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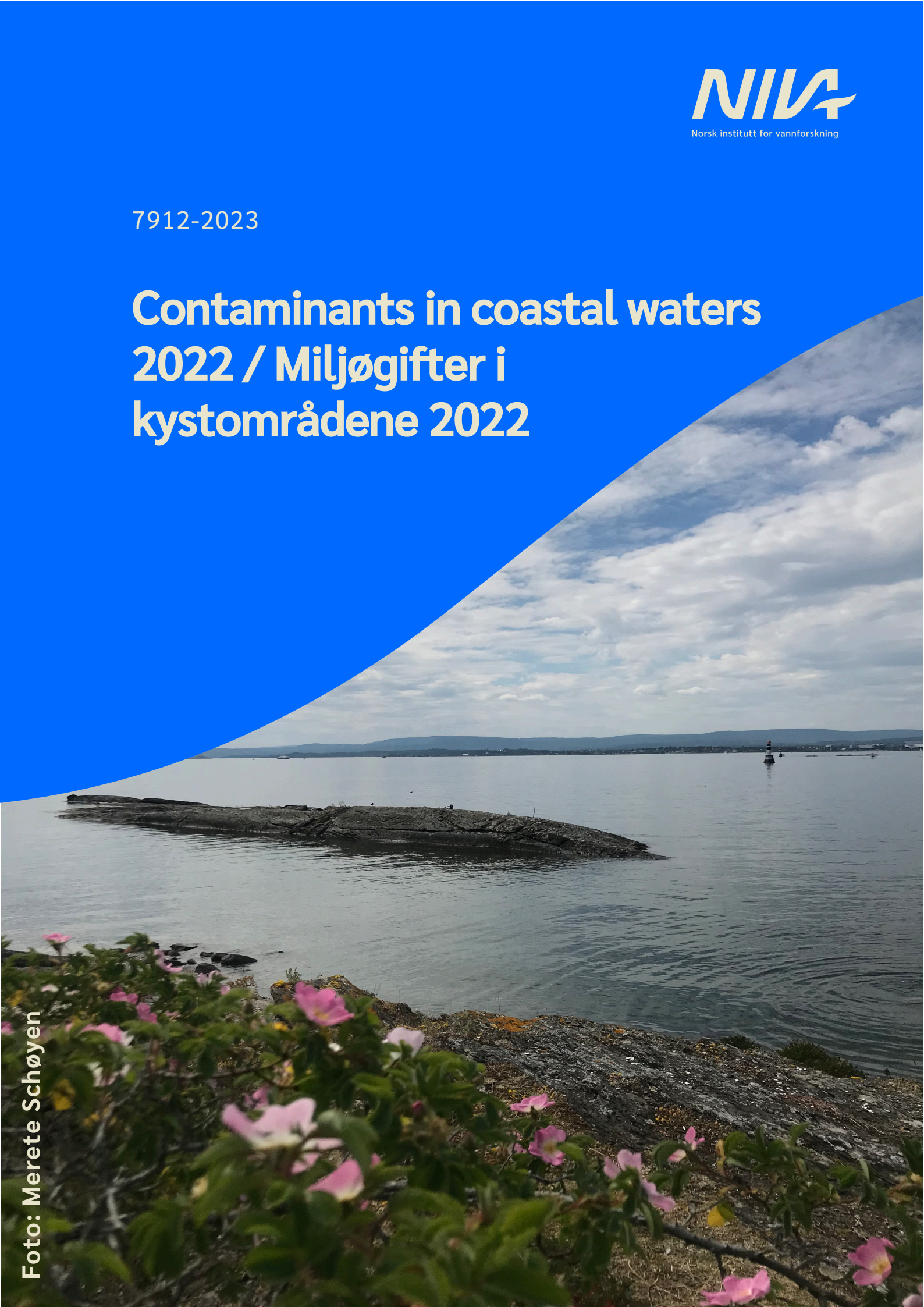
Contaminants in coastal waters 2022 / Miljøgifter i kystområdene 2022



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Norwegian Institute for Water Research

REPORT

Main Office
Økernveien 94
NO-0579 Oslo, Norway
Phone (47) 22 18 51 00

NIVA Region South
Jon Lilletuns vei 3
NO-4879 Grimstad, Norway
Phone (47) 22 18 51 00

NIVA Region East
Sandvikaveien 59
NO-2312 Ottestad, Norway
Phone (47) 22 18 51 00

NIVA Region West
Thormøhlensgate 53 D
NO-5006 Bergen Norway
Phone (47) 22 18 51 00

NIVA Denmark
Njalsgade 76, 4th floor
DK 2300 Copenhagen S, Denmark
Phone (45) 39 17 97 33

Internet: www.niva.no

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Summary

The Norwegian environmental monitoring programme “Contaminants in coastal waters” (Miljøgifter i kystområdene - MILKYS) examines the levels, trends, and effects of contaminants annually in biota along the Norwegian fjords and coastline including Svalbard. The 2022 investigation included analyses of more than 180 different contaminants or biological effect parameters in five species (blue mussel, cod, dogwhelk, common periwinkle, and common eider). The contaminants measured include metals, TBT, PCBs, PAHs, PBDEs, PFAS, HBCDs, chlorinated paraffins, siloxanes, and pesticides. Biological effect parameters investigated include imposex (VDSI) and intersex (ISI), PAH-metabolites, ALA-D, and EROD.

In this report, 37 contaminants and in addition, biological effect parameters were chosen for in-depth presentation. EQSs (Environmental Quality Standards) were exceeded in blue mussel (15%) and cod (36%) expressed as datapoints (contaminants x stations). Contaminants above EQSs were mercury (Hg), sumPCB7, sumPBDE6, and MCCP. The sum of exceedances (sum of risk quotients) was highest in cod from the Inner Oslofjord. PROREFs (Norwegian provisional high reference contaminant concentrations) were exceeded in blue mussel (38%) and cod (9%) expressed as datapoints (contaminants x stations), and exceedances were higher in mussel (up to 10-20x PROREF) than cod (5-10x PROREF). The sum of PROREFratio above background levels were highest in blue mussel from Akershuskaia in the harbour of the Inner Oslofjord. Significant decreasing time trends for specific contaminants/effects dominated both long-term (> 10 years) and short-term (≤ 10 years) where trends could be detected, still some significant increasing trends were observed for contaminants exceeding EQS and/or PROREF. Notably significant increasing short-term trends were found for lead (Pb), chromium (Cr), and some PCBs in blue mussel, and for mercury, and silver (Ag) in cod.

Four keywords	Fire emneord
1. Environmental contaminants in biota	1. Miljøgifter i biota
2. Levels and trends	2. Nivåer og trender
3. Biological effects	3. Biologiske effekter
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Merete Schøyen	*Sissel Ranneklev*	*Morten Jartun*
Project Manager/Main Author	Quality Assurance	Research Manager

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Contaminants in coastal waters 2022 / Miljøgifter i kystområdene 2022

Preface

The Norwegian environmental monitoring programme “Contaminants in coastal waters” (Miljøgifter i kystområdene - MILKYS) investigates contaminants in samples of blue mussel, cod, dogwhelk, common periwinkle, and common eider on a yearly basis. This report presents the findings from monitoring performed in 2022, the second year of a new five-year period (2021-2025). The coastal monitoring program for contaminants has been performed since 1981. The 2022 campaign was carried out by the Norwegian Institute for Water Research (NIVA) contracted by the Norwegian Environment Agency (NEA, Miljødirektoratet). Coordinator at NEA is Gunn Lise Haugestøl (deputy coordinator Bård Nordbø) and the project manager at NIVA is Merete Schøyen (deputy project manager Merete Grung).

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Thanks also go to the numerous fishermen and their boat crews for which we have had the pleasure of working with.

Oslo, 30 November 2023.
Merete Schøyen
Project Manager,
NIVA

Summary

The monitoring programme “Contaminants in coastal waters” (Miljøgifter i kystområdene - MILKYS) examines the levels, trends, and effects of contaminants along the Norwegian coast, fjords and Svalbard. The programme provides a basis for assessing the state of the environment in Norwegian coastal waters. The monitoring makes an important contribution to national administration and to the international organizations such as the Oslo-Paris Convention’s (Convention for the Protection of the Marine Environment of the North-East Atlantic, OSPAR), Coordinated Environmental Monitoring Programme (CEMP), the international Council for Marine Research (International Council for the Exploration of the Sea, ICES), and the European Environment Agency (EEA).

The 2022 investigation monitored the concentration of contaminants in blue mussel (*Mytilus edulis*) at 24 stations, Atlantic cod (*Gadus morhua*) at 18 stations, dogwhelk (*Nucella lapillus*) at eight stations, common periwinkle (*Littorina littorea*) at one station, and common eider (*Somateria mollissima*) at one station. The stations are located both in areas with known or presumed point sources of contaminants, in areas of diffuse loads of contaminants such as city harbour areas, and in more remote regions with presumed low exposure to pollution. In 2022 the following contaminants were monitored: metals (mercury (Hg), cadmium (Cd), lead (Pb), copper (Cu), zinc (Zn), silver (Ag), arsenic (As), nickel (Ni), chromium (Cr), and cobalt (Co)), tributyltin (TBT), polychlorinated biphenyls (PCBs), dichlorodiphenyltrichloroethane (DDT, using dichlorodiphenyldichloroethylene (p,p'-DDE) - principle metabolite of DDT as an indicator), hexachlorobenzene (HCB), pentachlorobenzene (QCB), octachlorostyrene (OCS), polycyclic aromatic hydrocarbons (PAHs), polybrominated diphenyl ethers (PBDEs), per- and polyfluoroalkyl substances (PFAS), hexabromocyclododecanes (HBCD), short and medium chained chlorinated paraffins (SCCP and MCCP), and siloxanes (the cyclic volatile methyl siloxanes, cVMS: D4, D5, and D6). Biological effect parameters were also monitored. These were imposex and intersex parameters in marine snails as biomarkers of TBT-exposure, OH-pyrene in cod bile as a marker of PAH-exposure, δ-aminolevulinic acid dehydrase inhibition (ALA-D) in red blood cells from cod as a marker of exposure to lead, and cytochrome P450 1A-activity (ethoxyresorufin-O-deethylase, EROD) in cod liver as a marker of exposure to planar PCBs, PAHs, and dioxins.

Significant decreasing time trends for contaminants dominated where trends could be detected, still some significant increasing trends were observed for contaminants exceeding EQS (Environmental Quality Standards) and/or PROREF (Norwegian provisional high reference contaminant concentrations). The main findings in 2022 can be summarized as follows:

Levels

- The EQS was exceeded in blue mussel (15%) and cod (36%) expressed as datapoints (contaminants x stations), and the contaminants above these limits were mercury (Hg), sumPCB7, sumPBDE6, and MCCP. The sum of exceedances (sum of risk quotients) was highest in cod from the Inner Oslofjord followed by urban areas (harbours).
- The PROREF was exceeded in blue mussel (38%) and cod (9%) expressed as datapoints (contaminants x stations), and exceedances were higher in blue mussel (up to 10-20x PROREF) than cod (5-10x PROREF). The sum of PROREFratio above background levels were highest in blue mussel from Akershuskaia in the Inner Oslofjord.

Time trends

- Decreasing time trends dominated both long-term (> 10 years) and short-term (≤ 10 years) where trends could be detected, and notably significant increasing short-term trends were found for lead, chromium, and some PCBs in blue mussel, and for mercury and silver in cod.

- Increasing long-term trend for mercury was found in cod fillet from the Inner Oslofjord where several contaminants occur at higher concentrations than other areas along the coast.

Effects

- Biological effect parameters (biomarker analysis) showed no effects of TBT in snails, but confirm exposure of PAH, lead, and planar organic compounds in cod.

EQS

A total of 293 assessments of EQSs for 20 contaminants have been evaluated. EQSs were exceeded in blue mussel (15%) and cod (36%) expressed as datapoints (contaminants x stations). No exceedances of EQS were observed in snails.

Contaminants often exceeding EQSs were mercury (there are no EQS value for other metals in biota), sumPBDE6 (sum of the following congeners: 28, 47, 99, 100, 153, and 154), and sumPCB7 (sum of the following congeners: 28, 52, 101, 118, 138, 153, and 180) for blue mussel and cod. Concentration of MCCP above EQS was observed at one station in cod.

Blue mussel in the Inner and Outer Oslofjord, in harbour areas and in areas like the Sørfjord had highest number of exceedances of EQS. For cod, EQSs were exceeded at all stations for sumPCB7 and sumPBDE6, and for almost all stations for mercury except on Svalbard.

PROREF

A total of 701 assessments for PROREFs have been made for the 25 contaminants presented in the extended summary. Blue mussel exceeded PROREF for 38% of the datapoints (contaminants x stations), and 10% of the datapoints (40 datapoints) could not be classified vs. PROREF since the limit of quantification (LOQ) was higher than PROREF. In cod, 91% of the samples were below PROREF, and the highest exceedances were lower for cod (2-5x PROREF) than for mussel (up to 10-20x PROREF).

The PROREFs were at higher concentrations in cod than in mussels (except for the three metals; cobalt, cadmium, and lead). For blue mussel, there were most exceedances of the PROREF for copper, mercury, lead, zinc, PCBs, and a PAH (pyrene, PYR). For cod, there were most exceedances for mercury, followed by silver, and PCBs.

For metals in blue mussel, the highest exceedances of PROREF were for lead at Kvalnes in the Sørfjord and for PCB118 at Akershuskaia in the Inner Oslofjord. Blue mussel stations in the Inner Oslofjord had many exceedances of PROREFs, and among these, Akershuskaia had the most and highest exceedances. PCBs had the highest exceedances, which were observed in several urban stations (harbours of Bodø, Ålesund, and Bergen). For cod, there were most exceedances of PROREF in the Inner Oslofjord and in the harbours of Bergen and Ålesund.

Time trends

A total of 782 time trends (long-term and short-term) were estimated for the contaminants presented in the extended summary. In general, there were fewer long-term trends (due to insufficient data), while no trends and decreasing trends dominated.

Long-term time trends

Long-term time trends (> 10 years) in blue mussel were dominated by no trends (37%) and decreasing trends (28%). Increasing trends were observed for 8% of data. A small number of data had insufficient count or data above LOQ for trends to be determined. The picture was similar for long-

term trends in cod compared to blue mussel, but the percentage of decreasing trends was somewhat higher (39%). No trends were observed for 20% of data for cod and increasing trends for 6%.

Short-term time trends
There were more datapoints (contaminants x stations) that could be determined for short-term time trends (≤ 10 years) than long-term trends for both blue mussel and cod. "No trend" dominated for blue mussel (44%), while decreasing short-term trends dominated for cod (45%). Increasing short-term trends were found both in mussels and cod (13% and 8%, respectively).

In blue mussel, there were instances of increasing short-term trends for most metals, but the increasing trends were dominated by PCB. However, some of these PCB trends are uncertain due to few data above the LOQ. For cod, there were increasing short-term trends for silver at seven stations and at four stations for mercury. Except for nickel, lead and chromium, there were instances of increasing short-term trends for all metals in cod.

The highest occurrence of increasing time trends were found at blue mussel stations in the Inner Oslofjord, and at cod stations at Lista and Lofoten.

Increasing time trends with exceedances of EQS and/or PROREF
Special attention needs to be paid on the increasing time trends for contaminants which at the same time exceed EQS and/or PROREF (>5 for mussel and >2 for cod). This was the case for 10 combinations of contaminants, stations, and species. In cod fillet from the Inner Oslofjord, Lista, and Bømlo, there were increasing time trends in addition to exceedance of EQS (RQ) and PROREF for mercury.

Biological effects
The 2022 data confirmed the annual results dating back to 2017 indicating no effects of TBT on dogwhelk (imposex parameter Vas Deferens Sequence Index, VDSI=0).

Median (non-normalized) OH-pyrene in cod bile concentrations was above the ICES/OSPAR assessment criterion (background assessment criteria, BAC) at all stations (Oslofjord, Sørfjord and Lista), except at Bømlo, the reference station, indicating exposure to PAH-compounds.

ALA-D activity in cod blood in the Inner Oslofjord appeared slightly lower than at the Bømlo reference station, however, this was not statistically significant. In the Inner Sørfjord, the median ALA-D activity was significantly lower than at the reference station. Reduced activities of ALA-D reflect higher exposure to lead. Higher concentrations of lead in cod liver have generally been observed in the Inner Oslofjord, as well as the Inner Sørfjord compared to Bømlo.

The median EROD activity appeared lower in cod liver at the Bømlo reference station, than in the Inner Oslofjord and Inner Sørfjord, suggesting exposure to planar PCBs, PAHs, and dioxins, however, this was not statistically significant. Median EROD activities were below the ICES/OSPAR assessment criterion (BAC) at all stations.

Sammendrag

Tittel: Miljøgifter i kystområdene 2022.
År: 2023.
Forfatter(e): Merete Schøyen, Merete Grung, Espen Lund, Dag Ø. Hjermann, Anders Ruus, Sigurd Øxnevad, Guttorm Christensen (Akvaplan-niva), Bjørnar Beylich, Marthe T. S. Jenssen, Lise Tveiten, Jarle Håvardstun, Veronica Eftevåg, og Kine Bæk.
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Overvåkingsprogrammet «Miljøgifter i kystområdene - MILKYS» (Contaminants in coastal waters) undersøker nivåer, trender og effekter av miljøgifter langs norskekysten, fjorder og på Svalbard. Programmet gir grunnlag for å vurdere miljøtilstanden i norske kystfarvann. Overvåkingen gir viktig bidrag til nasjonal forvaltning og til internasjonale organisasjoner som Oslo-Paris konvensjonen (Convention for the Protection of the Marine Environment of the North-East Atlantic, OSPAR) sitt koordinerte miljøovervåkingsprogram (Coordinated Environmental Monitoring Programme, CEMP), Det internasjonale havforskningsrådet (International Council for the Exploration of the Sea, ICES) og Det europeiske miljøbyrået (European Environment Agency, EEA).

I 2022 omfattet overvåkingen miljøgifter i blåskjell (*Mytilus edulis*) fra 24 stasjoner, torsk (*Gadus morhua*) fra 18 stasjoner, purpursnegl (*Nucella lapillus*) fra åtte stasjoner, strandsnegl (*Littorina littorea*) fra én stasjon og ærfugl (*Somateria mollissima*) fra én stasjon. Stasjonene er plassert i områder med kjente eller antatt kjente punktkilder for tilførsler av miljøgifter, i områder med diffus tilførsel av miljøgifter slik som byens havneområder, og i fjerntliggende områder med antatt lav eksponering for miljøgifter. Overvåkingen i 2022 omfattet analyser av bl.a. metaller (kvikksølv (Hg), kadmium (Cd), bly (Pb), kobber (Cu), sink (Zn), sølv (Ag), arsen (As), nikkel (Ni), krom (Cr) og kobolt (Co)), tributyltinn (TBT), polyklorerte bifenyler (PCBer), diklordifenyltrikloretan (DDT, bruker diklordifenyldikloretylen (DDE) metabolitt av DDT som indikator), heksaklorbenzen (HCB), pentaklorbenzen (QCB), oktaklorbenzen (OCB), polysykliske aromatiske hydrokarboner (PAHer), polybromerte difenyletere (PBDEer), perfluorerte alkylforbindelser (PFAS), heksabromsyklododekan (HBCD), korte- og mellomkjedete klorparafiner (SCCP og MCCP) og siloksaner (sykliske flyktige metylsiloksaner, cVMS: D4, D5 og D6). Det ble også gjort overvåking av biologiske effekt-parametere. Dette var imposex og intersex i marine snegler som biomarkører for TBT-eksponering, OH-pyren i torskegalle som markør for PAH-eksponering, d-aminolevulinsyre dehydrase (ALA-D) i røde blodceller fra torsk som markør for eksponering for bly og cytokrom P450 1A-aktivitet (ethoxyresorufin-O-deethylase, EROD) i torskelever som markør for eksponering for plane PCBer, PAHer og dioksiner.

Signifikante nedadgående tidstrender for miljøgifter dominerte der hvor trender kan påvises, likevel ble det observert signifikante økende trender for miljøgifter som overskred EQS (Environmental Quality Standards) og/eller PROREF (norsk provisorisk høy referansekonsentrasjon for miljøgifter). Hovedfunnene i 2022 kan oppsummeres som følger:

Nivåer

- Det var overskridelser av EQS i blåskjell (15%) og torsk (36%) målt ved datapunkter (miljøgifter x stasjoner), og miljøgiftene som overskred disse grensene var kvikksølv (Hg), sumPCB7, sumPBDE6, og MCCP. Summen av overskridelser (sum risikokvotienter) var høyest i torsk fra indre Oslofjord etterfulgt av urbane områder (havner).
- Det var overskridelser av PROREF i blåskjell (38%) og torsk (9%) målt ved datapunkter (miljøgifter x stasjoner), og overskridelsene var høyere i blåskjell (opptil 10-20x PROREF) enn torsk (5-10x PROREF). Summen av PROREFratio over bakgrunnsnivåer var høyest i blåskjell fra Akershuskaia i indre Oslofjord.

Tidstrender

- Nedadgående tidstrender dominerte både på lang sikt (> 10 år) og kort sikt (≤ 10 år), der hvor tidstrender kunne påvises, og i blåskjell ble signifikante oppadgående korttidstrender særlig påvist for bly, krom, og noen PCBer, og for kvikksølv og sølv i torsk.
- Oppadgående langtidstrend for kvikksølv ble funnet i torskefilét fra indre Oslofjord, hvor flere miljøgifter forekommer i høyere konsentrasjoner enn andre områder langs kysten.

Effekter

- For biologiske effektparametere (biomarkøranalyser) var det ingen effekter av TBT i snegler, men undersøkelsene bekrefter eksponering av PAH, bly og plane organiske forbindelser i torsk.

EQS

Det er gjort totalt 293 vurderinger av EQS for 20 miljøgifter. EQS ble overskredet i blåskjell (15%) og torsk (36%) målt ved datapunkter (miljøgifter x stasjoner). Det var ingen overskridelser av EQS i snegl.

Miljøgifter som ofte overskrider EQS var kvikksølv (det fines ikke EQS-verdier for andre metaller i biota), sumPBDE6 (sum av de følgende kongenere: 28, 47, 99, 100, 153, og 154) og sumPCB7 (sum av følgende kongenere: 28, 52, 101, 118, 138, 153, og 180) for blåskjell og torsk. Ett tilfelle av overskridelse av MCCP i blåskjell ble observert.

Blåskjell fra indre og ytre Oslofjord, i havneområder og i områder som Sørfjorden hadde flest overskridelser av EQS. For torsk var det overskridelser av EQS på alle stasjoner for sumPCB7 og sumPBDE6, og for de fleste stasjoner for kvikksølv unntatt på Svalbard.

PROREF

Det er gjort totalt 701 vurderinger for PROREF for de 25 utvalgte miljøgiftene som er utvalgt for presentasjon i det utvidede sammendraget. I blåskjell ble PROREF overskredet i 38% av datapunktene (miljøgifter x stasjoner), og 10% av datapunktene (40 datapunkter) kunne ikke klassifiseres vs. PROREF, fordi kvantifiseringsgrensen (limit of quantification, LOQ) var høyere enn PROREF. For torsk var 91% av prøvene under PROREF, og de høyeste overskridelsene var lavere for torsk (2-5x PROREF) enn for blåskjell (opptil 10-20x PROREF).

Konsentrasjoner for PROREF var høyere i torsk enn i blåskjell (unntatt for de tre metallene kobolt, kadmium og bly). For blåskjell var det flest overskridelser av PROREF for kobber, kvikksølv, bly, sink, PCB og én PAH-forbindelse (pyren, PYR). For torsk var det flest overskridelser for kvikksølv, etterfulgt av sølv og PCB.

For metaller i blåskjell var de høyeste overskridelsene av PROREF for bly på Kvalnes i Sørfjorden og for PCB118 på Akershuskaia indre Oslofjord. Blåskjellstasjoner i indre Oslofjord hadde mange

overskridelser av PROREF, og blant disse hadde Akershuskaia de fleste og høyeste overskridelsene. PCB hadde mange overskridelser som ble påvist i flere urbane stasjoner; havner i Bodø, Ålesund og Bergen. For torsk var det flest overskridelser av PROREF i indre Oslofjord og i havnene i Bergen og Ålesund.

Tidstrender
Totalt 782 tidstrender (både langtidstrender og korttidstrender) ble utregnet for miljøgifter presentert i det utvidede sammendraget. Generelt var det færre langtidstrender (på grunn av utilstrekkelig med data), mens ingen trender eller nedadgående trender dominerte.

Langtidstrender
I blåskjell var langtidstrender (> 10 år) dominert av ingen trender (37%) og nedadgående trender (28%). Oppadgående trender ble observert for 8% av dataene. Et lite antall data hadde utilstrekkelig antall eller data over LOQ for at trender kunne bestemmes. Bildet var likt for langtidstrender i torsk sammenliknet med blåskjell, men prosentandelen av nedadgående trender var høyere (39%). For torsk ble ingen trend observert for 20% av dataene, mens for 6% var det oppadgående trender.

Korttidstrender
For både blåskjell og torsk var det flere datapunkter (miljøgifter x stasjoner) hvor det kunne utregnes korttidstrender (≤ 10 år) enn langtidstrender. Det var «ingen trend» (44%) som dominerte for blåskjell, mens det var nedadgående korttidstrender (45%) som dominerte for torsk. Det ble påvist oppadgående korttidstrender i både blåskjell (13%) og torsk (8%).

For blåskjell ble det funnet tilfeller av oppadgående korttidstrender for de fleste metaller, men de oppadgående trendene var dominert av PCB. Noen av disse trendene for PCB er imidlertid usikre på grunn av få data over LOQ. For torsk var det oppadgående korttidstrender for sølv på syv stasjoner og for kvikksølv på fire stasjoner. Unntatt for nikkel, bly og krom, ble det funnet tilfeller av økende korttidstrender for alle metaller i torsk.

Høyest antall forekomst av oppadgående tidstrender ble funnet på blåskjellstasjoner i indre Oslofjord, og på torskestasjonene ved Lista og Lofoten.

Oppadgående tidstrender med overskridelser av EQS og/eller PROREF
Spesiell oppmerksomhet må gis ved økende tidstrender for miljøgifter som samtidig overskrider EQS og/eller PROREF (>5 for blåskjell og >2 for torsk). Dette var tilfellet for 10 kombinasjoner av miljøgifter, stasjoner og arter. I torskefilet fra indre Oslofjord, Lista og Bømlo var det oppadgående tidstrender i tillegg til overskridelse av EQS (RQ) og PROREF for kvikksølv.

Biologiske effekter
2022-dataene bekreftet resultatene siden 2017 om ingen effekter av TBT for purpursnegl (imposex parameter Vas Deferens Sequence Index, VDSI=0).

ICES/OSPARs vurderingskriterium for bakgrunnsnivå («background assessment criteria», BAC) ble overskredet for median (ikke-normalisert) OH-pyren i torskegalle fra alle stasjoner (indre Oslofjord, Lista og indre Sørfjorden), med unntak av referansestasjonen på Bømlo. Dette viser at fisken har vært eksponert for PAH.

ALA-D aktivitet i torskeblod fra indre Oslofjord var tilsynelatende noe lavere enn i torsk fra referansestasjonen på Bømlo, men ikke statistisk signifikant. I indre Sørfjorden var median ALA-D aktivitet signifikant lavere enn ved referansestasjonen. Redusert aktivitet av ALA-D tyder på høyere

eksponering for bly. Det har generelt vært høyere konsentrasjoner av bly i torskelever fra indre Oslofjord, og indre Sørfjorden, enn i torsk fra Bømlo.

Median EROD-aktivitet i lever av torsk var tilsynelatende lavere ved referansestasjonen på Bømlo, enn i indre Oslofjord og indre Sørfjorden, som kan tyde på eksponering for plane PCBer, PAHer, og dioksiner, men dette var ikke statistisk signifikant. Median EROD-aktivitet var lavere enn ICES/OSPARs bakgrunnsnivå (BAC) på alle stasjoner.

1 Introduction

1.1 Background

The national environmental monitoring programme “Contaminants in coastal waters” (Miljøgifter i kystområdene - MILKYS) is administered by the Norwegian Environment Agency (NEA), that monitors on the levels, trends, and effects of hazardous substances in fjords and coastal waters in Norway including Svalbard on an annually basis. The objective of this monitoring programme is to obtain updated information on levels and trends of selected environmental pollutants. The programme also provides a basis for assessing the state of the environment in Norwegian coastal waters. The monitoring contributes to the Oslo and Paris Commissions (OSPAR’s) Coordinated Environmental Monitoring Programme (CEMP). All the results in this report are considered part of the Norwegian contribution to the CEMP programme as well as to the European Environment Agency (EEA) as part of the assessment under the EU Water Framework Directive (WFD). NEA uses the data for international chemical regulation, reporting, and national knowledge dissemination. The results are also sent to the Norwegian Food Safety Authority (Mattilsynet) to assess warnings for seafood consumption.

1.2 Purpose

The main objective of this environmental monitoring programme is to provide an overview of the status and trends of environmental pollutants in Norwegian marine costal environment as well as to assess the importance of various sources of pollution.

MILKYS provides data to State of the Environment Norway (https://www.environment.no/) which provides the latest information about the state and development of the environment in Norway. This is important as input to Norway's national and international efforts to protect the environment against pollution and to reduce existing pollution. MILKYS data is part of the Norwegian contribution to CEMP which aims to deliver comparable data from across the OSPAR Maritime Area. These data can be used in assessments to address the specific questions raised in the OSPAR's Joint Assessment and Monitoring Programme, and is designed to address issues relevant to OSPAR (OSPAR, 2022) including also OSPAR priority substances[1,2]. The OSPAR Hazardous Substances Strategy is to prevent pollution by hazardous substances, by eliminating their emissions, discharges, and losses, to achieve levels that do not give rise to adverse effects on human health or the marine environment. Under OSPAR, data from MILKYS and other monitoring programmes support this strategy by:

1. Monitoring the levels of a selection of hazardous substances in biota.
2. Evaluating the bioaccumulation of priority hazardous substances in biota of coastal waters.
3. Provide a basis for assessing the effectiveness of previous remedial action.
4. Provide a basis for considering the need for additional remedial action.
5. Assessing the risk to biota in coastal waters.
6. Contribute with monitoring data that is reported in international environmental cooperation Norway is committed to.

[1] https://www.ospar.org/work-areas/hasec/hazardous-substances/priority-action
[2] https://www.ospar.org/work-areas/hasec/hazardous-substances/overview

MILKYS also contributes data to support the implementation of the Water Framework Directive (WFD) (EU, 2000) and the Environmental Quality Standards Directive (EQSD) (EU, 2013) to achieve good chemical status by assessing the results using EU EQSD in Norway. In this regard, Norway has supplemented the EQS with their own EQS for river basin specific pollutants assessed for ecological status. The results from MILKYS can also be useful in addressing aspects of the EU Marine Strategy Framework Directive (MSFD) (EU, 2008). One of the goals of the WFD and MSFD is to achieve concentrations of hazardous substances in the marine environment near background values for naturally occurring substances and close to zero for manmade synthetic substances. OSPAR has also adopted this goal[3].

The MILKYS programme investigates contaminants in blue mussel, cod, dogwhelk, common periwinkle, and common eider on a yearly basis. This report presents the findings from monitoring performed in 2022, the second year of a new five-year period (2021-2025). The program started in 1981 and has since been advanced. The reporting format has been changed from the 2020 investigation (Schøyen et al., 2021) to a shorter report for the 2021 investigation (Schøyen et al., 2022), to this even shorter report for the 2022 investigation due to financial cut and rejection of cut. More complementary information regarding previous programs, such as background history, abbreviations for contaminants, maps etc., can be found in the previous report (Schøyen et al., 2021).

[3] https://www.ospar.org/work-areas/hasec/hazardous-substances

2 Extended summary of MILKYS 2022

2.1 Samples, localities and chemical analyses

Location of stations sampled in MILKYS 2022 are shown in **Figure 1** and number of samples at each station are listed in **Table 1**. Overview of the contaminants selected for presentation of results in extended summary are listed in **Table 2**. The contaminants were selected because they represent the contaminant group, and also they reveal important exceedances of EQS and PROREF. This extended summary presents the main results. Many contaminants in addition to those discussed in the extended summary were analysed, and figures for those contaminants are shown, but not discussed any further in **Supplementary data**. The data is reported to Vannmiljø, ICES and OSPAR.

2.1.1 Samples and monitoring stations

Location of stations sampled in MILKYS 2022 are shown in **Figure 1** and number of samples at each station are listed in **Table 1**. **Table 2** gives an overview of contaminants that are assessed for exceedance of EQS (chapter 3.1) and exceedance of PROREF (chapter 3.2).

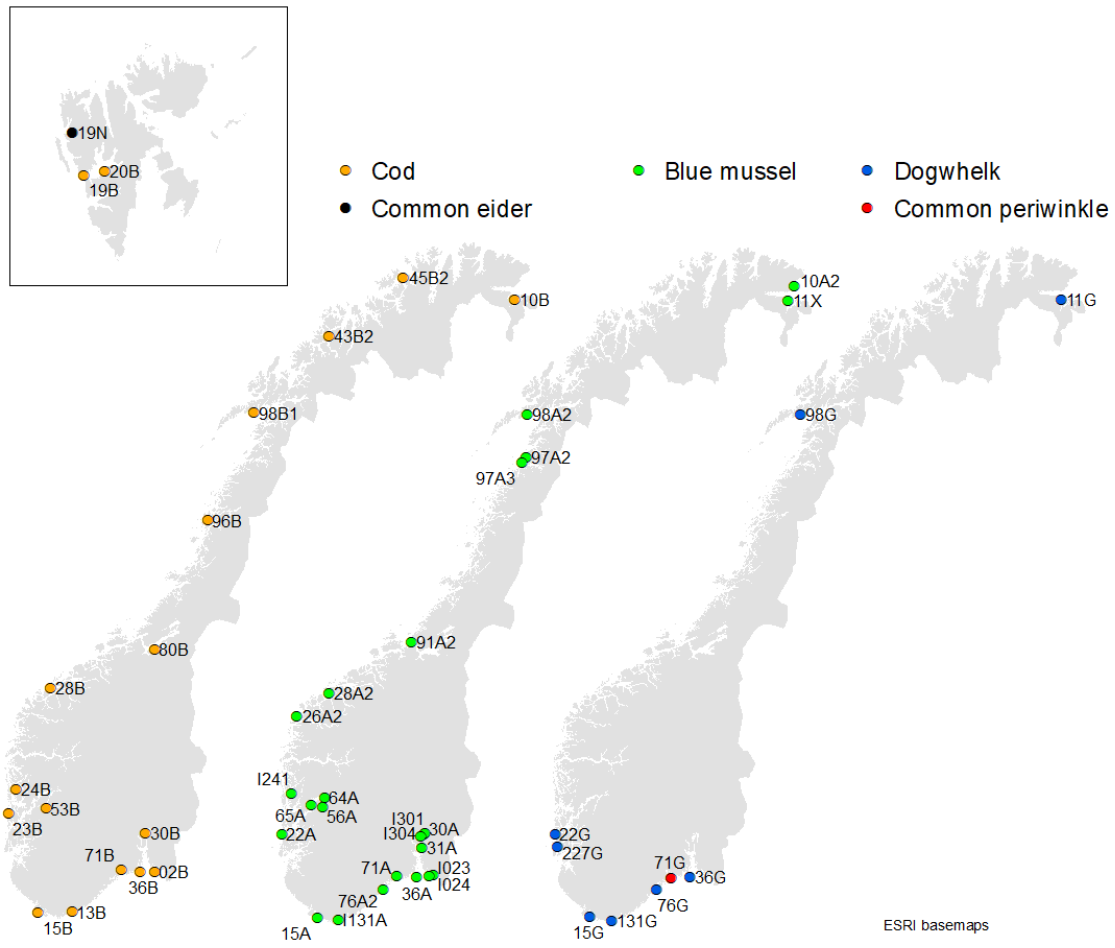


Figure 1. Stations where cod *(Gadus morhua*) and common eider (*Somateria mollissima*) (left), blue mussel (*Mytilus edulis*) (middle), dogwhelk (*Nucella lapillus*) and common periwinkle (*Littorina littorea*) (right) were sampled in Norway and Svalbard (inset) in 2022.

Table 1. Overview of number of samples of blue mussel (*Mytilus edulis*, pooled samples), cod (*Gadus morhua* (pooled for some liver samples)), eider (*Somateria mollissima*), dogwhelk (*Nucella lapillus*), and common periwinkle (*Littorina littorea*) taken at MILKYS stations 2022. All snail samples were pooled. The stations are ordered along the coastline starting north moving south. Due to short time after reversal of financial cuts for eider analysis, only the results for stable isotopes (SIA) are included in this report.

Species	Code	Station name	Latitude	Longitude	Bile	Blood	Egg	Liver	Muscle	Whole soft body
Blue mussel	11X	Brashavn, Varangerfjord	69.8993	29.741						3
	10A2	Skallnes, Varangerfjord	70.1373	30.3417						3
	98A2	Svolvær airport	68.2492	14.6627						3
	97A2	Mjelle, Bodø	67.4127	14.6219						3
	97A3	Bodø harbour	67.2963	14.3956						3
	91A2	Ørland airport	63.6514	9.5639						3
	28A2	Ålesund harbour	62.4659	6.2396						3
	26A2	Måløy, Nordfjord	61.9362	5.0488						3
	I241	Bergen harbour	60.4008	5.304						3
	56A	Kvalnes, Mid Sørfjord	60.2205	6.602						3
	65A	Vikingneset, Mid Hardangerfjord	60.2423	6.1527						3
	64A	Utne, Outer Sørfjord	60.4239	6.6223						3
	22A	Espevær, Bømlo	59.5871	5.152						3
	15A	Ullerøy, Farsund	58.0461	6.9159						3
	I131A	Lastad, Søgne	58.0556	7.7083						3
	76A2	Risøy, Risør	58.7327	9.281						3
	71A	Bjørkøya, Langesundfjord	59.0233	9.7537						2
	36A	Færder, Outer Oslofjord	59.0274	10.525						3
	I304	Gåsøya, Inner Oslofjord	59.8513	10.589						3
	I301	Akershuskaia, Inner Oslofjord	59.9053	10.7363						3
	30A	Gressholmen, Inner Oslofjord	59.8836	10.711						3
	31A	Solbergstrand, Mid Oslofjord	59.6155	10.6515						3
	I024	Kirkøy, Hvaler	59.0791	10.9873						3
	I023	Singlekalven, Hvaler	59.0951	11.1368						3
Cod	20B	Longyearbyen, Svalbard	78.2623	15.4795				15	15	
	19B	Isfjorden, Svalbard	78.17	13.46				10	15	
	10B	Varangerfjord	69.8162	29.7602				15	15	
	45B2	Hammerfest harbour	70.65	23.6333				14	15	
	43B2	Tromsø harbour	69.653	18.974				12	15	
	98B1	Lofoten	68.1858	14.7081				15	15	
	96B	Sandnessjøen	66.0444	12.5036				13	13	
	80B	Trondheim harbour	63.4456	10.3717				15	15	
	28B	Ålesund harbour	62.4678	6.0686				15	15	
	24B	Bergen harbour	60.3966	5.2707				3	7	
	53B	Inner Sørfjord	60.0973	6.5397	15	15		15	15	
	23B	Bømlo	59.8956	5.1086	15	16		15	15	
	15B	Lista	58.0514	6.7469	15			15	15	
	13B	Kristiansand harbour	58.1328	7.9885				2	7	
	71B	Langesundfjord	59.0465	9.7028				11	15	
	36B	Tjøme, Outer Oslofjord	59.0405	10.4358				15	15	
	30B	Inner Oslofjord	59.8127	10.5518	15	15		11	15	
	02B	Hvaler	59.0648	10.9735				15	15	
Dogwhelk	11G	Brashavn, Varangerfjord	69.8995	29.7419						1
	131G	Lastad, Søgne	58.0284	7.699						1
	15G	Ullerøy, Farsund	58.0493	6.9012						1
	227G	Mid Karmsund	59.3396	5.3122						1
	22G	Espevær, Bømlo	59.5837	5.1445						1
	36G	Færder, Outer Oslofjord	59.0278	10.5256						1
	76G	Risøya, Risør	58.728	9.2755						1
	98G	Svolvær airport	68.247	14.6664						1
Common periwinkle	71G	Fugløyskjær, Langesundfjord	58.985	9.8046						1
Common eider	19N	Kongsfjorden, Svalbard	79.004	12.11		15	15			

Table 2. List of parameters that will be shown and discussed in more detail in this report. Number of stations analysed in the species (blue mussel (*Mytilus edulis*), cod (*Gadus morhua*) and snail (*Nucella lapillus* and *Littorina littorea*) are provided and number of stations where the species are measured. An indication of which sections (3.1 EQS and/or 3.2 PROREF) in this report the data are presented is given. Time trends are shown in chapter 3.3 unless marked with "no" in this table.

Contaminant group	Contaminant	Blue mussel	Cod	Snails	Presented in sections	Presented in time trends
Metals	Ag	24	18	0	PROREF	
	As	24	18	0	PROREF	
	Cd	24	18	0	PROREF	
	Co	24	18	0	PROREF	
	Cr	24	18	0	PROREF	
	Cu	24	18	0	PROREF	
	Hg	24	18	0	EQS + PROREF	
	Ni	24	18	0	PROREF	
	Pb	24	18	0	PROREF	
	Zn	24	18	0	PROREF	
PFAS	PFOA	6	11	0	EQS + PROREF	
	PFOS	6	11	0	EQS + PROREF	
	PFOSA	6	11	0	PROREF	
PBDE	BDE47	11	12	0	PROREF	
	BDE100	11	12	0	PROREF	
	BDE154	11	12	0	PROREF	
	LB PBDE6[1]	11	12	0	EQS	no
PCB	CB118	23	18	0	PROREF	
	CB138	23	18	0	PROREF	
	CB153	23	18	0	PROREF	
	LB PCB7[2]	23	18	0	EQS	no
PAH	BAA	7	0	0	EQS + PROREF	
	BAP	7	0	0	EQS + PROREF	
	FLU	7	0	0	EQS + PROREF	
	PYR	7	0	0	PROREF	
	ANT	7	0	0	EQS	no
	NAP	7	0	0	EQS	no
Siloxanes	D5	0	13	0	EQS	
CP	MCCP	11	14	0	EQS	no
	SCCP	11	14	0	EQS	no
DDTs	p,p'-DDE	2	1	0	EQS	no
Pesticides	HCB	2	1	0	EQS	
	HCHG	2	1	0	EQS	no
	QCB	2	1	0	EQS	no
HBCDs	HBCDA	11	14	0	EQS	
TBT-related	TBT	0	0	9	EQS	no
	TPhT	0	0	9	EQS	no

[1] Lower bound of sumPBDE6, i.e. data below LOQ are set to 0 when sum is calculated.
[2] Lower bound of sumPCB7, i.e. data below LOQ are set to 0 when sum is calculated.

2.1.2 Detection frequencies of contaminants and history of Limit Of Quantifications (LOQs)

For this program, there have been changes in laboratories and methods the last 10 years. In the program period from 2012, the analytical provider was changed from NIVA to EF Moss. However, the methods were mainly the same, and only minor changes of the Limit Of Quantification (LOQ) occurred. From 2017, the organic pollutants were analysed at EF GFA, leading to discrepancies in both methods employed and LOQ. For PCBs, the LOQs were increased somewhat (except CB118 which was lowered). LOQ increased from 0.05 to 0.3 µg/kg, and this led to methodical results with artificial upward short-term trends for sumPCB7 in blue mussel for the 2020 survey (Schøyen et al., 2021). The same was also a problem for some stations this year, especially for analyses of blue mussel where the levels in general are lower than in cod liver. Also for cod livers with a high percentage of fat, the LOQ is higher with this method, and LOQs from 0.3 and up to 3 ng/g has been

obtained. For PBDE, the LOQs were mainly lowered somewhat, while for PAHs they were increased for some of the congener: metal analyses were moved to EF WEJ in 2019 and a different method was applied[4] (Green et al., 2020). The changed method had the same or lowered LOQs, except for Ag which had increased LOQ (0.004 mg/kg ww before and 0.05 mg/kg ww with the new methods). For the 2020 survey this led to one artificial upward short-term trend for Ag in blue mussel (Schøyen et al., 2021). This methodological change was accepted by the Norwegian Environment Agency.

The increased LOQs resulted in challenges with calculation of time trends for the contaminants. This is especially the case for stations where the concentrations are low (e.g. blue mussel stations with background concentrations). The current method is more sensitive to changes, and the results have been quality assured by using alternative statistical methods. Read more details in chapter 5.8. This quality assurance has only been done for contaminants in **Table 2**.

[4] Standard method prior to 2019 investigation was Standard method NS EN ISO 17294-2, and then Standard method NS EN ISO 15763 (2010) except for nickel, silver and zinc which then was Standard method NS EN ISO 17294-2-E29.

3 Summary of exceedances (EQS and PROREF) and time trends

Exceedances of EQSs, PROREF and time trends are shown in mosaic plots for species and contaminants. Assessments of EQSs have been done on the tissue as shown in **Table 1**. The EQSs refer to fish (concentrations in whole fish), except in the case of PAHs, where reference is made of crustaceans and mollusc (European Commission, 2014; Fliedner et al., 2018). Therefore, the EQS cannot be directly compared to concentrations found in specific tissues of fish or blue mussel. For example, we have in the present study measured mercury in fish fillet and other contaminants in liver, not in whole fish. Converting mercury concentrations in fish fillet to concentrations in whole fish is uncertain. Using fillet probably represents an overestimate of the whole fish concentration because mercury accumulates more in the fillet than in other tissues (Kwaśniak and Falkowska, 2012). It is assumed, for this exercise, that the same concentration is found in all fish tissue types. Also, contaminants measured in liver samples represents an overestimation compared to whole fish. Fliedner et al. (2018) found that for fillet concentrations, lipid soluble concentrations like PCBs and PBDEs were correlated with lipid concentrations in fillet compared to whole fish. The cod liver is lipid rich, and therefore assessing concentrations of contaminants to EQS in liver is conservative.
For mercury in cod, risk assessments vs. EQS and PROREF are done by using the concentrations measured directly. For the time plots and time trends, the concentrations have been converted to a cod (50 cm size) to account for variability in fish size between years (Ruus et al., 2017). The conversion makes the trends more robust against size variability over time.

How to read mosaic plots
Mosaic plots are a special type of stacked bar chart, where the width of the columns is proportional to the number of observations in each level of the variable plotted on the horizontal axis. The vertical length of the bars is proportional to the number of observations in the second variable (exceedances of EQSs and PROREFs, and time trends). Furthermore, heatmaps are illustrating exceedances and time trends for individual species and stations.

3.1 EQS

Assessment of exceedances of EQS have been done. The contaminants listed in **Table 3** have been determined in 2022, have an EQS in biota (Direktoratsgruppen vanndirektivet, 2018) and are therefore subject to assessment. A total of 293 assessments of EQSs have been done in 2022 (combination of contaminant × sample). Contaminants in eider will be reported in next years' report. Twenty contaminants determined in 2022 had EQSs (**Table 3**).

3.1.1 Species and contaminants

In MILKYS, exceedances of EQSs are considered by the *median* concentration for each station. The species groups blue mussel, cod, and snails were analysed for contaminants with an assigned EQS, and exceedances are shown in **Figure 2**. EQS were exceeded in 15% of all selected contaminants in blue mussel and 36% of all contaminants in cod, expressed as datapoints (contaminants x stations). No exceedances were observed for contaminants determined in snails.

Table 3. List of contaminants determined in 2022 for which an EQS exist. The EQSs are given in µg/kg (ng/g ww). The compound is a priority compound unless marked with "yes" in the column RBSP.

Contaminant Group	Contaminant	EQS (µg/kg ww)	River basin specific pollutants (RBSP)
Metals	Mercury (Hg)	20	
PFAS	Perfluorooctanoic acid (PFOA)	91	yes
	Perfluorooctanesulfonic acid (PFOS)	9.1	
PBDEs	Sum of PBDE congeners -28, -47, -99, -100, -153, -154 (sumPBDE6)	0.0085	
PAHs	Anthracene (ANT)	2,400	
	Benzo(a)anthracene (BAA)	300	yes
	Benzo(a)pyrene (BAP)	5	
	Fluoranthene (FLU)	30	
	Naphthalene (NAP)	2,400	
PCBs	Sum of PCB congeners -28, -52, -101, -118, -138, -153, and -180 (sumPCB7)	0.6	yes
Siloxanes	Decamethylcyclopentasiloxane (D5)	15,217	yes
CCPs	Chlorinated paraffins (MCCP (C14-C17))	170	yes
	Chlorinated paraffins (SCCP (C10-C13)	6,000	
HBCDs	Hexabromocyclododecane (HBCDD)	167	
DDTs	Dichlorodiphenyldichloroethylene (p,p'-DDE)	610	
Pesticides	Hexachlorobenzene (HCB)	10	
	Pentachlorobenzene (QCB)	50	
	Hexachlorocyclohexane (HCHG)	61	
TBT-related	Tributyltin (TBT)	150	
	Triphenyltin (TPhT)	150	yes

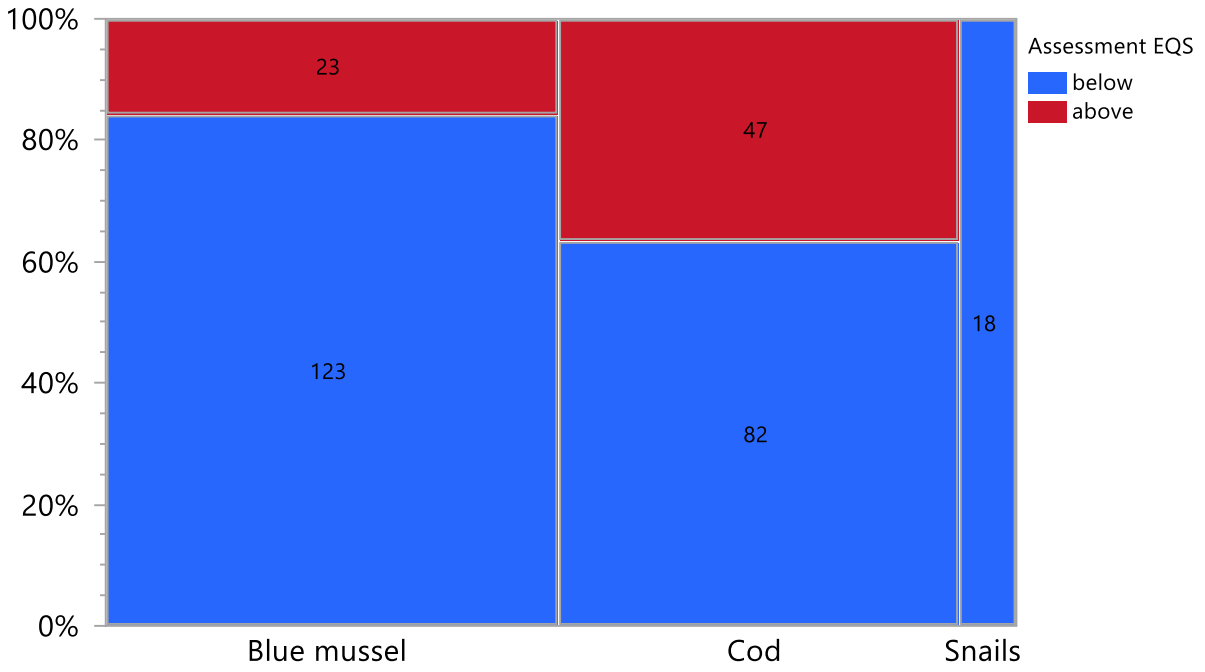


Figure 2. Exceedances of EQSs by species groups in a mosaic plot. The cells are labelled by the number of datapoints (contaminants x stations). The exceedances are considered by the median for each station and species. The colours represent below or above EQSs. The total area of the figures represents the 293 assessments of EQS.

To illustrate which contaminants that had concentrations exceeding EQS, **Figure 3** to **Figure 5** illustrate this for blue mussel, cod, and snails, respectively. Furthermore, heatmaps of concentration

exceedances at individual station and contaminant are shown in **Figure 6** and **Figure 7** for blue mussel and cod respectively.

In blue mussel (**Figure 3**), compounds with concentrations exceeding EQS were mercury (at three stations), sumPBDE6 (all but two stations exceeded EQS), and sumPCB7 (11 stations). The EQS for sumPBDE6 is very low to protect human health (European Commission, 2014).

In cod (**Figure 4**), all median concentrations of sumPBDE6 and sumPCB7 exceeded EQS. SumPCB7 is a RBSP, and is sometimes exceeded also in freshwater trout from supposedly pristine rivers in Norway (Moe et al., 2019, 2018; Sandin et al., 2021; Thrane et al., 2020). Only two stations did not exceed EQS for mercury. MCCP exceeded EQS at one station.

Two contaminants (TBT and TPhT) were analysed in snails (**Figure 5**), and no exceedances of EQSs were seen.

Contaminants often exceeding EQSs are therefore mercury, sumPBDE6 and sumPCB7 for blue mussel and cod. MCCP exceeded EQS at one station for cod.

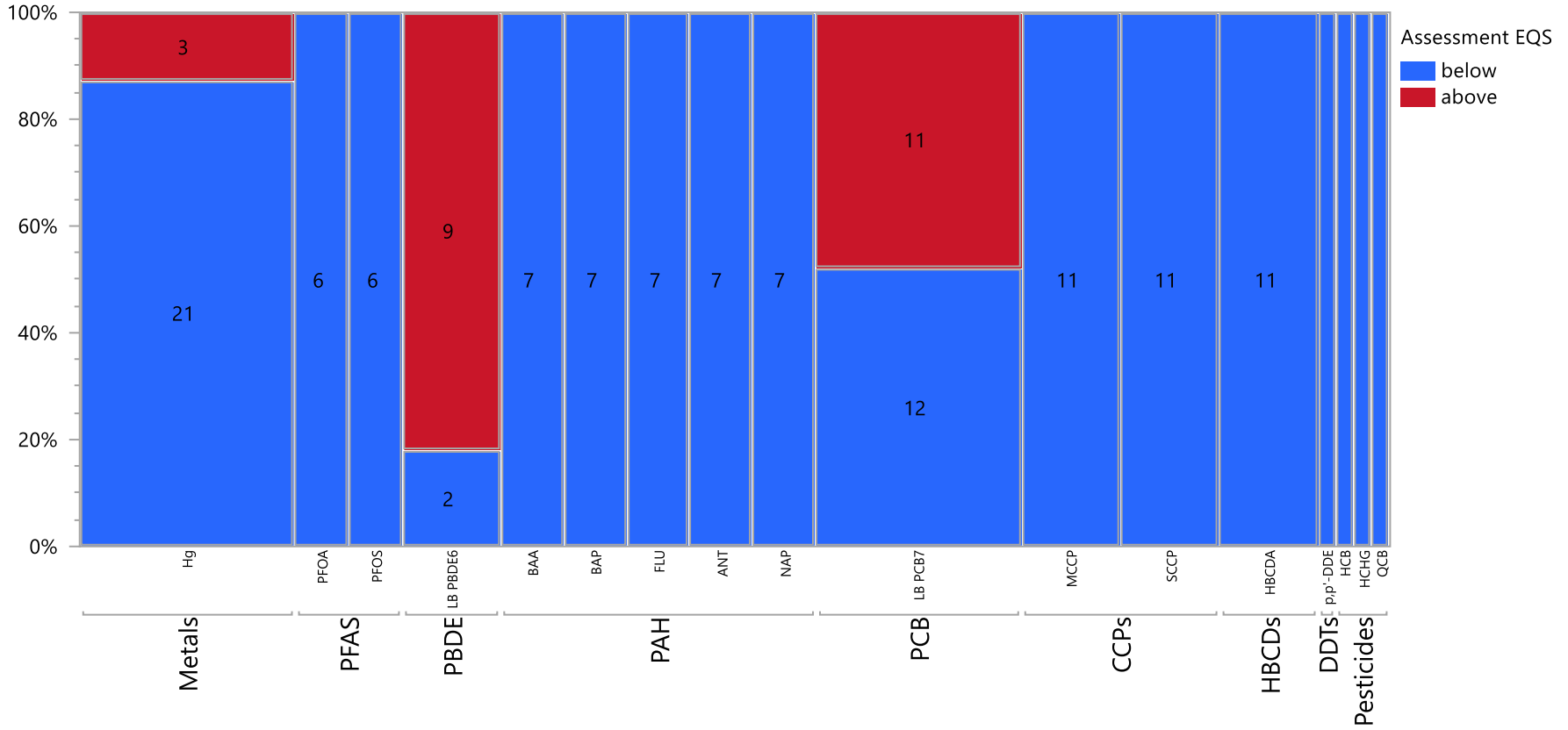


Figure 3. Exceedances of EQSs in blue mussel by contaminant and contaminant group. The cells are labelled by the number of stations in each category. The exceedances are considered by the median for each station. The colours represent below or above EQSs.

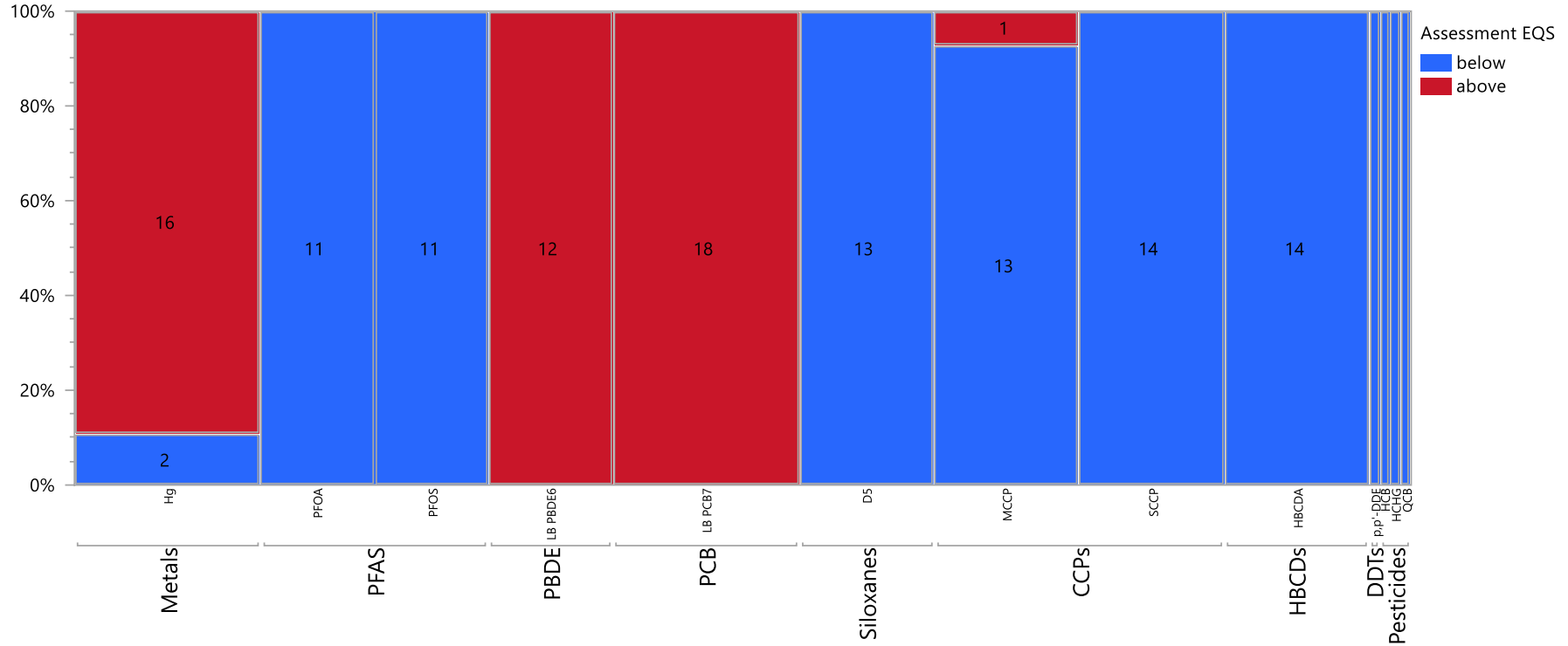


Figure 4. Exceedances of EQSs in cod by contaminant and contaminant group. The cells are labelled by the number of stations sampled. The exceedances are considered by the median for each station. The colours represent below or above EQSs.

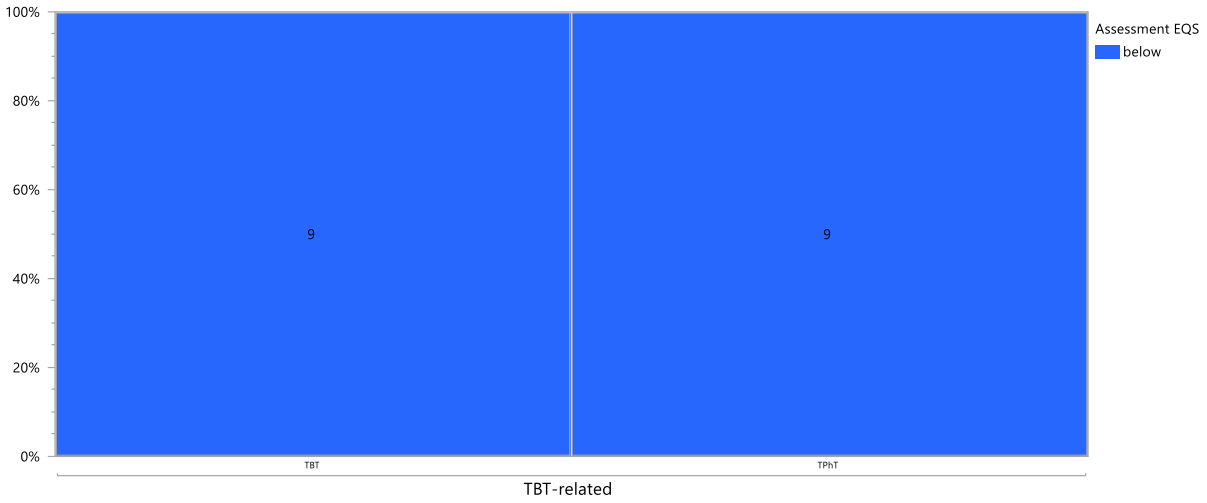


Figure 5. No exceedances of EQSs in snails (dogwhelk/common periwinkle) by contaminant. The number of stations in each group are labelled in the respective cell. The colour represents below EQSs.

3.1.2 Heatmaps for stations

To investigate potential pattern in stations exceeding EQSs, heatmaps for contaminants vs. stations are shown in **Figure 6** and **Figure 7**. No comments are made if we could not detect any special stations standing out compared to others.

In blue mussel, exceedances of mercury concentrations were observed at three stations (Kirkøy at Hvaler (I024), Bjørkøya in the Langesundfjord (71A), and Kvalnes in the Mid Sørfjord (56A)). Concentrations of sumPBDE6 exceeded EQSs at all but two stations investigated (Singlekalven at Hvaler (I023) and Gressholmen in the Inner Oslofjord (30A)). For sumPCB7, half of the stations (11 of 23) showed exceedances of EQSs. This is an improvement from 2021, for both sumPBDE6 and sumPCB7.

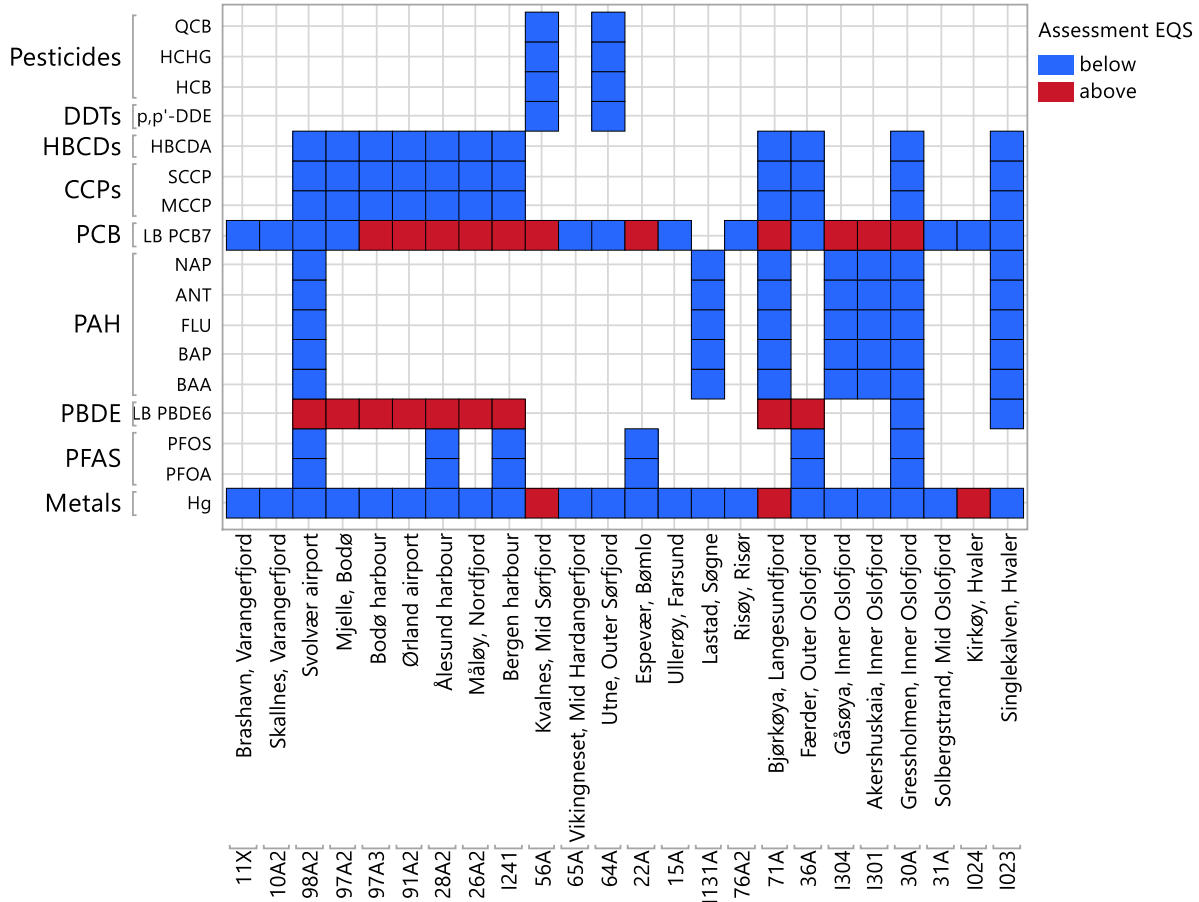


Figure 6. Heatmap of exceedances of EQSs in blue mussel. The exceedances are considered by the median for each station. The colours represent below or above EQSs. Empty "cells" mean that the contaminant was not analysed at the indicated station. Grey lines show the midpoint of each station and contaminant. The stations are ordered along the coastline starting north moving south.

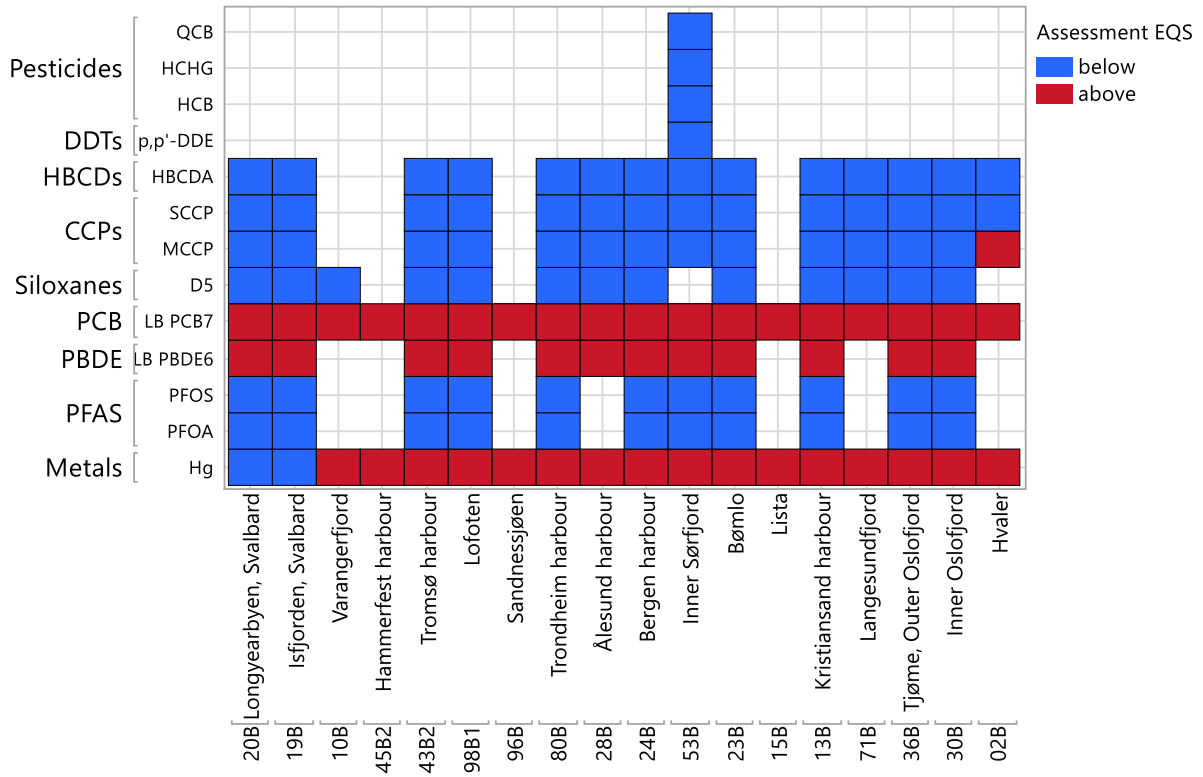


Figure 7. Heatmap of exceedance of EQSs in cod. The exceedances are considered by the median for each station. The colours represent below or above exceedance of EQSs. Empty "cells" mean that the contaminant was not analysed for at the indicated station. Grey lines show the midpoint of each station and contaminant, and darker lines have been inserted between contaminant groups. The stations are ordered along the coastline starting north moving south.

For cod stations, also mercury, sumPBDE6, and sumPCB7 were the compounds where exceedances of EQSs mainly were observed. Concentrations of MCCD exceeded EQS at one station (Kirkøy at Hvaler 02B). Mercury concentrations exceeded EQS at all stations but the two at Svalbard (Longyearbyen (20B) and Isfjorden (19B)).

3.1.3 Sum of risk quotients (RQ) - comparison between stations

Based on a publication (Backhaus and Faust, 2012), we have developed the sum of risk quotients (sum RQ, see details in chapter 5.6) as an aid for comparison of levels between stations. The RQ is the measured concentration divided by the PNEC for the contaminant. For a given station, sum RQ is the sum of all measured contaminants divided by their PNEC. The fish and blue mussel have been normalised to a "standard fish" with 5% lipid and 26% dry weight for fish, and "standard mussel" with 1% lipid and 8.3% dry weight as described in the guidance document (European Commission, 2014). For more information of the normalisation process, please read chapter 5.6.

A direct comparison of sum RQ is quite easy if the same contaminants have been analysed at all the stations. If the analytical repertoire is the same for all stations, the highest sum RQ identify the station(s) where the risks for environmental effects are highest. However, since the analytical repertoire was not the same at all stations, sum RQ is more difficult to use in our case. As can be seen in e.g. **Figure 7**, EQS for sumPBDE6 was exceeded for all cod stations where it was analysed. However, PBDEs were not analysed at all cod stations. Therefore, it is challenging to compare sum RQ at stations where PBDEs have been analysed to stations where PBDEs have not been analysed.

Four compound groups exceeded EQS in one or more stations; mercury, PBDE, PCB and CP (i.e. MCCP). Mercury was analysed at all stations, but the other compound groups exceeding EQS were not analysed at all stations. Of these, sumPBDE6 is the compound which is not analysed most often (11 of 24 mussel stations, and 12 of 18 cod stations). However, this also applies to a less extent to PCB (analysed at 23 of 24 mussel stations and all cod stations) and MCCP (analysed at 11 of 23 mussel stations, and 14 of 18 cod stations). For mussel stations, PAH were analysed only at seven stations. We have therefore chosen to present stations in three (cod) and four (blue mussel) categories for sum RQ. Compound groups that were *not analysed* at the stations are therefore used for grouping of stations in the sum RQ figures.

For mussel stations (**Figure 8**), the sum RQ varied between 26 and 0.13, with eight stations not exceeding sum RQ ≥ 1 (I023, 64A, I131A, 65A, 15A, 76A2, 11X, and 10A2). Four stations lacked analysis of PBDE, CP and PAH, while station Lastad in Søgne (I131A) lacked analysis of PBDE, PCB, and CP. The urban stations (Akershuskaia (I301), Gressholmen (30A), Ålesund (28A2), Bergen (I241), and Bodø harbour (97A3)) had the highest RQ. Akershuskaia in the Inner Oslofjord had the highest sum RQ despite that PBDE and CP was not analysed at the station.

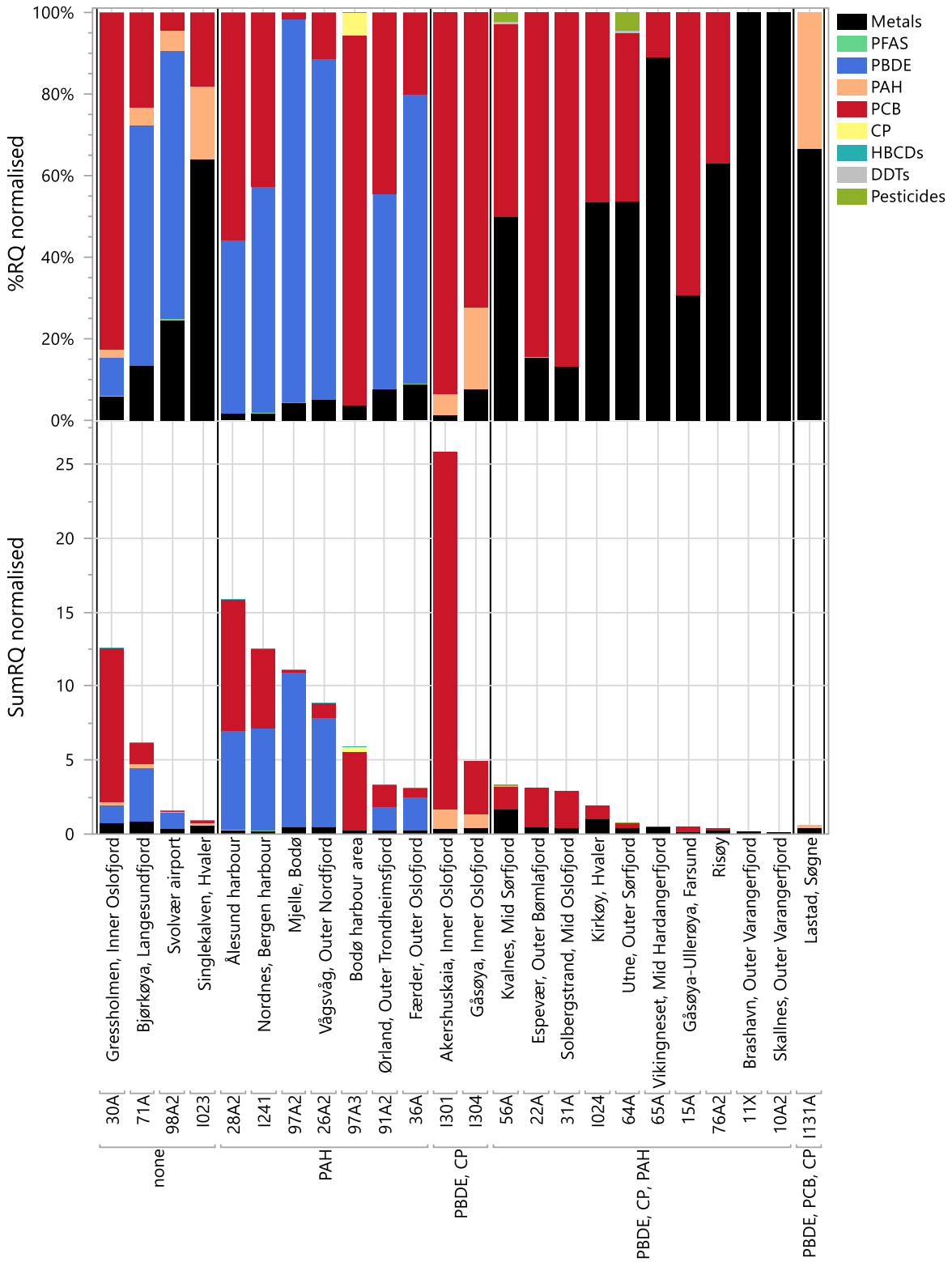


Figure 8. Stacked barplot of sum RQ for blue mussel stations grouped by analyses *not performed* for the stations (none: indicating that the stations were analysed for all compounds that had any exceedance of EQS). On the top, the % of sum RQ normalised for groups are shown, on the bottom the sum of RQ normalised.

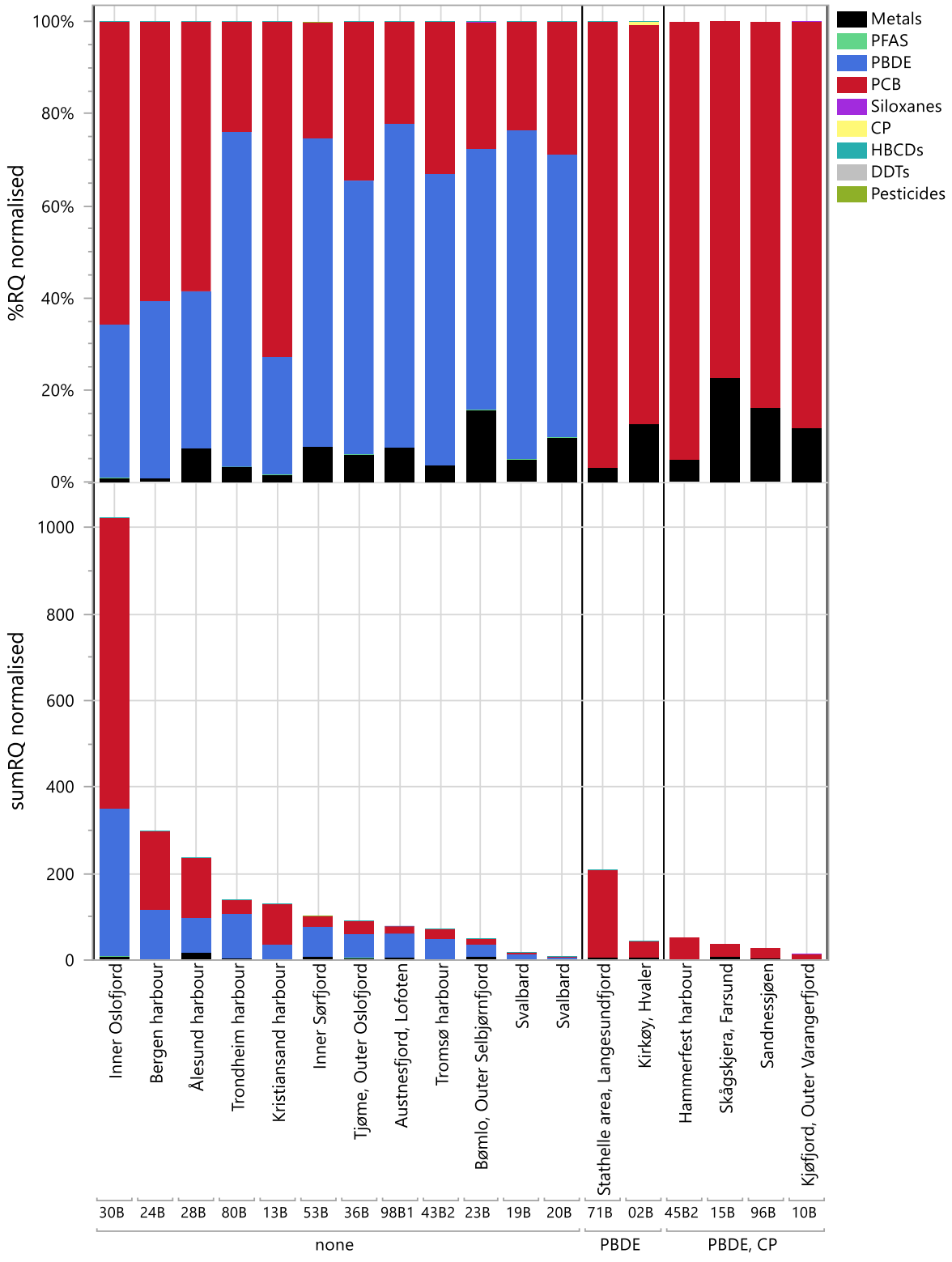


Figure 9. Stacked barplot of sum RQ for cod stations grouped by analyses *not performed* for the stations (none; indicating that the stations were analysed for all compounds that had any exceedance of EQS). On the top, the % of sum RQ normalised for groups is shown, on the bottom the sum RQ normalised.

For cod stations (**Figure 9**), the sum RQ varied between 1000 and 8.1, all stations exceed sum RQ ≥ 1. The two most important compounds contributing to exceedance were sumPCB7 and sumPBDE6, but also mercury contribute. All these compounds are shown to biomagnify in cod, which is the reason why the sum RQ is so much higher in cod than in mussel. The urban stations (Inner Oslofjord, Bergen, Ålesund Trondheim and Kristiansand harbour) had the highest sum RQ. Langesundfjord (71B) had the second highest RQ for PCB, only surpassed by Inner Oslofjord.

The median lipid% of cod liver stations was 47%, but median value for each station varied from 23 to 60%. For blue mussel, the lipid% was a median 0.91% (0.28-2.1%). The purpose of normalisation was to make a comparison between stations less dependent of the lipid%. Also, the contribution to risk of compounds that do not bioaccumulate in lipids (e.g. mercury, PFOS and PFOA) are more balanced.

3.2 PROREF

Concentrations of contaminants were compared to assumed reference levels, by a NIVA-developed tool denoted Norwegian provisional high reference contaminant concentration (PROREF, se chapter 5.7). PROREF is a comprehensive set of species-tissue-basis-specific contaminant concentrations that are statistically low when considering all MILKYS-results for the period 1991-2016. This tool sets reference concentrations for contaminants in an objective way, based on selecting stations with significantly lower values than other stations, instead of subjectively selecting stations in areas presumed remote from point sources of contamination. It thus provides a valuable method for assessing contaminants levels in addition to the risk based EQS. A total of 701 assessments for PROREF have been made for the 25 contaminants (**Table 4**) selected for presentation for 2022 data. There are minor changes in the selection of compounds vs. the 2021-report. BDE99 is not included in 2022, and instead of BDE153, BDE154 were selected in 2022. Results for other contaminants with PROREF, but not selected for presentation in 2022, are given in **Supplementary** data.

3.2.1 Species and contaminants

The PROREFs are at higher concentrations in cod than in mussels (except three metals; cobalt, cadmium and lead, **Table 4**). PROREFs have not been developed for PFAS in blue mussel yet due to low detection frequencies. PAHs are metabolised by cod and therefore PROREFs have not been developed for PAH in cod.

Table 4. List of contaminants selected in 2022 for which a PROREF exist. The PROREFs are given in mg/kg ww for metals and µg/kg ww (ng/g ww) for others. Data are given with two significant digits.

Contaminant group	Contaminant	Unit	PROREF blue mussel	PROREF cod
Metals	Silver (Ag)	mg/kg ww	0.0086	0.93
	Arsenic (As)		2.5	13
	Cadmium (Cd)		0.18	0.14
	Cobalt (Co)		0.08	0.06
	Chromium (Cr)		0.36	0.40
	Copper (Cu)		1.4	14
	Mercury (Hg)		0.012	0.056
	Nickel (N)i		0.29	0.65
	Lead (Pb)		0.20	0.05
	Zinc (Zn)		18	35
PFAS	Perfluorooctanoic acid (PFOA)	µg/kg ww		10
	Perfluorooctanesulfonic acid (PFOS)			10
	Perfluorooctanesulfonamide (PFOSA)			6.2
PBDEs	PBDE congener 47 (BDE47)		0.17	16
	PBDE congener 100 (BDE100)		0.05	2.6
	PBDE congener 154 (BDE154)		0.05	1.5
PAHs	Benzo(a)anthracene (BAA)		1.5	
	Benzo(a)pyrene (BAP)		1.2	
	Fluoranthene (FLU)		5.4	
	Pyrene (PYR)		1.0	
PCBs	PCB congener 118 (CB118)		0.07	100
	PCB congener 138 (CB138)		0.2	160
	PCB congener 153 (CB153)		0.26	190
HBCD	α-hexabromocyclododecane (HBCDA)		0.110	7
Pesticides	Hexachlorobenzene (HCB)		0.1	14

Exceedances of PROREF in different species are shown in **Figure 10**. Blue mussel exceeded PROREF less than cod. For mussel, 40 of the samples could not be classified vs. PROREF since LOQ was higher

than PROREF. The highest exceedances for mussels were 10-20x, while the highest exceedances for cod were 5-10x.

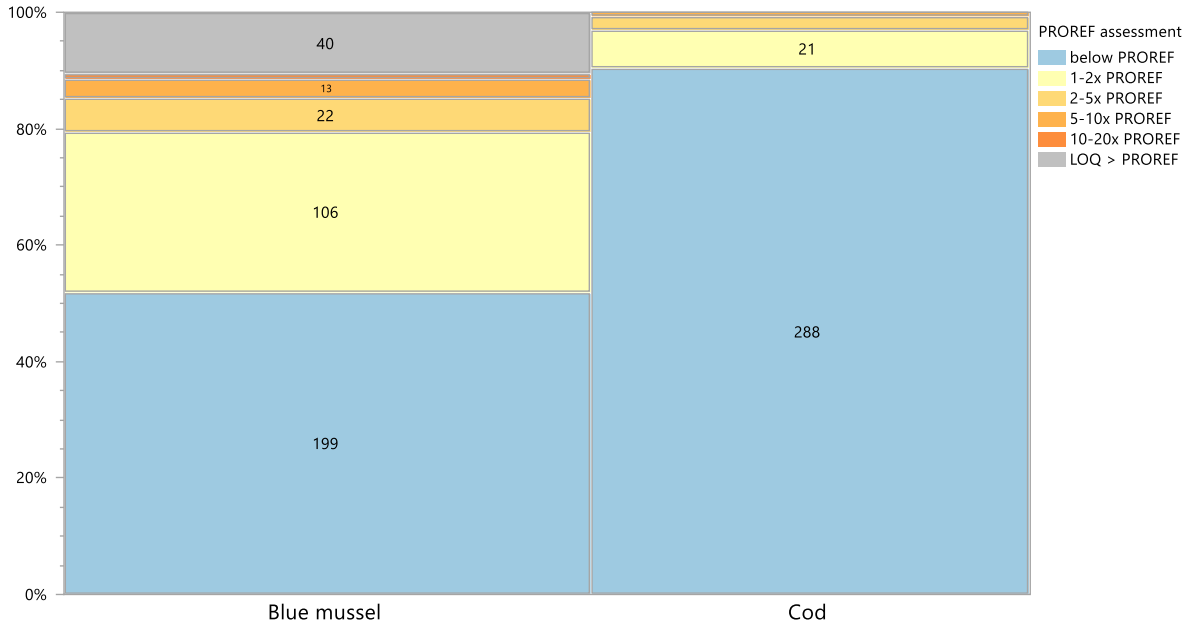


Figure 10. Exceedances of PROREF in a mosaic plot. The cells are labelled by the number of datapoints (contaminants x stations). The exceedances are considered by the median for each station and species. The colours represent below (blue) or above PROFEF (darker yellow to red), or that the PROREF was below LOQ, and therefore could not be classified (grey).

For mussels, all metals except silver had stations where concentrations exceeded PROREF (**Figure 11**). For silver, the LOQ was higher than PROREF, and therefore no assessment could be made. Among the metals, the highest exceedances were observed for lead (10-20x PROREF). The highest exceedances were seen for PCBs (CB118, CB138 and CB153). For CB138, no stations were below PROREF, but six stations had LOQs too high for assessing the concentrations vs. PROREF. PAHs were analysed in seven stations, and exceedances above PROREF were observed for benzo(a)anthracene, fluoranthene, and pyrene.

In cod, mercury was the contaminant exceeding PROREF the most (**Figure 12**). Among the metals, silver, arsenic, and cadmium also had concentrations exceeding PROREF. BDEs and CBs exceeded PROREF at a few stations each.

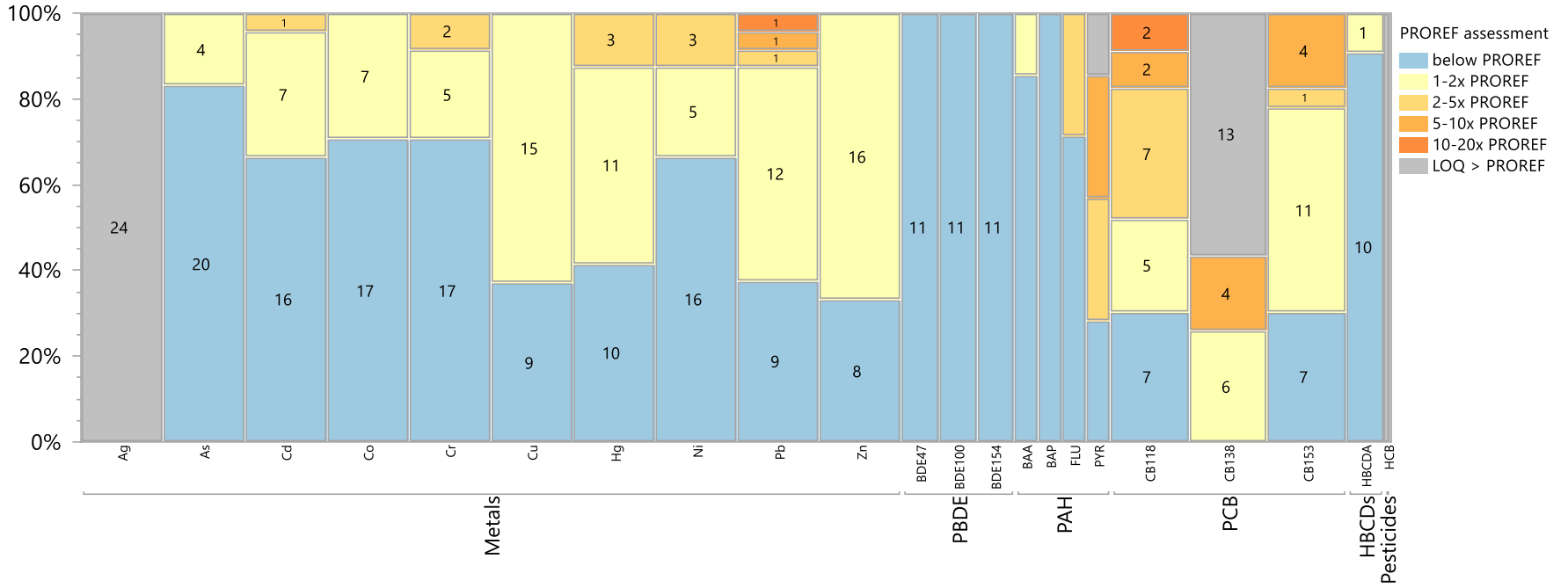


Figure 11. Exceedances of PROREF in blue mussel by contaminant and contaminant group. The cells are labelled by the number of stations sampled. The exceedances are considered by the median for each station. The colours represent below or above exceedance of PROFEF (darker yellow to red), or that the PROREF was below LOQ, and therefore could not be classified (grey).

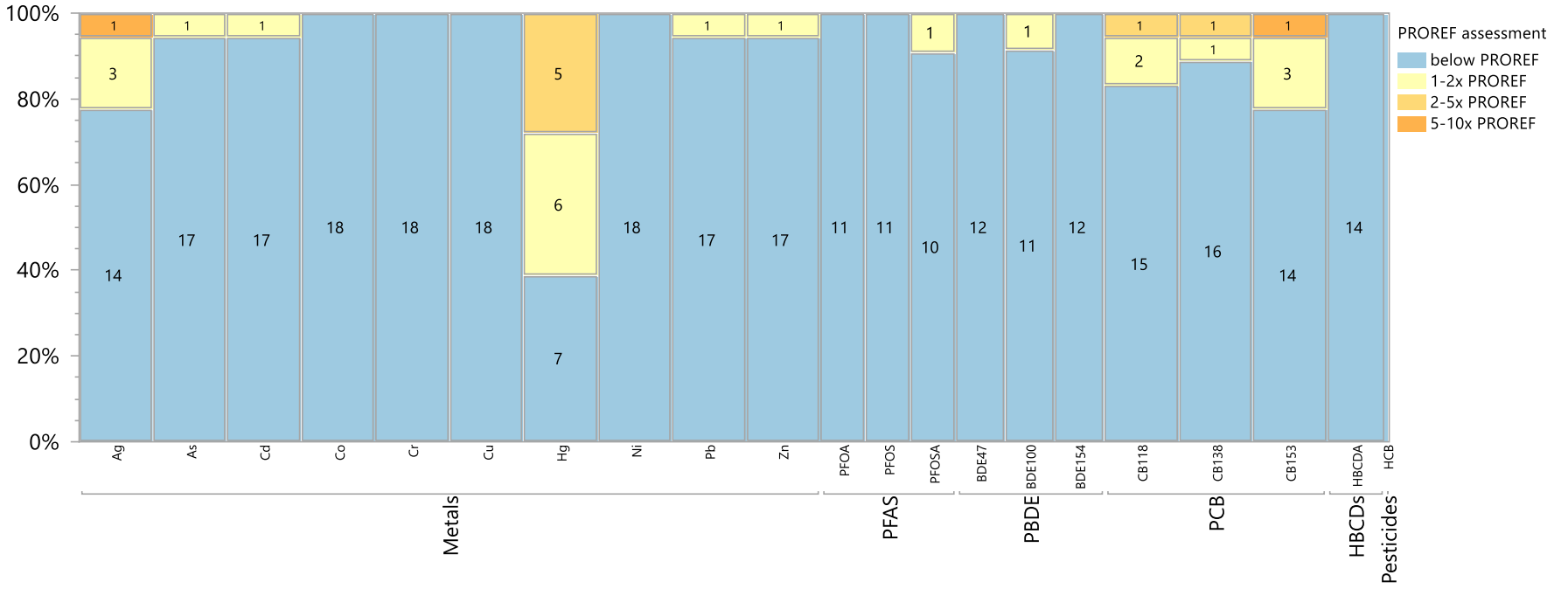


Figure 12. Exceedances of PROREF in cod by contaminant and contaminant group. The cells are labelled by the number of stations sampled. The exceedances are considered by the median for each station. The colours represent below or above exceedance of PROFEF (darker yellow to red), or that the PROREF was below LOQ, and therefore could not be classified (grey).

3.2.2 Heatmaps for stations

Blue mussel stations in the Inner Oslofjord (**Figure 13**) had many exceedances of PROREFs, and among these, Akershuskaia (I301) had the most and highest exceedances. PCBs had the highest exceedances, which were observed in several urban stations (harbours of Bodø (97A3), Ålesund (28A2), and Bergen (I241)). Brashavn (11X) in the Varangerfjord had only one compound (cadmium) exceeding PROREF, while Risøy (76A2) did not have any exceedances of PROREF.

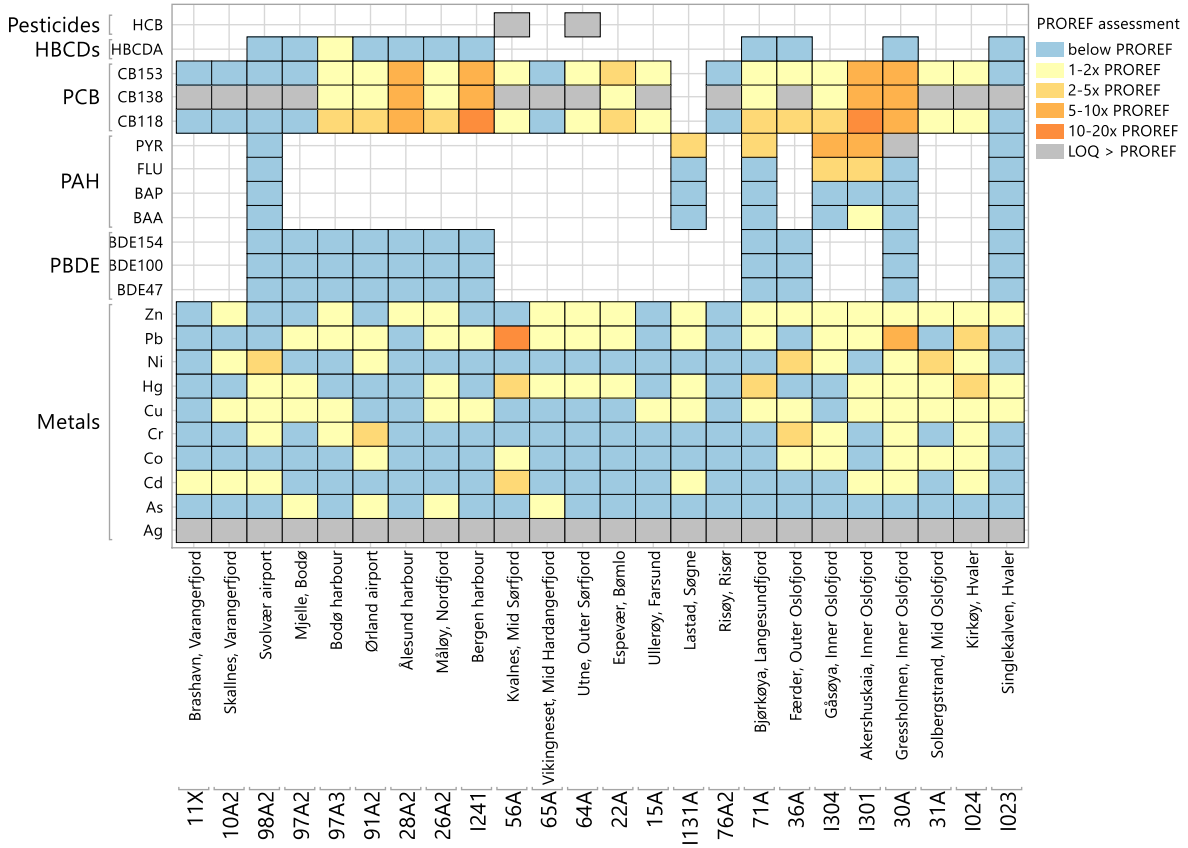


Figure 13. Heatmap of exceedances of PROREF in blue mussel. The colours represent below or above exceedance of PROREF. Empty "cells" mean that the contaminant was not analysed for at the indicated station. Grey lines show the midpoint of each station and contaminant. The stations are ordered along the coastline starting north moving south.

Exceedances of PROREF in cod (**Figure 14**) were most often observed at the stations in the Inner Oslofjord (30B) followed by Ålesund (28B), and Bergen (24B) harbours. Four stations did not have exceedances of PROREF for any compounds investigated (Longyearbyen (20B) and Isfjorden (19B) at Svalbard), the Varangerfjord (10B), and Tromsø harbour (43B2).

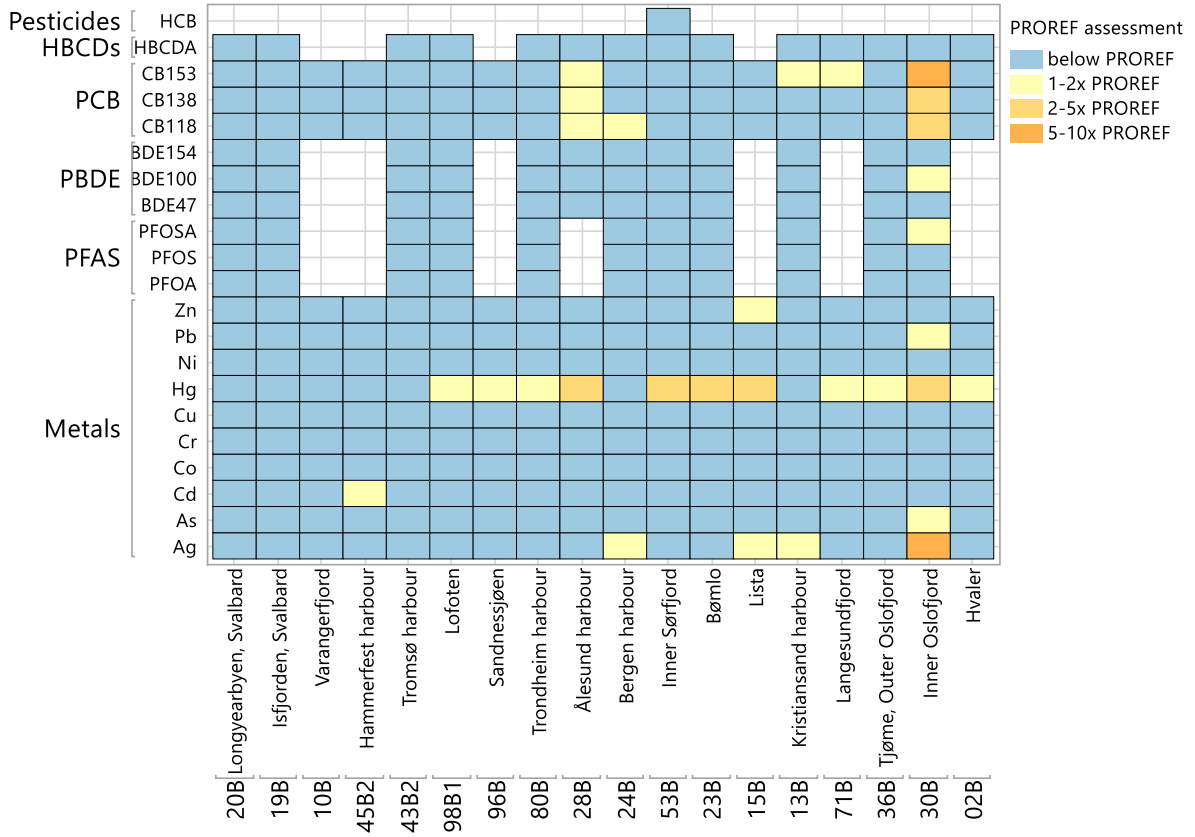


Figure 14. Heatmap of exceedances of PROREF in cod. The colours represent below or above exceedance of PROREF. Empty "cells" mean that the contaminant was not analysed for at the indicated station. Grey lines show the midpoint of each station and contaminant. The stations are ordered along the coastline starting north moving south.

3.2.3 Sum of PROREFratio above background level

As with sum RQ, we did a similar assessment of stations for PROREF. However, for sum PROREFratio, we only included contaminants above background levels (i.e. PROREF>1). As for sum RQ, the comparison between stations was hampered by different contaminant groups being analysed at each station. Stations are therefore grouped by the compounds *not analysed* at the station.

For blue mussel, one station (Risøy, 76A2) did not contain any groups above background levels and are therefore not shown in **Figure 15**. The sum of PROREFratio above background levels varied from 48 to 1.2 (Brashavn (11X) in the Varangerfjord). Akershuskaia (I301) in the Inner Oslofjord had the highest sum PROREFratio. PBDEs were not analysed at this station, but no exceedances of PROREF was observed for PBDE. Also, other stations in Oslofjord, Bergen and Ålesund harbour had high sum PROREFratio.

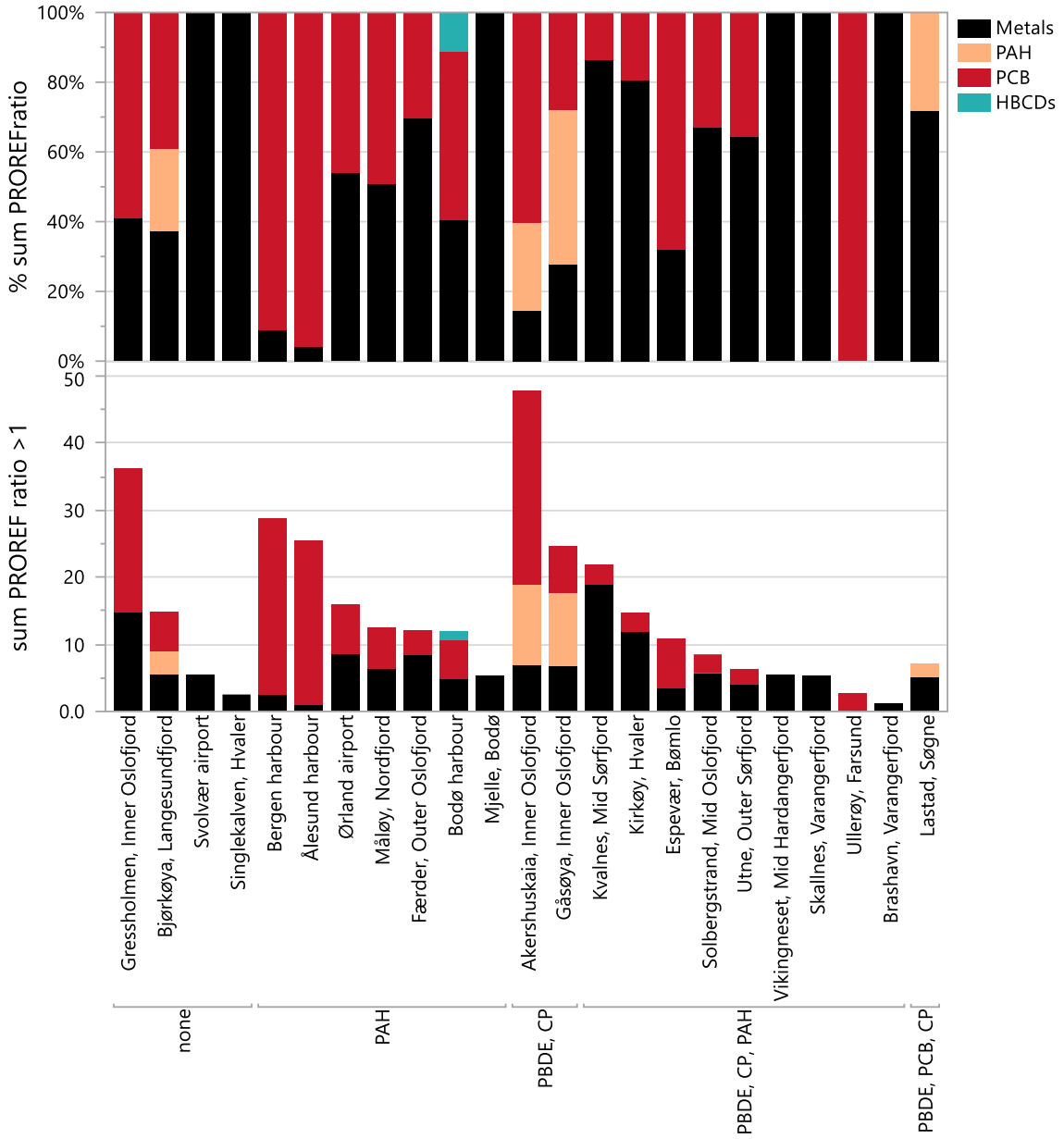


Figure 15. Stacked barplot of sum PROREFratio above background levels for blue mussel stations grouped by analyses *not performed* for the stations (CP have no derived PROREF yet). On the top, the % of sum PROREFratio>1 for individual groups is shown, on the bottom the sum PROREFratio>1.

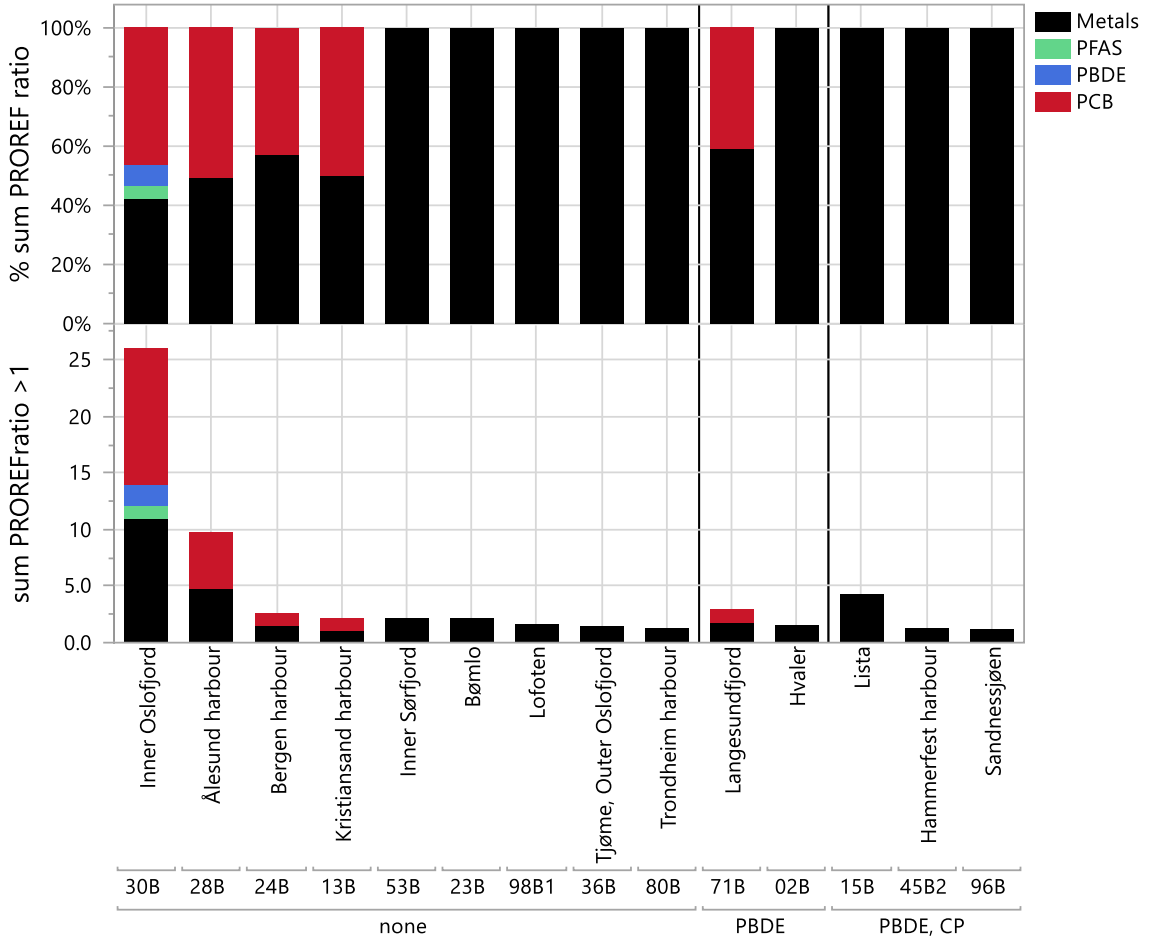


Figure 16. Stacked barplot of sum PROREFratio above background levels for cod stations grouped by analyses *not performed* for the stations (CP have no derived PROREF yet). On the top, the % of sum PROREF>1 for individual groups is shown, on the bottom the sum PROREFratio.

Four cod stations (Tromsø harbour (43B2), the Varangerfjord (10B), Isfjorden (19B) and Longyearbyen (20B)) had no exceedance of PROREF background levels, and are therefore not in **Figure 16**. The sum PROREFratio above background levels varied from 26 to 1.2. The Inner Oslofjord (30B) had sum PROREFratio higher than other stations, with contributions from metals, PFAS, PBDE and PCB.

3.3 Time trends

3.3.1 Species and contaminants

Time trends for selected contaminants (**Table 5**) were assessed. In total 782 time trends in each of two time span categories (short-term and long-term) were estimated (combination of selected contaminant × station × tissue). Figures for time trends for contaminants not selected for presentation in extended summary are shown (but not commented) in **Supplementary** data.

Table 5. Contaminants selected for describing time trends.

Group	Contaminant	Number of stations with trends (long-, and short-term)	
		Blue mussel	Cod
Metals	Ag	24	18
	As	24	18
	Cd	24	18
	Co	24	18
	Cr	24	18
	Cu	24	18
	Hg	24	18
	Ni	24	18
	Pb	24	18
	Zn	24	18
PFAS	PFOA	6	11
	PFOS	6	11
	PFOSA	6	11
PBDE	BDE47	11	12
	BDE100	11	12
	BDE154	11	12
PAH	BAA	7	0
	FLU	7	0
	PYR	7	0
	BAP	7	0
PCB	CB118	23	18
	CB138	23	18
	CB153	23	18
Siloxanes	D5	0	13
CP	MCCP	11	14
	SCCP	11	14
HBCDs	HBCDA	11	14
Pesticides	HCB	2	1
All	All	423	359

Time trends (long-term (>10 years) and short-term (≤ 10 years)) for blue mussel and cod are shown in **Figure 17** to **Figure 19**. Heatmaps of time trends (stations vs. contaminants) are shown in **Figure 20** and **Figure 21**. In this report we distinguish between no trend (i.e. a flat time trend during the period) and no change (there has been a nonlinear trend during the period, for instance an increase followed by a decrease, but when comparing the start and end, there is no change). For examples of this distinction, please refer to examples in chapter 5.8.

A part (20-30%) of the time trends could not be determined due to lacks in the dataset (too few data or trend estimation failure). Long-term time trends in blue mussel were dominated by no trend/no change followed by decreasing trends (**Figure 17**). However, also increasing trends were observed. In cod, decreasing trends dominated, followed by no trend (**Figure 17**).

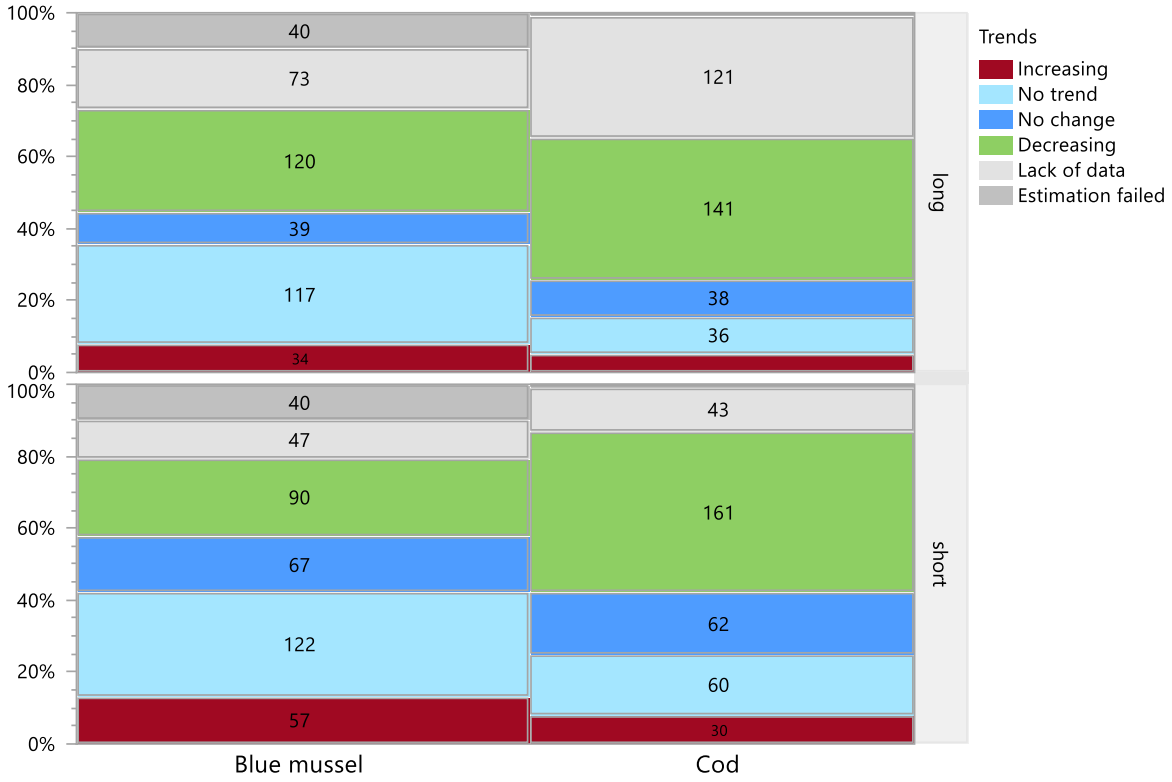


Figure 17. Mosaic plot of time trends for blue mussel and cod. Upper panel shows long-term trends, while lower panel shows short-term trends. The number of trends (stations × contaminants) are indicated in the respective cells. The total number of time trends in each category (long- and short-term) was 423 for blue mussel and 359 for cod.

In blue mussel (**Figure 18**), decreasing long-term trends were more frequent than increasing trends, and was found for most contaminants. Increasing long-term trends were found for all metals except cobalt, and were also found for PCB (CB118, CB138 and CB153) and PBDE (BDE47 and BDE100).

The overall picture was roughly the same for short-term trends, but more short-term increasing trends for PCBs were found. Some trends (both short- and long-term trends) have been quality assured with other methods because increased LOQs is challenging. In 2017 for PCB and in 2019 for silver, the LOQs of the chemical analyses were increased. The confidence in the time trends is therefore limited for silver and PCBs in blue mussel at low-concentration stations as the increased LOQ may affect the trend estimation. For PCB, such stations are Skallnes (10A2), Brashavn (11X), Vikingneset (65A), and Mjelle (97A2). For silver, such stations are Gressholmen (30A), Solbergstrand (31A), Færder (36A), Utne (64A), Bjørkøya (71A), Svolvær airport (98A2), and Akershuskaia (I301). For confirmation of these trends, analyses at a laboratory with lower LOQs are necessary. This is not planned for the program period 2021-2025.

Long-term trends in cod (**Figure 19**) were also dominated by decreasing time trends. Silver and mercury had the highest percentage of increasing time trends, both short-term and long-term. Short-term time trends in cod were also dominated by decreasing trends. Dominating decreasing trends were found for several contaminants (chromium, nickel, lead, PFOS, PFOSA, MCCP, SCCP, and HBCDA).

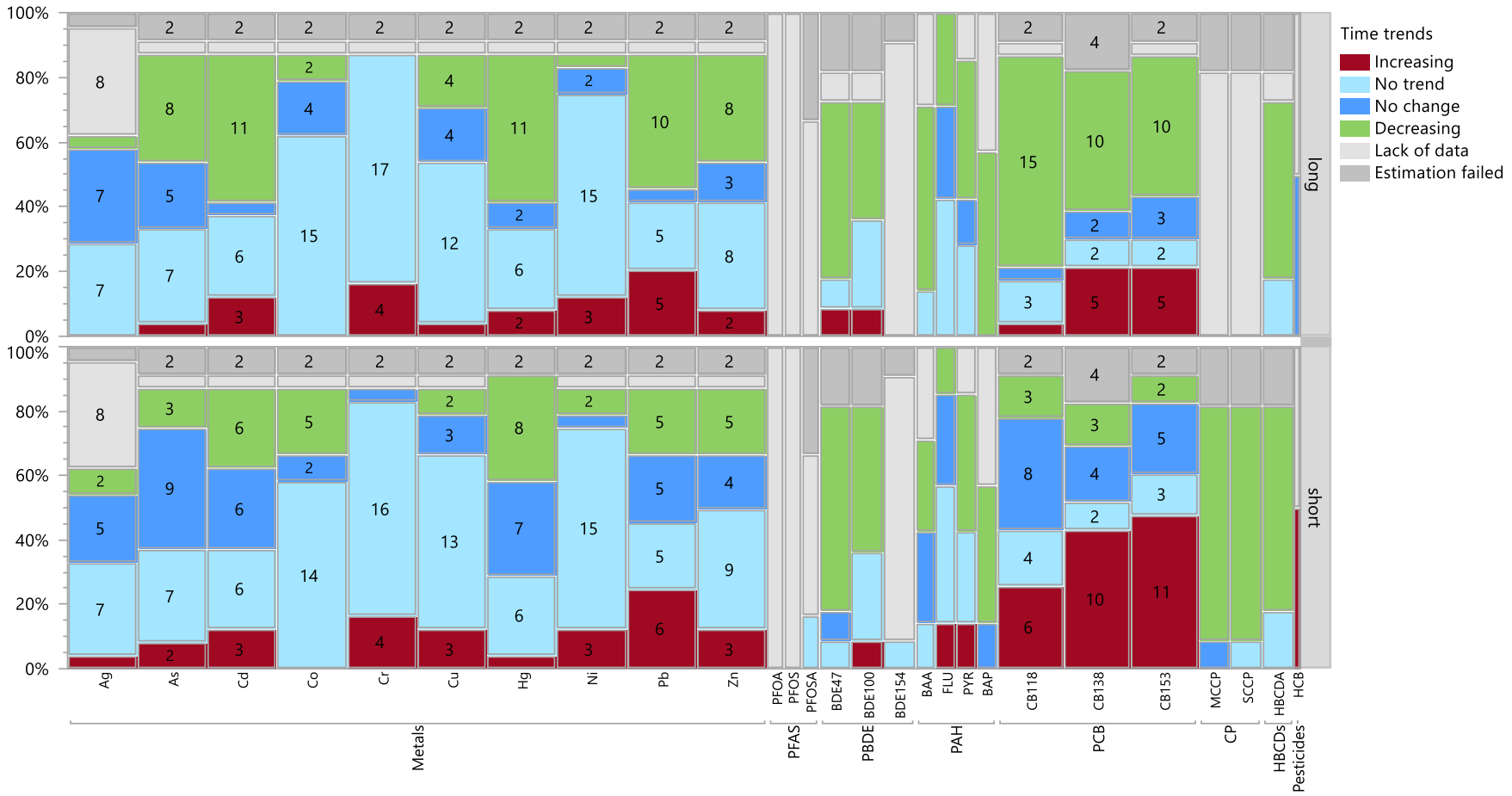


Figure 18. Time trends for blue mussel. Upper panel shows long-term trends, while lower panel shows short-term trends. The number of (stations × contaminants) are indicated in the respective cells.

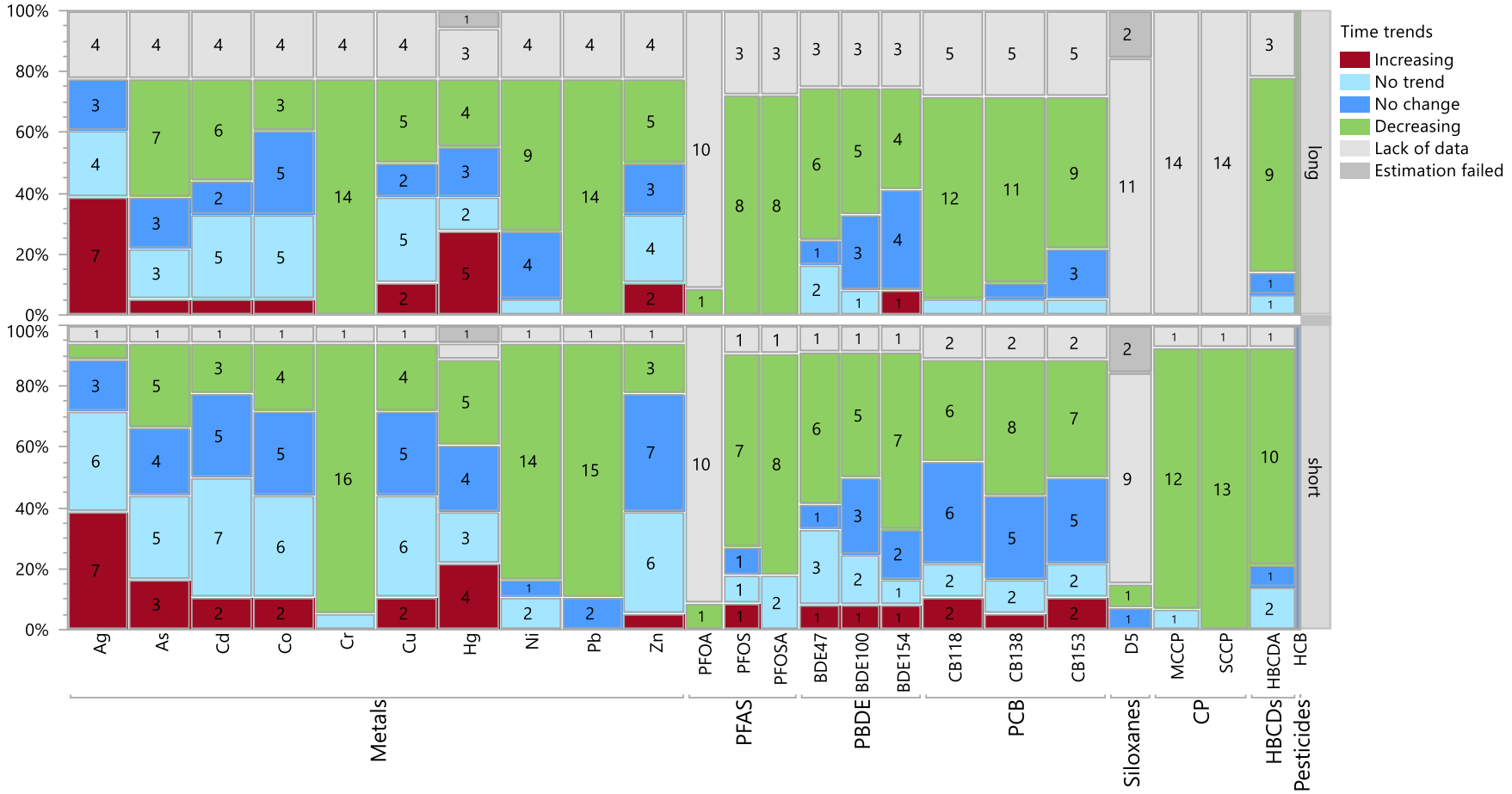


Figure 19. Time trends for cod. Upper panel shows long-term trends, while lower panel shows short-term trends. The number of (stations × contaminants) are indicated in the respective cells.

3.3.2 Heatmaps for stations

The increasing long- and short-term trends for metals in blue mussel (**Figure 20**) were mostly found at stations in the Oslofjord (Solbergstrand (31A), Gressholmen (30A), Akershuskaia (I301), Gåsøya (I304), and Færder (36A)).

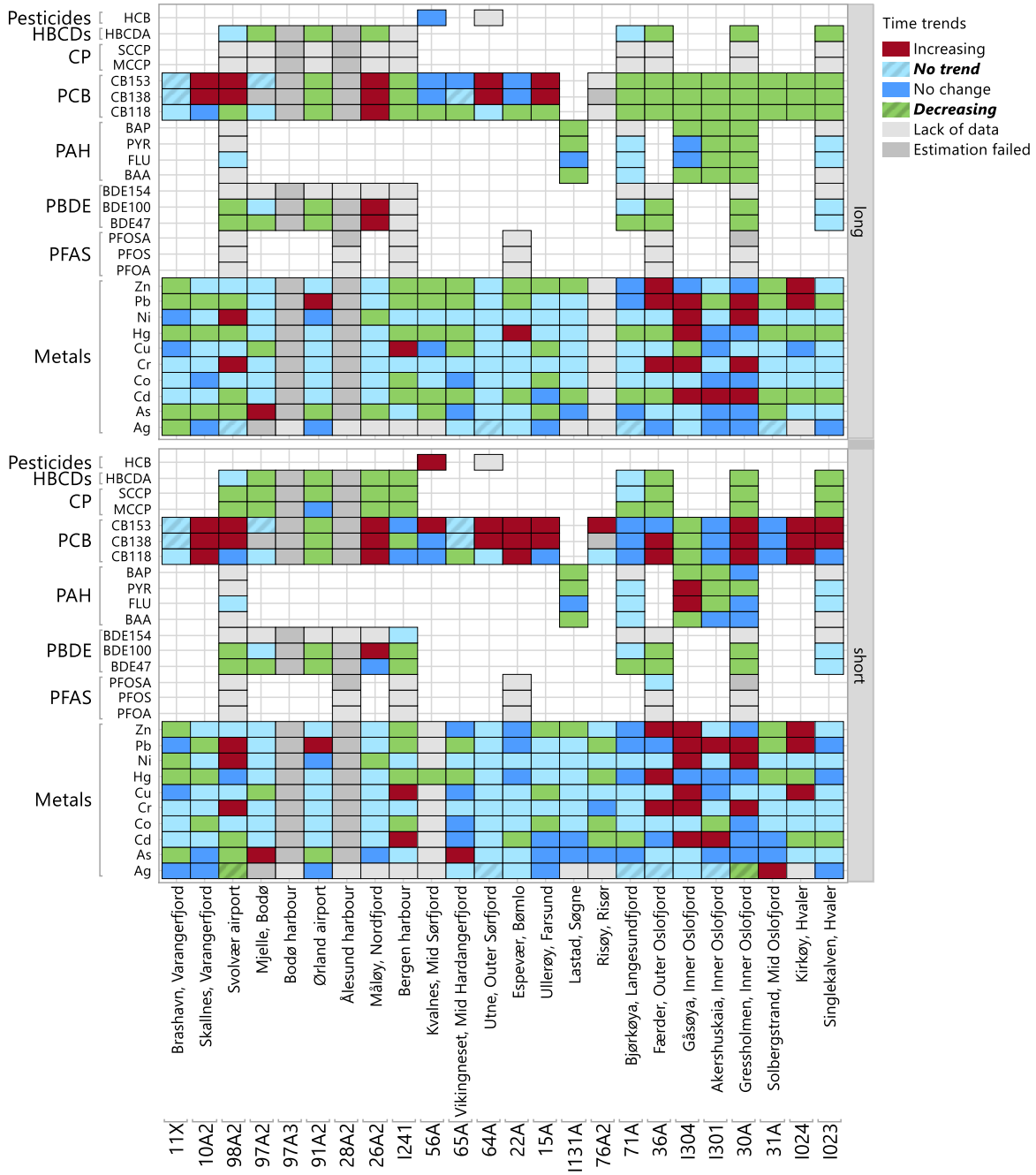


Figure 20. Heatmap of long-term and short-term time trends in blue mussel. The colours represent time trends observed at stations. Empty "cells" mean that the contaminant was not analysed. Grey lines show the midpoint of each station and contaminant. The stations are ordered along the coastline starting north moving south. Hatched cell pattern indicate that the original obtained trend was changed after manual QA.

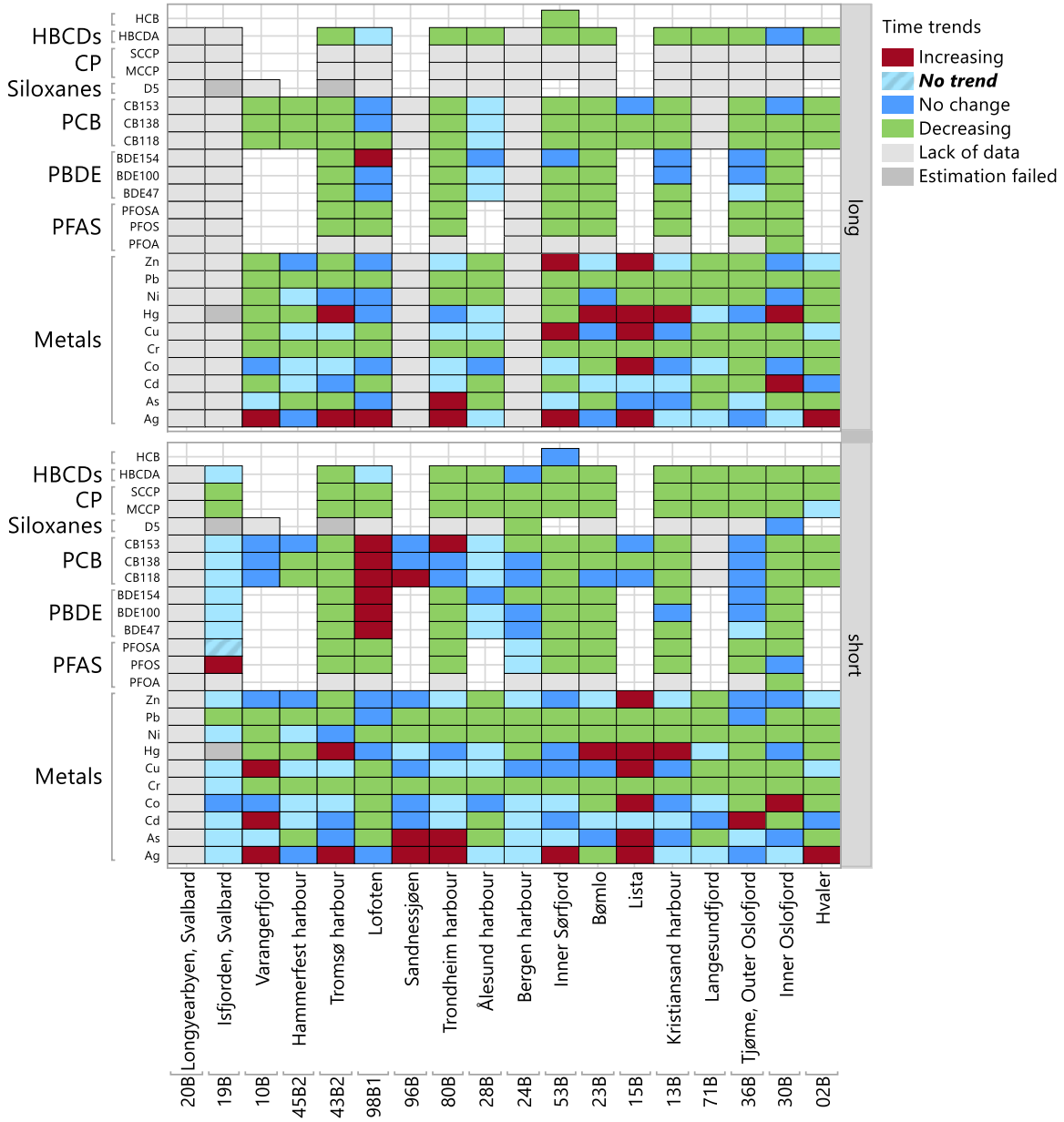


Figure 21. Heatmap of long-term trends in cod. The colours represent time trends observed at stations. Empty "cells" mean that the contaminant was not analysed. Grey lines show the midpoint of each station and contaminant. For mercury, cod have been length adjusted except for station 20B. The stations are ordered along the coastline starting north moving south. Hatched cell pattern indicate that the original obtained trend was changed after manual QA.

Also in cod, increasing long- and short-term trends for metals were dominating (**Figure 21**). Increasing short-term trends for PCB and PBDE were found at Lofoten (98B). For the first time, time trends at Svalbard (19B) could be estimated, and increasing short-term trend was found for PFOS.

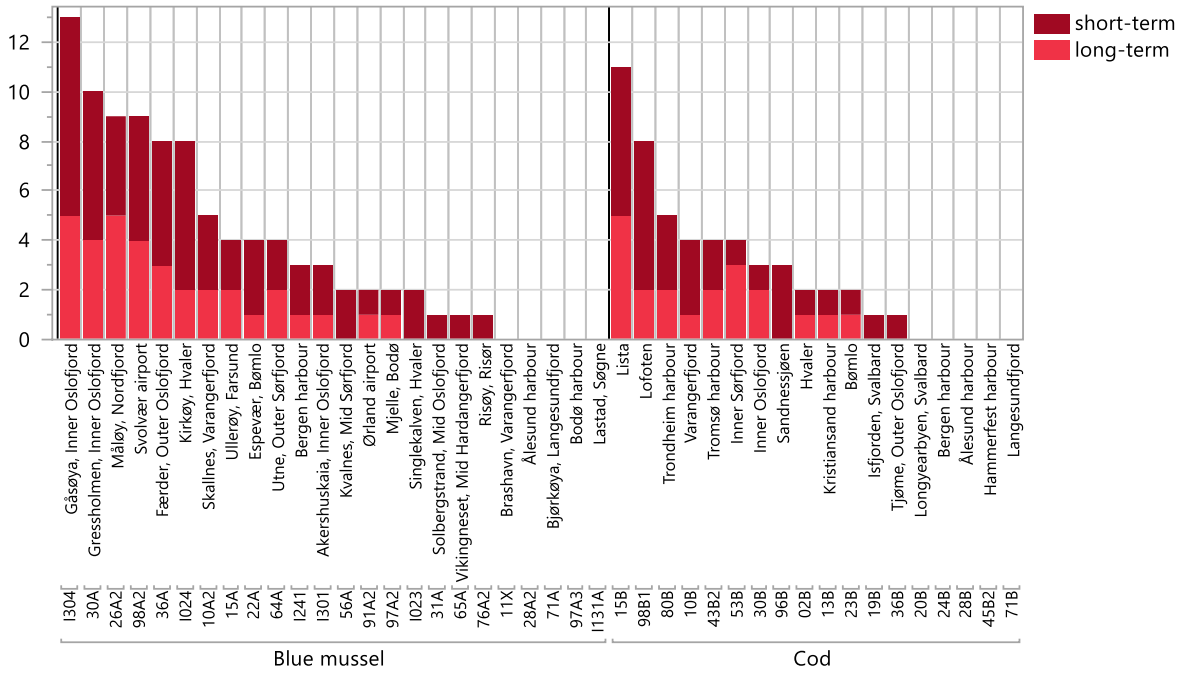


Figure 22. Number of increasing time trends for blue mussel and cod stations. Short-term trends and long-term trends are shown in different red colours.

The number of increasing time trends are shown in **Figure 22**. For blue mussel, the highest number of increasing trends were found in the Inner Oslofjord (I304 and 30A). For cod, the highest numbers were found at Lista (15B) and Lofoten (98B1).

3.3.3 Increasing time trends for stations that exceeded EQS and/or PROREF

Special attention needs to be paid for stations and contaminants with increasing trends which at the same time are exceeding EQS. This was the case for 10 combinations of contaminants, stations and species which are listed in **Table 6**. Note that EQS for sumPCB7 and sumBDE6 was exceeded in all cod stations. Also, the highest exceedances of PROREF were investigated, and for blue mussel we included exceedance higher or equal to 5xPROREF, while in cod higher than or equal to 2xPROREF (**Table 6**). The figures of the corresponding trends and relation to EQS are shown in **Figure 23** to **Figure 26**. Significant trends have a statement in the upper right corner of the figure indicating the annual percent change pr. year.

Table 6. Overview of contaminants with increasing trends and exceedance of EQS (RQ) and/or PROREF (>5 for mussel and >2 for cod). RQ and exceedance of PROREF are given with two significant digits.

Species	Contaminant	Station	Station name	RQ (concentration/EQS)	PROREFratio (concentration/ PROREF)	Shown in Figure
Cod	**sumPBDE6**	98B1	Lofoten	660		**Figure 23**
	sumPCB7	98B1	Lofoten	210		
	Hg	13B	Kristiansand harbour	1.6		**Figure 24**
		15B	Lista	6.5	2.3	
		23B	Bømlo	6.0	2.1	
		30B	Inner Oslofjord	7.0	2.5	
		43B2	Tromsø harbour	2.0		
Blue mussel	**sumPBDE6**	26A2	Måløy, Nordfjord	12		**Figure 25**
	sumPCB7	22A	Espevær, Bømlo	2.4		
		26A2	Måløy, Nordfjord	1.6		
	Pb	30A	Gressholmen, Inner Oslofjord		6.7	**Figure 26**
	PYR	I304	Gåsøya, Inner Oslofjord		8.0	
	CB118	30A	Gressholmen, Inner Oslofjord		9.7	
	CB138	30A	Gressholmen, Inner Oslofjord		6.0	
	CB153	30A	Gressholmen, Inner Oslofjord		5.8	

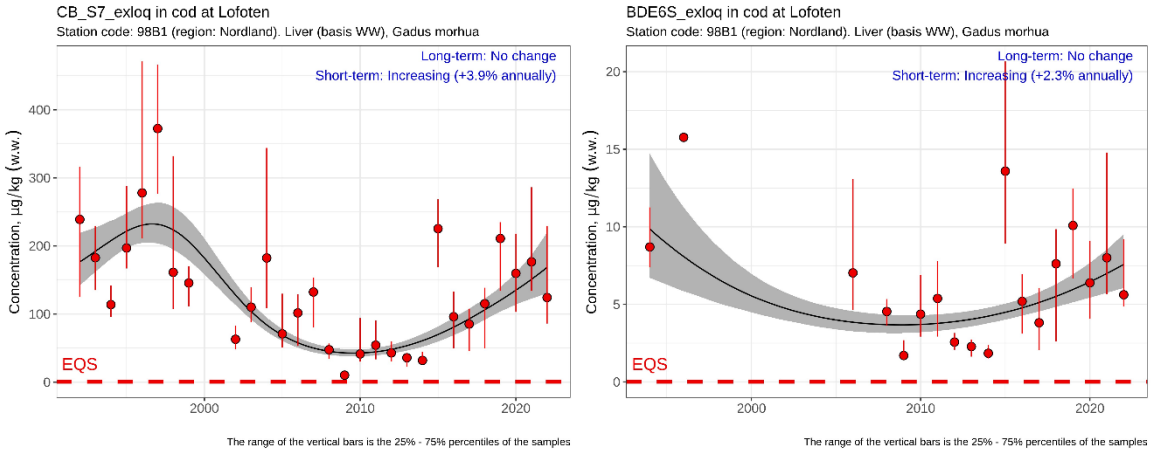


Figure 23. Cod station Lofoten (98B1) with increasing trends for sumPBDE6 and sumPCB7 and exceedance of EQS. Median concentrations are plotted against the year they were sampled and are shown as red circles, or triangles (where more than half of the data were below LOQ). For cod, the vertical red lines extending from the median concentrations indicate the percentile range (25%-75%), while for mussel they indicate the maximum and minimum concentrations. The model for the time trend is shown as a black line with the 95% confidence band in grey surrounding it. If applicable, the EQS is indicated with a red dashed line, while selected PROREF concentrations are indicated with dotted blue lines. In the upper right corner, the interpretations of the trends (long-term and short-term) are given with annual % change in parenthesis (if significant trend). Note that scales for the x axis and y axis can vary from figure to figure.

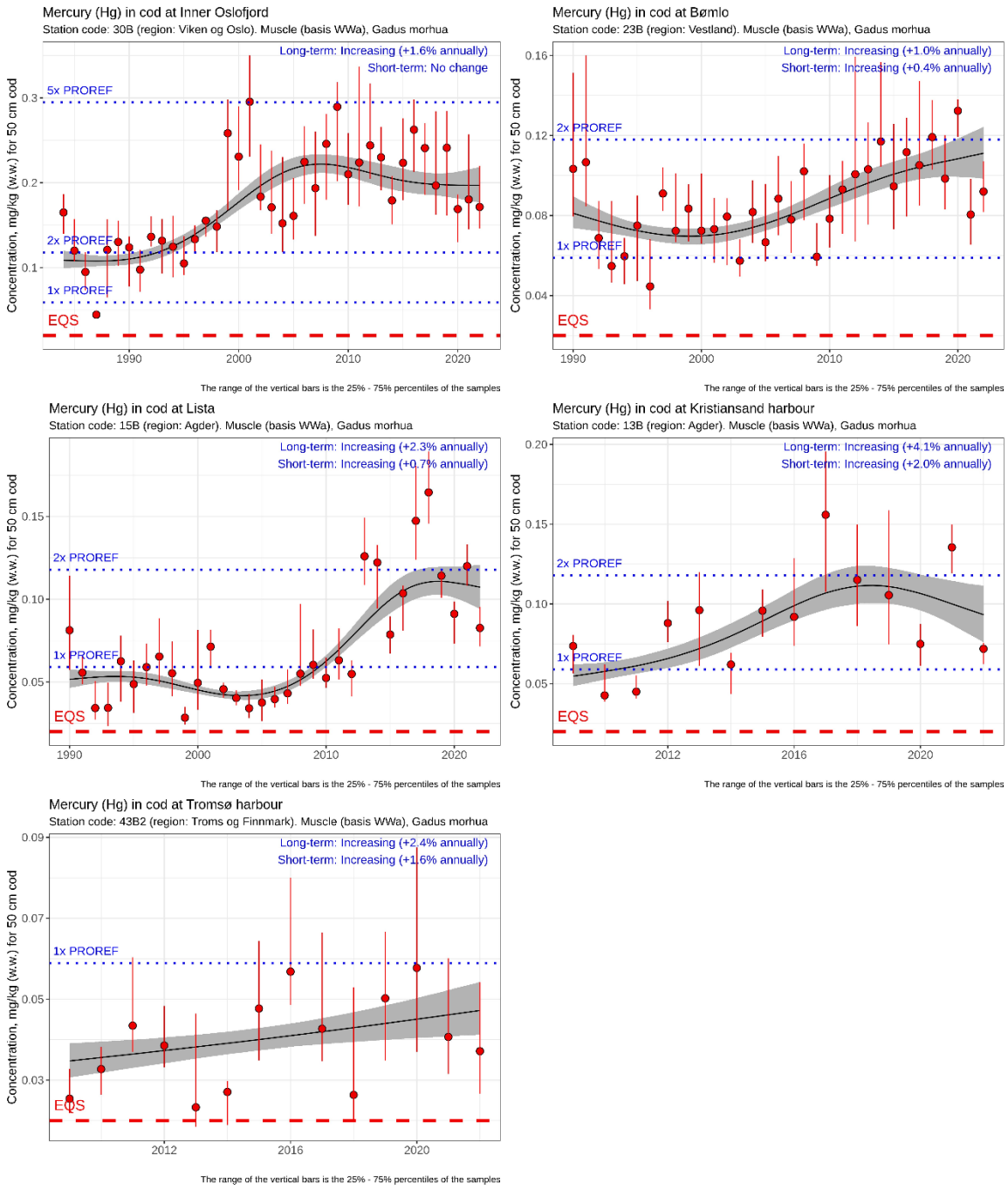


Figure 24. Cod stations in the Inner Oslofjord (30B), Bømlo (23B), Lista (15B), Kristiansand harbour (13B), and Tromsø harbour (43B2) with increasing trends for length adjusted mercury concentrations and for which the concentrations in cod are exceeding EQS and/or 2×PROREF or more.

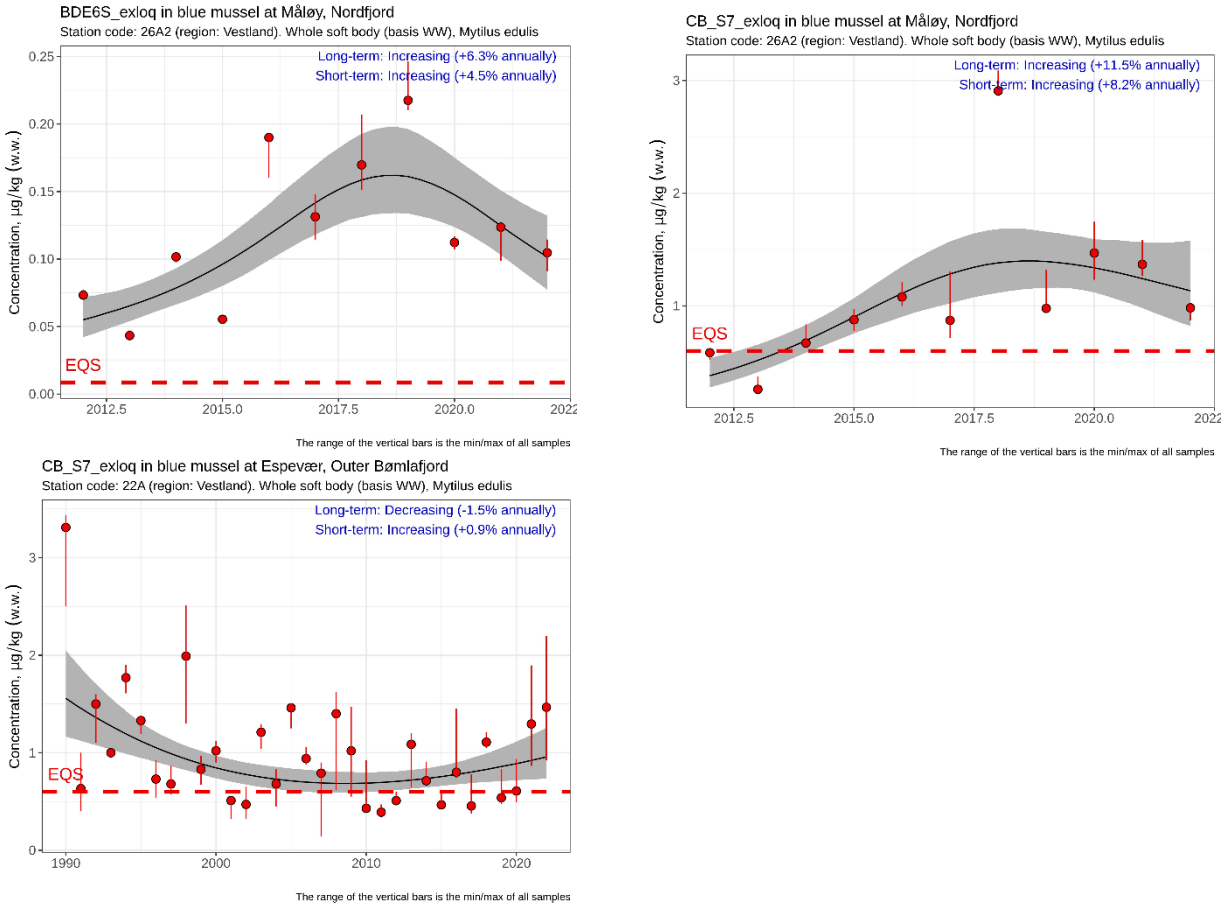


Figure 25. Mussel stations at Måløy in Nordfjord (26A2) and Espevær (22A) with increasing time trends for sumPBDE6 and sum PCB7 and exceedance of EQS.

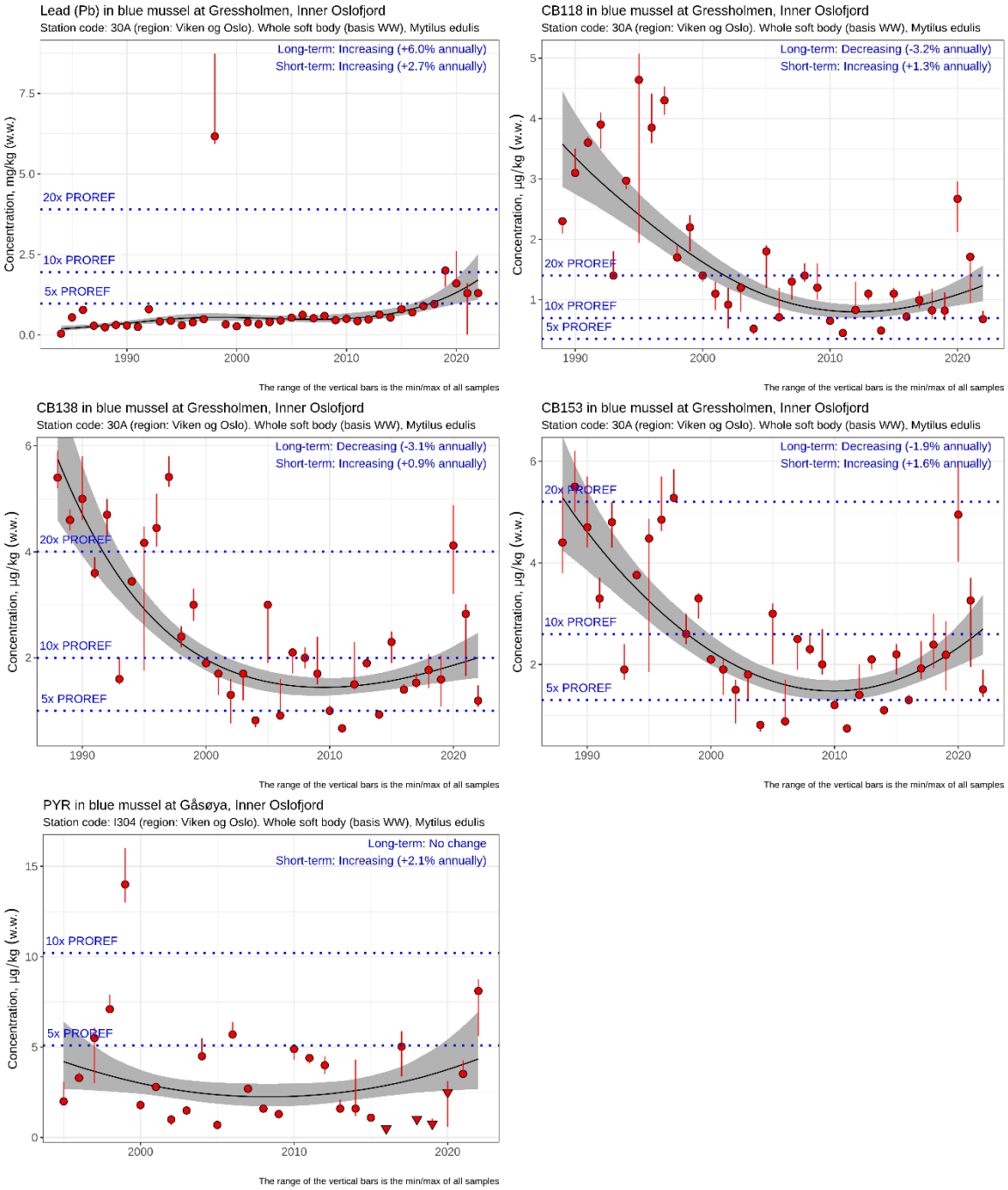


Figure 26. Mussel stations at Gressholmen (30A) and Gåsøya (I304) in the Inner Oslofjord showing increasing time trends and exceedance of 5×PROREF or more (lead, PCB 118, 138, 153, and pyrene).

4 Effect parameters, stable isotopes and cod at Svalbard

4.1 Biological effect parameters

4.1.1 Dogwhelk and common periwinkle

Tributyltin (TBT) and imposex/intersex

Tributyltin (TBT) is an organic compound of tin that was used as a biocide especially in marine antifouling paints until 2008, when it was banned globally. TBT is toxic to marine life and was first known to be used in the 1960s. Masculinized female marine snails was first described in the late sixties (Blaber, 1970). TBT induces male sex characters onto females, such as imposex in dogwhelk and intersex in common periwinkle. In female dogwhelk, the TBT effect causes a vas deference and a pseudopenis that are superimposed onto female genital structures. Sterility and even death of individuals occur in the most advanced stages. In female common periwinkle, the TBT effect causes a pathological alteration in the oviduct, development of spermatocytes in ovary or oocytes in the testis and/or penis. Sterility occurs in the most advanced stages. Common periwinkle is less sensitive to TBT than dogwhelk and may act as an alternative sentinel when dogwhelk is not found. In the present study, TBT was analysed in dogwhelk at eight stations and common periwinkle at one station (Fugløyskjær in Langesund, 71G). Imposex (Vas Deferens Sequence Index, VDSI) was investigated in dogwhelk and intersex (Intersex Stage Index, ISI) in common periwinkle.

EQS

When applying the EQS for TBT (150 µg/kg ww, **Table 3**) in biota ("for fish") on dogwhelk (< 3.2 µg/kg ww) and common periwinkle (1 µg/kg ww), all TBT-concentrations were below EQS. When applying the EQS for triphenyltin (TPhT) (150 µg/kg ww, **Table 3**) in biota on dogwhelk (<1.4 µg/kg ww) and common periwinkle (<0.49 µg/kg ww), all TPhT-concentrations were below EQS.

Time trends of TBT

There were significant decreasing long-term trends for TBT in dogwhelk at Færder (36G) in the Outer Oslofjord, Risøya (76G) at Risør, Ullerøy (15G) in Farsund, Espevær (22G) by Bømlo, Svolvær (98G) in Lofoten, and Brashavn (11G) in the Varangerfjord (**Figure S15**). There were significant decreasing short-term trends for TBT in dogwhelk at Espevær (22G) and Brashavn (11G).

Biological effects of TBT (imposex/VDSI) in dogwhelk

The effects of TBT measured by the imposex parameter VDSI were zero at all eight stations. All results were below the OSPARs Background Assessment Criteria (BAC=0.3) (OSPAR, 2008) and the OSPARs Ecotoxicological Assessment Criteria (EAC=2) (OSPAR, 2013a, 2013b).

Time trends of VDSI

In dogwhelk, both significant decreasing long- and short-term trends for VDSI were observed in the Mid Karmsund (227G) (**Figure 27**) and at Svolvær airport (98G) in Lofoten (**Figure S15**). Significant decreasing long-term trends were found at Færder (36G) in the Outer Oslofjord (**Figure 27**), Risøya (76G) at Risør, Lastad (131G) at Søgne, Ullerøy (15G) in Farsund, and at Espevær (22G) by Bømlo.

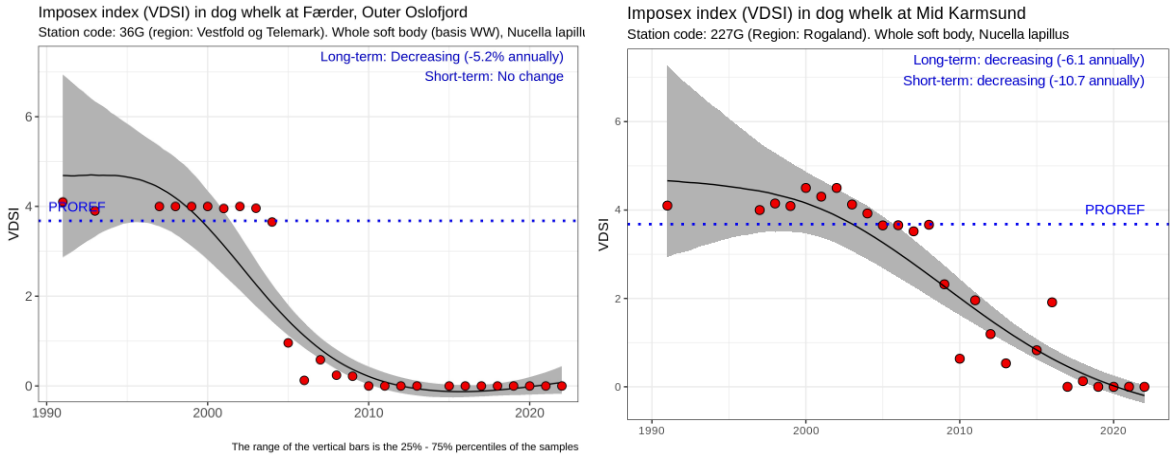


Figure 27. VDSI from 1991 to 2022 for dogwhelk from Færder (36G) in the Outer Oslofjord (left) and in the Mid Karmsund (227G) (right). For full explanation of figure see example in **Figure 23**.

Selected time trends

Two time trends for VDSI in dogwhelk are shown in **Figure 27**. VDSI in dogwhelk at Færder (36G) showed decreasing long-term trend (5.2% annually). In the Mid Karmsund (227G), there were both decreasing long- and short-term trends (6.1% and 10.7% annually).

The 2022 data confirmed the results since 2017 of no effects of TBT on dogwhelk (VDSI=0) (Schøyen et al., 2019).

Biological effects of TBT (intersex/ISI) in common periwinkle

The effect of TBT in common periwinkle, ISI, was zero at Fugløyskjær (71G) in the Langesundfjord (see **Figure S15**). ISI in common periwinkle is too sensitive for application of BAC and EAC (OSPAR, 2013a).

Time trends of ISI

The data of ISI in common periwinkle at Fugløyskjær (71G) showed a significant decreasing long-term trend (7.6% annually) (**Figure 28**, see **Figure S15**).

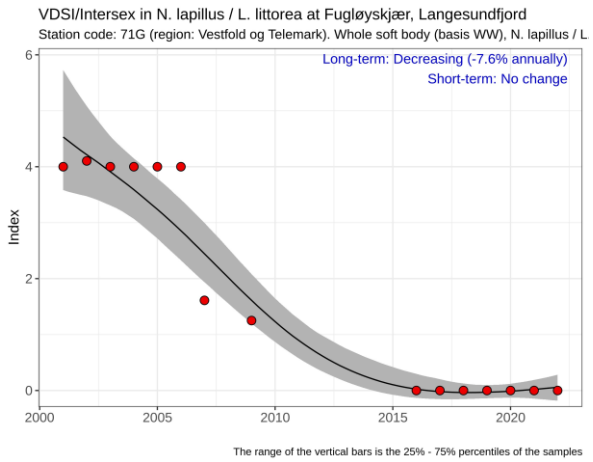


Figure 28. ISI from 2001 to 2022 for common periwinkle from Fugløyskjær in the Langesundfjord (71G). For full explanation of figure see example in **Figure 23**.

4.1.2 Cod

Biological effect methods (BEM) are included in the monitoring programme to assess the potential pollution effects on organisms. This can hardly be done solely on the basis of tissue concentrations of chemicals. There are three BEM methods applied on cod (including analyses of degradation products of PAH in bile). Each method is in theory specific for individual or groups of chemicals. One of the advantages of these methods used at the individual level is the ability to integrate biological and chemical endpoints, since both approaches are performed on the same individuals. The results can be interpreted in relation to established reference values (OSPAR, 2013a).

OH-pyrene metabolites in bile

Analysis of OH-pyrene in bile is not a measurement of biological effects, *per se*. It is included here, however, since it is a result of biological transformation (biotransformation) of PAHs and is thus a marker of exposure.

In 2022 the median (non-normalized [5]) concentration of OH-pyrene metabolites in bile from cod was above the ICES/OSPAR assessment criterion (background assessment criteria, BAC) at all stations monitored (Oslofjord, 30B, Sørfjord, 53B, and Lista, 15B), except at Bømlo (23B, reference station). Among the four stations, OH-pyrene concentrations were highest in the Inner Oslofjord (30B) and Sørfjord (53B), where concentrations were significantly different from those at Lista (15B) and Bømlo (23B; Tukey-Kramer HSD). The largest variation in OH-pyrene concentrations was observed at Lista (15B).

ALA-D in blood cells

Inhibited activity of ALA-D indicates exposure to lead. Although ALA-D inhibition is lead-specific, it is not possible to rule out interference by other metals or organic contaminants.

The median ALA-D activity in cod at the reference station (Bømlo; 23B) in 2022 appeared similar as in 2021, and thus as most previous years (since 2013). The median activity in the Inner Oslofjord (30B) in 2022 appeared slightly lower than at Bømlo (23B; reference station), however, this was not statistically significant (Tukey-Kramer HSD). In the Inner Sørfjord (53B), the median ALA-D activity was significantly lower than at the reference station (23B; Tukey-Kramer HSD). The largest variation in ALA-D activity was also observed in the Inner Sørfjord. Earlier frequent lower activities of ALA-D in cod from the Inner Sørfjord (53B), as well as the Inner Oslofjord (30B), have been attributed to lead contamination. Higher concentrations of lead in cod liver have generally been observed in the Inner Oslofjord and Inner Sørfjord, compared to Bømlo, though with a relatively large individual variation, as was also the case in 2022.

EROD activity

High activity of hepatic cytochrome P450 1A activity (EROD activity) normally occurs as a response to planar compounds such as certain PCBs, PCNs (polychlorinated naphthalenes), PAHs, or dioxins. In 2022, the median EROD activity appeared lower at Bømlo (23B, reference station), than in the Inner Oslofjord (30B) and Inner Sørfjord (53B), however this was not statistically significant (Tukey-Kramer HSD). Median EROD activities were below the ICES/OSPAR assessment criterion (background assessment criteria, BAC), at all stations.

[5] Not normalized to absorbance at 380 nm

4.2 Analysis of stable isotopes

Stable isotopes of carbon and nitrogen are useful indicators of food origin and trophic levels. $\delta^{13}C$ gives an indication of carbon source in the diet of a food web. For instance, it is in principle possible to detect differences in the importance of autochthonous (native marine) and allochthonous (watershed/origin on land) carbon sources in the food web, since the $\delta^{13}C$ signature of the land-based energy sources is lower (greater negative number) than the autochthonous. Also $\delta^{15}N$ (although to a lesser extent than $\delta^{13}C$) may be lower in allochthonous as compared to autochthonous organic matter (Helland et al., 2002), but more important, it increases in organisms with higher trophic level because of a greater retention of the heavier isotope (^{15}N). The relative increase of ^{15}N over ^{14}N ($\delta^{15}N$) is 3-5 ‰ per trophic level (Layman et al., 2012; Post, 2002). It thus offers a continuous descriptor of trophic position. As such, it is also the basis for Trophic Magnification Factors (TMFs). TMFs give the factor of increase in concentrations of contaminants per trophic level. If the concentration increase per trophic level can be expressed as:

Log Concentration = $a + b * (\text{Trophic Level})$

Then:

$TMF = 10^b$

TMFs has recently been amended to Annex XIII of the European Community Regulation on chemicals and their safe use (REACH) for possible use in weight of evidence assessments of the bioaccumulative potential of chemicals as contaminants of concern.

The results of the stable isotope analysis in 2022 generally show the same pattern as observed in previous years i.e., a continual geographical pattern, indicating a spatial trend persistent in time (**Figure 29**).

As previously, cod from the Sørfjord (53B) and Bergen harbour (24B; both in Vestland County) stand out with particularly low $\delta^{15}N$ signature (**Figure 29**). The same is shown for mussels from the Sørfjord (56A) and Bergen harbour (I241), indicating that the $\delta^{15}N$ baseline of the food web in these parts of Norway is lower. Likewise, isotope signatures of both cod (30B) and mussels (stations 30A and I304) are among the highest observed (**Figure 29**) indicating a high baseline.

The isotopic signatures in cod from Svalbard appear similar at the two stations (19B and 20B; **Figure 29**).

In 2019, the $\delta^{15}N$ data from the whole Norwegian coast were scrutinized further by deducing the trophic position of cod, based on a known baseline in the same area, given by the isotopic profile in blue mussel, inhabiting trophic position 2 (primary consumer, feeding on particulate matter; (Schøyen et al., 2021). This study showed that baseline adjusted trophic position of cod differed between stations along the Norwegian coast, suggesting that parts of the spatial differences in cod contaminant concentrations may be attributed to different trophic positions of the cod at the different stations, and not merely differences in environmental concentrations between stations.

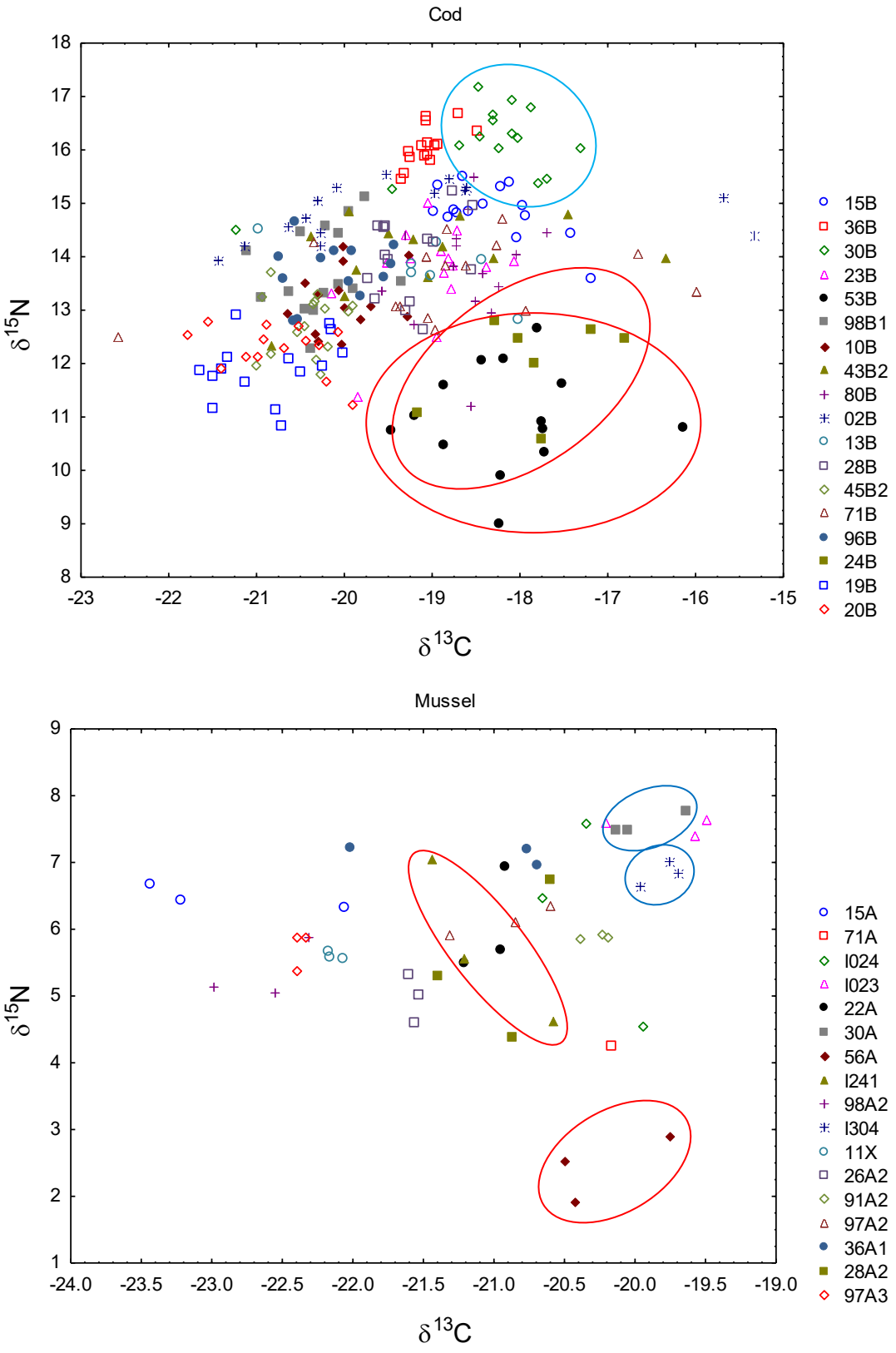


Figure 29. $\delta^{13}C$ plotted against $\delta^{15}N$ for cod and blue mussel. Blue ellipses indicate the position of the samples of cod and blue mussel from the Inner Oslofjord, while red ellipses indicate the position of the samples of cod and blue mussel from the Sørfjord and Bergen harbour.

δ^{15}N values in eiders from Svalbard (blood and egg) resembled those previously observed (Schøyen et al., 2021). The δ^{13}C values in the eiders differed between the two matrices (blood and egg; **Figure 30**), likely related to different lipid content, as lipids are δ^{13}C-depleted relative to proteins (Sweeting et al., 2006). Samples were not treated to remove carbonates or lipid prior to stable isotope analysis.

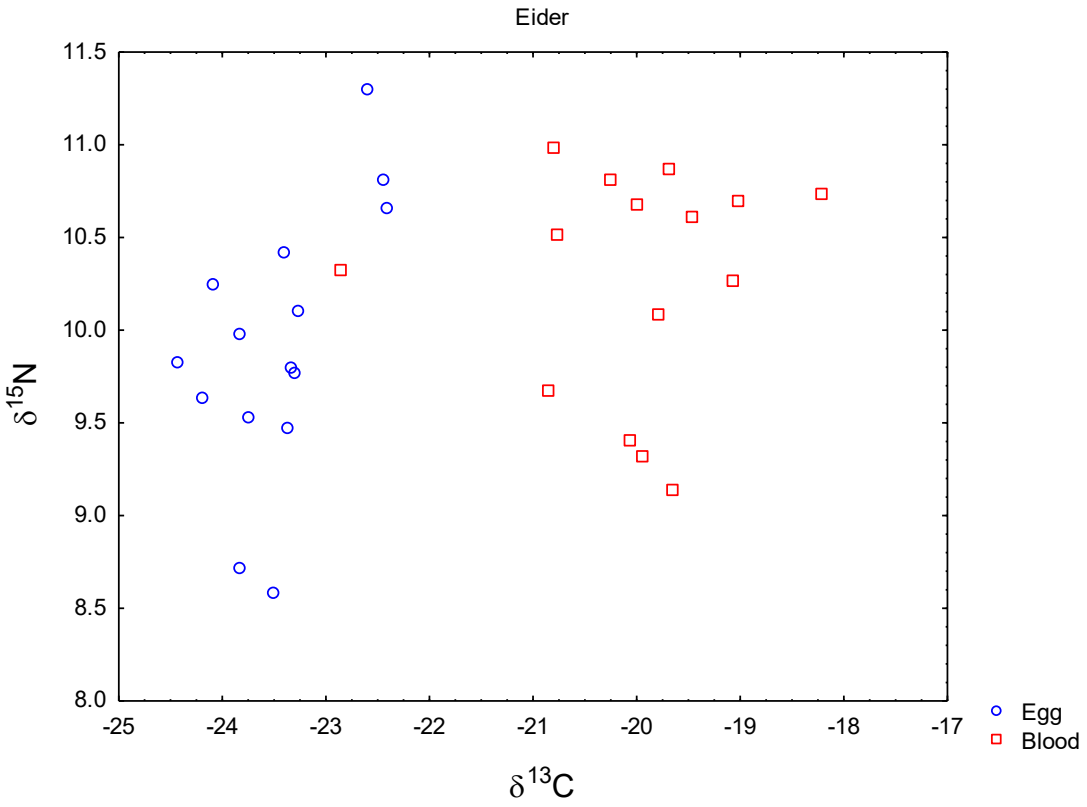


Figure 30. δ^{13}C plotted against δ^{15}N in blood (red squares) and egg (blue circles) of eider from Svalbard (19N).

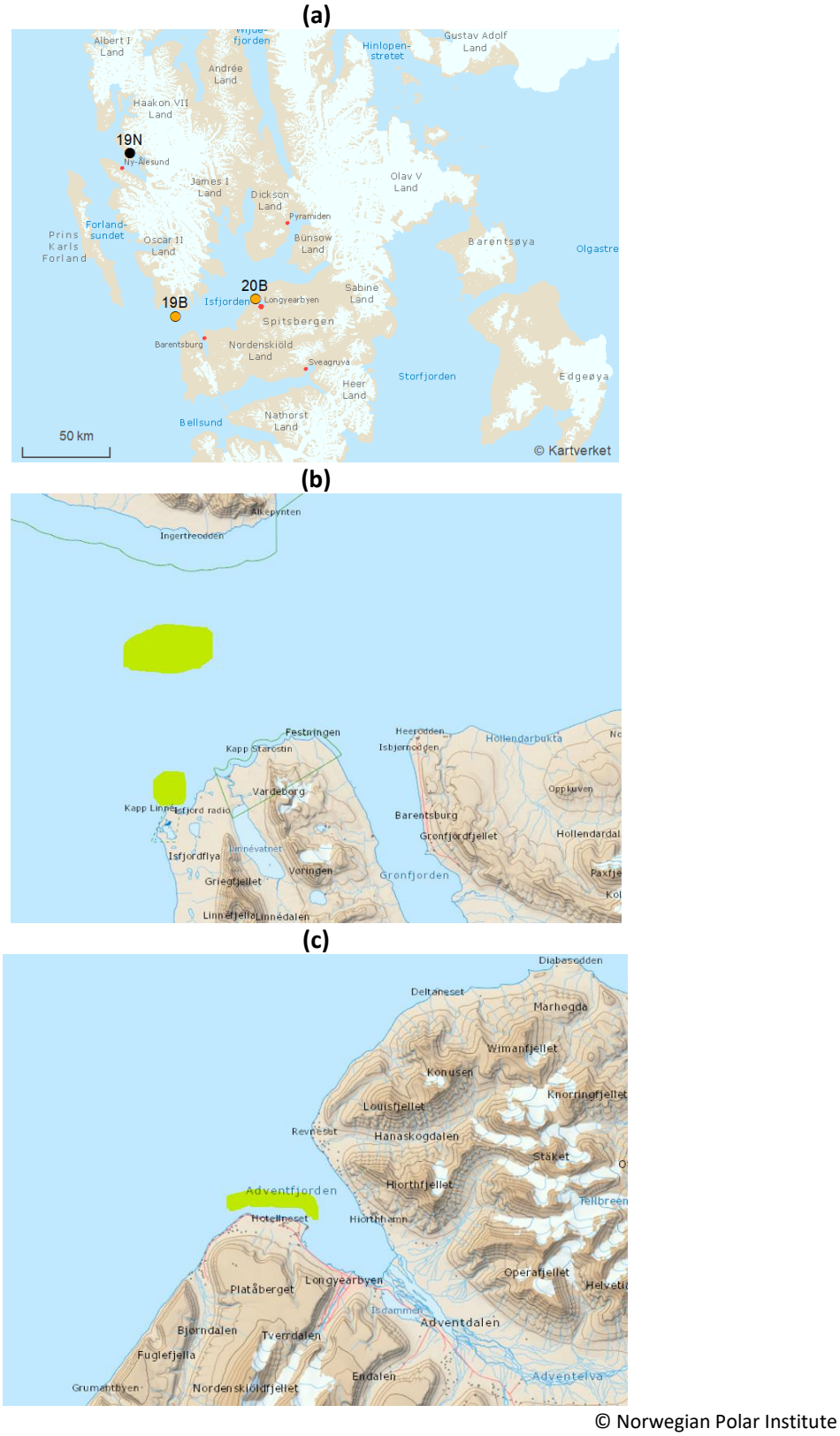
4.3 Cod at Svalbard

In the Barents Sea, there are two types of cod; coastal cod (CC) and North-East Arctic cod (NEAC)[6]. Coastal cod can be found from kelp belt down to a depth of 500 meters. It spawns deep in the fjords, but also in the same areas as North-East Arctic cod. Coastal cod reaches sexual maturity earlier, grows faster and migrates to lesser extent than North-East Arctic cod[7]. North-East Arctic cod lives most of its life in the Barents Sea, makes extensive migrations, and spawns mainly in Lofoten and Vesterålen. Cod caught along the coast may in some cases be North-East Arctic cod. Spotowitz et al. (2022) found that both North-East Arctic cod and coastal cod appear in Svalbard fjords, and revealed that 0-group and adult coastal cod differ genetically from those along the Norwegian coast, indicating a separation into a local Svalbard coastal cod population. Genetic analysis of cod collected at Svalbard in the MILKYS programme have not been done, neither a thorough study of cod otoliths regarding fish stock population. According to local people, fishing for cod is better in late summer (August/September) and further into autumn and there is little cod that are caught during spring and early summer. This indicates that the cod are migrating within the fjord system.

Cod from the Outer Isfjord (19B)
In 2017, cod from the Outer Isfjord (19B) was included in the programme (Green et al., 2018) (**Figure 31**). The station is about 13 km west of Barentsburg and about 50 km southwest of Longyearbyen. In 2022, the cod were sampled with a fishing rod at 50 m depth outside Kapp Linné in the Outer Isfjord. The cod was also caught by rod in 2020 (90 m) and 2018 (90 m). The cod was collected by trawl in 2021 (250 m), 2019 (150 m) and 2017 (260 m).

Cod from outside Longyearbyen (20B)
Investigations of cod outside Longyearbyen (20B) were included in 2021 (Schøyen et al., 2022). In 2022 and 2021, cod were sampled with a fishing rod at 15 to 80 m depth in the area near the airport outside Longyearbyen (**Figure 31**).

[6] https://miljostatus.miljodirektoratet.no/tema/hav-og-kyst/havindikatorer/barentshavet/fiskebestander/nordostarktisk-torsk-i-barentshavet/
[7] https://miljostatus.miljodirektoratet.no/tema/hav-og-kyst/havindikatorer/barentshavet/forurensende-stoffer/forurensning-i-torsk-i-barentshavet/



© Norwegian Polar Institute

Figure 31. Map of the two cod stations Isfjorden (19B) (a and b) and Longyearbyen (20B) (a and c), and the station for common eider in the Kongsfjord (19N) at Svalbard (a). The cod was sampled by a fishing rod and by trawl for different years at station Isfjorden (b), and by fishing rod at station Longyearbyen (c). The drain point from Longyearbyen to the Adventfjord is close to Hiorthavn (Moskushavn) at 50-60 m depth (c).

The cod lengths were significantly higher in the Isfjord (19B) than outside Longyearbyen (20B, both Tukey-Kramer HSD and Kruskal Wallis). The weights of both whole fish and gonads were significantly higher in the Isfjord (19B) than at outside Longyearbyen (20B) (Tukey-Kramer HSD).

There was no significant difference for fat content (%) for cod at the two stations (Tukey log-transformed data).

Contaminants

For selected contaminants, there were significant higher concentrations (Tukey-Kramer HSD log-transformed data) for cod from the Isfjord (19B, annual data from 2017 to 2022) than outside Longyearbyen (20B, data from 2021 and 2022) for mercury (not length adjusted), PFAS (PFOA, PFOS and PFOSA), PBDEs (BDE47, BDE100 and BDE154), PCBs (CB118, CB138 and CB153), siloxanes (D4, D5 and D6), CCPs (MCCP excl. LOQ), and HBCDs (HBCDA) (**Figure 32**). In **Figure 32** the univariate confidence limits are shown, which are slightly different from the confidence limits calculated by the Tukey-Kramer HSD test.

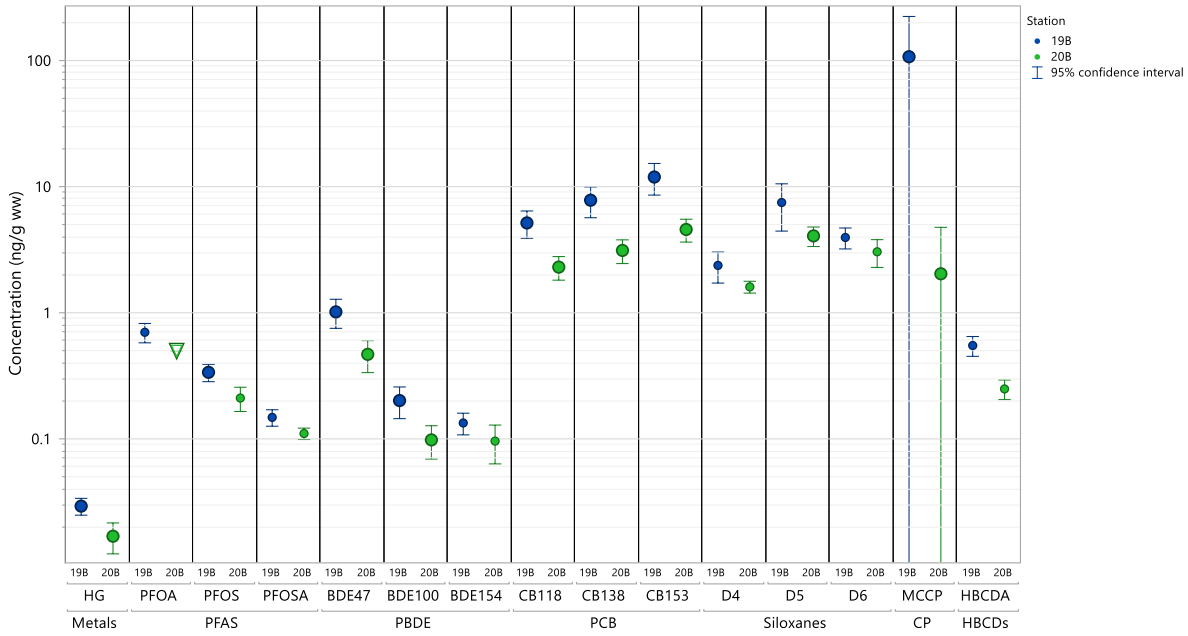


Figure 32. Significant higher concentrations of selected contaminants in cod from the Isfjord (19B) than outside Longyearbyen (20B). Mean concentrations for all fish sampled error bars representing 95% univariate confidence limits are shown. Data where all concentrations were below LOQ are indicated with a triangle.

The statistical tests used, only give an indication of differences since it has not been investigated that all assumptions for Tukey-Kramer HSD are present (e.g. no heteroscedasticity). In case of heterogeneity of variance, no non-parametric tests have been run when assumptions were not met.

Stable isotopes

There was no significant difference for $\delta^{13}C$ value in cod from the Isfjord (19B) and outside Longyearbyen (20B) (see also **Figure 29**). There were significantly higher $\delta^{15}N$ values in cod from the Isfjord (19B) than outside Longyearbyen (20B) (Tukey-Kramer HSD) (**Figure 33**).

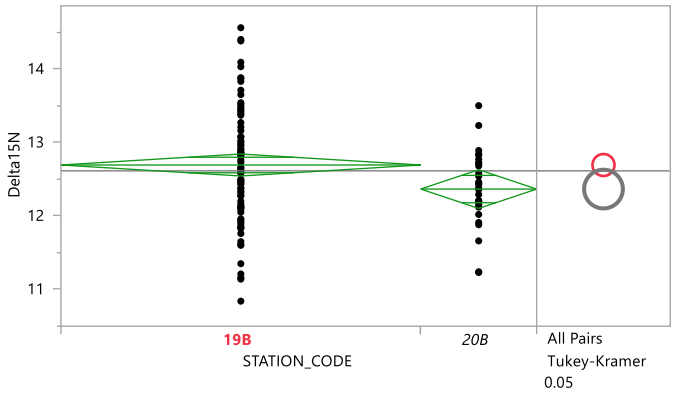


Figure 33. Tukey-Kramer HSD test indicate higher $\delta^{15}N$ values in cod from the Isfjord (19B) than outside Longyearbyen (20B).

Annual emission and discharges

There are many influencing factors such as long-range pollution through air and sea currents, local pollution from active and old sources, mining and coal power plant, settlements, wastewater from Longyearbyen into the Adventfjord, wastewater from Barentsburg into Grønfjorden, wastewater from Isfjord Radio at Kapp Linné, tourism and cruise traffic.

Longyearbyen

Wastewater from Longyearbyen is released uncleaned into the Adventfjord in a common waste pipe. Since 2008, all the sewage has been directed through a 3 km long pipeline on the north side of the Adventfjord, with a discharge point at Hiorthavn (Moskushavn) at 60 m water depth. With some local exceptions, no high levels of contaminants have been found in the recipient other than can be expected in the vicinity of coal activities like landfill, coal etc. (Cochrane and Evenset, 2020).

PFAS sources in Longyearbyen are firefighting foam used at the airport, WWTP, urban run off and landfill leachate [8]. For many years, PFOS and PFAS were used as firefighting foam at a now closed fire training field at Svalbard airport[9]. There are no reported annual emissions of PFOS to air and discharges to water from the airport at Svalbard at www.norskeutslipp.no from 1994 to 2022. Ali et al. (2021) found that the firefighting training stations (FFTS) at Svalbard airport and diffuse release from the local settlement were the major local PFAS sources. PFAS has been detected in several species of fish in this area other than cod. In 2018, Norconsult estimated an annual discharge to seawater of 5-7 g PFOS/year and 26 g sum PFAS/year spread from active fire training field via a discharge line from the airport terminal area (AVINOR, n.d.).

Outer Isfjord

Contamination in soil is suspected or proven at areas close to Kapp Linné and Isfjord Radio in the database for soil pollution (Grunnforurensning (miljodirektoratet.no). Cadmium, lead, zinc, PAH16 and organochlorine compounds are mentioned.

The ground in Barentsburg is contaminated by PCB, DDT, HCB, metal compounds and mineral oil/aliphatic, and is mainly due to the historical activities (Norconsult, 2017). Norconsult points out that investigations of PFAS will be relevant at localities where fire extinguishing agents have been used (e.g. helicopter base Kapp Heer close to Barentsburg) (Norconsult, 2017).

[8] NGI - Svalbard
[9] Svalbard Lufthavn: Må rydde opp PFAS-forurensning - Miljødirektoratet (miljodirektoratet.no)

It was somewhat unexpected that cod from the Outer Isfjord (19B) were significantly bigger (i.e. higher length, weight and gonad weight), and had higher concentrations of mercury, PFAS, PBDEs, PCBs, siloxanes, CCPs and HBCDs, and higher $\delta^{15}N$ values than cod from outside Longyearbyen (20B).

Recommendations

More research should be done for the 2024 survey at both cod stations in the Outer Isfjord (19B) and outside Longyearbyen (20B) in the Adventfjord to be able to determine which type of cod that is present, the coastal cod (CC) or/and North-East Arctic cod (NEAC). To be able to do that, samples for genetic analyses and otoliths study regarding fish stock population should be sampled and analysed. We assume that the cod station outside Longyearbyen (20B) are influenced by contaminants from the airport and settlement. Cod is now caught 3-7 km from the discharge point at Moskushavn, and the cod can be caught even closer. Fishing should be done at both stations in the same way by using fishing rod and not trawl (in the Outer Isfjord, 19B), within as short period of time to be comparable.

5 Materials and methods appendix

5.1 Sampling and matrices

5.1.1 Stations

Samples for the investigation of contaminants were collected along the Norwegian coast, from the Swedish border in the south and to the Russian border in the north, as well as Svalbard (**Figure 1**). The sampling involved blue mussel at 24 stations, dogwhelk at eight stations, common periwinkle at one station, cod at 18 stations, and the common eider at one station (**Table 1**).

Samples were collected during 2022 (early 2023 for some cod) and analysed according to OSPAR guidelines (OSPAR, 2021)[10] where these could be applied. The data was screened and submitted to ICES by agreed procedures (ICES, 1996) as well as to the national database Vannmiljø. Blue mussel (*Mytilus edulis*), dogwhelk (*Nucella lapillus*), common periwinkle (*Littorina littorea*) and Atlantic cod (*Gadus morhua*) are the target species selected for MILKYS to indicate the degree of contamination in the sea. Blue mussel is attached to shallow-water surfaces, thus reflecting exposure at a fixed point (local pollution). Mussels and snails are usually abundant, robust, and widely monitored in a comparable way. The species are, however, restricted to the shallow waters of the shoreline. Cod is widely distributed and commercially important fish species. It is a predator and, as such, will for hydrophobic compounds mainly reflect contamination levels in their prey. Recently, however, it has become increasingly difficult to catch sufficient numbers of adequate size of both blue mussel and cod. The 2022 programme also included investigation of contaminants in the common eider (*Somateria mollissima*).

Some details on methods applied in previous years of monitoring are provided in earlier reports (Green et al., 2008; Schøyen et al., 2022).

5.1.2 Blue mussel

Blue mussel has been proven as a promising indicator organism for contaminants (Beyer et al., 2017). In general, blue mussel is widely used for monitoring in controlled field studies (Schøyen et al., 2017).

A sufficient number of individuals for three pooled samples of blue mussel were found at nearly all the 24 stations (**Table 1**, **Figure 34**). The stations were chosen to represent highly polluted, or reference stations distributed along the Norwegian coast. It has been shown that the collected individuals are not all necessarily *Mytilus edulis* (Brooks and Farmen, 2013), but may be other *Mytilus* species (*M. trossulus* and *M. galloprovincialis*). Possible differences in contaminant uptake between *Mytilus* species were assumed to be small and they were not taken into account in the interpretations of the results for this investigation.

[10] See also http://www.ospar.org/work-areas/hasec

Figure 34. Blue mussel (photo: Janne Gitmark, NIVA).

The blue mussel samples were collected from 8th August to 18th October 2022. This is within the OSPAR guidelines and considered to be outside the mainly mussel spawning season.

Generally, blue mussel was not abundant on the exposed coastline from Lista (southern Norway) to the north of Norway. The mussel was more abundant in more protected areas and were collected from dock areas, buoys, or anchor lines. All blue mussels were collected by NIVA, except for some blue mussel stations collected by local contacts.

The method for collecting and preparing blue mussel was based on the National Standard for mussel collection (NS, 2017) Three pooled samples of approximately 50 individuals (size range of 3-5 cm) were collected at each station and kept frozen until later treatment. Shell length was measured by slide callipers. The blue mussel was scraped clean on the outside by using knives or scalpels before taking out the tissue for the analysis. Mussel samples were frozen (-20°C) for later analyses.

5.1.3 Dogwhelk and common periwinkle

Concentrations and effects of organotin on dogwhelk were investigated at eight stations and one station for common periwinkle (**Table 1**, **Figure 35**). TBT-induced development of irreversible male sex-characters in female dogwhelk, known as imposex, was quantified by the Vas Deferens Sequence Index (VDSI) analysed according to OSPAR-CEMP guidelines. The VDSI ranges from zero (no effect) to six (maximum imposex effect) (Gibbs et al., 1987). Detailed information about the chemical analyses of the animals is previously described (Følsvik et al., 1999).

Figure 35. Dogwhelk (left, photo: Jarle Håvardstun, NIVA) and common periwinkle (right, photo: Lise Tveiten, NIVA).

Dogwhelk lives on wave-exposed hard bottom areas in the tidal zone. Effects (imposex, (Gibbs, 1999)) and concentrations of organotin in dogwhelk were investigated using 50 individuals from each station. Individuals were kept alive in a refrigerator (at +4°C) until possible effects (imposex) were quantified, and about 25 females were analysed. The snail samples were collected from 12th September to 31st October 2022.

TBT-induced development of male sex-characters in female common periwinkle, known as intersex, was quantified by the intersex stage index (ISI) analysed according to guidelines (Bauer et al., 1995). The ISI ranges from zero (no effect) to four (maximum intersex effect).

5.1.4 Atlantic cod

Atlantic cod was caught from 18 stations (**Table 1**, **Figure 36**). The goal was to get a minimum of 15 cod from each station, but for some stations that was not possible. The cod was sampled from 16th August to 4th December 2022, except for cod collected until February 2023 in the Inner Sørfjord (53B). Cod was caught by Akvaplan-niva, and local fishermen except for the cod in the Inner Oslofjord (30B) which was collected by NIVA by trawling from the research vessel F/F Trygve Braarud owned and operated by the University of Oslo (UiO) (**Figure 36**). Instructions were given to the fishermen to catch coastal cod. Coastal cod is more attached to one place than open ocean cod which migrate considerably farther than coastal cod. Some spot checks were taken looking at the cross-section pattern of the otoliths. The otoliths are stored for further verification if necessary (Stransky et al., 2008). Tissue samples from each fish were prepared in the field and stored frozen (-20 °C) until analysis or the fish was frozen directly and prepared later at NIVA.

Figure 36. Trawling for cod in the Inner Oslofjord (30B). The cod population in the Oslofjord has been at a historically low level the recent years. NIVA has permission from the Norwegian Directorate of Fisheries to trawl for cod for research (photos: Merete Schøyen and Marthe T. S. Jenssen, NIVA).

The general lack of material was partially compensated for by making pooled samples of livers. The concerns using pooled samples or small sample size in cod are discussed in an earlier report (Green et al., 2015).

The age of the fish was determined by noting the number opaque and hyaline zones in otoliths (Vitale et al., 2019). These results, along with results from some other parameters (e.g., liver weight) are publicly available but not necessarily used for this report.

5.1.5 Common eider

Contaminants in the common eider were investigated at one station in the Kongsfjord at Svalbard (19N), which the present study considered as a reference station (**Table 1**, **Figure 37**). Blood samples were collected from 15 individuals (two subsamples from each) and eggs from 15 other individuals 5th June 2022. All samples are from adult nesting females. Ony data for stable isotopes is presented in this report due to short time after reversal of financial cuts.

Figure 37. Common eider (photo: Kjetil Sagerup, Akvaplan-niva).

5.2 Analytical procedures and information on quality assurance

The laboratories (NIVA, subcontractors EF and NILU) have participated in the Quality Assurance of Information for Marine Environmental Monitoring in Europe (QUASIMEME), International Food Analysis Proficiency Testing Services (FAPAS, BIPEA), international intercalibration exercises (EURL, JRC), and other proficiency testing relevant to chemical and imposex analyses. The results are acceptable. The quality assurance programme is corresponding to the analyses of the 2021 samples (Schøyen et al., 2021).

NIVA participated in the QUASIMEME Laboratory Performance Studies “imposex and intersex in Marine Snails BE1” in 2021. Females with imposex, penis-length-male, penis-length-female, average-shell-height, female-male-ratio, and VDSI were measured in two tests containing 40 samples. NIVA got the score satisfactory for all parameters except females with imposex, penis-length-female and VDSI in one test, which got the score questionable. This was due to lack of imposex-females in one of the tests.

In addition to the QUASIMEME exercises, certified reference materials (CRM) and in-house reference materials are analysed routinely with the MILKYS samples. It should be noted that for biota, the type of tissue used in the CRMs does not always match the target tissue for analysis. The Standard Reference Material (SRM) was ZRM 81 in mussel tissue. The in-house reference materials were apple juice, spiked fish oil, spiked fish meal and spiked fish liver.

The results are also quality checked before import to the database at NIVA and ICES using an interactive tool. In this tool, the new results are plotted together with the time series of the same

contaminant from the previous years, making it easier to pick out suspicious values. In addition, there is an automatic check of new values by comparison with previous year's values, so that stations/substances with values or LOQ values that differ greatly from previous years' values are automatically highlighted.

The laboratories used for the chemical testing are accredited according to ISO 17025[11].

Summary of quality control results

Standard Reference Materials (SRM) as well as in-house reference materials were analysed regularly (**Table 7**), and PAHs in blue mussel, as well as BDEs and HBCDDs in liver, was an internal reference (fish oil). Fish reference material was used as SRM for the quality assurance of PCBs in blue mussel and fish liver, and for tin organic compounds the reference material ZRM 81 was used as SRM mussel tissue. For the determination of the pesticides trans-nonachlor and DDTs in mussel and liver, internal reference materials provided by EF GfA Lab services were used, these consisted of fish meal and feeding stuff. For the quality assurance of chlorinated paraffines spiked fish was used as an in-house reference material, and spiked fish liver was used for quality control of per- and polyfluorinated chemicals (PFAS).

Table 7. Summary of the quality control of results for the 2022 biota samples analysed in 2022-2023. The SRM, in-house reference materials and quality assurance standards were analysed in series with the MILKYS samples and measured several times (N) over a number of weeks (W). The values are reported in the following units (in ww): metals (µg/kg), BDEs (pg/g), PCBs (ng/kg), DDTs (ng/kg), SCCPs and MCCPs (ng/sample), HBCDDs (ng/g), PAH (ng/kg), tin organic compounds (mg/kg), PFCs (% recovery) and trans-nonachlor (ng/g). Tissue types were: mussel soft body, snail (SB), fish liver (LI), and fish fillet (MU).

Code	Contaminant	Tissue type	SRM type	SRM value confidence interval	N	W	Mean value	Standard deviation
Ag	**Silver**	-	-	-	-	-		-
As	**Arsenic**	SB/LI	Apple juice	109 ± 22	45	8	108	9,80
Cd	**Cadmium**	SB/LI	Apple juice	95 ± 29	45	8	94,3	5,40
Cr	**Chromium**	SB/LI	Apple juice	103 ± 30	45	8	107	7,97
Co	**Cobalt**	-	-	-	-	-		-
Cu	**Copper**	SB/LI	Apple juice	4796 ± 1439	45	8	4716	259
Hg	**Mercury**	SB/MU	Apple juice	18,4 ± 4,8	45	8	17,0	1,10
Ni	**Nickel**	SB/LI	Apple juice	112 ± 34	45	8	108	11,1
Pb	**Lead**	SB/LI	Apple juice	95 ± 20	45	12	97,8	6,20
Zn	**Zinc**	SB/LI	Apple juice	5163 ± 1549	45	8	5160	299
Sn	**Tin**	-	-	-	-	-		-
BDE28	**2,2,4' Tribromodiphenylether**	SB	Internal RM (fish oil)	102 ± 178	7	16	103,8	4
BDE47	**2,2,4,4',-Tetrabromodiphenylether**	SB	Internal RM (fish oil)	1030 ± 84	7	16	1045,1	23,8
BDE100	**2,2',4,4',6-Pentabromodiphenylether**	SB	Internal RM (fish oil)	296 ± 52	7	16	280,3	24,2
BDE99	**2,2',4,4',5-Pentabromodiphenylether**	SB	Internal RM (fish oil)	138 ± 16	7	16	141,1	6,2
BDE154	**2,2',4,4',5,6'-Hexabromodiphenylether**	SB	Internal RM (fish oil)	353 ± 54	7	16	314,8	41,8
BDE153	**2,2',4,4'5,5'-Hexabromodiphenylether**	SB	Internal RM (fish oil)	174 ± 23	7	16	182,6	3,9
BDE209	**Decabromodiphenylether**	SB	Internal RM (fish oil)-	415± 254-	6	16	449,6	40,4
BDE49	**2,2',4,5'-tetrabromodiphenyleter**	SB	Internal RM (fish oil)	323 ± 60	7	16	329,8	29

[11] ISO/IEC 17025. General requirements for the competence of testing and calibration laboratories

Code	Contaminant	Tissue type	SRM type	SRM value confidence interval	N	W	Mean value	Standard deviation
BDE66	2,3',4,4'-Tetrabromodiphenyleter	-	Internal RM (fish oil	79,1 ± 16,2 -	7-	16	82,2	6,9
BDE119	2,3',4,4',6-Pentabromodiphenyl ether	SB	Internal RM (fish oil)	91 ± 18	7	16	84,5	6,1
CB77	PCB congener CB77	-	-	-	-	-	-	-
CB52	PCB congener CB52	SB/LI	Internal RM (fish)	426 ± 128	23	28	419	32,3
CB28	PCB congener CB28	SB/LI	Internal RM (fish)	255 ± 76	23	28	250	22,3
CB189	PCB congener CB189	-	-	-	-	-	-	-
CB180	PCB congener CB180	SB/LI	Internal RM (fish)	4555 ± 1367	23	28	4506,1	510,8
CB169	PCB congener CB169	-	-	-	-	-	-	-
CB167	PCB congener CB167	-	-	-	-	-	-	-
CB157	PCB congener CB157	-	-	-	-	-	-	-
CB156	PCB congener CB156	-	-	-	-	-	-	-
CB153	PCB congener CB153	SB/LI	Internal RM (fish)	4839 ± 1452	23	28	5198,2	540,6
CB138	PCB congener CB138	SB/LI	Internal RM (fish)	3578 ± 1073	23	28	3459,1	297,3
CB126	PCB congener CB126	-	-	-	-	-	-	-
CB123	PCB congener CB123	-	-	-	-	-	-	-
CB118	PCB congener CB118	SB/LI	Internal RM (fish)	832 ± 250	23	28	837,4	55,3
CB114	PCB congener CB114	-	-	-	-	-	-	-
CB105	PCB congener CB105	-	-	-	-	-	-	-
CB101	PCB congener CB101	SB/LI	Internal RM (fish)	1501 ± 450	23	28	1570,1	135,2
DDEOP	o,p'-DDE	SB/LI	Internal RM (feed)	0,11 ± 0,03	3	3	0,09	0,02
TDEOP	o,p'-DDD	SB/LI	Internal RM (feed)	0,267 ± 0,08	3	3	0,26	0,04
DDTOP	o,p'-DDT	SB/LI	Internal RM (feed)	0,259 ± 0,08	2	3	0,23	0,04
DDEPP	p,p'-DDE	SB/LI	Internal RM (feed)	5,01 ± 1,50	2	3	5,74	0,58
TDEPP	p,p'-DDD	SB/LI	Internal RM (feed)	1,73 ± 0,50	3	3	1,36	0,25
DDTPP	p,p'-DDT	SB/LI	Internal RM (feed)	0,613 ± 0,20	3	3	0,60	0,06
SCCP	Short-chain chlorinated Paraffins (C10-C13)	SB/LI	Internal RM (spiked fish)	10000	17	13	10540	1062
MCCP	Medium-chain chlorinated Paraffins (C14-C17)	SB/LI	Internal RM (spiked fish)	10000	17	13	10140	1453
α-HBCDD	α-Hexabromocyclododecane	SB	Internal RM (fish oil)	1,3 ± 0,3	7	16	1,35	0,124
β-HBCDD	β- Hexabromocyclododecane	SB	Internal RM (fish oil)	0,5 ±0,1	7	16	0,51	0,047
γ-HBCDD	γ- Hexabromocyclododecane	SB	Internal RM (fish oil)	0,5 ± 0,1	7	16	0,52	0,062
BGHIP	Benzo[ghi]perylene	SB	Internal RM (fish oil)	46 ± 23	3	5	36	-
ICDP	Indeno[1,2,3-cd]pyrene	SB	Internal RM (fish oil)	53 ± 64	1	5	41	2
BBJF	Benzo[b+j]fluoranthene	SB	Internal RM (fish oil)	93 ± 65	3	5	49	5,2
DBA3A	Dibenzo[ac,ah]anthracene	-	-	-	-	-	-	-
BKF	Benzo[k]fluoranthene	SB	Internal RM (fish oil)	54 ± 50	3	5	44	3,8
ACNLE	Acenaphthylene	SB	Internal RM (fish oil)	38 ± 11	3	5	38	3,5
ANT	Anthracene	SB	Internal RM (fish oil)	49 ± 26	3	5	44	1,4
BAA	Benzo[a]anthracene	SB	Internal RM (fish oil)	49 ± 15	3	5	44	0,9
BAP	Benzo[a]pyrene	SB	Internal RM (fish oil)	42 ± 23	3	5	37	3,7
CHR	Chrysene	SB	Internal RM (fish oil)	49 ± 23	3	5	86	3,9
FLU	Fluoranthene	SB	Internal RM (fish oil)	42 ± 13	3	5	44	1,5
FLE	Fluorene	SB	Internal RM (fish oil)	58 ± 38	3	5	48	1,6
NAP	Naphthalene	SB	Internal RM (fish oil)	61 ±32	3	5	40	4,6
PA	Phenanthrene	SB	Internal RM (fish oil)	47 ± 21	3	5	37	3,5

Code	Contaminant	Tissue type	SRM type	SRM value confidence interval	N	W	Mean value	Standard deviation
PYR	**Pyrene**	SB	Internal RM (fish oil)	42 ± 12	2	5	47	4,5
ACNE	**Acenaphthene**	SB	Internal RM (fish oil)	41 ± 14	3	5	54	7,3
TBBPA	**Tetrabromobisphenol-A**	-	-	-	-	-	-	-
BPA	**Bisphenol-A**	-	-	-	-	-	-	-
BPA	**Bisphenol-A**	-	-	-	-	-	-	-
BPA	**Bisphenol-A**	-	-	-	-	-	-	-
APO	**4-tert-oktylfenol**	-	-	-	-	-	-	-
APO	**4-n-oktylfenol**	-	-	-	-	-	-	-
APO	**4-n-nonylfenol**	-	-	-	-	-	-	-
MBT	**Monobutyltin (MBT)**	SB	ZRM 81 (mussel)	1,3 ± 0,2	2	1	1,38	0,12
DBT	**Dibutyltin (DBT)**	SB	ZRM 81 (mussel)	1,6 ± 0,3	2	1	1,75	0,07
TBT	**Tributyltin (TBT)**	SB	ZRM 81 (mussel)	2,1 ± 0,12	2	1	1,89	0,12
TPhT	**Triphenyltin (TPhT)**	SB	ZRM 81 (mussel)	1,5 ± 0,3	2	1	1,38	0,09
PFBS	**Perfluorobutane sulphonate**	LI	In-house spiked liver	100%[1)]	10	20	90,6	2,80%
PFHxA	**Perfluorohexane acid**	LI	In-house spiked liver	100%[1)]	10	20	90,5	8,36%
PFHpA	**Perfluoroheptane acid**	LI	In-house spiked liver	100%[1)]	10	20	90,5	2,71%
PFOA	**Perfluorooctane acid**	LI	In-house spiked liver	100%[1)]	10	20	90,7	5,26%
PFNA	**Perfluorononane acid**	LI	In-house spiked liver	100%[1)]	10	20	94,8	3,69%
PFOS	**Perfluorooctane sulphonate**	LI	In-house spiked liver	100%[1)]	10	20	133*	4,43%
PFOSA	**Perfluorooctane sulphone amide**	LI	In-house spiked liver	100%[1)]	10	20	103	8,24%
PFHxS	**Perfluorohexane sulphonate**	LI	In-house spiked liver	100%[1)]	10	20	87,0	5,35%
PFDA	**Perfluorodecanoic acid**	LI	In-house spiked liver	100%[1)]	10	20	96,5	4,69%
PFUDA	**Perfluoroundecanoic acid**	LI	In-house spiked liver	100%[1)]	10	20	113	3,64%
PFTrDA	**Perfluorotridecanoic acid**	LI	In-house spiked liver	100%[1)]	10	20	98,1	5,62%
PFDS	**Perfluorodecanesulphonate**	LI	In-house spiked liver	100%[1)]	10	20	75,5	5,50%
	Dieldrin	SB	Internal RM (feed-	2,05 ± 0,6	3	3	1,54	0,20
	Trans-Nonachlor	SB	Internal RM (feed)	1,39 ± 0,40	1	3	1,22	-

* The spiked in-house liver is known to contain approximately 0,7 ng/g of PFOS, which gave a higher recovery resulting in 133%.

1) Recovery of spiked control sample.

Subcontractor NILU has analysed fish liver from Atlantic cod (*Gadus morhua*) in this programme. The laboratory has participated in Quality Assurance of Information for Marine Environmental Monitoring in Europe (QUASIMEME, 2021) and Food Analysis Proficiency Testing Services (FOOD 2021/2022) for the testing of PCBs. The Standard Reference Materials (SRM) in these tests were EDF-2525 in blue mussel, fish liver and fish fillet. For the quality assurance of chlorinated paraffines the reference material was certified through the European Commission Joint Research Centre (JRC, 2021).

5.3 QA/QC

Additional to the general quality assurance (QA) done by the individual laboratory, all the results from EF, NILU and NIVA are transferred into NIVAs laboratory information management system (LIMS). An extra quality control is then performed by trained NIVA personnel. In this quality assurance, trends and variations within the different stations are also considered. NIVA has developed an app in R (R Statistical Software, see chapter 5.8) to make this control easier and more efficient. Here trends from the last years will appear and deviating results are marked (example in **Figure 38**). A manual assessment is then performed before the results are validated and reported to the project manager and automatically imported in to NIVAs database for further treatment. When the results are questionable, a deviation are registered to NIVAs internal control system, and a complaint are reported to the relevant laboratory. For the 2022 data, nine mussel samples showed low results for arsenic (As) and a deviation was sent to the subcontractor. There was not enough material for reanalysis, but the data was inspected with no sign to mistake was found. Two deviations where also sent for two mussel samples due to high zinc (Zn) levels, and there were found mistakes in the calculations and new results were reported. All activity is recorded in NIVAs deviation/control system.

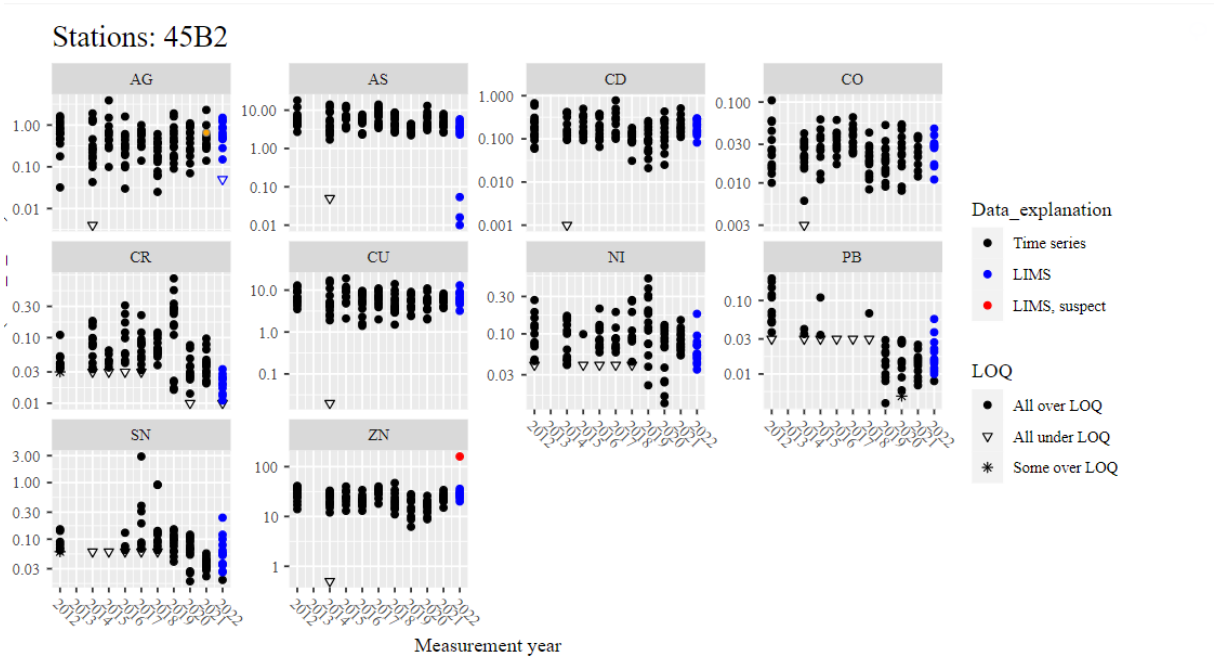


Figure 38. Screenshot of QA app used for manual assessment of chemical concentrations every year. The screenshot shows the results for a single station and for one substance group. The concentrations to perform QA on (last year's results, red and blue dots) are shown together with previous year's results (black dots) in order to make it easier to identify suspect data. In addition, the app aids manual QA by automatically "flagging" possibly suspect data (red dots) following a set of rules, comparing using both this year's data as well as previous year's data. The person performing the QA can then decide whether or not the flagged results should be treated as a deviation and a complaint should be sent to the relevant laboratory.

5.4 LOQ

The proportion over LOQ (detection frequency, in %) of the various compounds for each tissue is given in **Figure 39. Figure 40** gives the observed LOQ (median values) in blue mussel in the various compounds since 2002.

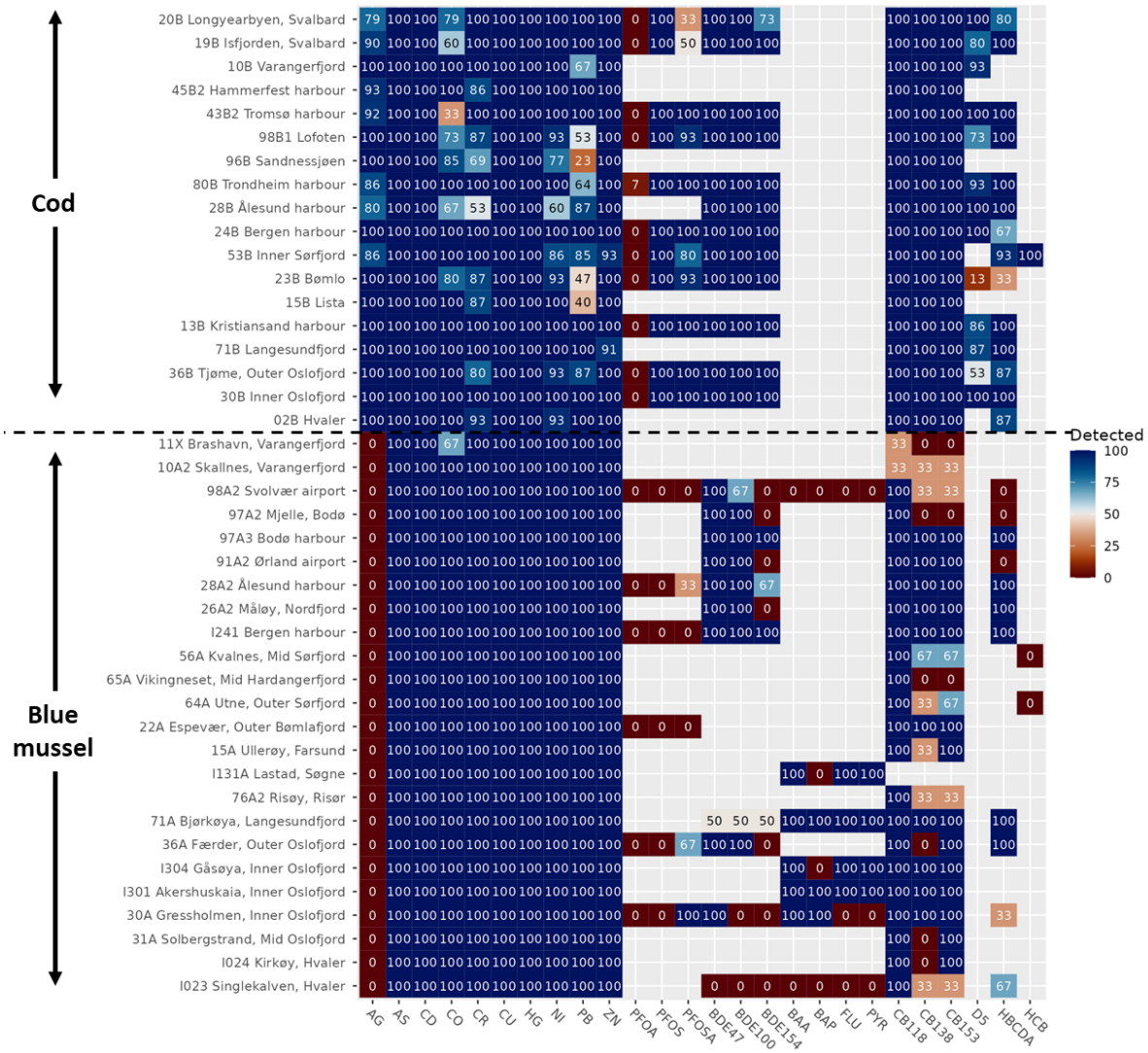


Figure 39. Proportion over LOQ (detection frequency, in %) of the compounds for each tissue. Within each species, the stations are ordered along the coastline starting north moving south.

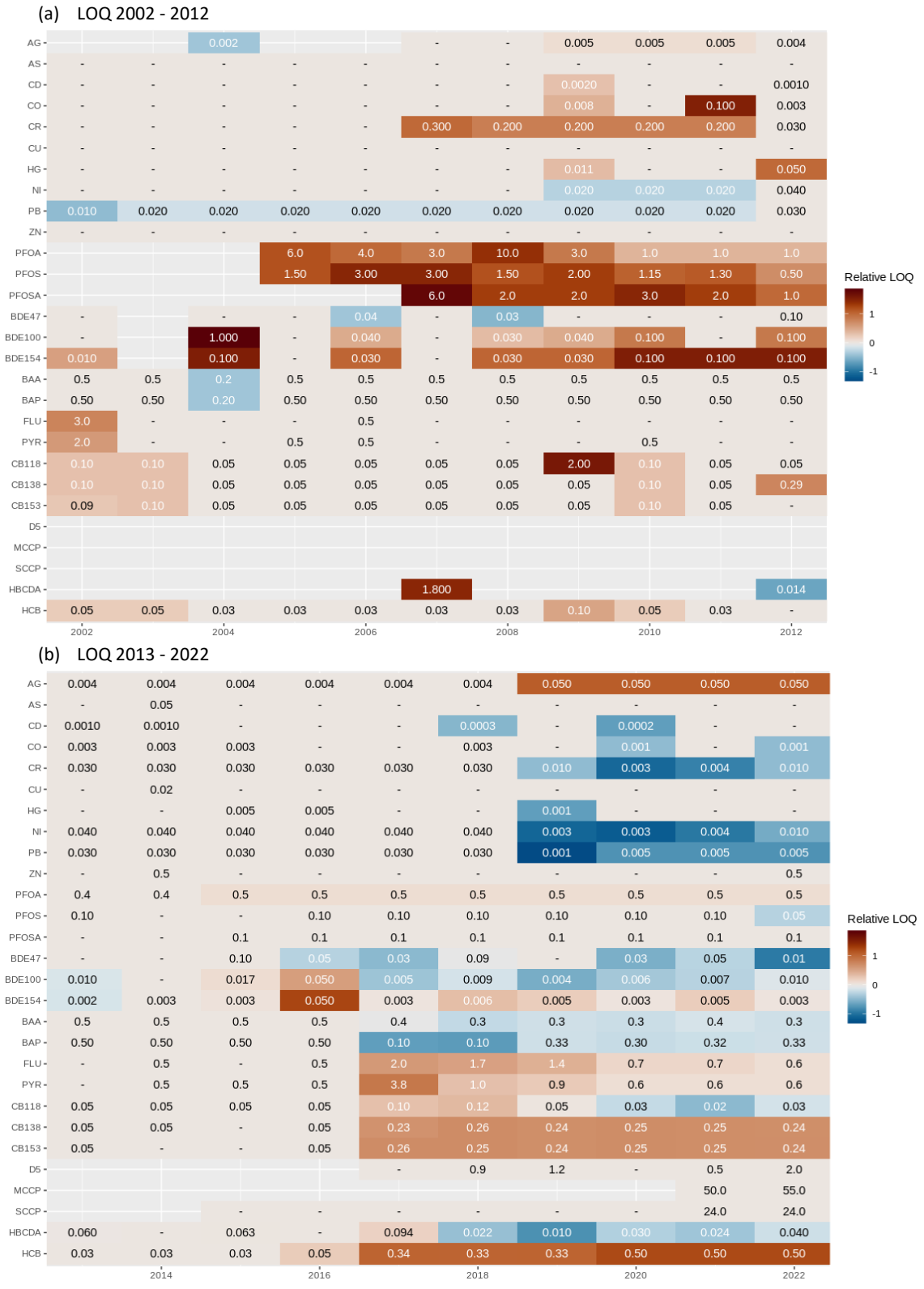


Figure 40. Changes in LOQ values over time. The numbers in the table show observed LOQ (median values, mg/kg ww for metals incl. Hg, and ug/kg ww for others). In blue mussel for various compounds in the periods 2002-2012 (a) and 2013-2022 (b). The colors indicate the "relative LOQ" compared to the reference period 2013-2015 (median LOQ versus the median LOQ during the years 2013-2015), with blue colors showing lower (better) LOQ and red colors showing higher LOQ compared to the reference period. For some groups, e.g. PFAS, there were no measurements in the reference period (shown in light grey).

5.5 Classification of environmental quality (EQS and PROREF)

There are several systems that can be used to classify the concentrations of contaminants observed. No system is complete in that it covers all the contaminants and target species-tissues investigated in this programme. Up to and including 2015 investigations, MILKYS relied largely on a national classification system prepared by the NEA as described in a report (Molvær, J. et al., 1997). This system was based on high background concentrations derived from an array of national and international monitoring programme and investigative literature.

With the ratification of EU Water Framework Directive (WFD) (EU, 2000) by Norway in 2007 and the subsequent application of the daughter directive on EQS (EU, 2013) the assessment of the environment using EQS became imperative. The daughter directive outlines 45 priority substances or groups of substances. Several of these substances are monitored by MILKYS. The EQS apply to concentrations in water, and for fifteen substances it also applies to concentrations in biota (see **Table 3** for contaminants in MILKYS). There is a provision in this daughter directive which allows a country to develop their own EQS for water, sediment and biota provided these offer the same level of protection as the EQS set for water. Norway used this approach and developed their own EQS for biota, water and sediments for river basin specific pollutants not otherwise accounted for by the EU directives (Direktoratsgruppen vanndirektivet, 2018).

Assessing the risk to human consumption from elevated concentrations of contaminants in seafood has not been the task of this programme and hence, the EU foodstuff limits have not been applied. However, it should be noted that the NEA communicates the results to the Norwegian health authorities. Also, it should be noted that the background dossiers for the EQS (EU, 2013) as well as the national environmental quality standards (Miljødirektoratet, 2016) applied foodstuff limits if these are lower than the limits found by assessing risk of secondary poisoning of marine organisms.

Both EU and national standards are referred to collectively in this report as EQSs. Both standards are risk-based, i.e., exceedances of EQSs are interpreted as potentially harmful to the environment and or humans and remedial action should be considered.

The application of these standards has been discussed previously (Green et al., 2016), and three main challenges were noted. The first is that the standards for biota are generally not species or tissue specific but refer to whole organisms. The second is that the standards are often in large conflict with the system based on background concentrations (see chapter 3.8.3 in the report (Green et al., 2016)). And lastly, the standards do not address all the contaminants in all the tissues that are monitored, for example, there are no EQSs for metals in biota except for mercury. To address this issue for this report, and in dialogue with the NEA, Norwegian provisional high reference contaminant concentrations (PROREF) were derived and used in parallel with the risk-based standards (see method description below).

This report of the 2022 investigations addresses the principle cases primarily where median concentrations exceeded EQS and secondarily where median concentrations exceeded PROREF (**Table 4**). Exceedances of PROREF (see derivation explained in chapter 3.5.1 (Green et al., 2016)) were grouped in six factor-intervals: <PROREF, 1-2x (between PROREF and two times PROREF), 2-5x, 5-10x, 10-20x and >20x.

The EQS and PROREF as well as time trend analyses use concentrations on a wet weight (ww) basis. The choice of basis (i.e. concentrations on a wet weight, dry weight, or fat weight basis) follows the OSPAR approach aimed at meeting several considerations: scientific validity, uniformity for groups of contaminants for specific tissues and a minimum loss of data. As to the latter, the choice of basis will affect the number of data that can be included in the assessment, depending on available information on dry weights, wet weights, and lipid weights.

A few contaminants have both an EQS and a PROREF, and the relationship between them are depicted in **Figure 41**. Different organisms have different PROREF. In blue mussel, the EQS are much higher than PROREF (1.6 times to 3000 times higher), while for cod the EQS are more similar (0.36-24 times PROREF). The differences illustrate the difference in purposes between the two classification systems.

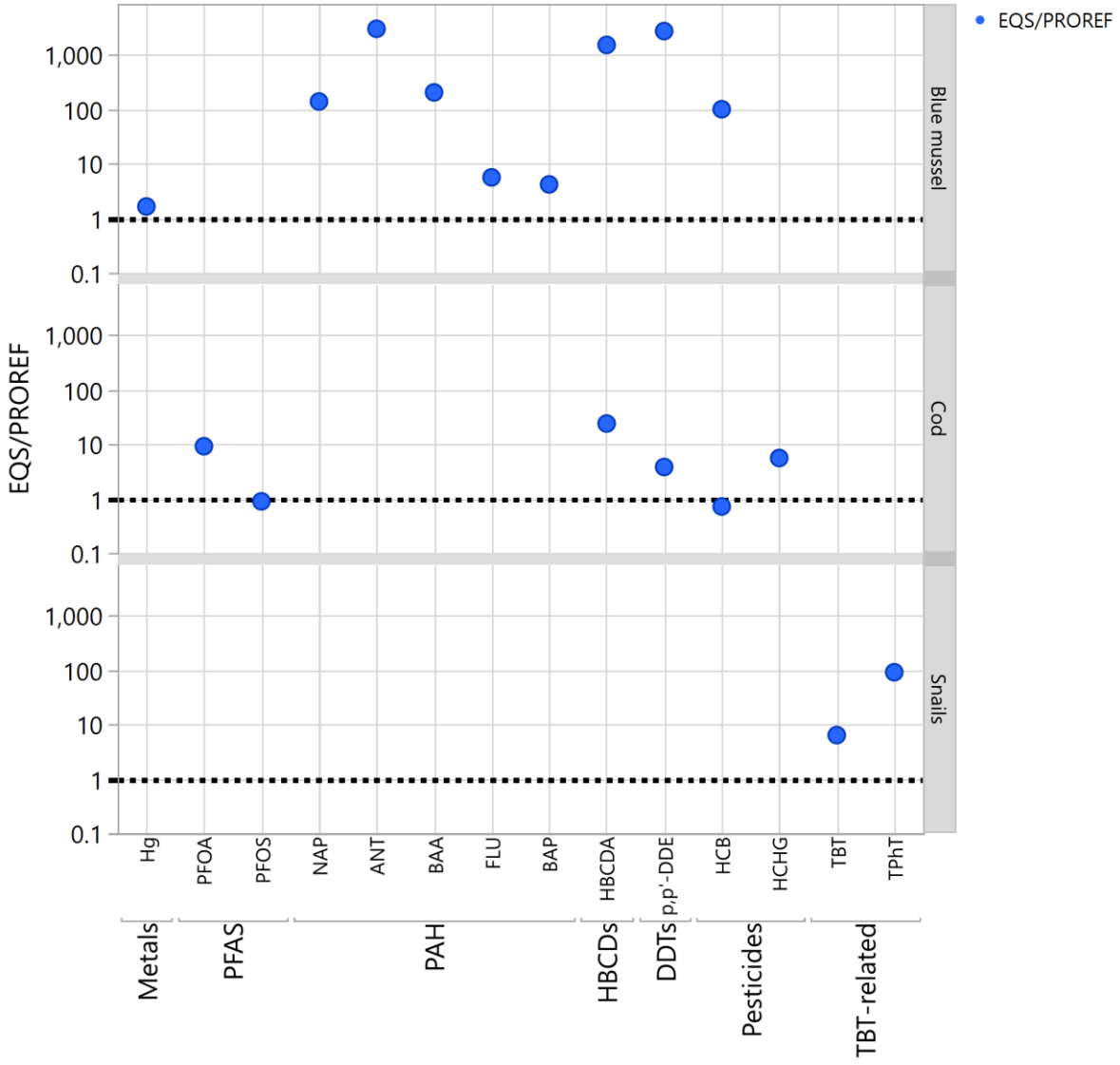


Figure 41. Relationship between EQS and PROREF (EQS/PROREF). A line where EQS=PROREF (i.e. where the ratio is 1) is indicated. The ratio is shown pr species since PROREFs are on a species basis.

5.6 Sum RQ and sum PROREFratio>1

For each station and species, the measured environmental concentration (MEC) was divided by the EQS (or predicted no-effect concentration PNEC). In ecotoxicology, this ratio is denoted RQ (risk quotient (Eq 1).

$$RQ_i = \frac{MEC_i}{EQS_i}$$

Furthermore, a sum of RQ for each station and species is estimated as a sum of each RQ at the given station for the specific species (Eq. 2) (Backhaus and Faust, 2012).

$$\text{sum RQ} = \sum_{i=1}^{n} RQ_i$$

The sum RQ can be used to assess which stations are "worst". However, the analytical repertoire varied between stations, therefore it is difficult to compare all stations with each other. However, we believe that this is a start for evaluating mixture toxicity. However, it must be kept in mind that the EQSs are based on different protection measures, and as such do not necessarily describe toxicity to the same organism. Sum RQ is often a tier 1 of assessing mixture toxicity, followed by more refined assessments.

Most of the contaminants with an EQS in this study accumulate in lipids of organisms (except mercury, PFOS and PFOA). For cod, the fatty rich liver is analysed, and therefore the results of assessment vs. EQS are very conservative. For these substances, the implementation strategy for WFD (European Commission, 2014) recommend that measured concentrations in fish should be normalised to fish with 5% lipid content. We have chosen to do this normalisation only for the sum RQ-approach. Also for blue mussels the sum RQ for blue mussel was normalised to a 1% lipid content to make the sum RQs comparable between stations with different lipid% in blue mussel.

For mercury, PFOS and PFOA that do not accumulate in lipids, the recommended practice is to normalise against another parameter such as dry weight. The default dry weight content for fish is approximately 26% while for blue mussels a suggested default dry weight content is 8.3%. In MILKYS, the lipid content of mussel and livers of cod and the dry weigh of mussel are measured. However, the dry weight of cod has not been done since 2020. But based on historic measurements (1981-2020) of dry weight of cod livers, a good correlation between lipid% and dry weight of cod liver was found. The correlation is presented in **Figure 42**. For cod muscle, the median dry weight% was 20, see **Figure 42**. The median dry weight of 20% for cod muscle was therefore used for normalisation of data to 26% dry weight. Mussel samples were also normalised to a dry weight of 8.3%.

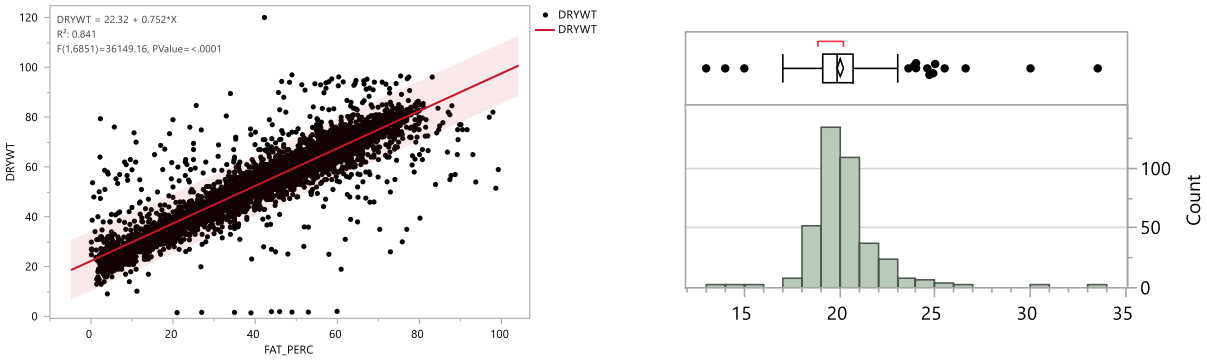


Figure 42. To the left: correlation between lipid% and dry weight% of cod liver in the period 1981-2020. The linear regression line is shown in red, while the uncertainty of the prediction is shown in transparent red colour. The prediction formula, the R^2 and F-test is shown in the upper left corner. The number of data points for lipid% and dry weight % was 6,853. **To the right**: histogram of dry weight% of cod muscle in the period 1981-2020. The median and mean dry weight% was 20% with upper and lower 95% mean from 19.89 to 20.23%. The number of muscle samples was 378.

Based on the sum RQ-approach, we also calculated sum PROREFratio. PROREF is an estimation of concentrations above background levels when PROREF exceeds 1. Sum PROREFratio was calculated the same way as RQ and sum RQ *with one important difference*. Only PROREFratio >1 was included in the sum PROREFratio. Therefore, the sum PROREFratio for a station with only background concentrations will be 0.

5.7 PROREF

The MILKYS programme and its forerunners have since 1981 generated over 400 000 analyses on concentrations of over 100 contaminants in biota alone, mostly for blue mussel and cod. This unique dataset was used to define and determine a reference value, Norwegian provisional high reference contaminant concentrations (PROREF). PROREF is a comprehensive set of species-tissue-basis-specific contaminant concentrations that are statistically low when considering all MILKYS-results for the period 1991-2016. This tool sets reference concentrations for contaminants, mostly in areas presumed remote from point sources of contamination, and thus provides a valuable method for assessing contaminants levels in addition to the risk based EQS. The PROREF value can be interpreted as *the upper range of contaminant concentrations in reference (or background) stations* - i.e., stations far from point sources of contamination. The PROREF is calculated for each species/tissue separately and was calculated for 177 combinations of contaminant and species/tissue in 2017, with a revision in 2019 (which in only four cases changed the value by >20%). We use the same values in this report.

The selection of background stations is objective and reproducible, based solely on concentration data (i.e., not based on expert judgment; see below). The derivation is done independently for each contaminant/species/tissue, taking into account that different contaminants may have different geographic patterns and therefore different stations should be considered to be "background". We see PROREF as a valuable method of assessment of levels of contaminants along the coast of Norway both in impacted and less impacted areas in addition to EQSs.

The derivation of PROREF has two basic steps: first, determine which stations that are reference stations, and secondly, to determine the upper range of concentrations at those stations (i.e., the PROREF value). In more detail, this is the procedure followed for a given contaminant in each species/tissue, measured on a given basis (wet-weight, dry-weight etc.):

1. Selection of reference stations:
    a. Only data from 1991 to 2016 were considered (25 years) on the general assumption that prior to this time, important discharge reductions were not in place.
    b. For each station, calculate annual median concentrations (i.e. 25 numbers per station, if the time series is complete).
    c. For each station, discard the highest 10% of the values from b (i.e., remove possible "outlier years").
    d. Discard stations with less than five years of data, counting only years with at least two analysed samples for blue mussel stations and 10 analysed samples for cod stations.
    e. For each remaining station, calculate the logarithm of the median of the values from c.
    f. Set values below the limit of quantification (LOQ) to a random value between 0.5*LOQ and 1*LOQ.
    g. Order stations by concentration, from the lowest to the highest.
    h. Test the difference between station 1 and station 2 using a t-test.
    i. If station 1 is not statistically different from station 2 (at level P = 0.05), combine the values of both stations, and test the difference between station 1+2 and station 3 (again, using a t-test).
    j. If station 1+2 is not statistically different from station 3, combine 1,2 and 3 and test the difference between station 1+2+3 and station 4.
    k. Continue this procedure until a statistically significant difference is encountered. The reference stations are defined as all stations that were not statistically different.
2. Determine the upper range of concentrations at the reference stations.
    a. Combine the concentrations (raw data, i.e. concentrations at sample level) from the reference stations.
    b. Calculate the upper 95 percentile of these concentrations.
3. Determine the PROREF value.
    a. If all concentrations are above LOQ, the outcome of 2b equals the PROREF value.
    b. If some concentrations are below LOQ, repeat step 1 and 2 *n* times (in order to minimize the effect of the random value selection in step 1f). This results in *n* values (outcomes of step 2b). PROREF is defined as the median value of these values. We used *n* = 21.

The PROREF values applied in this report are shown in **Table 4** of the MILKYS report for 2020 data (Schøyen et al., 2021).

5.8 Statistical time trend analysis

The statistical time trend analysis follows the method used in OSPAR for contaminants in biota[12] as closely as possible (there has been changes to the OSPAR methodology every year since 2014[13]). The concentrations are log transformed and changes in the log concentrations over time are modelled using a linear or a non-linear (spline) model:

A. No change over time: mean concentration = a
B. Linear change over time: mean concentration = a + b*Year
C. Non-linear change over time: mean concentration = s(Year),
   where *s* is a smoother with either 2, 3 or 4 degrees of freedom (denoted C2, C3 and C4).

For every time series, several models may be fitted, and the model that fits the data best (the most parsimonious model) is used. The type of models that are considered depend on the number of years of data, counting only years with at least one concentration over LOQ[14]:

- 1-4 years: no model is fitted
- 5-6 years: models A and B
- 7-9 years: models A, B, and C2
- 10-14 years: models A, B, C2, and C3
- 15 years or more: models A, B, C2, C3, and C4

Following OSPAR, we used thin plate regression splines for the non-linear models. Also following OSPAR, three more refinements (described in OSPAR 2022c) to the selection of "accepted" years were performed. This was done to prevent over-fitting if there are many less-thans or if the less-thans are unevenly distributed across the time series, for instance avoiding that time series start with years with only values under LOQ.

The model is fitted by maximum likelihood assuming each of the random effects are independent and normally distributed. The analysis takes into account that the analytical error (the uncertainty in the chemical determination of concentrations), adjusting the likelihood correspondingly. This error varies from 5 – 50% depending on substance and laboratory. The analytical error was assumed to be known, based on information from the laboratories. The likelihood was also depending both on over-LOQ and under-LOQ values, where the latter likelihood was taken as the likelihood of values being below LOQ (given a proposed model and coefficients). In principle, both our approach and OSPAR's time series approach are similar, as both uses a maximum likelihood approach. This is expected to be a better approach than "workaround" approaches, such as replacing values under LOQ with ½ LOQ or random numbers between ½ LOQ and 1 LOQ. The technical approach to estimating model parameters differ between our approach and OSPAR: we used a Bayesian approach with non-informative priors using the JAGS program through R, while OSPAR uses the optim() function in R. For time-series with concentrations under LOQ (i.e., left-censored values), we divided the data set in two (values over and under LOQ), and estimated the total log-likelihood as the sum of the log-likelihoods for the two parts (Qi et al., 2022). While we expect our method and OSPAR's method to be similar, there may be differences between the two approaches due to the differences in estimation techniques.

[12] https://dome.ices.dk/ohat/trDocuments/2022/help_methods_biota_contaminants.html
[13] https://dome.ices.dk/ohat/trDocuments/2022/help_methods_changes.html
[14] https://dome.ices.dk/ohat/trDocuments/2022/help_methods_less_thans.html

Using every sample measurement instead of the only annual medians (as in the previous years' analysis until the 2020 survey) results in higher sample size and thereby higher statistical power (lower p-values). Including analytical error instead of assuming there is no analytical error results in lower statistical power. In most cases, the effect of sample size dominates over the effect of analytical error, resulting in higher statistical power; therefore, more time trends are detected than before (see example in **Figure 43**). The results are now much more in line with OSPAR's results, shown in OSPAR's OHAT tool (https://dome.ices.dk/ohat). Analyses were performed using R version 4.1.3 and JAGS 4.3.0 with the R packages runjags 2.2.1-7, rjags 4.13, mgcv 1.8-40 and leftcensored 0.0.0.9000[15].

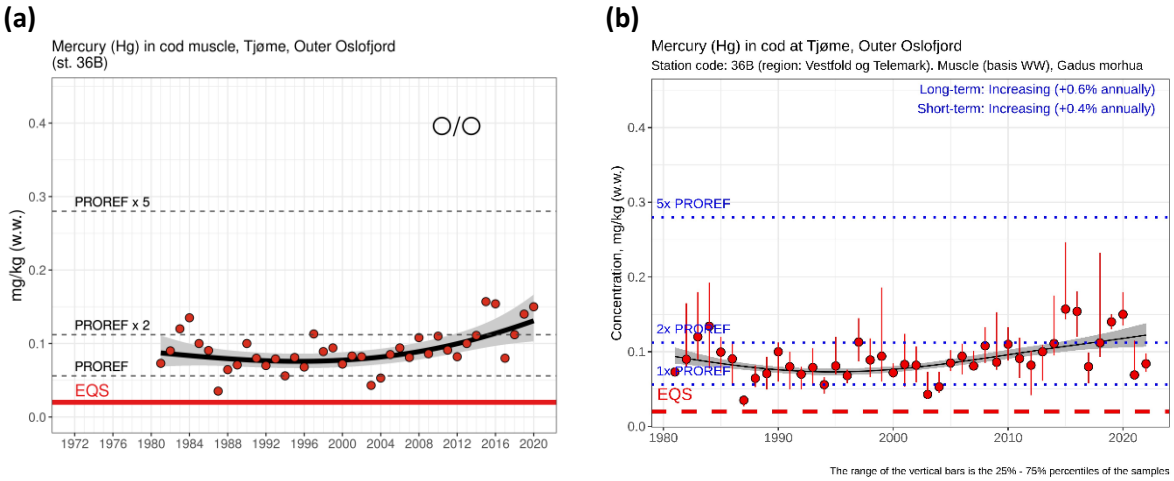


Figure 43. Differences in detection of time trends for mercury in cod muscle at Tjøme (36B). (a) The previous time series approach (until the 2020 survey), using only median values. Neither short- or long-term time trends were detected (P > 0.18). (b) The updated statistical method utilising all data measurements (lines shows 25th-75th percentiles), but taking analytical error into account. Both short- or long-term time trends were detected (P < 0.001). Both time trends are also detected in OSPAR's analyses (figure not shown here; see https://dome.ices.dk/ohat). For full explanation of figure see example in **Figure 23**.

When there is a significant non-linear time trend, we classified the trend results for a given period (i.e., either the whole series or the last 10 years) by comparing the height of the time series curve at only the start and end of the period (taking into account the uncertainty of these two times). If there is a non-linear trend, these two points may not differ even if the time trend in itself is signficantly different (**Figure 44**). We have denoted these cases as “no change” as short for “non-linear trend but no net change over the time period”. Thus, these differ from “no trend” (where no linear or non-linear change can be detected). Again, this follows the practice of OSPAR (OSPAR 2022c).

[15] https://github.com/DagHjermann/leftcensored

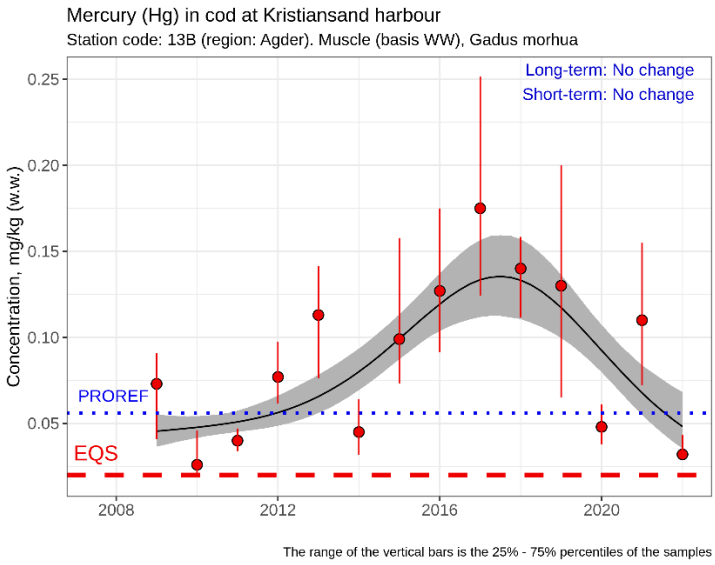


Figure 44. Example of time series classified as "no change" (as opposed to "no trend"). The time series has a significant non-linear trend, but there is no significant difference between the start point and last point of the curve.

The statistical analysis of time trends was carried out on all the results, including those for biological effect parameters. These analyses as well as the figures similar to that performed using R Statistical Software[16] version 4.0.2 with the packages nlme (nonlinear mixed effects, version 3.1-148) and mgcv (Generalized Linear Models including Generalized Additive Models and Generalized Additive Mixed Models, version 1.8-31).

Increased LOQ for a few contaminants has posed challenges for the method we employ, especially for stations and species with concentrations near the LOQ. For these contaminants, we have chosen to do an individual quality assessment of the data. In these cases, we compared our method to:

- linear regression using the method developed by Helsel (2011) for left-censored data (i.e. data below LOQ)
- OSPAR- reported time trends (https://dome.ices.dk/ohat)

When conflicting results were obtained by the three methods, we have reported results based on a "weight of evidence" and our best judgement for the three methods.

5.9 Sum parameters

In MILKYS, we have changed the practice for reporting sum parameters including the reporting of data for the survey in 2021 (Schøyen et al., 2022). The method is described in a NIVA note (Grung, M. et al., 2023).

There are two estimates for reporting sum parameters below LOQ; EFSA (European Food Safety Authority) describes these as lower bound[17] (LB) and upper bound[18] (UB). When summing several isomers, the partial sum of *not* quantified partial sums can either be set to 0 (LB) or to LOQ (UB). LB will be the lowest estimate for the sum parameter while UB will be the highest estimate. According

[16] https://www.r-project.org/
[17] lower bound estimate | EFSA (europa.eu)
[18] upper bound estimate | EFSA (europa.eu)

to the Water Frame Directive, LB must be used for total parameters (EU, 2009). For sum parameters (e.g. sumPCB7, sumPAH16, sumPBDE6, and MCCP/SCCP) the concentrations in MILKYS (including reporting of the survey in 2020; (Schøyen et al., 2021)) have mainly been reported as UB – i.e. data below LOQ were set to LOQ for the subtotal. The exception was MCCP and SCCP which were analysed by NILU (eider from Svalbard), where the sum parameters were reported as LB (the pragmatic reason for this is that NILU and EF reports data in different ways).

5.10 Other statistical analyses

JMP[19] statistical software (version 17.1.0) was used for data treatment after initial treatment in R. Mosaic plots, heatmaps, stacked barplots and tables used in extended summary data were produced using JMP.

[19] https://www.jmp.com/

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Supplementary data

Assessments of exceedances of PROREF for contaminants not selected for presentation in 2022 are presented here. The contaminants are listed in **Table S1**.

Table S1. List of contaminants *not selected* in 2022 for which a PROREF exist. The PROREFs are given in µg/kg (ng/g ww), except for Tin (Sn) given in mg/kg ww, and VDSI (index). PROREF is given with two significant digits.

		PROREF for contaminants				
	Contaminant	**Unit**	**Blue mussel**	**Cod**	**Dogwhelk**	**Common periwinkle**
Metals	Tin (Sn)	mg/kg ww	0.3	0.3		
PFAS	Perfluorobutane sulfonate (PFBS)	µg/kg ww		8		
	Perfluorononanoic acid (PFNA)			5		
PBDEs	PBDE congener -28 (BDE28)			1.4		
	PBDE congener -49 (BDE49)			4.0		
	PBDE congener -66 (BDE66)			0.6		
	PBDE congener -71 (BDE71)			0.4		
	PBDE congener -77 (BDE77)			1.7		
	PBDE congener -85 (BDE85)			1.7		
	PBDE congener -99 (BDE99)		0.06	0.75		
	PBDE congener -126 (BDE126)		0.05	0.1		
	PBDE congener -138 (BDE138)			0.3		
	PBDE congener -153 (BDE153)		0.05	0.15		
	PBDE congener -183 (BDE183)		0.3	0.6		
	PBDE congener -196 (BDE196)		0.3	1		
	PBDE congener -209 (BDE209)		1.3	2		
PAHs	Naphthalene (NAP)		17			
	Acenaphthene (ACNE)		0.8			
	Acenaphthylene ACNLE		1			
	Fluorene (FLE)		1.6			
	Anthracene (AN)T		0.8			
	Phenanthrene (PA)		2.3			
	Benzo[k]fluoranthene (BKF)		1.5			
	Benzo[b+j]fluoranthene (BBJF)		6.2			
	Benzo[ghi]perylene (BGHIP)		2.1			
	Dibenz[a,c/a,h]anthracene (DBA3A)		0.5			
	Indeno[1,2,3-cd]pyrene (ICDP)		1.7			
PCBs	PCB congener 28 (CB28)		0.12	8		
	PCB congener 52 (CB52)		0.2	16		
	PCB congener 101 (CB101)		0.2	32		
	PCB congener 180 (CB180)		0.1	46		
HBCDs	β-hexabromocyclododecane (HBCDB)		0.02	0.4		
	γ-hexabromocyclododecane (HBCDG)		0.03	0.89		
DDTs	p,p'-DDE (a DDT metabolite)		0.22	160		
	p,p'-DDD (TDEPP)		0.1	32		
Pesticides	α HCH = alpha HCH (HCHA)			8		
	Lindane, γ HCH = gamma			11		
TBT-related compounds	Dibutyltin (DBT)					2.0
	Dioctyltin (DOT)				1.2	
	Monobutyltin (MBT)					1.3
	Monooctyltin (MOT)				1.2	
	Tributyltin (TBT)				24	
	Tricyclohexyl-stannylium (TCHT)				2.3	
	Triphenyltin (TPhT)				1.7	
	Tetrabutyltin (TTBT)				1	
Biomarkers	Vas Deferens Sequence Index (VDSI)	index			3.7	

The figures with exceedances of PROREF are shown in **Figure S1** to **Figure S6**.

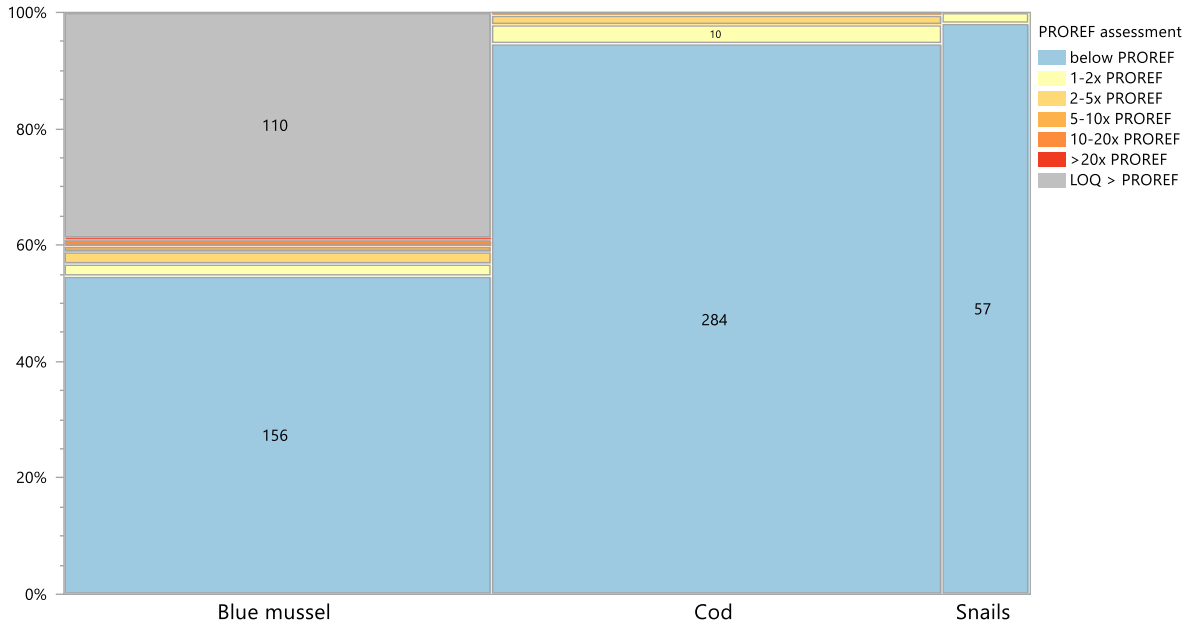

Figure S1. Exceedances of PROREF for contaminants *not selected* in 2022 in a mosaic plot. The cells are labelled by the number of stations and parameters. The exceedances are considered by the median for each station and species. The colours represent below or above exceedance of PROFEF (darker yellow to red), or that the PROREF was below LOQ, and therefore could not be classified (grey).

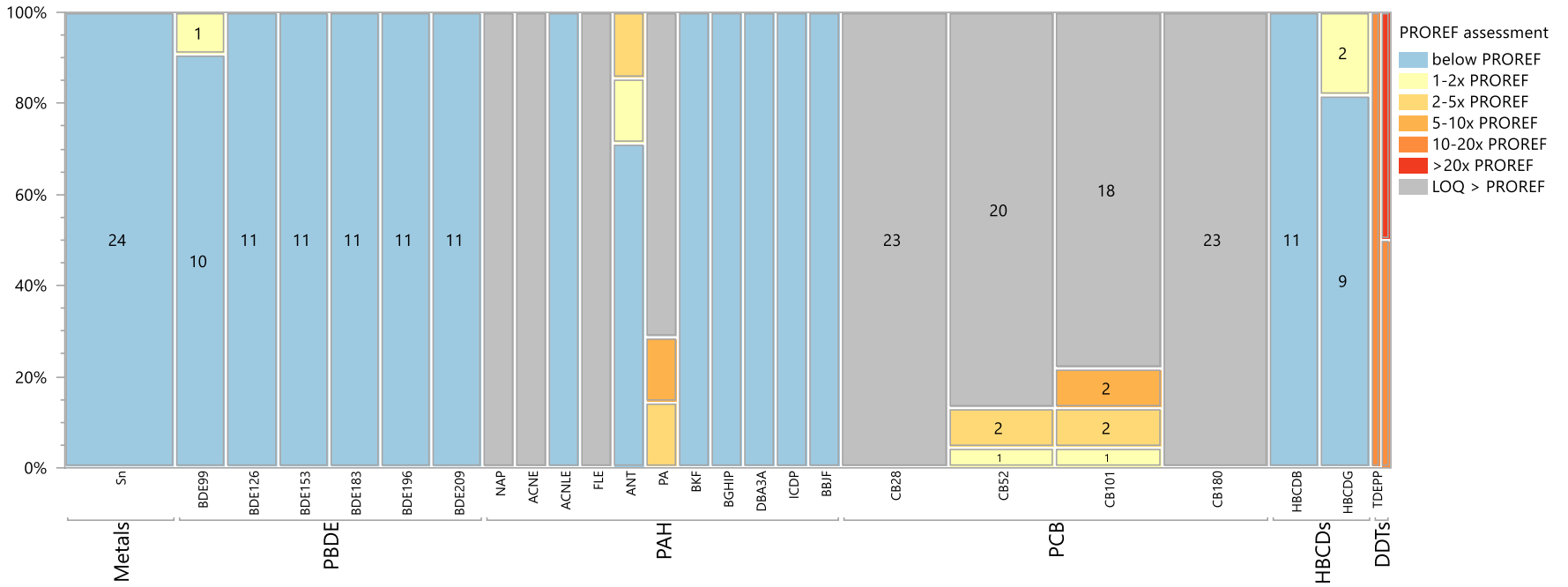

Figure S2. Exceedances of PROREF for contaminants *not selected* in 2022 in blue mussel by contaminant and group of contaminants. The cells are labelled by the number of stations sampled. The exceedances are considered by the median for each station. The colours represent below or above exceedance of PROFEF (darker yellow to red), or that the PROREF was below LOQ, and therefore could not be classified (grey).

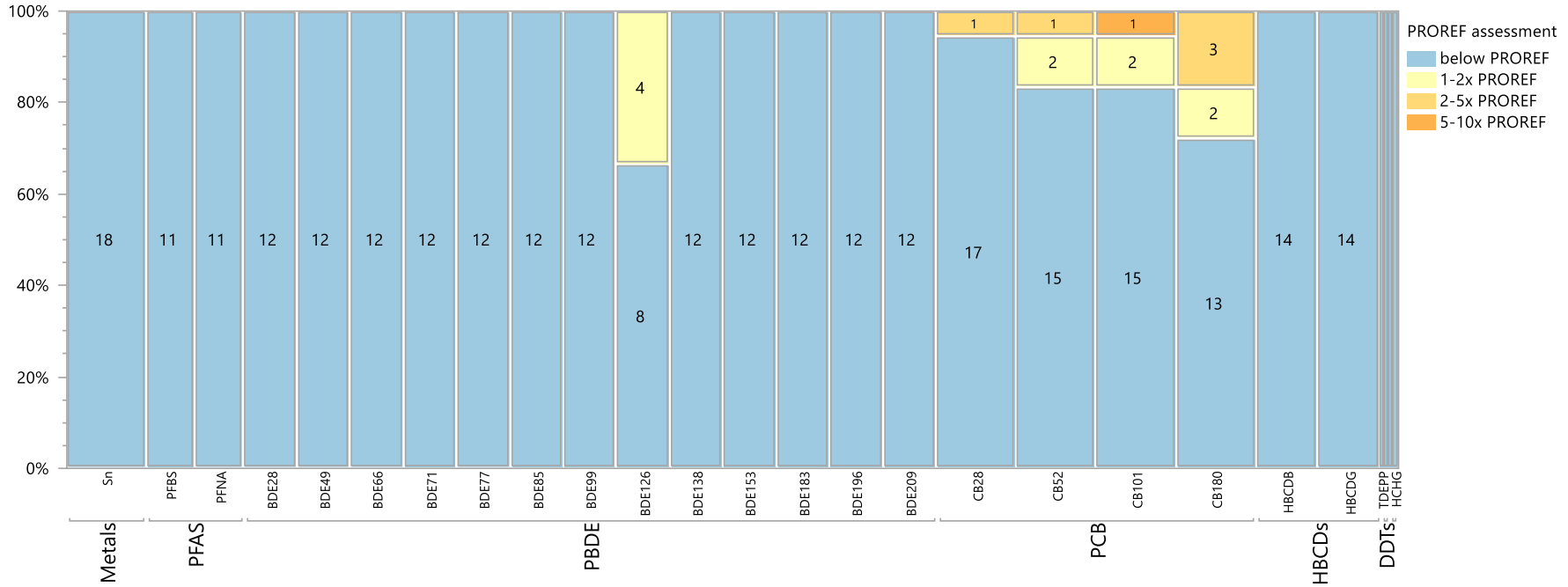

Figure S3. Exceedances of PROREF for contaminants *not selected* in 2022 in cod by parameter and group of contaminants. The cells are labelled by the number of stations sampled. The exceedances are considered by the median for each station. The colours represent below or above exceedance of PROFEF (darker yellow to red), or that the PROREF was below LOQ, and therefore could not be classified (grey).

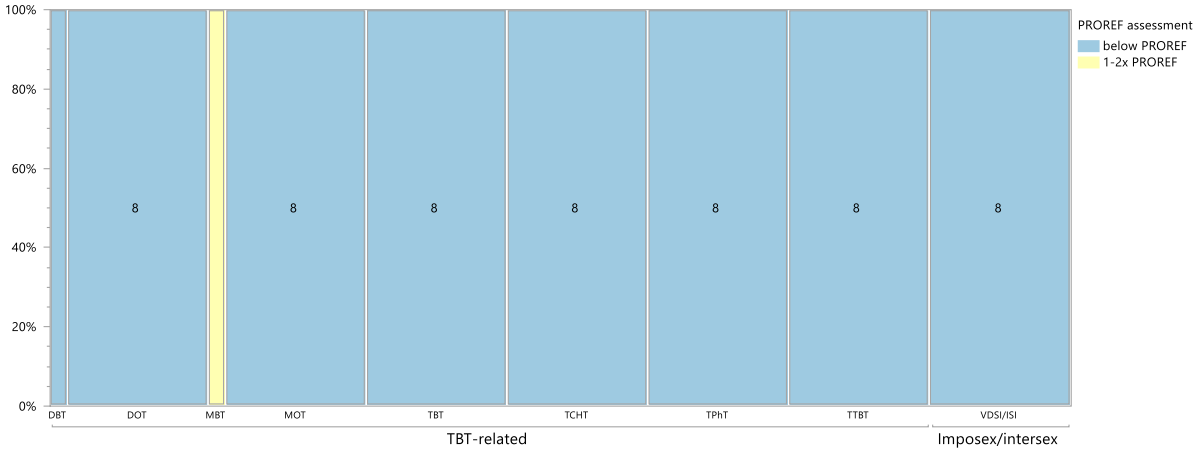

Figure S4. Exceedances of PROREF for contaminants *not selected* in 2022 in snails by parameter and group of parameters. The cells are labelled by the number of stations sampled. The exceedances are considered by the median for each station. The colours represent below or above exceedance of PROFEF (darker yellow to red), or that the PROREF was below LOQ, and therefore could not be classified (grey).

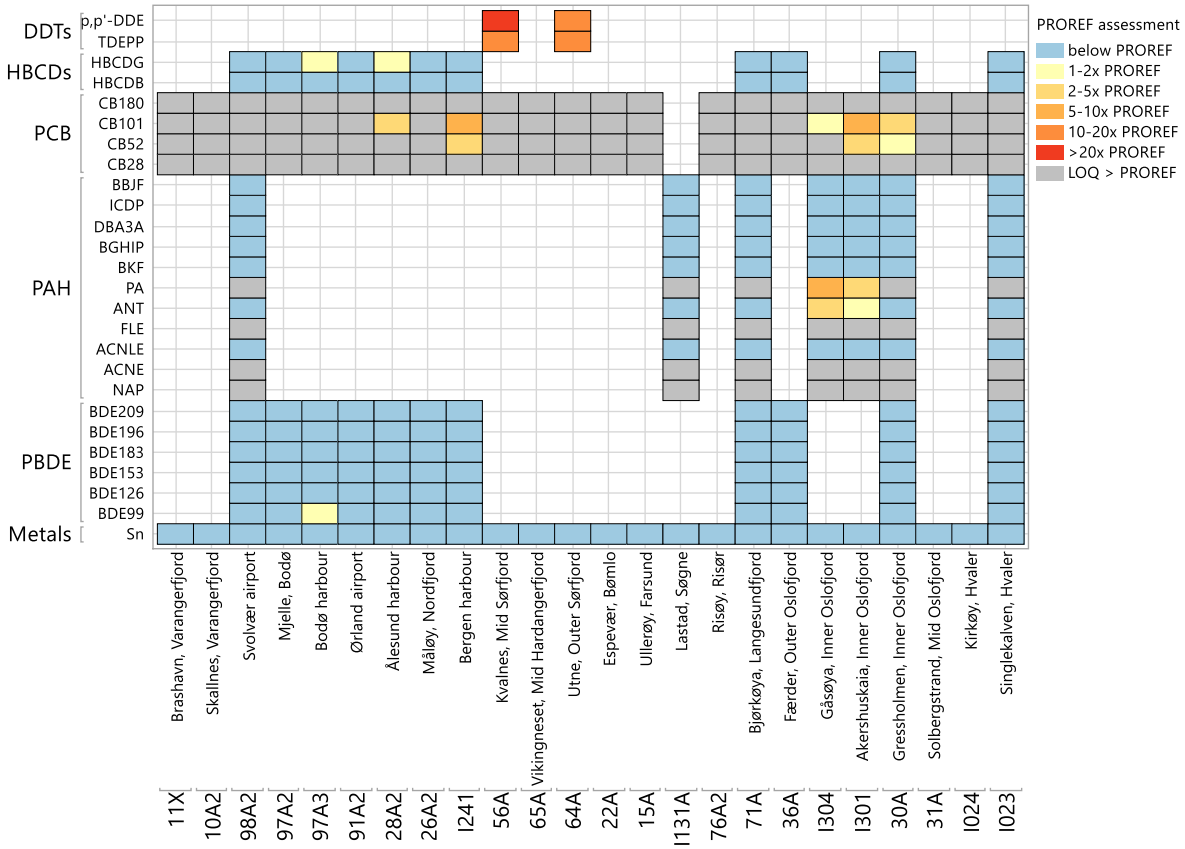

Figure S5. Heatmap of exceedances of PROREF in mussel for contaminants *not selected* in 2022. The colours represent below or above exceedance of PROREF. Empty "cells" mean that the contaminant was not analysed for at the indicated station. Grey lines show the midpoint of each station and contaminant.

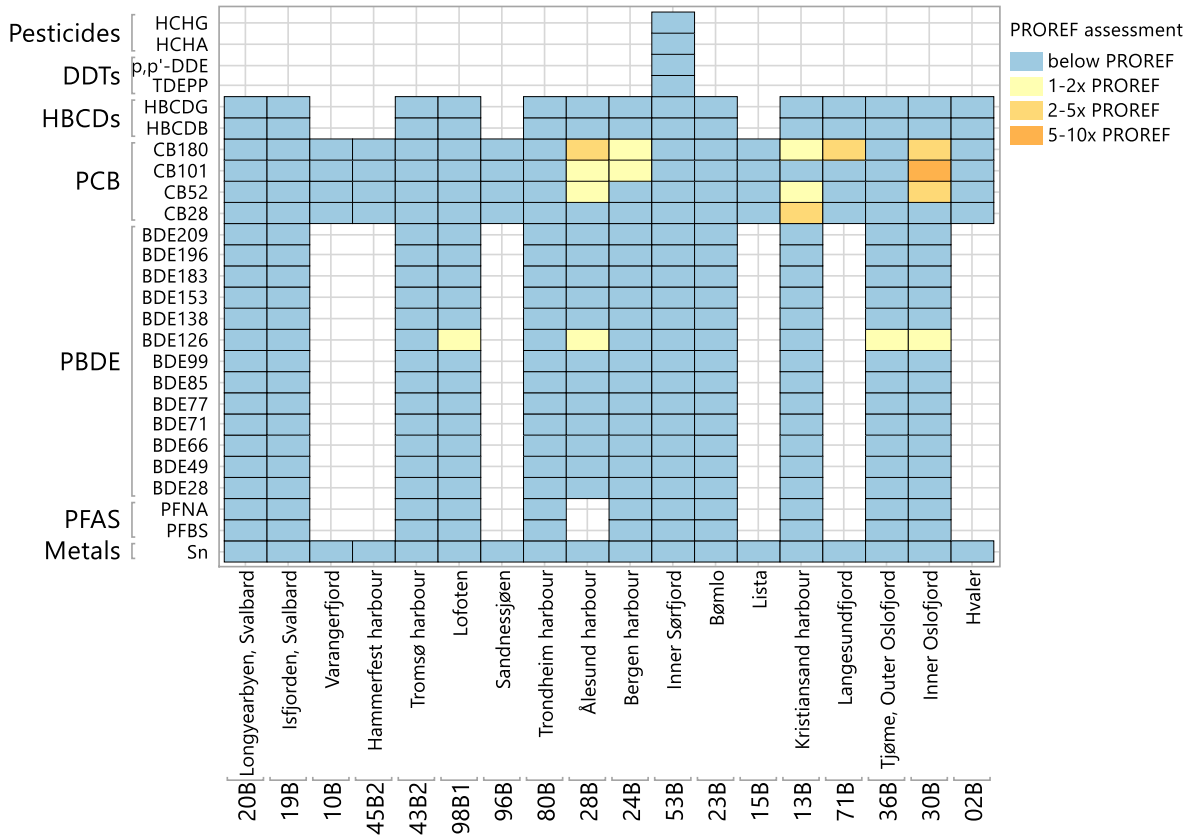

Figure S6. Heatmap of exceedances of PROREF in cod for contaminants *not selected* in 2022. The colours represent below or above exceedance of PROREF. Empty "cells" mean that the contaminant was not analysed for at the indicated station. Grey lines show the midpoint of each station and contaminant.

Time trends (combinations of contaminant × station) have also been estimated for contaminants not selected for presentation in 2022. The contaminants are listed in **Table S2**. The figures with time trends are shown in **Figure S7** to **Figure S15**. The time trends presented for selected contaminants (chapter 3.3) have been manually quality assured, but the time trends presented in this chapter has not undergone a manual quality assurance. Care must therefore be taken, especially for increasing time trends in cases where the LOQ has increased in the later years (**Figure 40**). Examples of such contaminants are silver and PCB.

Table S2. List of parameters for which a time trend is shown in supplementary data.

Contaminant group	Contaminant	Blue mussel	Cod	Snails
Metals	Sn	24	18	0
PFAS	PFBS	6	11	0
	PFDA	6	11	0
	PFDS	6	11	0
	PFHpA	6	11	0
	PFHxA	6	11	0
	PFHxS	6	11	0
	PFNA	6	11	0
	PFTrA	6	11	0
	PFUnDA	6	11	0
PBDE	LB sumPBDE6	10	12	0
	BDE28	11	12	0
	BDE49	11	12	0
	BDE66	11	12	0
	BDE71	11	12	0
	BDE77	11	12	0
	BDE85	11	12	0
	BDE99	11	12	0
	BDE119	11	12	0
	BDE126	11	12	0
	BDE138	11	12	0
	BDE153	11	12	0
	BDE183	11	12	0
	BDE196	11	12	0
	BDE209	11	12	0
	BDE156	11	12	0
	BDE17	11	12	0
	BDE184	11	12	0
	BDE191	11	12	0
	BDE197	11	12	0
	BDE206	11	12	0
	BDE207	11	12	0
PAH	NAP	7	0	0
	ACNE	7	0	0
	ACNLE	7	0	0
	FLE	7	0	0
	ANT	7	0	0
	PA	7	0	0
	C	7	0	0
	BKF	7	0	0
	BGHIP	7	0	0
	DBA3A	7	0	0
	ICDP	7	0	0
	BBJF	7	0	0
PCB	CB28	23	18	0
	CB52	23	18	0
	CB101	23	18	0
	CB180	23	18	0
	LB PCB7	23	18	0
Siloxanes	D4	0	8	0
	D6	0	8	0
HBCDs	HBCDB	11	14	0
	HBCDG	11	14	0
DDTs	TDEPP	2	0	0
	o,p'-DDD	2	0	0
	o,p'-DDE	2	0	0
	o,p'-DDT	2	0	0
	p,p'-DDE	2	1	0
Pesticides	Aldrin	2	0	0
	Dieldrin	2	0	0
	Endrin	2	0	0
	HCHA	2	0	0

Contaminant group	Contaminant	Blue mussel	Cod	Snails
Pesticides	HCHB	2	0	0
	HCHD	2	0	0
	HCHG	2	1	0
	Heptachlor	2	0	0
	Heptachlor epoxide	2	0	0
	Mirex	2	0	0
	Nonachlor, trans-	2	0	0
	Oxychlordan	2	0	0
	PROTV	0	2	0
	QCB	2	1	0
	Toksafen Parlar 26	2	0	0
	Toksafen Parlar 50	2	0	0
	Toksafen Parlar 62	2	0	0
	alfa-Chlordan (cis)	2	0	0
	gamma-chlordan (trans)	2	0	0
	trans-Heptachlor epoxide	2	0	0
TBT-related	DBT	0	0	9
	DOT	0	0	9
	MBT	0	0	9
	MOT	0	0	9
	TBT	0	0	9
	TCHT	0	0	9
	TPhT	0	0	9
	TTBT	0	0	9
ALA-D	ALAD	0	1	0
EROD	EROD	0	1	0
Imposex/intersex	VDSI/ISI	0	0	9
PAH metabolites	1-OH-naftalen	0	2	0
	2-OH-fenantren	0	2	0
	2-OH-naftalen	0	2	0
	3-OH-fenantren	0	2	0
	4-OH-fenantren	0	2	0
	BAP3OH	0	4	0
	PA1OH	0	2	0
	PYR1OH	0	2	0

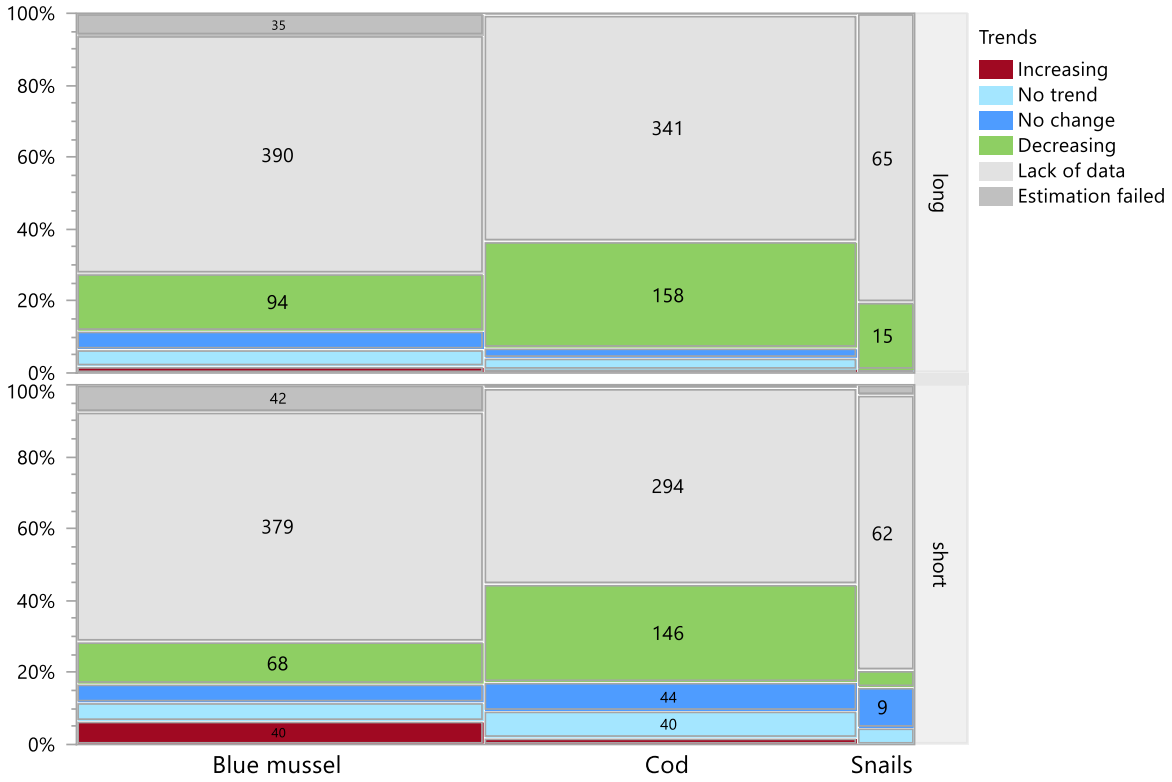

Figure S7. Mosaic plot of time trends for blue mussel, cod, and snails for contaminants *not selected* in 2022. Upper panel shows long-term trends, while lower panel shows short-term trends. The number of stations/species/tissues are indicated in the respective cells.

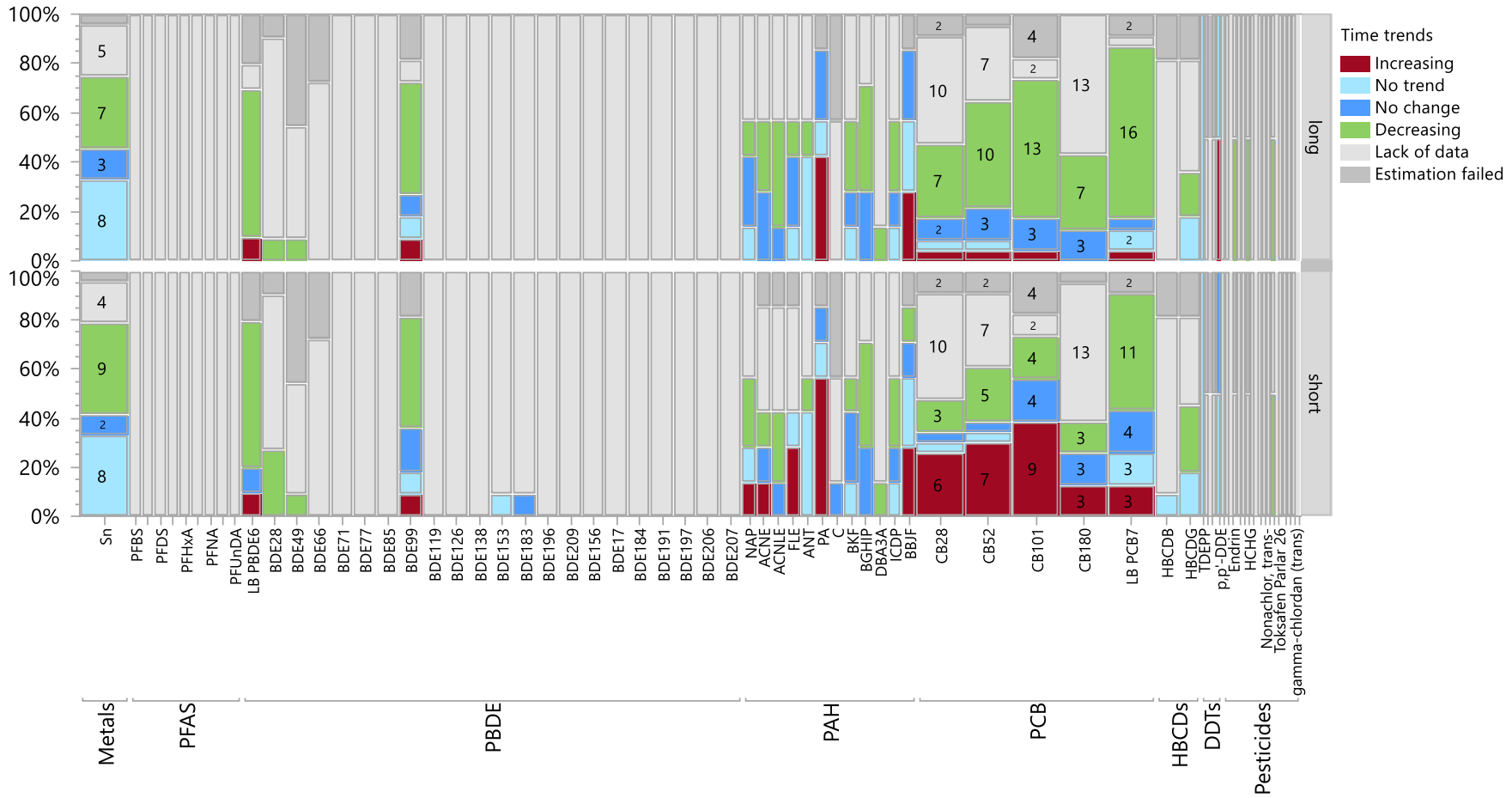

Figure S8. Time trends for blue mussel for contaminants *not selected* in 2022. Upper panel shows long-term trends, while lower panel shows short-term trends. The number of stations is indicated in the respective cells. For contaminant names not visible in this figure, please see **Figure S11** and **Figure S12**.

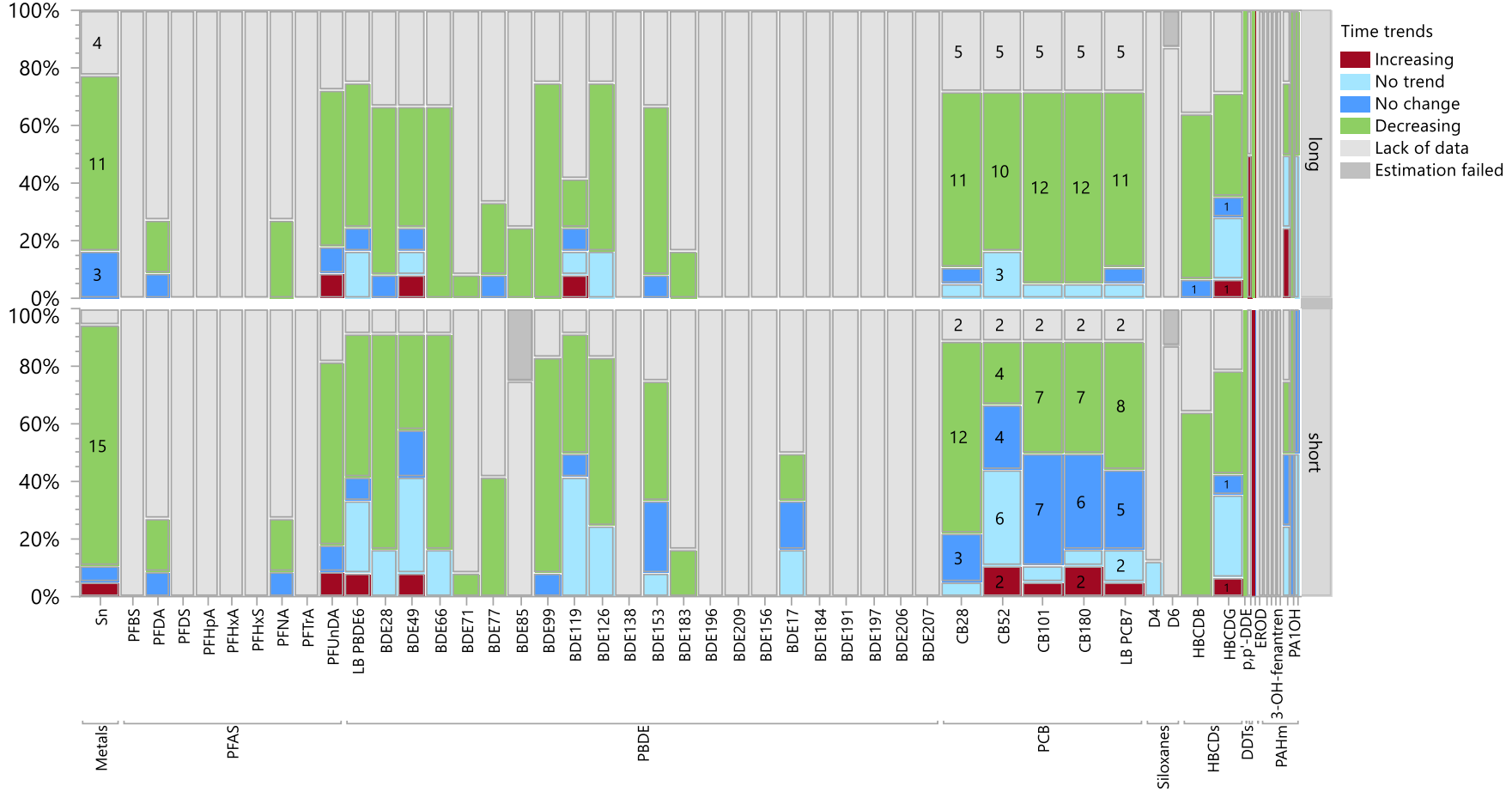

Figure S9. Time trends for cod for contaminants *not selected* in 2022. Upper panel shows long-term trends, while lower panel shows short-term trends. The number of stations is indicated in the respective cells. For contaminant names not visible in this figure, please see **Figure S13** and **Figure S14**.

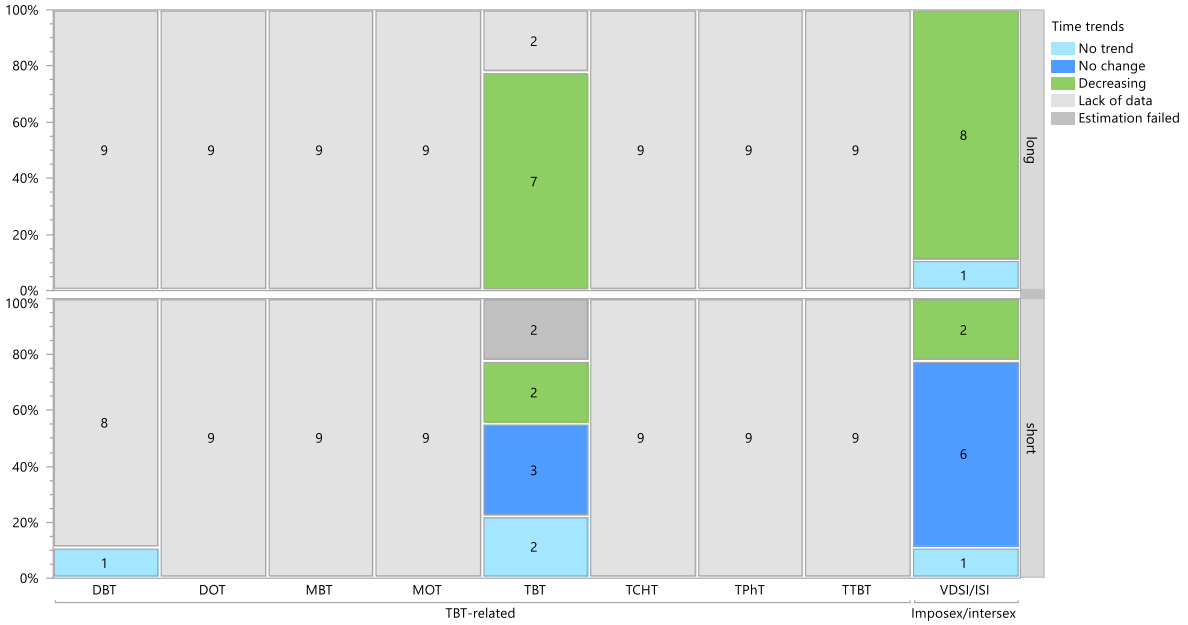

Figure S10. Time trends in snails for contaminants *not selected* in 2022. Upper panel shows long-term trends, while lower panel shows short-term trends. The number of stations is indicated in the respective cells.

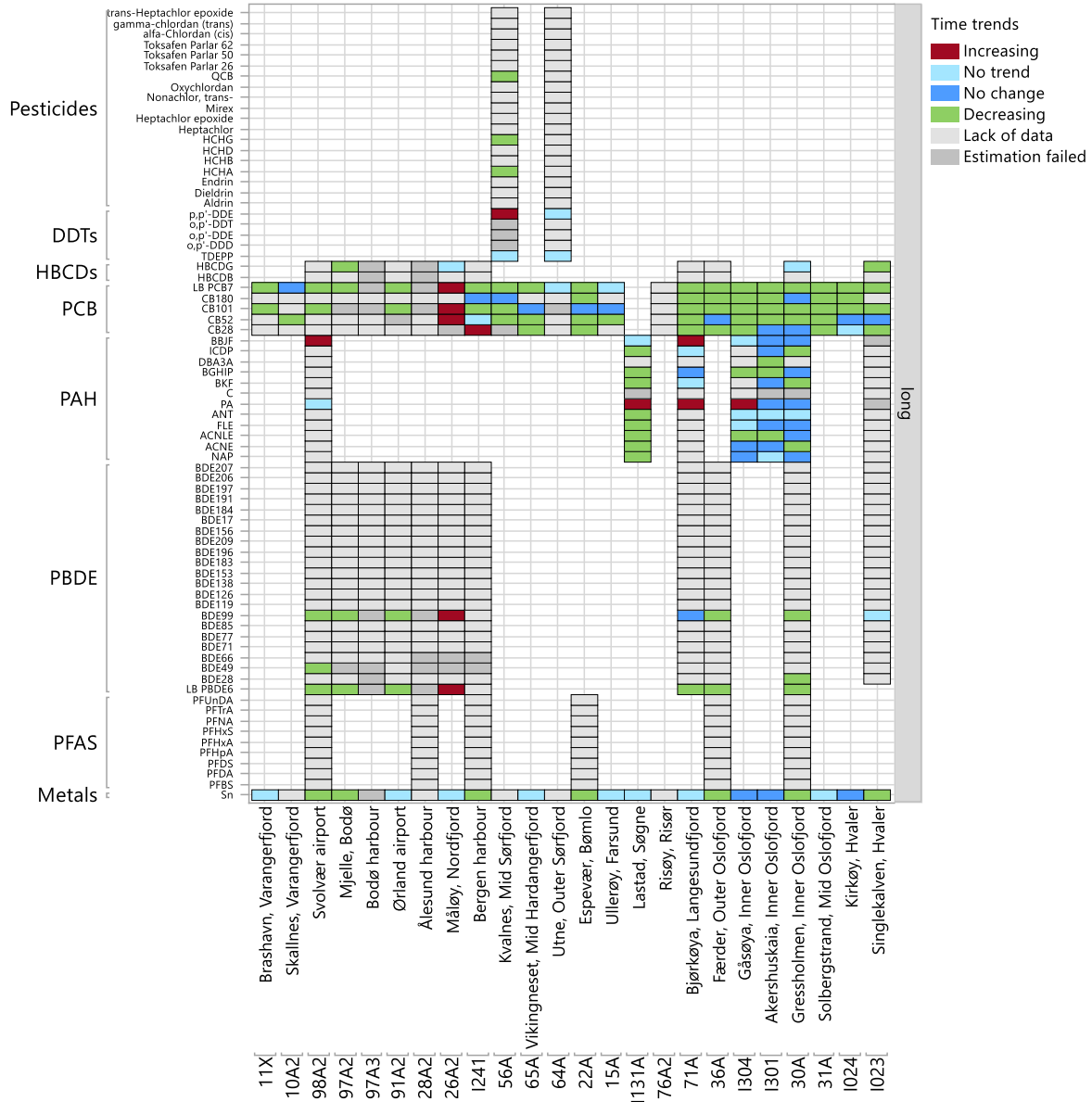

Figure S11. Heatmap for long-term time trends blue mussel for contaminants *not selected* in 2022. The colours represent time trends observed at stations. Empty "cells" mean that the contaminant was not analysed for at the indicated station. Grey lines show the midpoint of each station and contaminant.

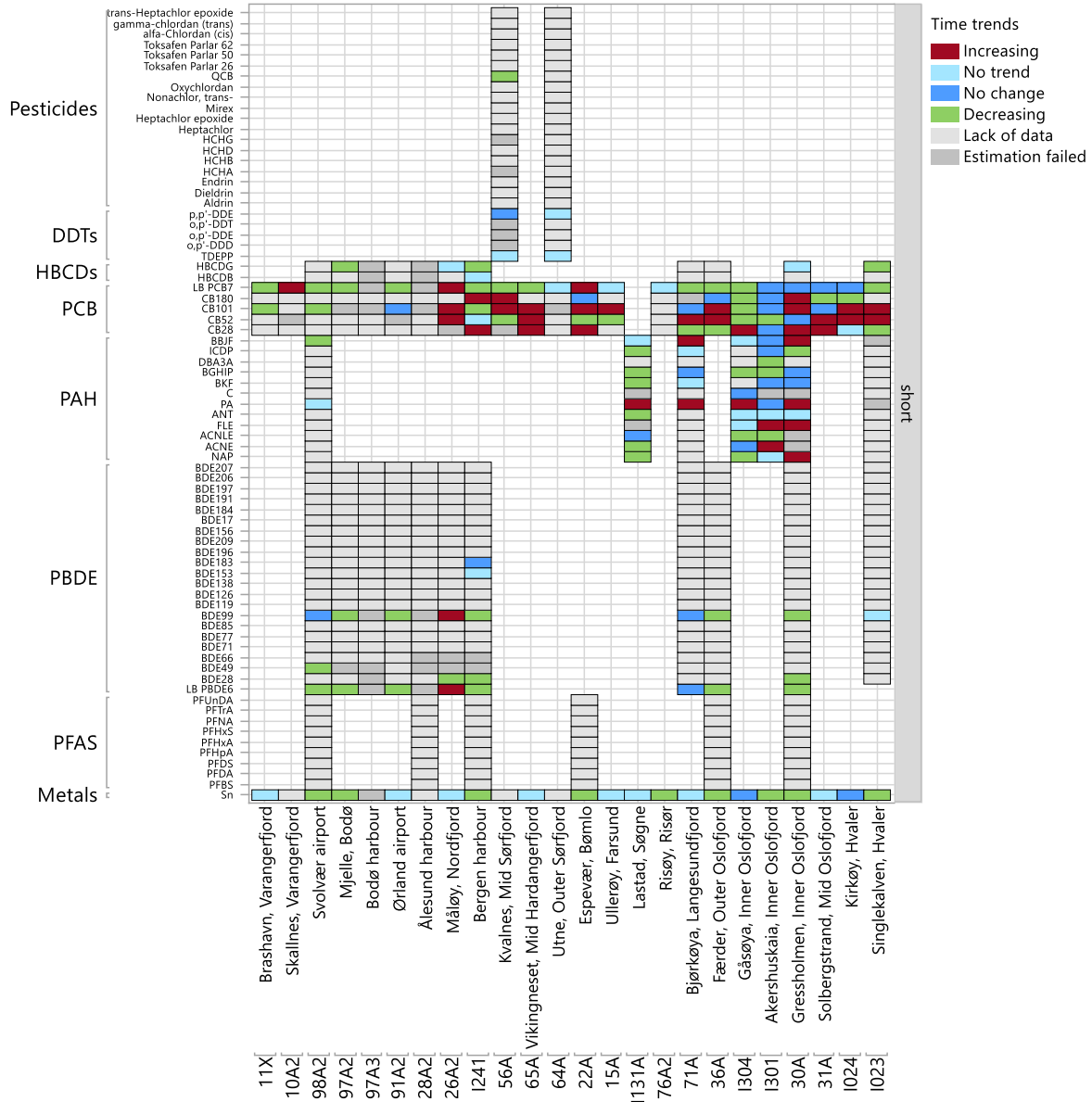

Figure S12. Heatmap for short-term time trends blue mussel for contaminants *not selected* in 2022. The colours represent time trends observed at stations. Empty "cells" mean that the contaminant was not analysed for at the indicated station. Grey lines show the midpoint of each station and contaminant.

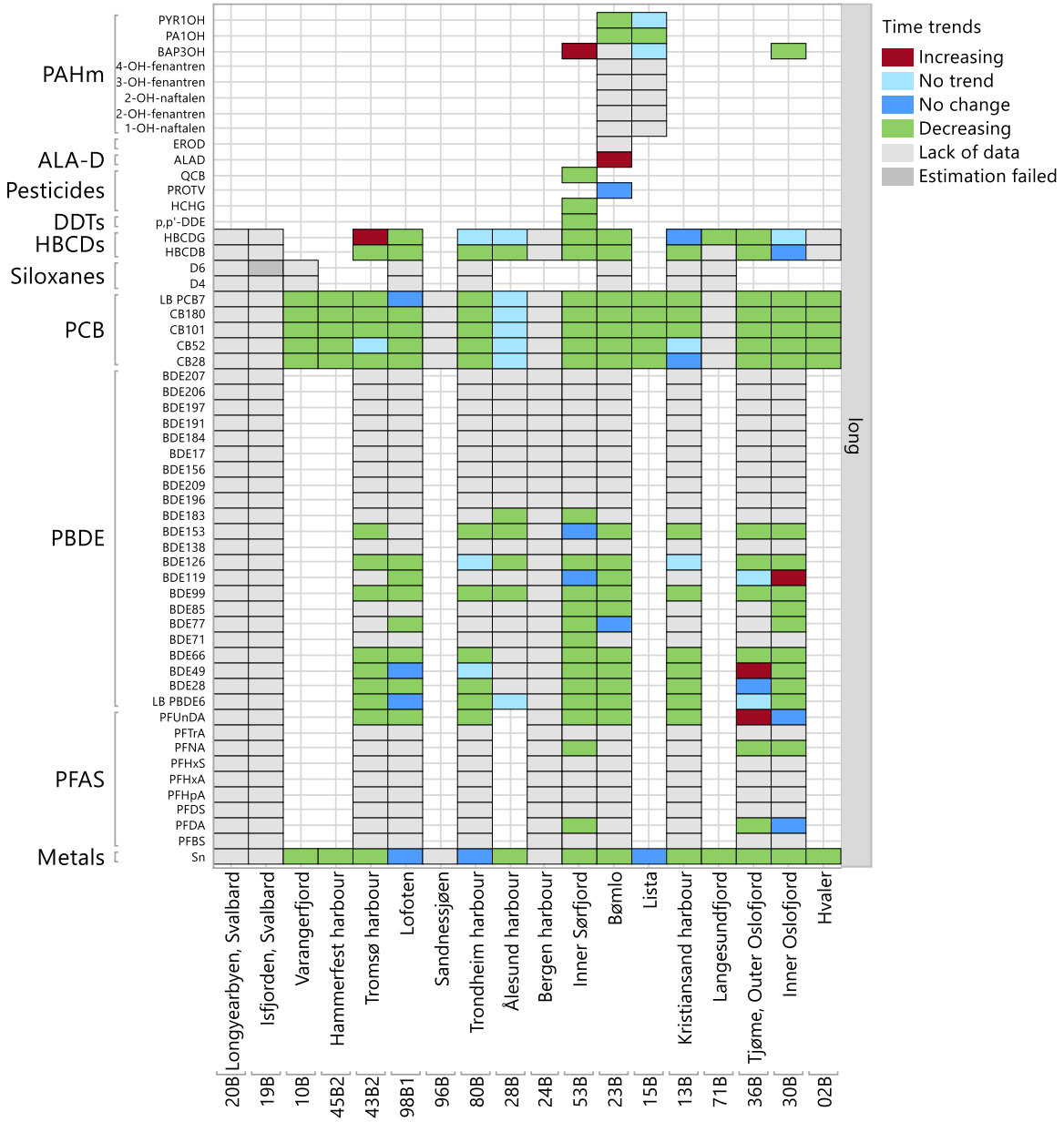

Figure S13. Heatmap for long-term time trends in cod for contaminants *not selected* in 2022. The colours represent time trends observed at stations. Empty "cells" mean that the contaminant was not analysed for at the indicated station. Grey lines show the midpoint of each station and contaminant.

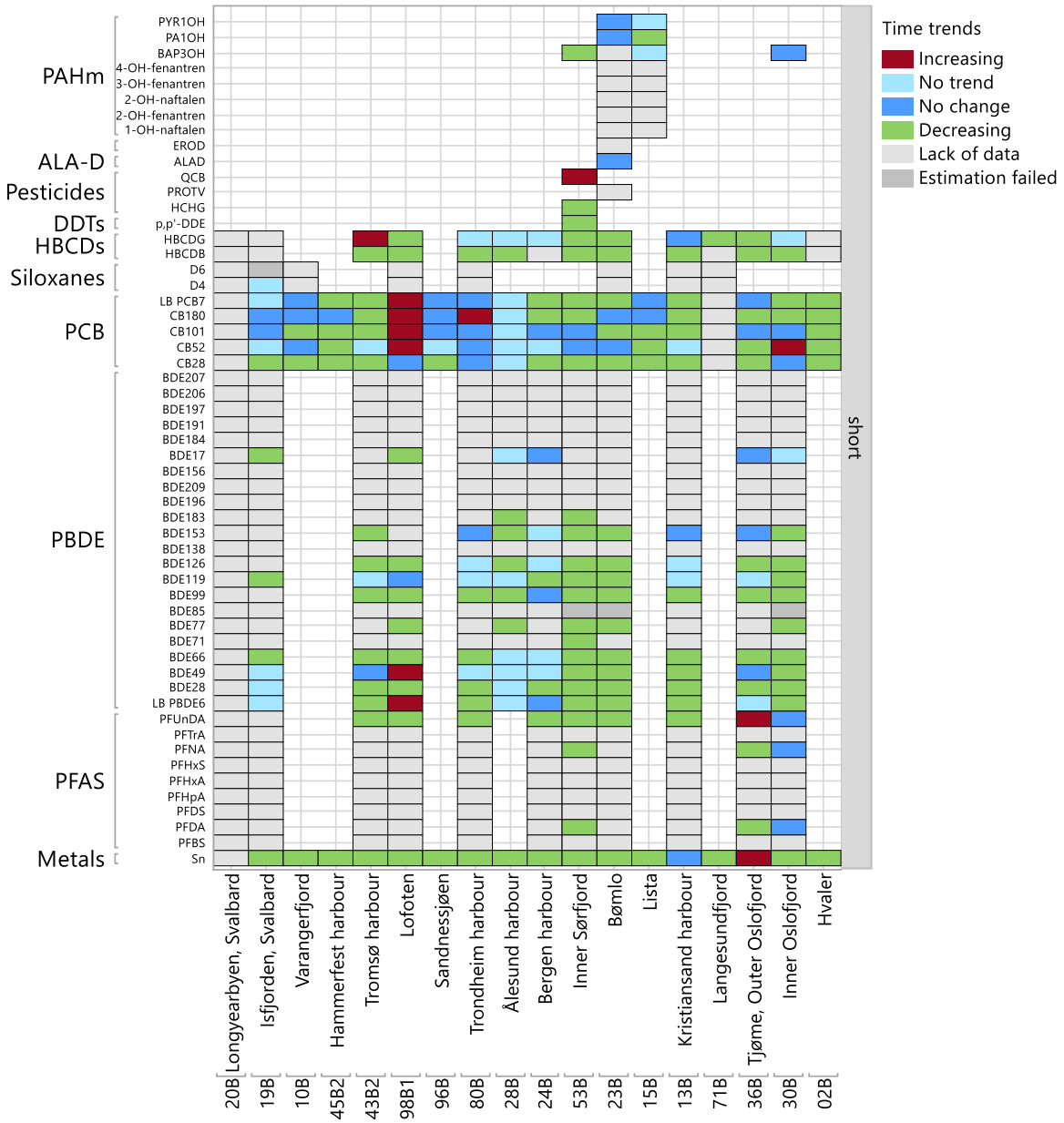

Figure S14. Heatmap for short-term time trends cod for contaminants *not selected* in 2022. The colours represent time trends observed at stations. Empty "cells" mean that the contaminant was not analysed for at the indicated station. Grey lines show the midpoint of each station and contaminant.

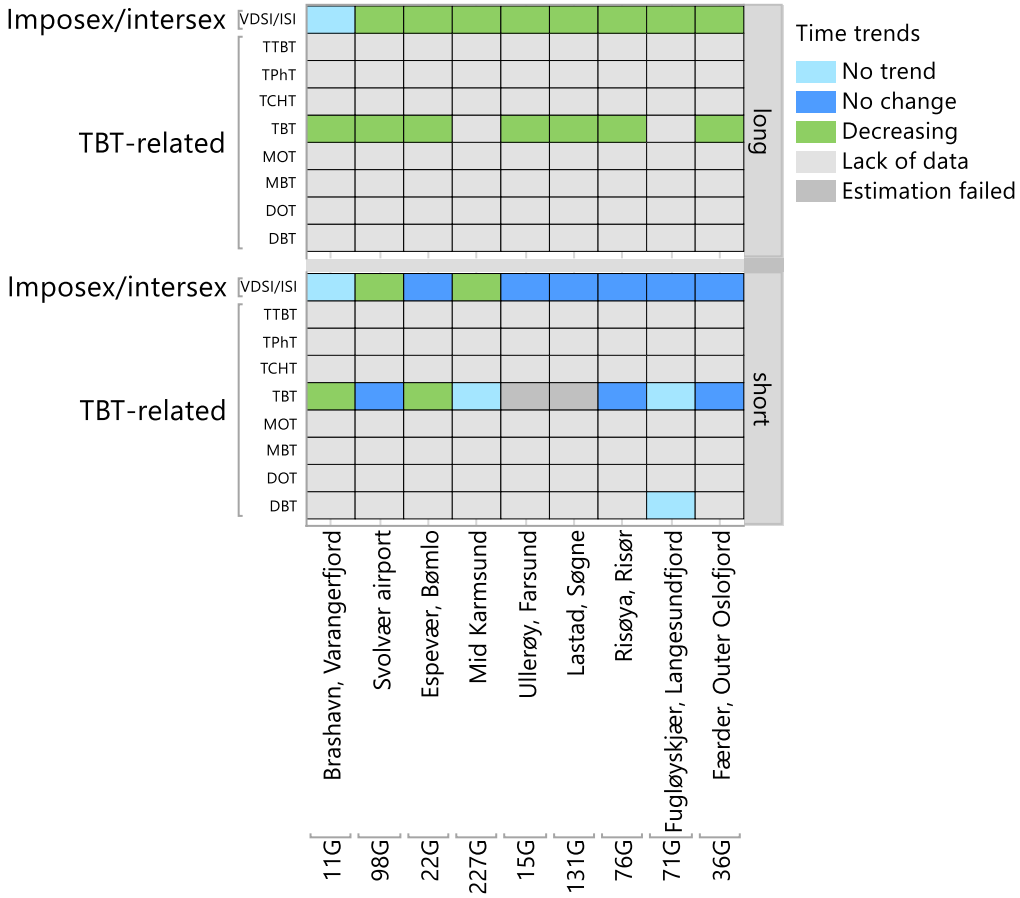

Figure S15. Heatmap for time trends in snails for contaminants *not selected* in 2022. The colours represent time trends observed at stations.

Norges ledende kompetansesenter på vannmiljø

Norsk institutt for vannforskning (NIVA) er Norges viktigste miljøforskningsinstitutt for vannfaglige spørsmål, og vi arbeider innenfor et bredt spekter av miljø, klima- og ressursspørsmål. Vår forskerkompetanse kjennetegnes av en solid faglig bredde, og spisskompetanse innen mange viktige områder. Vi kombinerer forskning, overvåkning, utredning, problemløsning og rådgivning, og arbeider på tvers av fagområder.