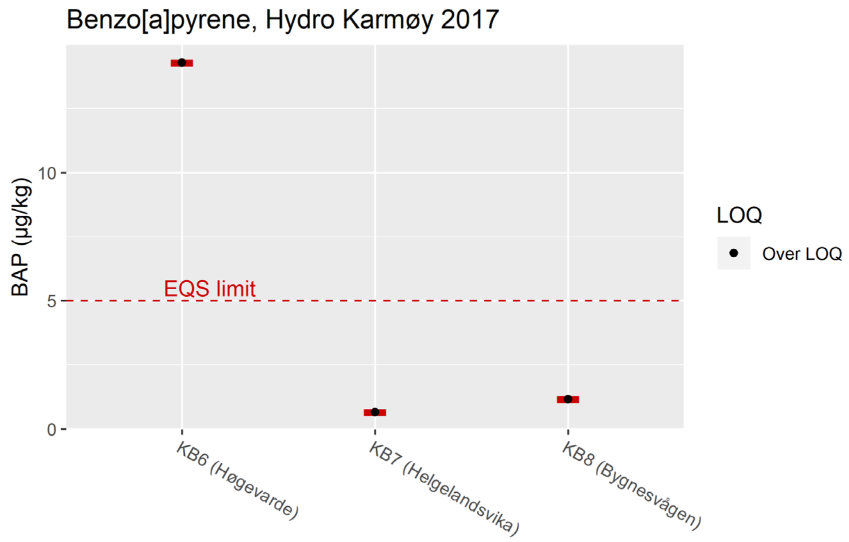
b. Lista (Blue mussel)



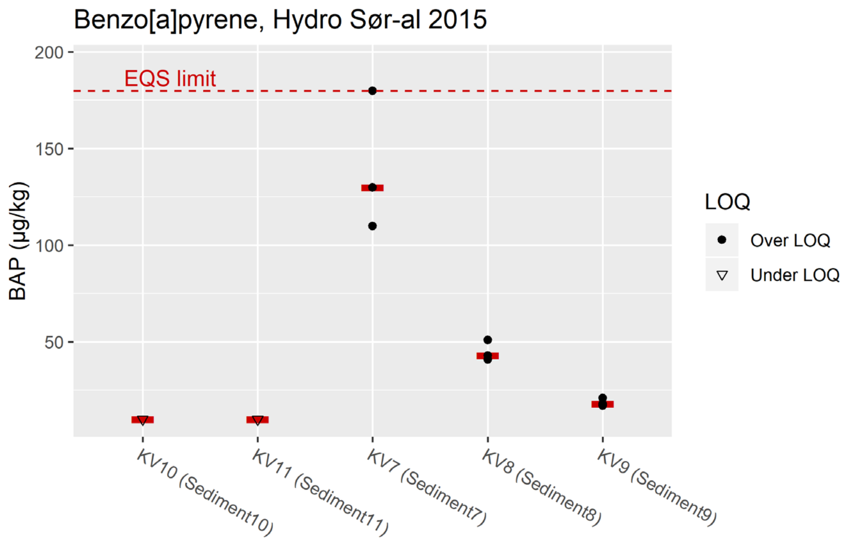
c. Karmøy (Sediment)



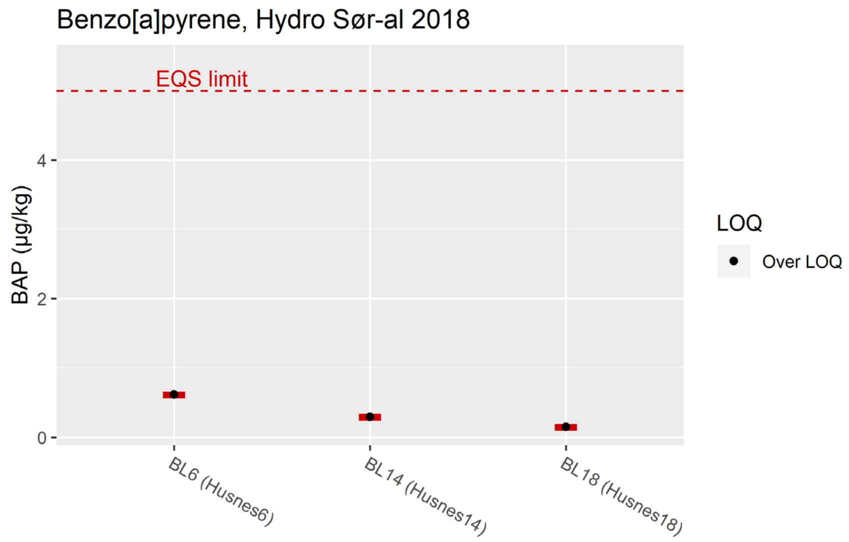
d. Karmøy (Blue mussel)



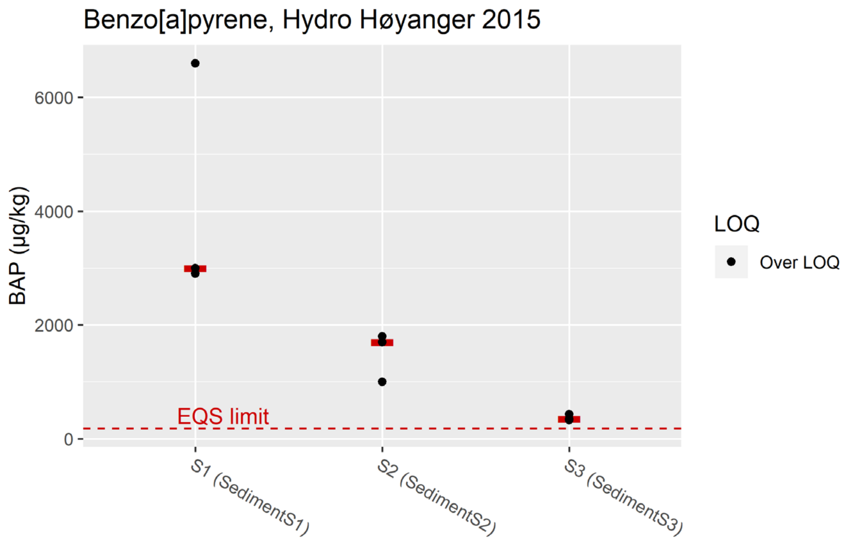
e. Sør-Norge Aluminium, Husnes (Sediment)



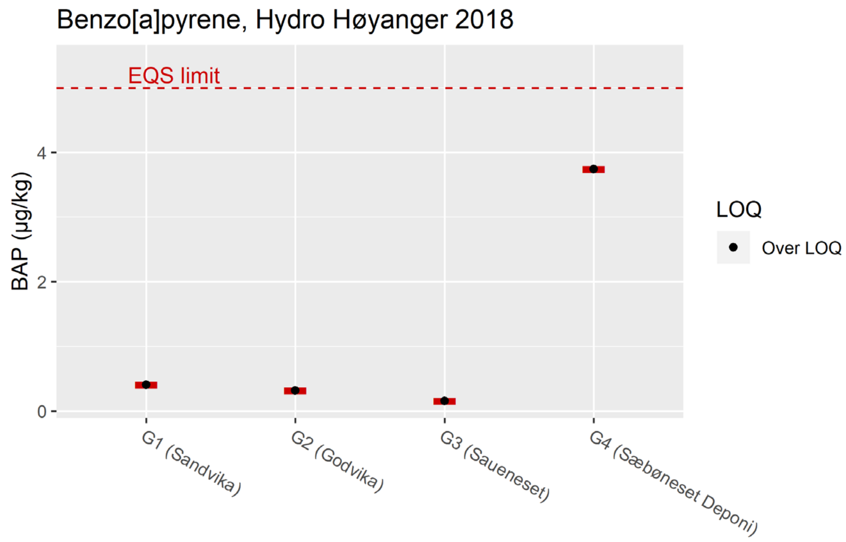
f. Sør-Norge Aluminium, Husnes (Blue mussel)



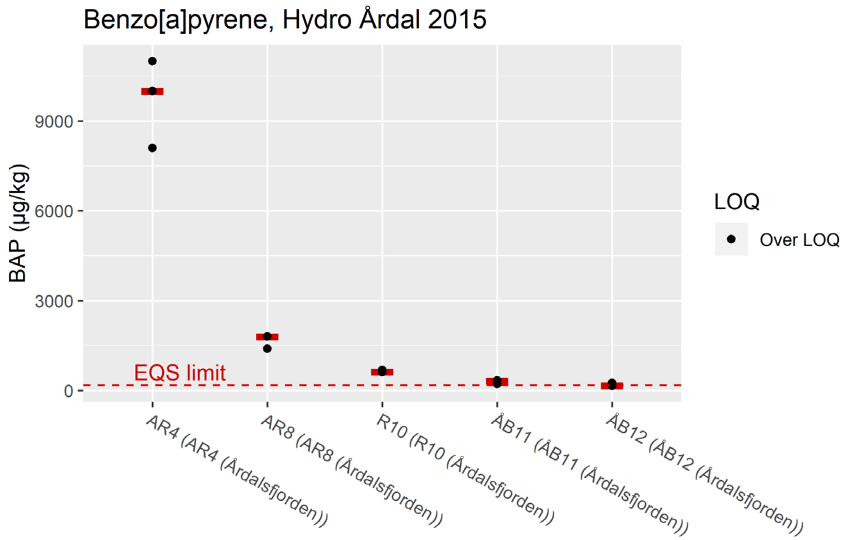
g. Høyanger (Sediment)



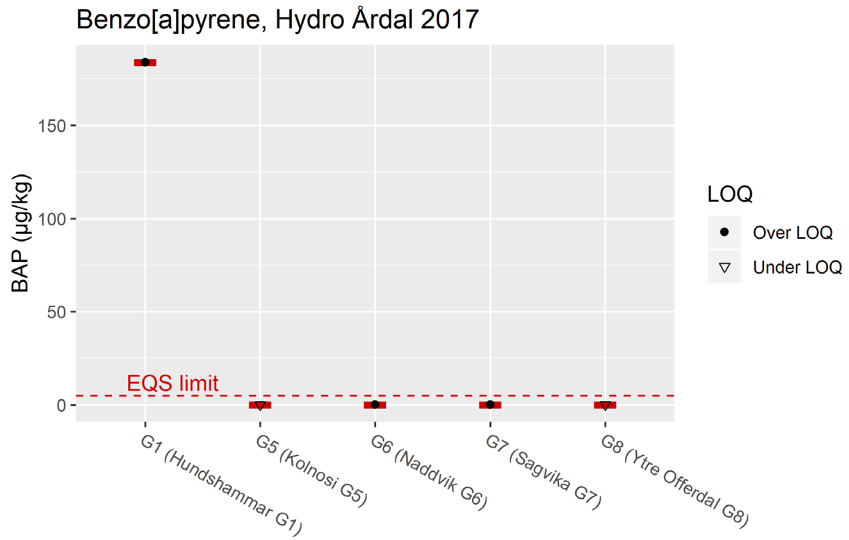
h. Høyanger (Blue mussel)



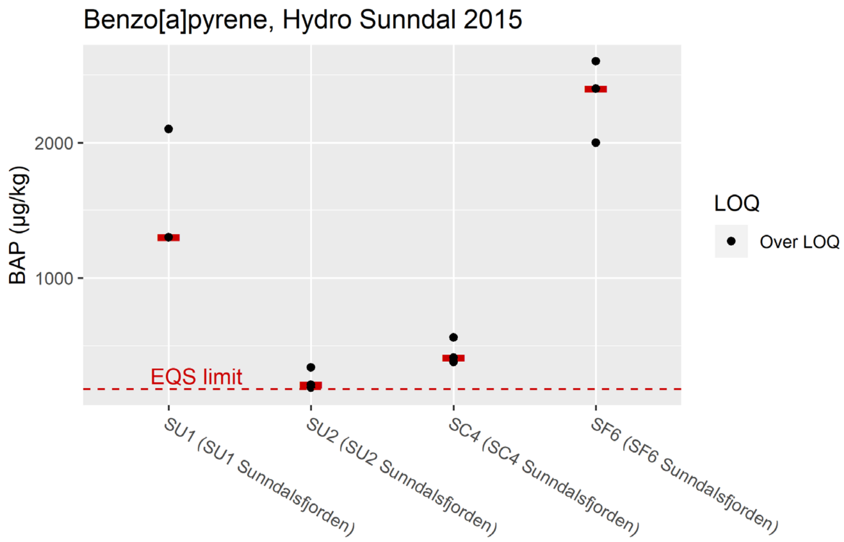
i. Årdal (Sediment)



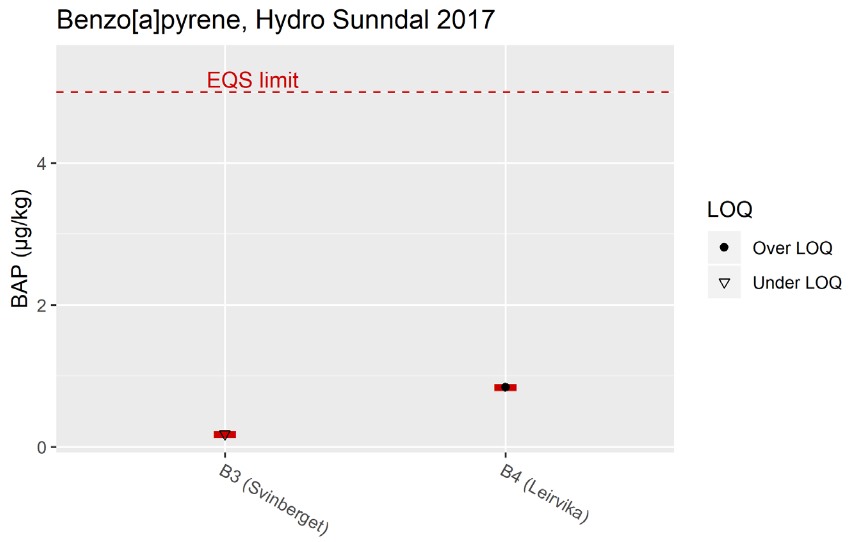
j. Årdal (Blue mussel)



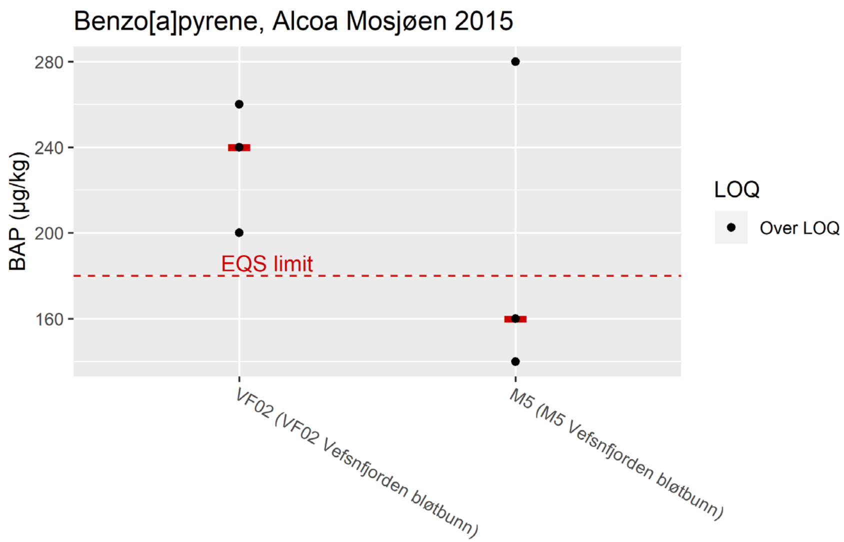
k. Sunndal (Sediment)



l. Sunndal (Blue mussel)



m. Mosjøen (Sediment)



n. Mosjøen (Blue mussel)

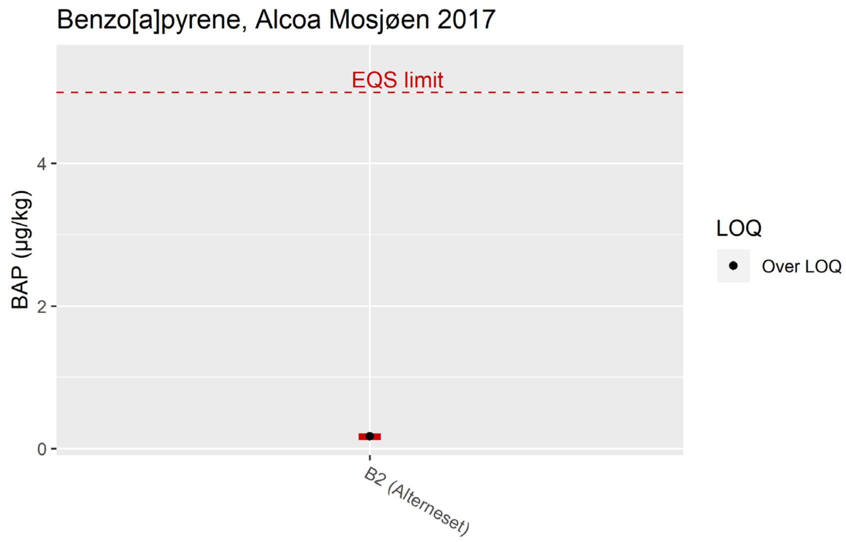
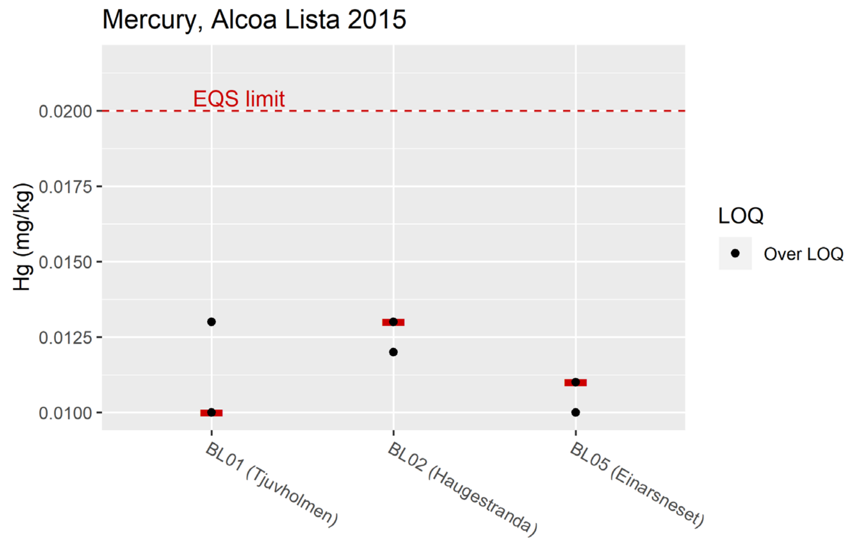


Figure 21. The latest measured concentrations of mercury in sediment (left; a, c, e, g, i, k, m) and blue mussel * (right; b, d, f, h, j, l, n) at the Norwegian smelter sites (Lista, a and b; Karmøy, c and d; Husnes, e and f; Høyanger g and h; Årdal i and j; Sunndal, k and l; Mosjøen m and n). The year of sampling/analysis is given above each graph. Furthermore, Environmental quality standards are depicted for indication of environmental status. Note: Different scales on axes. Median (-) and replicate values (over/under limit of quantification, LOQ, is indicated) are given.
* Horsemussel for mercury at Sunndal.

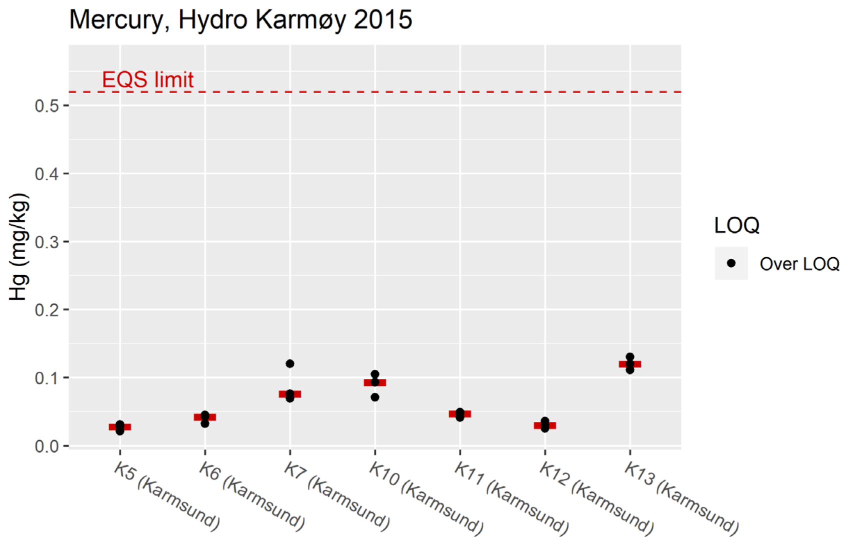
a. Lista (Sediment)

No data

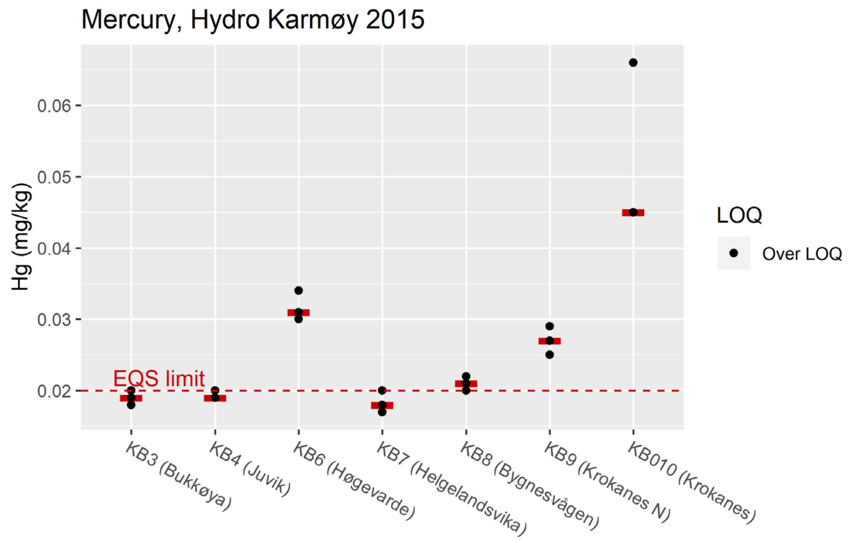
b. Lista (Blue mussel)



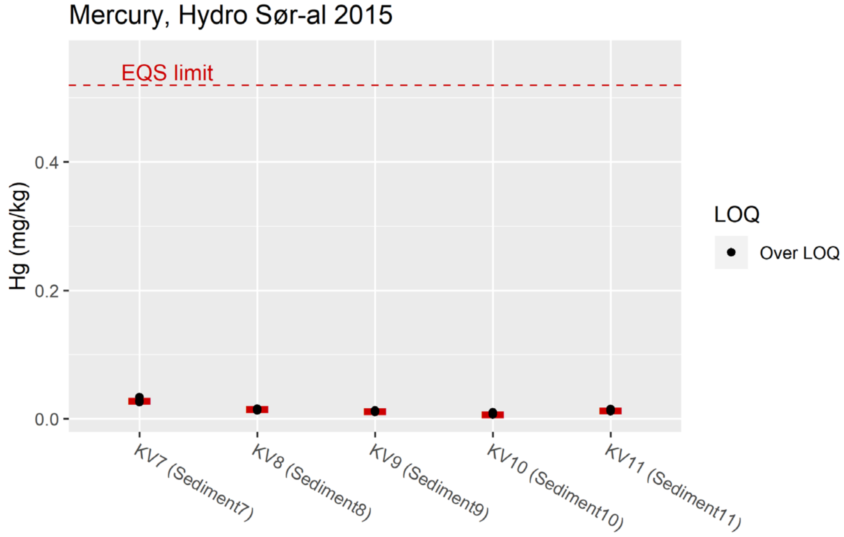
c. Karmøy (Sediment)



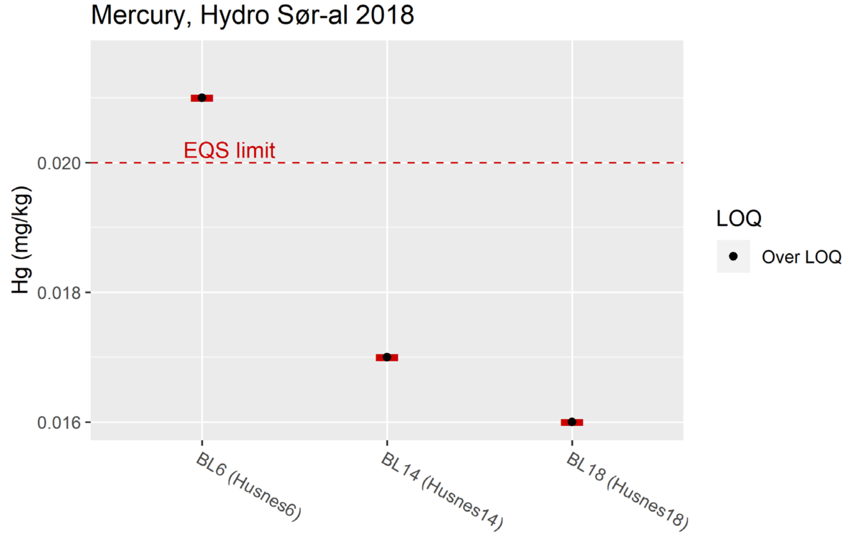
d. Karmøy (Blue mussel)



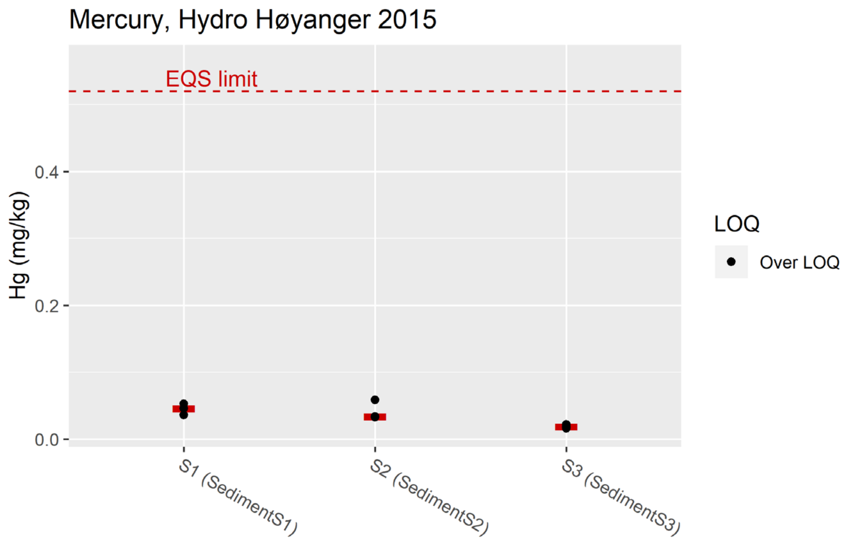
e. Sør-Norge Aluminium, Husnes (Sediment)



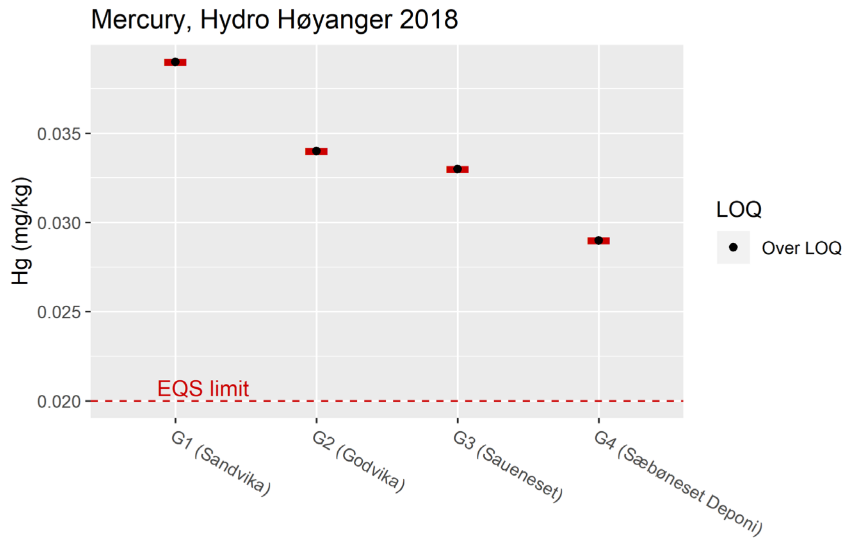
f. Sør-Norge Aluminium, Husnes (Blue mussel)



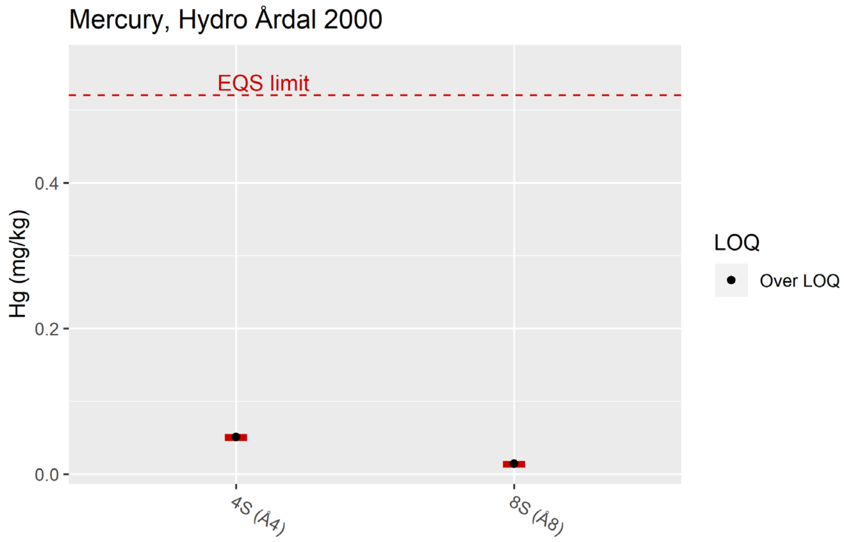
g. Høyanger (Sediment)



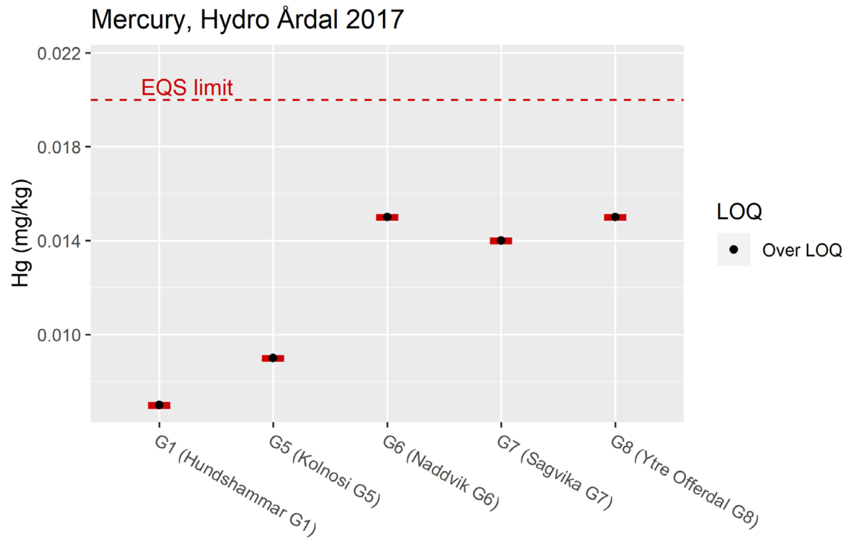
h. Høyanger (Blue mussel)



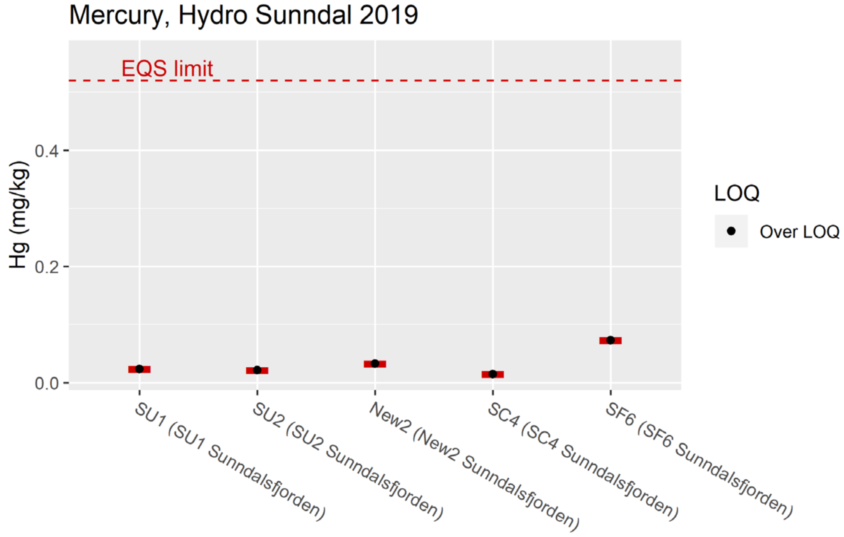
i. Årdal (Sediment)



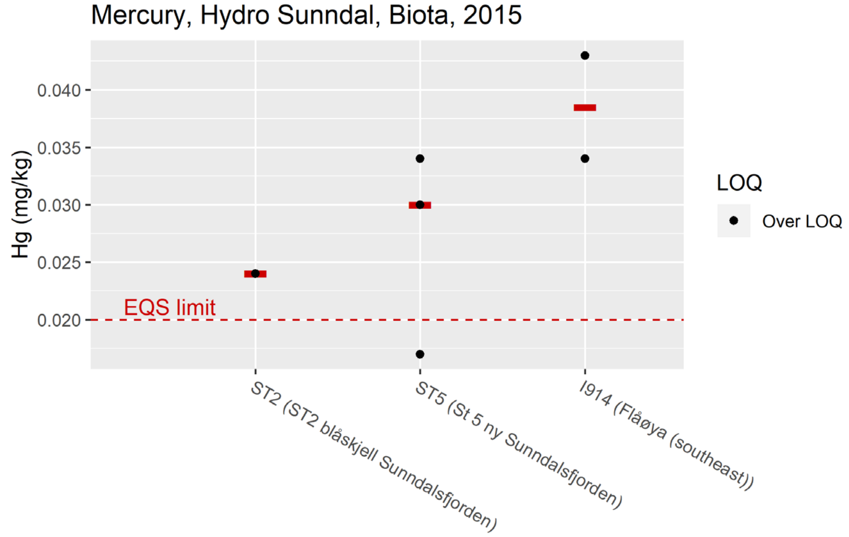
j. Årdal (Blue mussel)



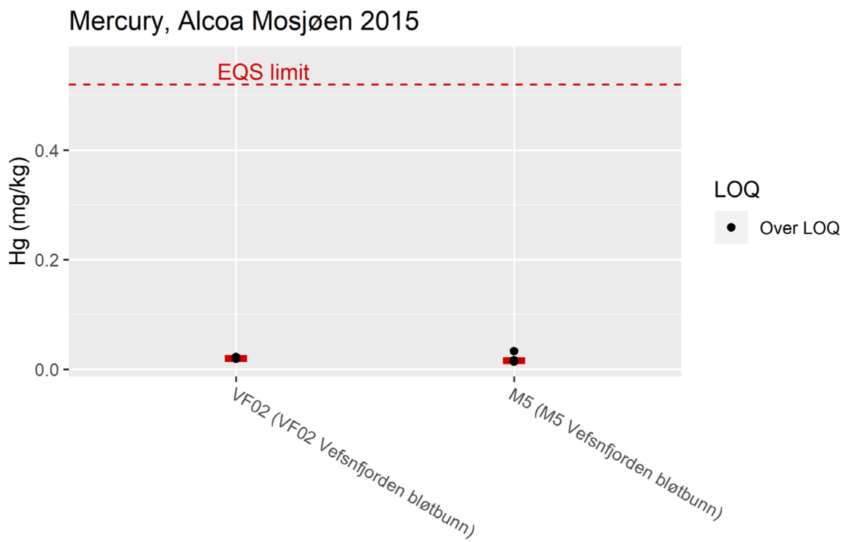
k. Sunndal (Sediment)



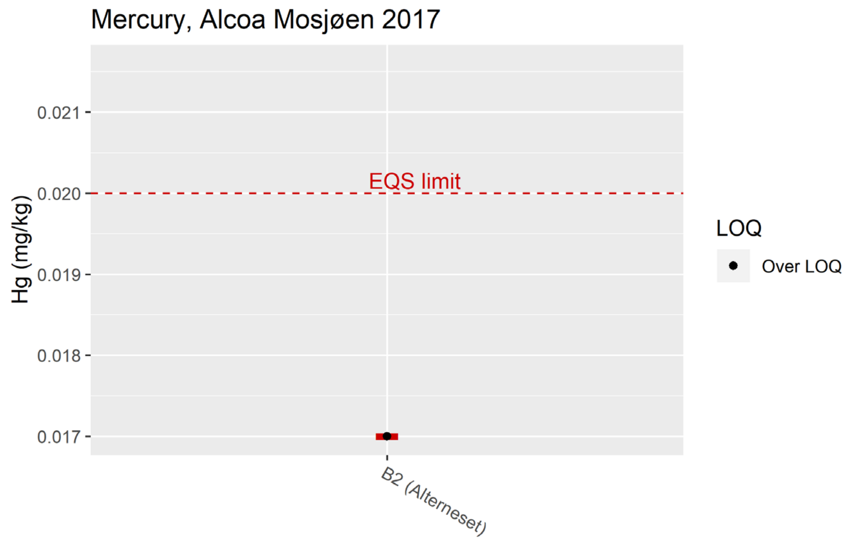
l. Sunndal (Horsemussel)



m. Mosjøen (Sediment)



n. Mosjøen (Blue mussel)



The figures show that there are still excess concentrations of benzo(a)pyrene in sediments at the smelter sites (exception: Husnes), although not necessary reflected in the mussels at many stations. Excess concentrations in mussels are conspicuous at Lista and at Årdal, station G1, the station closest to Hydro aluminium.

The opposite is the trend for mercury: There are no concentrations above EQS in sediments, but excess concentrations in mussels at several stations at Karmøy, Høyanger and Sunndal. For mercury, there are other likely sources than the smelters. In Karmsundet, there are also other industries and urban areas, and at Høyanger there are also other metal processing industries (Nyrstar Høyanger). Furthermore, these excess concentrations of mercury in blue mussel reflect the low EQS for Hg in biota (because of the high oral toxicity of methyl mercury), contributing to exceedances of EQS in mussels at several stations along the coast of Norway (Green et al. 2019).

Table 6 Suggests an improvement/reduction in terms of mercury contamination in biota, as approximately half of the concentrations measured (all data in the database; i.e. all Norwegian sites) in the last decade exceeded the current EQS, while all concentrations previous to this exceeded the EQS. The table also shows that no mercury concentrations in sediment have ever exceeded the EQS. An improvement/reduction in the concentrations of benzo(a)pyrene is also suggested, as the percentage of concentrations in biota exceeding the EQS is reduced from 73% (in 1977-1989) to 16% (in 2010-2019). In sediment, however, 79% (90 of 114) still exceeded the EQS in the last decade (2010-2019).

Table 6. Fraction (and percentage, %) of concentrations in biota and sediment, respectively, in the database exceeding the current Environmental Quality Standards (EQS) in the previous four decades.

Biota	**1977-1989**	**1990-1999**	**2000-2009**	**2010-2019**
PCB7	4/4 (100%)	1/1 (100%)	12/12 (100%)	3/3 (100%)
DDEPP	0/2 (0%)	0/1 (0%)	0/12 (0%)	0/3 (0%)
TPTIN	-	21/21 (100%)	10/10 (100%)	-
TCDD	0/1 (0%)	0/6 (0%)	0/3 (0%)	-
TCDDN	0/1 (0%)	0/6 (0%)	0/3 (0%)	-
HCHG	-	0/1 (0%)	0/12 (0%)	0/3 (0%)
Hg	8/8 (100%)	-	12/12 (100%)	76/140 (54%)
TBT	-	-	2/26 (8%)	0/6 (0%)
HCB	0/4 (0%)	0/1 (0%)	0/12 (0%)	0/3 (0%)
QCB	-	0/1 (0%)	0/7 (0%)	0/3 (0%)
ANT	0/40 (0%)	0/91 (0%)	0/124 (0%)	0/198 (0%)
BAA	26/58 (45%)	0/93 (0%)	2/124 (2%)	9/198 (5%)
BAP	38/52 (73%)	43/97 (44%)	19/124 (15%)	31/198 (16%)
FLU	55/61 (90%)	59/105 (56%)	20/124 (16%)	31/198 (16%)
NAP	0/5 (0%)	0/61 (0%)	0/124 (0%)	0/198 (0%)

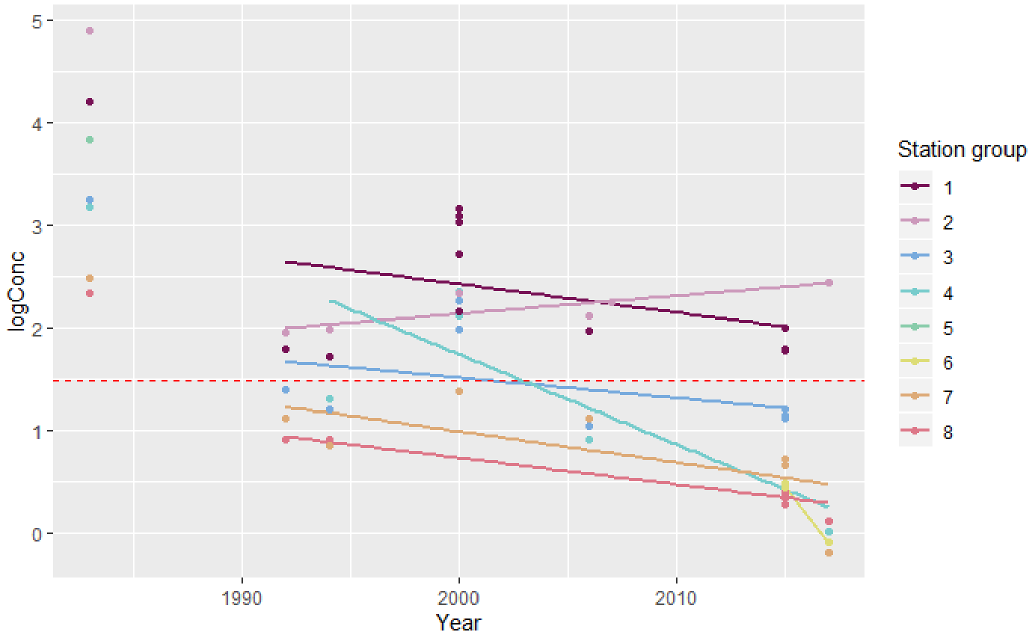
Sediment	**1977-1989**	**1990-1999**	**2000-2009**	**2010-2019**
PCB7	3/5 (60%)	2/4 (50%)	8/14 (57%)	19/30 (63%)
As	-	1/2 (50%)	-	3/119 (3%)
Cr	0/27 (0%)	0/8 (0%)	0/25 (0%)	0/149 (0%)
Cu	23/71 (32%)	1/8 (12%)	23/27 (85%)	32/149 (21%)
Hg	0/71 (0%)	0/14 (0%)	0/4 (0%)	0/121 (0%)
Zn	16/71 (23%)	3/11 (27%)	19/27 (70%)	22/149 (15%)
TBT	-	-	2/2 (100%)	19/19 (100%)
HCB	0/2 (0%)	0/3 (0%)	0/2 (0%)	-
DBAHA	15/22 (68%)	-	-	96/114 (84%)
ACNE	17/51 (33%)	1/4 (25%)	0/18 (0%)	45/114 (39%)
ACNLE	2/2 (100%)	0/3 (0%)	0/18 (0%)	1/114 (1%)
ANT	49/49 (100%)	4/4 (100%)	8/8 (100%)	114/114 (100%)
BAA	69/70 (99%)	3/3 (100%)	7/18 (39%)	99/114 (87%)
BAP	65/71 (92%)	4/4 (100%)	5/18 (28%)	90/114 (79%)
BBF	46/46 (100%)	-	3/12 (25%)	99/114 (87%)
BGHIP	66/69 (96%)	4/4 (100%)	9/18 (50%)	97/114 (85%)
BKF	-	-	6/18 (33%)	85/114 (75%)
CHR	14/18 (78%)	-	5/6 (83%)	-
FLE	32/44 (73%)	0/3 (0%)	0/18 (0%)	24/114 (21%)
FLU	66/71 (93%)	2/3 (67%)	5/18 (28%)	69/114 (61%)
ICDP	65/65 (100%)	3/3 (100%)	11/18 (61%)	98/114 (86%)
NAP	16/22 (73%)	2/3 (67%)	10/18 (56%)	69/114 (61%)
PA	52/71 (73%)	0/3 (0%)	0/18 (0%)	37/114 (32%)
PYR	69/71 (97%)	3/3 (100%)	9/18 (50%)	100/114 (88%)

3.5 Prospects

Where time series exist for concentrations in biota (multiple concentration measurements in time at the same station), where there is a decreasing trend, but where there are stations where the latest concentrations still exceed EQS, one can make crude prospects of the future development based on the slope of linear temporal trend curves, by extrapolation. This was possible for concentrations of fluoranthene in mussels in Årdalsfjorden (**Figure 22**). Here data prior to 1990 were omitted, as they were much higher than those after 1990 and would be too dominating on the trends, and thus obscure the development in the years after 1990. Furthermore, relaxation of the n=5 criterion (see chapter 2.2), was necessary to cover the stations of interest (i.e. extrapolations done on trendlines independently of the above mention statistical trend analysis). The result of this exercise is summarized in **Figure 22 b**: where concentrations below EQS are still not obtained, it is either impossible to make a crude estimate (as there is no downward trend to extrapolate), or concentrations equal to EQS can be expected within the next couple of decades, given a continuation of the concentration reductions observed after 1990. It is important to note the very high uncertainty in these estimates.

A similar exercise can be performed for concentrations of benzo(a)pyrene in sediment in Årdalsfjorden (**Figure 23**). Note that these extrapolations are based on even fewer measurements (2 or 3) from the years 1983, 1989 and 2015, introducing even higher uncertainty. Furthermore, it is important to note that these prospects/predictions do not account for any changes in discharges, or any remedial action. They are merely extrapolations of the trends that culminated in the latest measurements. They do, however, suggest that the time to reach concentrations of benzo(a)pyrene in sediment equal to EQS is substantially longer than for concentrations of fluoranthene in mussels.

a.



b.

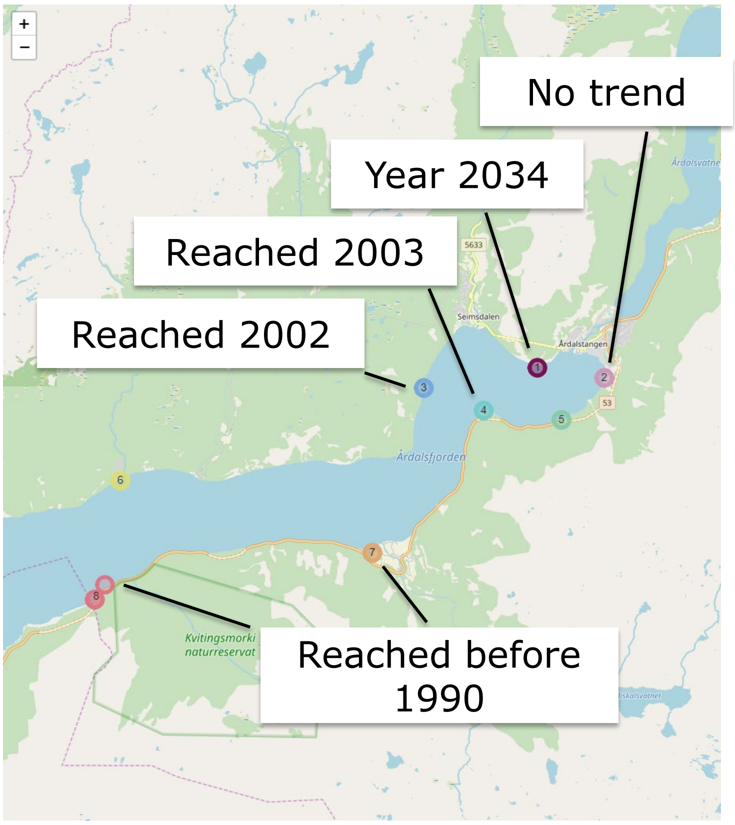


Figure 22. Temporal trends for concentrations of fluoranthene in mussels (horsemussel, except for in 2017, where blue mussel is analysed; also blue mussel at station 2 in 2007) (**a.**), and the year of reaching concentrations equal to the fluoranthene environmental quality standard (EQS), extrapolating these trends, at the stations (Årdalsfjorden) shown in map (**b.**).

Sediment: BaP i Årdalsfjorden

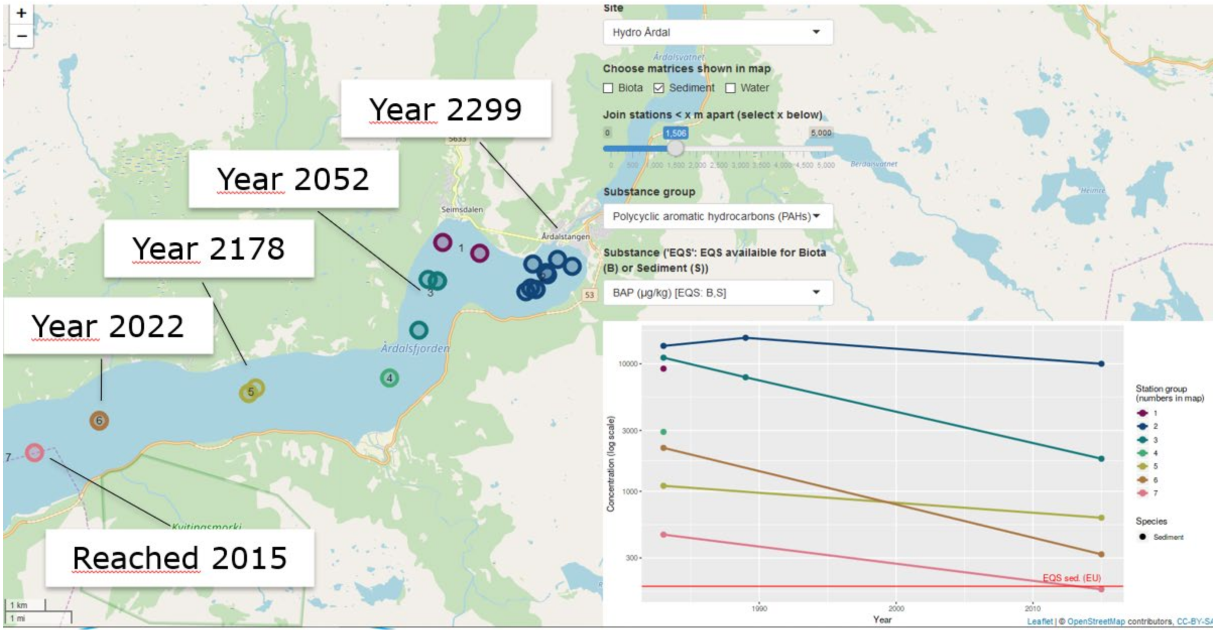


Figure 23. Temporal trends for concentrations of benzo(a)pyrene in sediment, and the year of reaching concentrations equal to the benzo(a)pyrene environmental quality standard (EQS), extrapolating these trends, at the stations (Årdalsfjorden; stations less than 1300 m apart are merged) shown in map. Note the few observations as basis of trends.

4 Concluding remarks

The main outcomes and findings of this ESPIAL work package can be listed as follows:

- Polycyclic aromatic hydrocarbons (PAHs) constitute most data in the database.
- There is a decent amount of data on sediment samples in the data base, however these do not form time series.
- Most time series available in the data base are on blue mussel from Sunndal and northern horsemussel from Årdal.
- Most time series show a declining tendency.
- All statistically significant PAH time trends show declining concentrations.

Other concluding remarks:

- The database is likely not complete, and more data could be available.
- The graphical data displaying tool is a promising aid for instant insight in concentrations and time series of different analytes in environmental samples, historically collected in the vicinity of the smelters.
- The graphical data displaying tool makes it possible to view concentrations and trends in relation to the EQSs, giving insight in chemical status and sites of interest, in terms of possible need of remedial action or natural recovery.
- Extended monitoring continuing established stations and matrices will increase/improve the basis for time trend analysis and thus also predictions of environmental concentrations.

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