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Contaminants in coastal waters 2021 / Miljøgifter i kystområdene 2021

Norwegian Institute for Water Research

REPORT

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Summary

The Norwegian environmental monitoring programme “Contaminants in coastal waters” (Miljøgifter i kystområdene - MILKYS) examines levels, trends, and effects of contaminants in biota. The 2021 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, 27 contaminants and in addition, biological effect parameters were chosen for in-depth presentation. EQSs (environmental quality standard) were exceeded in samples of blue mussel (21%), cod (36%), and eider (23%). Contaminants above EQSs were sumPBDE6, mercury (Hg), sumPCB7, and fluoranthene. PROREF (Norwegian provisional high reference contaminant concentration) was exceeded for 35% in blue mussel and 11% in cod, and exceedances were higher in mussel (up to >20x PROREF) than cod (2-5x PROREF). Even though decreasing time trends dominated both long-term (>10 years) and short-term (≤ 10 years) where trends could be detected, notably increasing trends were observed which are discussed here. Notably increasing short-term trends for blue mussel were lead (Pb), chromium (Cr), arsenic (As) and some PCBs, and for cod were mercury (Hg), and silver (Ag).

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Contaminants in coastal waters 2021

Preface

The Norwegian environmental monitoring programme "Contaminants in coastal waters" (Miljøgifter i kystområdene - MILKYS) 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 2021, the first year of a new five-year period (2021-2025). The program started in 1981 and has since been continued. The 2021 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, 24 November 2022.
Merete Schøyen
Project Manager,
NIVA

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Summary

The monitoring programme "Contaminants in coastal waters" (Miljøgifter i kystområdene - MILKYS) examines the levels, trends, and effects of contaminants along the Norwegian coast from the Swedish to the Russian border, as well as on 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 2021 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 2021 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.

The main findings in 2021 can be summarized as follows:

Levels

- Most of the contaminant concentrations that could be assessed against the EQSs (Environmental Quality Standards) were below these limits. EQSs were exceeded in samples of blue mussel (21%), cod (36%), and eider (23%). Contaminants above EQSs were sumPBDE6, mercury, sumPCB7, and fluoranthene.
- Most of the contaminant concentrations that could be assessed against the PROREFs (Norwegian provisional high reference contaminant concentrations) were below these limits, and exceedances were higher in mussel (up to >20x PROREF) than cod (2-5x PROREF). The PROREFs were exceeded by a factor greater than 20 in blue mussel for the PAH compound pyrene at Akershuskaia in the Oslo harbour, for CB118 at Gressholmen in the Inner Oslofjord, and for DDT at Kvalnes in the Mid Sørfjord.

Trends

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

- In the Inner Oslofjord more contaminants have higher concentrations than in other areas along the coast and this area warrants special concern. Furthermore, in the Inner Oslofjord the investigation found a significant increasing long-term trend for mercury in cod fillet (adjusted to 50 cm length since mercury concentration is strongly linked to fish length), but for the last ten years (short-term trend) the concentrations varied, and the trend has levelled off.

Effects

- Biological effect parameters (biomarker analysis) found no effects of TBT in snails, but confirm exposure of PAH, lead and planar PCBs, PAHs, and dioxins in cod.

EQSs

A total of 315 assessments of EQSs for 20 contaminants have been evaluated in 2021. EQSs were exceeded in 21% of all contaminants in blue mussel and 36% of all contaminants in cod. No exceedances were observed for contaminants in snails. For eider, both eggs and blood were analysed, and 23% of data (samples × tissue) exceeded EQSs.

Contaminants often exceeding EQSs were mercury, 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, cod, and eider. One instance of exceeding of a PAH (fluoranthene, FLU) in blue mussel were observed.

For blue mussel, EQSs were mostly exceeded in the Inner and Outer Oslofjord, in harbour areas and in areas like the Sørfjord. For cod, EQSs were exceeded at all stations for sumPCB7 and sumPBDE6, and for almost all stations for mercury.

PROREF

A total of 724 assessments for PROREFs have been made for the 26 contaminants presented in the extended summary. The concentrations are compared to assumed reference levels, by a NIVA-developed tool denoted PROREF, which 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. Blue mussel exceeded PROREF for 35% of the samples, and 9% of the samples (35 samples) could not be classified vs. PROREF since Limit of Quantification (LOQ) was higher than PROREF. In cod, 89% of the samples were below PROREF, and the highest exceedances were lower for cod (2-5x PROREF) than for mussel (up to >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 mercury, nickel, lead, zinc, PCBs, and a PAH (pyrene, PYR). For cod, there were most exceedances for mercury, followed by silver, cadmium, and PCBs.

For metals in blue mussel, the highest exceedances of PROREF were for lead at Kvalnes in the Sørfjord and at Gressholmen in the Inner Oslofjord. PAHs and PCBs also stand out with highest exceedances of PROREF in the Inner Oslofjord at Akershuskaia and Gressholmen, and in Bergen and Ålesund. For cod, there were most exceedances of PROREF in the Inner Oslofjord an in the harbours of Bergen and Trondheim.

Time trends
A total of 801 time trends (short-term and long-term) were estimated for the contaminants presented in the extended summary. In general, there were fewer long-term trends (because data before 2012 were lacking), while no trends and decreasing trends dominated.

Long-term time trends
Long-term time trends (> 10 years) in blue mussel were dominated by decreasing trends (26%) and no trends (26%). Increasing trends were observed for 6% 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 (30%). No trends were observed for 17% of data for cod and increasing trends for 6%.

Short-term time trends
There were more datapoints (station × contaminant) that could be determined for short-term time trends (≤ 10 years) than long-term trends for both mussel and cod. No trend dominated for short-term trends for mussel (48%) while decreasing trends dominated for cod (38%). Increasing short-term trends were found both in mussels and cod (14% and 10%, respectively). No short-term trends could be determined for eider.

In blue mussel, there were found instances of increasing short-term trends for most metals and for some BDEs, but the increasing trends were dominated by some PCBs. However, some of these trends are uncertain due to few data above LOQ. For cod, there were increasing short-term trends for silver at eight stations and at four stations for mercury. Except for nickel and chromium, there were found instances of increasing short-term trends for all metals in cod. There were increasing short-term trends for BDE47 and BDE100 in Lofoten, and for PCBs in Trondheim harbour, Sandnessjøen and Lofoten.

There were found increasing short-term trends at almost all blue mussel stations. In general, the increasing short-term trends for PCBs were evenly distributed, while the increasing short-term trends for metals were concentrated in the Oslofjord. There are increasing short-term trends for PCBs in blue mussel at many stations, also in the remote areas in the Varangerfjord. Increasing short-term trends for BDEs in mussel were only found at Måløy in the Nordfjord.

Biological effects
The 2021 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 OH-pyrene bile concentrations was above the ICES/OSPAR assessment criterion (background assessment criteria, BAC) in cod from the Inner Oslofjord and Inner Sørfjord, indicating exposure to PAH-compounds.

ALA-D activity in the Inner Oslofjord appeared slightly lower than at the Bømlo reference station, however, this was not statistically significant. 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 was higher in the Inner Oslofjord, compared to the reference station at Bømlo indication exposure to planar PCBs, PAHs, and dioxins. Median EROD-activities were below the ICES/OSPAR assessment criterion (background assessment criteria, BAC) at all stations.

Sammendrag

Tittel: Miljøgifter i kystområdene 2021
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Forfatter(e): Merete Schøyen, Merete Grung, Espen Lund, Dag Ø. Hjermann, Anders Ruus, Sigurd Øxnevad, Bjørnar Beylich, Marthe T. S. Jenssen, Lise Tveiten, Jarle Håvardstun, Anne Luise Ribeiro, Isabel Doyer 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 fra svensk til russisk grense, 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 2021 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 2021 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.

Hovedfunnene i 2021 kan oppsummeres som følger:

Nivåer

- De fleste miljøgiftkonsentrasjonene som kunne vurderes i forhold til miljøkvalitetsstandarder EQSer (Environmental Quality Standards) var under disse grenseverdiene. EQS ble overskredet i prøver av blåskjell (21%), torsk (36%) og ærfugl (23%). EQS ble overskredet for sumPBDE6, kvikksølv, sumPCB7 og fluoranten.
- De fleste miljøgiftkonsentrasjonene som kunne vurderes i forhold til PROREF (norsk provisorisk høy referansekonsentrasjon for miljøgifter) var under disse grenseverdiene, og overskridelsene var høyere i blåskjell (opptil >20x PROREF) enn torsk (2-5x PROREF). PROREF ble overskredet med en faktor større enn 20 i blåskjell for PAH-forbindelsen pyren ved

Akershuskaia i Oslo havn, for CB118 ved Gressholmen i indre Oslofjord, og for DDT ved Kvalnes i midtre Sørfjord.

Trender

- Nedadgående tidstrender dominerte både på lang sikt (>10 år) og kort sikt (≤ 10 år), der tidstrender kunne påvises. I blåskjell ble oppadgående korttidstrender særlig påvist for bly, krom, arsen, og noen PCBer, og for torsk var det for kvikksølv og sølv.
- Indre Oslofjord peker seg ut som et område der flere miljøgifter har relativt høye konsentrasjoner sammenliknet med andre områder langs kysten. Dette gir grunnlag for bekymring og behov for nærmere undersøkelser. I indre Oslofjord var det en signifikant oppadgående langtidstrend for kvikksølv i torskefilét (justert til 50 cm lengde siden kvikksølvkonsentrasjon er sterkt avhengig av fiskelengde), men for de siste ti årene (korttidstrend) varierte konsentrasjonene og trenden har flatet ut.

Effekter

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

EQS

Det er gjort totalt 315 vurderinger av EQS for 20 miljøgifter i 2021. EQS ble overskredet for 21% av alle miljøgiftene i blåskjell og for 36% av miljøgiftene i torsk. Det var ingen overskridelser av EQS i snegl. Både egg og blod ble analysert i ærfugl, og 23% av dataene (prøver × vev) overskred EQS.

Miljøgifter som ofte overskrider EQS var kvikksølv, 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, torsk, og ærfugl. Ett tilfelle av overskridelse av PAH (fluoranten, FLU) i blåskjell ble observert.

For blåskjell var det stort sett overskridelser av EQS i indre og ytre Oslofjord, i havneområder og i områder som Sørfjorden. For torsk var det overskridelser av EQS på alle stasjoner for sumPCB7 og sumPBDE6, og for de fleste stasjoner for kvikksølv.

PROREF

Det er gjort totalt 724 vurderinger for PROREF for de 26 utvalgte miljøgiftene som er utvalgt for presentasjon i det utvidede sammendraget. Konsentrasjonene vurderes i forhold til antatte referansenivåer, ved et NIVA-utviklet verktøy betegnet PROREF, som er et omfattende sett med arts-vev-basis-spesifikke miljøgiftkonsentrasjoner som er statistisk lave når alle MILKYS-resultater for perioden 1991 til 2016 tas i betraktning. Dette verktøyet angir referansekonsentrasjoner for miljøgifter, hovedsakelig i områder som antas fjernt fra punktkilder, og er dermed en verdifull metode for å vurdere nivåer av miljøgifter i tillegg til de risikobaserte EQS. I blåskjell ble PROREF overskredet i 35% av prøvene, og 9% av prøvene (35 prøver) kunne ikke klassifiseres vs. PROREF, fordi kvantifiseringsgrensen (Limit of Quantification, LOQ) var høyere enn PROREF. For torsk var 89% av prøvene under PROREF, og de høyeste overskridelsene var lavere for torsk (2-5x PROREF) enn for blåskjell (opptil >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 kvikksølv, nikkel, bly, sink, PCB og én PAH-forbindelse (pyren, PYR). For torsk var det flest overskridelser for kvikksølv, etterfulgt av sølv, kadmium og PCB.

For metaller i blåskjell var de høyeste overskridelsene av PROREF for bly på Kvalnes i Sørfjorden og på Gressholmen i indre Oslofjord. PAH og PCB skiller seg også ut med høyeste overskridelser av PROREF i indre Oslofjord ved Akershuskaia og Gressholmen, og i Bergen og Ålesund. For torsk var det flest overskridelser av PROREF i indre Oslofjord og i havnene i Bergen og Trondheim.

Tidstrender
Totalt 801 tidstrender (både langtidstrender og korttidstrender) ble utregnet for miljøgifter presentert i det utvidede sammendraget. Generelt var det færre langtidstrender (fordi data før 2012 mangler), mens ingen trender eller nedadgående trender dominerte.

Langtidstrender
I blåskjell var langtidstrender (> 10 år) hovedsakelig nedadgående (26%) og av ingen trender (26%). Oppadgående trender ble observert for 6% 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, men prosentandelen av nedadgående trender var høyere (30%). For torsk ble ingen trend observert for 17% av dataene, mens for 6% var det oppadgående trender.

Korttidstrender
For både blåskjell og torsk var det flere datapunkter (stasjon x miljøgift) hvor det kunne utregnes korttidstrender (≤ 10 år) enn langtidstrender. For korttidstrender var det ingen trend (48%) som dominerte for blåskjell, mens det var nedadgående trender (38%) som dominerte for torsk. Det ble påvist oppadgående korttidstrender i både blåskjell (14%) og torsk (10%). Det ble ikke påvist korttidstrender for ærfugl.

For blåskjell ble det funnet tilfeller av oppadgående korttidstrender for de fleste metaller og for noen BDEer, men de oppadgående trendene var dominert av noen PCBer. Noen av disse trendene er imidlertid usikre på grunn av få data over LOQ. For torsk var det oppadgående korttidstrender for sølv på åtte stasjoner og for kvikksølv på fire stasjoner. Unntatt for nikkel og krom, ble det funnet tilfeller av økende korttidstrender for alle metaller i torsk. Det var oppadgående korttidstrender for BDE47 og BDE100 i Lofoten, og for PCBer i Trondheim havn, Sandnessjøen og Lofoten.

Det ble funnet oppadgående korttidstrender ved nesten alle blåskjellstasjonene. Generelt var de oppadgående korttidstrendene for PCB jevnt fordelt, mens de oppadgående korttidstrendene for metaller var konsentrert i Oslofjorden. For blåskjell var det oppadgående korttidstrender for PCB på mange stasjoner, også i de fjerntliggende områdene i Varangerfjorden. Det ble kun påvist oppadgående korttidstrender for BDEer i blåskjell ved Måløy i Nordfjorden.

Biologiske effekter
2021-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) for OH-pyren i torskegalle ble overskredet i indre Oslofjord og indre Sørfjorden. Dette viser at fisken har vært eksponert for PAH.

ALA-D aktivitet i torsk fra indre Oslofjord virket lavere enn i torsk fra Bømlo, men dette var ikke statistisk signifikant. 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 fra indre Oslofjord var høyere enn i torsk fra referansestasjonen på Bømlo som indikerer eksponering for plane PCBer, PAHer, og dioksiner. EROD-aktiviteten 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).

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.

MILKYS also contributes data to support the implementation of the Water Framework Directive (WFD) (EU 2000) and its daughter directive the Environmental Quality Standards Directive (EQSD) (EU 2013) to achieve good chemical status by assessing the results using EU EQSD in Norway. In this

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

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 2021, the first 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 this short report for the 2021-investigation. More complementary information regarding previous programs, such as background history, abbreviations for contaminants, maps etc., can be found in (Schøyen et al. 2021).

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

2 Extended summary of MILKYS 2021

2.1 Samples, localities and chemical analyses

Overview of the contaminants selected for presentation of results in extended summary are listed in **Table 1**. The contaminants were selected with an expectation that these contaminants represent the contaminant group. More metals have been chosen for selection this year than previous years. This extended summary presents the main results, while more details data can be found in **Chapter 3.2** (factsheets for selected contaminants). 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 reportet to Vannmiljø, ICES and OSPAR. Location of stations sampled in MILKYS 2021 are shown in **Figure 1** and number of samples at each station are listed in **Table 2**.

2.1.1 Samples and locations

Location of stations sampled in MILKYS 2021 are shown in **Figure 1** and number of samples at each station are listed in **Table 2**.

Table 1. List of parameters that will be shown in more detail in the report, which species (blue mussel (*Mytilus edulis*), cod (*Gadus morhua*, and eider (*Somateria mollissima*) they are analysed in and how many stations are measured for each species. Eider numbers are for tissues (blood and eggs) – not number of stations.

Contaminant group	Contaminant	*Blue mussel*	*Cod*	*Eider*	Number of stations
Metals	Silver (Ag)	24	18	2	44
	Arsenic (As)	24	18	2	44
	Cadmium (Cd)	24	18	2	44
	Cobalt (Co)	24	18	2	44
	Chromium (Cr)	24	18	2	44
	Copper (Cu)	24	18	2	44
	Mercury (Hg)	24	18	2	44
	Nickel (Ni)	24	18	2	44
	Lead (Pb)	24	18	2	44
	Zinc (Zn)	24	18	2	44
PFAS	Perfluorooctanoic acid (PFOA)	6	11	2	19
	Perfluorooctanesulfonic acid (PFOS)	6	11	2	19
	Perfluorooctanesulfonamide (PFOSA)	6	11	2	19
PBDEs	PBDE congener 47 (BDE47)	11	12	2	25
	PBDE congener 99 (BDE99)	11	12	2	25
	PBDE congener 100 (BDE100)	11	12	2	25
	PBDE congener 153 (BDE153)	11	12	2	25
PCBs	PCB congener 118 (CB118)	23	18	2	43
	PCB congener 138 (CB138)	23	18	2	43
	PCB congener 153 (CB153)	23	18	2	43
PAHs	Benzo(a)anthracene (BAA)	7	0	0	7
	Benzo(a)pyrene BAP)	7	0	0	7
	Fluoranthene (FLU)	7	0	0	7
	Pyrene (PYR)	7	0	0	7
Siloxanes	Decamethylcyclopentasiloxane (D5)	0	13	2	15
HBCD	α-hexabromocyclododecane (HBCDA)	11	14	2	27
Pesticides	Hexachlorobenzene (HCB)	2	1	2	5
All	**All**	**412**	**343**	**46**	**801**

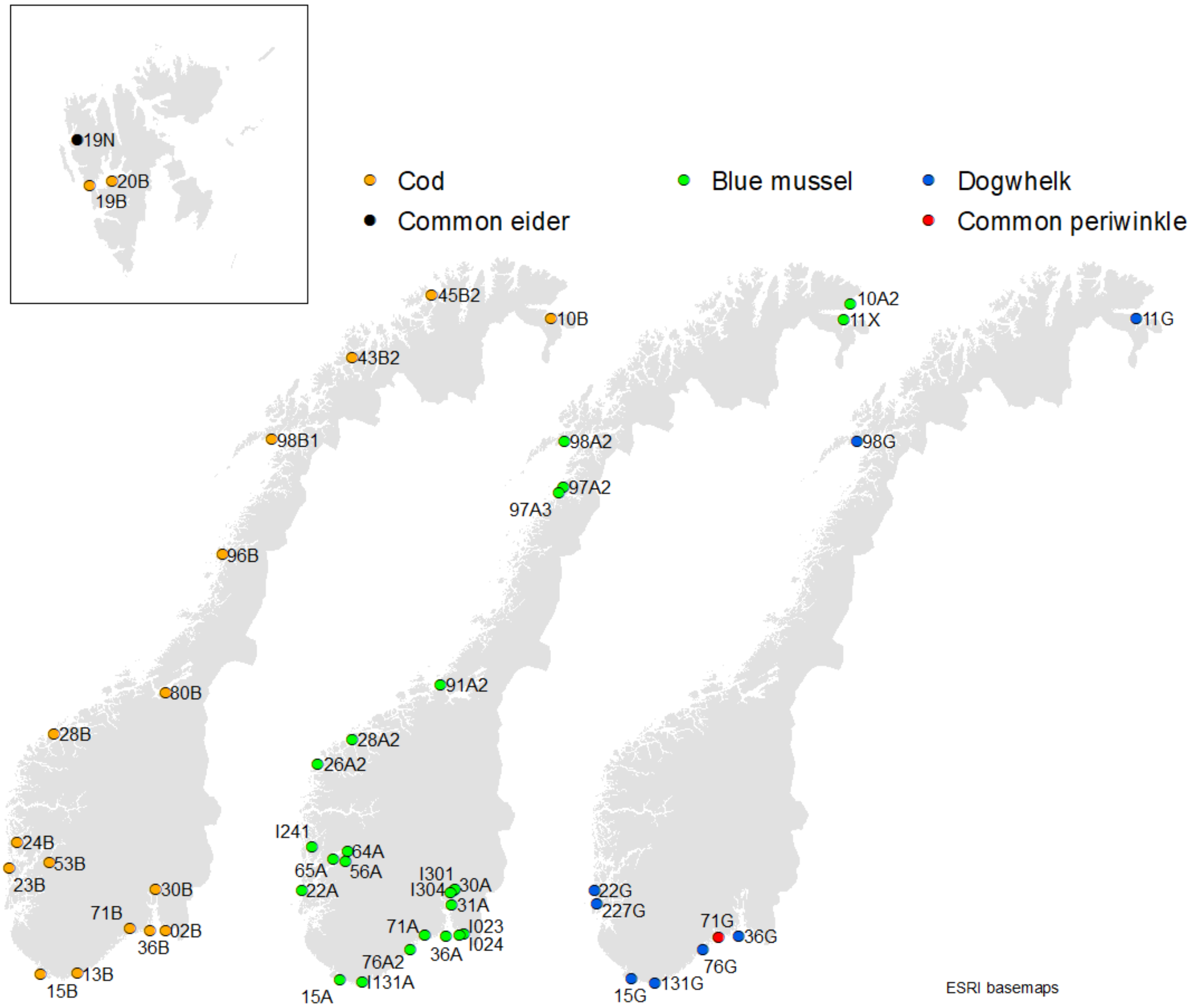


Figure 1. Stations where cod *(Gadus morhua*) and common eider (*Somateria mollissima*) (left), blue mussel (*Mytilus edulis*) (middle), dogwhelk (*Nucella lapillus*) and common periwinkle (*Littorina littorea*) (right) were sampled in Norway and Svalbard (inset) in 2021.

Table 2. 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 2021. All snail samples were pooled. The stations are ordered along the coastline starting north moving south.

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						1
	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				15	15	
	10B	Varangerfjord	69.8162	29.7602				12	15	
	45B2	Hammerfest harbour	70.65	23.6333				15	15	
	43B2	Tromsø harbour	69.653	18.974				15	15	
	98B1	Lofoten	68.1858	14.7081				14	15	
	96B	Sandnessjøen	66.0444	12.5036				15	15	
	80B	Trondheim harbour	63.4456	10.3717				3	3	
	28B	Ålesund harbour	62.4678	6.0686				15	15	
	24B	Bergen harbour	60.3966	5.2707				15	15	
	53B	Inner Sørfjord	60.0973	6.5397	15	15		15	15	
	23B	Bømlo	59.8956	5.1086	16	16		16	15	
	15B	Lista	58.0514	6.7469	15			15	15	
	13B	Kristiansand harbour	58.1328	7.9885				6	15	
	71B	Langesundfjord	59.0465	9.7028				15	15	
	36B	Tjøme, Outer Oslofjord	59.0405	10.4358				10	15	
	30B	Inner Oslofjord	59.8127	10.5518	14	15		15	15	
	02B	Hvaler	59.0648	10.9735				6	7	
Eider	19N	Kongsfjorden, Svalbard	79.004	12.11		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

2.1.2 Detection frequencies of contaminants and history of 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 Eurofins Moss. However, the methods were mainly the same, and only minor changes of the LOQ occurred. For the program period (starting 2017) the organic pollutants were analysed at Eurofins GFA, leading to discrepancies in both methods employed and LOQs. For PCBs, the LOQs were increased somewhat (except CB118 which was lowered). For BDE, the LOQs were mainly lowered somewhat, while for PAHs they were increased for some of the congeners. The metal analyses were changed in 2019 where most LOQs were lowered, while for Ag the LOQ was increased.

Figure 2 gives the proportion over LOQ (detection frequency, in %) of the various compounds for each tissue and **Figure 3** gives the observed LOQ (median values) in blue mussel in the various compounds since 2001.

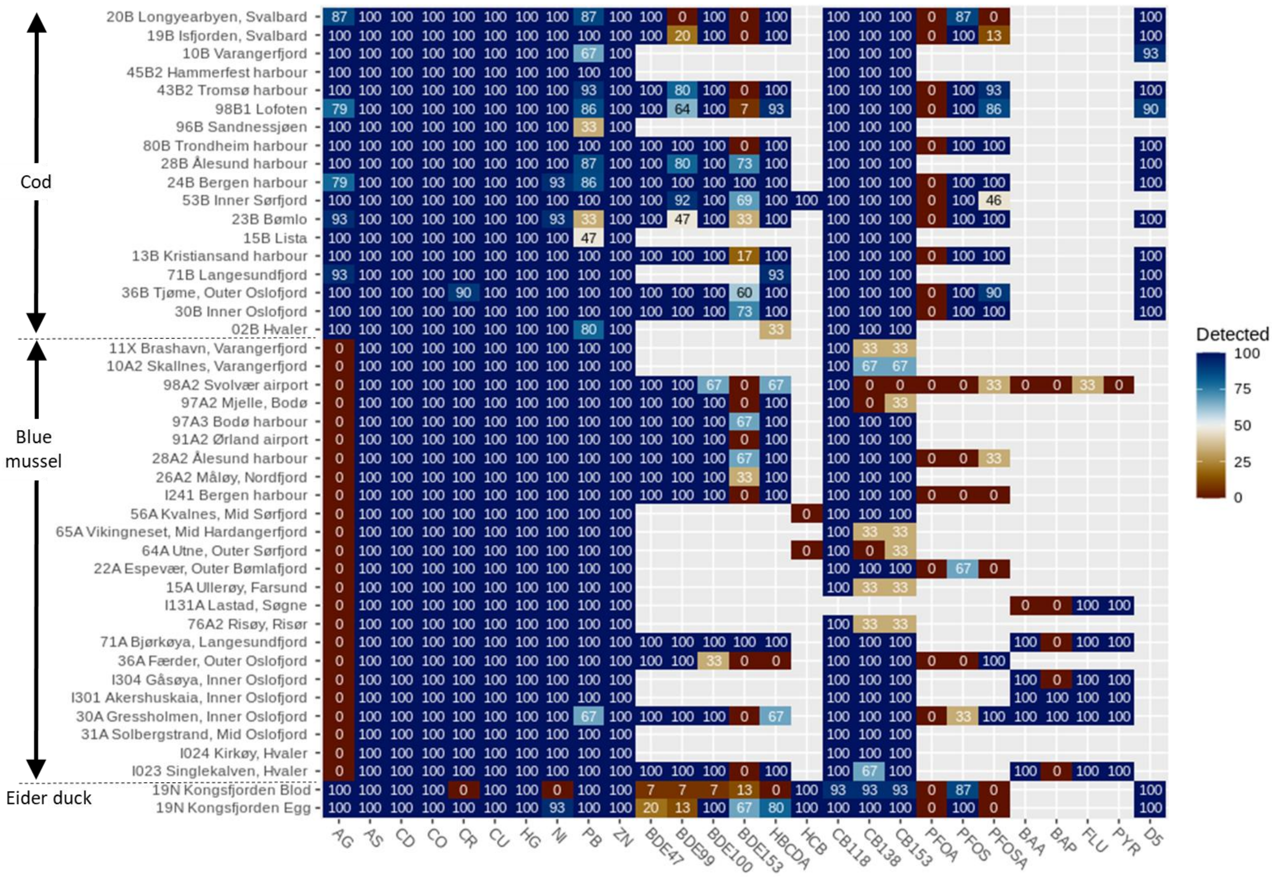


Figure 2. Proportion over LOQ (detection frequency, in %) of the compounds for each tissue. The stations are ordered along the coastline starting north moving south.

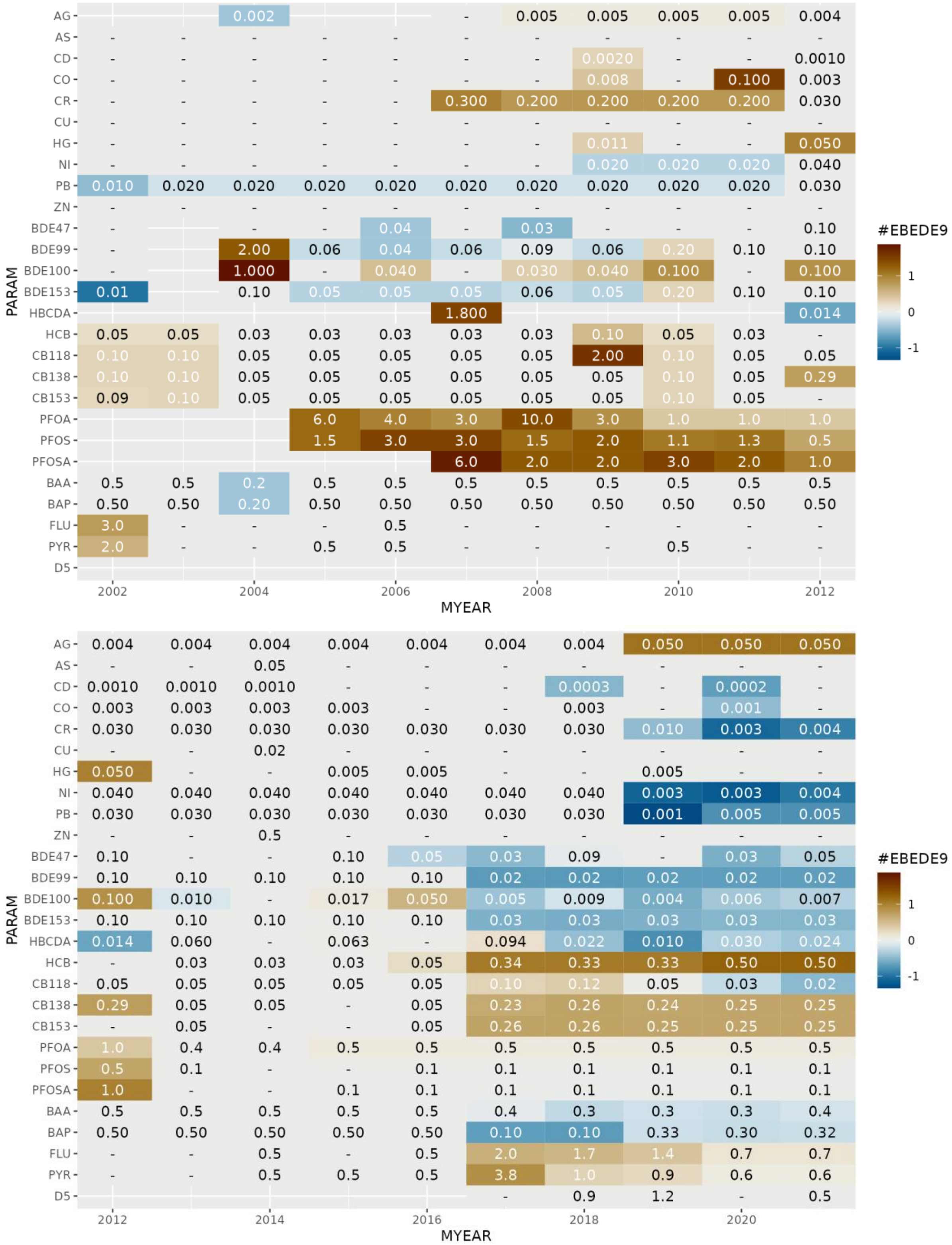


Figure 3. Observed LOQ (median values) in blue mussel of the various compounds since 2001. The colors are LOQ versus the median LOQ during the years 2013-2015, with blue colors showing improvement (lower LOQ) and brown colors showing increased LOQ. For some groups, e.g. PFAS, there were no measurements in these years (shown in light grey).

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

3.1 Exceedances 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 that the organism was analysed in (see **Table 2**). 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, 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.

For mercury in cod, risk assessments vs. EQS and PROREF are done by using the concentrations measured directly. For the tile 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.

3.1.1 EQS

Assessment of exceedances of concentrations of contaminants for which an EQS exist have been done. The contaminants listed in **Table 3** have been determined in 2021, have an EQS in biota (Direktoratsgruppen vanndirektivet 2018) and are therefore subject to assessment. A total of 315 assessments of EQSs have been done in 2021 (combination of contaminant × station × tissue (tissue is only relevant for samples of eider (eggs and blood)). Twenty contaminants determined in 2021 had EQSs (**Table 3**).

In MILKYS, exceedances of EQSs are considered by the *median* concentration for each station. The four species groups blue mussel, cod, snails, and eider were analysed for contaminants with an assigned EQS, and exceedances are shown in **Figure 4**. EQS were exceeded in 21% of all selected contaminants in blue mussel and 36% of all contaminants in cod. No exceedances were observed for contaminants determined in snails. For eider, both eggs and blood were analysed, and 23% of data (contaminants × tissues) exceeded in eider.

Table 3. List of contaminants determined in 2021 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 contaminant (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

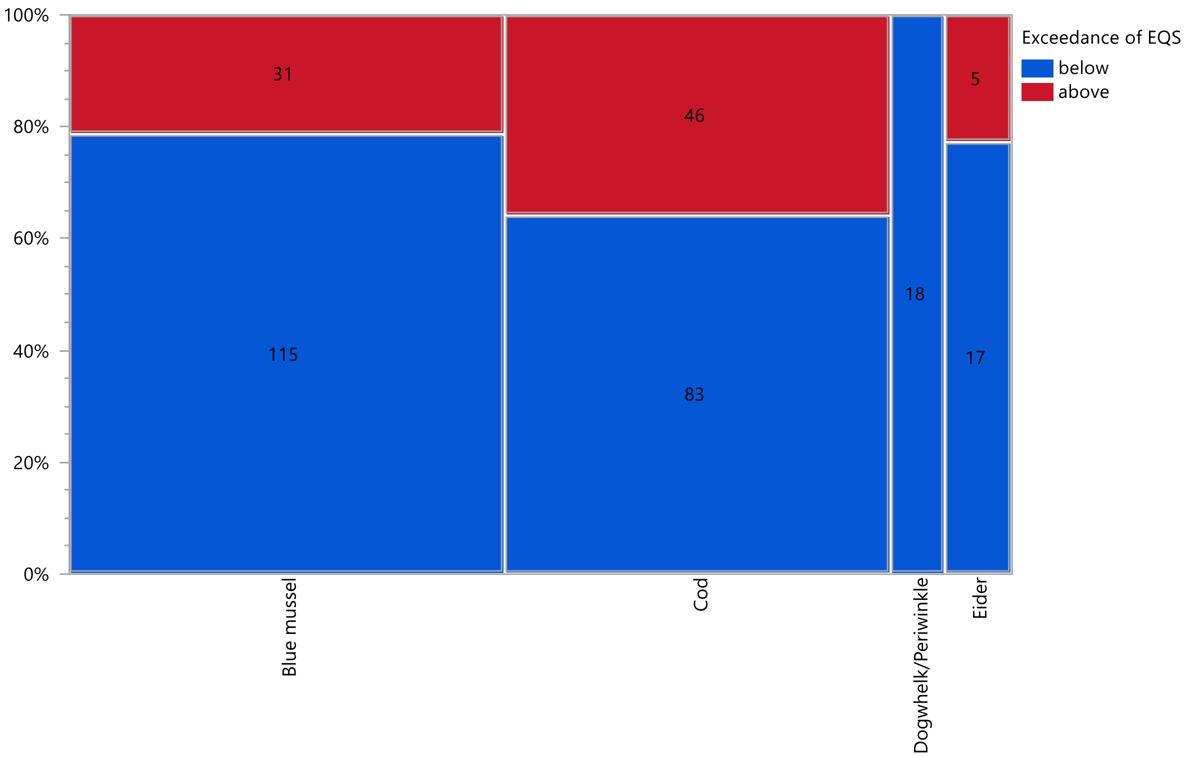


Figure 4. Exceedances of EQSs by species groups in a mosaic plot. The cells are labelled by the number of datapoints (stations × contaminants (for eider one station × 2 tissues)). The exceedances are considered by the median for each station and species. The colours represent below or above EQSs. The total area of the figures represents the 315 assessments of EQS.

To illustrate which contaminants that had concentrations exceeding EQS, **Figure 5** to **Figure 7** to illustrate this for blue mussel, cod, and snails, respectively. Furthermore, heatmaps of concentration exceedances at individual station and contaminant are shown in **Figure 8** to **Figure 10** for blue mussel, cod, and eider, respectively.

In blue mussel, compounds with concentrations exceeding EQS were mercury (at 3 stations, 13%), sumPBDE6 (all stations exceeded EQS), sumPCB7 (16 stations (70%) and fluoranthene (FLU, at 1 station, 14%). The EQS for sumPBDE6 is very low to protect human health (European Commission 2014).

In cod, 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. 2018; Moe et al. 2019; Thrane et al. 2020; Sandin et al. 2021). Only two stations (11%) did not exceed EQS for mercury.

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

Both blood and eggs were analysed for contaminants in eider. Mercury concentrations exceed EQSs in both tissues, while sumPBDE6 and sumPCB7 concentrations were exceeded in eider eggs (**Figure 10**).

Contaminants often exceeding EQSs are therefore mercury, sumPBDE6 and sumPCB7 for blue mussel, cod, and eider. One instance of exceeding of a PAH (FLU) in blue mussel were observed.

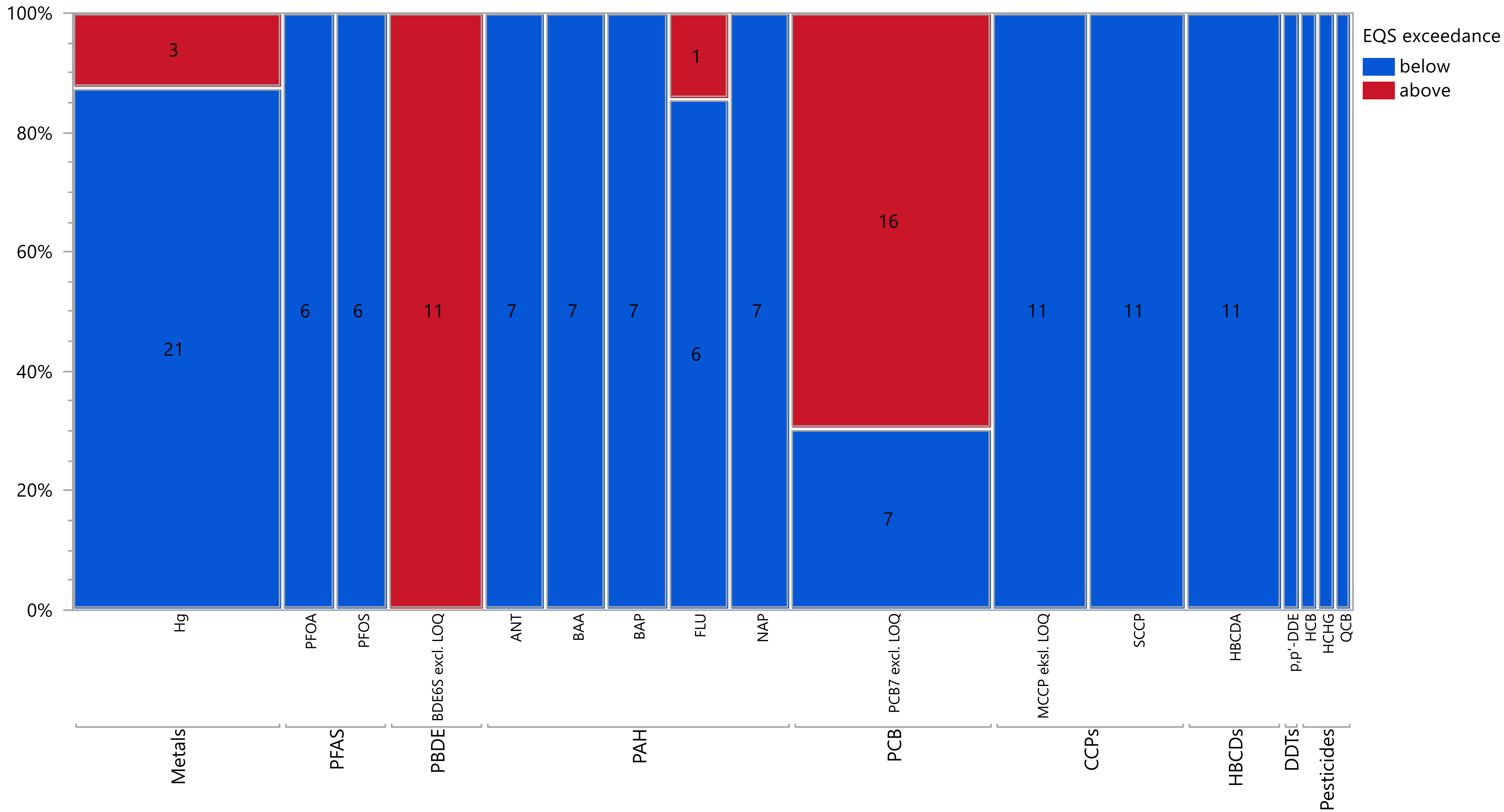


Figure 5. Exceedances of EQSs in blue mussel by contaminant and contaminant group. The cells are labelled by the number of stations in each category. The exceedances are considered by the median for each station. The colours represent below or above EQSs.

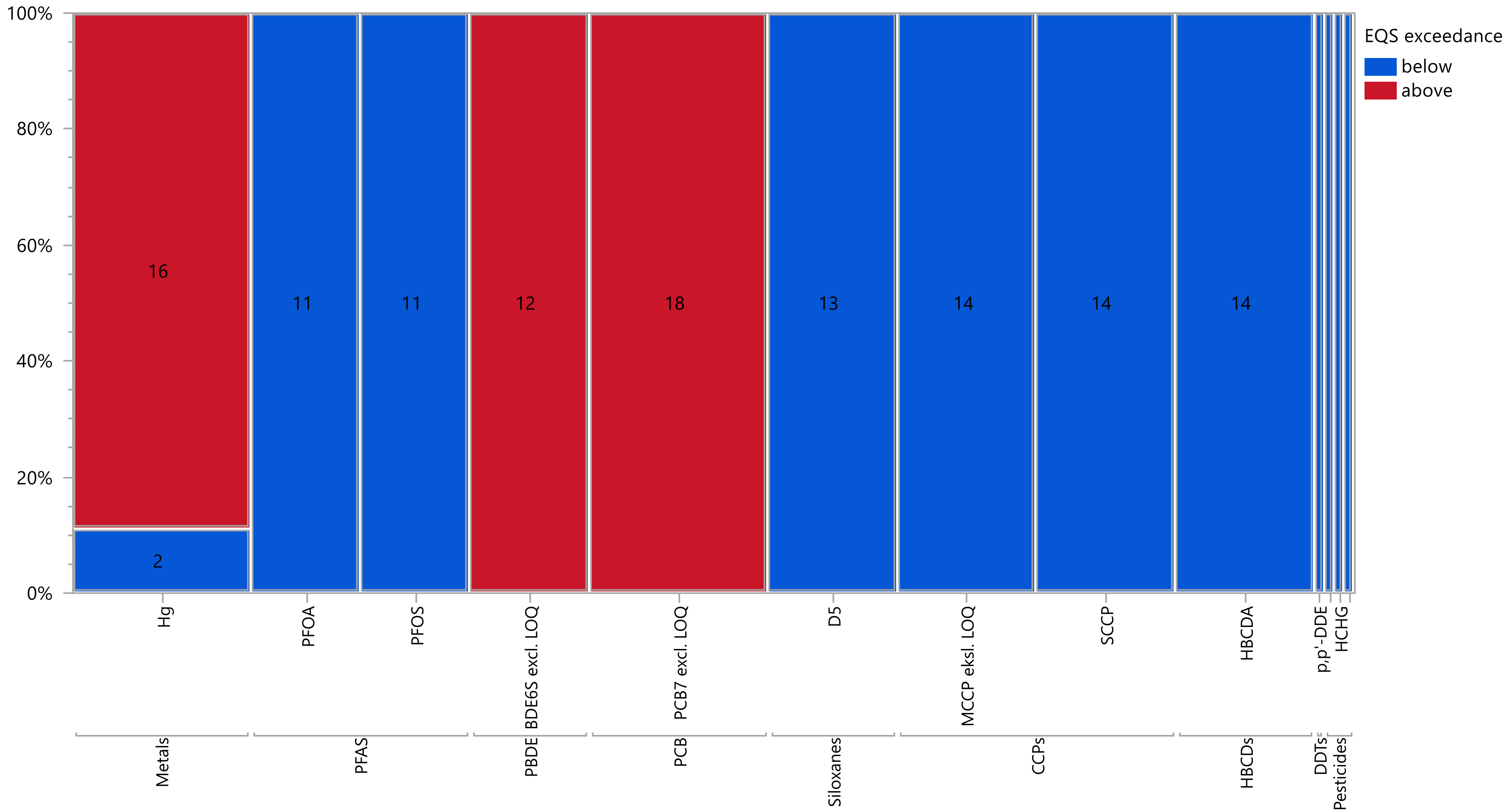


Figure 6. Exceedances of EQSs in cod by contaminant and contaminant group. The cells are labelled by the number of stations sampled. The exceedances are considered by the median for each station. The colours represent below or above EQSs.

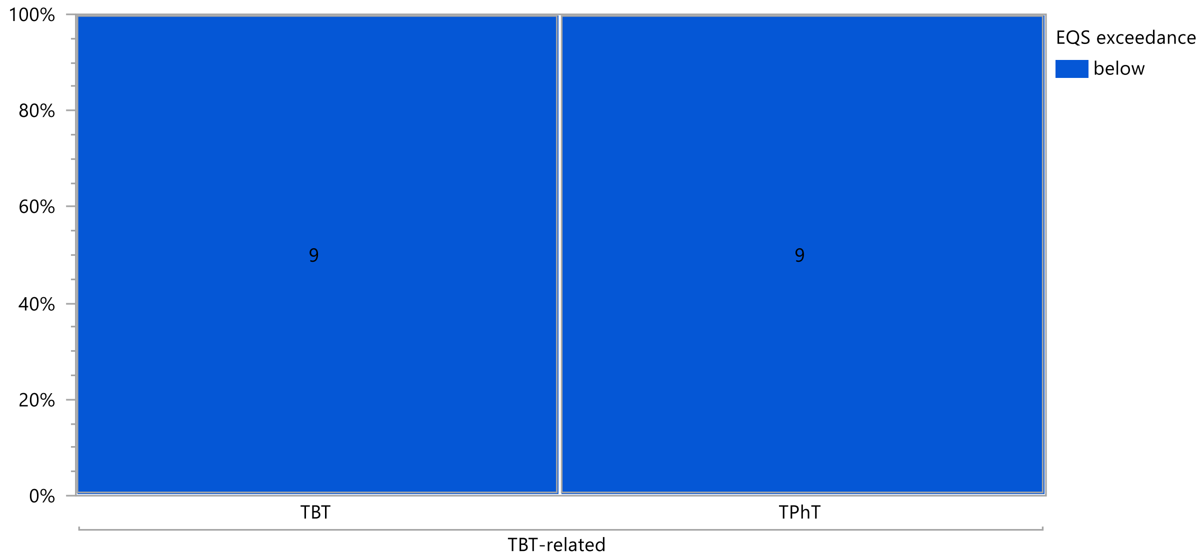


Figure 7. No exceedances of EQSs in snails (dogwhelk/common periwinkle) by contaminant. The number of stations in each group are labelled in the respective cell. The colour represents below EQSs.

3.1.1.1 Heatmaps for EQSs

To investigate potential pattern in stations exceeding EQSs, heatmaps for contaminants vs. stations are shown in **Figure 8** to **Figure 10**. 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, Hvaler (I024), Bjørkøya, Langesundfjord (71A) and Kvalnes, Mid Sørfjord (56A)). Concentrations of FLU was exceeded at Akershuskaia in the Inner Oslofjord (I301) which is not surprising since historically Akershuskaia had high levels of PAHs in blue mussel, and there is advice against eating seafood from the Oslofjord due to high concentrations of PAHs in blue mussel and mercury in cod[4]. SumPBDE6 exceeded EQSs at all stations investigated. For sumPCB7 frequent exceedances of EQSs were seen with exception of seven stations in 2021.

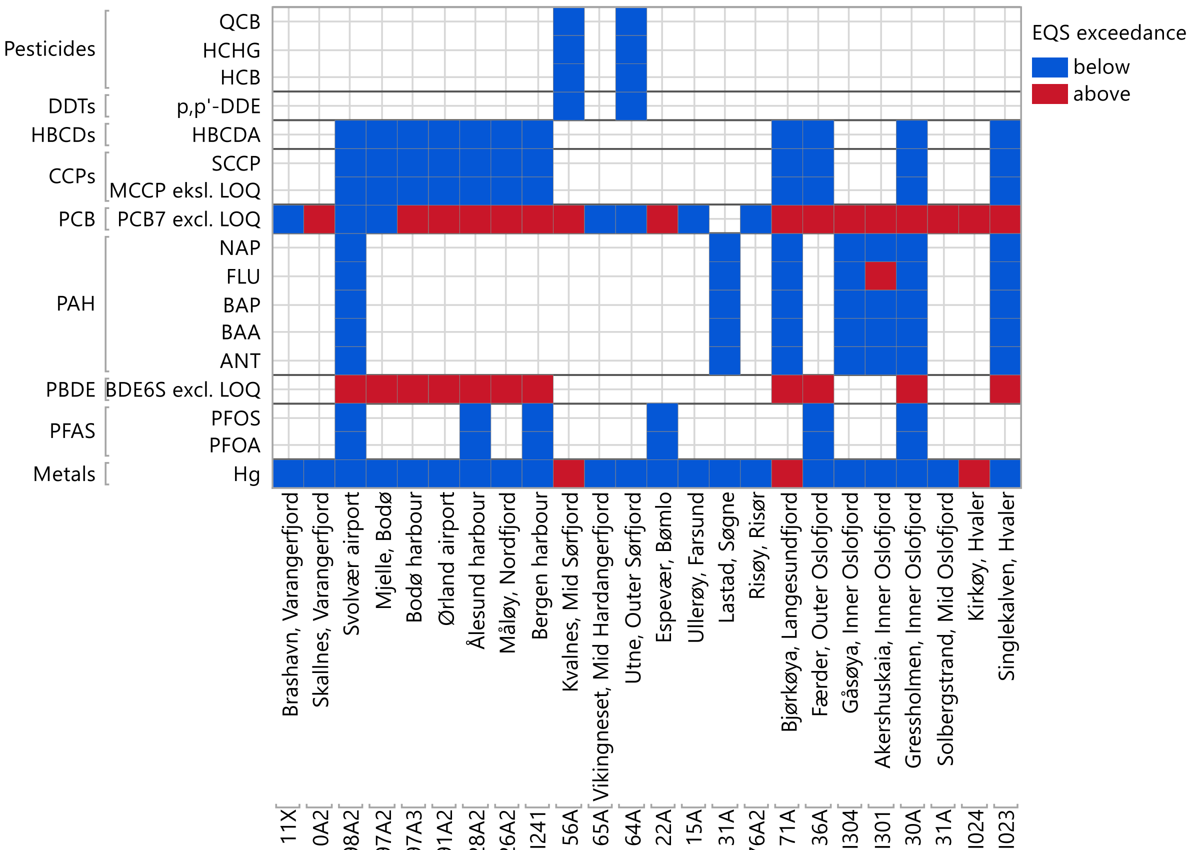


Figure 8. Heatmap of exceedances of EQSs in blue mussel. The exceedances are considered by the median for each station. The colours represent below or above EQSs. Empty "cells" mean that the contaminant was not analysed for at the indicated station. Grey lines show the midpoint of each station and contaminant, and darker lines have been inserted between contaminant groups. The stations are ordered along the coastline starting north moving south.

[4] https://miljostatus.miljodirektoratet.no/Sjomatadvarsel-for-Indre-Oslofjord

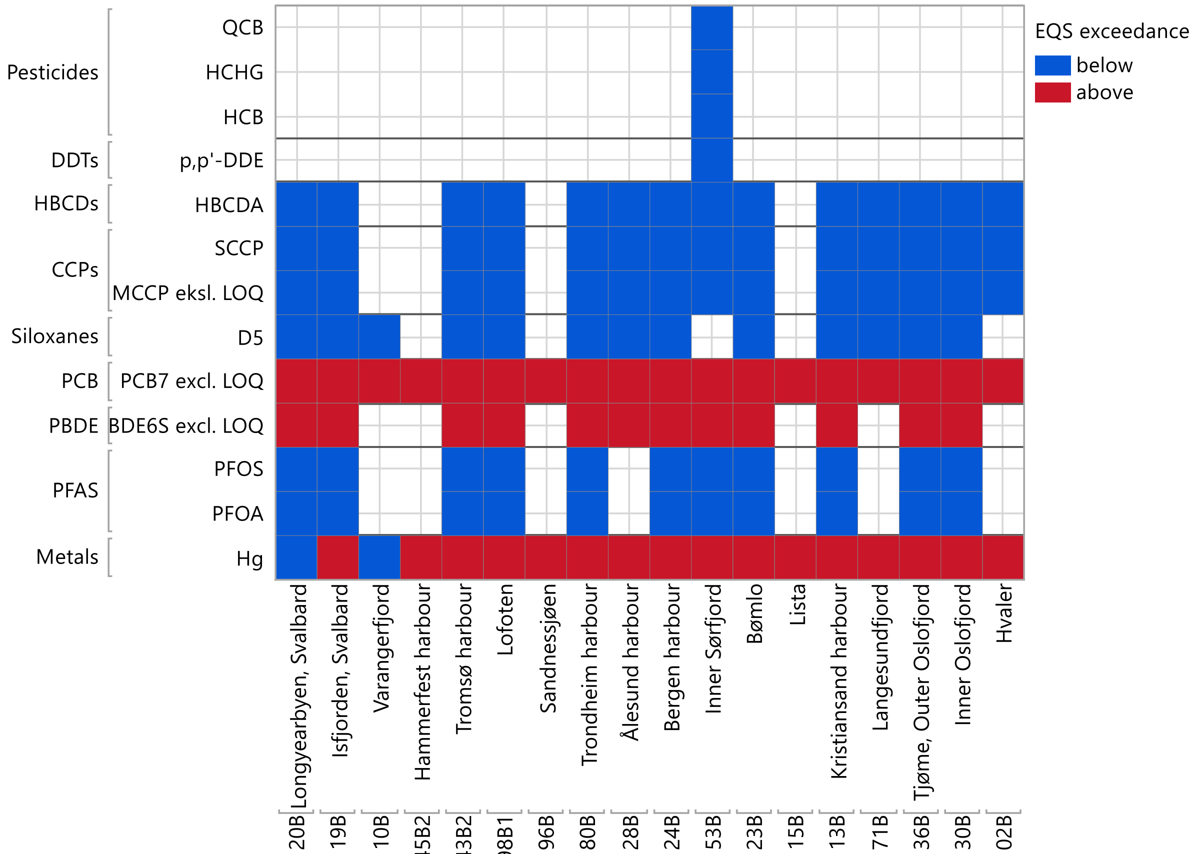


Figure 9. Heatmap of exceedance of EQSs in cod. The exceedances are considered by the median for each tissue. The colours represent below or above exceedance of EQSs. Empty "cells" mean that the contaminant was not analysed for at the indicated station. Grey lines show the midpoint of each station and 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 were observed. With exception of two stations for mercury (the Varangerfjord (10B) and Longyearbyen at Svalbard (20B)), median concentrations were exceeded at all stations analysed for the compounds.

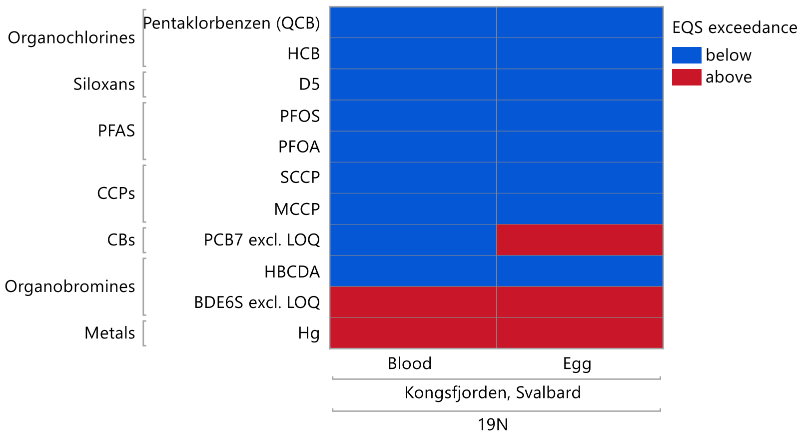


Figure 10. Heatmap of exceedance of EQSs in eider. The exceedances are considered by the median for each tissue. The colours represent below or above exceedance of EQSs.

3.1.2 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 4.5**). 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. A total of 724 assessments for PROREF have been made for the 26 contaminants (**Table 4**) selected for presentation for 2021 data. Results for other contaminants with PROREF, but not selected for presentation in 2021, are given in **Supplementary data**.

The PROREFs are at higher concentrations in cod than in mussels (except three metals; Co, Cd and Pb, **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 2021 for which a PROREF exist. The PROREFs are given in µg/kg ww (ng/g ww). Data are given with two significant digits.

Contaminant group	Contaminant	PROREF blue mussel (µg/kg ww)	PROREF cod (µg/kg ww)
Metals	Silver (Ag)	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)		10
	Perfluorooctanesulfonic acid (PFOS)		10
	Perfluorooctanesulfonamide (PFOSA)		6.2
PBDEs	PBDE congener 47 (BDE47)	0.17	16
	PBDE congener 99 (BDE99)	0.06	0.75
	PBDE congener 100 (BDE100)	0.05	2.6
	PBDE congener 153 (BDE153)	0.05	0.15
PAHs	Benzo(a)anthracene (BAA)	1.5	
	Benzo(a)pyrene (BAP)	1.2	
	Fluoranthene (FLU)	5.6	
	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 11**. Blue mussel exceeded PROREF for 35% of the samples, and 9% of the samples (35 samples) could not be classified vs. PROREF since LOQ was higher than PROREF. In cod, 89% of the samples were not exceeding PROREF, and the highest exceedances were lower for cod (2-5x PROREF) than for mussel (up to >20x PROREF).

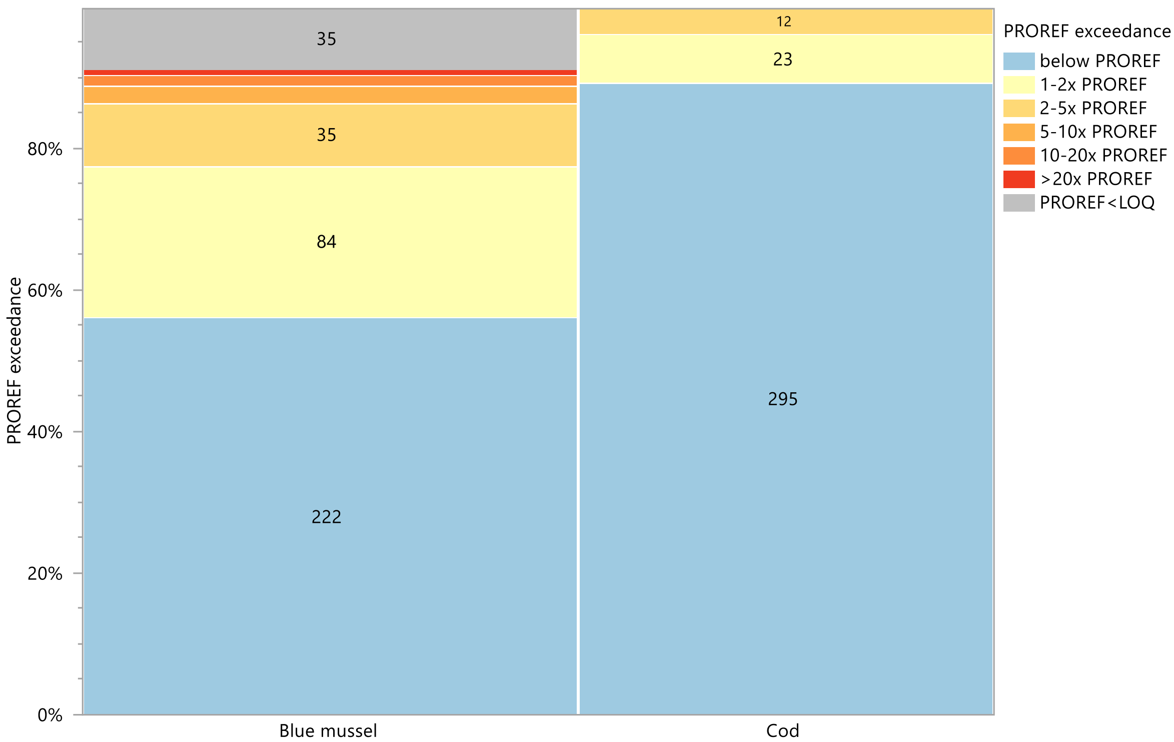


Figure 11. Exceedances of PROREF in a mosaic plot. The cells are labelled by the number of datapoints data points (stations × contaminants). The exceedances are considered by the median for each station and species. The colours represent below (blue) or above PROFEF (darker yellow to red), or that the PROREF was below LOQ, and therefore could not be classified (grey).

In mussel, the highest exceedances were seen for PCBs (CB118, CB138 and CB153, **Figure 12**). PCBs were exceeding PROREF at high percentages of stations; 83% of stations exceeded PROREF for CB118 with exceedances up more than 20x PROREF observed. For CB138, no stations were below PROREF, but 9 stations (39%) had LOQs too high for assessing the concentrations vs. PROREF. CB153 exceeded PROREF at 70% of stations. Only one exceedance of PROREF was observed for BDEs (BDE99), BDEs reported here were BDE47 BDE99, BDE100 and BDE153. Also, one exceedance of PROREF for HBCDA was observed. PAHs were analysed only in seven stations, where exceedances were observed in 1-2 stations of these for BAA, BAP, and FLU. PYR had exceedances of PROREF in all but one station. LOQs for silver and HCB were higher than PROREF, and this also applied to nine stations for CB138. Among the metals, lead and mercury exceeded PROREF at 58% of stations, with the highest exceedances observed for lead (10-20x PROREF). Except mercury, metal concentrations exceeded PROREF in general for 6-10 stations. However, for Cu concentrations only one station exceeded PROREF.

In cod, mercury was the contaminant exceeding PROREF the most (67% of stations exceeded, **Figure 13**). Among the metals, silver, arsenic, cadmium, and copper also had exceedances of PROREF (6-22% of stations). BDEs and CBs exceeded PROREF at a few stations each, maximum 22% of stations exceeded.

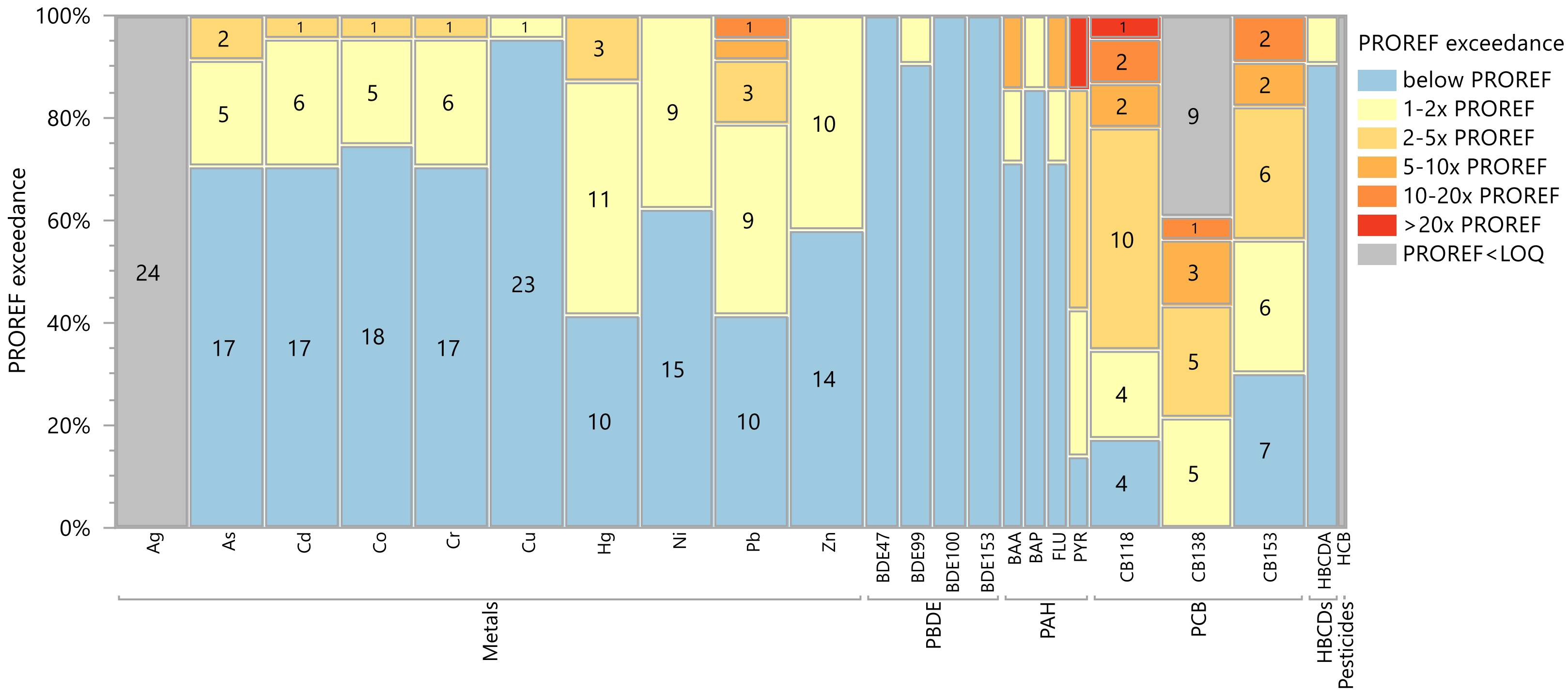


Figure 12. Exceedances of PROREF in blue mussel by contaminant and contaminant group. The cells are labelled by the number of stations sampled. The exceedances are considered by the median for each station. The colours represent below or above exceedance of PROFEF (darker yellow to red), or that the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

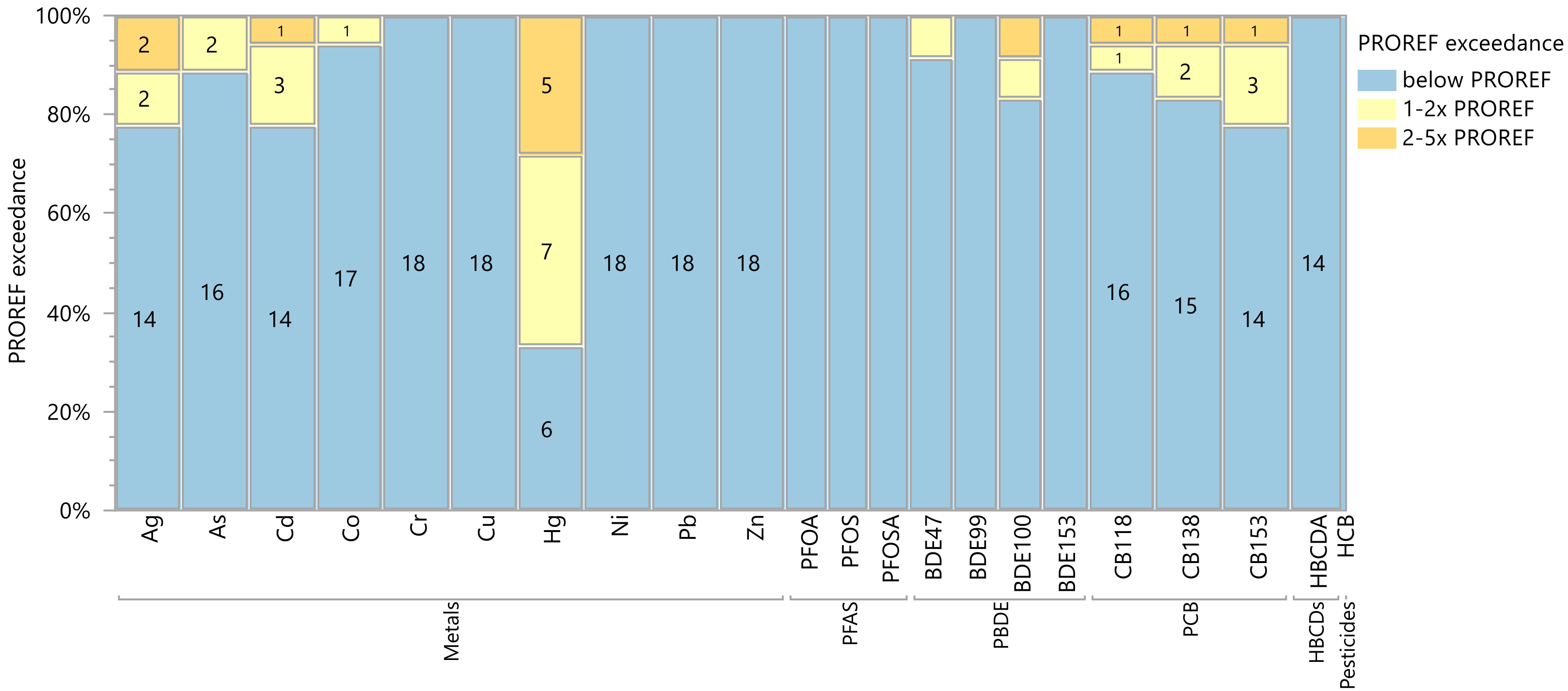


Figure 13. Exceedances of PROREF in cod by contaminant and contaminant group. The cells are labelled by the number of stations sampled. The exceedances are considered by the median for each station. The colours represent below or above exceedance of PROFEF (darker yellow to red), or that the PROREF was below LOQ, and therefore could not be classified.

3.1.2.1 Heatmaps for PROREF

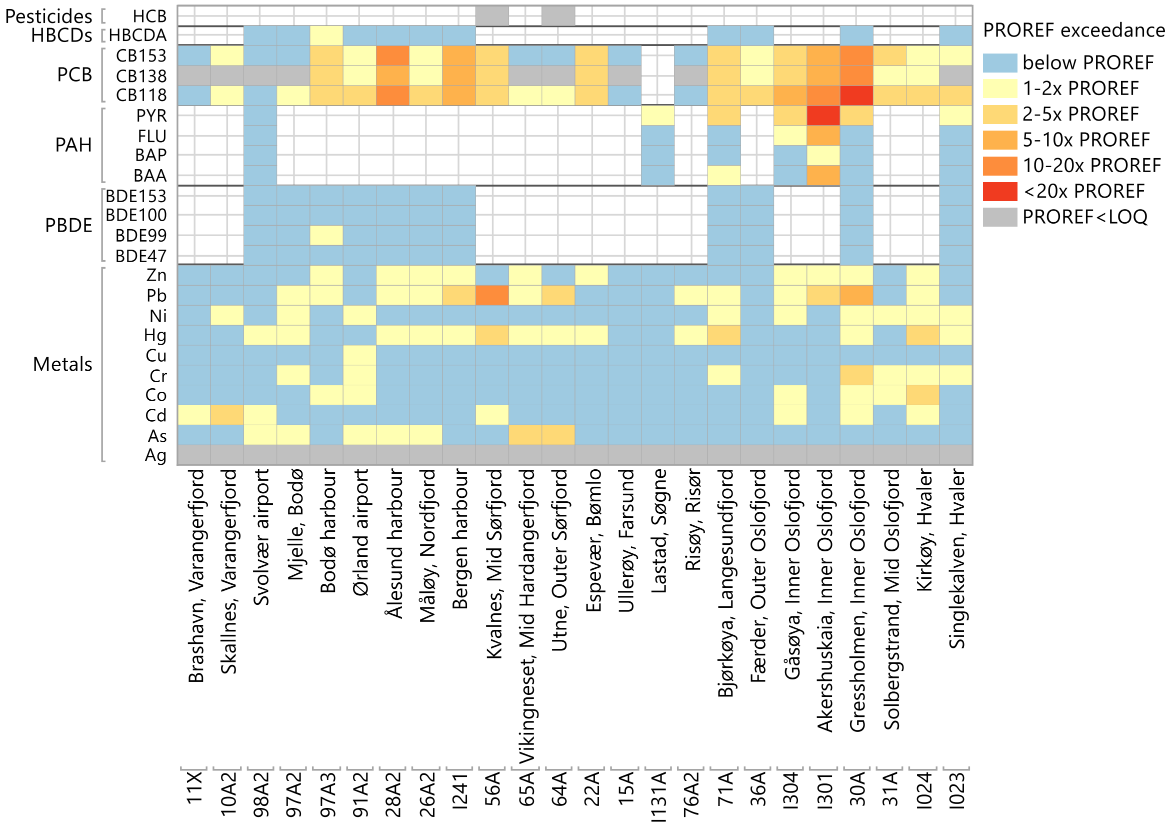


Figure 14. Heatmap of exceedances of PROREF in mussel. The colours represent below or above exceedance of PROREF. Empty “cells” mean that the contaminant was not analysed for at the indicated station. Grey lines show the midpoint of each station and contaminant, and darker lines have been inserted between contaminant groups. The stations are ordered along the coastline starting north moving south.

Blue mussel stations in the Oslofjord (**Figure 14**) had many exceedances of PROREFs. For CB118 in mussel at Gressholmen (30A) and PYR in mussel at Akershuskaia (I301) in the Inner Oslofjord, the exceedances were highest (>20x PROREF). PCBs were not exceeding PROREFs at stations in the south (Risøy at Risør (76A2) and Ullerøy at Farsund (15A)) and north (Svolvær airport (98A2) and Brashavn in the Varangerfjord (11X)). For PAHs, the only station investigated with no exceedances of the PAHs selected was 98A2. Exceedances of PROREF were not observed for organobromines except BDE99 and HBCDA at Bodø harbour (97A3). Metals were often exceeding PROREF, but the stations at Færder in the Outer Oslofjord (36A), Lastad at Søgne (I131A) and Ullerøy at Farsund (15A) had no exceedances of any metals investigated.

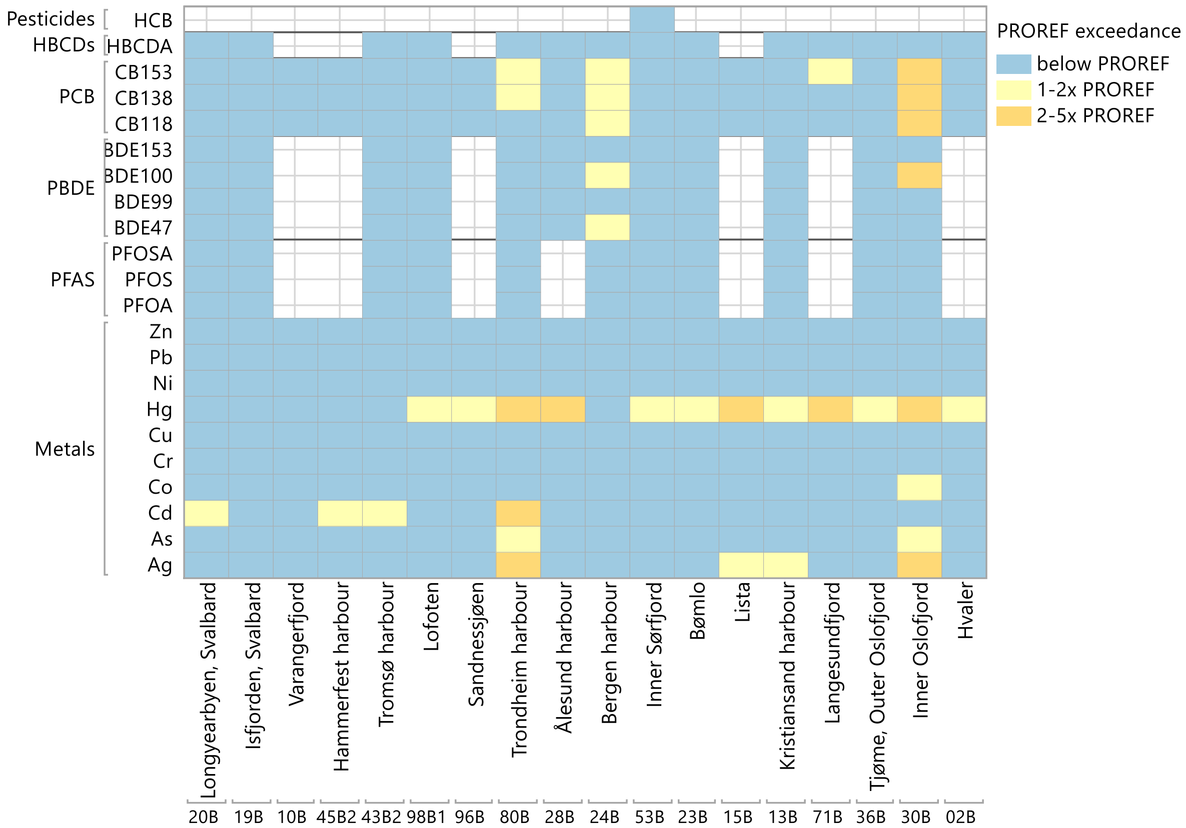


Figure 15. Heatmap of exceedances of PROREF in cod. The colours represent below or above exceedance of PROREF. Empty "cells" mean that the contaminant was not analysed for at the indicated station. Grey lines show the midpoint of each station and contaminant, and darker lines have been inserted between contaminant groups. The stations are ordered along the coastline starting north moving south.

Exceedances of PROREF in cod (**Figure 15**) were most often observed at the stations in the Inner Oslofjord (30B) (CBs, BDE100, Hg, Co, As and Ag) followed by Trondheim harbour (80B) (CB138 and 118, Hg, Cd, As and Ag) and Bergen harbour (24B) (CBs, BDE47 and 99). Two stations (the Varangerfjord (10B) and the Isfjord at Svalbard (19B)) had no exceedances of PROREFs investigated.

3.1.3 Time trends

Time trends for selected contaminants (**Table 1**) were assessed, in total 801 time trends (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.**

Time trends (long-term (>10 years) and short-term (≤ 10 years)) for blue mussel and cod are shown in **Figure 16** to **Figure 18**. For eider, only short-term time trends exist and are shown only in **Figure 16** (lower panel). Heatmaps of time trends (stations vs. contaminants) are shown in **Figure 19** to **Figure 26**.

A substantial part of the long-term trends could not be determined because data before 2012 were not existing. Otherwise, long-term time trends in blue mussel were dominated by decreasing trends (26%) and no trend (26%). However, also increasing trends were observed for 6% 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, but the percentage of decreasing trends was higher (30%). No trend was observed for 17% of data for cod and increasing trend for 6%.

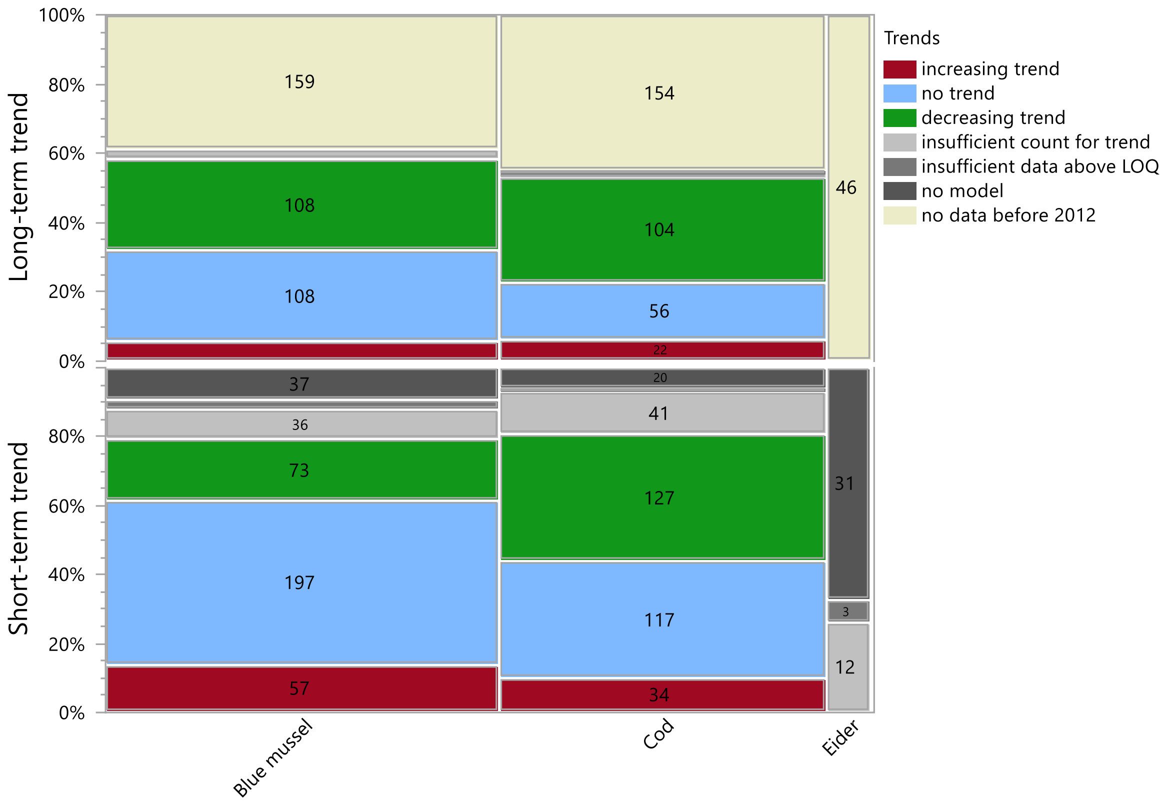


Figure 16. Mosaic plot of time trends for blue mussel, cod, and eider. Upper panel shows long-term trends, while lower panel shows short-term trends. The number of (stations × contaminants (and × tissue for eider)) are indicated in the respective cells.

There were more datapoints (station × contaminant) that could be determined for short-term trends than long-term trends for both mussel and cod. However, the percentage of insufficient count/data/model was also higher for short-term trends (**Figure 16** lower panel). No trend dominated for short-term trends for mussel (48%) while decreasing trends dominated for cod (38%).

Increasing short-term trends were found both in mussel and cod (14% and 10% respectively). No short-term trends could be determined for eider.

In blue mussel (**Figure 17**), decreasing long-term trends were dominating and was found for most contaminants. Increasing long-term trends were found for all metals (apart from Ag) were also found for two PCBs (CB138 and CB153).

The overall picture was roughly the same for short-term trends, but two metals (Ag and Cr), BDE153, PFOS, and PFOSA did not have any decreasing short-term trends in mussel. All selected metals apart from Ag, and BDEs, and PCBs had increasing short-term trends. The highest percentages of increasing short-term trends were seen for CB153 and CB138 and Ag (57% and 52% respectively). Due to increased LOQ for Ag the last three years, the confidence of the time trends is limited.

Long-term trends in cod (**Figure 18**) were also dominated by decreasing time trends which was observed for all but three selected compounds (Ag, HBCDA and D5). A few of the selected contaminants had increasing trends (only metals; Ag, As, Cd, Co, Cu, Hg, and Zn). For Ag and Hg increasing trends were dominating (39% and 33%, respectively).

Short-term time trends in cod were also dominated by decreasing trends, which were found for all but two selected compounds (D5 and HCB). Dominating decreasing trends were found for several contaminants (Cr, Ni, Pb, BDE47, BDE99, BDE199, HBCDA, PFOS and PFOSA) from 72% (Cr and Ni) to 42% (BDE100). Most selected metals except Cr and Ni had increasing short-term trends. For Ag, the increasing short-term trend was the most common finding (44%). Also, BDEs (BDE47 and BDE100) and PCBs had increasing trends.

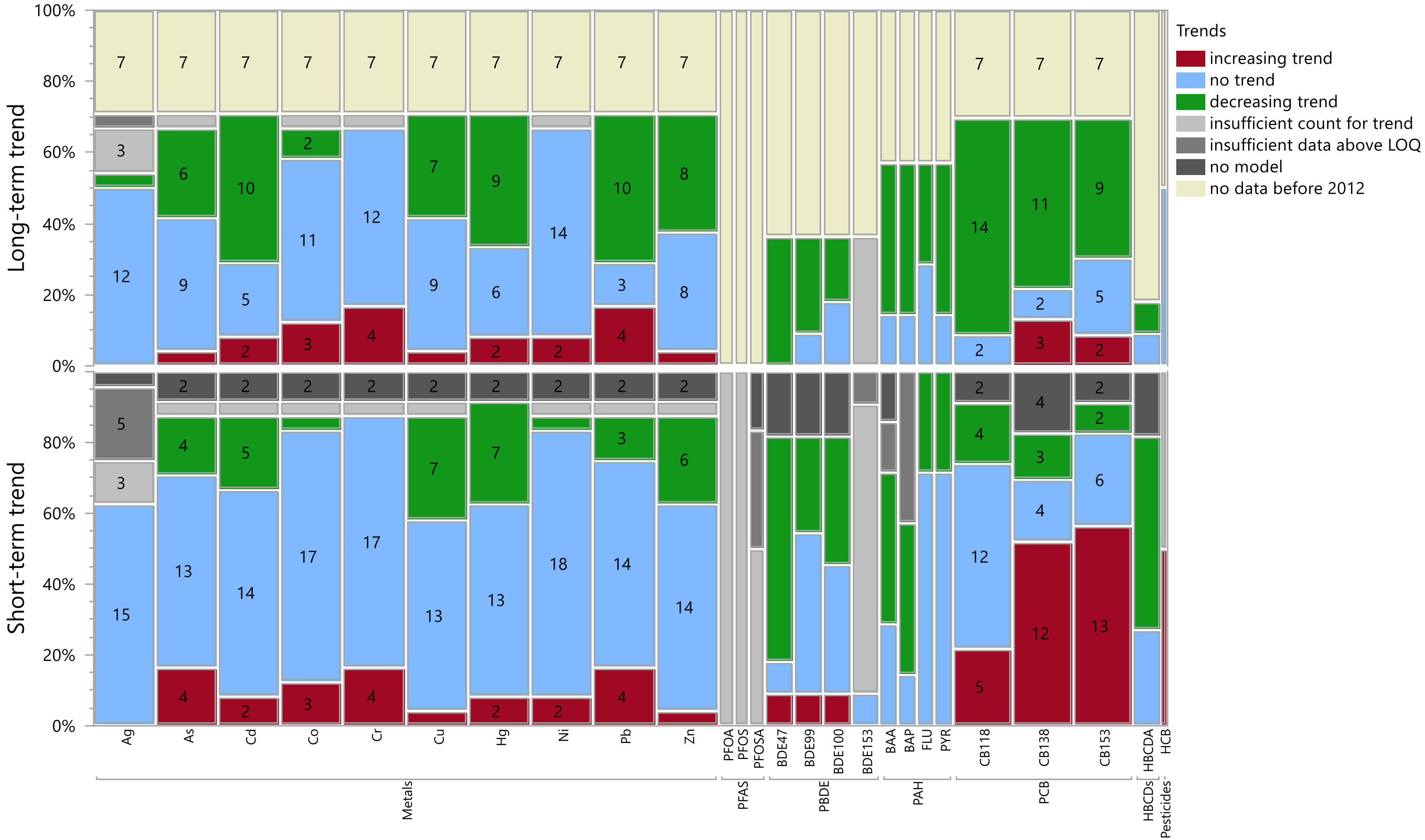


Figure 17. Time trends for blue mussel. Upper panel shows long-term trends, while lower panel shows short-term trends. The number of (stations × contaminants) are indicated in the respective cells.

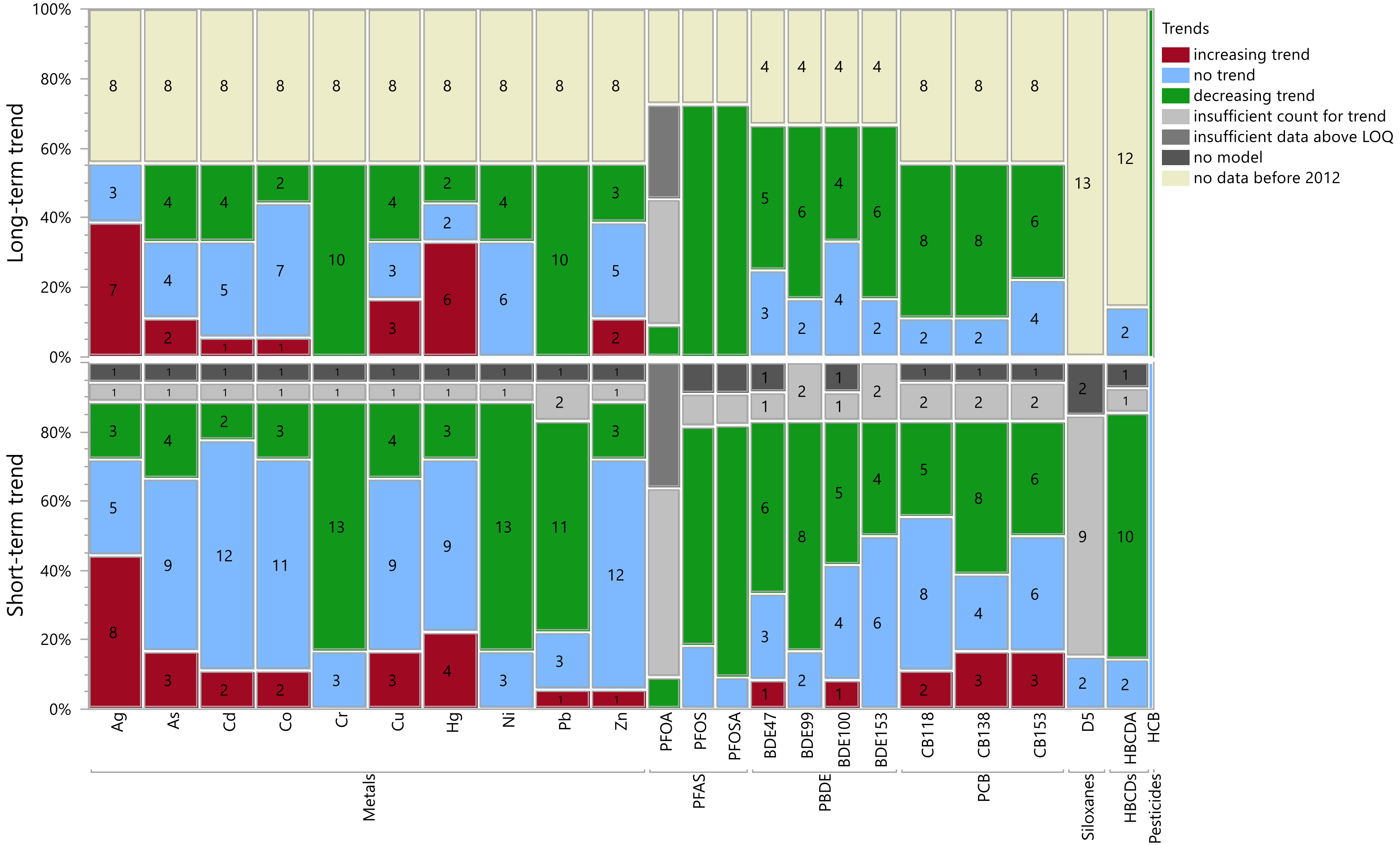


Figure 18. Time trends for cod. Upper panel shows long-term trends, while lower panel shows short-term trends. The number of (stations × contaminants) are indicated in the respective cells.

3.1.3.1 Heatmaps for long term trends

The increasing long-term trends for metals in mussel (**Figure 19**) were mostly found at stations in the Oslofjord (Solbergstrand (31A), Gressholmen (30A), Akershuskaia (I301), Gåsøya (I304), and Færder (36A)). In addition to these, the stations at Kirkøy at Hvaler (I024), Bjørkøya in the Langesundfjord (71A), Espevær at Bømlo (22A), Vikingneset in the Mid Hardangerfjord (65A), Bergen harbour (I241), Svolvær airport (98A2) and Brashavn in the Varangerfjord (11X) had increasing long-term trends. The stations with most increasing trends were Gåsøya in the Inner Oslofjord (I304) (Cd, Co, Cr, Hg, Ni and Pb) and Gressholmen in the Inner Oslofjord (30A) (the same compounds except Hg). Decreasing time trends were more often seen in other stations than in the Oslofjord for metals. For PCBs, all stations in the Oslofjord had decreasing trends, but increasing trends were seen at Ullerøy at Farsund (15A), Vikingneset in the Mid Hardangerfjord (65A), Skallnes in the Varangerfjord (10A2), and Brashavn in the Varangerfjord (11X).

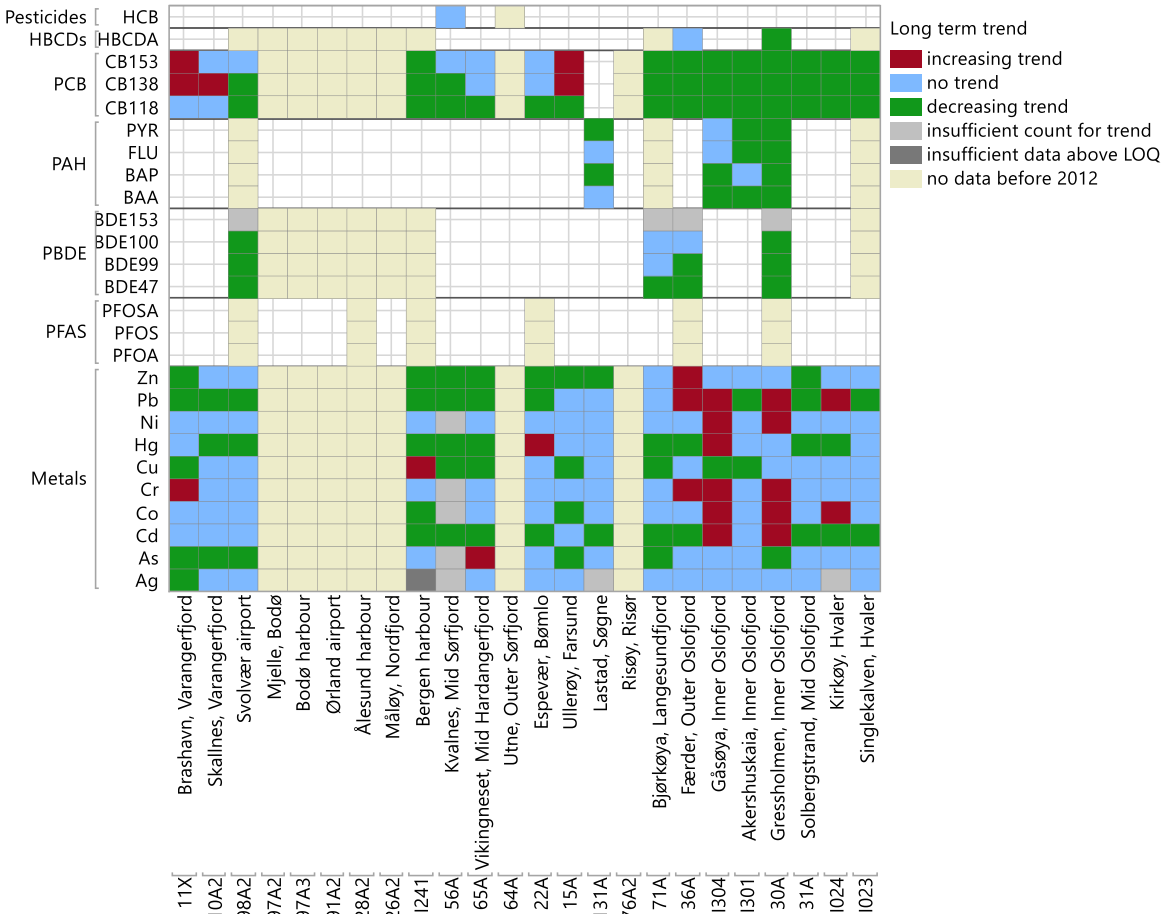


Figure 19. Heatmap of long-term time trends in blue mussel. The colours represent time trends observed at stations. Empty "cells" mean that the contaminant was not analysed. Grey lines show the midpoint of each station and contaminant, and darker lines have been inserted between contaminant groups. The stations are ordered along the coastline starting north moving south.

For selected organic contaminants (PFAS, PCBs, BDEs), no increasing long-term trends were observed in cod (**Figure 20**). Cod from Lista (15B) had six metals with increasing time trends. Increasing time trends were observed for one or more metals at all stations where metals were analysed.

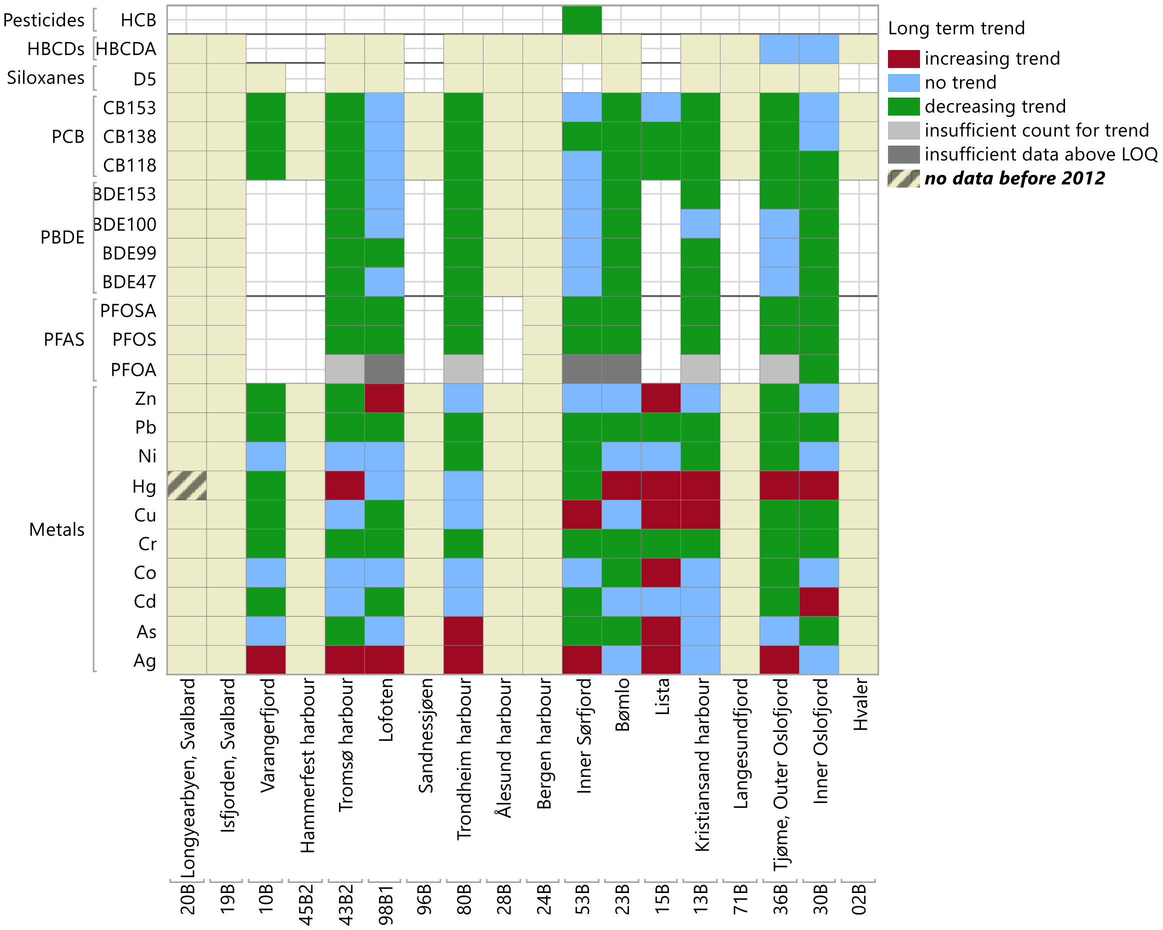


Figure 20. Heatmap of long-term trends in cod. The colours represent time trends observed at stations. Empty "cells" mean that the contaminant was not analysed. Grey lines show the midpoint of each station and contaminant, and darker lines have been inserted between contaminant groups. For mercury, cod have been length adjusted except for station marked (patterned, station 20B). The stations are ordered along the coastline starting north moving south.

3.1.3.2 Heatmaps for short-term trends

While only a few increasing long-term time trends for PCBs in mussel were observed, 61% (14) of stations had increasing short-term trends for one or more PCBs (**Figure 21**). Only two stations (Bergen harbour (I241) and Ørland airport (91A2)) had decreasing short-term trends for the selected PCBs. The stations in the Inner Oslofjord Gressholmen (30A), Akershuskaia (I301) and Gåsøya (I304) had high proportions of decreasing trends for PAHs, and no increasing trends were observed for PAHs or PFAS. BDEs had decreasing trends at station Gressholmen (30A) and increasing trends at Måløy in the Nordfjord (26A2).

Station Gressholmen in the Inner Oslofjord (30A) had the most increasing time trends for metals (six metals), but also Gåsøya in the Inner Oslofjord (I304) and Færder (36A) in the Outer Oslofjord had five metals with increasing time trends. Nine stations had no increasing short-term time trends for metals (Singlekalven at Hvaler (I023), Risøy at Risør (76A2), Lastad at Søgne (I131A), Ullerøy at Farsund (15A), Espevær at Bømlo (22A), Kvalnes in the Mid Sørfjord (56A), Måløy in the Nordfjord (26A2), Ørland airport (91A2), and Skallnes in the Varangerfjord (10A2)).

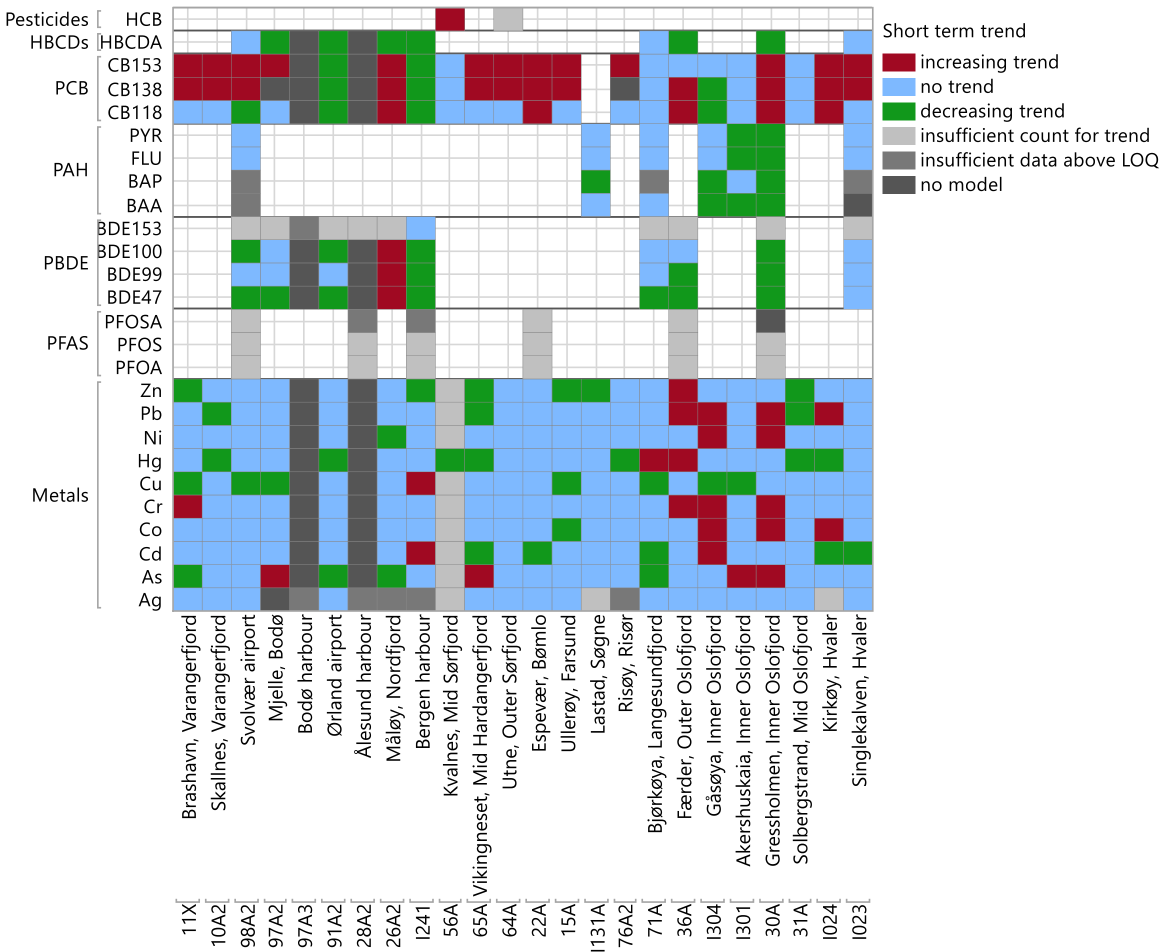


Figure 21. Heatmap of short-term trends in blue mussel. The colours represent time trends observed at stations. Empty "cells" mean that the short-term trend could not be estimated or that the contaminant was not analysed. Grey lines show the midpoint of each station and contaminant, and darker lines have been inserted between contaminant groups. **The time trends for silver are more uncertain due to higher LOQ for silver the last three years. Trends estimated for silver are therefore more uncertain than the other time trends.** The stations are ordered along the coastline starting north moving south.

In cod, short-term time trends for organic contaminant were mostly decreasing or had no trends (**Figure 22**). Increasing trends were only seen at three stations (Trondheim harbour (80B), Sandnessjøen (96B) and Lofoten (98B1)) for PCBs and for two BDEs (BDE47 and BDE100) at Lofoten (98B1). For metals, only a few stations did not have any stations with increasing trends (Langesundfjord (71B), Bergen harbour (24B), Ålesund harbour (28B), Lofoten (98B1), and Hammerfest harbour (45B2)).

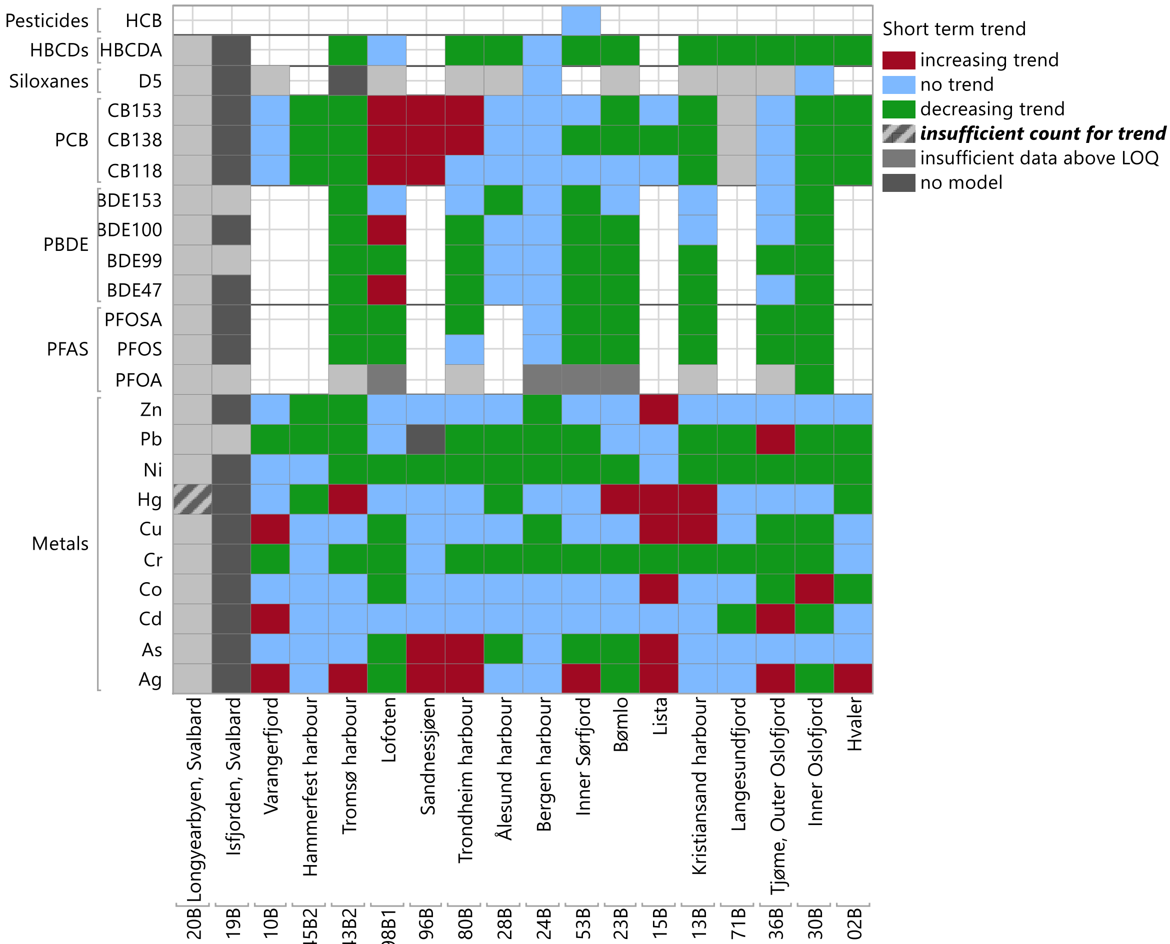


Figure 22. Heatmap of short-term trends in cod. The colours represent time trends observed at stations. Empty "cells" mean that the short-term trend could not be estimated or that the contaminant was not analysed. Grey lines show the midpoint of each station and contaminant, and darker lines have been inserted between contaminant groups. For mercury, cod have been length adjusted except for station marked (patterned, station 20B). The stations are ordered along the coastline starting north moving south.

3.2 Factsheets of selected contaminants

Factsheets for selected contaminants are built in the same way for contaminants in **chapter 3.2.1** (metals) and **chapter 3.2.2** (organic contaminants). Example figures are shown here with full figure text explanation. In the rest of the chapter, only shorter figure texts are given to avoid repetition.

A heatmap with assessments of exceedances of EQS (if there is an EQS for the contaminant) and PROREF (if PROREF has been calculated) for both blue mussel and cod/eider are given for each contaminant. Note that PROREF can vary depending on species, tissue, and contaminant. The ratings for PROREF are under (by a factor of lower than 0.5, 0.5-0.75, 0.75-0.9 and 0.9-1), over (by a factor of 1-2, 2-5, 5-10, 10-20 and greater than 20), and no PROREF. In addition, the time trends for both long-term (>10 years) and short-term (≤ 10 years) trends are shown to the right of the heatmap. In the time trends figures, the confidence interval (95%) shows the statistical confidence of the trends. An example of a heatmap with explanation is presented in **Figure 23**.

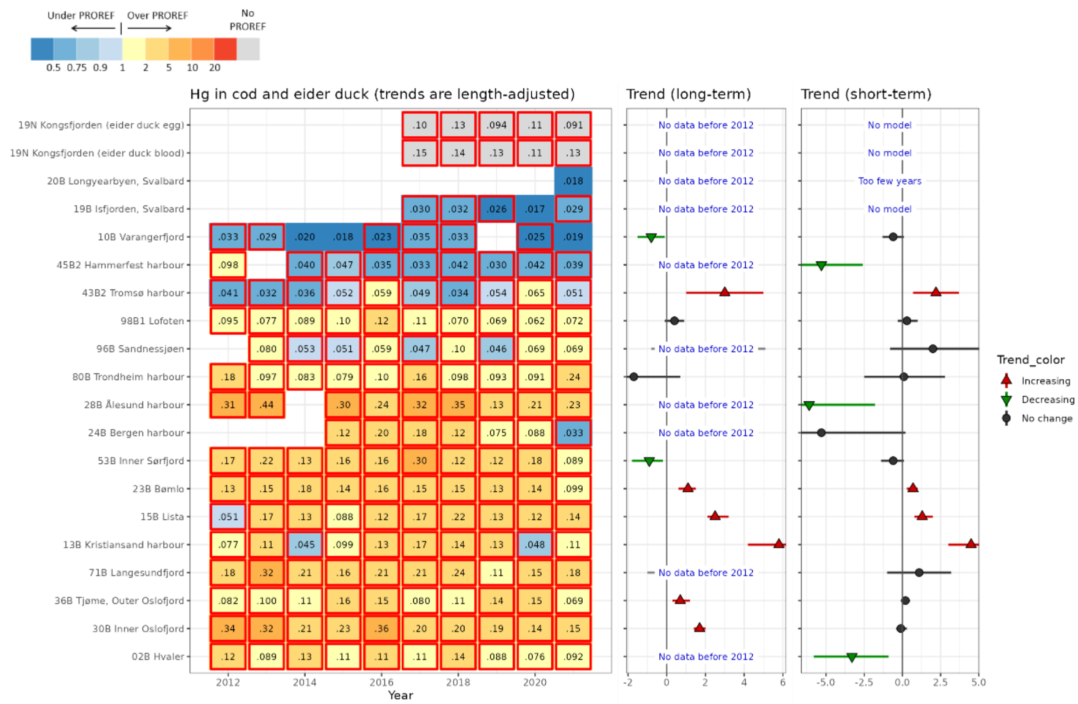


Figure 23. Example (mercury in cod and eider) of heatmap and time trends presented in chapters 3.2.1 and 3.2.2. The heatmaps to the left show median concentrations vs. years (last 10 years) for stations investigated in 2021. Concentrations are presented in µg/kg ww. Numbers are given with two significant digits if space allows, while numbers below zero are given without the 0 in front of the point to save space (i.e. 0.10 are written ".10"). The stations to the left are sorted according to placement along the Norwegian coast including Svalbard from north moving south. The cells have background coloured according to exceedances of PROREF and exceedances of EQSs indicated with a red rectangle enclosing the cells (station/year) if there is an EQS for the contaminant. Where "cells" are empty, the station was not analysed that year. To the right are long-term (>10 years) and short-term (≤ 10 years) trends indicated with medians and 95% confidence interval for each station. Statistically increasing/decreasing trends are indicated in red/green triangles respectively (confidence intervals not including 0), and non-significant trends in black circles (confidence intervals including 0). The scale on the time trends x-axes is % changes/year. The text in the trends indicates reasons for missing trends.

Thereafter, the concentrations in blue mussel and cod are compared to EQS and PROREF. This is done in a similar manner to risk assessments, i.e. the measured concentrations are divided by EQS or PROREF to get a ratio ("risk quotient"). If the ratio is above 1, then EQS/PROREF is exceeded. Higher ratio means higher exceedance of EQS/PROREF. Very high exceedances indicate that environmental concentrations are far higher than the chronic toxicity (for exceedance of EQS) and far higher than reference conditions (for PROREF). Conversely, if the ratio of concentration/EQS or PROREF is far below 1, then the concentration measured is considered non-toxic (for EQS) or below PROREF. **Figure 24** gives an example of concentration/EQS.

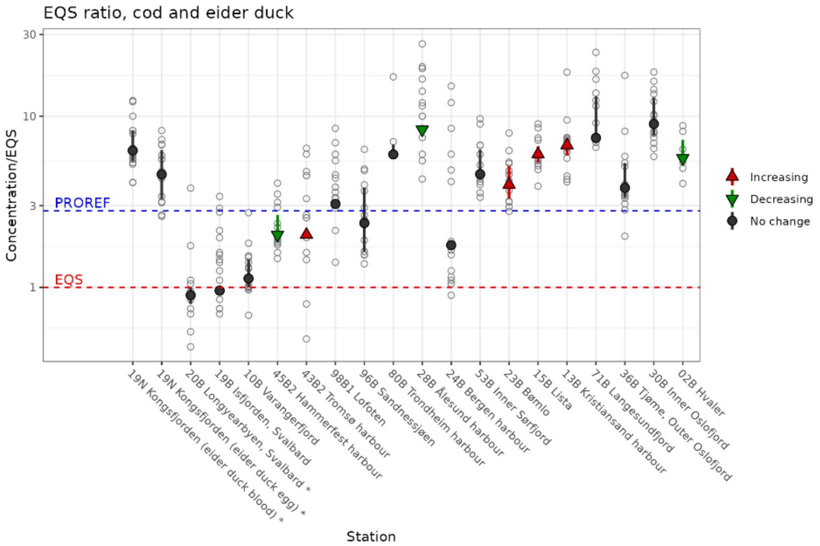


Figure 24. Example (mercury in cod and eider) of a concentration/EQS figure presented in chapters 3.2.1 and 3.2.2. The ratios presented in later chapters can be either vs EQS or PROREF, text along the y-axis indicate whether EQS or PROREF ratio is presented. For each station (ordered from north to south), the median concentrations are presented as points or triangles representing the *short-term time trends* for the station (see **Figure 23** for explanation). The vertical line extending from the point extends from maximum to minimum for blue mussel and interquartile range (25%-75%) for cod/eider. The data points for individual samples>LOQ are presented as outlined grey circles, while samples <LOQ are presented as triangles at LOQ. EQS is indicated with a dashed horizontal red line, while PROREF is indicated with a dashed horizontal blue line. Where ratios vs. EQS is presented, the relative position of PROREF (vs. EQS) is indicated with a dashed blue line if possible. In this figure, the PROREF is found at ca. 3 times higher concentration than the EQS. The EQS for mercury in cod is 0.02 µg/kg ww, and PROREF is 0.056 µg/kg. Note that scales for the x axis and y axis can vary from figure to figure.

Lastly, for each selected contaminant, one or more selected time trends for one or more stations are selected for presentation of the whole time-series. An example of a time trend figure is given in **Figure 25**.

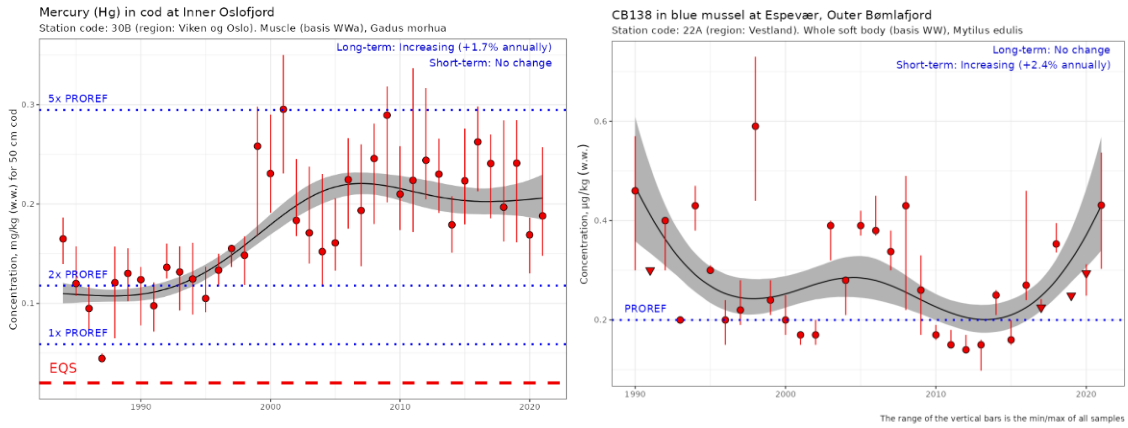


Figure 25. Example of two time trend figures, in this case for mercury in cod (left) and CB138 in blue mussel (right). Median concentrations are plotted against the year they were sampled and are shown as red circles, or triangles (where more than half of the data were below LOQ). For cod, the vertical 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 any). Note that scales for the x axis and y axis can vary from figure to figure.

3.2.1 Metals

3.2.1.1 Mercury (Hg)

Mercury (Hg) is a heavy metal belonging to the transition element series of the periodic table. Mercury can be organic (methylmercury and dimethylmercury), inorganic (Hg^{2+}) or elemental (Hg^{0}) and has toxic effects on *inter alia* the nerve system. The toxic substance can be transported by water and air over long distances and end up in the environment in completely different parts of the globe than where it was released. With a few exceptions, there is a general prohibition on the use of Hg in products in Norway. In the present study, total Hg (organic and inorganic, here abbreviated to Hg), was analysed in blue mussel at 24 stations, in cod fillet at 18 stations, and in eider blood and eggs at one station (**Table 1**). For cod, the data for time trends are based on length adjusted cod (50 cm) (Ruus et al. 2017).

EQS

Mercury concentrations exceeded EQS (20 µg/kg ww, **Table 3**) in three mussel stations which they also did in 2020 (**Figure 26**). Median concentrations at all but two cod stations exceeded EQS for (**Figure 26** and **Figure 27**). **Figure 27** shows that EQS was exceeded more in cod (up to 12x for median concentration) than in blue mussel. EQS was exceeded by one or more fish in *all* cod stations. Mercury concentrations also exceeded EQS in eider blood and eggs.

PROREF

The same three mussel stations that exceeded EQS had the highest ratio (3-5x PROREF, **Figure 28**). Eleven mussel stations exceeded PROREF (up to 2.5x). Five cod stations exceeded PROREF 2-3x, while seven exceeded 1-2x PROREF. PROREF was exceeded by one or more cod in *all* cod stations.

Long-term trends

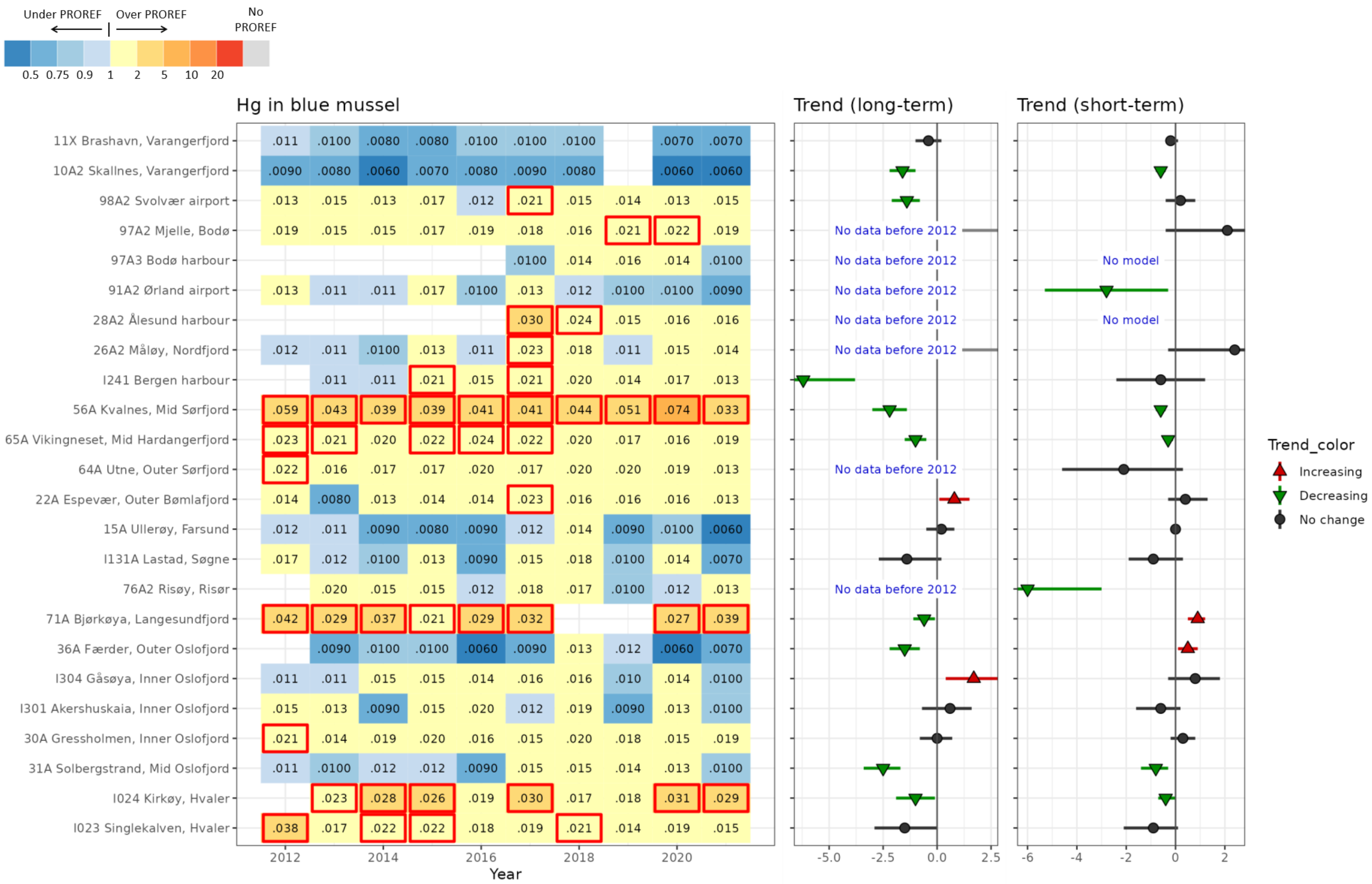
For mussel stations, the dominating long-term time trends were significant decreasing trends at nine stations (**Figure 26**), while two (Espevær close to Bømlo (22A) and Gåsøya in the Inner Oslofjord (I304)) had significant increasing trend.

Ten cod stations had enough data to estimate long-term time trends. Six cod stations had significant increasing long-term trends and two had significant decreasing trend.

Short-term trends

Seven mussel stations had significant decreasing short-term time trends, two of these had no long-term trends previously due to no data prior to 2012. Two mussel stations had significant increasing trends, and both these stations had significant decreasing long-term trends.

Three cod stations had significant increasing short-term trends and these three also had significant increasing long-term trends. Four stations had significant decreasing short-term trends.



Under PROREF
Over PROREF
No PROREF
0.5 0.75 0.9 1 2 5 10 20
Hg in blue mussel
Trend (long-term)
Trend (short-term)
11X Brashavn, Varangerfjord
10A2 Skallnes, Varangerfjord
98A2 Svolvær airport
97A2 Mjelle, Bodø
97A3 Bodø harbour
91A2 Ørland airport
28A2 Ålesund harbour
26A2 Måløy, Nordfjord
I241 Bergen harbour
56A Kvalnes, Mid Sørfjord
65A Vikingneset, Mid Hardangerfjord
64A Utne, Outer Sørfjord
22A Espevær, Outer Bømlafjord
15A Ulleröy, Farsund
I131A Lastad, Søgne
76A2 Risøy, Risør
71A Bjørkøya, Langesundfjord
36A Færder, Outer Oslofjord
I304 Gåsøya, Inner Oslofjord
I301 Akershuskaia, Inner Oslofjord
30A Gressholmen, Inner Oslofjord
31A Solbergstrand, Mid Oslofjord
I024 Kirkøy, Hvaler
I023 Singlekalven, Hvaler
No data before 2012
No model
Year
Trend_color
Increasing
Decreasing
No change

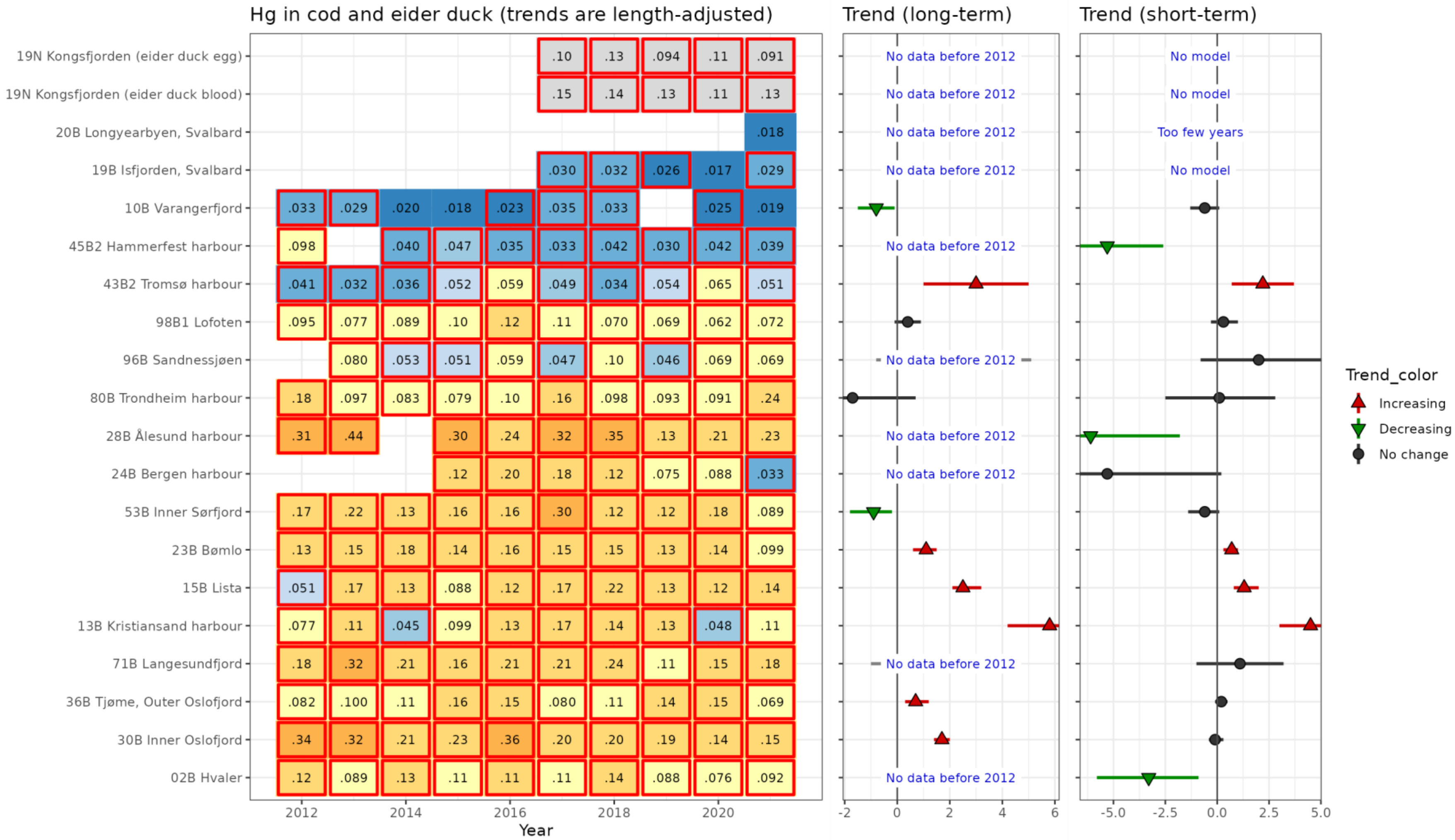
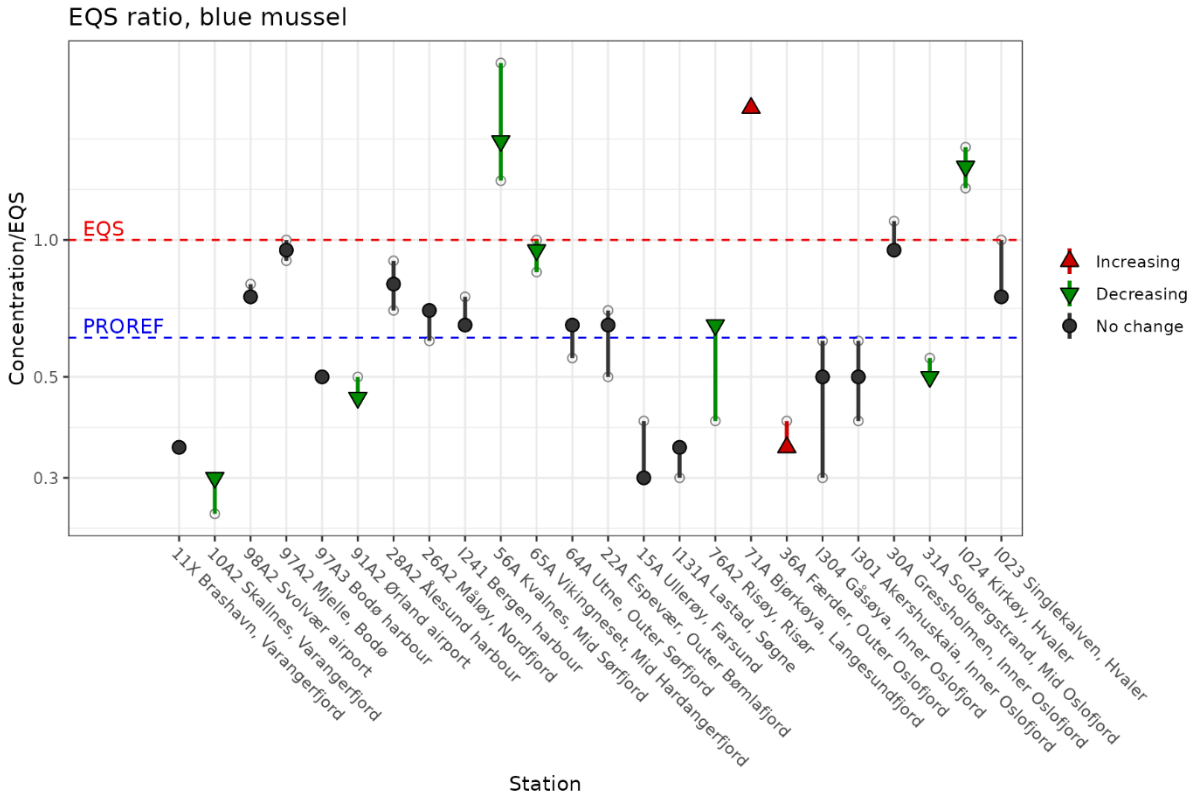


Figure 26. Heatmap and time trends of mercury in (upper panel) blue mussel and (lower panel) cod (length adjusted (50 cm)) and eider. One station (20B) could not be length adjusted. For full explanation of figure see example **Figure 23**.



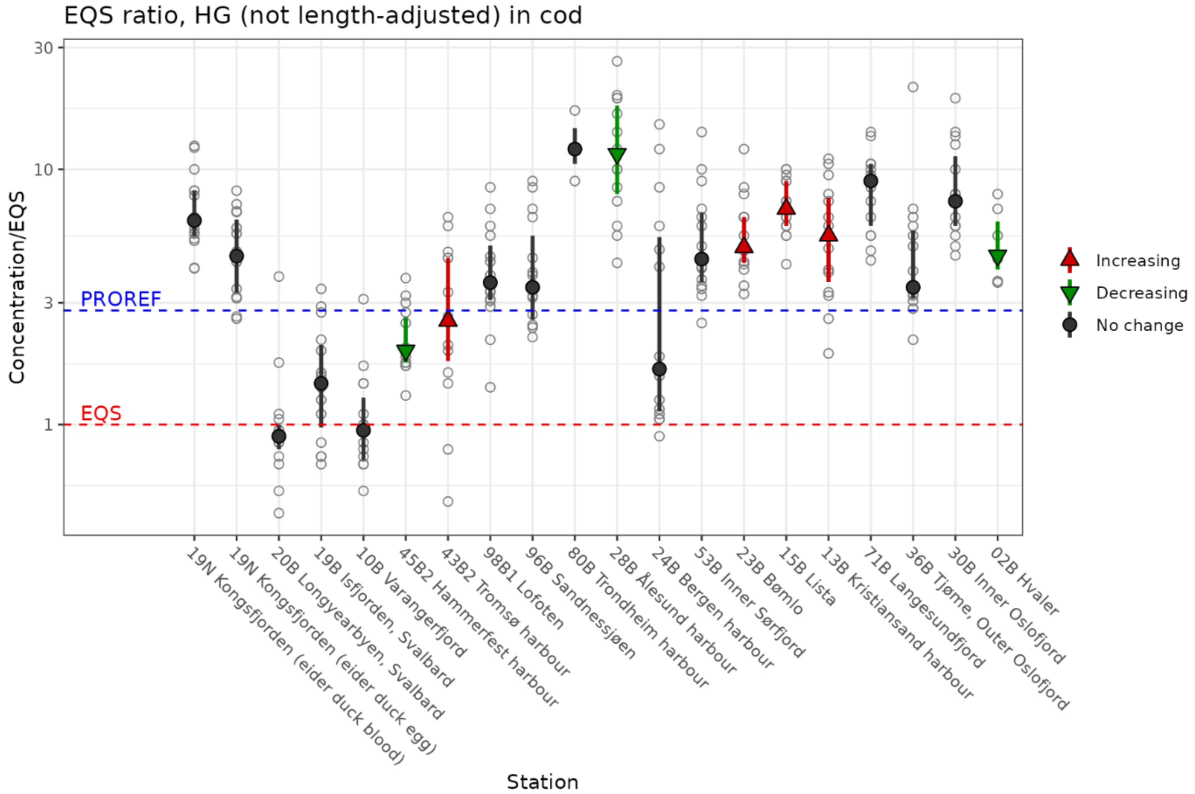


Figure 27. Ratio (concentrations divided by EQS) for mercury in (upper panel) blue mussel and (lower panel) cod and eider. The EQS ratios for cod are from fish which has not been length adjusted, while the time-trend symbols are for cod that have been length adjusted. *The y-axes for both mussel and cod are on a natural log scale*. The PROREF in lower panel is for cod, not eider, EQS applies both to cod and eider. For full explanation of figure see example **Figure 24**.

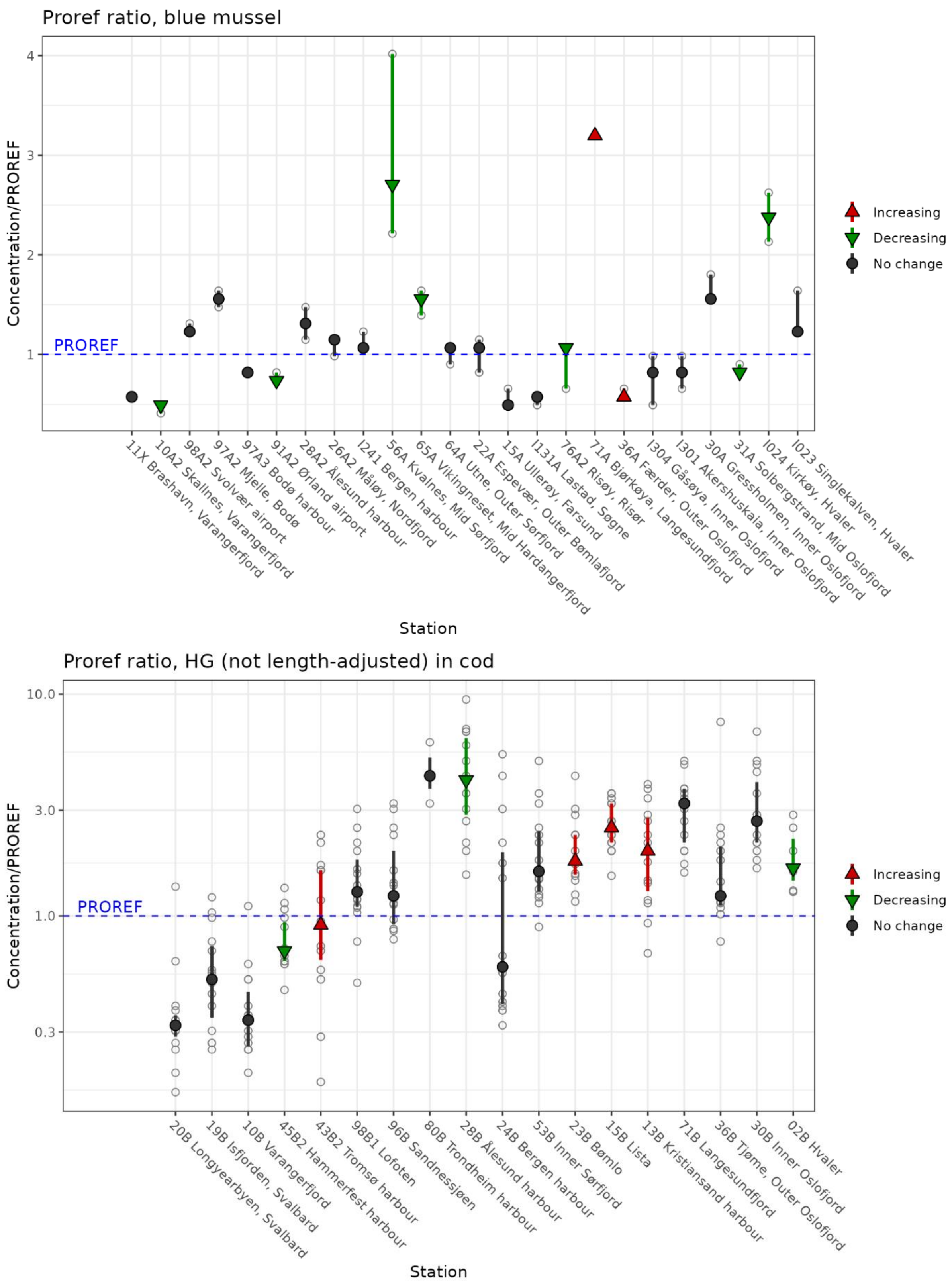


Figure 28. Ratio (concentrations divided by PROREF) for mercury in (upper panel) blue mussel and (lower panel) cod. The PROREF ratios for cod are from fish which has not been length adjusted, while the time-trend symbols are for cod that have been length adjusted. *The y-axis for cod is on a natural log scale*. For full explanation of figure see example **Figure 24**.

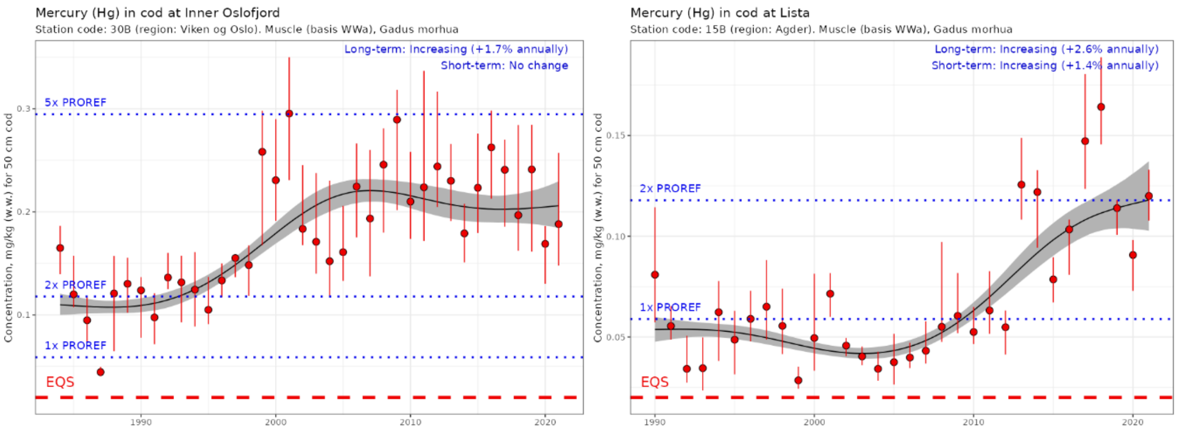


Figure 29. Two selected time trends for mercury in length adjusted cod. Left, mercury in cod at Inner Oslofjord (30B) and right, mercury in cod at Lista (15B). For full explanation of figure see example **Figure 25**.

Selected time trends

Figure 29 shows two selected time trends for length adjusted cod from the Inner Oslofjord (30B) and Lista (15B). At station 30B, the mercury concentration has increased 1.7% annually since mid-1980s, but the last ten years there has been no significant trend. At station 15B, there has been a higher annual percent increase, but starting from a lower concentration, the levels are lower at this station than 30B. The annual increase for mercury concentration at 15B long-term was 2.6% annually, and there is an ongoing increase (short-term trend) of 1.4% increase annually. The time trend shows that the long-term change did not start until after mid-2000s.

3.2.1.2 Cadmium (Cd)

Cadmium (Cd) is a naturally occurring heavy metal. Sources are agricultural, industrial emissions and long-range air pollutants and Cd is naturally found in small quantities in the earth's crust. In the present study, Cd was analysed in blue mussel at 24 stations, in cod liver at 18 stations, and in eider blood and eggs at one station (**Table 1**, **Figure 30**).

EQS

There is no EQS for cadmium in biota (**Table 3**).

PROREF

For mussel stations, concentrations of cadmium were mostly below PROREF, but seven stations had median concentrations exceeding PROREF between 1-2x (**Figure 31**). Station Skallnes in the Varangerfjord (10A2) had the highest median exceedances (2-3 times).

For cod stations it was the similar picture as for mussels, the concentrations of cadmium were mostly below PROREF, but four stations had median concentrations exceeding PROREF 1-2x. Station Trondheim harbour (80B) had the highest exceedances of PROREF (2x).

Long-term trends

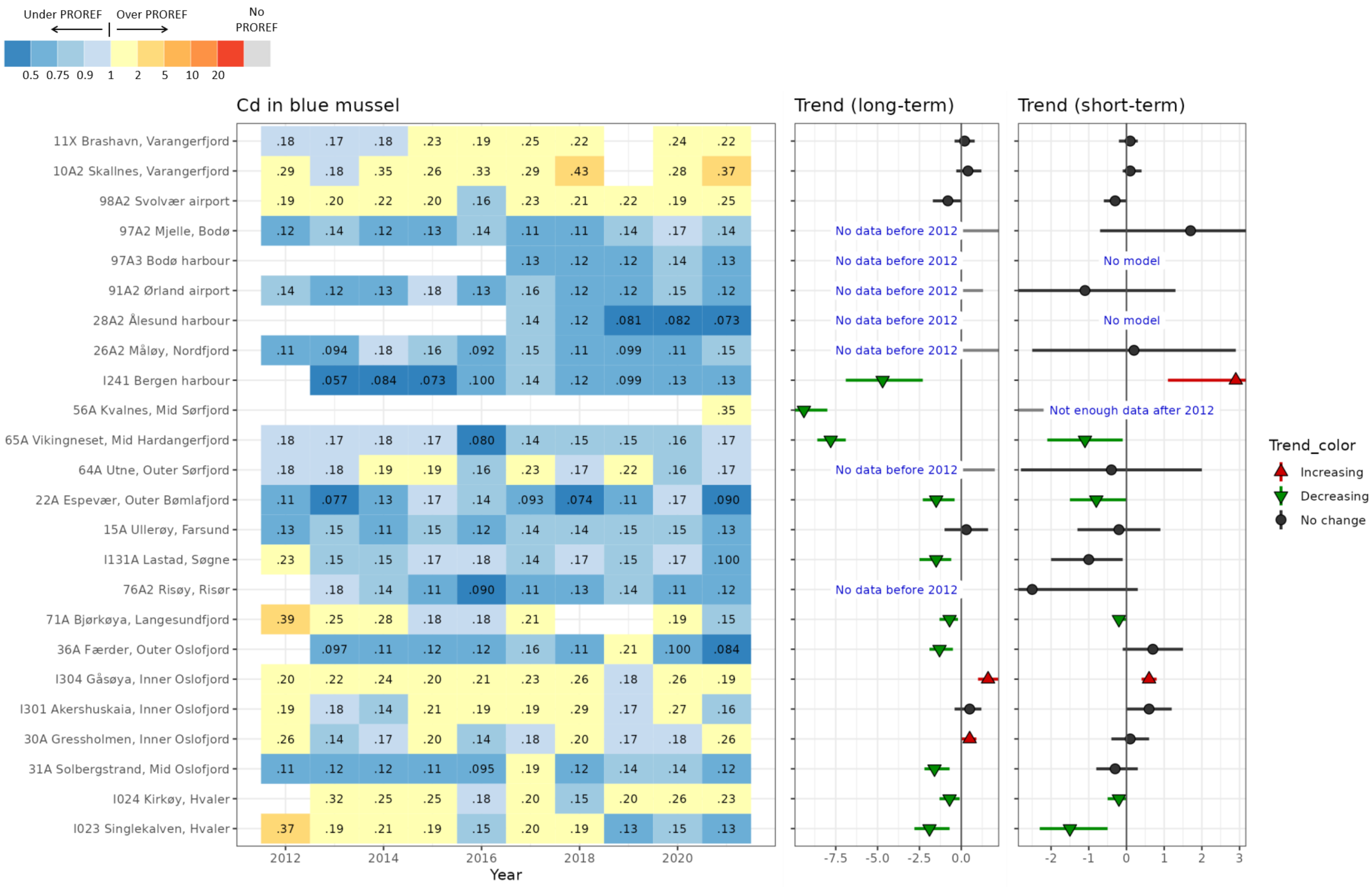
Significant long-term trends in mussel were mostly decreasing (10 stations), but two increasing long-term trends were also observed (Gressholmen (30A) and Gåsøya (I304) in the Inner Oslofjord (**Figure 30**).

Significant decreasing long-term trends in cod were observed at four stations (Tjøme in the Outer Oslofjord (36B), the Inner Sørfjord (53B), Lofoten (98B1) and the Varangerfjord (10B), while one station (the Inner Oslofjord (30B)) had significant increasing long-term trend.

Short-term trends

For short-term time trends in mussel, mostly no trends were observed, but six decreasing trends were observed as well as two increasing trends (Bergen harbour (I241) and Gåsøya in the Inner Oslofjord (I304), **Figure 30**).

Two decreasing short-term trends in cod were estimated (the Inner Oslofjord (30B) and Langesundfjord (71B)), while two stations (Tjøme in the Outer Oslofjord (36B) and the Varangerfjord (10B)) had significant increasing short-term for concentrations in cod. Higher short-term changes were observed at two other stations (Sandnessjøen (96B) and Trondheim harbour (80B) with almost 4% and 5% annual increase). However, these changes were not statistically significant due to wide confidence bands for the trend models. The confidence band for both are crossing the 0-line (no changes), indicating that zero (no) change cannot be ruled out.



Station	2012	2013	2014	2015	2016	2017	2018	2019	2020	2021
11X Brashavn, Varangerfjord	.18	.17	.18	.23	.19	.25	.22		.24	.22
10A2 Skallnes, Varangerfjord	.29	.18	.35	.26	.33	.29	.43		.28	.37
98A2 Svolvær airport	.19	.20	.22	.20	.16	.23	.21	.22	.19	.25
97A2 Mjelle, Bodø	.12	.14	.12	.13	.14	.11	.11	.14	.17	.14
97A3 Bodø harbour						.13	.12	.12	.14	.13
91A2 Ørland airport	.14	.12	.13	.18	.13	.16	.12	.12	.15	.12
28A2 Ålesund harbour						.14	.12	.081	.082	.073
26A2 Måløy, Nordfjord	.11	.094	.18	.16	.092	.15	.11	.099	.11	.15
I241 Bergen harbour		.057	.084	.073	.100	.14	.12	.099	.13	.13
56A Kvalnes, Mid Sørfjord										.35
65A Vikingneset, Mid Hardangerfjord	.18	.17	.18	.17	.080	.14	.15	.15	.16	.17
64A Utne, Outer Sørfjord	.18	.18	.19	.19	.16	.23	.17	.22	.16	.17
22A Espevær, Outer Bømlafjord	.11	.077	.13	.17	.14	.093	.074	.11	.17	.090
15A Ulleröy, Farsund	.13	.15	.11	.15	.12	.14	.14	.15	.15	.13
I131A Lastad, Søgne	.23	.15	.15	.17	.18	.14	.17	.15	.17	.100
76A2 Risøy, Risør		.18	.14	.11	.090	.11	.13	.14	.11	.12
71A Bjørkøya, Langesundfjord	.39	.25	.28	.18	.18	.21			.19	.15
36A Færder, Outer Oslofjord		.097	.11	.12	.12	.16	.11	.21	.100	.084
I304 Gåsøya, Inner Oslofjord	.20	.22	.24	.20	.21	.23	.26	.18	.26	.19
I301 Akershuskaia, Inner Oslofjord	.19	.18	.14	.21	.19	.19	.29	.17	.27	.16
30A Gressholmen, Inner Oslofjord	.26	.14	.17	.20	.14	.18	.20	.17	.18	.26
31A Solbergstrand, Mid Oslofjord	.11	.12	.12	.11	.095	.19	.12	.14	.14	.12
I024 Kirkøy, Hvaler		.32	.25	.25	.18	.20	.15	.20	.26	.23
I023 Singlekalven, Hvaler	.37	.19	.21	.19	.15	.20	.19	.13	.15	.13

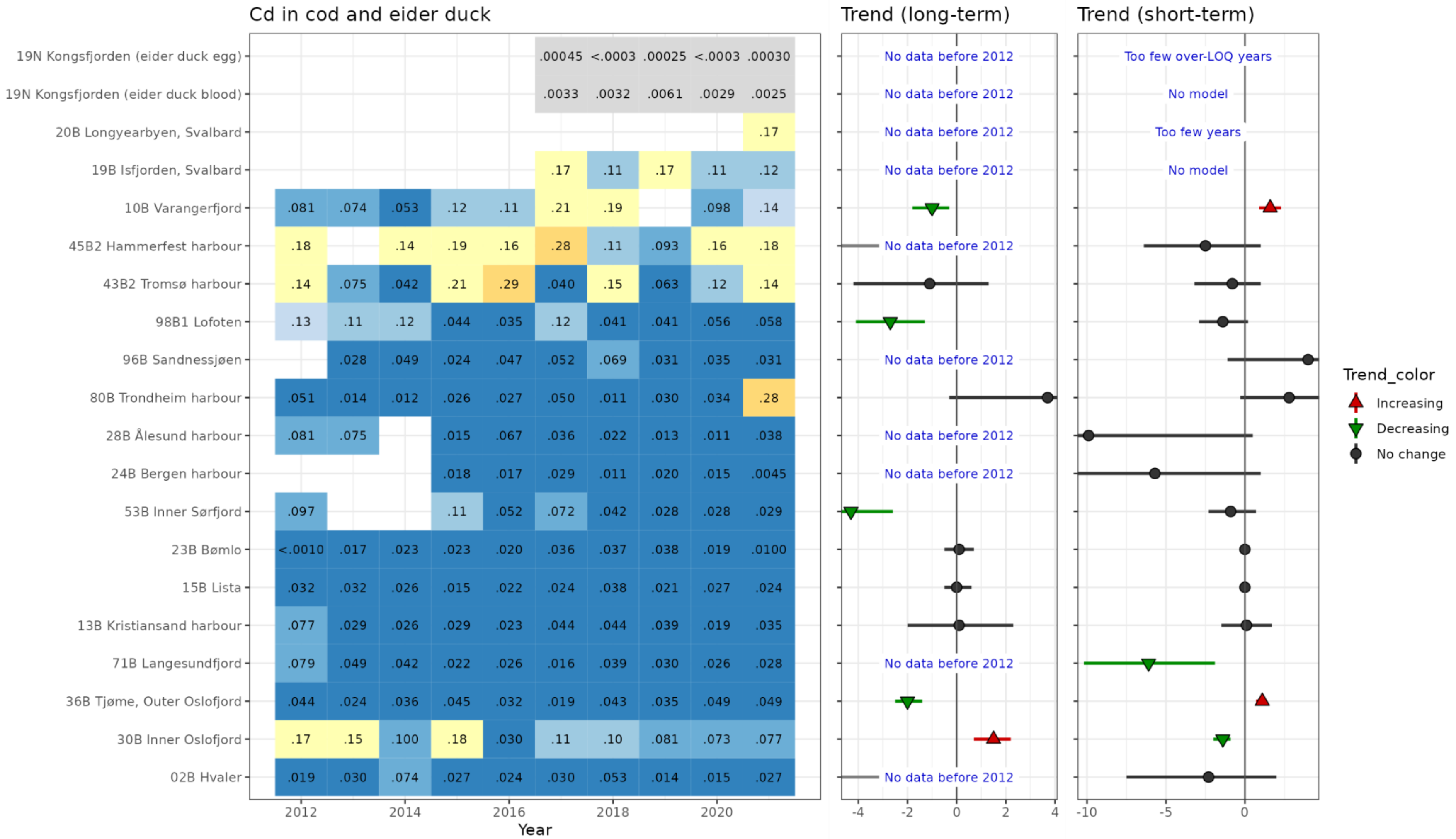


Figure 30. Heatmap and time trends of cadmium in (upper panel) blue mussel and (lower panel) cod and eider. For full explanation of figure see example **Figure 23**.

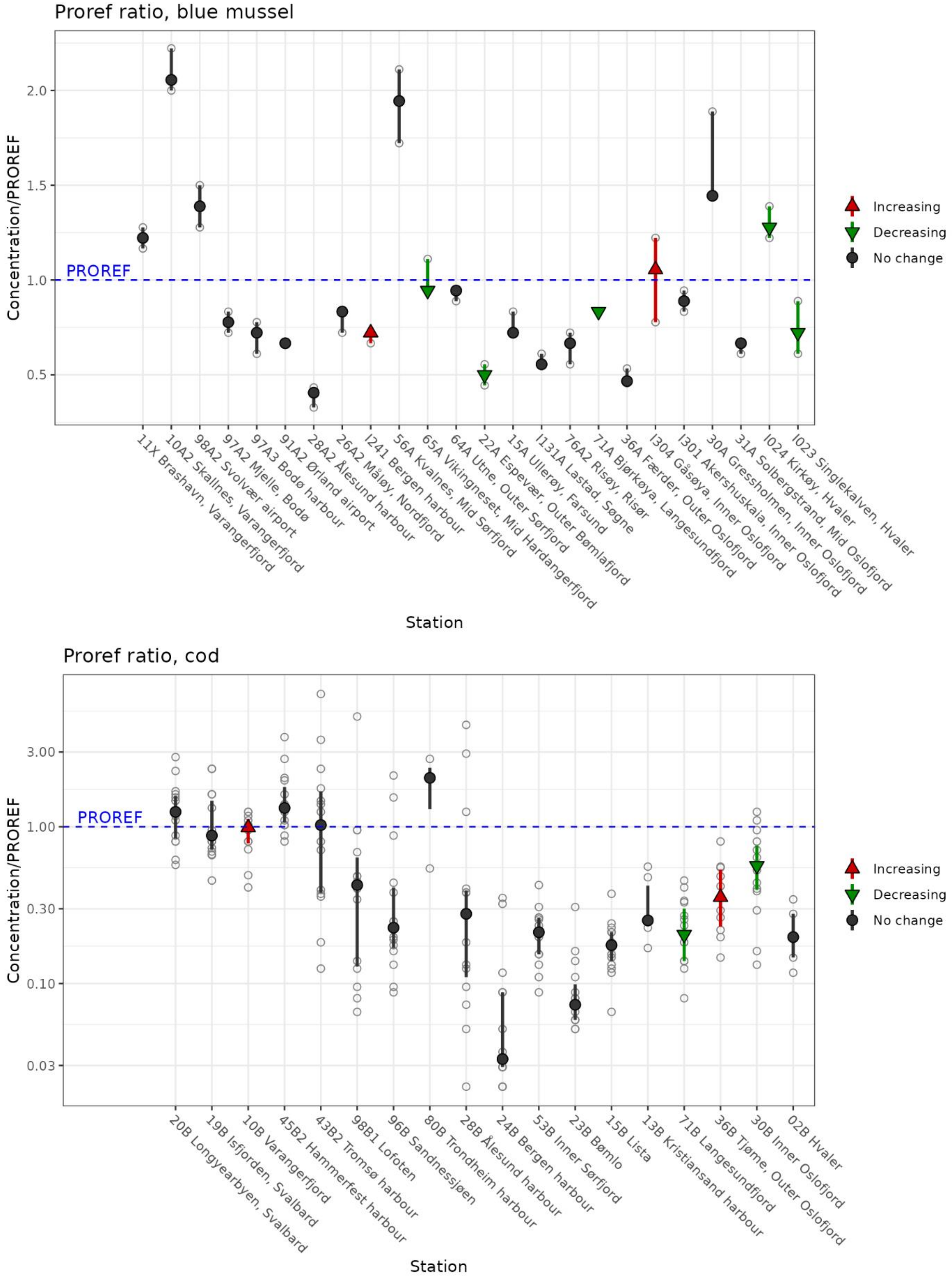


Figure 31. Ratio (concentrations/PROREF) for cadmium in (upper panel) blue mussel and (lower panel) cod. *The PROREF ratio for cod is on a natural log scale.* For full explanation of figure see example **Figure 24**.

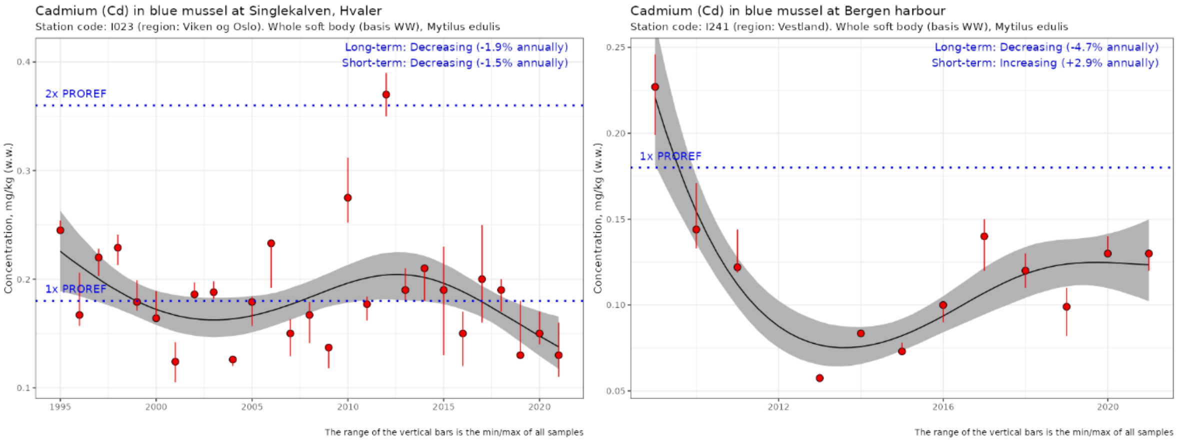


Figure 32. Two selected time trends for cadmium. For full explanation of figure see example **Figure 25**.

Selected time trends

Two selected time trends for cadmium concentrations in mussel are shown in **Figure 32**, at Singlekalven in Hvaler (I023) and Bergen harbour (I241). The long-term change at Hvaler was a long-term decrease of 1.9% annually since mid-1990. However, there was a temporary increase in concentrations peaking in 2013 (according to the model). After this temporary peak, a decreasing short-term trend was observed the last 10 years, with annual decrease of 1.5%. At Bergen harbour, the long-term trend was decreasing, but the short term was increasing since 2013 (2.9% annually).

3.2.1.3 Lead (Pb)

Lead (Pb) is a naturally occurring heavy metal present in small amounts in the earth's crust. Anthropogenic activities such as fossil fuels burning, mining, and manufacturing contributes to the release of high concentrations of lead. Lead has many different industrial, agricultural, and domestic applications. In the present study, Pb was analysed in blue mussel at 24 stations, in cod liver at 18 stations, and in eider blood and eggs at one station (**Table 1**, **Figure 33**).

EQS

There is no EQS for lead in biota (**Table 3**).

PROREF

For mussels, the dominating finding was that concentrations of lead were exceeding PROREF (14 stations, **Figure 34**). PROREF was exceeded the most (ca. 13x) at station Kvalnes in the Mid Sørfjord (56A), but also station Gressholmen in the Inner Oslofjord (30A) was many times higher than other stations.

At cod stations, no exceedances of PROREF were found. The highest concentrations were found at station Inner Oslofjord (30B).

Long-term trends

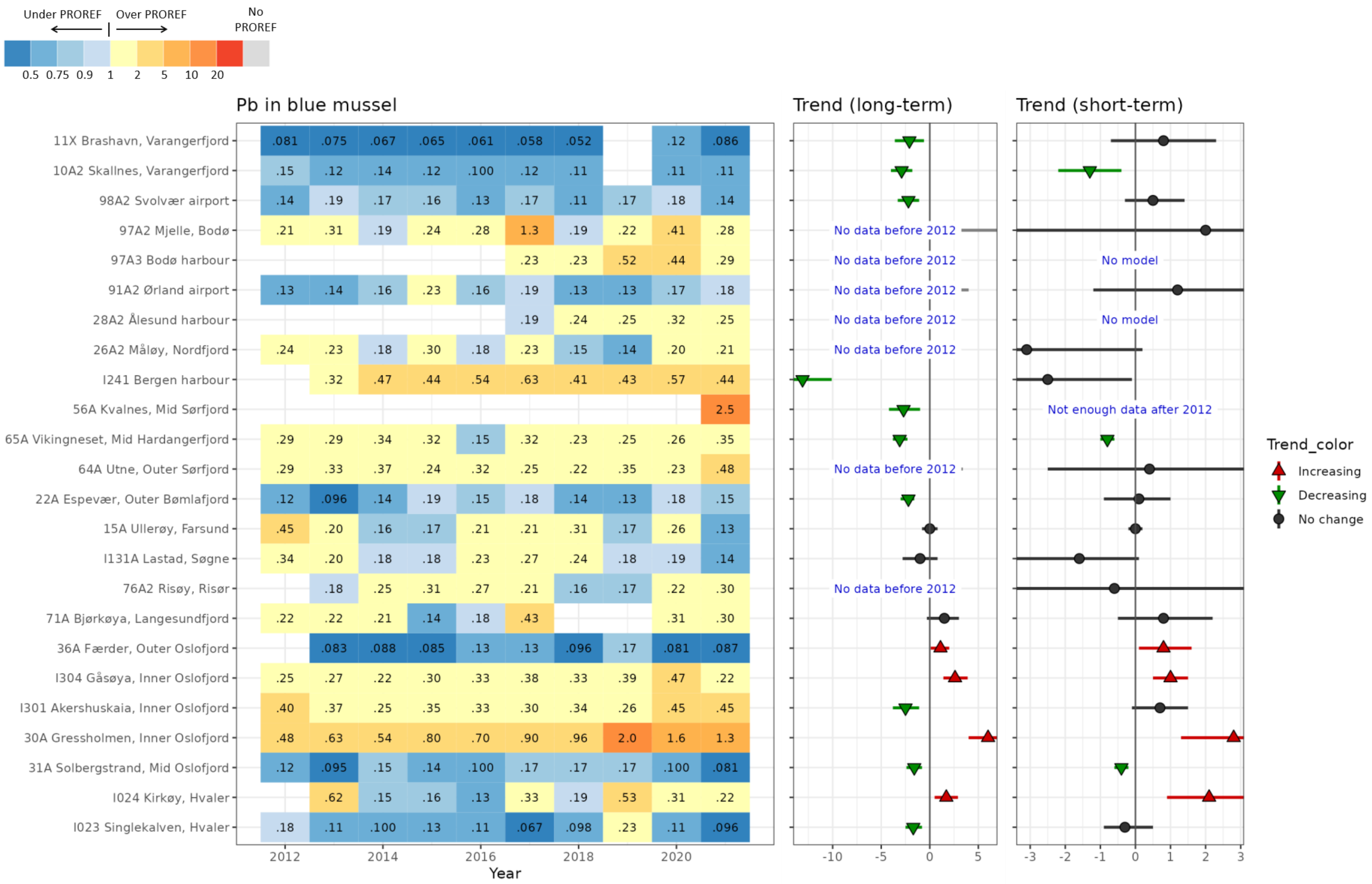
Long-term trends in mussel were mostly decreasing or no trends were observed (**Figure 33**), but a few increasing trends were observed at four stations (Færder in the Outer Oslofjord (36A), Gåsøya in the Inner Oslofjord (I304), Gressholmen in the Inner Oslofjord (30A) and Kirkøy at Hvaler (I024)). The increasing trends were highest at Gressholmen (30A) (50% annually).

Long-term trends in cod were only decreasing for stations where trends could be determined.

Short-term trends

Short-term trends in mussel were mostly no change (**Figure 33**), but a few decreasing trends were observed (Skallnes in the Varangerfjord (10A2), Kvalnes in the Mid Sørfjord (56A) and Vikingneset in the Mid Hardangerfjord (65A)). The same four stations that had increasing long-term trends, also had increasing short-term trends (Færder in the Outer Oslofjord (36A), Gåsøya in the Inner Oslofjord (I304), Gressholmen in the Inner Oslofjord (30A) and Kirkøy at Hvaler (I024)).

Trends in cod were mostly decreasing, but one increasing trend was found at station Tjøme in the Outer Oslofjord (36B), with increasing annual trend of almost 4%. However, the levels were still far below PROREF.


Under PROREF
Over PROREF
No PROREF
0.5 0.75 0.9 1 2 5 10 20
Pb in blue mussel
Trend (long-term)
Trend (short-term)
11X Brashavn, Varangerfjord
.081 .075 .067 .065 .061 .058 .052 .12 .086
10A2 Skallnes, Varangerfjord
.15 .12 .14 .12 .100 .12 .11 .11 .11
98A2 Svolvær airport
.14 .19 .17 .16 .13 .17 .11 .17 .18 .14
97A2 Mjelle, Bodø
.21 .31 .19 .24 .28 1.3 .19 .22 .41 .28
97A3 Bodø harbour
.23 .23 .52 .44 .29
91A2 Ørland airport
.13 .14 .16 .23 .16 .19 .13 .13 .17 .18
28A2 Ålesund harbour
.19 .24 .25 .32 .25
26A2 Måløy, Nordfjord
.24 .23 .18 .30 .18 .23 .15 .14 .20 .21
I241 Bergen harbour
.32 .47 .44 .54 .63 .41 .43 .57 .44
56A Kvalnes, Mid Sørfjord
2.5
65A Vikingneset, Mid Hardangerfjord
.29 .29 .34 .32 .15 .32 .23 .25 .26 .35
64A Utne, Outer Sørfjord
.29 .33 .37 .24 .32 .25 .22 .35 .23 .48
22A Espevær, Outer Bømlafjord
.12 .096 .14 .19 .15 .18 .14 .13 .18 .15
15A Ullerøy, Farsund
.45 .20 .16 .17 .21 .21 .31 .17 .26 .13
I131A Lastad, Søgne
.34 .20 .18 .18 .23 .27 .24 .18 .19 .14
76A2 Risøy, Risør
.18 .25 .31 .27 .21 .16 .17 .22 .30
71A Bjørkøya, Langesundfjord
.22 .22 .21 .14 .18 .43 .31 .30
36A Færder, Outer Oslofjord
.083 .088 .085 .13 .13 .096 .17 .081 .087
I304 Gåsøya, Inner Oslofjord
.25 .27 .22 .30 .33 .38 .33 .39 .47 .22
I301 Akershuskaia, Inner Oslofjord
.40 .37 .25 .35 .33 .30 .34 .26 .45 .45
30A Gressholmen, Inner Oslofjord
.48 .63 .54 .80 .70 .90 .96 2.0 1.6 1.3
31A Solbergstrand, Mid Oslofjord
.12 .095 .15 .14 .100 .17 .17 .17 .100 .081
I024 Kirkøy, Hvaler
.62 .15 .16 .13 .33 .19 .53 .31 .22
I023 Singlekalven, Hvaler
.18 .11 .100 .13 .11 .067 .098 .23 .11 .096
2012 2014 2016 2018 2020
Year
No data before 2012
No model
Not enough data after 2012
-10 -5 0 5
-3 -2 -1 0 1 2 3
Trend_color
Increasing
Decreasing
No change

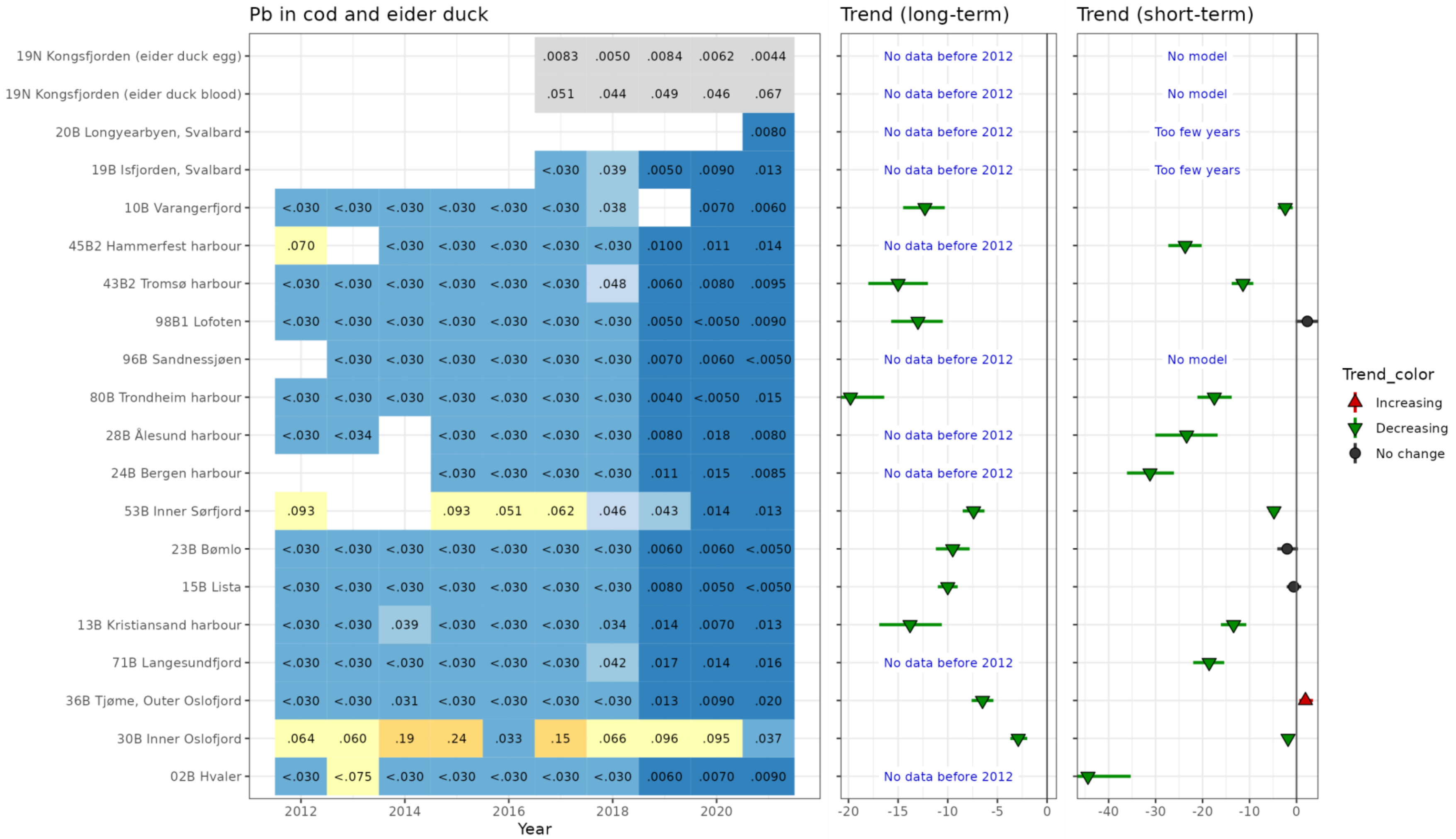


Figure 33. Heatmap and time trends of lead in (upper panel) blue mussel and (lower panel) cod and eider. For full explanation of figure see example **Figure 23**.

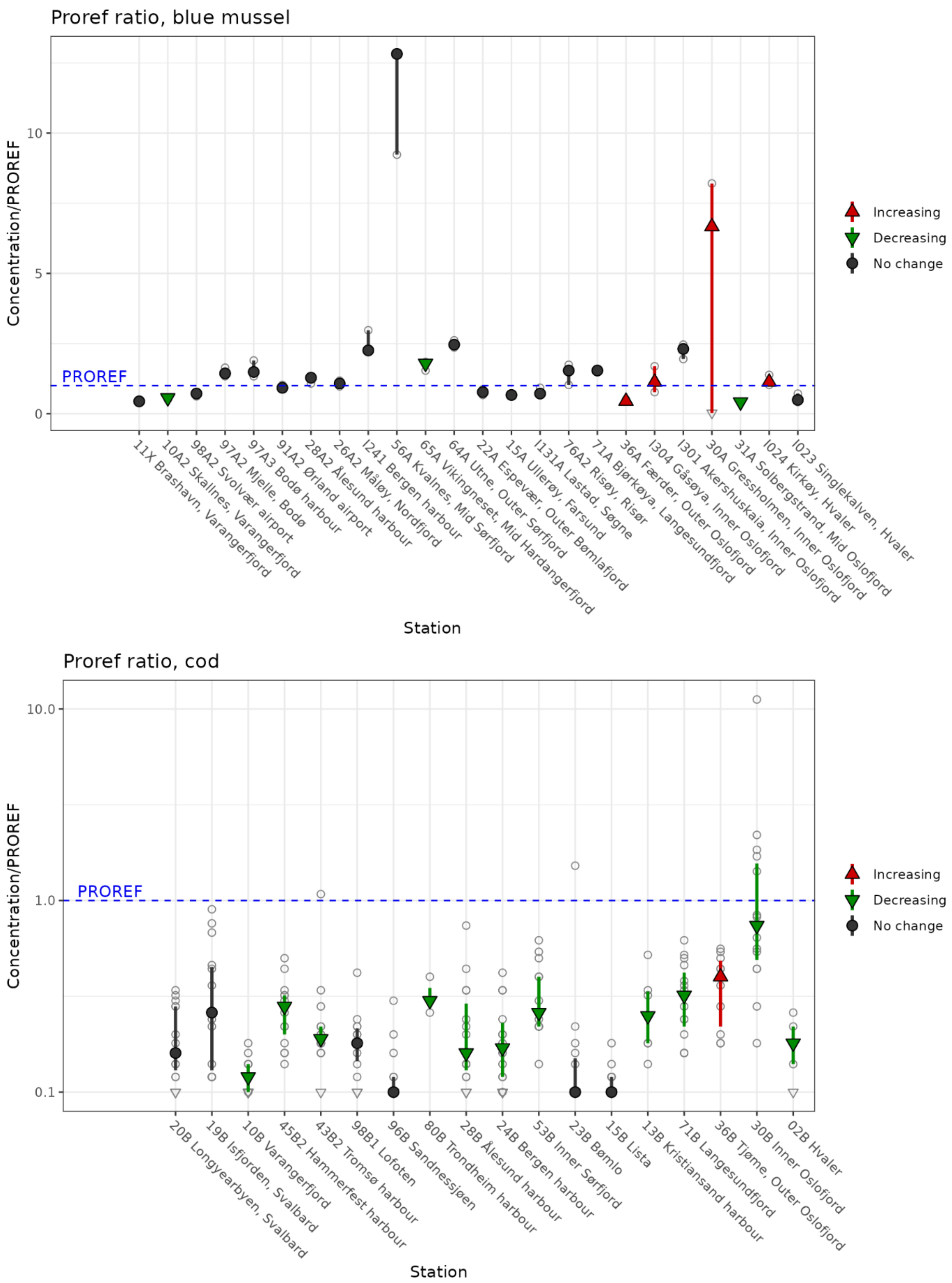


Figure 34. Ratio (concentrations/PROREF) for lead in (upper panel) blue mussel and (lower panel) cod. *The scale for cod is on a natural log scale.* For full explanation of figure see example **Figure 24**.

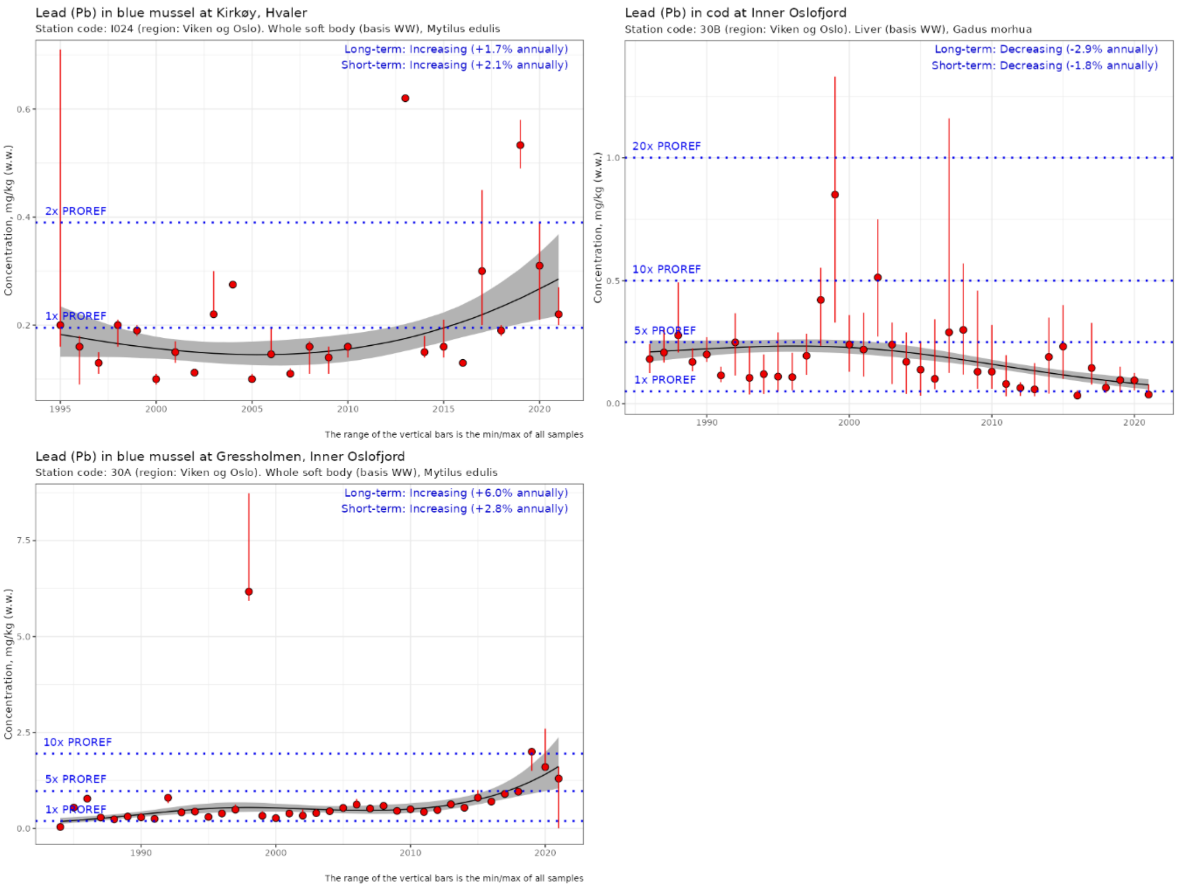


Figure 35. Three selected time trends for lead. The time trend for station Gressholmen (30A) is on a *natural log scale*. For full explanation of figure see example **Figure 25**.

Selected time trends

Three selected time trends are shown in **Figure 35**. Lead in mussel at station Kirkøy at Hvaler (I204) shows that the concentration has had an annual increase in both long-term and short-term (1.7% and 2.1%, respectively). Both decreasing long- and short-term trends observed in cod at station 30B mean that Inner Oslofjord in 2021 were below PROREF. The annual short-term decrease was ca. 2% annually. The mussel at station Gressholmen in the Inner Oslofjord (30A) increased the most (6% long-term annual increase), and concentrations are now approaching 10x PROREF.

3.2.1.4 Arsenic (As)

Arsenic (As) is a heavy metal that is detected at low concentrations in virtually all environmental matrices. Environmental pollution by arsenic occurs as a result of natural phenomena such as volcanic eruptions and soil erosion, and anthropogenic activities. Several arsenic-containing compounds are produced industrially and have been used to manufacture products with agricultural applications such as insecticides, herbicides, fungicides, algicides, sheep dips, wood preservations and dyestuff. The use of arsenic and arsenic compounds to prevent fouling of ships and equipment in water, for the treatment of water in industry, for wood impregnation and sale of treated wood, is prohibited through the REACH Regulation (Annex XVII, item 19). From 2002, it was forbidden to produce and sell chromated copper arsenate (CCA) impregnated wood in Norway, but chromium, copper and arsenic continue to leak from old wood. Therefore, it is assumed that impregnated wood is still the largest source of arsenic emissions in Norway. Large quantities are still found in, among other places, wharves, terrace floors and play equipment, and the emissions are therefore still significant. As evidenced from national monitoring activities, atmospheric long-range environmental transport of arsenic to Norway has decreased sharply since the 1970s (State of the Environment Norway[5]). In the present study, As was analysed in blue mussel at 24 stations, in cod liver at 18 stations, and in eider blood and eggs at one station (**Table 1**, **Figure 36**).

EQS

There is no EQS for arsenic in biota (**Table 3**).

PROREF

Most mussel stations were below PROREF, but seven stations exceeded PROREF. Highest exceedances were observed at Utne in the Outer Sørfjord (64A) and Vikingneset in the Mid Hardangerfjord (65A, **Figure 36** and **Figure 37**).

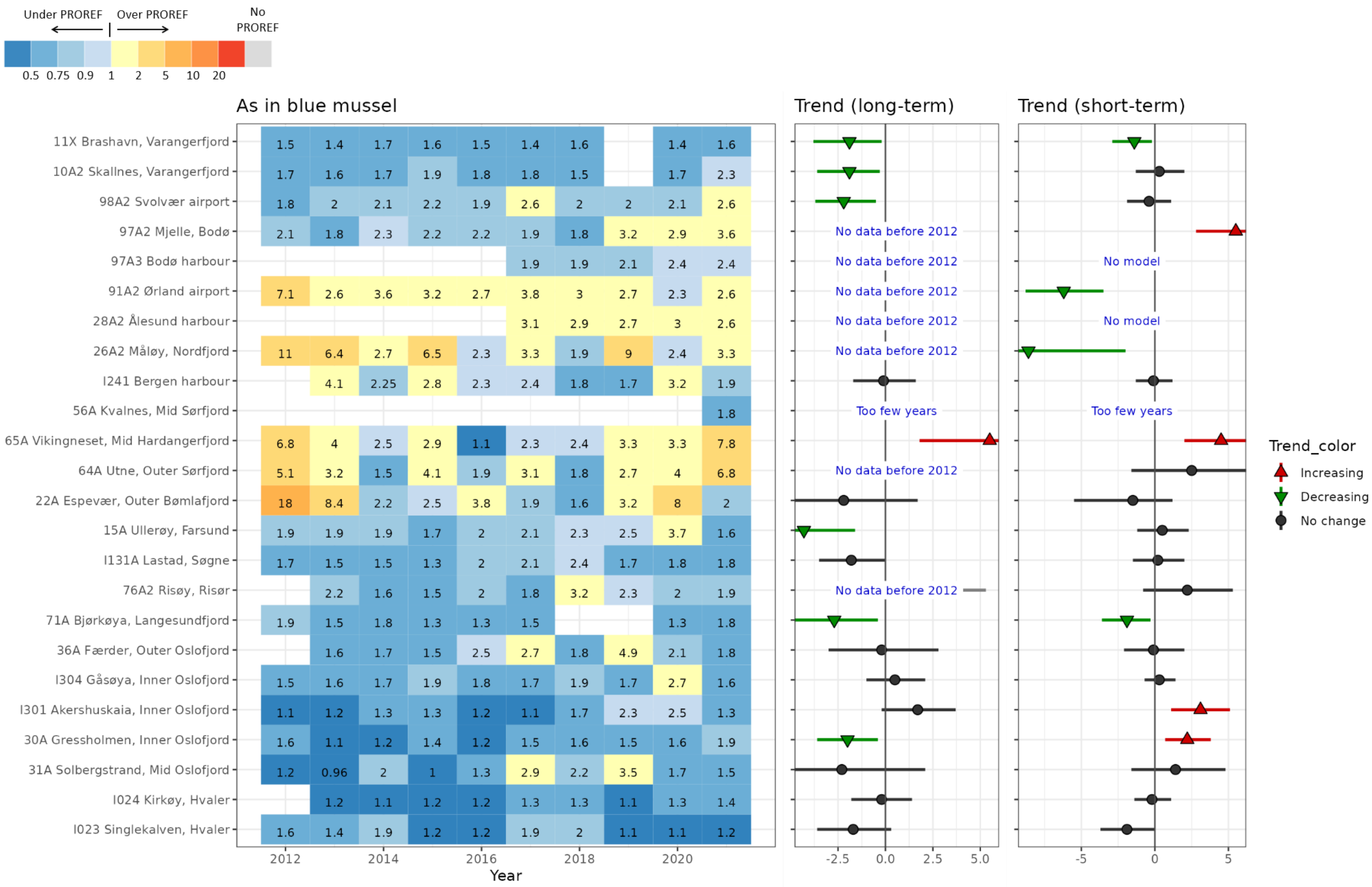
Concentrations of arsenic in cod did not exceed PROREF except at the two stations in the Inner Oslofjord (30B) and Trondheim harbour (80B).

Time trends

Long-term trends in mussels were mostly no trends or significant decreasing trends (six stations, **Figure 36**). One mussel station had significant increasing long-term trend (Vikingneset in the Mid Hardangerfjord (65A)). Short-term time trends were also mostly no trends. Three stations had significant increasing short-term time trends (Gressholmen in the Inner Oslofjord (30A), Akershuskaia in the Inner Oslofjord (I301) and Vikingneset in the Mid Hardangerfjord (65A)), while also tree stations had significant decreasing time trends (Bjørkøya in the Langesundfjord (71A), Måløy in the Nordfjord (26A2) and Ørland airport (91A2)).

For arsenic concentrations in cod there were four stations with no trends and four stations with significant decreasing time trends (the Inner Oslofjord (30B), Bømlo (23B), the Inner Sørfjord (53B) and Tromsø harbour (43B2)). No trends dominated the short-term time trends in cod, but three stations had increasing short-term time trends (Lista (15B), Trondheim harbour (80B) and Sandnessjøen (96B)) and four had decreasing short-term trends (Bømlo (23B), the Inner Sørfjord (53B), Ålesund harbour (28B) and Lofoten (98B1)).

[5] https://miljostatus.miljodirektoratet.no/tema/miljogifter/prioriterte-miljogifter/arsen-og-arsenforbindelser/



Under PROREF
Over PROREF
No PROREF
0.5 0.75 0.9 1 2 5 10 20
As in blue mussel
Trend (long-term)
Trend (short-term)
11X Brashavn, Varangerfjord
10A2 Skallnes, Varangerfjord
98A2 Svolvær airport
97A2 Mjelle, Bodø
97A3 Bodø harbour
91A2 Ørland airport
28A2 Ålesund harbour
26A2 Måløy, Nordfjord
I241 Bergen harbour
56A Kvalnes, Mid Sørfjord
65A Vikingneset, Mid Hardangerfjord
64A Utne, Outer Sørfjord
22A Espevær, Outer Bømlafjord
15A Ullerøy, Farsund
I131A Lastad, Søgne
76A2 Risøy, Risør
71A Bjørkøya, Langesundfjord
36A Færder, Outer Oslofjord
I304 Gåsøya, Inner Oslofjord
I301 Akershuskaia, Inner Oslofjord
30A Gressholmen, Inner Oslofjord
31A Solbergstrand, Mid Oslofjord
I024 Kirkøy, Hvaler
I023 Singlekalven, Hvaler
No data before 2012
No model
Too few years
2012 2014 2016 2018 2020
Year
-2.5 0.0 2.5 5.0
-5 0 5
Trend_color
Increasing
Decreasing
No change

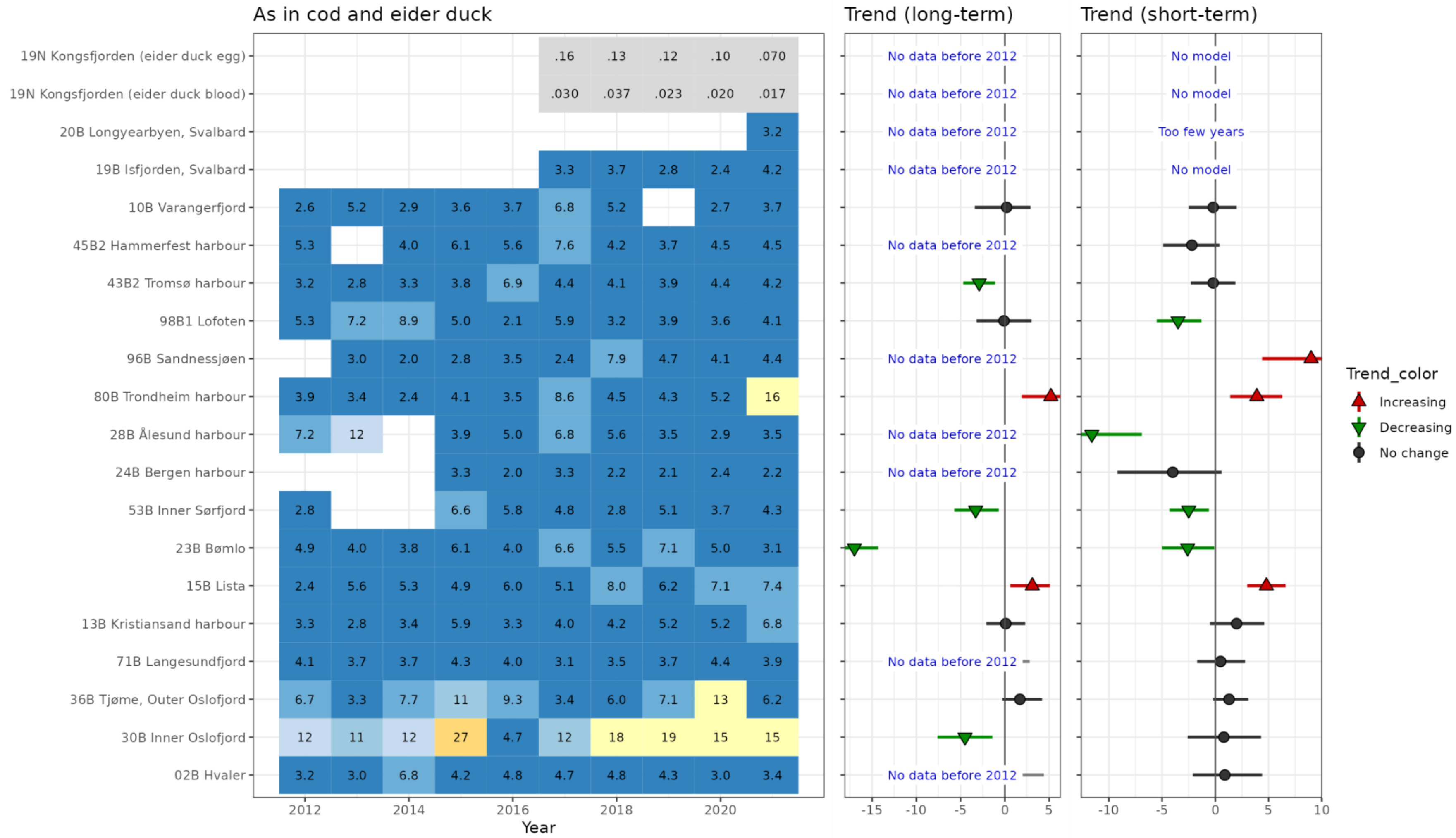


Figure 36. Heatmap and time-trends of arsenic in (upper panel) blue mussel and (lower panel) cod and eider. For full explanation of figure see example **Figure 23**.

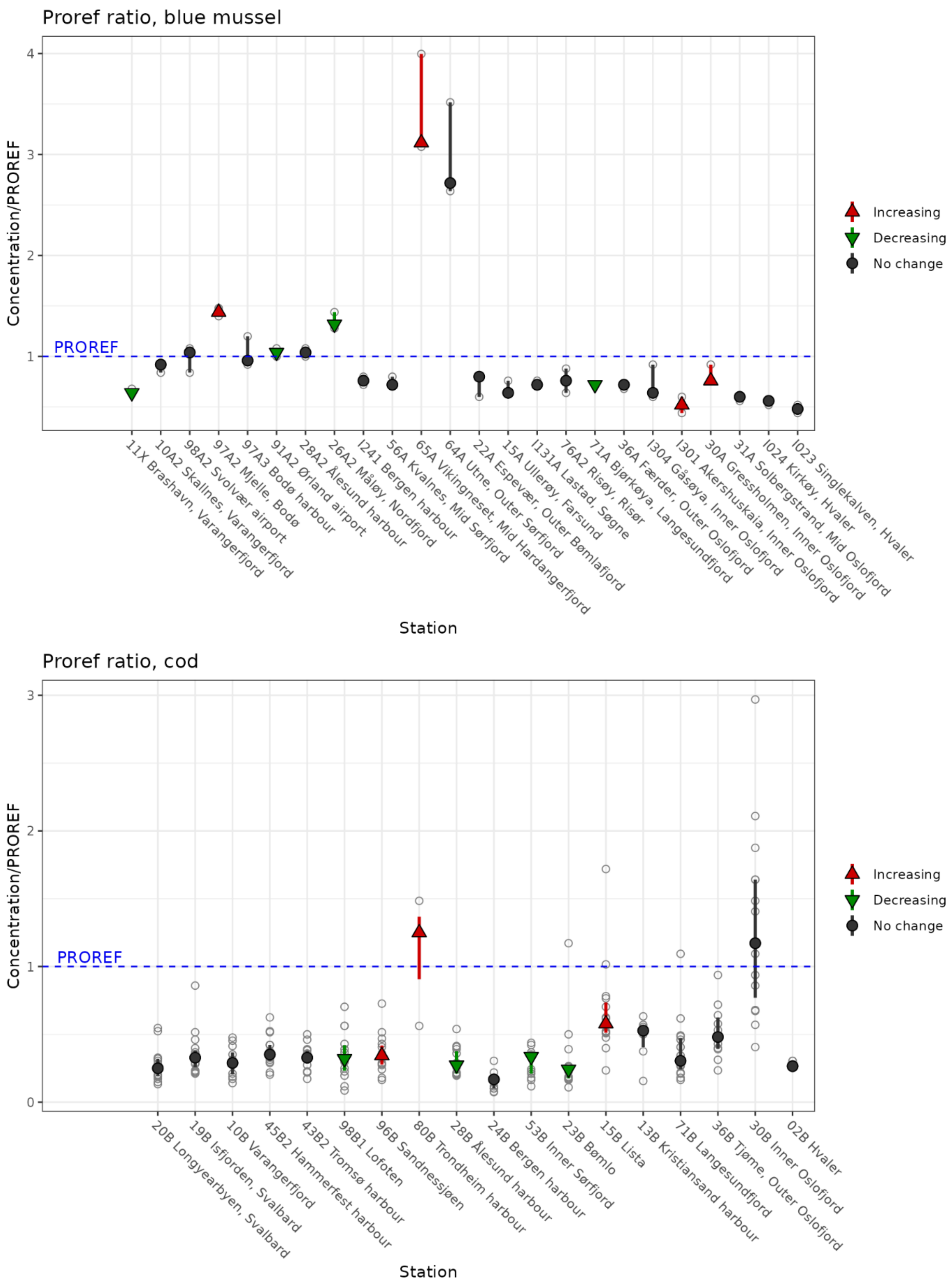


Figure 37. Ratio (concentrations/PROREF) for arsenic in (upper panel) blue mussel and (lower panel) cod. For full explanation of figure see example **Figure 24**.

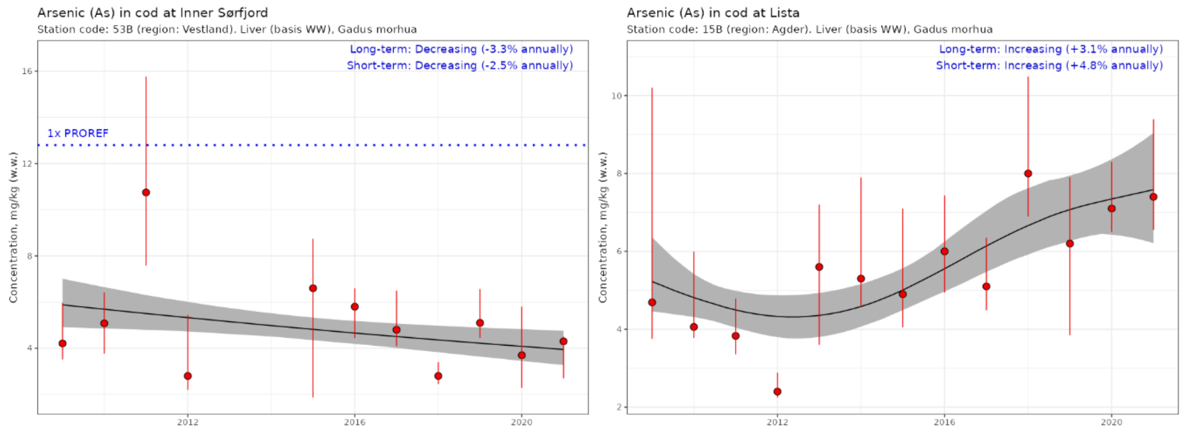


Figure 38. Two selected time trends for arsenic. For full explanation of figure see example **Figure 25**.

Selected time trends

Two time trends for arsenic in cod are shown in **Figure 38**. Arsenic in cod at station Inner Sørfjord (53B) showed decreasing trends, both long-term and short-term (3.3% and 2.5% annually). In cod at Lista (15B), there were both increasing long-term and short-term trends (3.1% and 4.8% annually).

3.2.1.5 Silver (Ag)

Silver (Ag) is an element. Possible sources are the iron and steel industry, cement industry, mining, and landfills. Silver is used as active substance in biocidal products and in treated articles. Under the biocidal product regulations all active substances must be authorized to be permitted to be placed on the market. Only silver uses which show acceptable risks, get authorized. Evaluation of silver as an active substance within the biocidal product regulations is ongoing. Pending the outcome of this evaluation, four silver compounds are permitted used as biocides in the EU/ EEA-region. Discharges of wastewater treatment plants (WWTP) and discharges from mine tailings are considered major and important sources for Ag to the aquatic environment (Tappin et al. 2010). The Ag nanoparticles from consumer products is important in terms of inputs to wastewater treatment plants (Nowack 2010). In the present study, Ag was analysed in blue mussel at 24 stations, in cod liver at 18 stations, and in eider blood and eggs at one station (**Table 1**, **Figure 39**).

EQS

There is no EQS for silver in biota (**Table 3**).

PROREF

Silver concentrations in mussels could not be assessed for exceedance of PROREF since the LOQ was higher than the PROREF.

In cod, silver concentrations exceeded PROREF in four stations (the Inner Oslofjord (30B), Kristiansand harbour (13B), Lista (15B) and Trondheim harbour (80B), most at 30B (3x) and 80B (4x) (**Figure 40**).

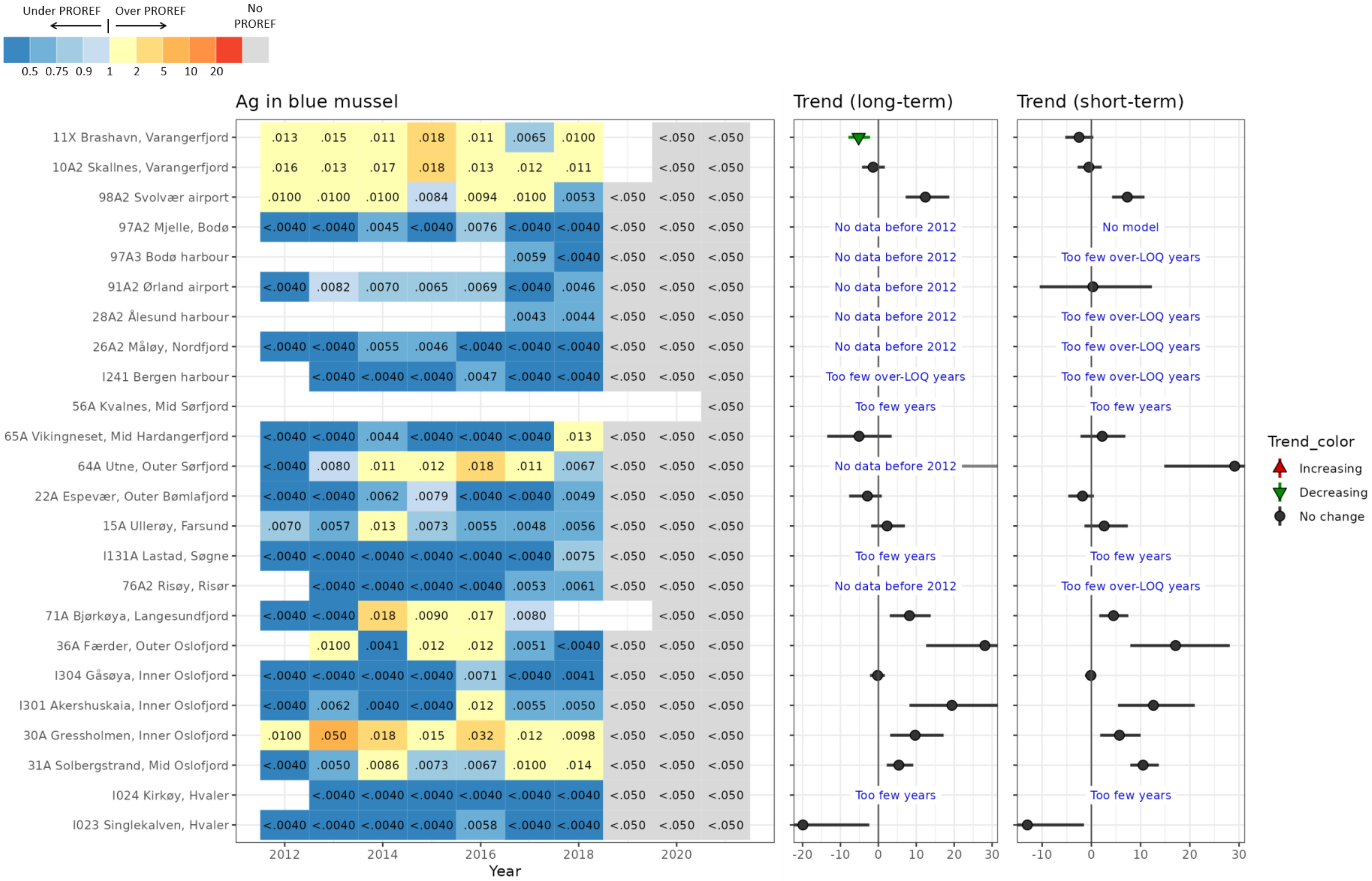
Long-term trends

One station had significant decreasing long-term trend (Brashavn in the Varangerfjord (11X)), while most stations had no trends. Three increasing trends were estimated, but since the LOQ have increased since 2019, the time trends in blue mussel are uncertain.

Cod concentrations had increasing long-term trends at seven stations (the Inner Oslofjord (30B), Lista (15B), the Inner Sørfjord (53B), Trondheim harbour (80B), Lofoten (98B1), Tromsø harbour (43B2) and the Varangerfjord (10B)), a few had no trends and no stations had decreasing trends.

Short-term trends

Since the LOQ for silver has increased since 2019, the time trends in blue mussel are uncertain. Silver concentrations in cod increased for short-term trends at eight stations while no trends were observed at five stations. Decreasing short-term trends were observed at three stations, but decreases were small compared to increases.


Under PROREF
Over PROREF
No PROREF
0.5 0.75 0.9 1 2 5 10 20
Ag in blue mussel
Trend (long-term)
Trend (short-term)
11X Brashavn, Varangerfjord
10A2 Skallnes, Varangerfjord
98A2 Svolvær airport
97A2 Mjelle, Bodø
97A3 Bodø harbour
91A2 Ørland airport
28A2 Ålesund harbour
26A2 Måløy, Nordfjord
I241 Bergen harbour
56A Kvalnes, Mid Sørfjord
65A Vikingneset, Mid Hardangerfjord
64A Utne, Outer Sørfjord
22A Espevær, Outer Bømlafjord
15A Ullerøy, Farsund
I131A Lastad, Søgne
76A2 Risøy, Risør
71A Bjørkøya, Langesundfjord
36A Færder, Outer Oslofjord
I304 Gåsøya, Inner Oslofjord
I301 Akershuskaia, Inner Oslofjord
30A Gressholmen, Inner Oslofjord
31A Solbergstrand, Mid Oslofjord
I024 Kirkøy, Hvaler
I023 Singlekalven, Hvaler
No data before 2012
No model
Too few over-LOQ years
Too few years
Year
2012 2014 2016 2018 2020
-20 -10 0 10 20 30
-10 0 10 20 30
Trend_color
Increasing
Decreasing
No change

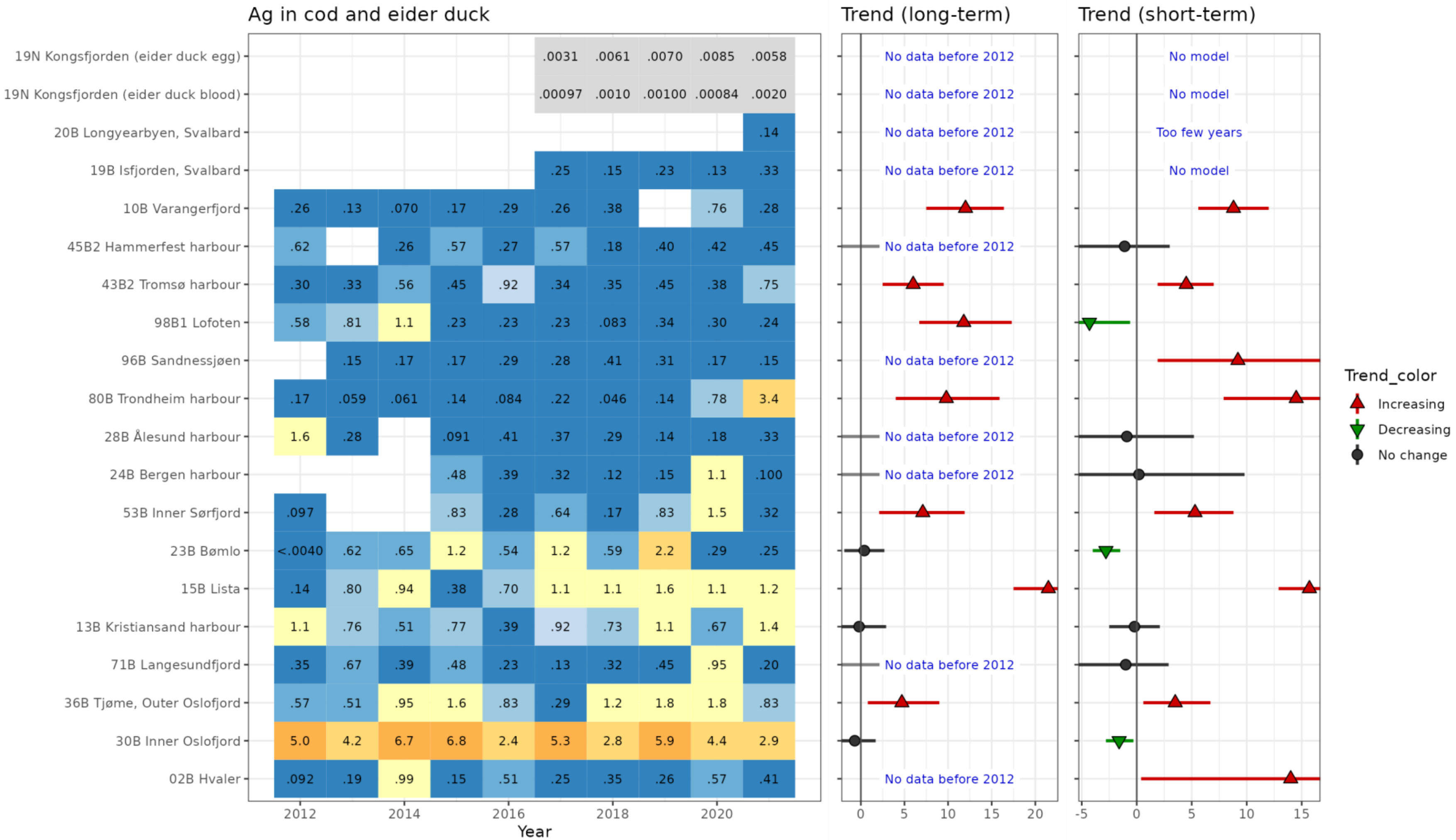


Figure 39. Heatmap and time trends of silver in (upper panel) blue mussel and (lower panel) cod and eider. For full explanation of figure see example **Figure 23**.

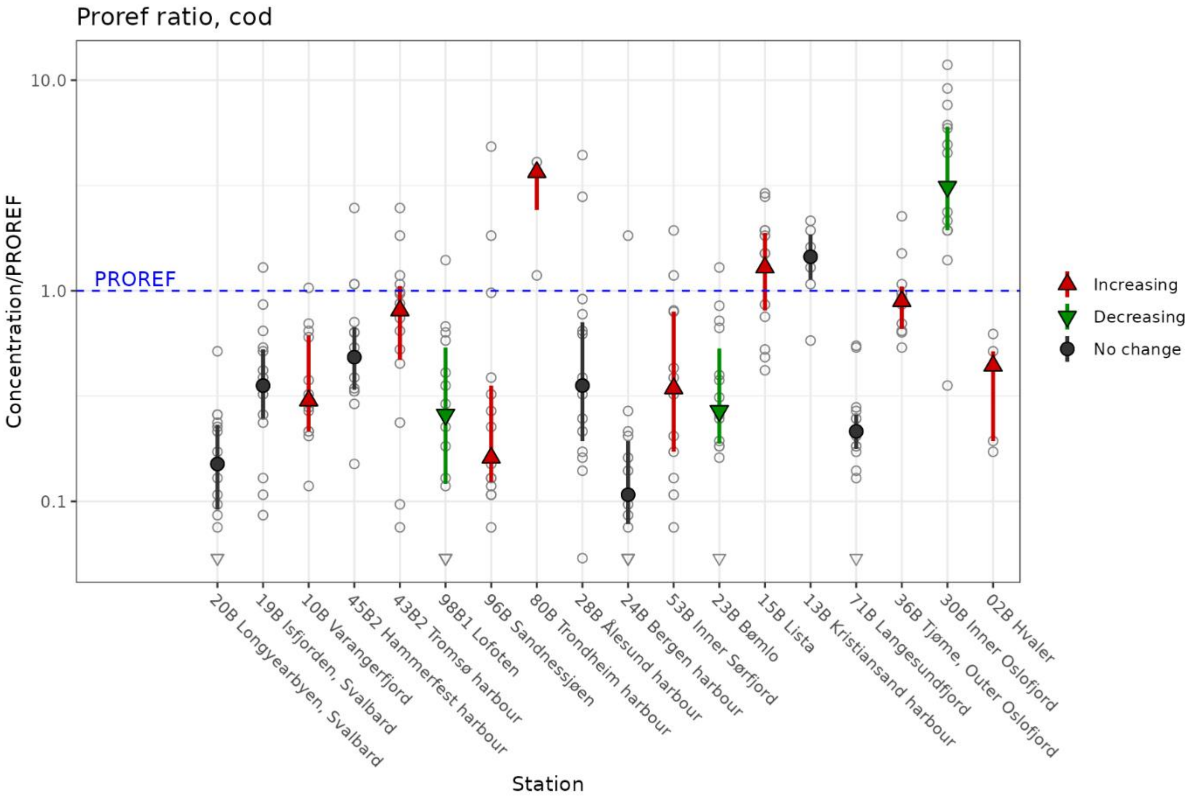


Figure 40. Ratio (concentrations/PROREF) for silver in cod (data for blue mussel are not shown since all were below LOQ). *The y-axis is on a natural log scale.* For full explanation of figure see example **Figure 24**.

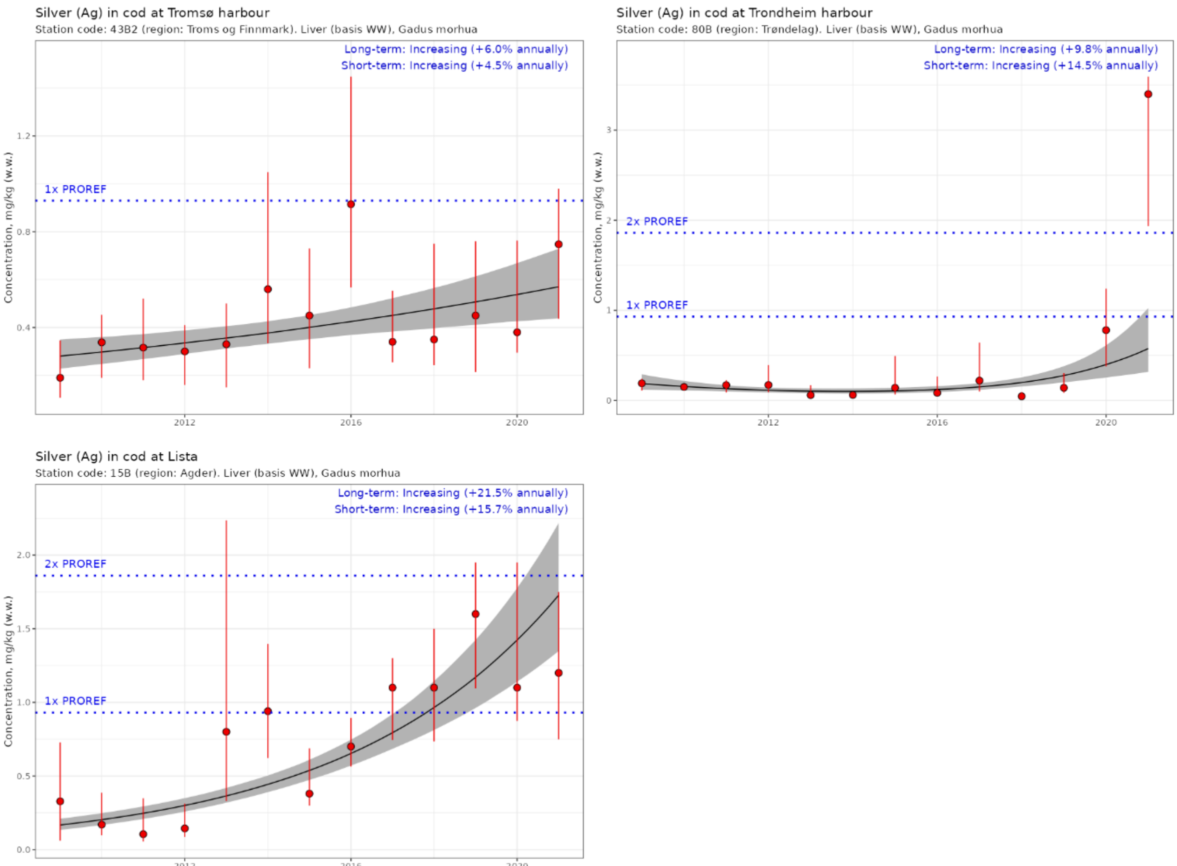


Figure 41. Three selected time trends for silver. For full explanation of figure see example **Figure 25**.

Selected time trends

Three selected time trends for silver concentrations in cod are shown in **Figure 41**. Silver concentrations are increasing in cod long-term and short-term for all stations (Tromsø harbour (43B2), Trondheim harbour (80B), and Lista (15B). The increases were highest at Lista (21% long-term and 16% short-term annually). At Trondheim harbour this year's median was higher than previous years and could be because only three cod were caught in 2021. The trends should therefore be treated with caution until results can be confirmed next year.

3.2.1.6 Chromium (Cr)

Chromium (Cr) is an element found in several forms that have different toxicities. In the past, wood was often impregnated with Cr. From 2002, it was forbidden to produce and sell CCA-impregnated wood in Norway, but Cr, copper and arsenic continue to leak from old wood. Impregnated wood is therefore still the largest source of Cr emissions in Norway, accounting for around 63% of emissions in 2019. In the present study, Cr was analysed in blue mussel at 24 stations, in cod liver at 18 stations, and in eider blood and eggs at one station (**Table 1**, **Figure 42**).

EQS

There is no EQS for chromium in biota (**Table 3**).

PROREF

No exceedance of PROREF dominated chromium concentrations in mussels (**Figure 42**, **Figure 43**). Six stations had chromium concentrations exceeding PROREF, with station Gressholmen in the Inner Oslofjord (30A) having the highest exceedances.

No median concentrations of chromium in cod exceeded PROREF, except for a single sample at station Sandnessjøen (96B).

Long-term trends

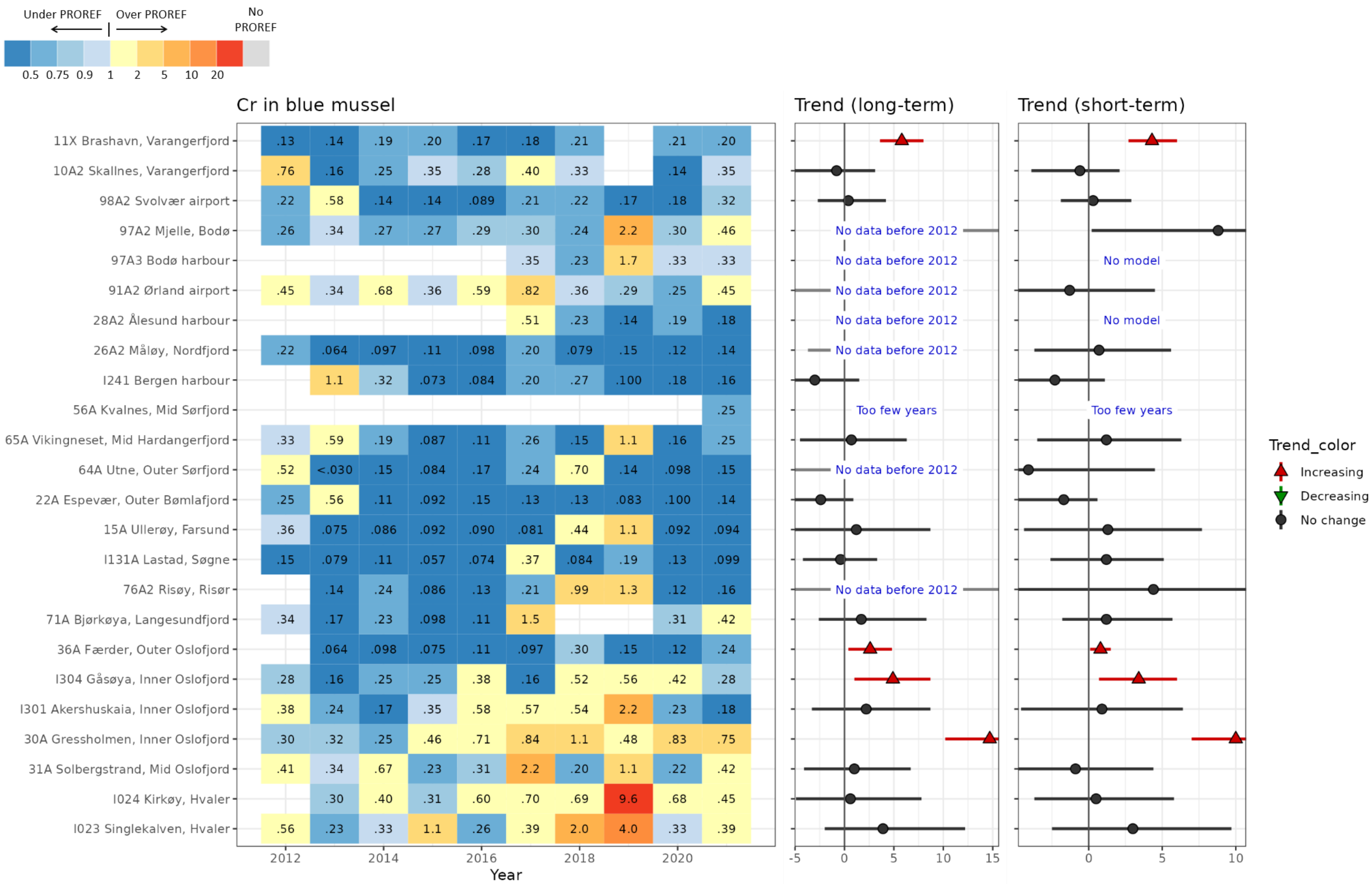
For mussel stations, concentrations of chromium had mostly no long-term trends (**Figure 42**). Four stations however, had increasing long-term trends (Gressholmen (30A) and Gåsøya in the Inner Oslofjord (I304), Færder in the Outer Oslofjord (36A) and Brashavn in the Varangerfjord (11X)).

Chromium concentrations in cod had significant decreasing long-term trends at all stations.

Short-term trends

The same four stations had significant increasing short-term trends (Gressholmen (30A), Gåsøya (I304), Færder (36A) and Brashavn (11X)).

For decreasing short-term trends in cod dominated, but no trends were observed at three stations.


Under PROREF
Over PROREF
No PROREF
0.5 0.75 0.9 1 2 5 10 20
Cr in blue mussel
Trend (long-term)
Trend (short-term)
11X Brashavn, Varangerfjord
10A2 Skallnes, Varangerfjord
98A2 Svolvær airport
97A2 Mjelle, Bodø
97A3 Bodø harbour
91A2 Ørland airport
28A2 Ålesund harbour
26A2 Måløy, Nordfjord
I241 Bergen harbour
56A Kvalnes, Mid Sørfjord
65A Vikingneset, Mid Hardangerfjord
64A Utne, Outer Sørfjord
22A Espevær, Outer Bømlafjord
15A Ullerøy, Farsund
I131A Lastad, Søgne
76A2 Risøy, Risør
71A Bjørkøya, Langesundfjord
36A Færder, Outer Oslofjord
I304 Gåsøya, Inner Oslofjord
I301 Akershuskaia, Inner Oslofjord
30A Gressholmen, Inner Oslofjord
31A Solbergstrand, Mid Oslofjord
I024 Kirkøy, Hvaler
I023 Singlekalven, Hvaler
No data before 2012
No model
Too few years
Year
2012 2014 2016 2018 2020
Trend_color
Increasing
Decreasing
No change

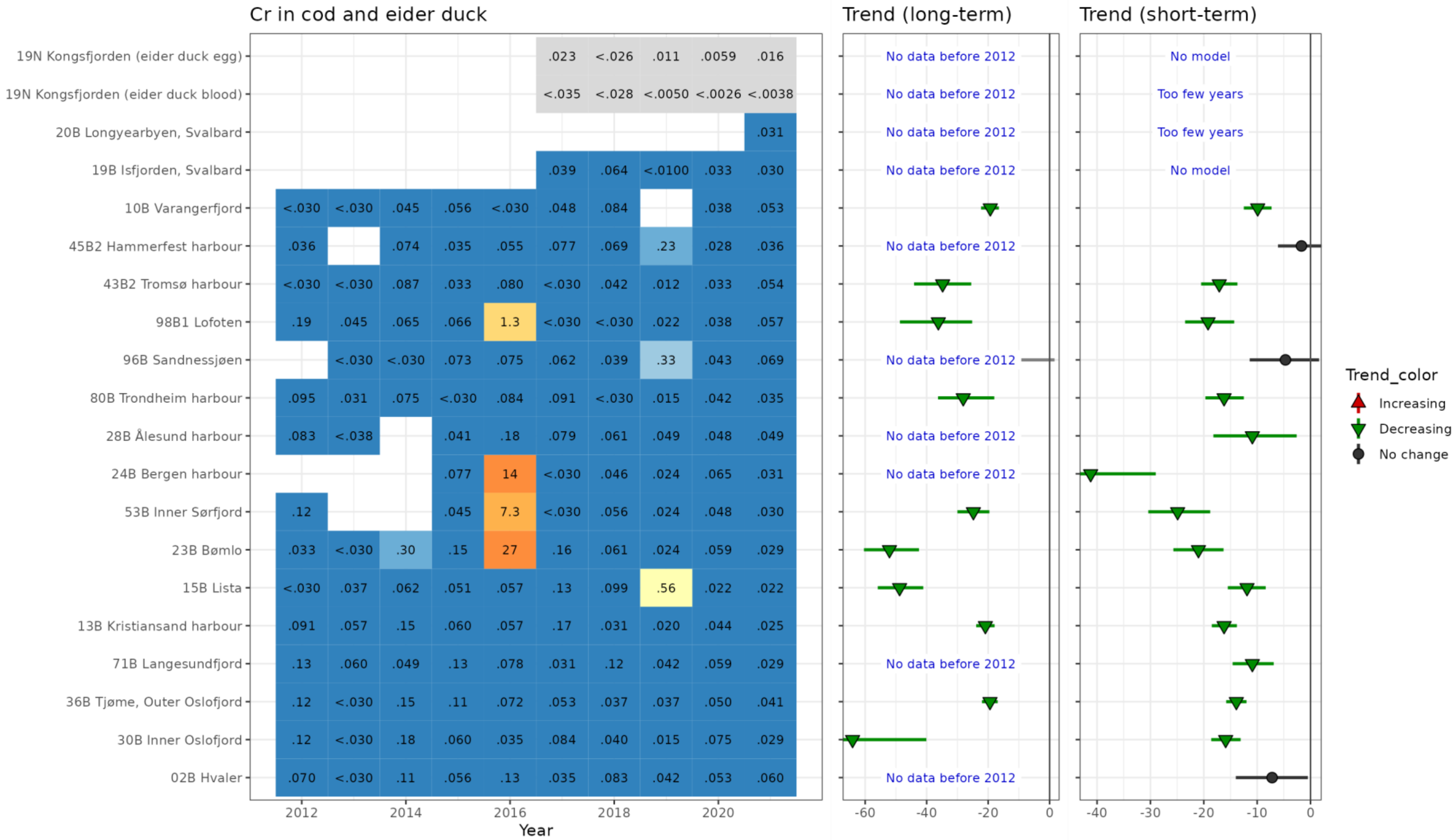
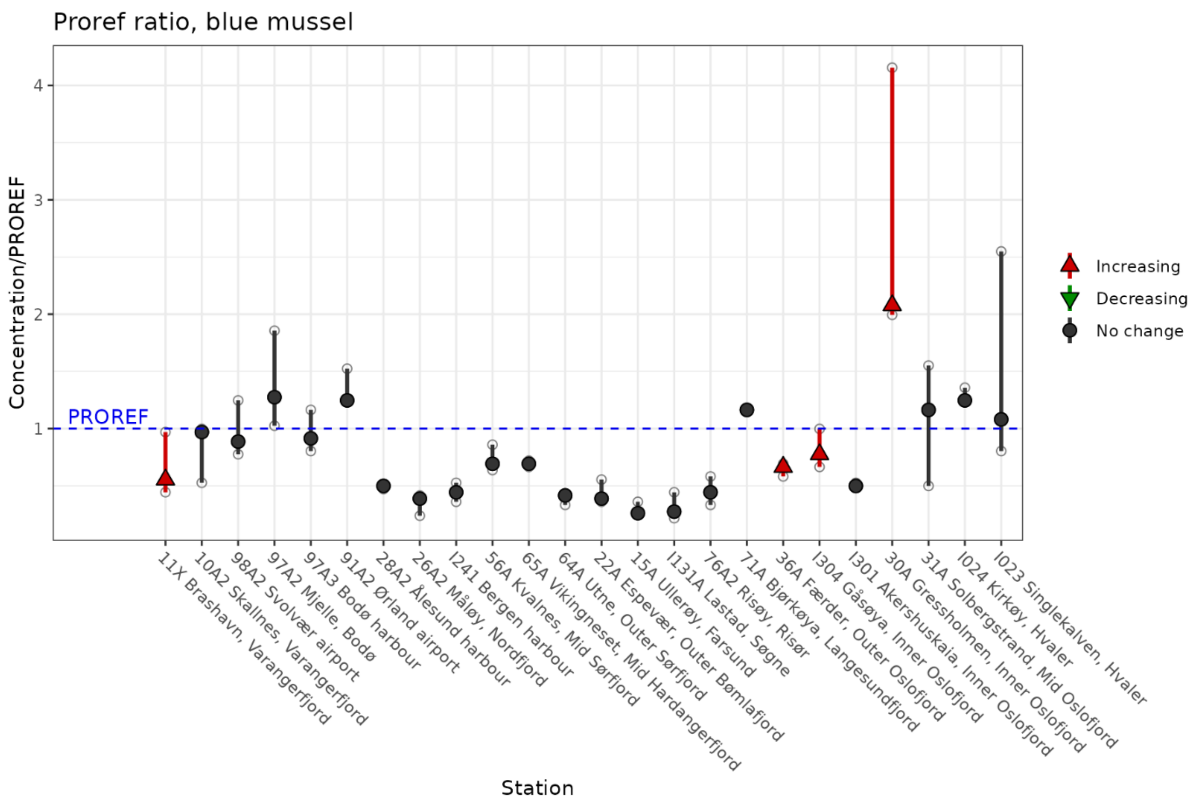


Figure 42. Heatmap and time-trends of chromium in (upper panel) blue mussel and (lower panel) cod and eider. For full explanation of figure see example **Figure 23**.



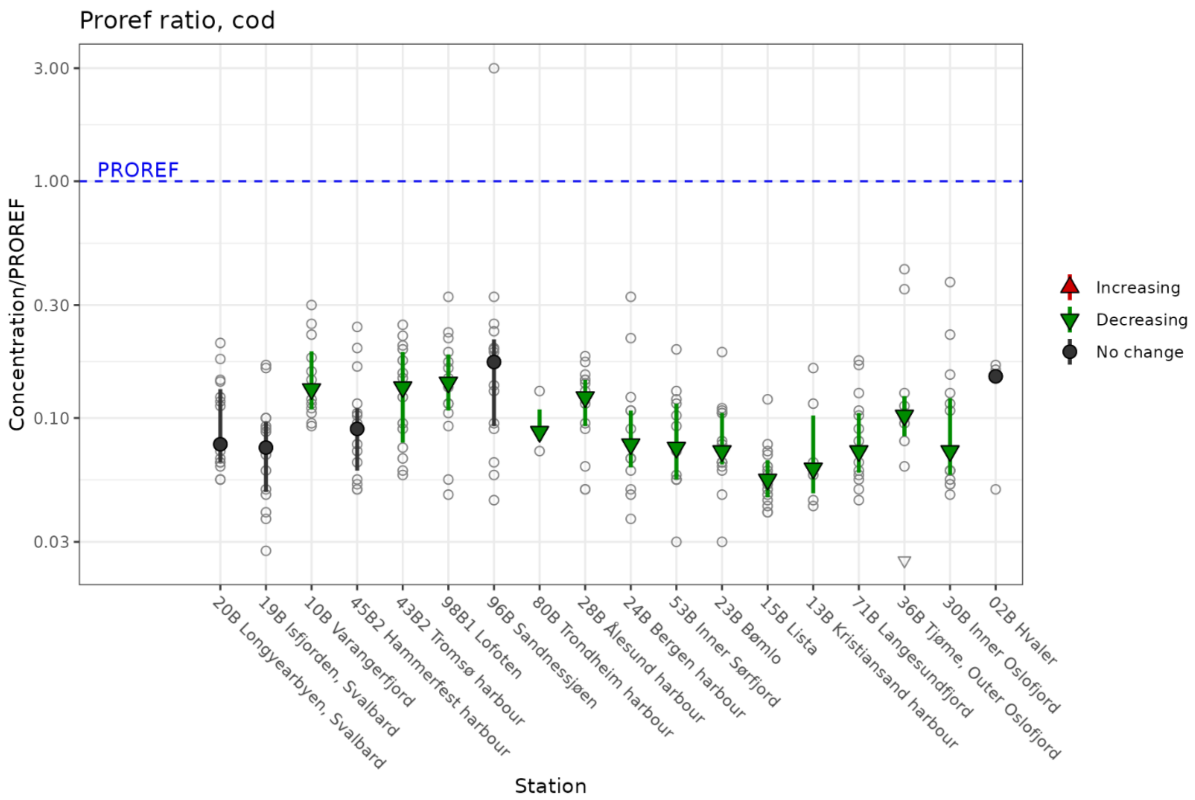


Figure 43. Ratio (concentration/PROREF) for chromium in mussel (upper panel) and cod (lower panel). *The y-axis for cod is on a natural log scale.* For full explanation of figure see example **Figure 24**.

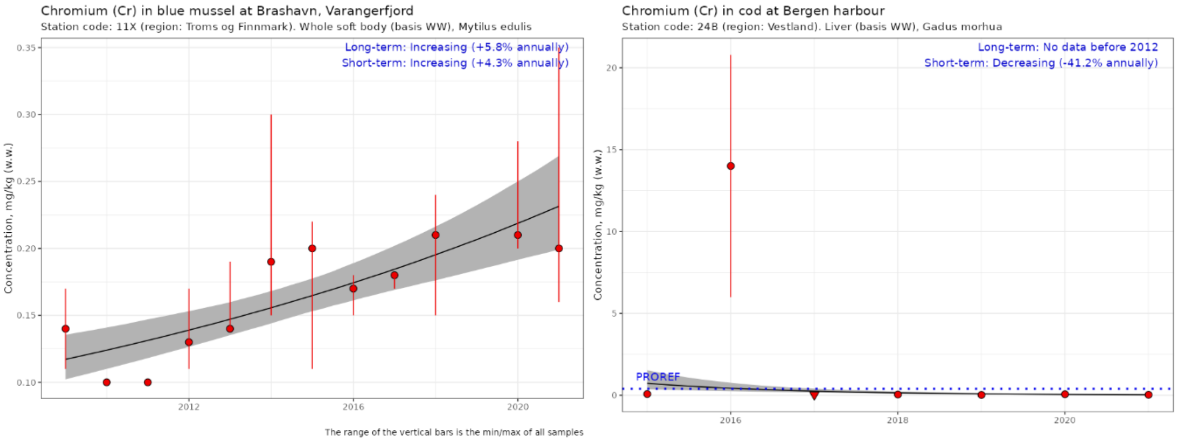


Figure 44. Two selected time trends for chromium. For full explanation of figure see example **Figure 25**.

Selected time trends

Two selected time trends are shown in **Figure 44**, one for concentrations in mussel (Brashavn in the Varangerfjord (11X)) and one for cod (Bergen harbour (24B). In mussels from 11X, concentrations were increasing both long-term and short-term, but concentrations were still below PROREF. In cod from station 24B, there is only an annual short-term decreasing trend (40%). Concentrations from 2016 were much higher than other concentrations but had little impact on the model (narrow confidence band after 2016). The concentrations are now below PROREF.

3.2.1.7 Cobalt (Co)

Cobalt (Co) is a trace metal involved in photosynthesis and nitrogen fixation detected most oceans basins and is a limiting micronutrient for phytoplankton and cyanobacteria. Sources of cobalt for many ocean bodies include rivers and terrestrial runoff with some input from hydrothermal vents. Cobalt is considered toxic for marine environments at high concentrations. In the present study, cobalt was analysed in blue mussel at 24 stations, in cod liver at 18 stations, and in eider blood and eggs at one station (**Table 1**, **Figure 45**).

EQS

There is no EQS for cobalt in biota (**Table 3**).

PROREF

Most mussel stations had concentration below PROREF (**Figure 45** and **Figure 46**). Six mussel stations had cobalt concentration above PROREF, and the stations with the highest exceedances of PROREF was at Kirkøy at Hvaler (I024) (2.5x). Only one cod station had cobalt concentration that exceeded PROREF (the Inner Oslofjord (30B)).

Long-term trends

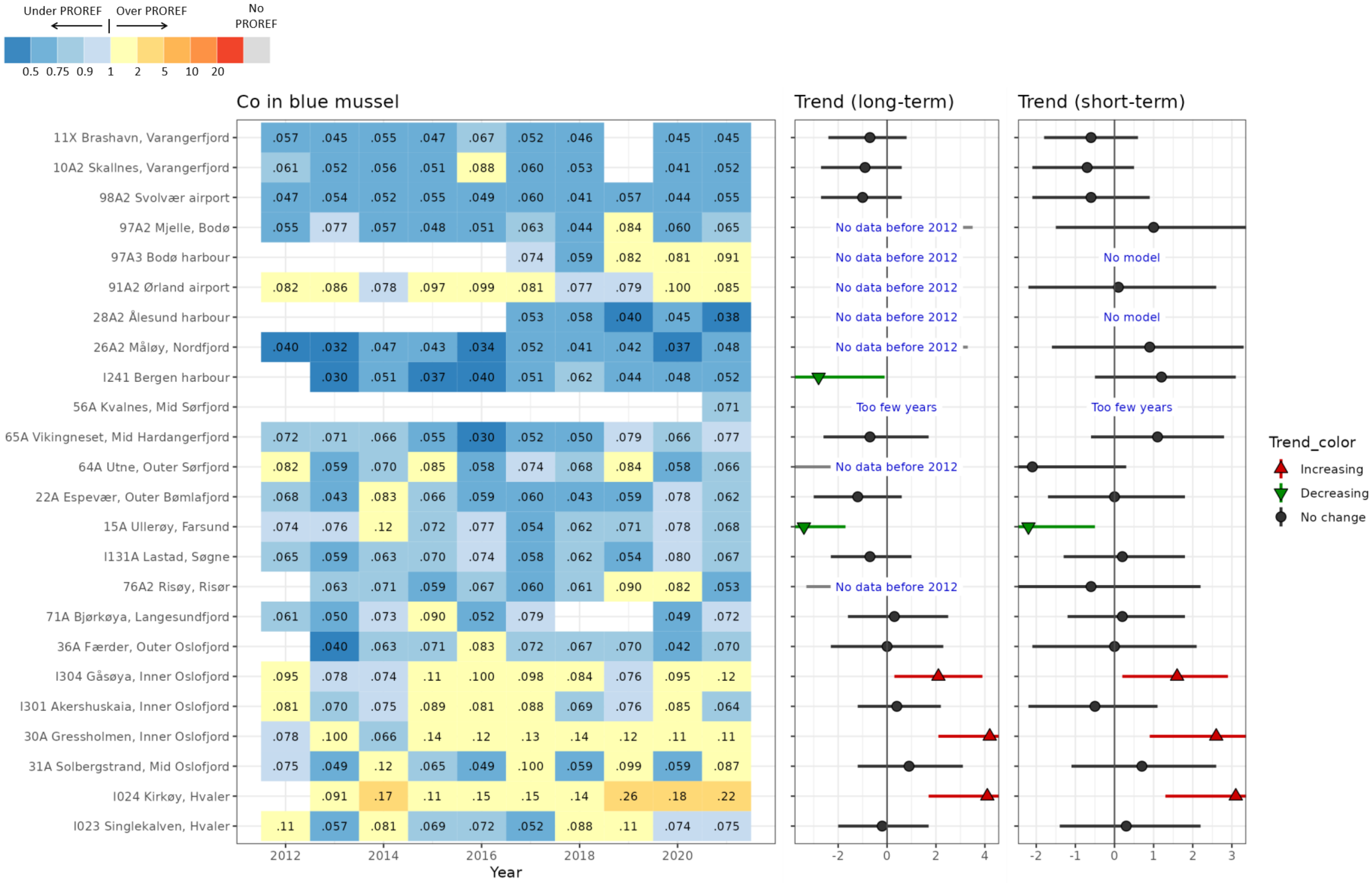
Most mussel stations had no long-term trends (**Figure 45**). Three stations had significant increasing long-term (Kirkøy at Hvaler (I024), Gressholmen in the Inner Oslofjord (30A) and Gåsøya in the Inner Oslofjord (I304)) and one station had significant decreasing long-term trend (Ullerøy at Farsund (15A)).

Most cod stations had no long-term trends. One station had significant increasing time trend (Lista (15B)) and two had significant decreasing time trends (Tjøme in the Outer Oslofjord (36B) and the Inner Sørfjord (53B)).

Short-term trends

The same mussel stations that had increasing long-term trends had increasing short-term trends, and those that had decreasing long-term trends had decreasing short-term trends.

For short-term trends in cod, station Lista (15B) also had significant increasing short-term trend, and in addition station Inner Oslofjord (30B) had significant increasing short-term time trend. Station Tjøme in the Outer Oslofjord (36B) had both significant decreasing long-term and short-term time trends. In addition, Hvaler (02B) and Lofoten (98B1) had significant decreasing short-term time trends.


Under PROREF
Over PROREF
No PROREF
0.5 0.75 0.9 1 2 5 10 20
Co in blue mussel
Trend (long-term)
Trend (short-term)
11X Brashavn, Varangerfjord .057 .045 .055 .047 .067 .052 .046 .045 .045
10A2 Skallnes, Varangerfjord .061 .052 .056 .051 .088 .060 .053 .041 .052
98A2 Svolvær airport .047 .054 .052 .055 .049 .060 .041 .057 .044 .055
97A2 Mjelle, Bodø .055 .077 .057 .048 .051 .063 .044 .084 .060 .065
97A3 Bodø harbour .074 .059 .082 .081 .091
91A2 Ørland airport .082 .086 .078 .097 .099 .081 .077 .079 .100 .085
28A2 Ålesund harbour .053 .058 .040 .045 .038
26A2 Måløy, Nordfjord .040 .032 .047 .043 .034 .052 .041 .042 .037 .048
I241 Bergen harbour .030 .051 .037 .040 .051 .062 .044 .048 .052
56A Kvalnes, Mid Sørfjord .071
65A Vikingneset, Mid Hardangerfjord .072 .071 .066 .055 .030 .052 .050 .079 .066 .077
64A Utne, Outer Sørfjord .082 .059 .070 .085 .058 .074 .068 .084 .058 .066
22A Espevær, Outer Bømlafjord .068 .043 .083 .066 .059 .060 .043 .059 .078 .062
15A Ullerøy, Farsund .074 .076 .12 .072 .077 .054 .062 .071 .078 .068
I131A Lastad, Søgne .065 .059 .063 .070 .074 .058 .062 .054 .080 .067
76A2 Risøy, Risør .063 .071 .059 .067 .060 .061 .090 .082 .053
71A Bjørkøya, Langesundfjord .061 .050 .073 .090 .052 .079 .049 .072
36A Færder, Outer Oslofjord .040 .063 .071 .083 .072 .067 .070 .042 .070
I304 Gåsøya, Inner Oslofjord .095 .078 .074 .11 .100 .098 .084 .076 .095 .12
I301 Akershuskaia, Inner Oslofjord .081 .070 .075 .089 .081 .088 .069 .076 .085 .064
30A Gressholmen, Inner Oslofjord .078 .100 .066 .14 .12 .13 .14 .12 .11 .11
31A Solbergstrand, Mid Oslofjord .075 .049 .12 .065 .049 .100 .059 .099 .059 .087
I024 Kirkøy, Hvaler .091 .17 .11 .15 .15 .14 .26 .18 .22
I023 Singlekalven, Hvaler .11 .057 .081 .069 .072 .052 .088 .11 .074 .075
No data before 2012
No model
Too few years
2012 2014 2016 2018 2020
Year
-2 0 2 4
-2 -1 0 1 2 3
Trend_color
Increasing
Decreasing
No change

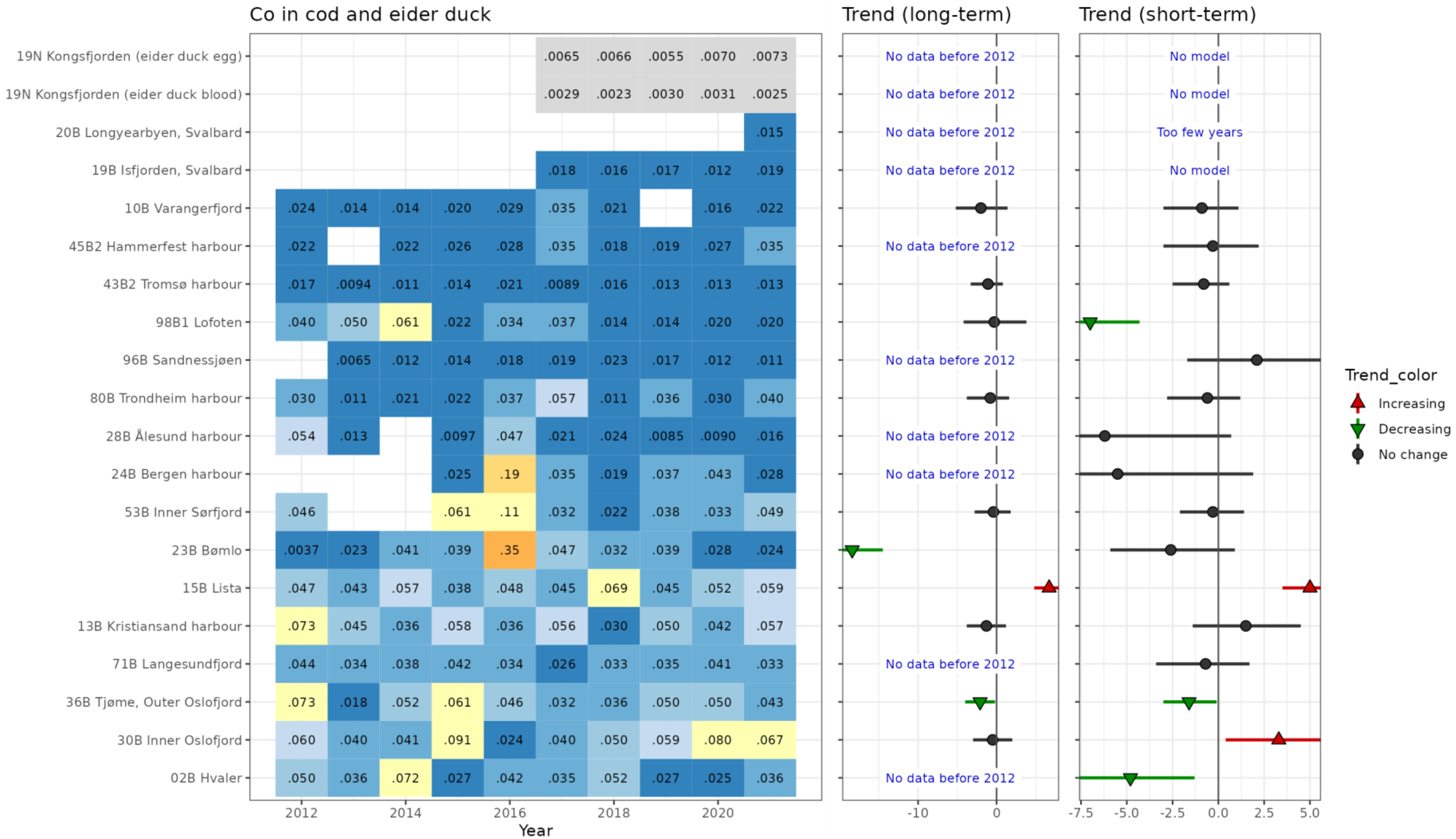


Figure 45. Heatmap and time trends of cobalt in (upper panel) blue mussel and (lower panel) cod and eider. For full explanation of figure see example **Figure 23**.

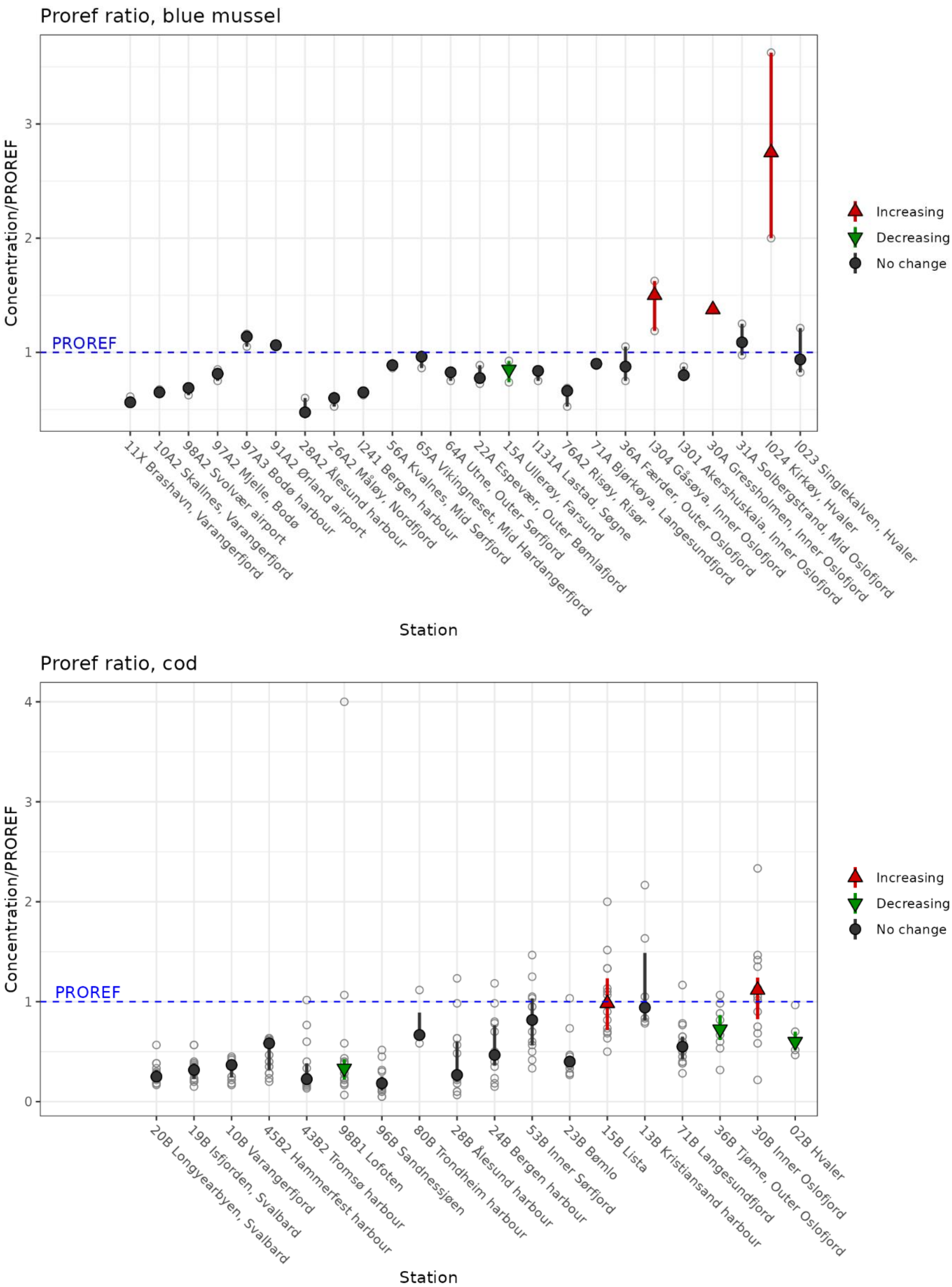


Figure 46. Ratio (concentration/PROREF) for cobalt in mussel (upper panel) and cod (lower panel). For full explanation of figure see example **Figure 24**.

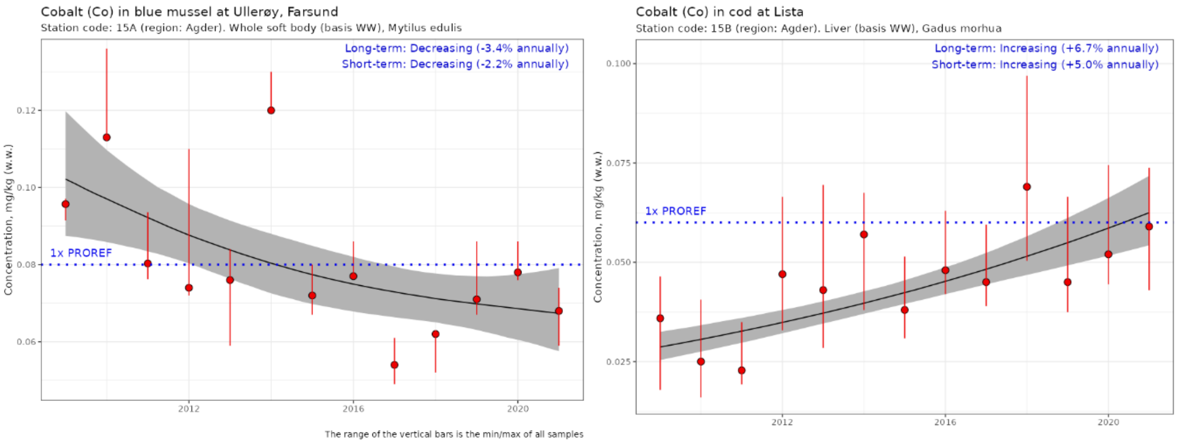


Figure 47. Two selected time trends for cobalt. For full explanation of figure see example **Figure 25**.

Selected time trends

Two selected time trends for cobalt concentrations are shown in **Figure 47**, mussel station at Ullerøy, Farsund (15A) and cod at Lista (15B). Cobalt in mussels at station 15A has decreased long-term (3.4% annually) and short-term (2.2% annually). The concentrations are now below PROREF, while they were modelled to be above PROREF until 2014. The situation is opposite for cobalt at Lista, where long-term and short-term trends in cod are increasing (6.7% and 5% annually). The median concentration in 2021 was just below PROREF.

3.2.1.8 Nickel (Ni)

Nickel (Ni) is a transition element extensively distributed in the environment, air, water, and soil. It may derive from natural sources and anthropogenic activity. Although nickel is ubiquitous in the environment, its functional role as a trace element for animals and human beings has not been yet recognized. Environmental pollution from nickel may be due to industry, the use of liquid and solid fuels, as well as municipal and industrial waste. Nickel contact can cause a variety of side effects on human health, such as allergy, cardiovascular and kidney diseases, lung fibrosis, lung and nasal cancer (text from (Genchi et al. 2020)). In the present study, Ni was analysed in blue mussel at 24 stations, in cod liver at 18 stations, and in eider blood and eggs at one station (**Table 1**, **Figure 48**).

EQS

There is no EQS for nickel in biota (**Table 3**).

PROREF

PROREF at mussel stations were mostly not exceeded, but exceedances were seen at nine stations (**Figure 48**, **Figure 49**). The highest exceedances were seen at Gressholmen in the Inner Oslofjord (30A) (2x PROREF).

For cod, PROREF was not exceeded at any stations, and concentrations were far lower than PROREF.

Long-term trends

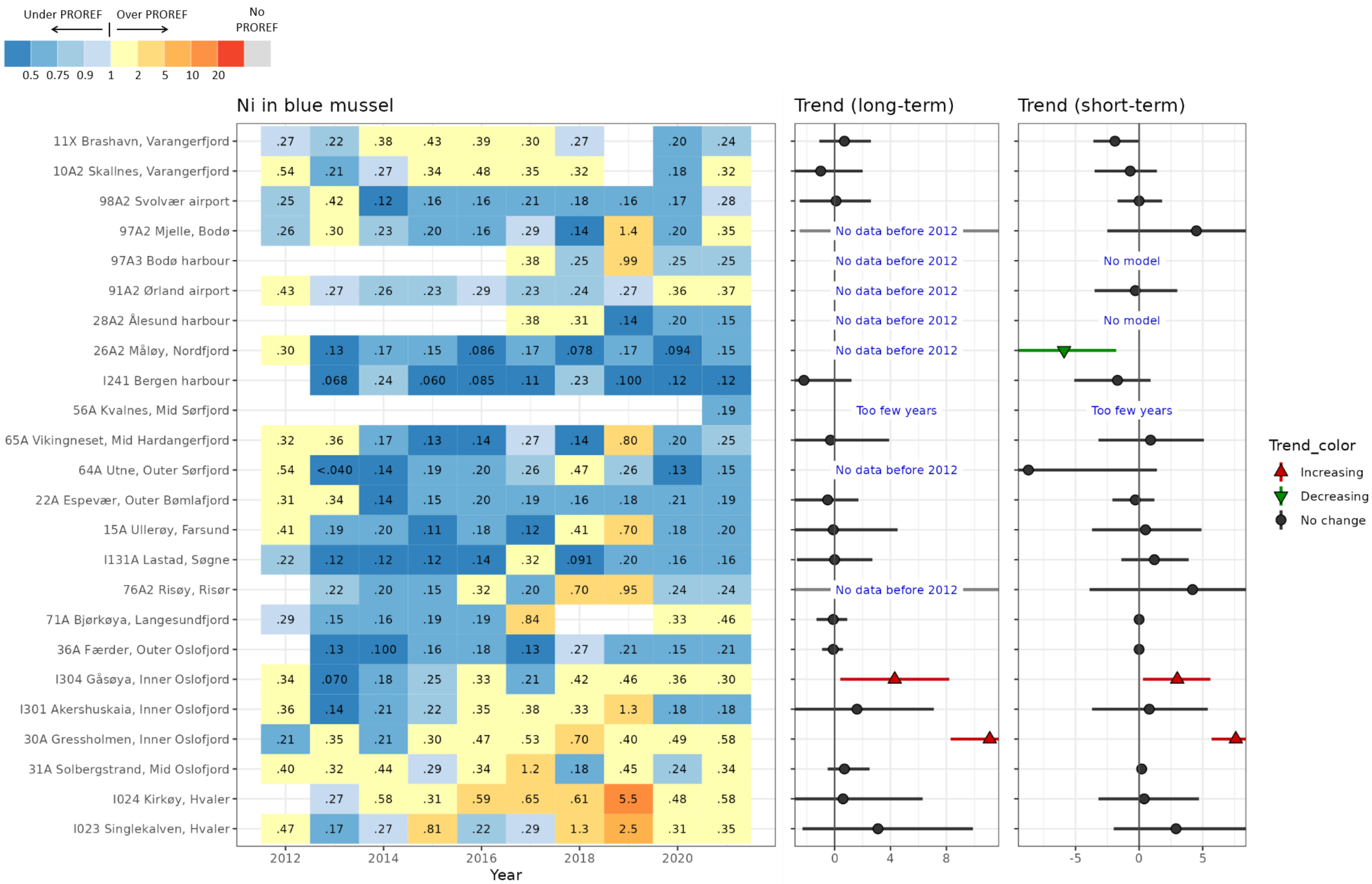
For long-term trends in mussel, no trend was dominating, but increasing trends were observed at two stations (Gressholmen (30A) and Gåsøya (I304) in the Inner Oslofjord (**Figure 48**).

Significant long-term decreasing time trends were observed in cod at four stations (Tjøme in the Outer Oslofjord (36B), the Inner Oslofjord (53)B, Kristiansand harbour (13B), and Trondheim harbour (80B)).

Short-term trends

Two significant increasing short-term time trends in mussel were found at the same stations that also had increasing long-term trends (Gressholmen (30A) and Gåsøya (I304)). One decreasing trend was observed (Måløy in the Nordfjord (26A2)).

For short-term trends in cod, decreasing trends dominated and were found at 13 stations.


Under PROREF
Over PROREF
No PROREF
0.5 0.75 0.9 1 2 5 10 20
Ni in blue mussel
Trend (long-term)
Trend (short-term)
11X Brashavn, Varangerfjord .27 .22 .38 .43 .39 .30 .27 .20 .24
10A2 Skallnes, Varangerfjord .54 .21 .27 .34 .48 .35 .32 .18 .32
98A2 Svolvær airport .25 .42 .12 .16 .16 .21 .18 .16 .17 .28
97A2 Mjelle, Bodø .26 .30 .23 .20 .16 .29 .14 1.4 .20 .35
97A3 Bodø harbour .38 .25 .99 .25 .25
91A2 Ørland airport .43 .27 .26 .23 .29 .23 .24 .27 .36 .37
28A2 Ålesund harbour .38 .31 .14 .20 .15
26A2 Måløy, Nordfjord .30 .13 .17 .15 .086 .17 .078 .17 .094 .15
I241 Bergen harbour .068 .24 .060 .085 .11 .23 .100 .12 .12
56A Kvalnes, Mid Sørfjord .19
65A Vikingneset, Mid Hardangerfjord .32 .36 .17 .13 .14 .27 .14 .80 .20 .25
64A Utne, Outer Sørfjord .54 <.040 .14 .19 .20 .26 .47 .26 .13 .15
22A Espevær, Outer Bømlafjord .31 .34 .14 .15 .20 .19 .16 .18 .21 .19
15A Ullerøy, Farsund .41 .19 .20 .11 .18 .12 .41 .70 .18 .20
I131A Lastad, Søgne .22 .12 .12 .12 .14 .32 .091 .20 .16 .16
76A2 Risøy, Risør .22 .20 .15 .32 .20 .70 .95 .24 .24
71A Bjørkøya, Langesundfjord .29 .15 .16 .19 .19 .84 .33 .46
36A Færder, Outer Oslofjord .13 .100 .16 .18 .13 .27 .21 .15 .21
I304 Gåsøya, Inner Oslofjord .34 .070 .18 .25 .33 .21 .42 .46 .36 .30
I301 Akershuskaia, Inner Oslofjord .36 .14 .21 .22 .35 .38 .33 1.3 .18 .18
30A Gressholmen, Inner Oslofjord .21 .35 .21 .30 .47 .53 .70 .40 .49 .58
31A Solbergstrand, Mid Oslofjord .40 .32 .44 .29 .34 1.2 .18 .45 .24 .34
I024 Kirkøy, Hvaler .27 .58 .31 .59 .65 .61 5.5 .48 .58
I023 Singlekalven, Hvaler .47 .17 .27 .81 .22 .29 1.3 2.5 .31 .35
No data before 2012
Too few years
No model
2012 2014 2016 2018 2020
Year
0 4 8
-5 0 5
Trend_color
Increasing
Decreasing
No change

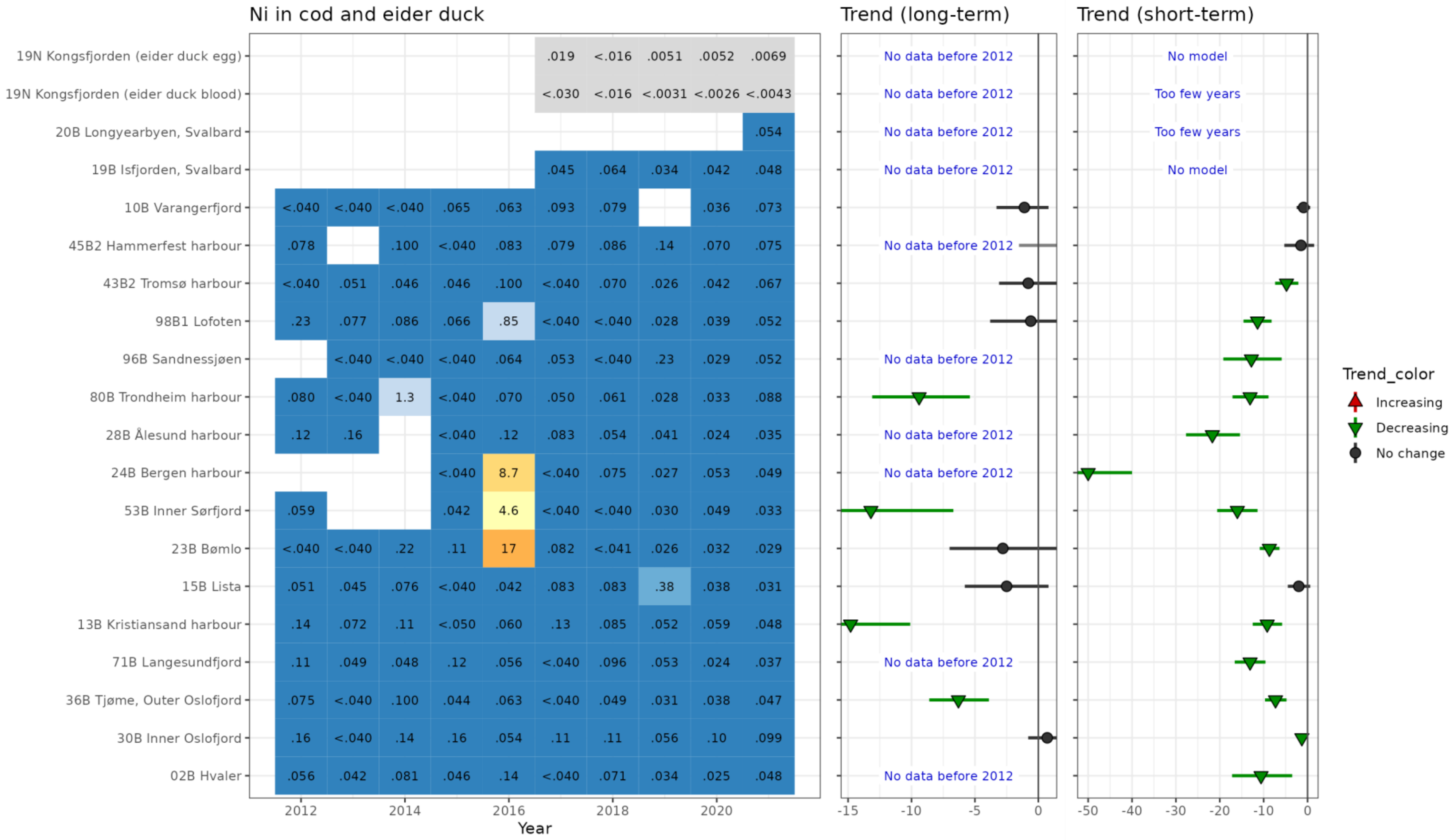
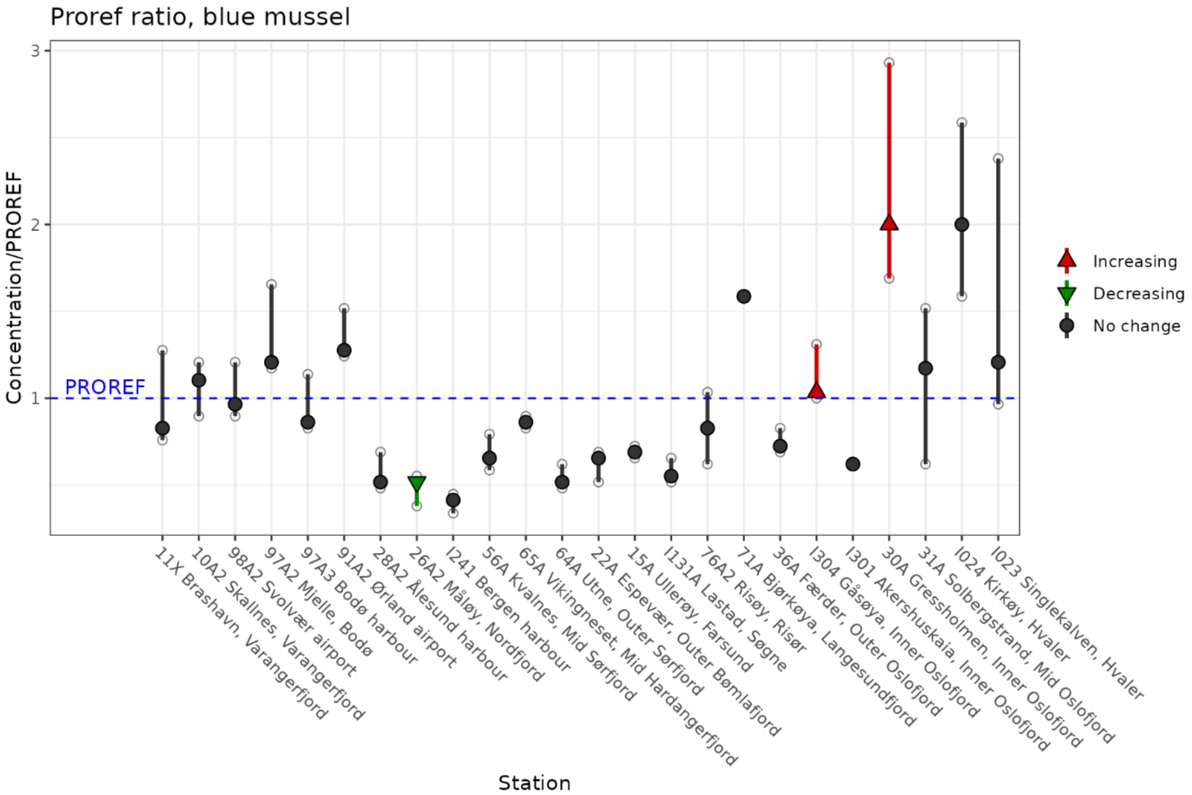


Figure 48. Heatmap and time trends of nickel in (upper panel) blue mussel and (lower panel) cod and eider. For full explanation of figure see example **Figure 23**.



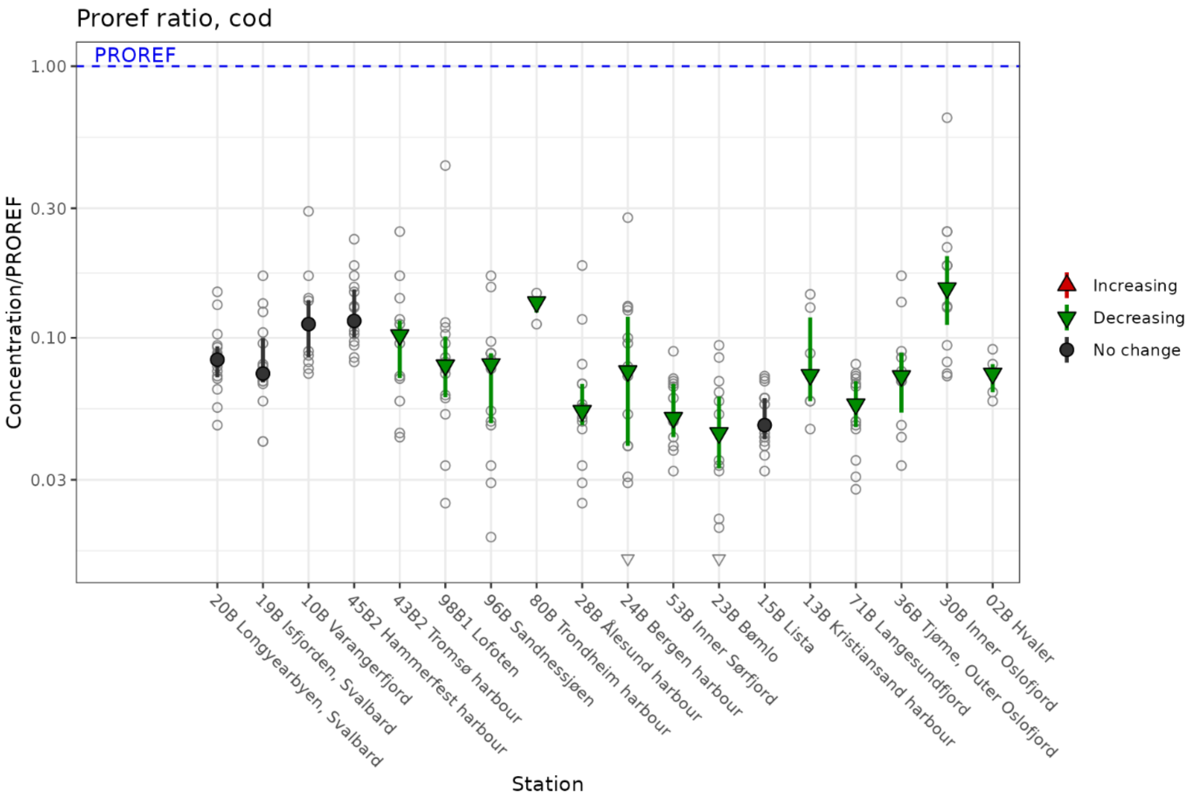


Figure 49. Ratio (concentration/PROREF) for nickel in blue mussel (upper panel) and cod (lower panel). *The y-axis for cod is on a natural log scale.* For full explanation of figure see example **Figure 24**.

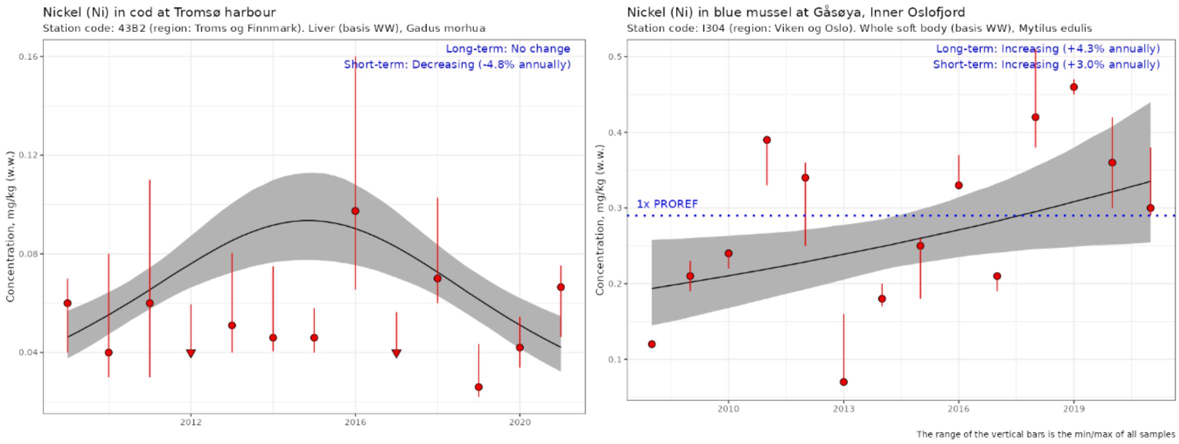


Figure 50. Two selected time trends for nickel. For full explanation of figure see example **Figure 25**.

Selected time trends

Two selected time trends are shown in **Figure 50**; nickel concentration in cod at Tromsø harbour (43B2) and mussel concentration at Gåsøya in the Inner Oslofjord (I304). The cod from Tromsø had very low concentrations which increased around 2016. The long-term trend is therefore no trend, while the short-term trend arises because of the temporary increase.

Nickel concentrations at Gåsøya had and increasing trend, both long-term and short-term. The increase was annually 4.3% and 3%, respectively. The PROREF was now exceeding PROREF at Gåsøya since 2018.

3.2.1.9 Zinc (Zn)

Zinc (Zn) is one of the most common elements in the earth's crust. It is found in air, soil, and water, and is present in all foods. Pure zinc is a bluish-white shiny metal. Zinc has many commercial uses as coatings to prevent rust, in dry cell batteries, and mixed with other metals to make alloys like brass, and bronze. Zinc combines with other elements to form zinc compounds. Common zinc compounds found at hazardous waste sites include zinc chloride, zinc oxide, zinc sulfate, and zinc sulfide. Zinc compounds are widely used in industry to make paint, rubber, dyes, wood preservatives, and ointments (text from ATSR[6]). In the present study, Zn was analysed in blue mussel at 24 stations, in cod liver at 18 stations, and in eider blood and eggs at one station (**Table 1**, **Figure 51**).

EQS

There is no EQS for zinc in biota (**Table 3**).

PROREF

PROREF was exceeded in mussels at 10 stations, while in cod no exceedance of PROREF was observed (**Figure 51**, **Figure 52**).

Long-term trends

Long-term decreasing trends in mussels were most frequently observed and was found at eight stations (**Figure 51**). One mussel station had significant increasing long-term trend (Færder in the Outer Oslofjord (36A)).

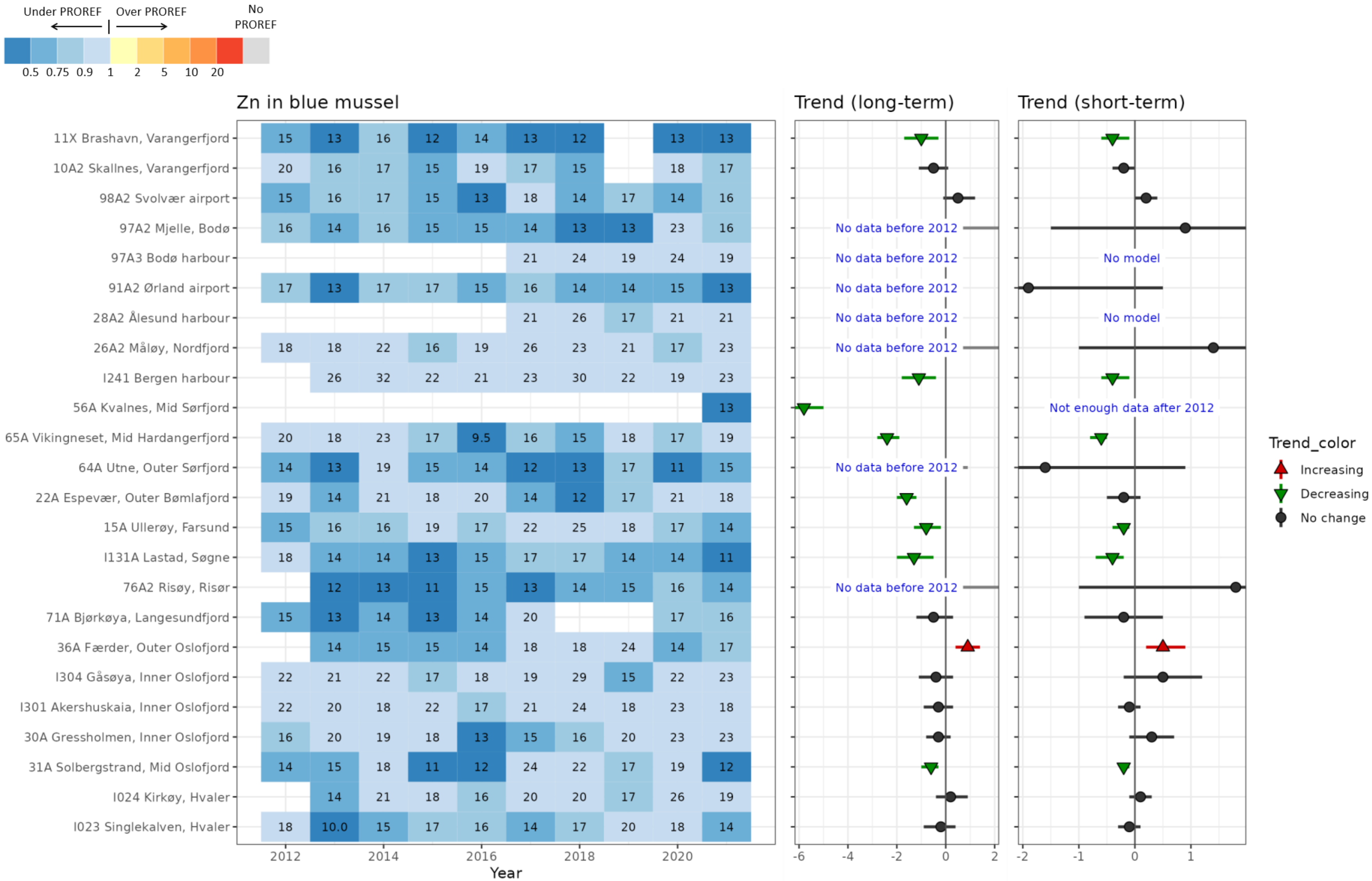
Long-term trends in cod were mostly no trends, but two stations had significant increasing time trends (Lista (15B) and Lofoten (98B1)), while three had decreasing time trends (Tjøme in the Outer Oslofjord (36B), Tromsø harbour (43B2) and Varangerfjord (10B)).

Short-term trends

There were mostly no short-term trends in cod, but significant increasing time trend was found at Lista (15B). Short-term time trends were observed at Bergen harbour (24B), Tromsø harbour (43B2) and Hammerfest harbour (45B2).

For short-term trends, the picture was roughly the same, with seven of the same stations showing decreasing time trends. Station Færder in the Outer Oslofjord (36A) also had significant increasing short-term trend.

[6] https://wwwn.cdc.gov/TSP/substances/ToxSubstance.aspx?toxid=54



Under PROREF
Over PROREF
No PROREF
0.5 0.75 0.9 1 2 5 10 20
Zn in blue mussel
Trend (long-term)
Trend (short-term)
11X Brashavn, Varangerfjord
10A2 Skallnes, Varangerfjord
98A2 Svolvær airport
97A2 Mjelle, Bodø
97A3 Bodø harbour
91A2 Ørland airport
28A2 Ålesund harbour
26A2 Måløy, Nordfjord
I241 Bergen harbour
56A Kvalnes, Mid Sørfjord
65A Vikingneset, Mid Hardangerfjord
64A Utne, Outer Sørfjord
22A Espevær, Outer Bømlafjord
15A Ullerøy, Farsund
I131A Lastad, Søgne
76A2 Risøy, Risør
71A Bjørkøya, Langesundfjord
36A Færder, Outer Oslofjord
I304 Gåsøya, Inner Oslofjord
I301 Akershuskaia, Inner Oslofjord
30A Gressholmen, Inner Oslofjord
31A Solbergstrand, Mid Oslofjord
I024 Kirkøy, Hvaler
I023 Singlekalven, Hvaler
No data before 2012
No model
Not enough data after 2012
Year
Trend_color
Increasing
Decreasing
No change

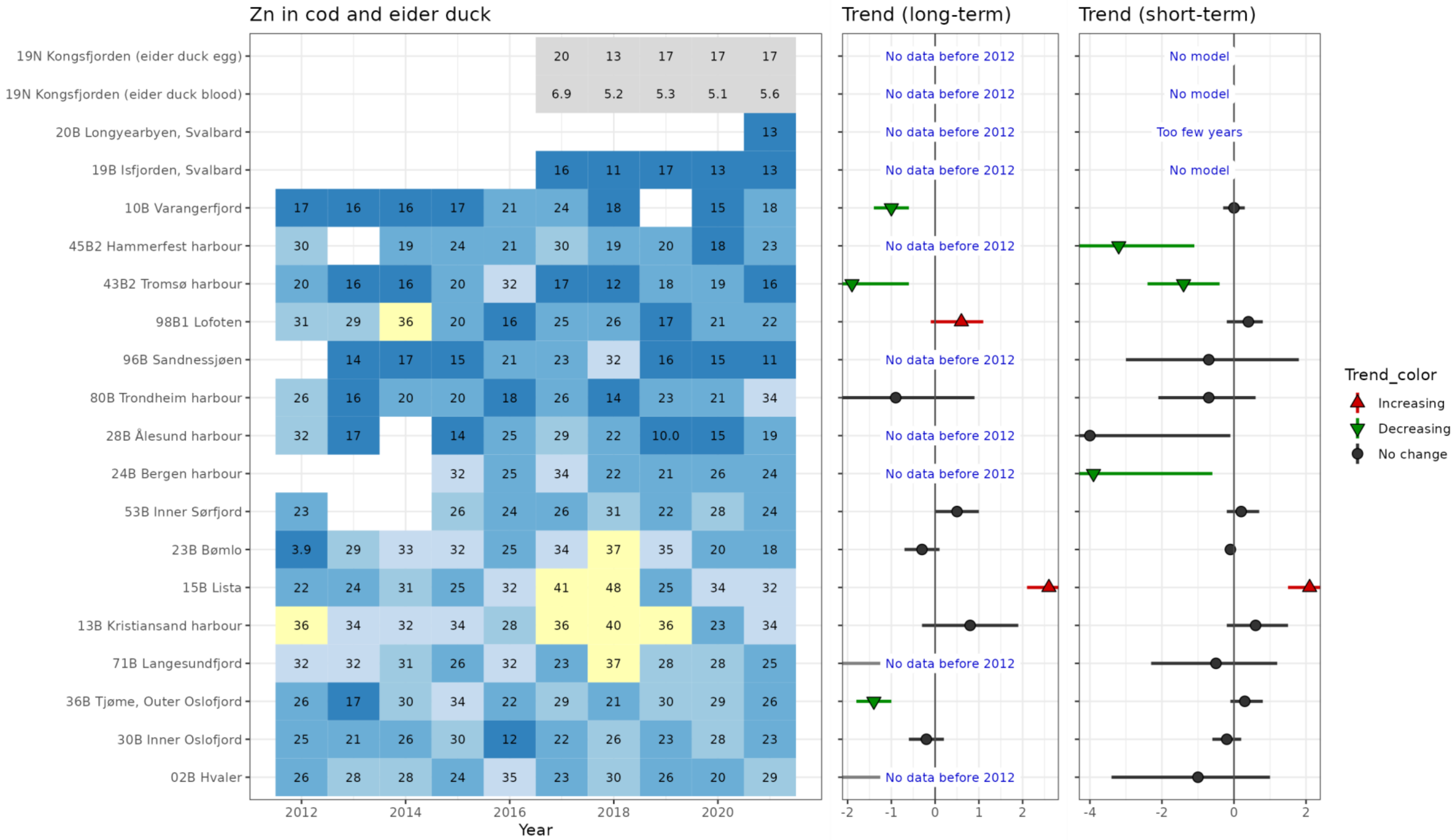


Figure 51. Heatmap and time-trends of zinc in (upper panel) blue mussel and (lower panel) cod and eider. For full explanation of figure see example **Figure 23**.

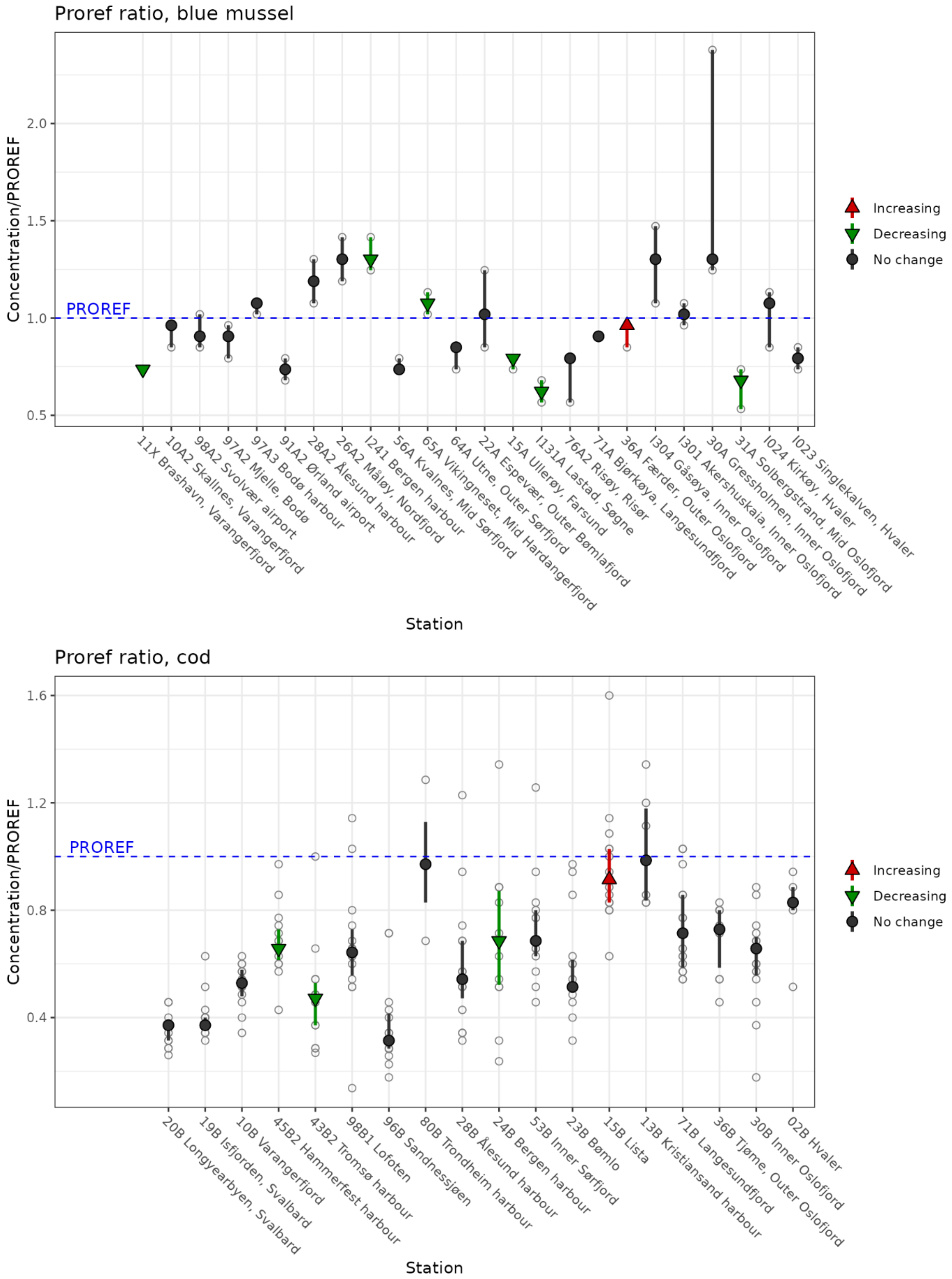


Figure 52. Ratio (concentration/PROREF) for zinc in blue mussel (upper panel) and cod (lower panel). For full explanation of figure see example **Figure 24**.

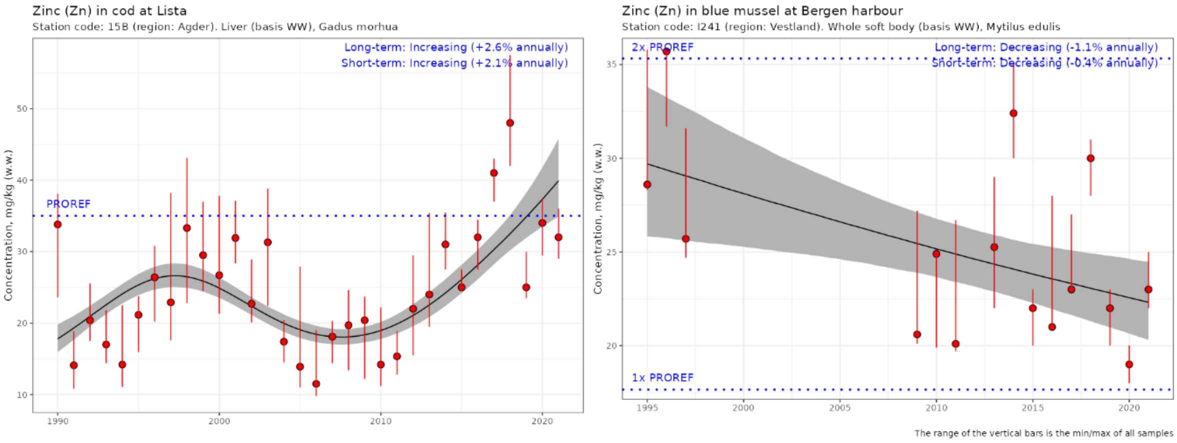


Figure 53. Two selected time trends for zinc. For full explanation of figure see example **Figure 25**.

Selected time trends

Two selected time trends for zinc concentrations are presented in **Figure 53**, cod at station Lista (15B) and blue mussel at Bergen harbour (I241). At Lista there were both a long-term and short-term increase of zinc. The median concentration was only a little below PROREF in 2021.

For blue mussel at Bergen harbour (I241), the concentrations have been decreasing, both long-term and short-term.

3.2.1.10 Copper (Cu)

Copper (Cu) is an element. In the past, wood was often impregnated with Cu. Today such use is prohibited, and the use has been significantly reduced. Under the Biocidal Products Regulation however dicopper oxide and copper thiocyanate are still permitted as active substances in antifouling agents in Norway. When copper from metallic copper, copper thiocyanate or cuprous oxide leaches into marine water in presence of oxygen, the predominant form of the copper is the active substance, the cupric ion, Cu^{2+} [7]. In the present study, Cu was analysed in blue mussel at 24 stations, in cod liver at 18 stations, and in eider blood and eggs at one station (**Table 1**, **Figure 54**).

EQS

There is no EQS for copper in biota (**Table 3**).

PROREF

Copper concentrations in mussel were exceeding PROREF in one mussel station (91A2), while no cod stations exceeded PROREF (**Figure 54**, **Figure 55**).

Long-term trends

Long-term trends in mussel were dominated by no trends, but seven decreasing trends were observed and one increasing trend (Bergen harbour (I241)) (**Figure 54**).

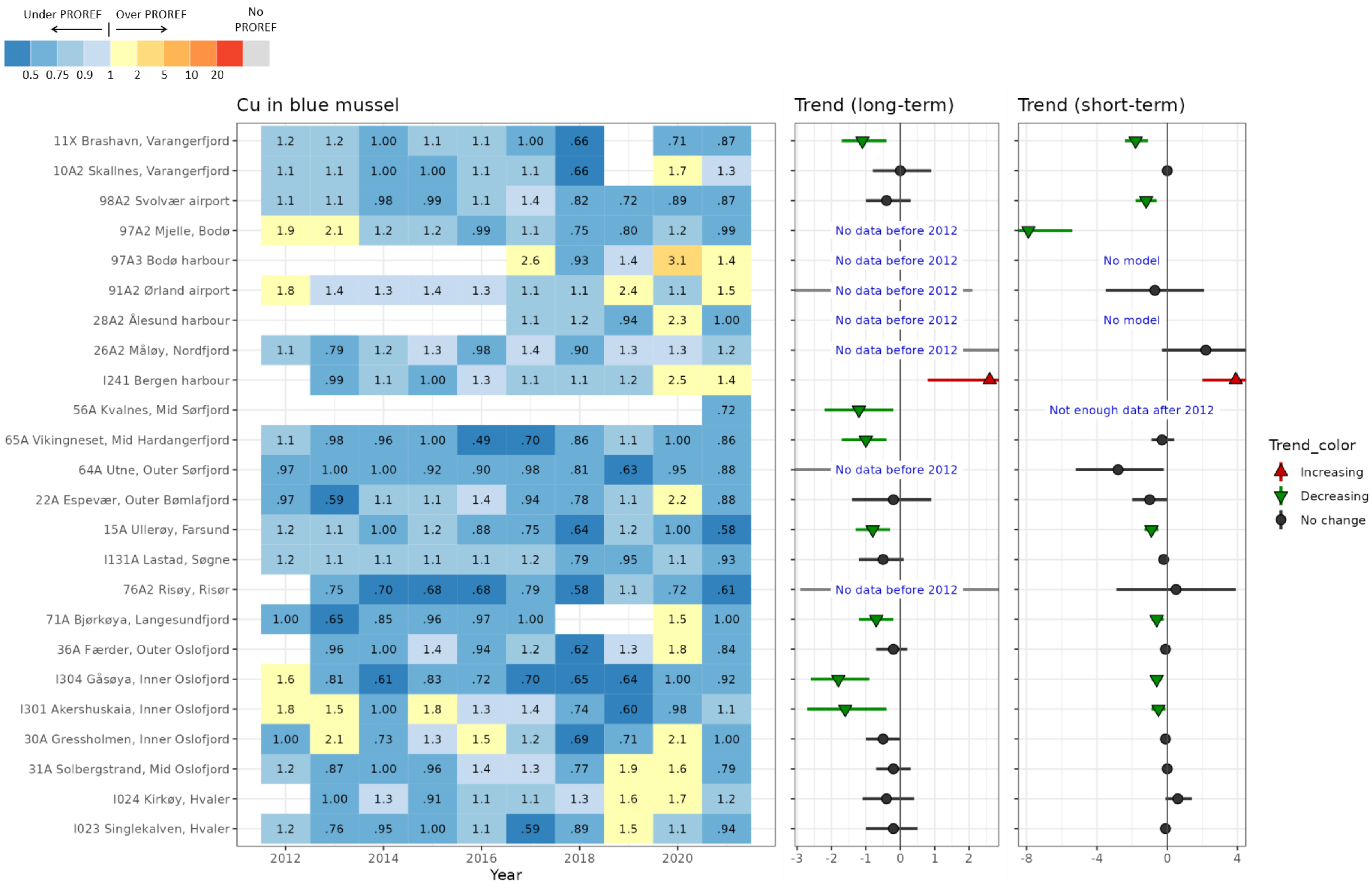
Copper had most decreasing long-term trends in cod (4 stations), but also two stations had increasing trends (Kristiansand harbour (13B), Lista (15B) and the Inner Sørfjord (53B)).

Short-term trends

Blue mussel at Bergen harbour (I241) had also increasing short-term trend, and most of the eight decreasing short-term trends were for the same stations that had decreasing long-term trends.

For short-term trends in cod, no trends dominated. Four decreasing short-term time trends were observed, mostly for the same stations that had long-term trends, but station Varangerfjord (10B) had decreasing long-term trend and increasing short-term trend. Stations Kristiansand harbour (13B) and Lista (15B) had significant increasing short-term time trends.

[7] https://echa.europa.eu/documents/10162/7417a7be-8032-c2d1-08d2-f780b50b3751


Under PROREF
Over PROREF
No PROREF
0.5 0.75 0.9 1 2 5 10 20
Cu in blue mussel
Trend (long-term)
Trend (short-term)
11X Brashavn, Varangerfjord
10A2 Skallnes, Varangerfjord
98A2 Svolvær airport
97A2 Mjelle, Bodø
97A3 Bodø harbour
91A2 Ørland airport
28A2 Ålesund harbour
26A2 Måløy, Nordfjord
I241 Bergen harbour
56A Kvalnes, Mid Sørfjord
65A Vikingneset, Mid Hardangerfjord
64A Utne, Outer Sørfjord
22A Espevær, Outer Bømlafjord
15A Ulleroy, Farsund
I131A Lastad, Søgne
76A2 Risøy, Risør
71A Bjørkøya, Langesundfjord
36A Færder, Outer Oslofjord
I304 Gåsøya, Inner Oslofjord
I301 Akershuskaia, Inner Oslofjord
30A Gressholmen, Inner Oslofjord
31A Solbergstrand, Mid Oslofjord
I024 Kirkøy, Hvaler
I023 Singlekalven, Hvaler
No data before 2012
No model
Not enough data after 2012
Year
Trend_color
Increasing
Decreasing
No change

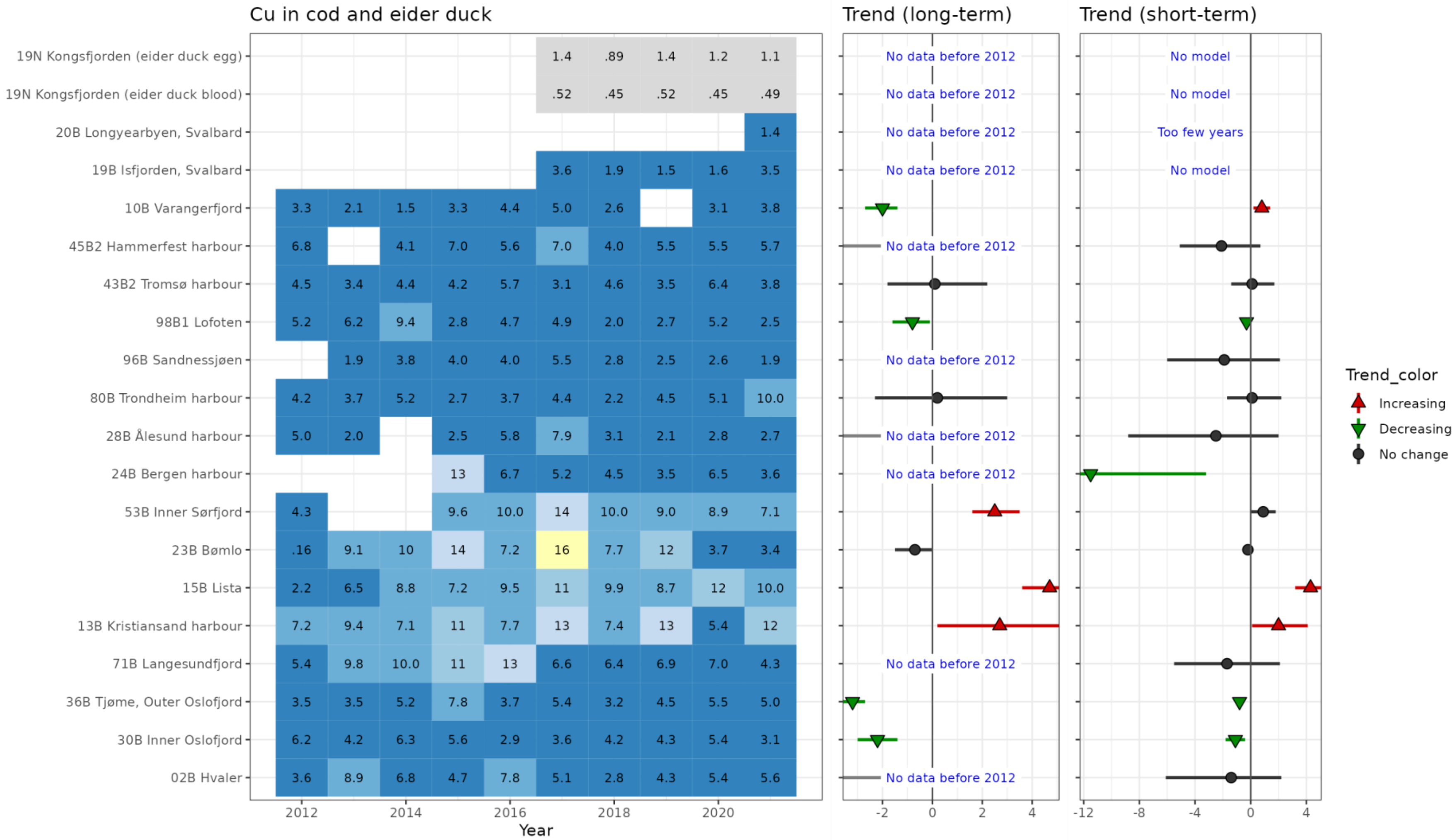


Figure 54. Heatmap and time-trends of copper in (upper panel) blue mussel and (lower panel) cod and eider. For full explanation of figure see example **Figure 23**.

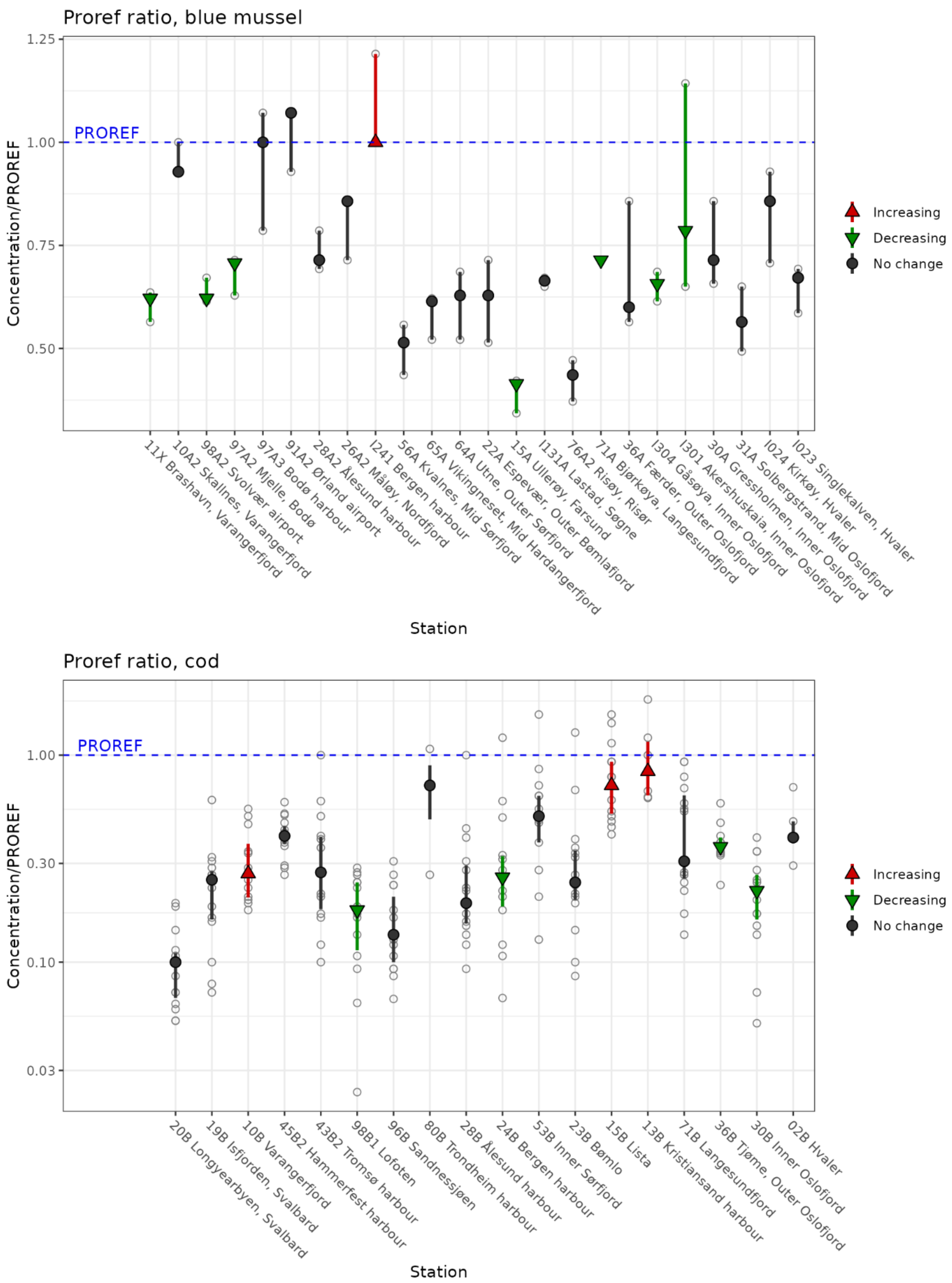


Figure 55. Ratio (concentration/PROREF) for copper in blue mussel (upper panel) and cod (lower panel). For full explanation of figure see example **Figure 24**.

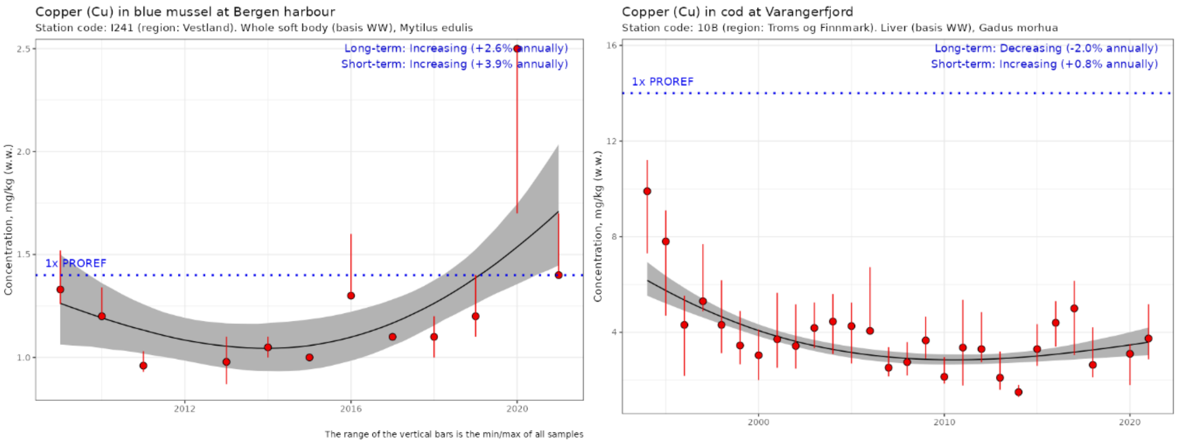


Figure 56. Two selected time trends for cupper. For full explanation of figure see example **Figure 25**.

Selected time trends

Two selected time trends for copper concentrations are shown in **Figure 56** in blue mussel at Bergen harbour (I241) and in cod in the Varangerfjord (10B). Copper in mussel at Bergen harbour (I241) are increasing both long-term and short-term. The model is possibly influenced a lot from high copper concentrations measured in 2020, and data next year will be interesting to see.

Copper concentrations in cod in the Varangerfjord (10B) are well below PROREF and have decreased both long-term and short-term.

3.2.2 Organic contaminants

3.2.2.1 PFAS

PFAS are fluorinated substances that contain at least one fully fluorinated methyl or methylene carbon atom (without any H/Cl/Br/I atom attached to it), i.e., with a few noted exceptions. Any chemical with at least a perfluorinated methyl group (−CF3) or a perfluorinated methylene group (−CF2−) is a PFAS compound (Wang et al. 2021). With this new definition, OECD has expanded the PFAS numbers of substances substantially and updated number of PFAS can be found at PubChem[8]. PFAS are often used as oil-, stain- and water-repellent surfactants and in many other applications. Firefighting foam is the largest source to PFOS in the Norwegian environment[9]. In the present study, PFAS were analysed in blue mussel at six stations, cod liver at 11 stations, and in eider blood and eggs at one station (**Table 1**, **Figure 57**). PFAS have been analysed annually in cod liver since 2005, as well as in 1993 for the Inner Oslofjord (30B) and Bømlo (23B).

In mussel samples, very few detections of any PFAS were done. These results are therefore not presented here, and only figures of concentrations in cod are shown.

EQS

There were no exceedances of EQSs of PFOS (9.1 µg/kg ww, **Table 3**) and PFOA (91 µg/kg ww) for median concentrations either in blue mussel or cod or eider (**Figure 57**, **Figure 58**). One cod sampled in the Inner Oslofjord (30B) had concentration exceeding EQS and PROREF.

PROREF

Cod stations have established PROREFs for three PFAS (PFOS, PFOA, and PFOSA). PFAS in cod were below PROREF for all three PFAS. PROREF was exceeded for PFOSA for some individual cod, but the median was below PROREF (**Figure 59**).

Long-term trends

For blue mussel there were no data for long term time trends (**Figure 57**).

In cod, PFOSA and PFOS concentrations had significant decreasing long-term trends for all stations for which time trends could be estimated. Long term time trends could not be estimated for PFOA.

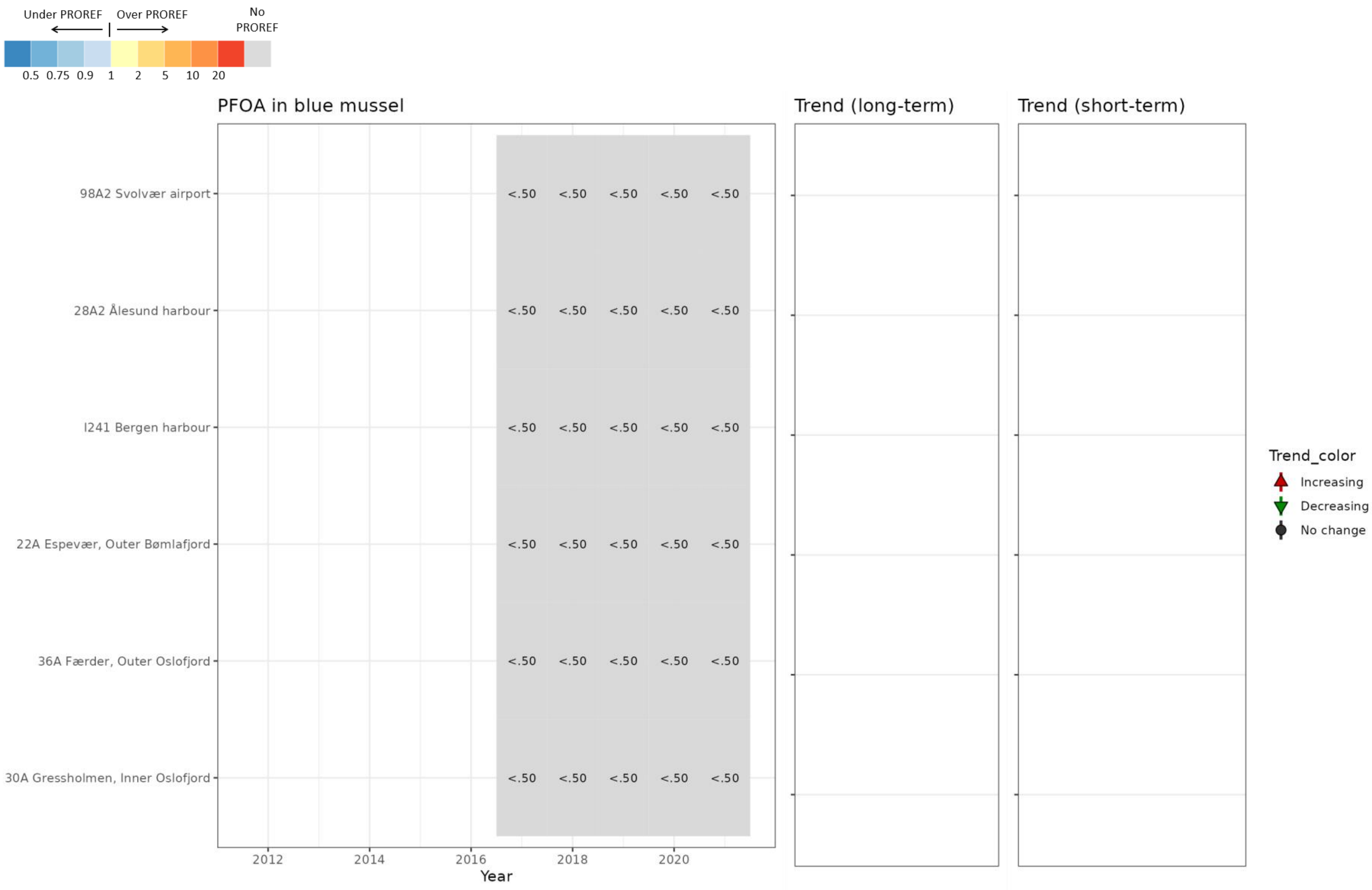
Short-term trends

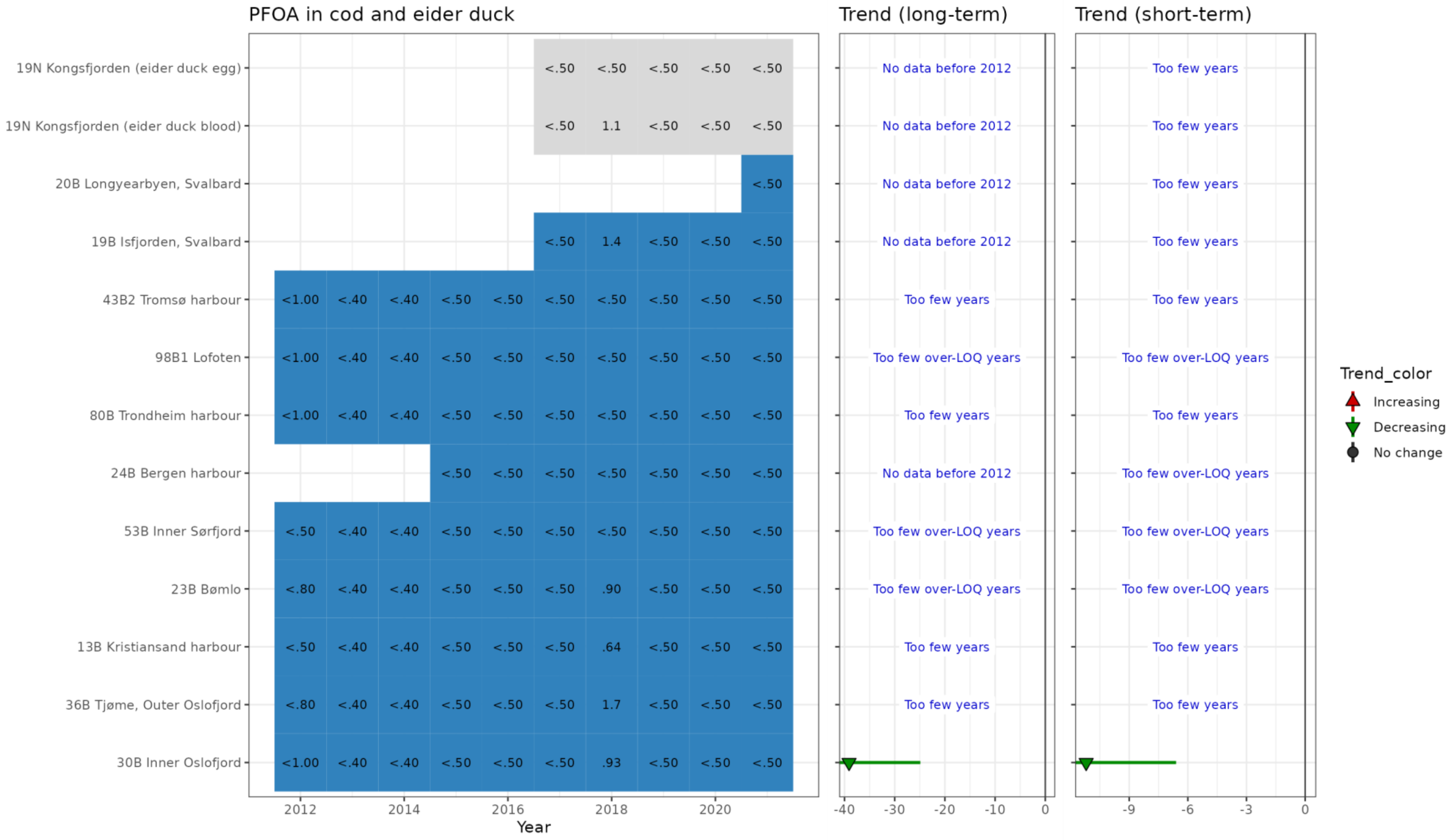
No short-term trends could be estimated in mussels.

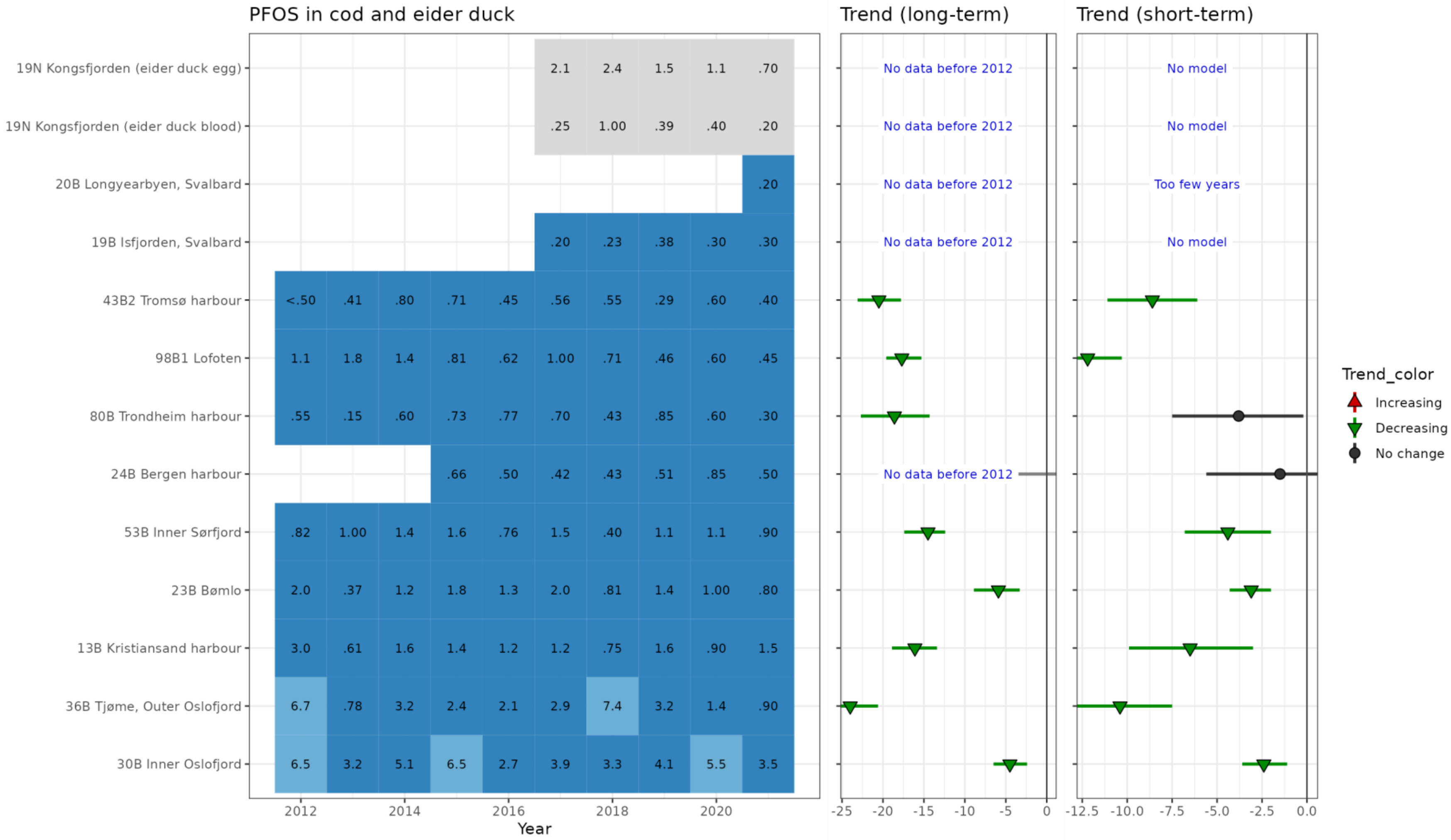
For short-term trends in cod, significant decreasing time trends were dominating for PFOSA and PFOS. Eight stations had significant decreasing short-term time trends for PFOSA and five for PFOS. For PFOA no short-term time trends could be estimated.

[8] https://pubchem.ncbi.nlm.nih.gov/classification/#hid=120

[9] https://miljostatus.miljodirektoratet.no/tema/miljogifter/prioriterte-miljogifter/perfluorerte-stoffer-pfos-pfoa-og-andre-pfas-er/


Under PROREF
Over PROREF
No PROREF
0.5 0.75 0.9 1 2 5 10 20
PFOA in blue mussel
Trend (long-term)
Trend (short-term)
98A2 Svolvær airport
28A2 Ålesund harbour
I241 Bergen harbour
22A Espevær, Outer Bømlafjord
36A Færder, Outer Oslofjord
30A Gressholmen, Inner Oslofjord
<.50 <.50 <.50 <.50 <.50
2012 2014 2016 2018 2020
Year
Trend_color
Increasing
Decreasing
No change


PFOA in cod and eider duck
Trend (long-term)
Trend (short-term)
19N Kongsfjorden (eider duck egg)
19N Kongsfjorden (eider duck blood)
20B Longyearbyen, Svalbard
19B Isfjorden, Svalbard
43B2 Tromsø harbour
98B1 Lofoten
80B Trondheim harbour
24B Bergen harbour
53B Inner Sørfjord
23B Bømlo
13B Kristiansand harbour
36B Tjøme, Outer Oslofjord
30B Inner Oslofjord
Year
No data before 2012
Too few years
Too few over-LOQ years
Trend_color
Increasing
Decreasing
No change



PFOS in cod and eider duck
Trend (long-term)
Trend (short-term)
19N Kongsfjorden (eider duck egg)
19N Kongsfjorden (eider duck blood)
20B Longyearbyen, Svalbard
19B Isfjorden, Svalbard
43B2 Tromsø harbour
98B1 Lofoten
80B Trondheim harbour
24B Bergen harbour
53B Inner Sørfjord
23B Bømlo
13B Kristiansand harbour
36B Tjøme, Outer Oslofjord
30B Inner Oslofjord
2.1 2.4 1.5 1.1 .70
.25 1.00 .39 .40 .20
.20
.20 .23 .38 .30 .30
<.50 .41 .80 .71 .45 .56 .55 .29 .60 .40
1.1 1.8 1.4 .81 .62 1.00 .71 .46 .60 .45
.55 .15 .60 .73 .77 .70 .43 .85 .60 .30
.66 .50 .42 .43 .51 .85 .50
.82 1.00 1.4 1.6 .76 1.5 .40 1.1 1.1 .90
2.0 .37 1.2 1.8 1.3 2.0 .81 1.4 1.00 .80
3.0 .61 1.6 1.4 1.2 1.2 .75 1.6 .90 1.5
6.7 .78 3.2 2.4 2.1 2.9 7.4 3.2 1.4 .90
6.5 3.2 5.1 6.5 2.7 3.9 3.3 4.1 5.5 3.5
2012 2014 2016 2018 2020
Year
No data before 2012
No model
Too few years
-25 -20 -15 -10 -5 0
-12.5 -10.0 -7.5 -5.0 -2.5 0.0
Trend_color
Increasing
Decreasing
No change

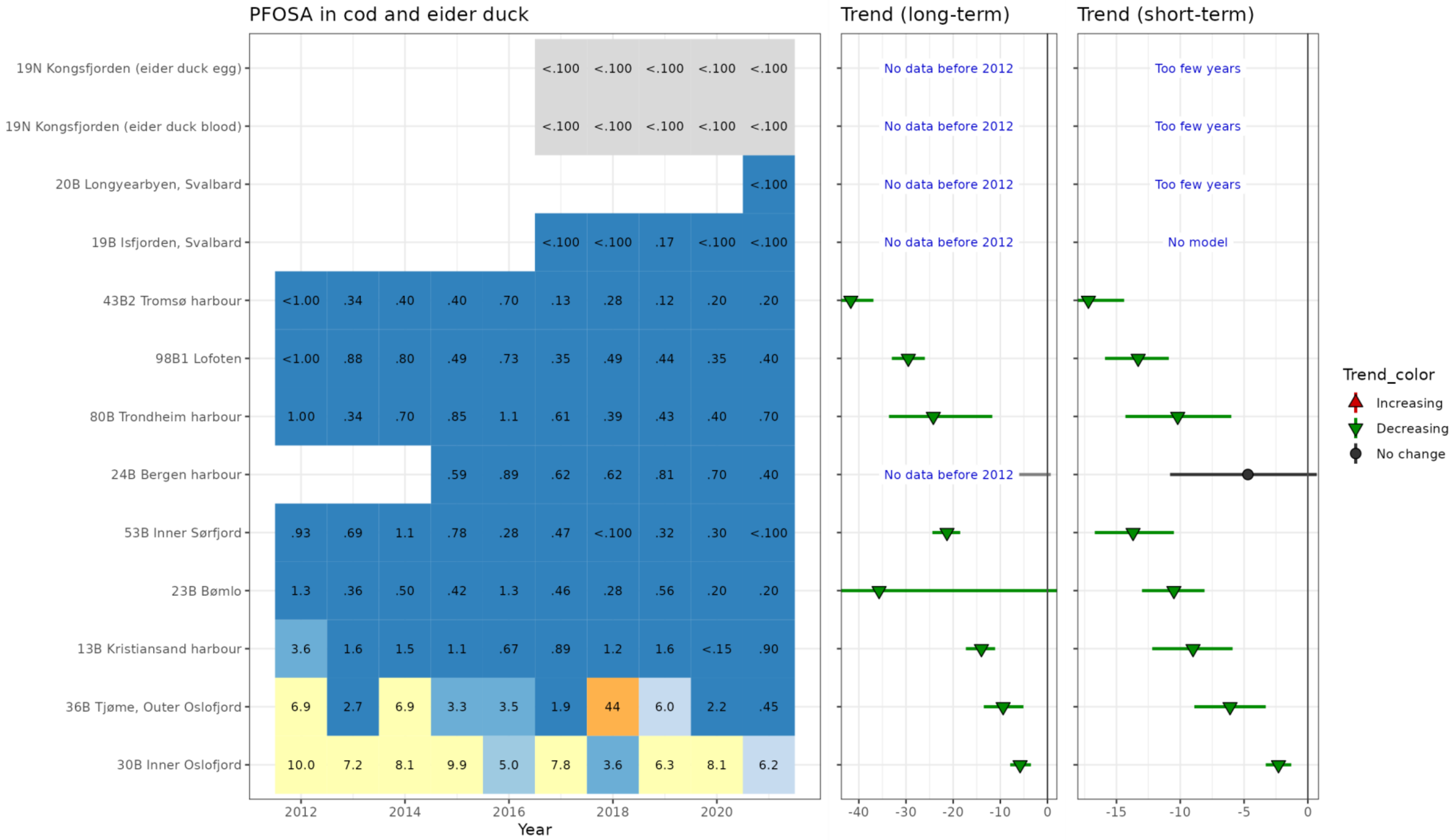


Figure 57. Heatmap and time trends of PFOS, PFOA and PFOSA in cod and eider. For full explanation of figure see example **Figure 23**.

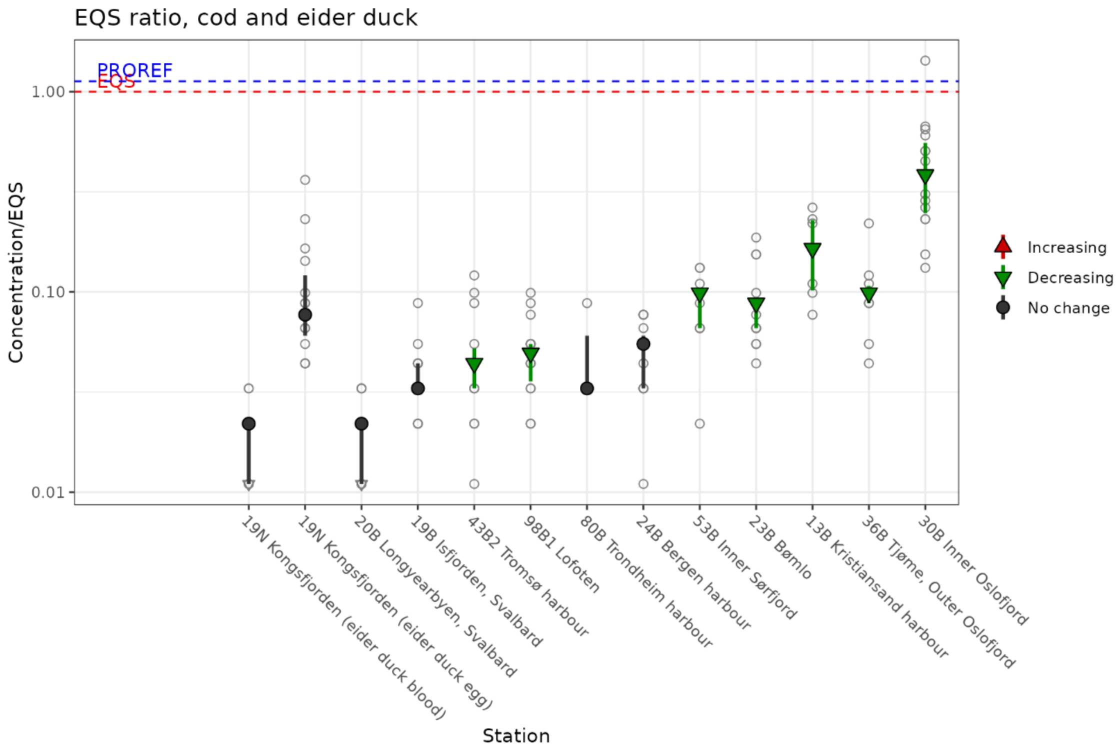


Figure 58. Ratio (concentrations/EQS) for PFOS in cod. *The y-scale is on a natural log scale.* For full explanation of figure see example **Figure 24**.

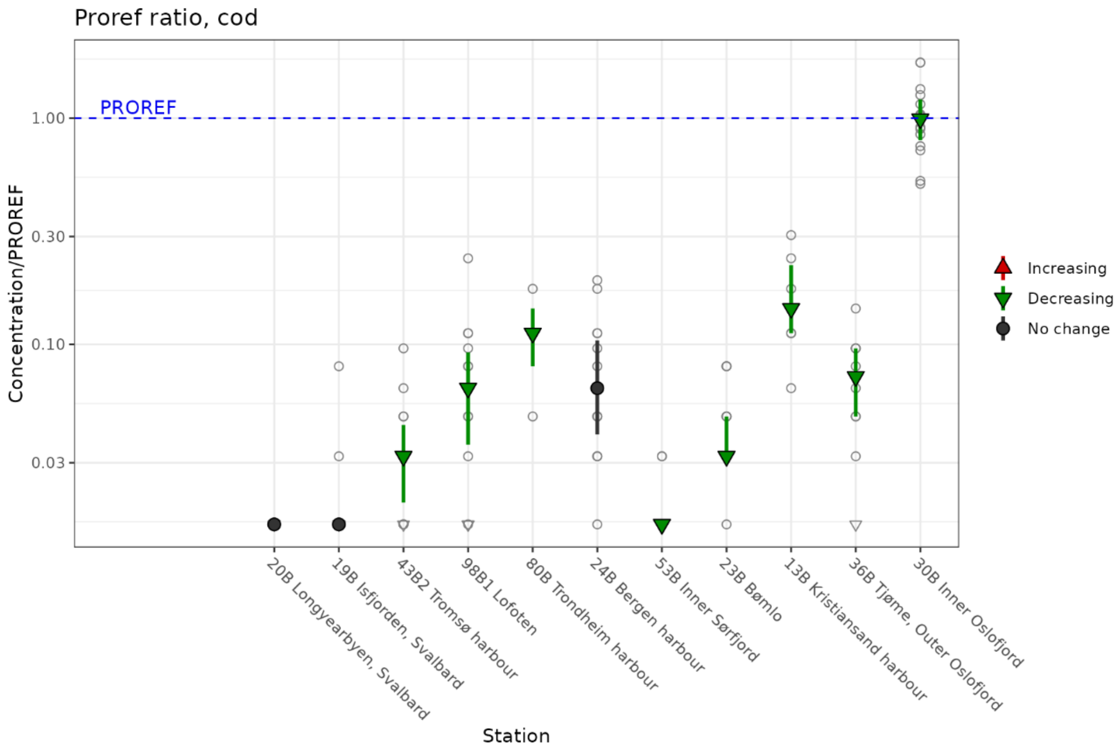


Figure 59. Ratio (concentrations/PROREF) for PFOSA in cod. *The y-scale is on a natural log scale.* For full explanation of figure see example **Figure 24**.

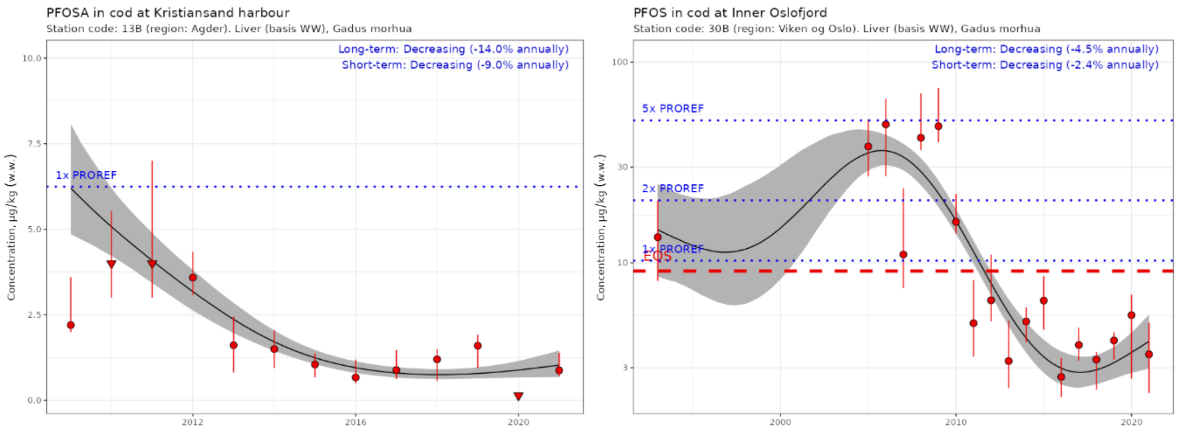


Figure 60. Two selected time trends for PFAS (PFOSA in cod at Kristiansand harbour and PFOS in cod in Inner Oslofjord). For full explanation of figure see example **Figure 25**.

Selected time trends

Two selected time trends are presented in **Figure 60**. PFOSA in cod at Kristiansand harbour (13B), and PFOS in cod from the Inner Oslofjord (30B). PFOSA was decreasing both long-term and short-term (14% and 9% annual decrease respectively). The same was observed for PFOS in cod from Inner Oslofjord (roughly 4.5% and 2,4% annually). In 2009, median concentration of PFOS in liver of cod from Inner Oslofjord was 48 µg/kg ww. Since 2009 the concentration of PFOS has been decreasing, and below EQS since 2011. Even though both trends were declining, it will be important to follow these trends as the decreases seem to have been reduced in the past few years. In a recent publication, Cousins et al. (2022) state that “we are now out of the planetary boundary for PFAS” which can be found in rainwater globally. A new field study from Norway confirms that sea spray aerosol is a source of perfluoroalkyl acids (PFAAs) to the atmosphere (Sha et al. 2022).

3.2.2.2 PBDEs

Polybrominated diphenyl ethers (BDEs) are a group of brominated flame retardants used in a variety of consumer products. They are used in electrical and electronic products, textiles, and cars. In 2013, the consumption of brominated flame retardants in Norway was estimated to 280 tons[10]. The most important commercial PBDE mixtures are banned globally by their listing in the Stockholm Convention. In Norway, production, imports, placing on the market and use of PBDEs is banned. Regulations are also in place to ensure proper management of PBDE containing wastes and stockpiles. In the present study, BDEs were analysed in blue mussel at 11 stations, cod liver at 12 stations and in eider blood and eggs at one station (**Table 1**, **Figure 61**).

EQS

There is an EQS for the sumPBDE6 (0.0085 µg/kg ww, **Table 3**), which was exceeded in all 11 blue mussel stations and 12 cod stations where BDEs were analysed. **Figure 61** and **Figure 62** show exceedances of the EQS of BDE47 as a proxy for sumPBDE6. BDE47 is one of the 6 BDEs in sumPBDE6. The exceedances are therefore correct, but non-exceedances are not necessarily correct since sumPBDE6 could exceed while only BDE47 may not.

PROREF

Except for BDE99 at station Bodø harbour (97A3), all selected BDE concentrations (BDE47, BDE99, BDE100 and BDE153) at all mussel stations were below PROREFs. The highest concentrations for BDE47 were observed at Bodø harbour (97A3) (15x EQS, but below PROREF **Figure 62**). BDE47 represents the other BDEs quite well.

For cod stations, BDE100 exceeded PROREF 2-3 times in the Inner Oslofjord (30B) (**Figure 63**), where also sumPBDE6 were exceeded (1-2 times PROREF). In Bergen harbour (24B), PROREF was exceeded (1-2 times) for BDE47 and BDE100 (**Figure 62**)).

Long-term trends

For mussels, there are few data before 2012 and therefore only a few long-term trends (**Figure 61**). Only significant decreasing time trends or no trends were found. Station Gressholmen in the Inner Oslofjord (30A) had decreasing time trends for BDE47, 99, and 100.

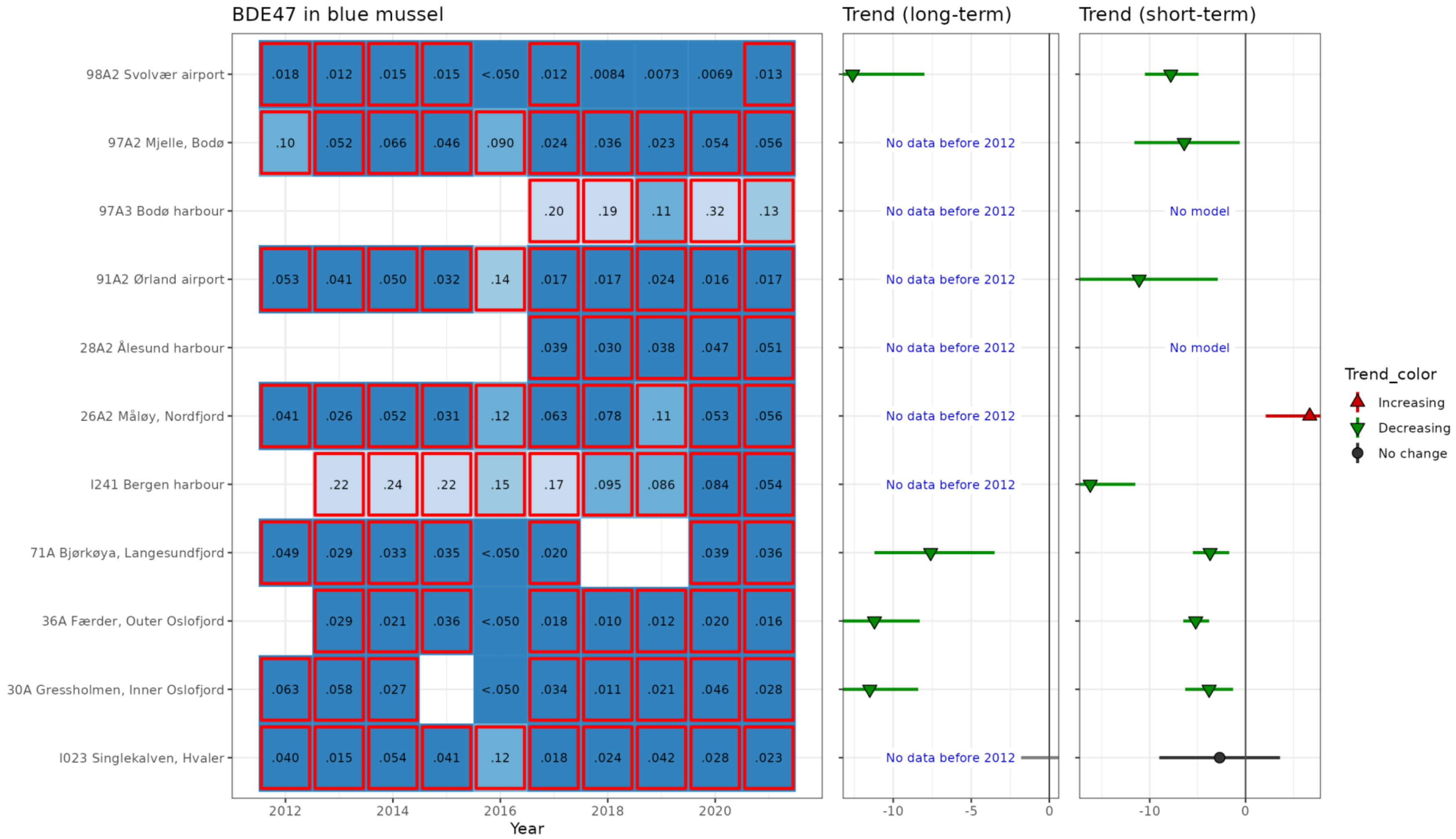
For cod, significant decreasing long-term trends were dominating for BDEs. Stations Bømlo (23B) and Tromsø harbour (43B2) had decreasing trends for all selected isomers. Stations Inner Oslofjord (30B) and Trondheim harbour (80B) had decreasing trends for all isomers that had a trend, while the other stations had a mix of decreasing or no time trends. No significant increasing long-term trends were observed.

Short-term trends

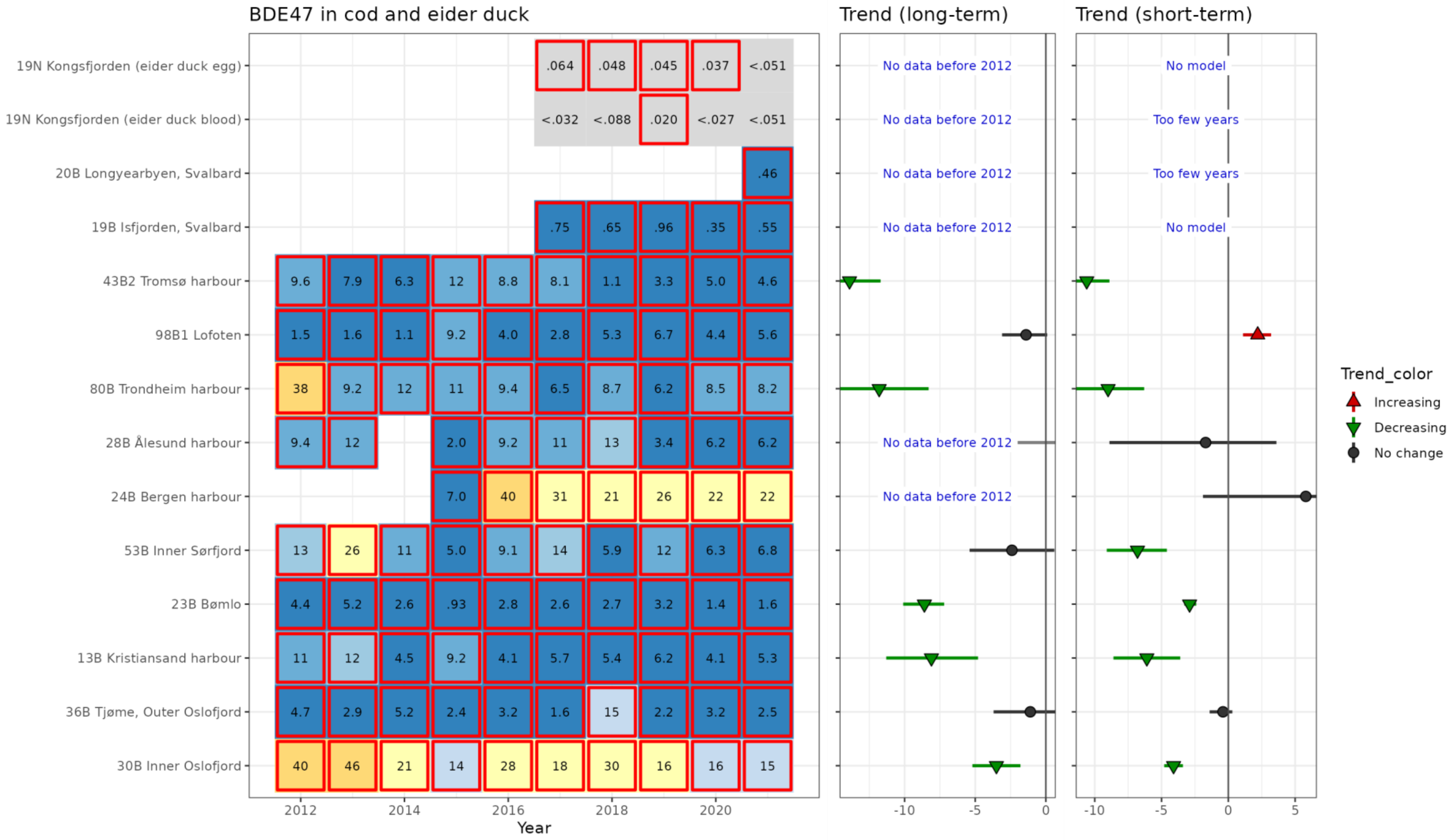
For short-term trends in blue mussel, significant decreasing time trends were observed for BDE47 at seven stations, BDE99 at three stations and BDE100 at four stations. No short-term trends could be determined in mussels for BDE153. Significant increasing short-term trends were observed at one station (Måløy in the Nordfjord (26A2)) for most BDEs (BDE47, BDE99, and BDE100).

Short-term decreasing time trends were dominating for all BDE isomers except BDE153 where only three stations had significant decreasing time trends. Significant increasing short-term time trends were observed at station Lofoten (98B1) for sumPBDE6, BDE47, and BDE100.

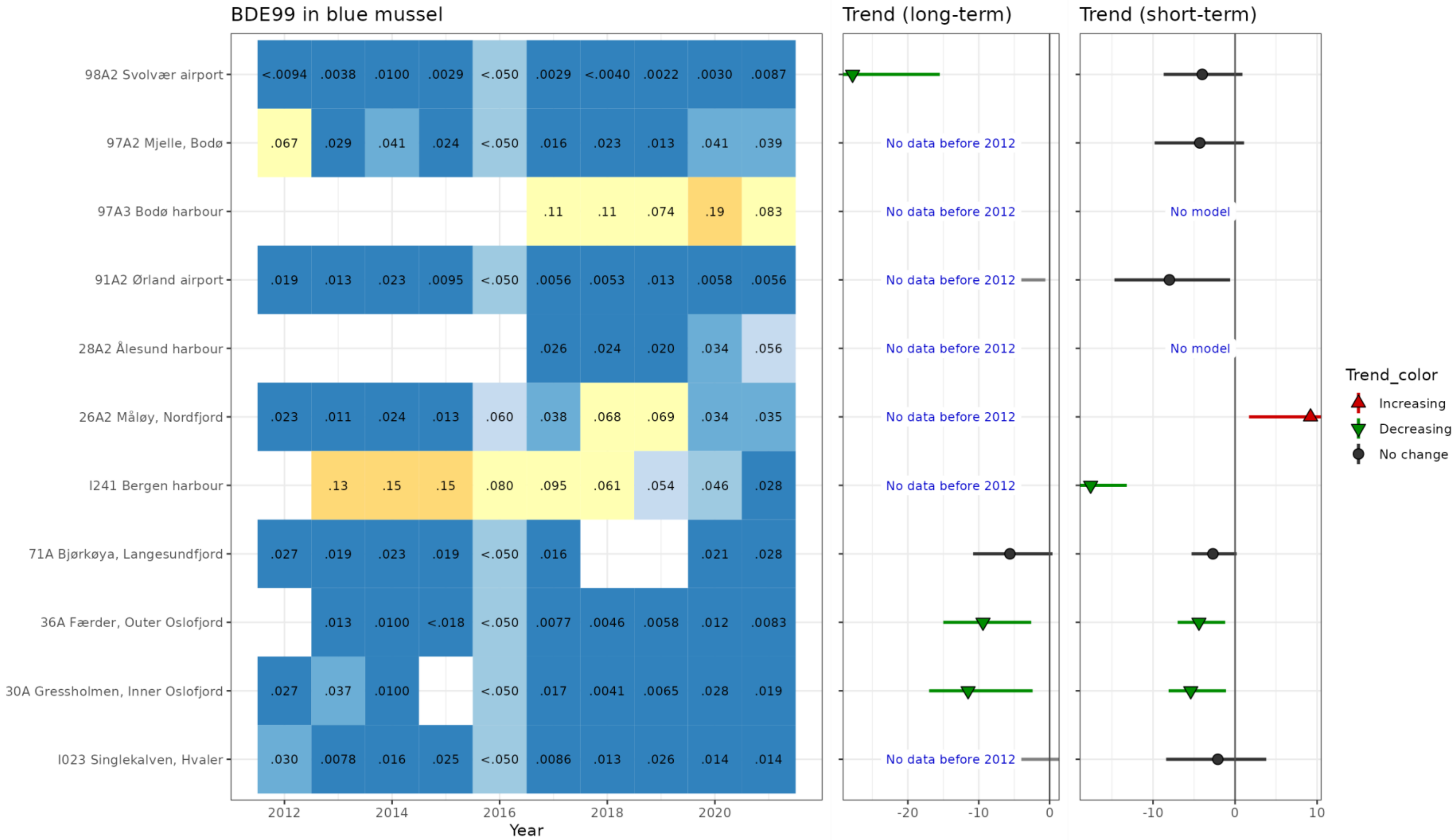
[10] https://miljostatus.miljodirektoratet.no/tema/miljogifter/prioriterte-miljogifter/bromerte-flammehemmere/

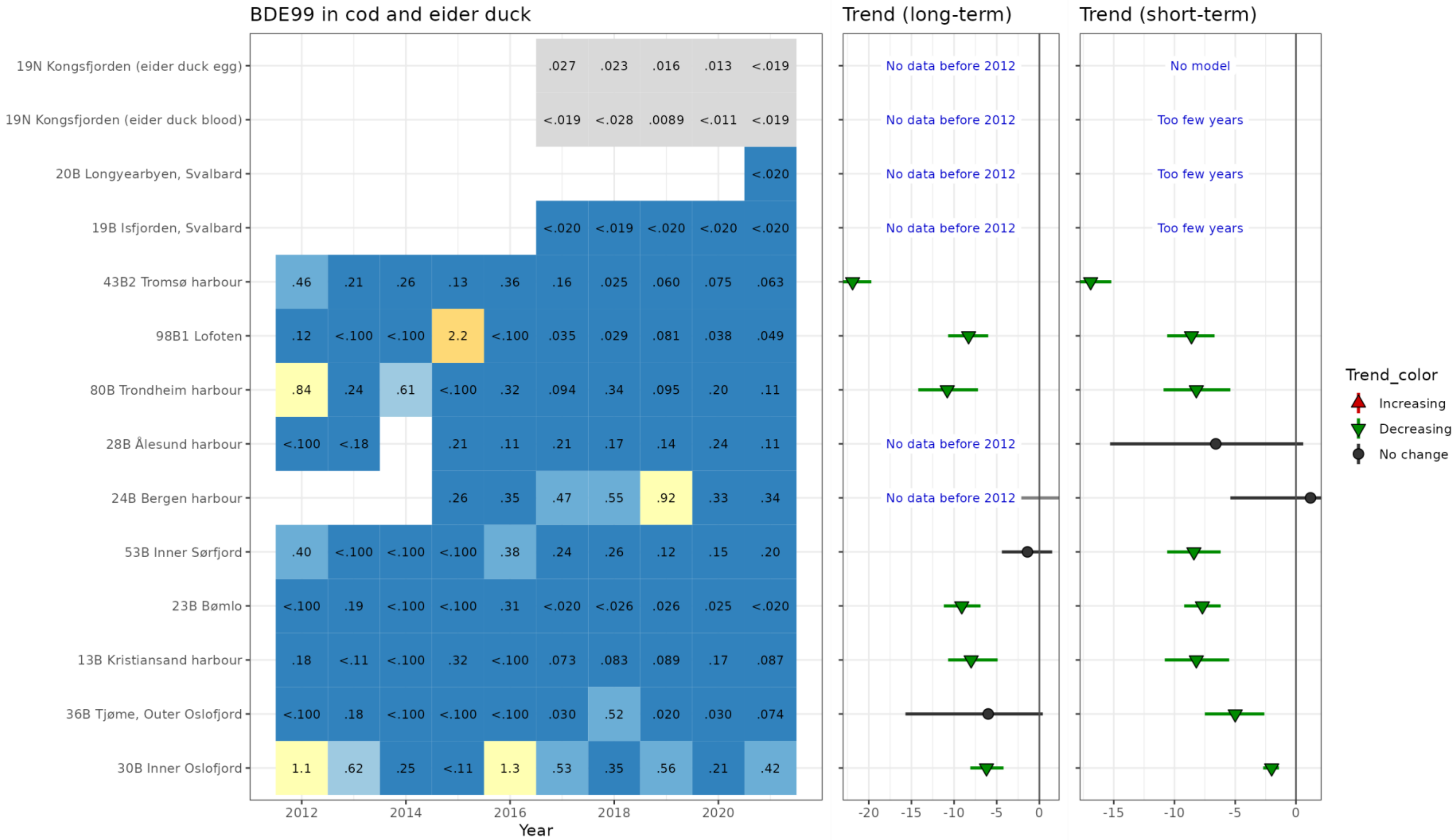


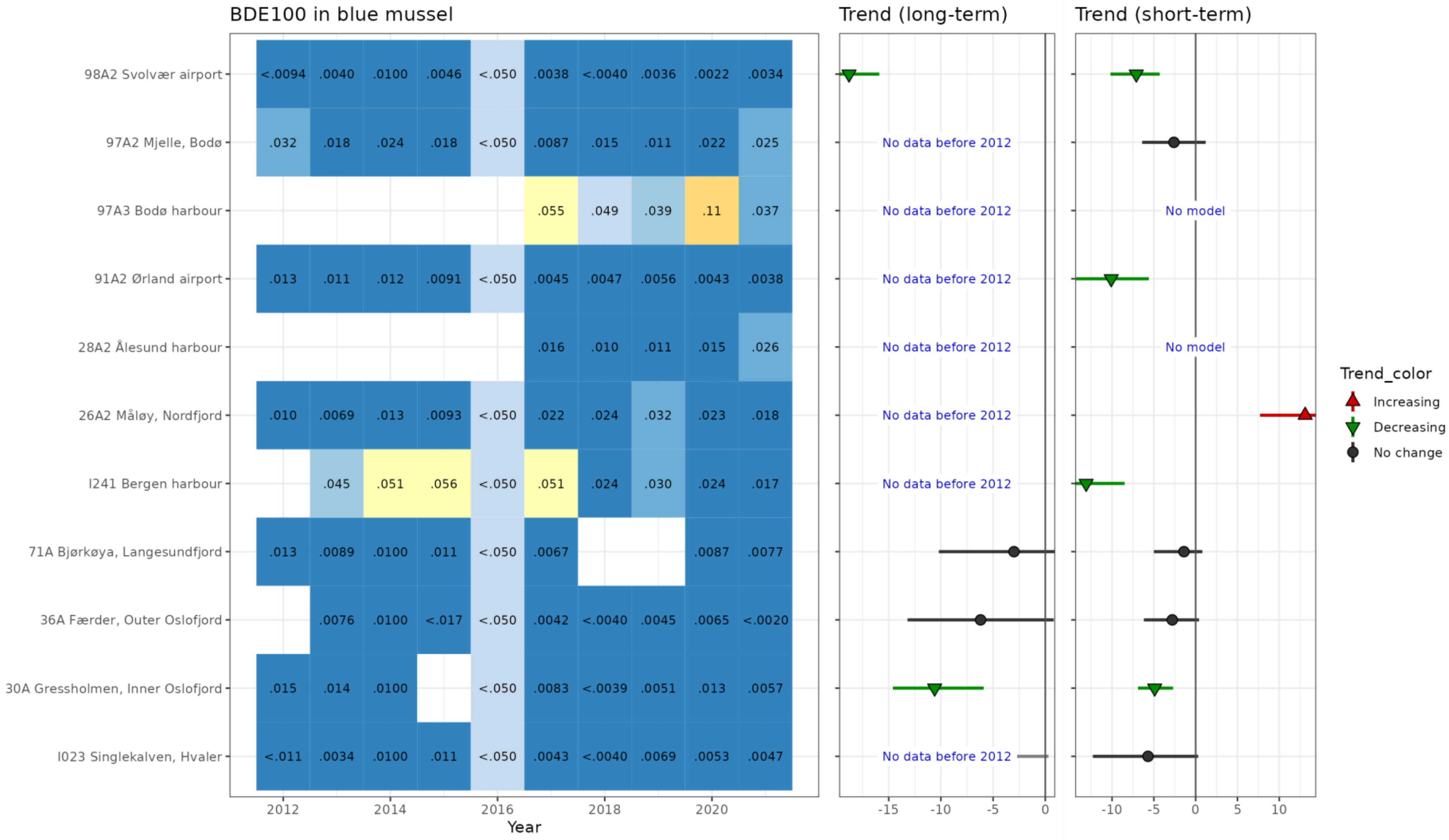
BDE47 in blue mussel
Trend (long-term)
Trend (short-term)
98A2 Svolvær airport
97A2 Mjelle, Bodø
97A3 Bodø harbour
91A2 Ørland airport
28A2 Ålesund harbour
26A2 Måløy, Nordfjord
I241 Bergen harbour
71A Bjørkøya, Langesundfjord
36A Færder, Outer Oslofjord
30A Gressholmen, Inner Oslofjord
I023 Singlekalven, Hvaler
.018 .012 .015 .015 <.050 .012 .0084 .0073 .0069 .013
.10 .052 .066 .046 .090 .024 .036 .023 .054 .056
.20 .19 .11 .32 .13
.053 .041 .050 .032 .14 .017 .017 .024 .016 .017
.039 .030 .038 .047 .051
.041 .026 .052 .031 .12 .063 .078 .11 .053 .056
.22 .24 .22 .15 .17 .095 .086 .084 .054
.049 .029 .033 .035 <.050 .020 .039 .036
.029 .021 .036 <.050 .018 .010 .012 .020 .016
.063 .058 .027 <.050 .034 .011 .021 .046 .028
.040 .015 .054 .041 .12 .018 .024 .042 .028 .023
No data before 2012
No model
2012
2014
2016
2018
2020
Year
-10
-5
0
Trend_color
Increasing
Decreasing
No change

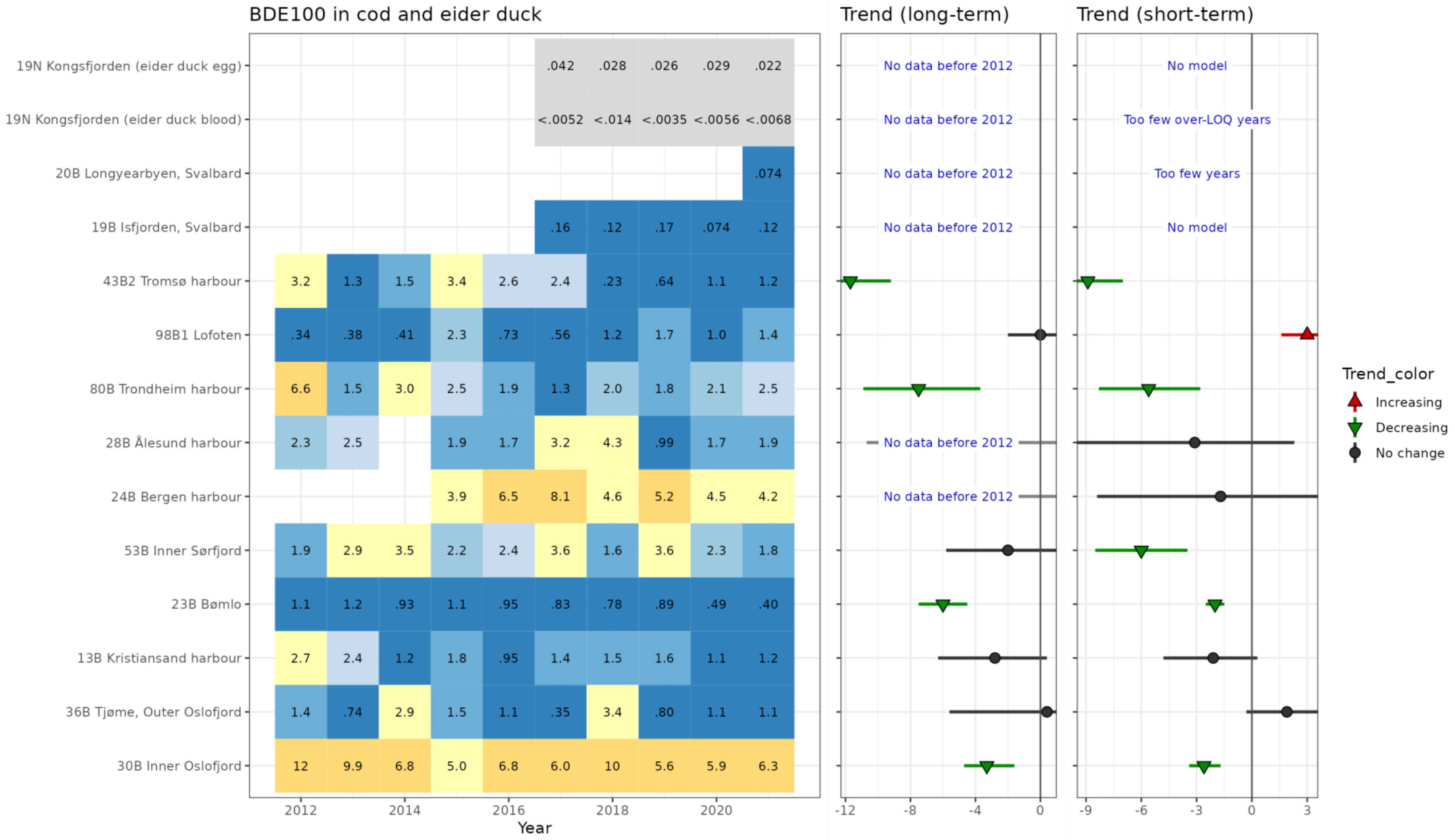


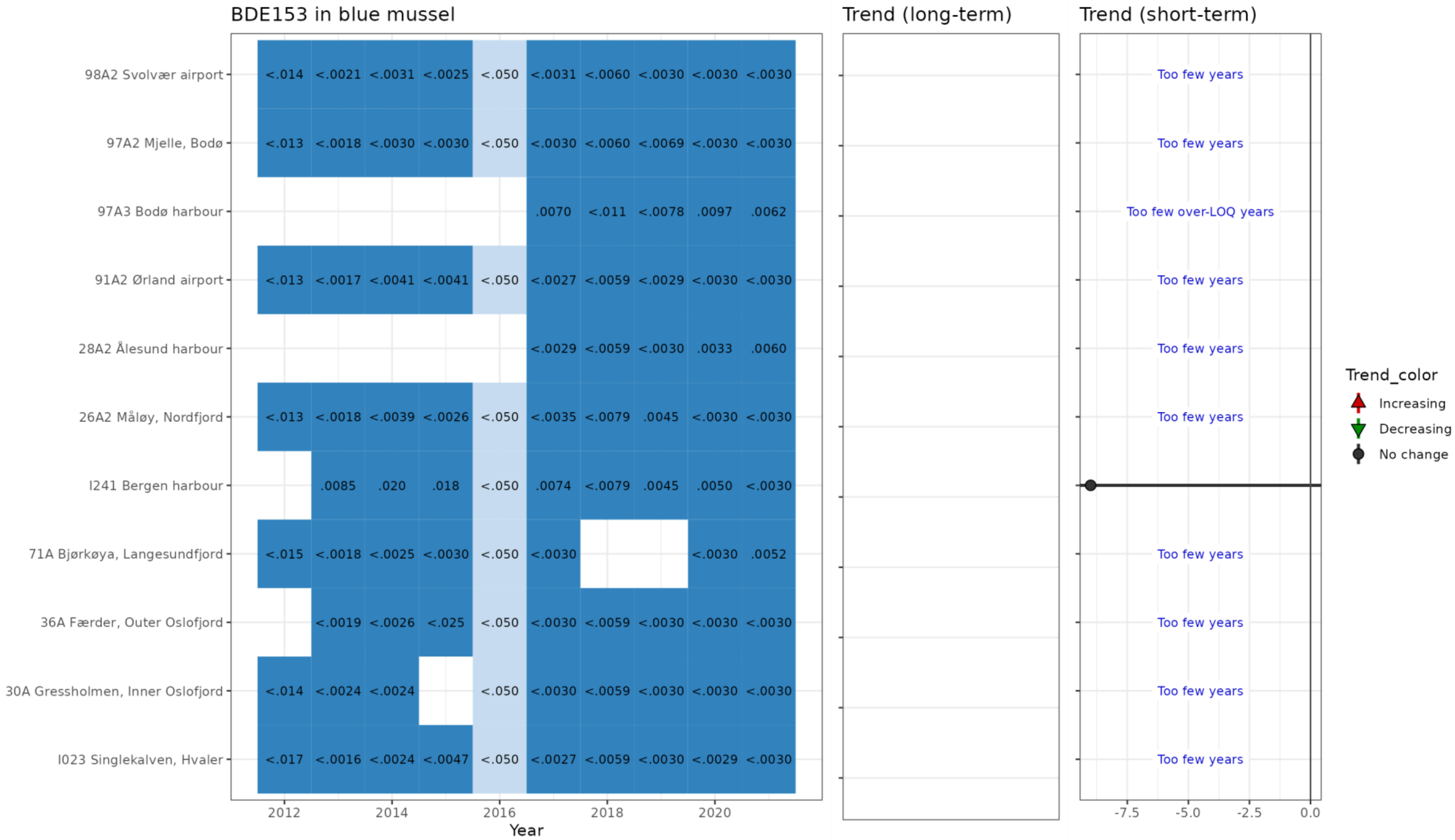
BDE47 in cod and eider duck
Trend (long-term)
Trend (short-term)
19N Kongsfjorden (eider duck egg)
19N Kongsfjorden (eider duck blood)
20B Longyearbyen, Svalbard
19B Isfjorden, Svalbard
43B2 Tromsø harbour
98B1 Lofoten
80B Trondheim harbour
28B Ålesund harbour
24B Bergen harbour
53B Inner Sørfjord
23B Bømlo
13B Kristiansand harbour
36B Tjøme, Outer Oslofjord
30B Inner Oslofjord
No data before 2012
No model
Too few years
Year
Trend_color
Increasing
Decreasing
No change


BDE99 in blue mussel
Trend (long-term)
Trend (short-term)
98A2 Svolvær airport
97A2 Mjelle, Bodø
97A3 Bodø harbour
91A2 Ørland airport
28A2 Ålesund harbour
26A2 Måløy, Nordfjord
I241 Bergen harbour
71A Bjørkøya, Langesundfjord
36A Færder, Outer Oslofjord
30A Gressholmen, Inner Oslofjord
I023 Singlekalven, Hvaler
<.0094 .0038 .0100 .0029 <.050 .0029 <.0040 .0022 .0030 .0087
.067 .029 .041 .024 <.050 .016 .023 .013 .041 .039
.11 .11 .074 .19 .083
.019 .013 .023 .0095 <.050 .0056 .0053 .013 .0058 .0056
.026 .024 .020 .034 .056
.023 .011 .024 .013 .060 .038 .068 .069 .034 .035
.13 .15 .15 .080 .095 .061 .054 .046 .028
.027 .019 .023 .019 <.050 .016 .021 .028
.013 .0100 <.018 <.050 .0077 .0046 .0058 .012 .0083
.027 .037 .0100 <.050 .017 .0041 .0065 .028 .019
.030 .0078 .016 .025 <.050 .0086 .013 .026 .014 .014
No data before 2012
No model
2012 2014 2016 2018 2020
Year
-20 -10 0
-10 0 10
Trend_color
Increasing
Decreasing
No change


BDE99 in cod and eider duck
Trend (long-term)
Trend (short-term)
19N Kongsfjorden (eider duck egg)
19N Kongsfjorden (eider duck blood)
20B Longyearbyen, Svalbard
19B Isfjorden, Svalbard
43B2 Tromsø harbour
98B1 Lofoten
80B Trondheim harbour
28B Ålesund harbour
24B Bergen harbour
53B Inner Sørfjord
23B Bømlo
13B Kristiansand harbour
36B Tjøme, Outer Oslofjord
30B Inner Oslofjord
.027 .023 .016 .013 <.019
<.019 <.028 .0089 <.011 <.019
<.020
<.020 <.019 <.020 <.020 <.020
.46 .21 .26 .13 .36 .16 .025 .060 .075 .063
.12 <.100 <.100 2.2 <.100 .035 .029 .081 .038 .049
.84 .24 .61 <.100 .32 .094 .34 .095 .20 .11
<.100 <.18 .21 .11 .21 .17 .14 .24 .11
.26 .35 .47 .55 .92 .33 .34
.40 <.100 <.100 <.100 .38 .24 .26 .12 .15 .20
<.100 .19 <.100 <.100 .31 <.020 <.026 .026 .025 <.020
.18 <.11 <.100 .32 <.100 .073 .083 .089 .17 .087
<.100 .18 <.100 <.100 <.100 .030 .52 .020 .030 .074
1.1 .62 .25 <.11 1.3 .53 .35 .56 .21 .42
2012 2014 2016 2018 2020
Year
No data before 2012
No model
Too few years
-20 -15 -10 -5 0
-15 -10 -5 0
Trend_color
Increasing
Decreasing
No change


BDE100 in blue mussel
Trend (long-term)
Trend (short-term)
98A2 Svolvær airport
97A2 Mjelle, Bodø
97A3 Bodø harbour
91A2 Ørland airport
28A2 Ålesund harbour
26A2 Måløy, Nordfjord
I241 Bergen harbour
71A Bjørkøya, Langesundfjord
36A Færder, Outer Oslofjord
30A Gressholmen, Inner Oslofjord
I023 Singlekalven, Hvaler
<.0094 .0040 .0100 .0046 <.050 .0038 <.0040 .0036 .0022 .0034
.032 .018 .024 .018 <.050 .0087 .015 .011 .022 .025
.055 .049 .039 .11 .037
.013 .011 .012 .0091 <.050 .0045 .0047 .0056 .0043 .0038
.016 .010 .011 .015 .026
.010 .0069 .013 .0093 <.050 .022 .024 .032 .023 .018
.045 .051 .056 <.050 .051 .024 .030 .024 .017
.013 .0089 .0100 .011 <.050 .0067 .0087 .0077
.0076 .0100 <.017 <.050 .0042 <.0040 .0045 .0065 <.0020
.015 .014 .0100 <.050 .0083 <.0039 .0051 .013 .0057
<.011 .0034 .0100 .011 <.050 .0043 <.0040 .0069 .0053 .0047
No data before 2012
No model
2012 2014 2016 2018 2020
Year
-15 -10 -5 0
-10 -5 0 5 10
Trend_color
Increasing
Decreasing
No change


BDE100 in cod and eider duck
Trend (long-term)
Trend (short-term)
19N Kongsfjorden (eider duck egg)
19N Kongsfjorden (eider duck blood)
20B Longyearbyen, Svalbard
19B Isfjorden, Svalbard
43B2 Tromsø harbour
98B1 Lofoten
80B Trondheim harbour
28B Ålesund harbour
24B Bergen harbour
53B Inner Sørfjord
23B Bømlo
13B Kristiansand harbour
36B Tjøme, Outer Oslofjord
30B Inner Oslofjord
Year
No data before 2012
No model
Too few over-LOQ years
Too few years
Trend_color
Increasing
Decreasing
No change



Station	2012	2013	2014	2015	2016	2017	2018	2019	2020	2021
98A2 Svolvær airport	<.014	<.0021	<.0031	<.0025	<.050	<.0031	<.0060	<.0030	<.0030	<.0030
97A2 Mjelle, Bodø	<.013	<.0018	<.0030	<.0030	<.050	<.0030	<.0060	<.0069	<.0030	<.0030
97A3 Bodø harbour						.0070	<.011	<.0078	.0097	.0062
91A2 Ørland airport	<.013	<.0017	<.0041	<.0041	<.050	<.0027	<.0059	<.0029	<.0030	<.0030
28A2 Ålesund harbour						<.0029	<.0059	<.0030	.0033	.0060
26A2 Måløy, Nordfjord	<.013	<.0018	<.0039	<.0026	<.050	<.0035	<.0079	.0045	<.0030	<.0030
I241 Bergen harbour		.0085	.020	.018	<.050	.0074	<.0079	.0045	.0050	<.0030
71A Bjørkøya, Langesundfjord	<.015	<.0018	<.0025	<.0030	<.050	<.0030			<.0030	.0052
36A Færder, Outer Oslofjord		<.0019	<.0026	<.025	<.050	<.0030	<.0059	<.0030	<.0030	<.0030
30A Gressholmen, Inner Oslofjord	<.014	<.0024	<.0024		<.050	<.0030	<.0059	<.0030	<.0030	<.0030
I023 Singlekalven, Hvaler	<.017	<.0016	<.0024	<.0047	<.050	<.0027	<.0059	<.0030	<.0029	<.0030

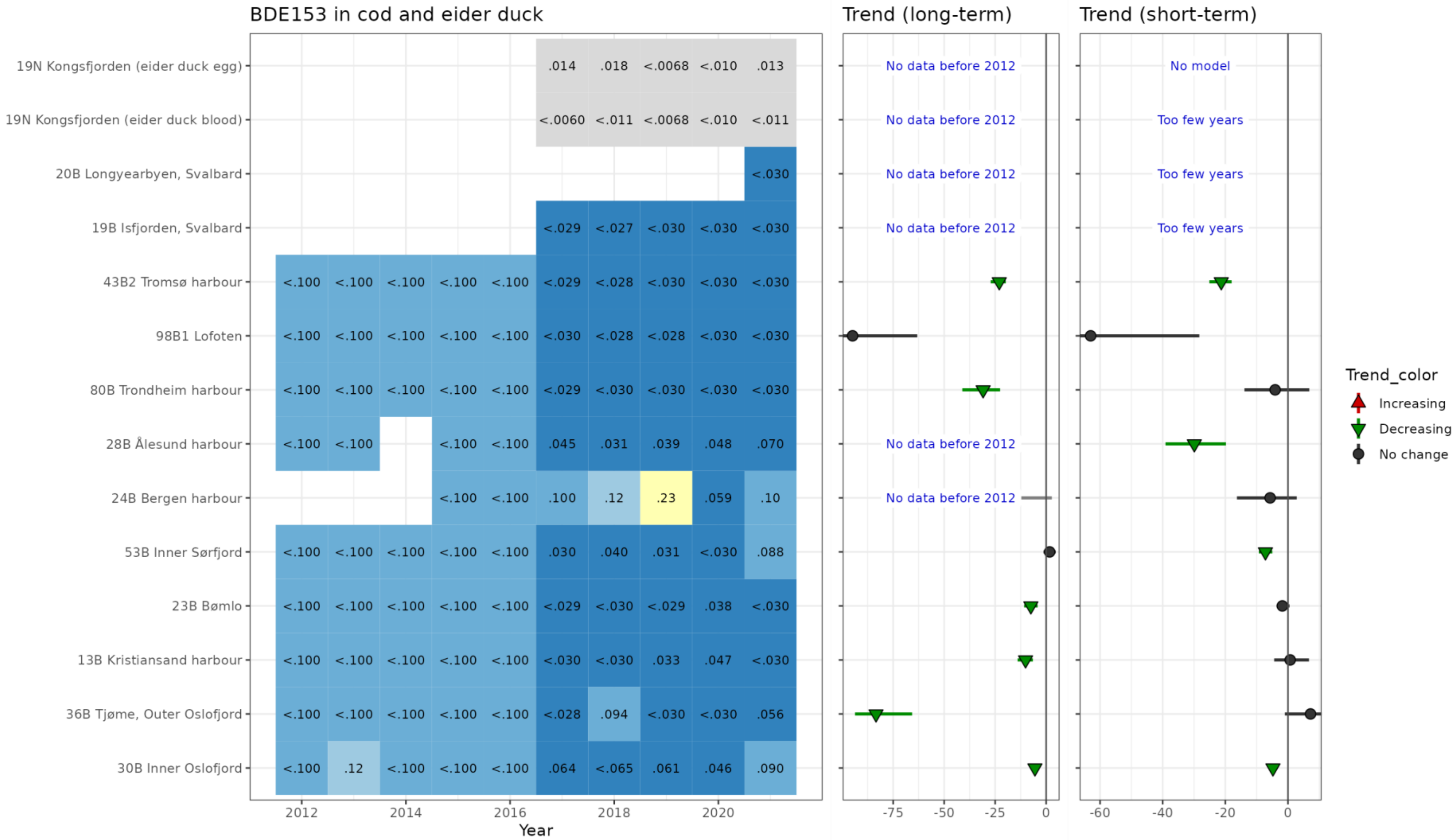


Figure 61. Heatmap and time trends of BDE47, BDE99, BDE100 and BDE153 in blue mussel, and cod and eider. For full explanation of figure see example **Figure 23**.

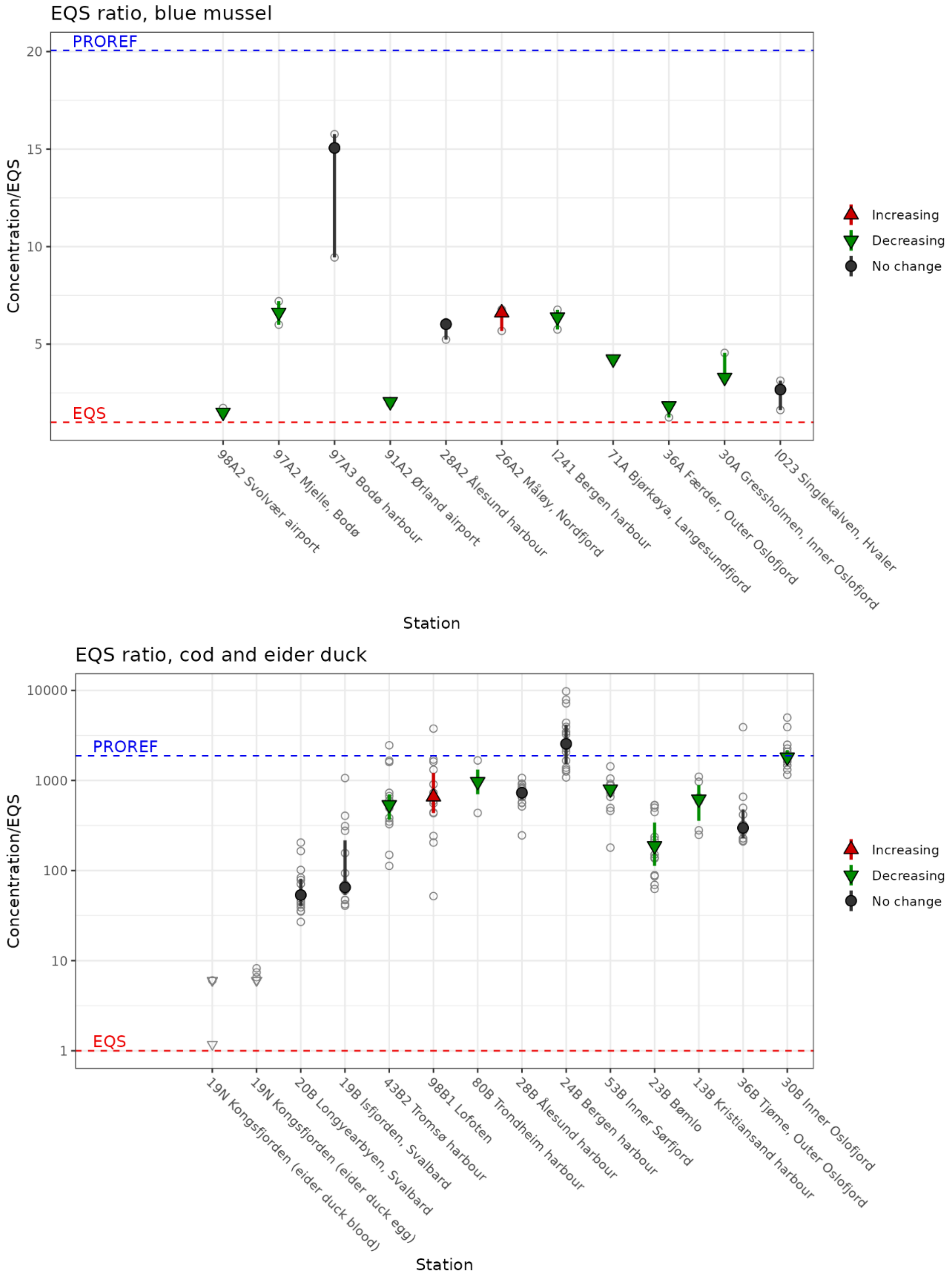


Figure 62. Ratios (concentrations/EQS) for BDE47 in blue mussel (upper panel) and in cod (lower panel). **The EQS is for sumPBDE6, therefore all exceedances indicated are correct, but lack of exceedance is not necessarily correct**. *The y-axis for cod is on a natural log scale.* For full explanation of figure see example **Figure 24**.

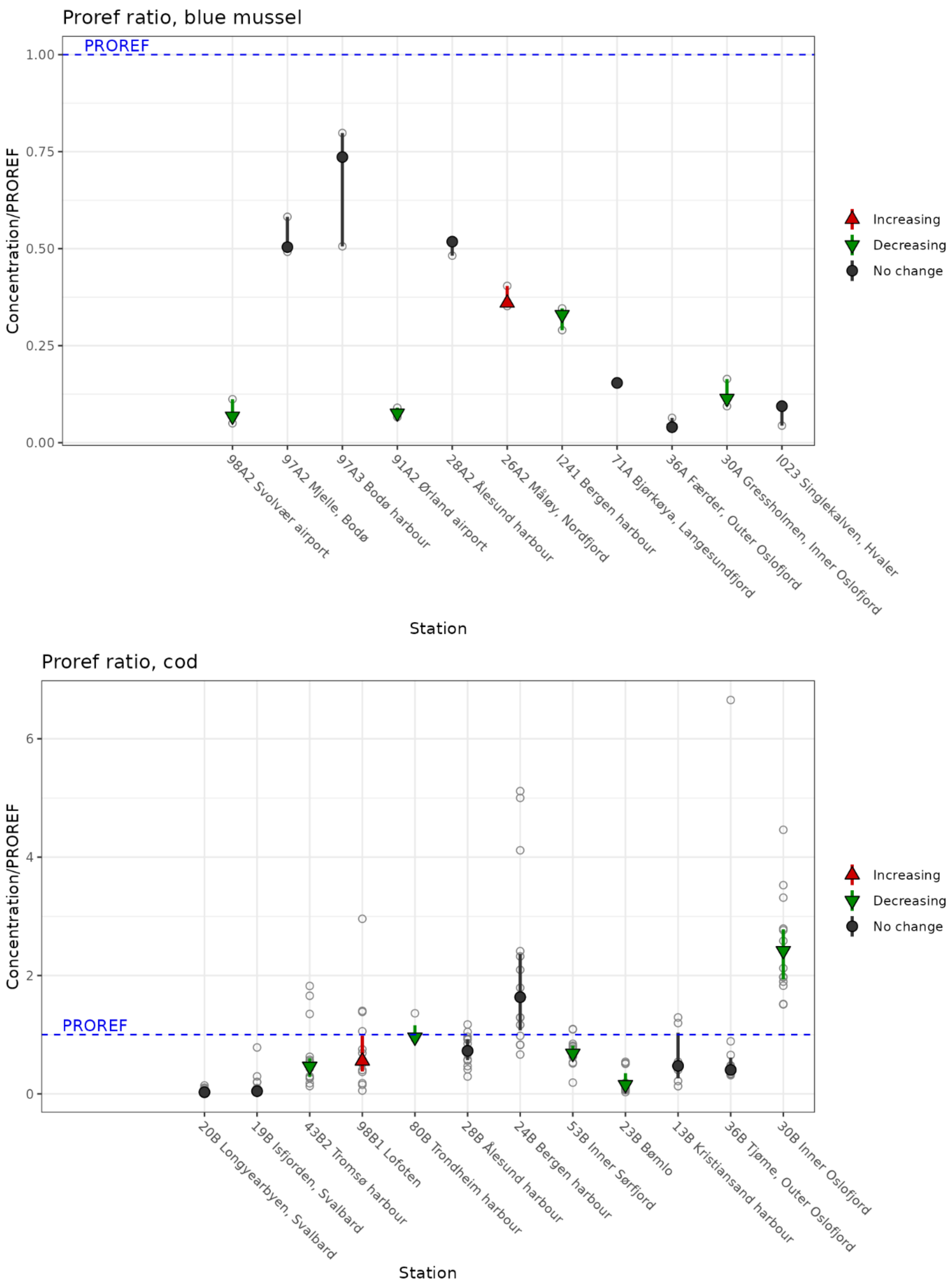


Figure 63. Ratios (concentration/PROREF) for BDE100 in blue mussel (upper panel) and in cod (lower panel). For full explanation of figure see example **Figure 24**.

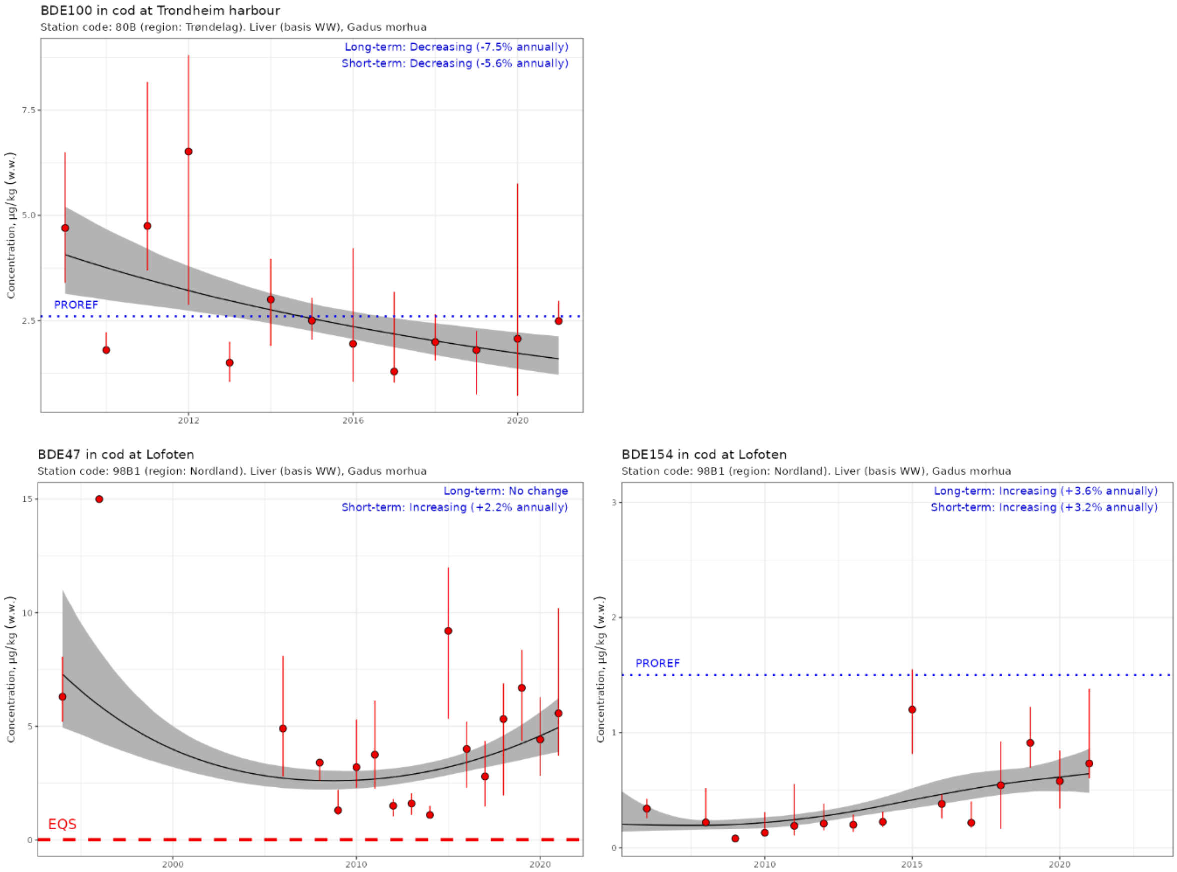


Figure 64. Three selected time trends for BDEs. The time scale for BDE154 (lower panel) has been shortened, and only shows data from 2006 and onwards. For full explanation of figure see example **Figure 25**.

Selected time trends

Three selected time trends are shown in **Figure 64**. BDE100 in cod at Trondheim harbour (80B) shows the most common picture that BDE concentrations are decreasing over time. In 2012, the median concentration of BDE100 in cod liver from Trondheim Harbour was 6.55 µg/kg ww and has had a significant decrease since 2012 (5.6% annual decrease). BDE47 concentrations in cod at Lofoten (98B1) are however, increasing short-term. Also, BDE154 at the same station were increasing. BDE154 was not selected contaminant in 2021, but data for time trends are presented in **Figure S9**, Supporting data.

3.2.2.3 PAHs

Polycyclic aromatic hydrocarbons (PAHs) are a class of organic compounds produced by incomplete combustion or high-pressure processes. PAHs form when complex organic substances are exposed to high temperatures or pressures. The main sources of PAH in coastal waters include discharges from smelting industry and waste incinerators. Creosote impregnated wood is also an important source. In 2017, 77 tons of PAH was released in Norway, and there has been an 70% reduction in discharges of PAH since 1995[11]. The Convention on Long-range Transboundary Air Pollution (LRTAP) impose parties to introduce measures to control emissions of PAH to air from major stationary sources. However, emissions and releases continue in Norway and other countries. High PAH levels are therefore reported in air in Norway, with three to four times higher concentrations in Southern Norway than in the Arctic, at Svalbard (Bohlin-Nizzetto et al. 2020). In the present study, PAHs were analysed in blue mussel at seven stations (**Table 1**, **Figure 65**).

EQS

Five PAHs have EQSs in biota; naphthalene (NAP) (2 400 µg/kg ww, **Table 3**), anthracene (ANT) (2 400 µg/kg ww), fluoranthene (FLU) (30 µg/kg ww), benzo[a]pyrene (BAP) (5 µg/kg ww), and benzo(a)anthracene (BAA) (300 µg/kg ww). In mussel stations, only FLU concentrations exceeded EQS at one station (Akershuskaia in the Inner Oslofjord (I301)). NAP has an EQS, and therefore a heatmap is shown even though NAP is not among the selected compounds. NAP is volatile, and therefore concentrations are more variable than heavier PAHs.

PAHs are metabolised by cod, and therefore there are no measurements of PAHs in cod. However, PAH-metabolites in cod bile are analysed at some stations, see **chapter 3.2.3.2.**

PROREF

For exceedance of PROREF, selected PAHs are; FLU, BAP, BAA, and PYR. The main picture for exceedances of PROREF is that one station (Akershuskaia (I301)) and PYR concentrations had most of the exceedances. Station Akershuskaia (I301) exceeded PROREF for all PAHs, highest exceedances were seen for PYR (>10, **Figure 67**), but FLU (**Figure 66**), BAA (**Figure 68**, and ANT (**Figure 69**) exceeded PROREF up to 10 times. FLU, BAA, and ANT had concentrations exceeding PROREF at one station in addition to I301. PYR concentrations were above PROREF at all measured mussel stations except one.

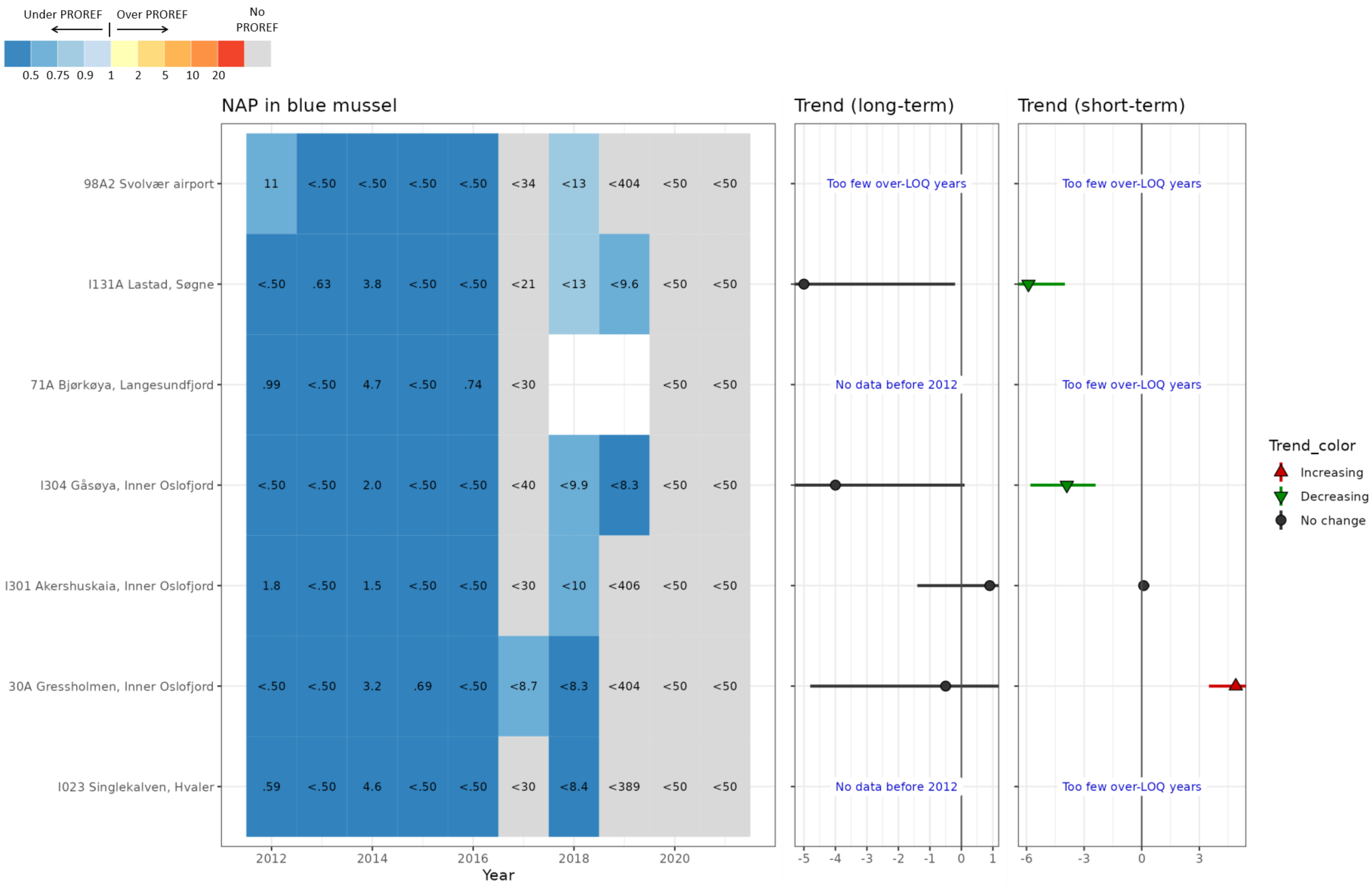
Long-term trends

Significant long-term trends in mussel were only decreasing, but no time trends were also observed. Four stations had significant decreasing time trends, Gressholmen (30A), Akershuskaia (I301) and Gåsøya (I304) in the Inner Oslofjord, and at Lastad at Søgne (I131A).

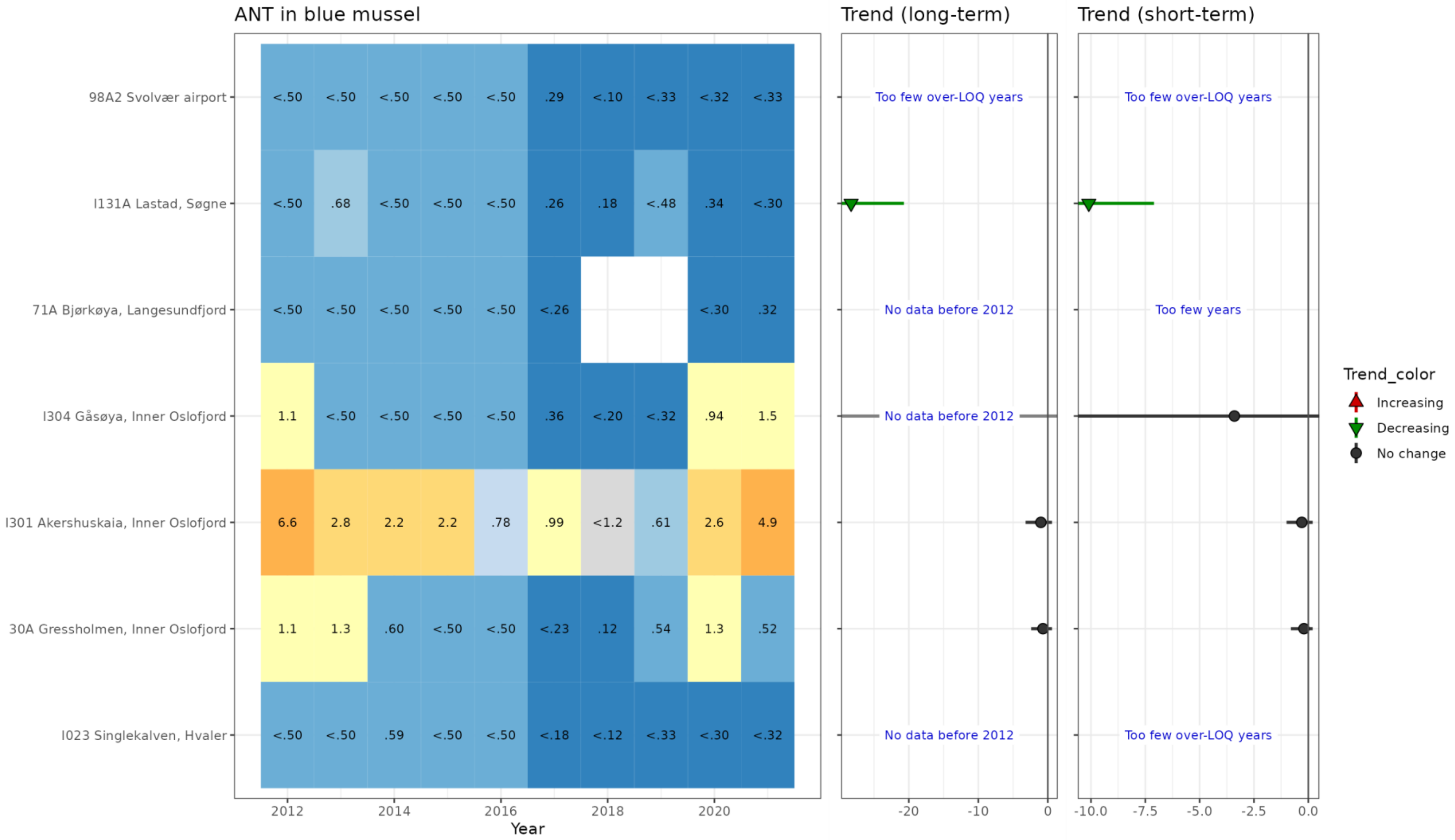
Short-term trends

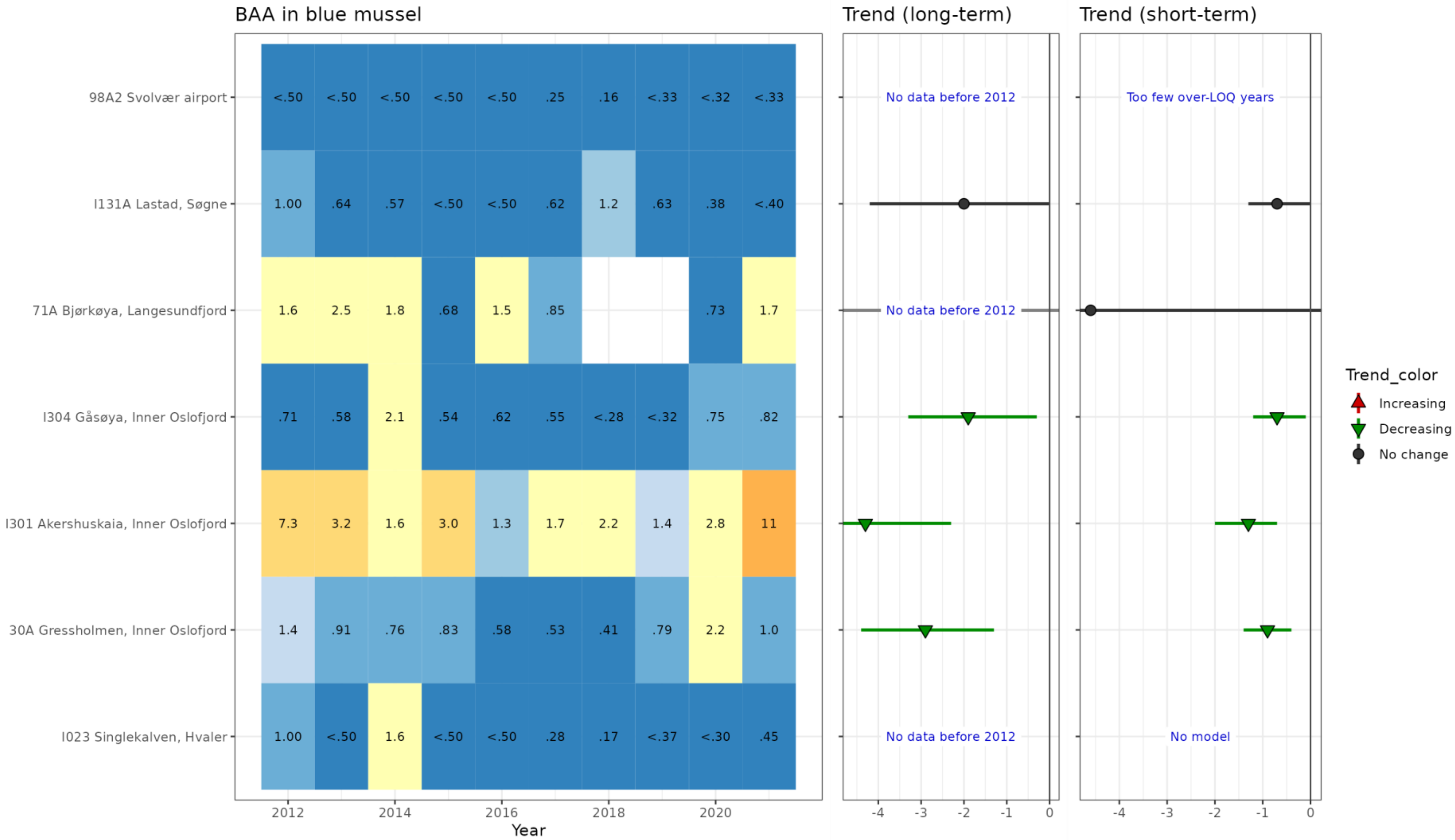
Short term trends were dominated by no trends, but some significant decreasing time trends were observed at four stations (30A, I301, I304, and I131A).

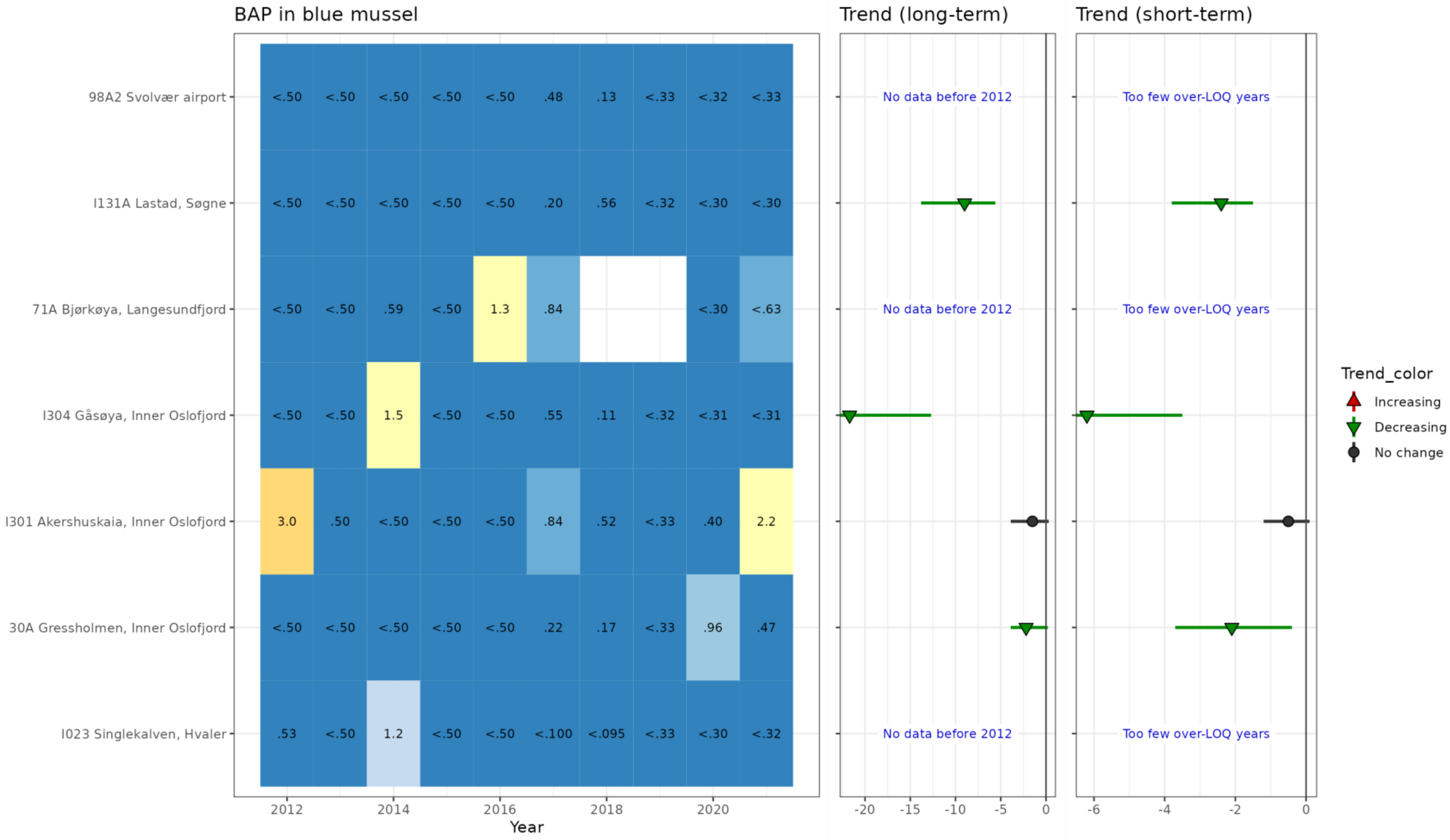
[11] https://miljostatus.miljodirektoratet.no/tema/miljogifter/prioriterte-miljogifter/polysykliske-aromatiske-hydrokarboner-pah/

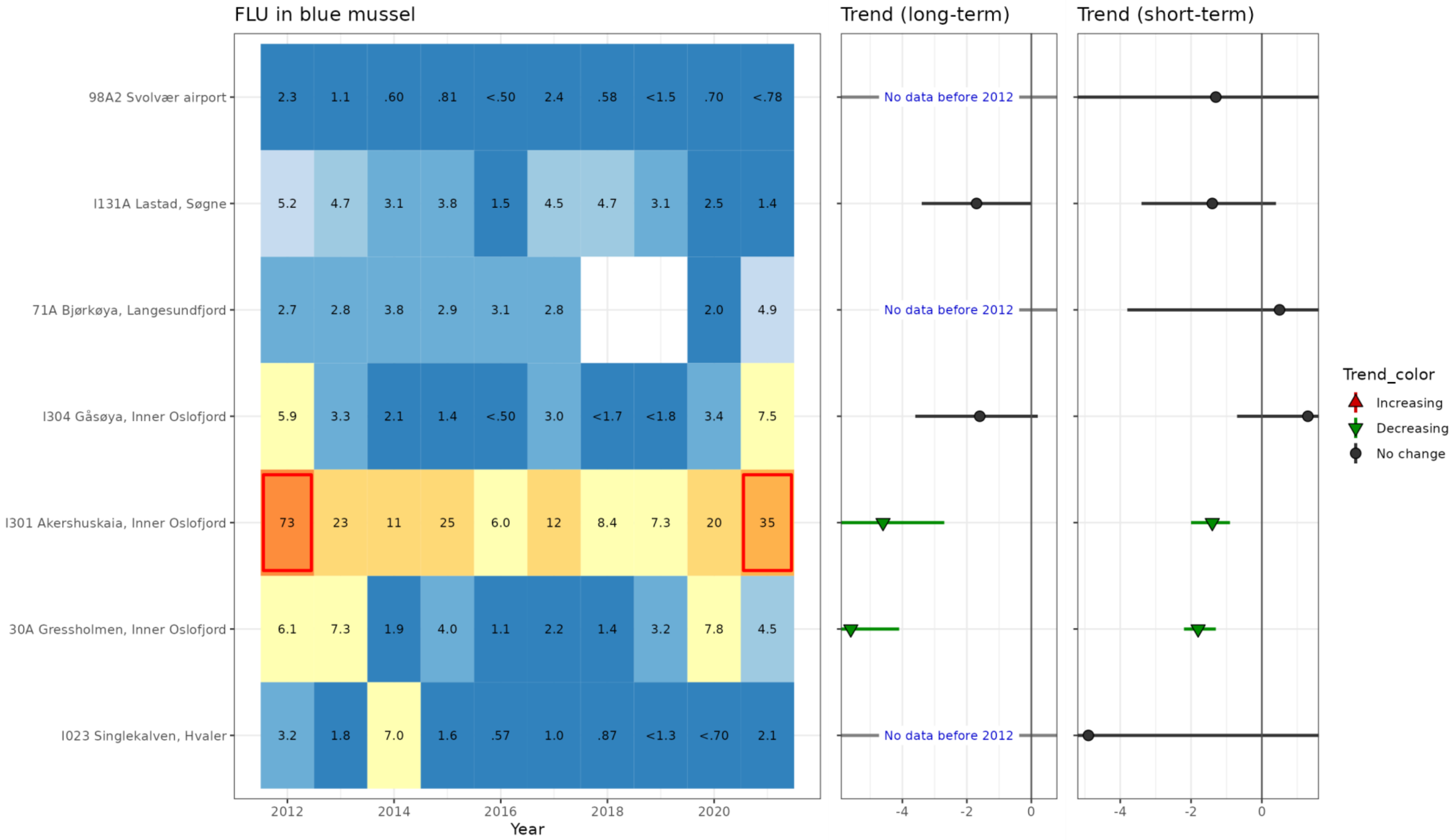


Under PROREF
Over PROREF
No PROREF
0.5 0.75 0.9 1 2 5 10 20
NAP in blue mussel
Trend (long-term)
Trend (short-term)
98A2 Svolvær airport
I131A Lastad, Søgne
71A Bjørkøya, Langesundfjord
I304 Gåsøya, Inner Oslofjord
I301 Akershuskaia, Inner Oslofjord
30A Gressholmen, Inner Oslofjord
I023 Singlekalven, Hvaler
11 <.50 <.50 <.50 <.50 <34 <13 <404 <50 <50
<.50 .63 3.8 <.50 <.50 <21 <13 <9.6 <50 <50
.99 <.50 4.7 <.50 .74 <30 <50 <50
<.50 <.50 2.0 <.50 <.50 <40 <9.9 <8.3 <50 <50
1.8 <.50 1.5 <.50 <.50 <30 <10 <406 <50 <50
<.50 <.50 3.2 .69 <.50 <8.7 <8.3 <404 <50 <50
.59 <.50 4.6 <.50 <.50 <30 <8.4 <389 <50 <50
Too few over-LOQ years
No data before 2012
2012 2014 2016 2018 2020
Year
-5 -4 -3 -2 -1 0 1
-6 -3 0 3
Trend_color
Increasing
Decreasing
No change


ANT in blue mussel
Trend (long-term)
Trend (short-term)
98A2 Svolvær airport
I131A Lastad, Søgne
71A Bjørkøya, Langesundfjord
I304 Gåsøya, Inner Oslofjord
I301 Akershuskaia, Inner Oslofjord
30A Gressholmen, Inner Oslofjord
I023 Singlekalven, Hvaler
Too few over-LOQ years
No data before 2012
Too few years
Year
Trend_color
Increasing
Decreasing
No change


BAA in blue mussel
Trend (long-term)
Trend (short-term)
98A2 Svolvær airport
I131A Lastad, Søgne
71A Bjørkøya, Langesundfjord
I304 Gåsøya, Inner Oslofjord
I301 Akershuskaia, Inner Oslofjord
30A Gressholmen, Inner Oslofjord
I023 Singlekalven, Hvaler
<.50 <.50 <.50 <.50 <.50 .25 .16 <.33 <.32 <.33
1.00 .64 .57 <.50 <.50 .62 1.2 .63 .38 <.40
1.6 2.5 1.8 .68 1.5 .85 .73 1.7
.71 .58 2.1 .54 .62 .55 <.28 <.32 .75 .82
7.3 3.2 1.6 3.0 1.3 1.7 2.2 1.4 2.8 11
1.4 .91 .76 .83 .58 .53 .41 .79 2.2 1.0
1.00 <.50 1.6 <.50 <.50 .28 .17 <.37 <.30 .45
2012 2014 2016 2018 2020
Year
No data before 2012
Too few over-LOQ years
No model
-4 -3 -2 -1 0
Trend_color
Increasing
Decreasing
No change


BAP in blue mussel
Trend (long-term)
Trend (short-term)
98A2 Svolvær airport
I131A Lastad, Søgne
71A Bjørkøya, Langesundfjord
I304 Gåsøya, Inner Oslofjord
I301 Akershuskaia, Inner Oslofjord
30A Gressholmen, Inner Oslofjord
I023 Singlekalven, Hvaler
Year
No data before 2012
Too few over-LOQ years
Trend_color
Increasing
Decreasing
No change


FLU in blue mussel
Trend (long-term)
Trend (short-term)
98A2 Svolvær airport
I131A Lastad, Søgne
71A Bjørkøya, Langesundfjord
I304 Gåsøya, Inner Oslofjord
I301 Akershuskaia, Inner Oslofjord
30A Gressholmen, Inner Oslofjord
I023 Singlekalven, Hvaler
2.3 1.1 .60 .81 <.50 2.4 .58 <1.5 .70 <.78
5.2 4.7 3.1 3.8 1.5 4.5 4.7 3.1 2.5 1.4
2.7 2.8 3.8 2.9 3.1 2.8 2.0 4.9
5.9 3.3 2.1 1.4 <.50 3.0 <1.7 <1.8 3.4 7.5
73 23 11 25 6.0 12 8.4 7.3 20 35
6.1 7.3 1.9 4.0 1.1 2.2 1.4 3.2 7.8 4.5
3.2 1.8 7.0 1.6 .57 1.0 .87 <1.3 <.70 2.1
No data before 2012
2012 2014 2016 2018 2020
Year
-4 -2 0
Trend_color
Increasing
Decreasing
No change

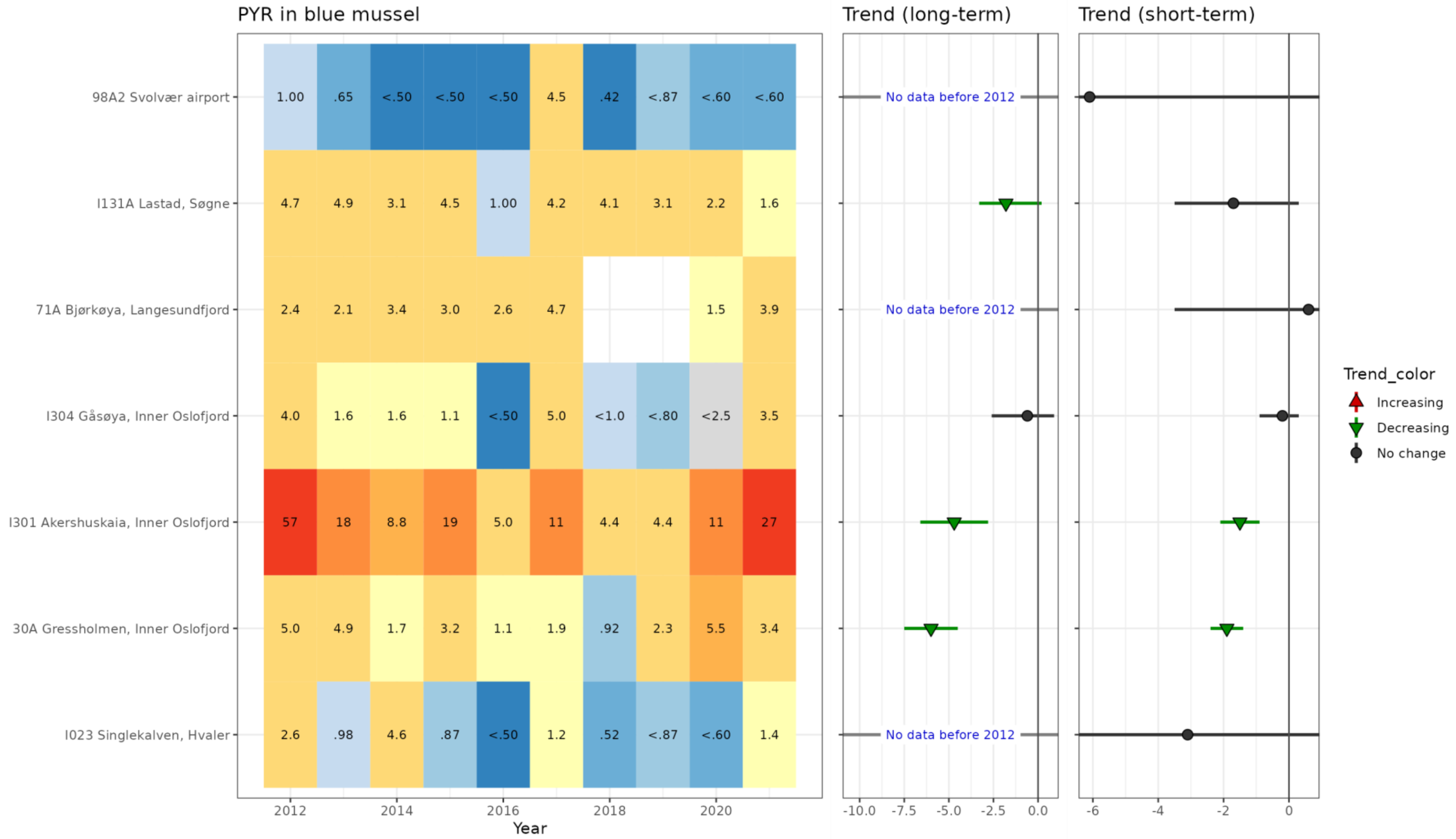


Figure 65. Heatmap and time trends of NAP, ANT, BAA, BAP, FLU and PYR in blue mussel. For full explanation of figure see example **Figure 23**.

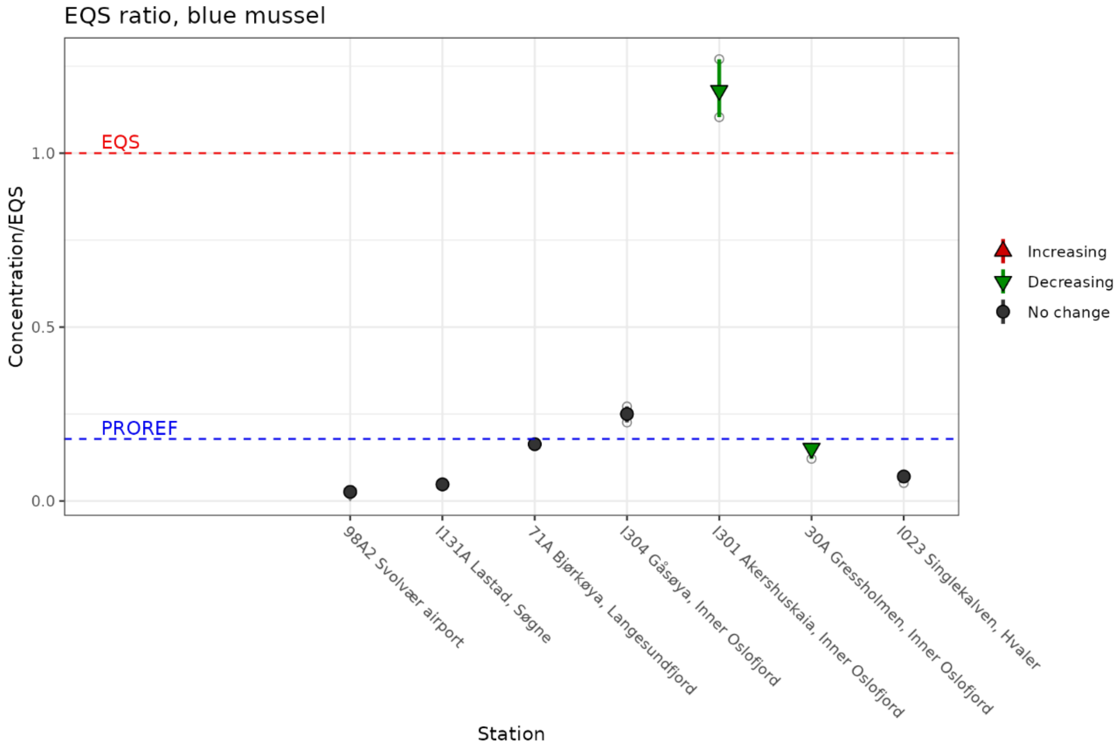


Figure 66. Ratio (concentration/EQS) for fluoranthene (FLU) in blue mussel. For full explanation of figure see example **Figure 24**.

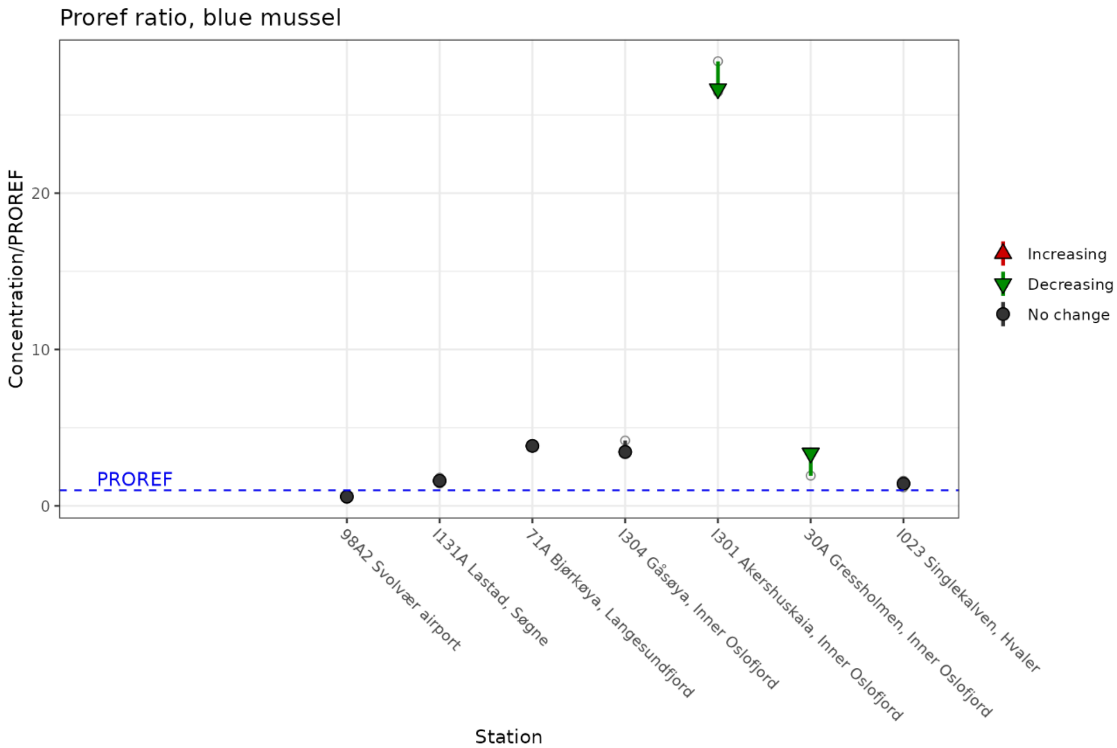


Figure 67. Ratio (concentration/PROREF) for pyrene (PYR) in blue mussel. For full explanation of figure see example **Figure 24**.

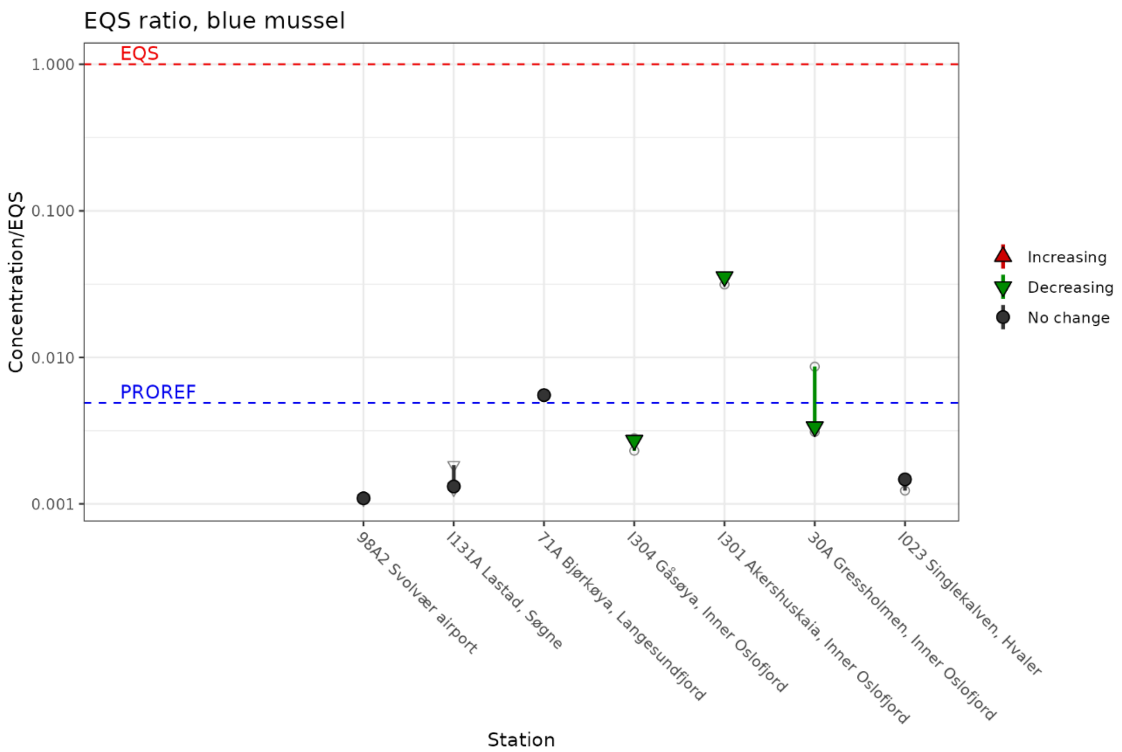


Figure 68. Ratio (concentration/EQS) for benzo(a)anthracene (BAA) in blue mussel. The y-axis is on a *natural log basis.* For full explanation of figure see example **Figure 24**.

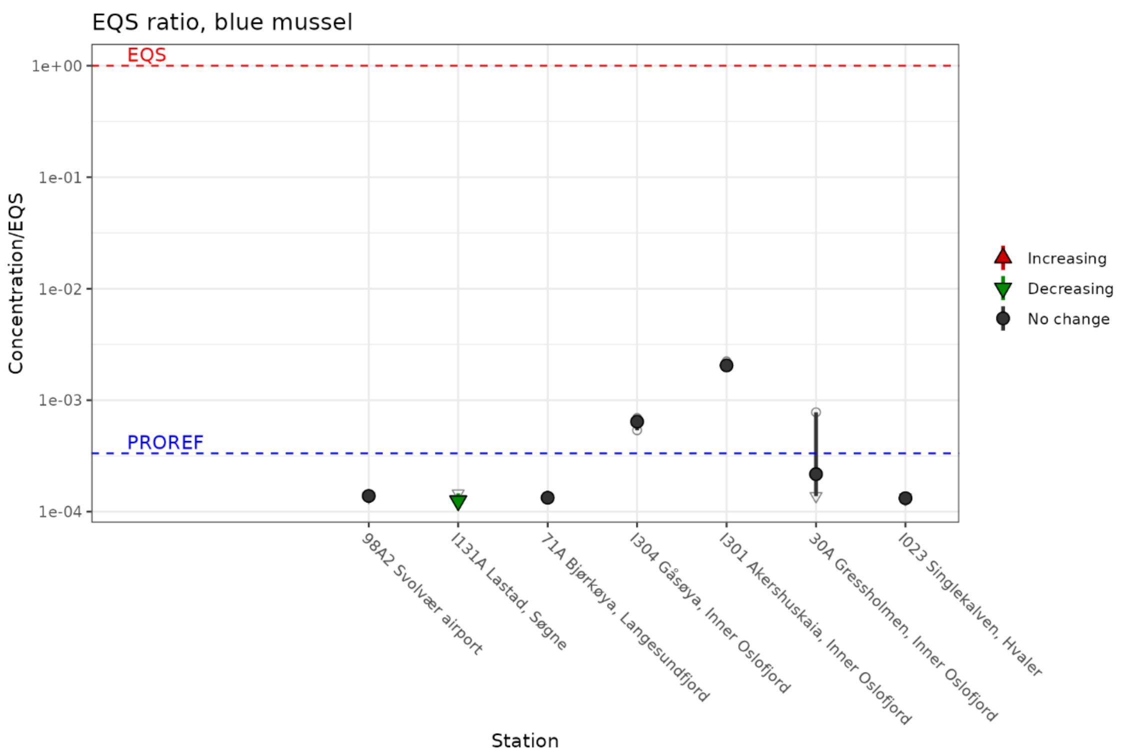


Figure 69. Ratio (concentration/EQS) anthracene (ANT) in blue mussel. The y-axis is on a *natural log basis.* For full explanation of figure see example **Figure 24**.

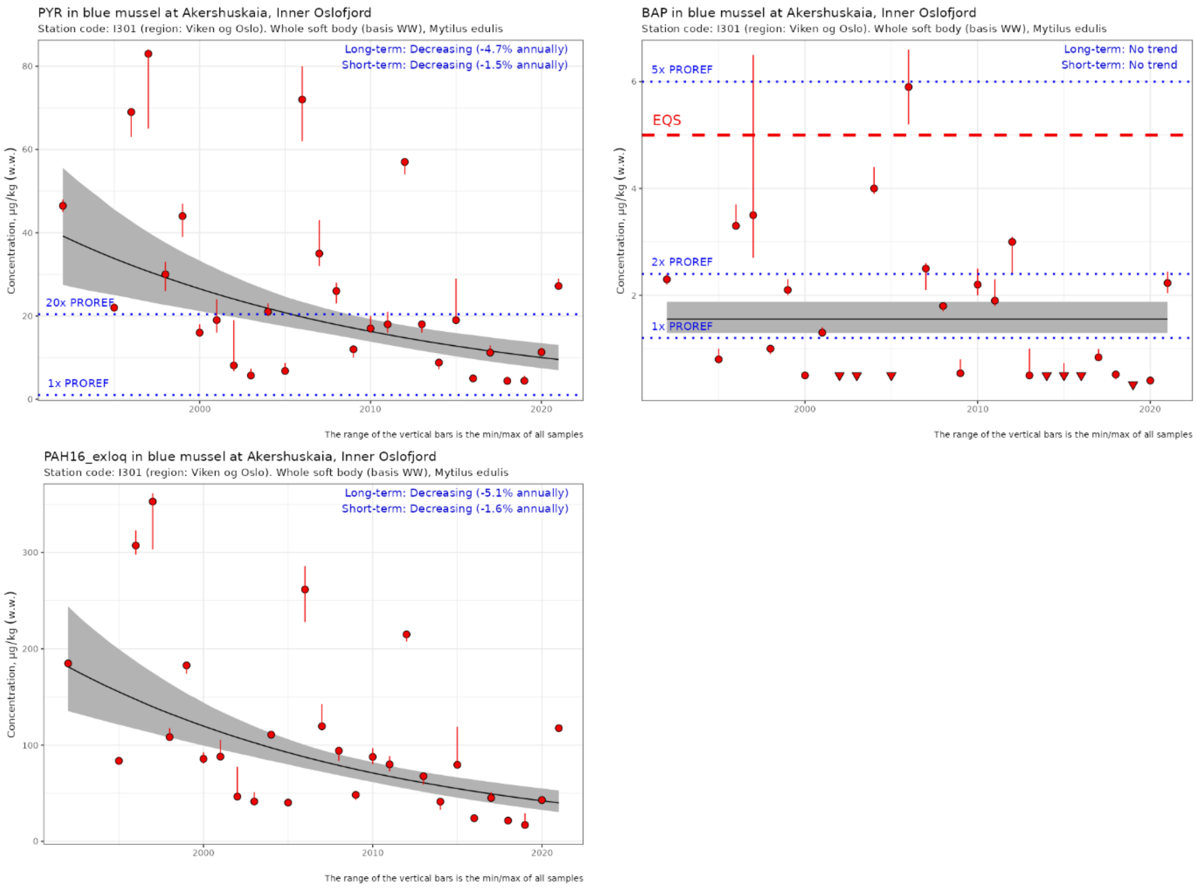


Figure 70. Three selected time trends for PAHs. For full explanation of figure see example **Figure 25**.

Selected time trends

Three selected time trends for pyrene (PYR), benzo(a)pyrene (BAP), and PAH16 in mussel at Akershuskaia in the Inner Oslofjord (I301) are shown in **Figure 70**. PYR and PAH16 have roughly the same trends, both long-term and short-term decreasing trends). In 1997, median concentration of PAH16 in blue mussel from Akershuskaia was 353 µg/kg ww. Since then, the concentrations have been decreasing. PAH16 in blue mussel from this station has a long-term decreasing trend (5,1% annually) and a short-term decreasing trend (1.6% annually).

It has been shown by Arp et al. (2011) that PYR is a good indicator for sumPAH16 concentrations in pore water. It seems that it also may work well for predicting sumPAH16 in blue mussel. PYR concentrations were previously many times exceeding PROREF (up to >80x) prior to 2000, it was 30x PROREF in 2021, and might decrease further.

BAP on the other hand had no trends neither long-term nor short-term.

3.2.2.4 PCBs

Polychlorinated biphenyls are a group of chlorinated organic compounds that previously had a broad industrial and commercial application. There are 209 different theoretical PCB analogues (congeners). It is estimated that 1300 tons of PCBs were used in products and buildings in Norway in 1980, and that 100 tons remains in products and buildings today[12]. In the present study, PCBs were analysed in blue mussel at 23 stations, in cod liver at 18 stations, and in eider blood and eggs at one station (**Table 1**). In the following, focus is on CB153, in addition to CB138 and CB118, in terms of PROREF and time trends, since they are major constituents of the sum of PCBs, and sumPCB7 (ΣPCB_7), and therefore appropriate proxies/ representatives. The EQS is given for sumPCB7, which is a river-basin specific pollutant. CB118 is the only mono-*ortho* Cl-substituted constituent of sumPCB7, and therefore the only with some dioxin-like toxicity in terms of toxic equivalency factors (TEF; Van den Berg et al. 2006).

EQS

SumPCB7 concentrations exceeded EQS (0.6 µg/kg ww, **Table 3**) at 16 mussel stations (*inter alia* in the Inner Oslofjord, Hvaler and Langesundfjord; **Figure 71** and Figure 72), as most previous years. All cod stations exceeded the EQS (**Figure 71** and **Figure 72**), as all previous years. Eggs from eider from Svalbard also contained concentrations of sumPCB7 above the EQS.

PROREF

Blue mussel at seven stations had concentrations of CB153 below PROREF. Six stations exceeded PROREF by 1-2 times, and six stations exceeded PROREF by 2-5 times (**Figure 73** and **Figure 74**). As in previous years, the highest exceedances (5-10 or 10-20 times) of PROREF were observed for mussels from Bergen (I241), Ålesund (28A2), and the Inner Oslofjord (I301 and 30A) (**Figure 73** and **Figure 74**).

Cod at 14 stations had concentrations of CB153 below PROREF. Three stations exceeded PROREF by 1-2 times (Langesundfjord (71B), Bergen (24B) and Trondheim (80B)), while cod from the Inner Oslofjord (30B) exceeded PROREF by 2-5 times (**Figure 73** and **Figure 74**).

CB138 and CB118 give the same overall picture, as CB153, in terms of which localities are more or less contaminated by PCBs (**Figure 73**-**Figure 76**).

Long-term trends

For mussels, there were nine stations showing decreasing long-term trends in CB153 concentrations (stations from the Oslofjord (36A, I304, I301, 30A, and 31A), Hvaler (I024 and I023), Langesundfjord (71A), and Bergen (I241)), five stations showing no trend, and two stations showing increasing long-term trends (Farsund (15A) and Varangerfjord (11X)).

Six cod stations showed decreasing long-term trends in CB153 concentration, four stations showed no trend, and no increasing long-term trends were observed.

Short-term trends

For mussels, there were 13 stations showing increasing short-term trends in CB153 concentrations, six stations showing no trend, and two stations showing decreasing short-term trends. At two stations, no trend-model could be established.

[12] https://miljostatus.miljodirektoratet.no/tema/miljogifter/prioriterte-miljogifter/polyklorerte-bifenyler-pcb/

Six cod stations showed decreasing short-term trends in CB153 concentrations, six stations showed no trend, and three stations showed increasing short-term trends. At one station, no trend-model could be established, and on two stations the time-series is too short to calculate trends.

CB138 and CB118 give the same overall picture, as CB153, in terms of long- and short-term trends (**Figure 73**). However, CB118 generally shown slightly more decreasing trends and slightly less increasing trends.

Some trends (CB-138 at Brashavn, 11X, Skallnes, 10A2, and Utne, 64A) are however very uncertain because of many non-detects and large variability, in combination with the nature of the trend-fitting (see chapter 4.6), and should thus be “taken with a pinch of salt”.

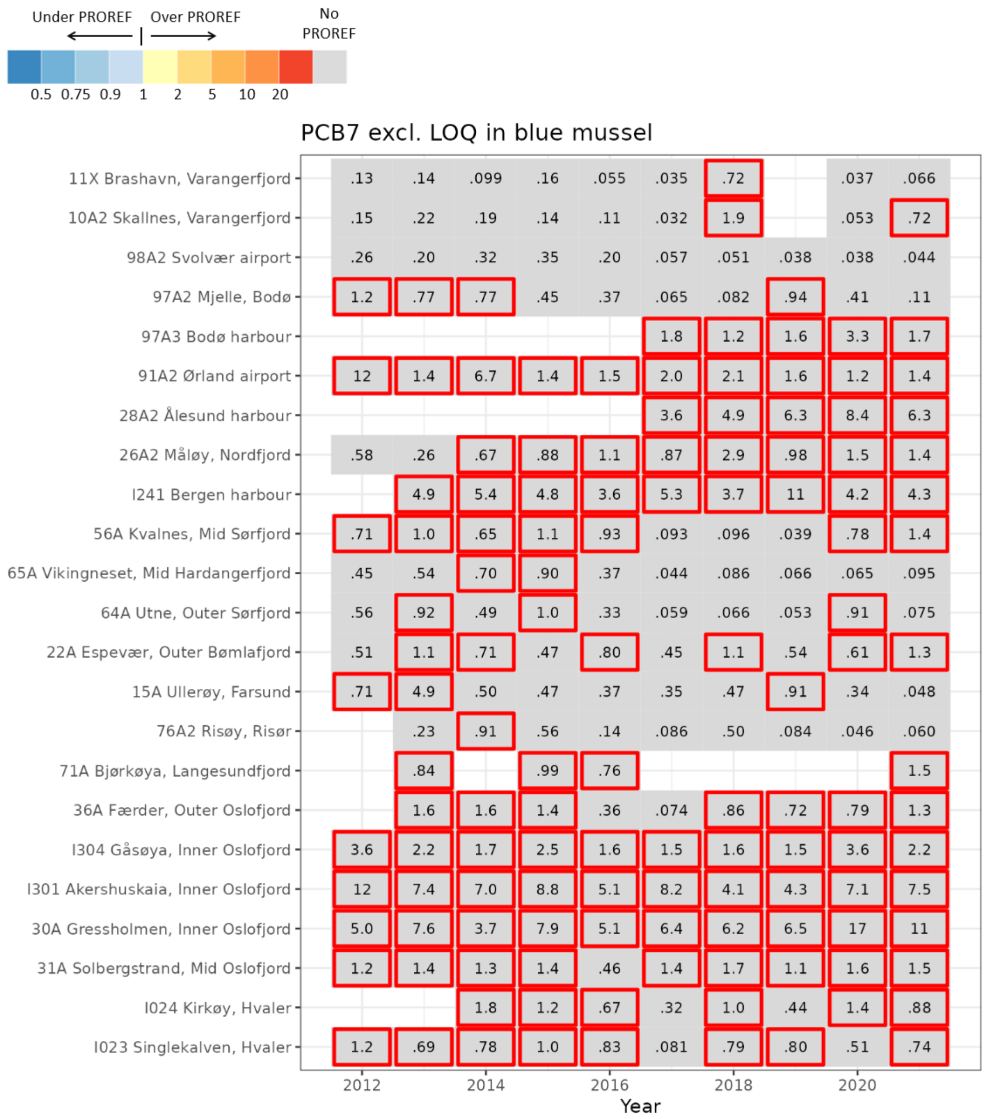
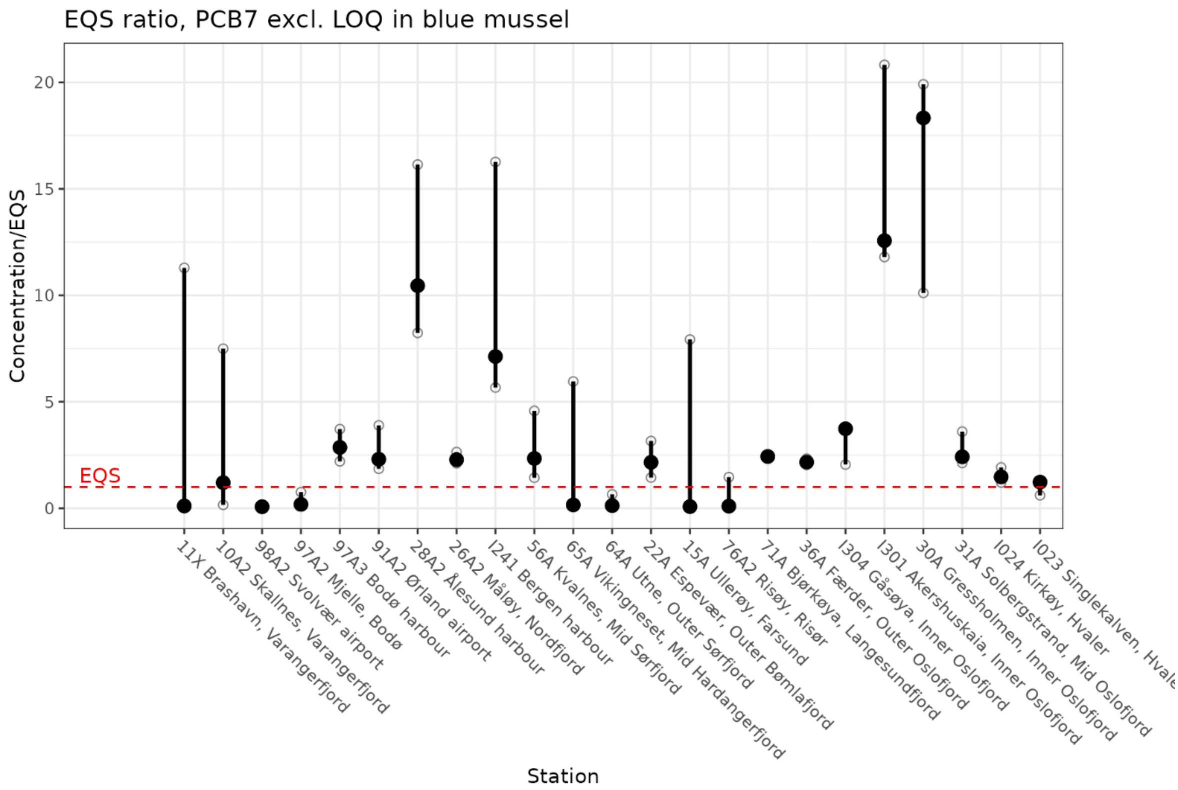

Under PROREF
Over PROREF
No PROREF
0.5 0.75 0.9 1 2 5 10 20
PCB7 excl. LOQ in blue mussel
11X Brashavn, Varangerfjord .13 .14 .099 .16 .055 .035 .72 .037 .066
10A2 Skallnes, Varangerfjord .15 .22 .19 .14 .11 .032 1.9 .053 .72
98A2 Svolvær airport .26 .20 .32 .35 .20 .057 .051 .038 .038 .044
97A2 Mjelle, Bodø 1.2 .77 .77 .45 .37 .065 .082 .94 .41 .11
97A3 Bodø harbour 1.8 1.2 1.6 3.3 1.7
91A2 Ørland airport 12 1.4 6.7 1.4 1.5 2.0 2.1 1.6 1.2 1.4
28A2 Ålesund harbour 3.6 4.9 6.3 8.4 6.3
26A2 Måløy, Nordfjord .58 .26 .67 .88 1.1 .87 2.9 .98 1.5 1.4
I241 Bergen harbour 4.9 5.4 4.8 3.6 5.3 3.7 11 4.2 4.3
56A Kvalnes, Mid Sørfjord .71 1.0 .65 1.1 .93 .093 .096 .039 .78 1.4
65A Vikingneset, Mid Hardangerfjord .45 .54 .70 .90 .37 .044 .086 .066 .065 .095
64A Utne, Outer Sørfjord .56 .92 .49 1.0 .33 .059 .066 .053 .91 .075
22A Espevær, Outer Bømlafjord .51 1.1 .71 .47 .80 .45 1.1 .54 .61 1.3
15A Ullerøy, Farsund .71 4.9 .50 .47 .37 .35 .47 .91 .34 .048
76A2 Risøy, Risør .23 .91 .56 .14 .086 .50 .084 .046 .060
71A Bjørkøya, Langesundfjord .84 .99 .76 1.5
36A Færder, Outer Oslofjord 1.6 1.6 1.4 .36 .074 .86 .72 .79 1.3
I304 Gåsøya, Inner Oslofjord 3.6 2.2 1.7 2.5 1.6 1.5 1.6 1.5 3.6 2.2
I301 Akershuskaia, Inner Oslofjord 12 7.4 7.0 8.8 5.1 8.2 4.1 4.3 7.1 7.5
30A Gressholmen, Inner Oslofjord 5.0 7.6 3.7 7.9 5.1 6.4 6.2 6.5 17 11
31A Solbergstrand, Mid Oslofjord 1.2 1.4 1.3 1.4 .46 1.4 1.7 1.1 1.6 1.5
I024 Kirkøy, Hvaler 1.8 1.2 .67 .32 1.0 .44 1.4 .88
I023 Singlekalven, Hvaler 1.2 .69 .78 1.0 .83 .081 .79 .80 .51 .74
2012 2014 2016 2018 2020
Year

Figure 71. Heatmap of sumPCB7 (non-detected congeners were assigned values of zero when calculating sum) in (upper panel) blue mussel and (lower panel) cod/eider. For full explanation of figure see example Figure 23.



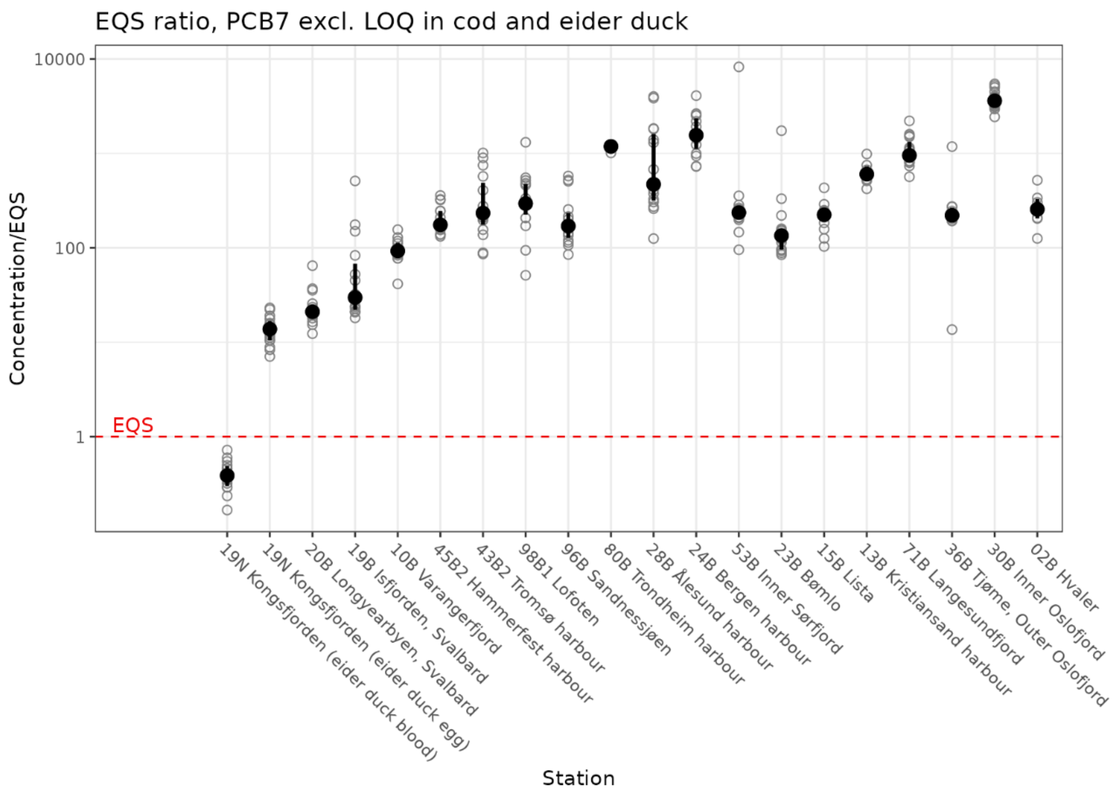
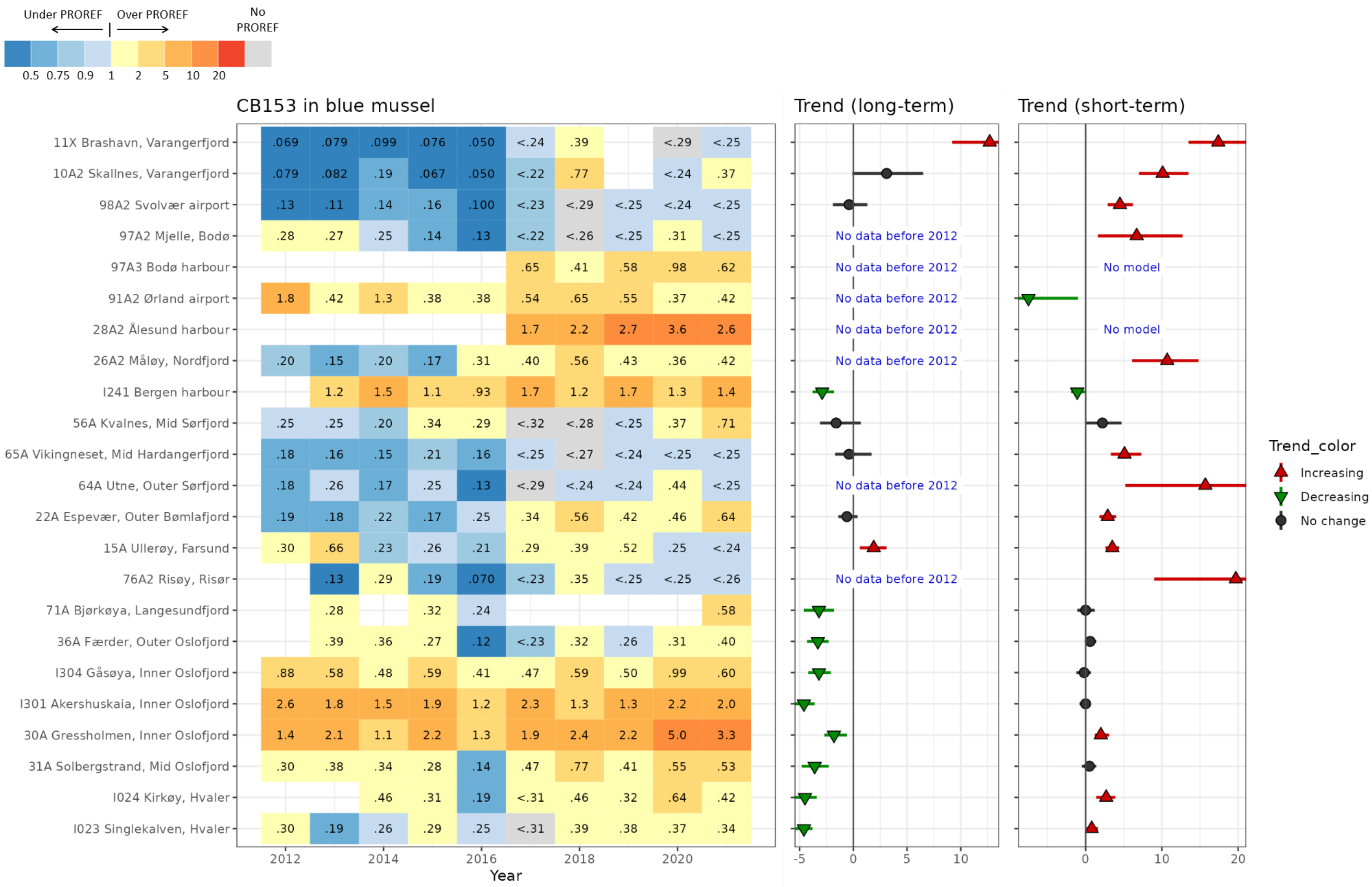
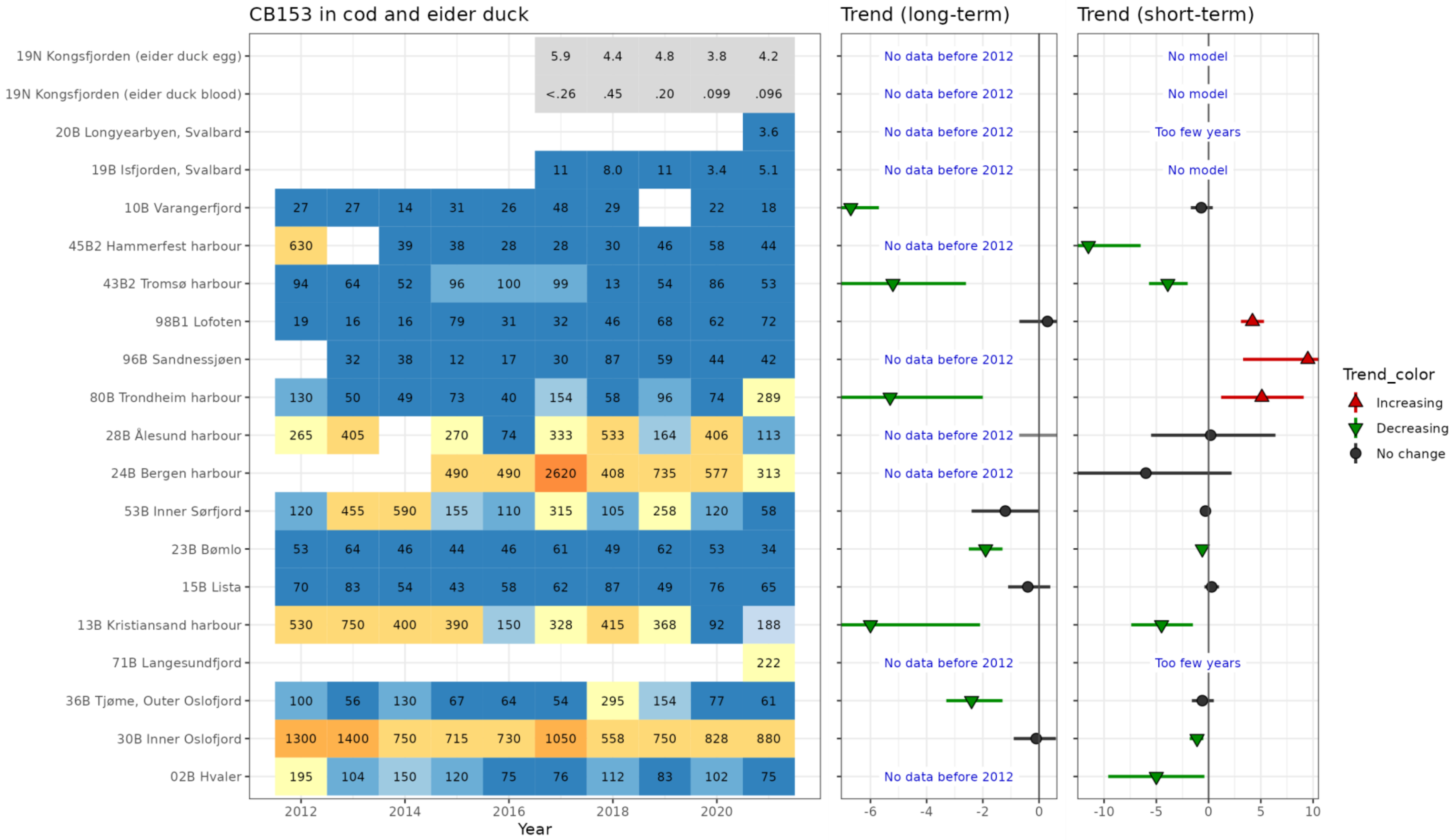
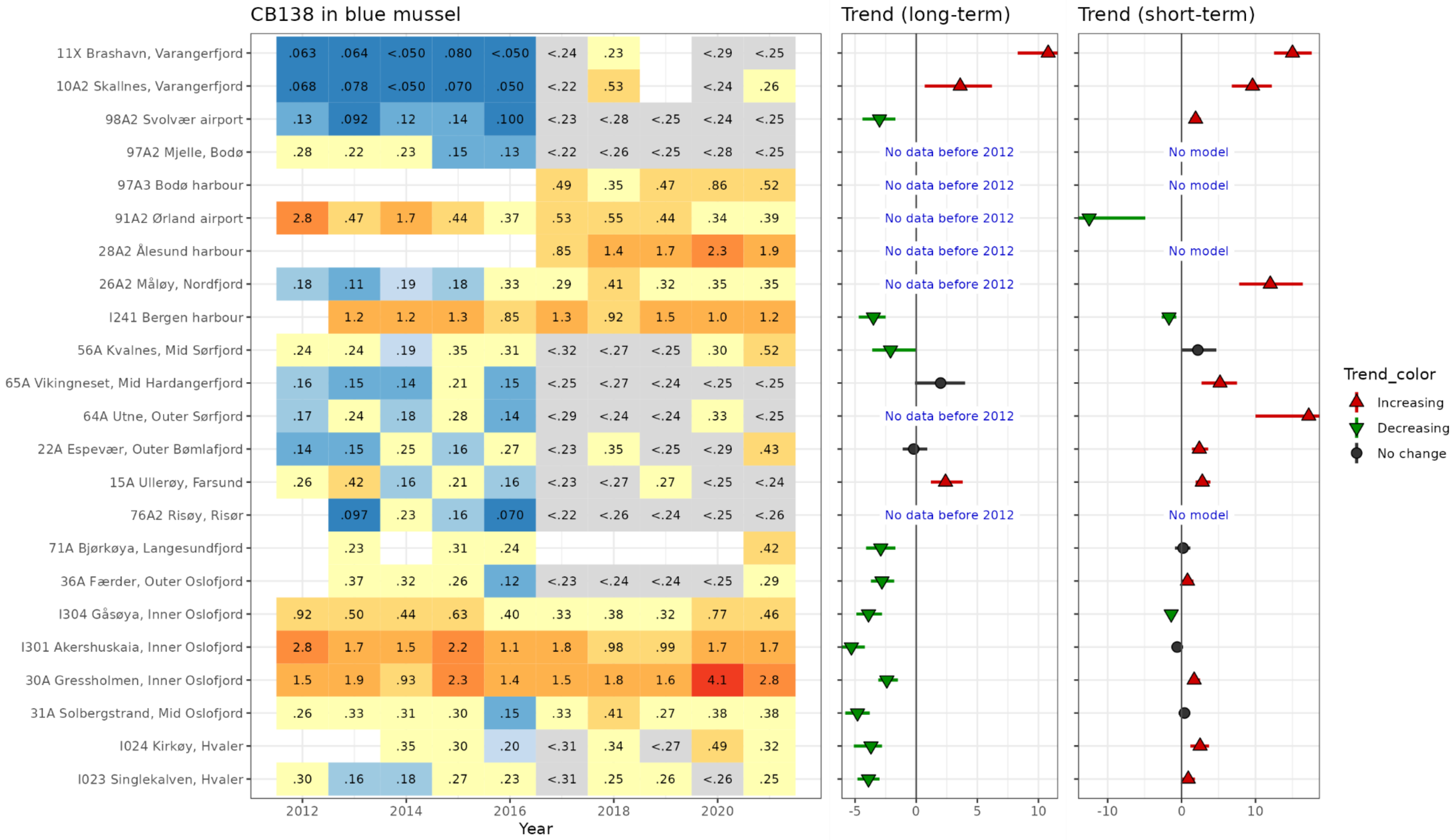


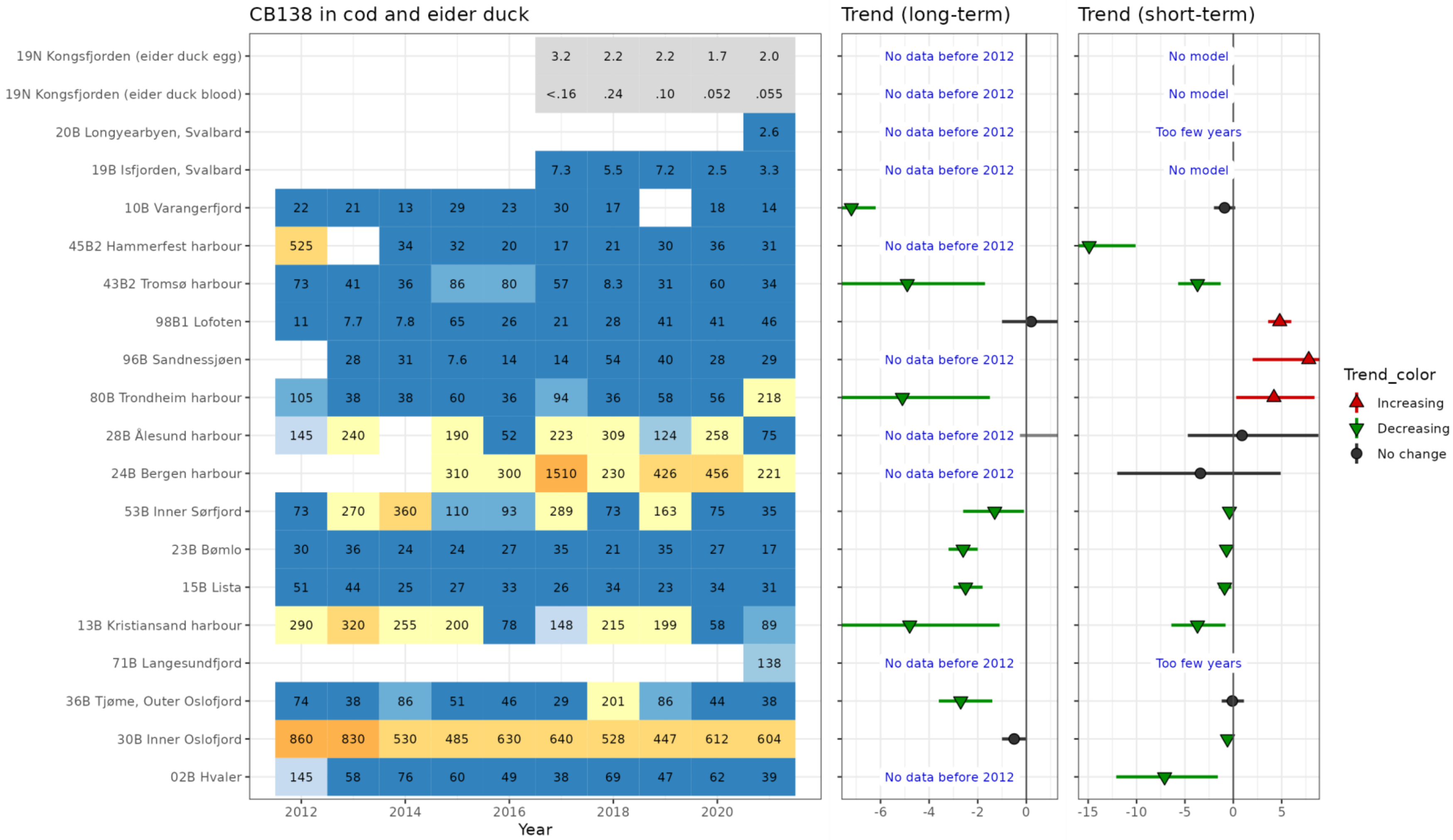
Figure 72. Ratio (concentrations/EQS) for sumPCB7 in (upper panel) blue mussel and (lower panel) cod and eider. *The y-axis for cod is on* log scale. For full explanation of figure see example Figure 24.

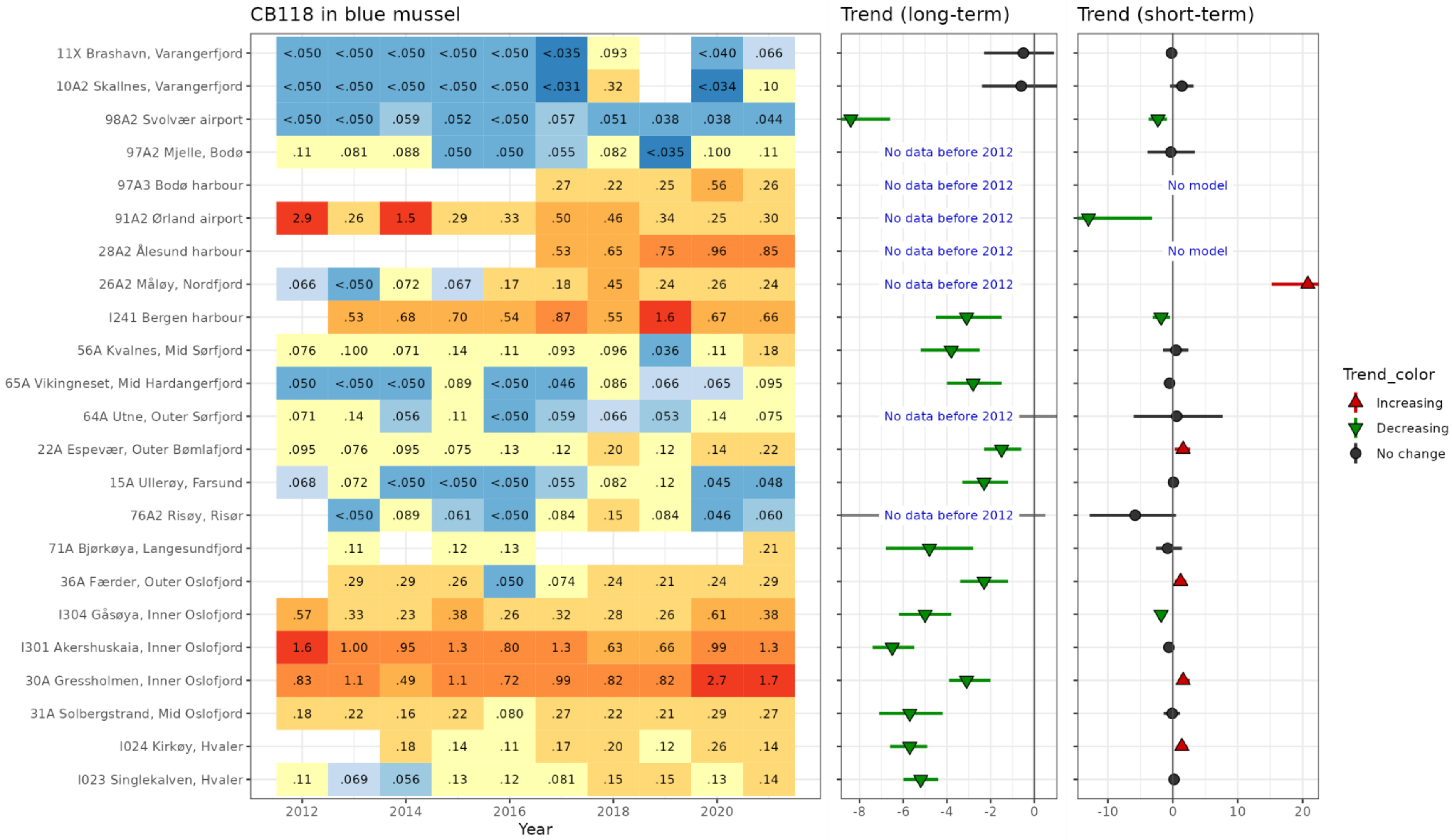


Under PROREF
Over PROREF
No PROREF
0.5 0.75 0.9 1 2 5 10 20
CB153 in blue mussel
Trend (long-term)
Trend (short-term)
11X Brashavn, Varangerfjord
10A2 Skallnes, Varangerfjord
98A2 Svolvær airport
97A2 Mjelle, Bodø
97A3 Bodø harbour
91A2 Ørland airport
28A2 Ålesund harbour
26A2 Måløy, Nordfjord
I241 Bergen harbour
56A Kvalnes, Mid Sørfjord
65A Vikingneset, Mid Hardangerfjord
64A Utne, Outer Sørfjord
22A Espevær, Outer Bømlafjord
15A Ulleroy, Farsund
76A2 Risøy, Risør
71A Bjørkøya, Langesundfjord
36A Færder, Outer Oslofjord
I304 Gåsøya, Inner Oslofjord
I301 Akershuskaia, Inner Oslofjord
30A Gressholmen, Inner Oslofjord
31A Solbergstrand, Mid Oslofjord
I024 Kirkøy, Hvaler
I023 Singlekalven, Hvaler
No data before 2012
No model
Year
2012 2014 2016 2018 2020
Trend_color
Increasing
Decreasing
No change


CB153 in cod and eider duck
Trend (long-term)
Trend (short-term)
19N Kongsfjorden (eider duck egg) 5.9 4.4 4.8 3.8 4.2
19N Kongsfjorden (eider duck blood) <.26 .45 .20 .099 .096
20B Longyearbyen, Svalbard 3.6
19B Isfjorden, Svalbard 11 8.0 11 3.4 5.1
10B Varangerfjord 27 27 14 31 26 48 29 22 18
45B2 Hammerfest harbour 630 39 38 28 28 30 46 58 44
43B2 Tromsø harbour 94 64 52 96 100 99 13 54 86 53
98B1 Lofoten 19 16 16 79 31 32 46 68 62 72
96B Sandnessjøen 32 38 12 17 30 87 59 44 42
80B Trondheim harbour 130 50 49 73 40 154 58 96 74 289
28B Ålesund harbour 265 405 270 74 333 533 164 406 113
24B Bergen harbour 490 490 2620 408 735 577 313
53B Inner Sørfjord 120 455 590 155 110 315 105 258 120 58
23B Bømlo 53 64 46 44 46 61 49 62 53 34
15B Lista 70 83 54 43 58 62 87 49 76 65
13B Kristiansand harbour 530 750 400 390 150 328 415 368 92 188
71B Langesundfjord 222
36B Tjøme, Outer Oslofjord 100 56 130 67 64 54 295 154 77 61
30B Inner Oslofjord 1300 1400 750 715 730 1050 558 750 828 880
02B Hvaler 195 104 150 120 75 76 112 83 102 75
2012 2014 2016 2018 2020
Year
No data before 2012
No model
Too few years
-6 -4 -2 0
-10 -5 0 5 10
Trend_color
Increasing
Decreasing
No change


CB138 in blue mussel
Trend (long-term)
Trend (short-term)
11X Brashavn, Varangerfjord
10A2 Skallnes, Varangerfjord
98A2 Svolvær airport
97A2 Mjelle, Bodø
97A3 Bodø harbour
91A2 Ørland airport
28A2 Ålesund harbour
26A2 Måløy, Nordfjord
I241 Bergen harbour
56A Kvalnes, Mid Sørfjord
65A Vikingneset, Mid Hardangerfjord
64A Utne, Outer Sørfjord
22A Espevær, Outer Bømlafjord
15A Ulleroøy, Farsund
76A2 Risøy, Risør
71A Bjørkøya, Langesundfjord
36A Færder, Outer Oslofjord
I304 Gåsøya, Inner Oslofjord
I301 Akershuskaia, Inner Oslofjord
30A Gressholmen, Inner Oslofjord
31A Solbergstrand, Mid Oslofjord
I024 Kirkøy, Hvaler
I023 Singlekalven, Hvaler
No data before 2012
No model
2012
2014
2016
2018
2020
Year
-5
0
5
10
-10
Trend_color
Increasing
Decreasing
No change


CB138 in cod and eider duck
Trend (long-term)
Trend (short-term)
19N Kongsfjorden (eider duck egg)
19N Kongsfjorden (eider duck blood)
20B Longyearbyen, Svalbard
19B Isfjorden, Svalbard
10B Varangerfjord
45B2 Hammerfest harbour
43B2 Tromsø harbour
98B1 Lofoten
96B Sandnessjøen
80B Trondheim harbour
28B Ålesund harbour
24B Bergen harbour
53B Inner Sørfjord
23B Bømlo
15B Lista
13B Kristiansand harbour
71B Langesundfjord
36B Tjøme, Outer Oslofjord
30B Inner Oslofjord
02B Hvaler
Year
No data before 2012
No model
Too few years
Trend_color
Increasing
Decreasing
No change


CB118 in blue mussel
Trend (long-term)
Trend (short-term)
Year
No data before 2012
No model
Trend_color
Increasing
Decreasing
No change

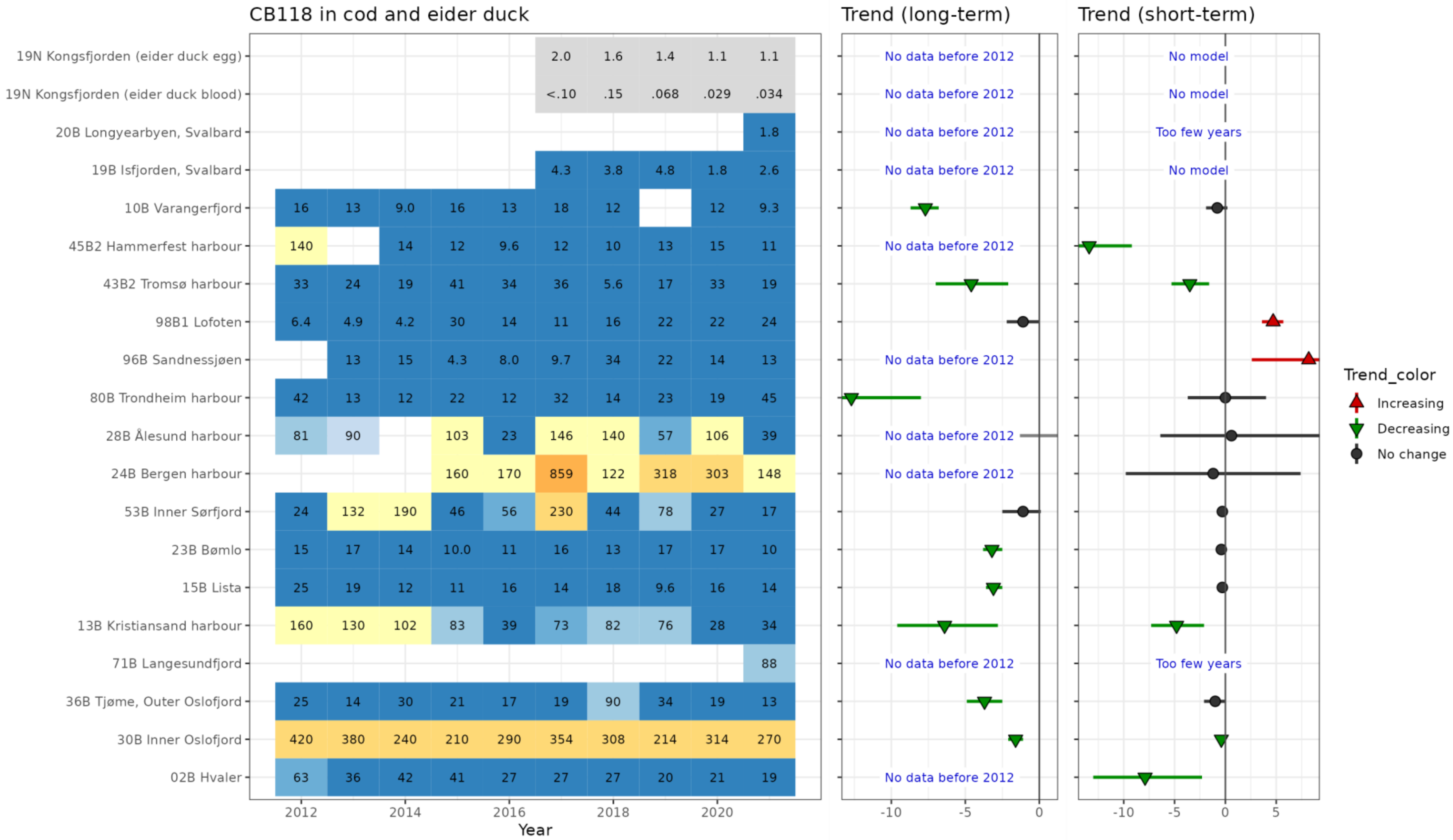
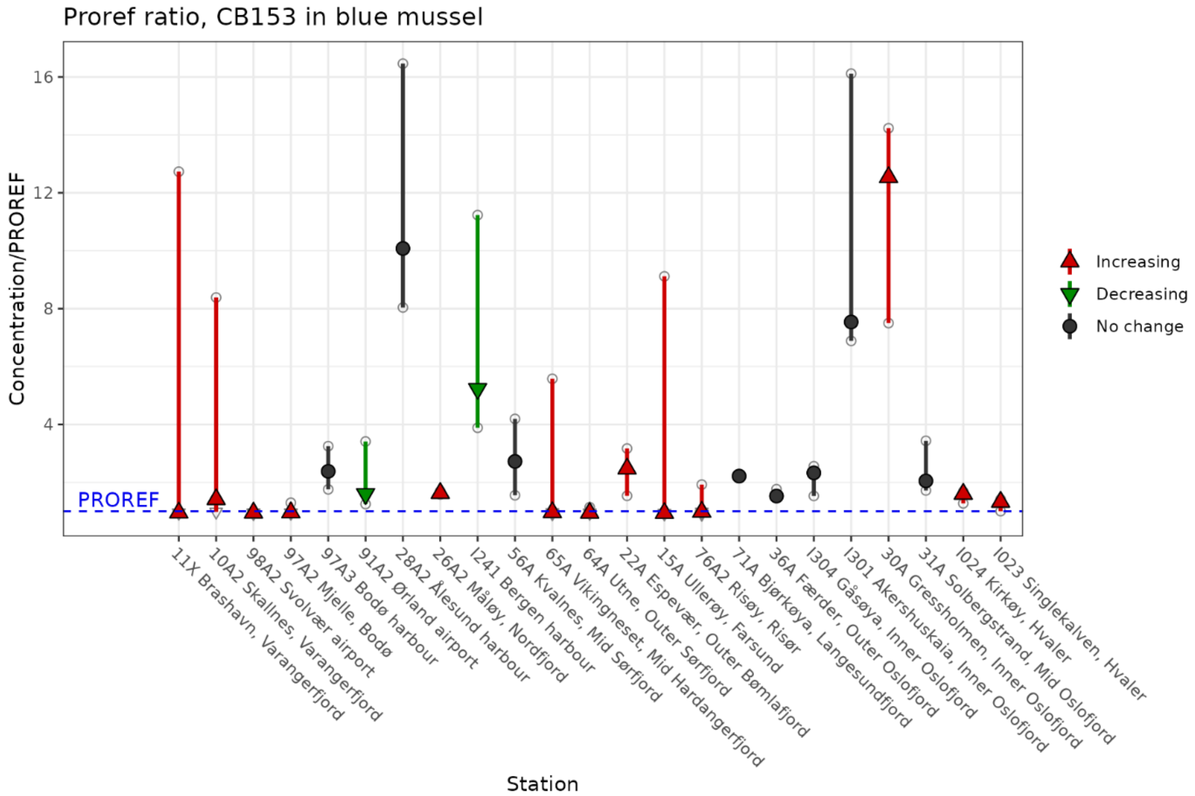


Figure 73. Heatmap and time trends of CB153, CB138 and CB118 in blue mussel, and cod and eider. For full explanation of figure see example Figure 23.



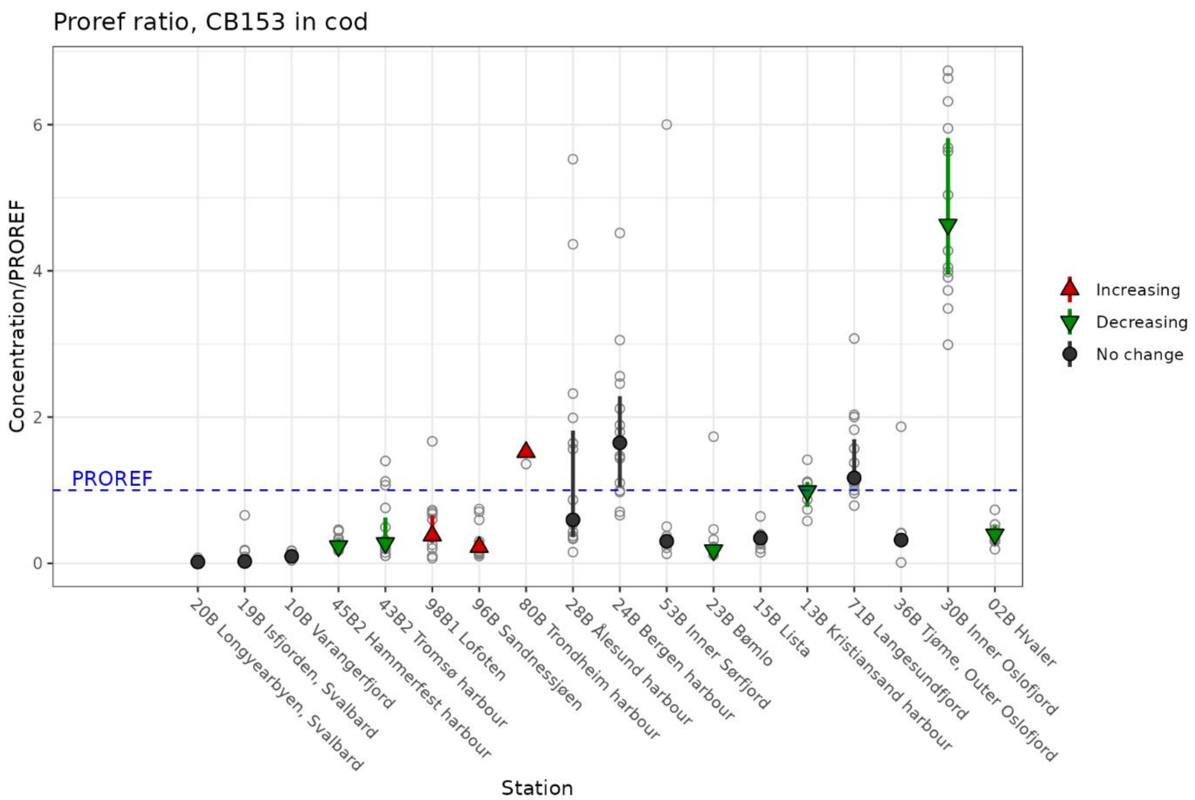
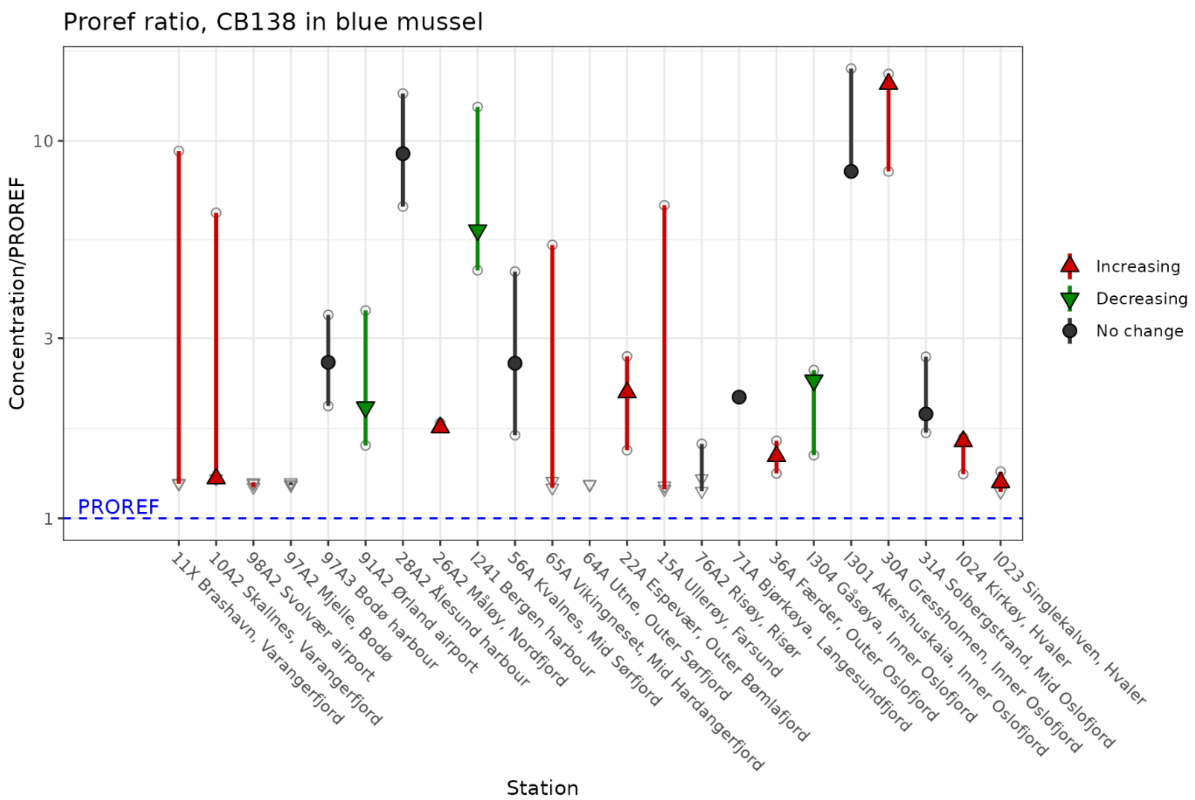


Figure 74. Ratio (concentrations/PROREF) for CB153 in (upper panel) blue mussel and (lower panel) cod. For full explanation of figure see example Figure 24.



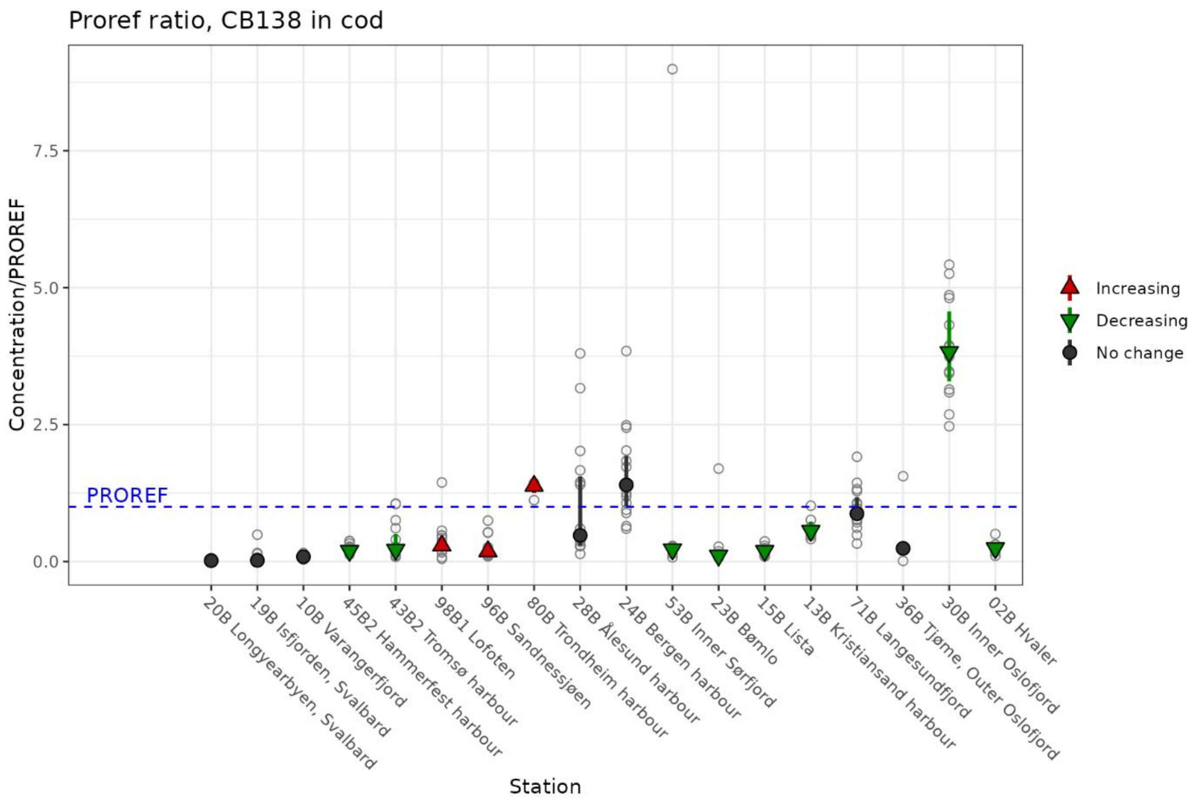
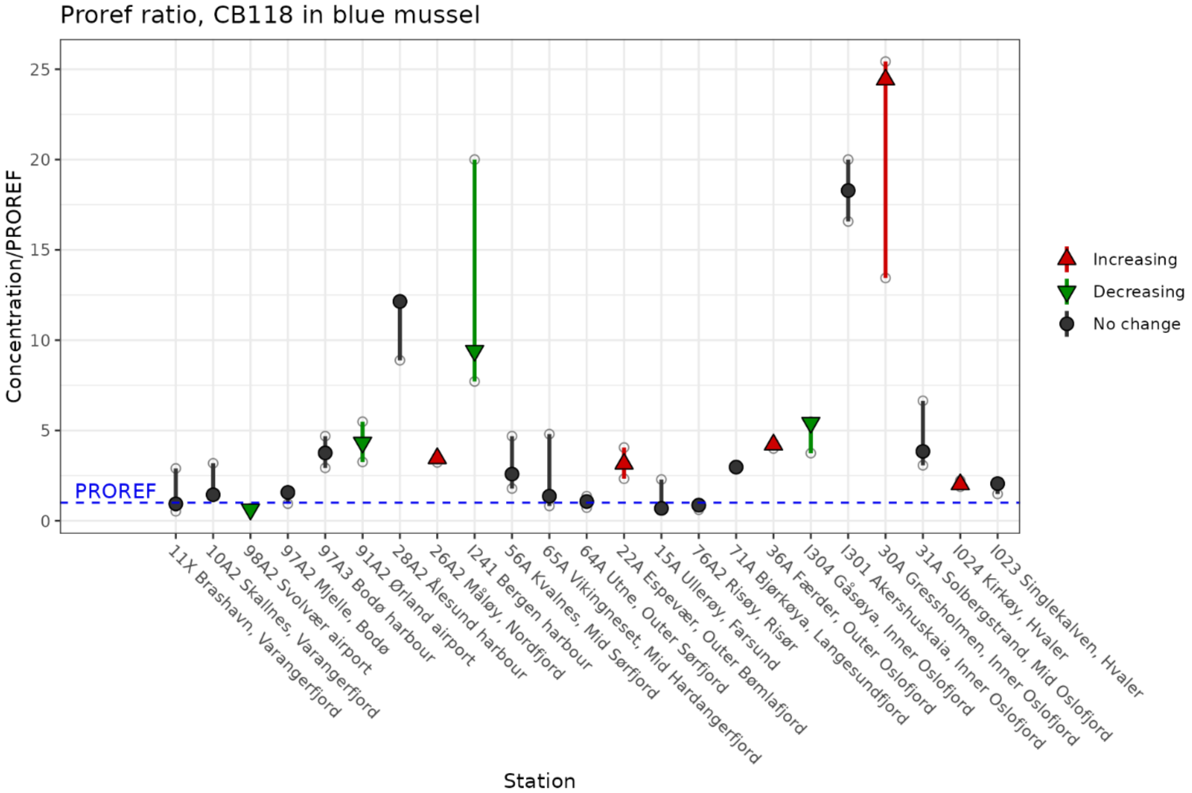


Figure 75. Ratio (concentrations/PROREF) for CB138 in (upper panel) blue mussel and (lower panel) cod. For full explanation of figure see example **Figure 24**.



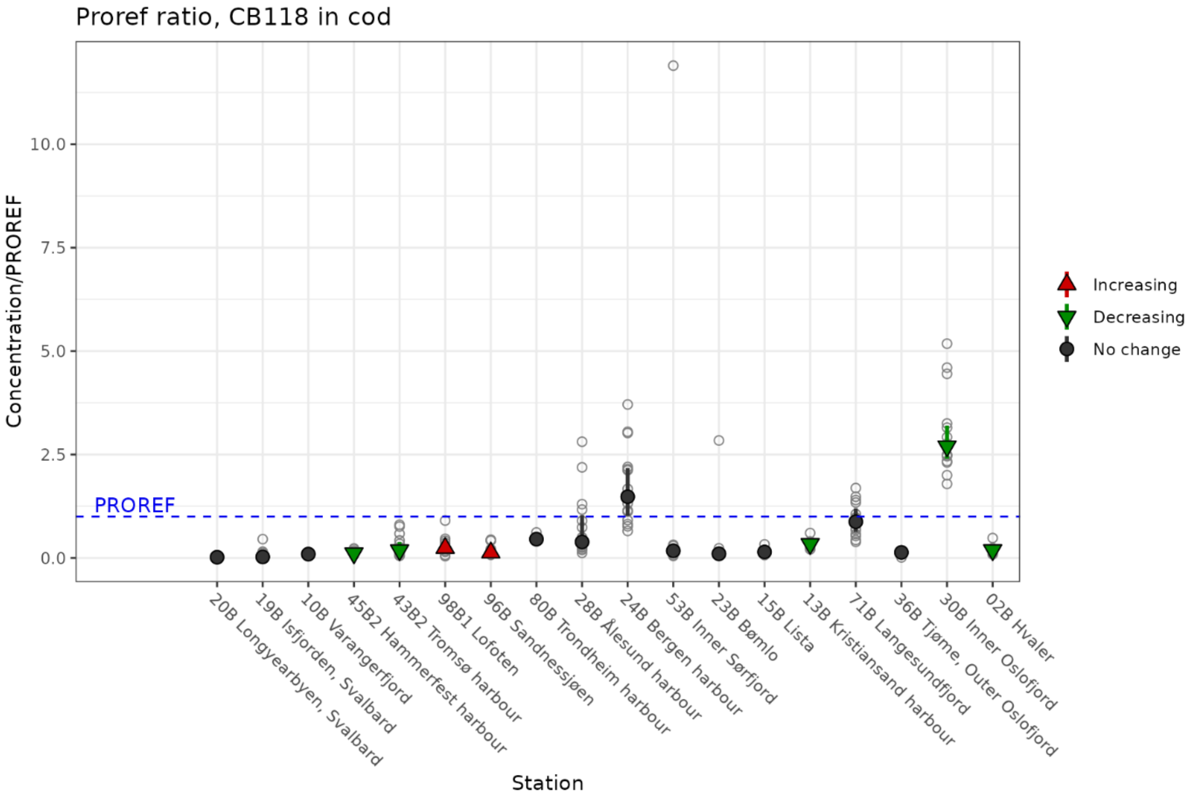


Figure 76. Ratio (concentrations/PROREF) for CB118 in (upper panel) blue mussel and (lower panel) cod. For full explanation of figure see example **Figure 24**.

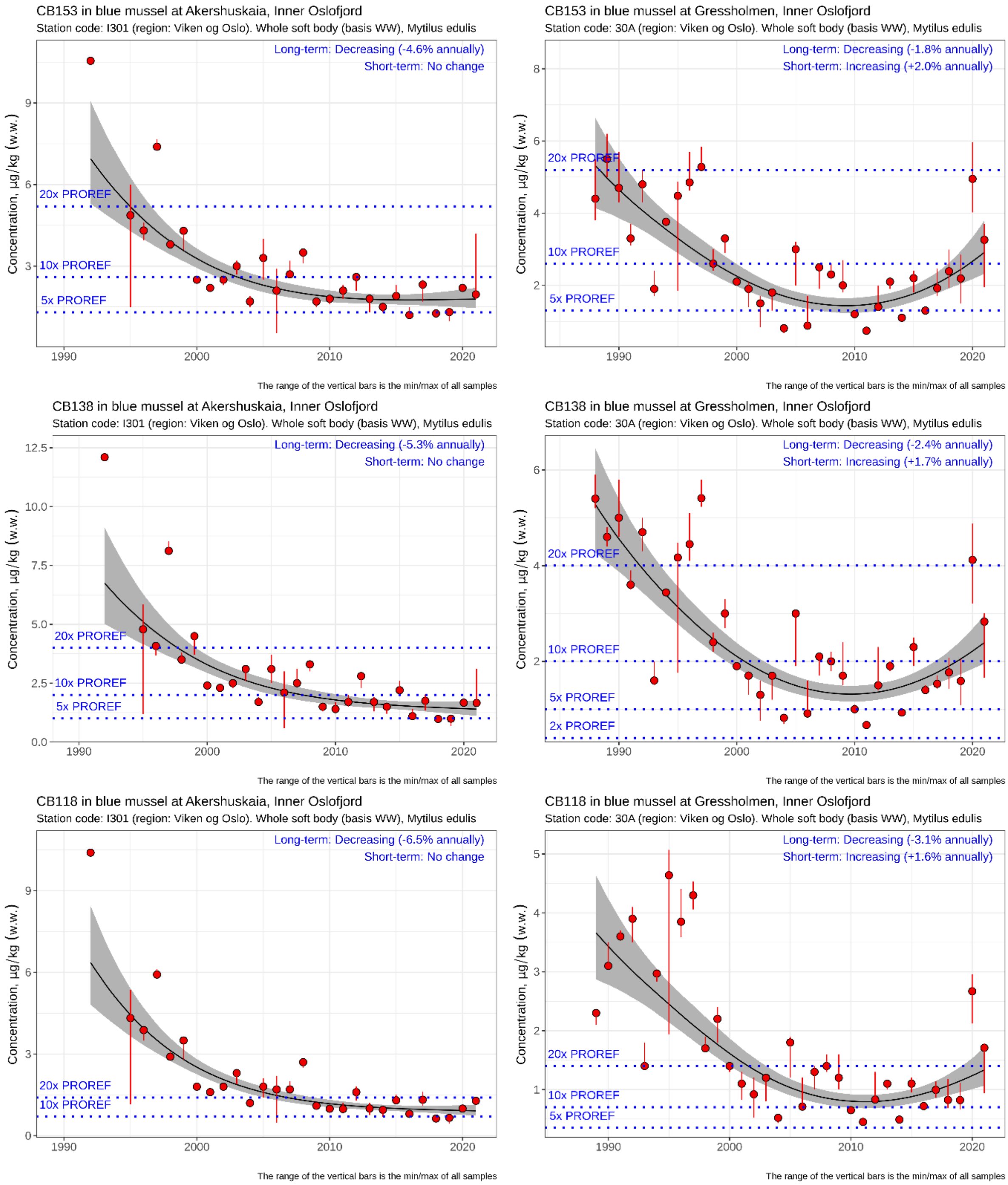


Figure 77. Six selected time trends for PCBs in blue mussel: CB153 (top), CB138 (middle) and CB118 (bottom) from the Inner Oslofjord (Akershuskaia, left, and Gressholmen, right). For full explanation of figure see example Figure 25.

Selected time trends

Six selected time trends for blue mussel are depicted in **Figure 77**, all from the Inner Oslofjord. CB153, CB138, and CB118 all show decreasing long-term trends at both stations. At Akershuskaia (I301) no short-term trends in either of the PCB congeners could be observed, while at Gressholmen (30A), there are increasing short-term trends. Cod from the inner Oslofjord (30B) shows downward short-term trends for the same three PCB congeners, as well as long-term trend for CB118 (**Figure 78**).

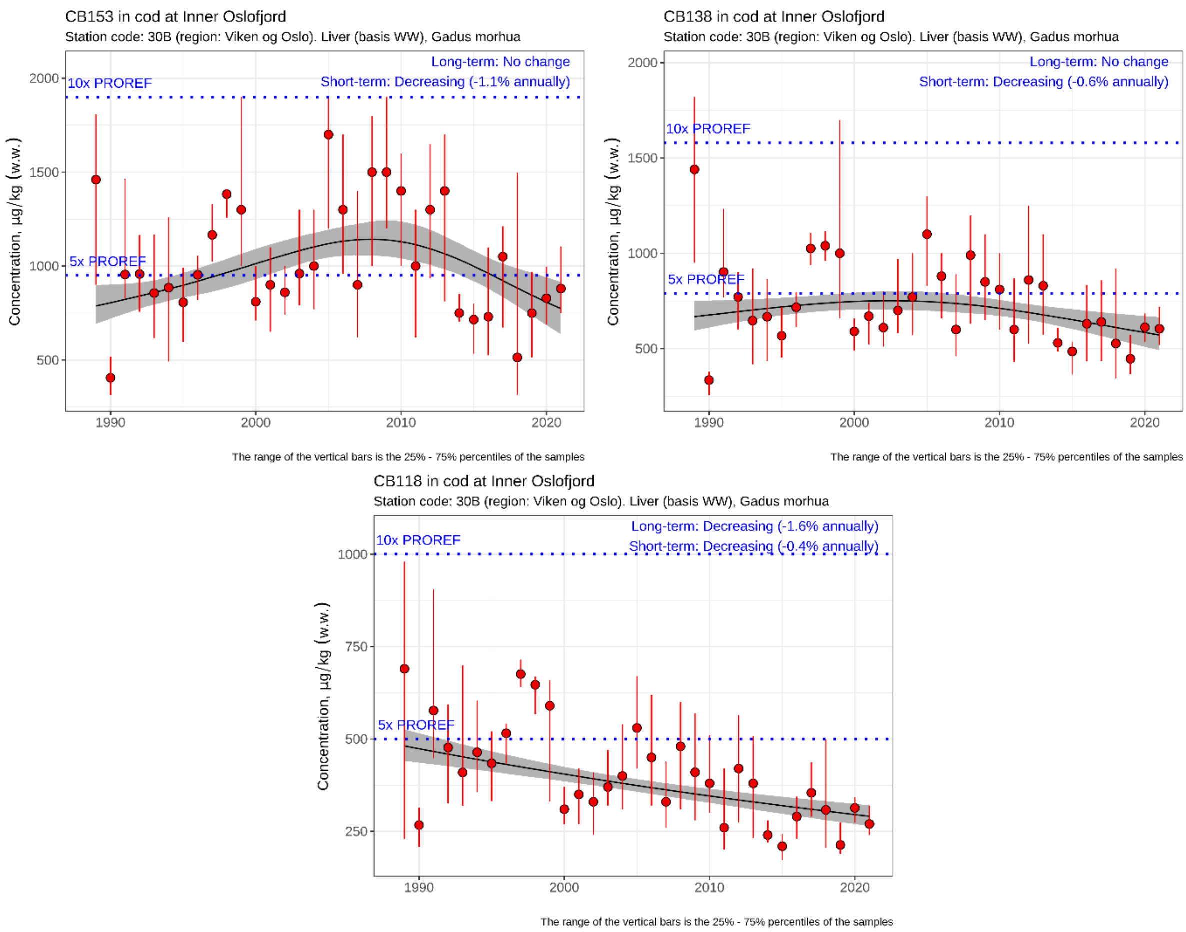


Figure 78. Time trends for PCBs in cod liver from the Inner Oslofjord (30B): CB153 (top left), CB138 (top right) and CB118 (bottom). For full explanation of figure see example Figure 25.

3.2.2.5 Siloxanes (D4, D5 and D6)

Siloxanes are chemical compounds consisting of silicon and oxygen substituted with various organic side chains, and they exist both as linear and cyclic substances. Siloxanes are chemicals used as synthetic intermediates in silicone polymer productions and are ingredients in e.g. cosmetic and personal care products. Siloxanes have properties that affect the consistency of personal care products such as deodorants, skin, and hair products to facilitate their use. The chemicals are also used in mechanical fluids and lubricants, biomedical products, cleaning and surface treatment agents, paint, insulation materials, and cement. Since 1. February 2020, there are restrictions for D4 and D5 for wash-off cosmetic products in concentrations above 0.1%[13] in the EU/EEA. Siloxanes, i.e. the cyclic volatile methyl siloxanes (cVMS) octamethylcyclotetrasiloxane (D4), decamethylcyclopentasiloxane (D5), and dodecamethylcyclohexasiloxane (D6) were analysed in cod liver at 13 stations and in eider blood and eggs at one station in Kongsfjorden at Svalbard (19N).

EQS

When applying the EQS for D5 (15 217 µg/kg ww, **Table 3**) in biota, the concentrations were below EQS in cod liver (**Figure 9**) and in eider blood and eggs (**Figure 10**; **Figure 79**). **Figure 80** also shows that all median concentrations in cod and eider were below EQS.

In cod liver, D5 was the most dominant cVMS in the Inner Oslofjord (30B) (741 µg/kg ww). Median D5-concentrations in cod liver were lowest at Tjøme in the Outer Oslofjord (1.8 µg/kg ww). In eider, D5 was the most dominant cVMS in eggs in Kongsfjorden at Svalbard (19N) (1.8 µg/kg ww), while D6 was the most dominant in blood (0.60 µg/kg ww).

PROREF

No PROREF for siloxanes are calculated for neither cod nor eider.

Time trends

No trends were found for D4, D5 or D6 in cod at the two stations in the Inner Oslofjord (30B) and Bergen harbour (24B) where short-term trends could be calculated (**Figure 79**; **Figure 81**).

[13] https://www.miljodirektoratet.no/aktuelt/nyheter/2020/februar-2020/nytt-forbud-mot-bruk-av-miljogifter/

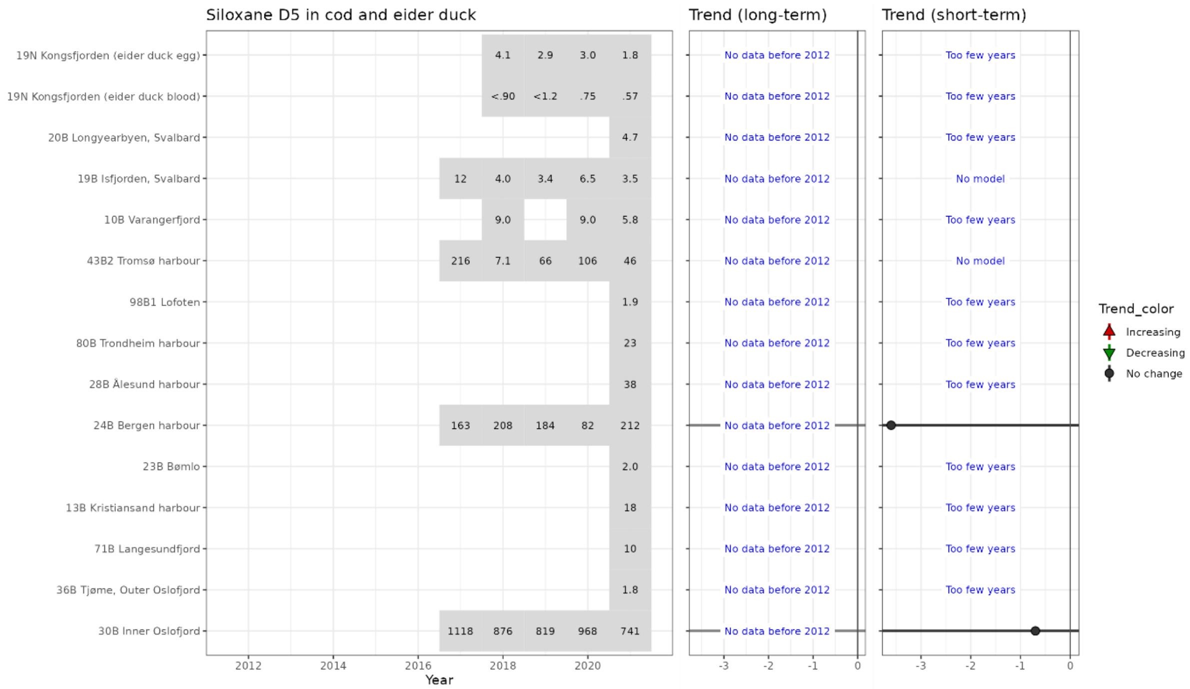


Figure 79. Heatmap and time trends of D5 in eider and cod. For full explanation of figure see example **Figure 23**.

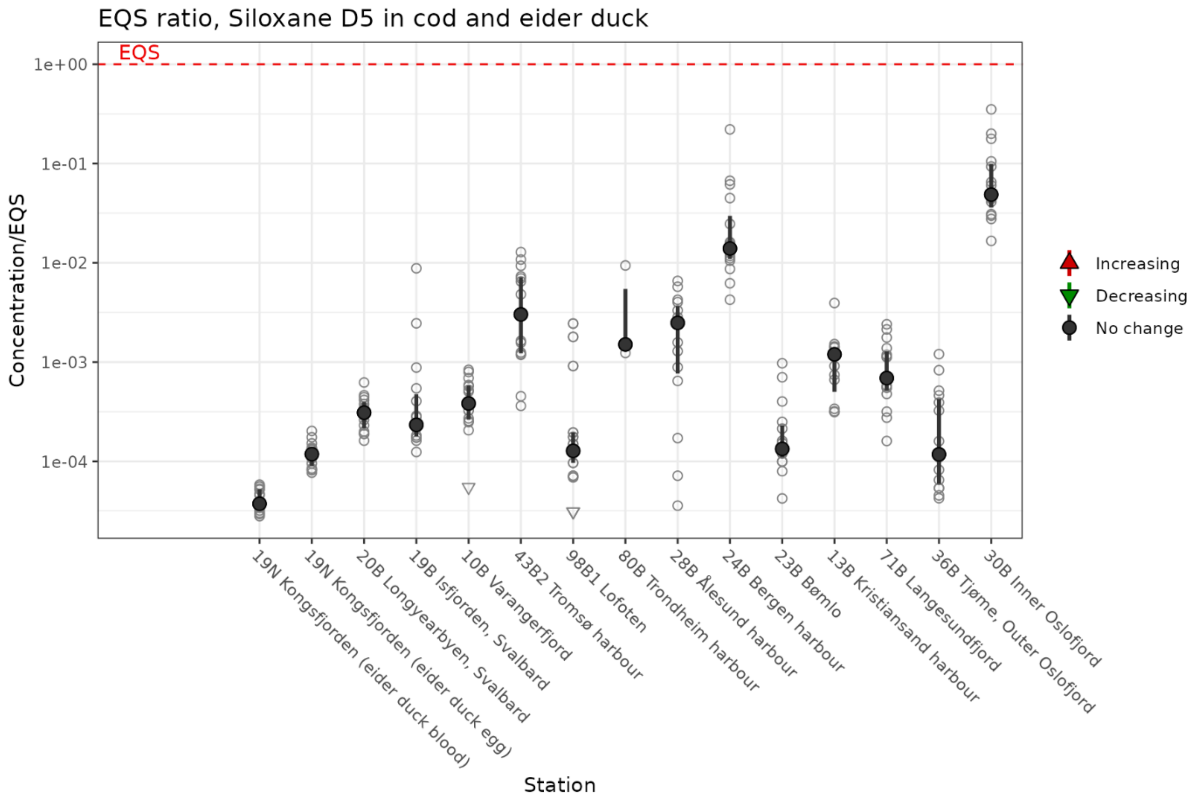


Figure 80. Ratio (concentrations/EQS) for D5 in cod and eider. EQS applies both to cod and eider. For full explanation of figure see example Figure 24**.**

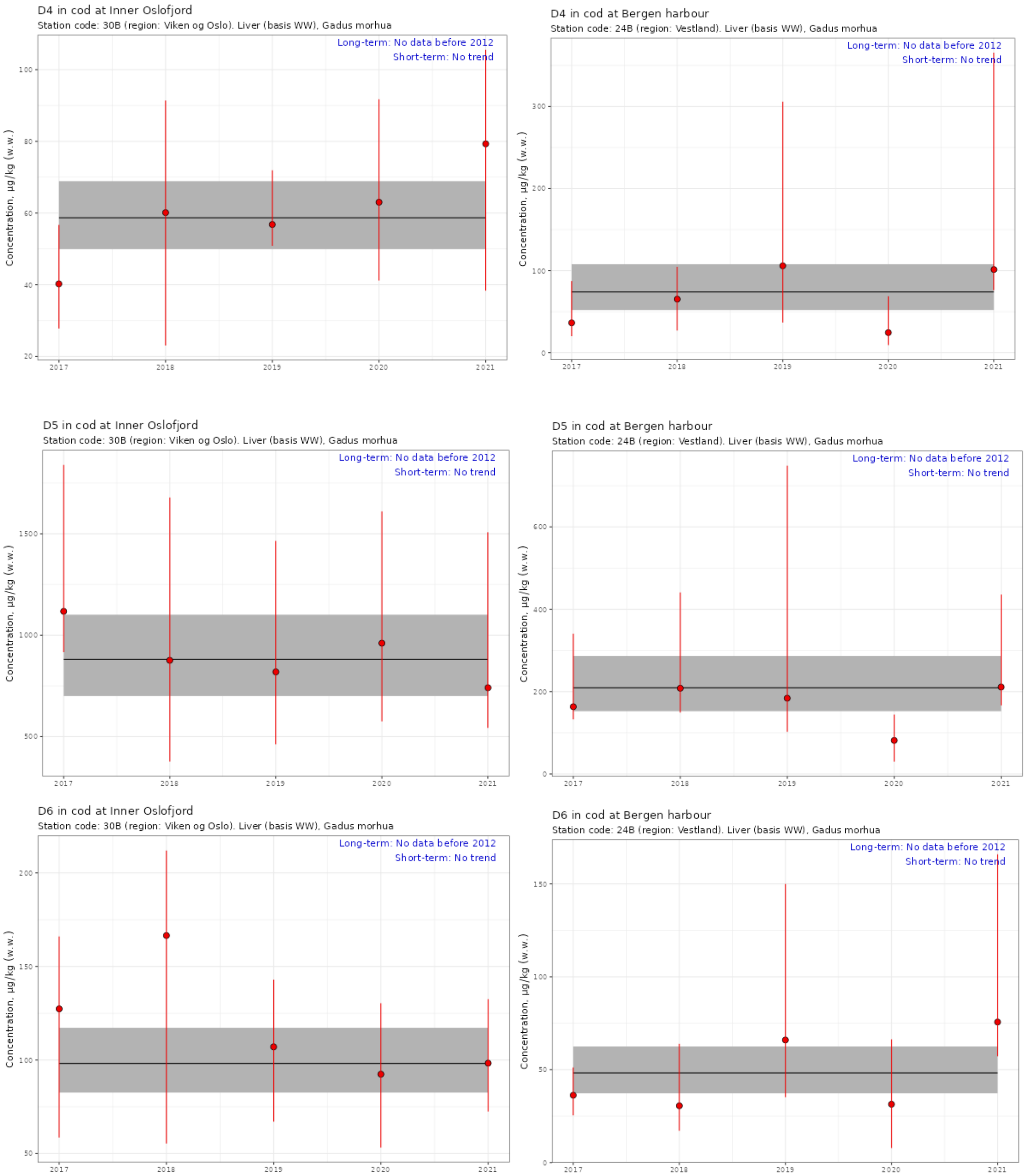


Figure 81. D4, D5 and D6 in cod from the Inner Oslofjord (30B) and Bergen harbour (24B). For full explanation of figure see example **Figure 25**.

3.2.2.6 Chlorinated paraffins (SCCPs and MCCPs)

Chlorinated paraffins are complex mixtures of polychlorinated organic compounds. They are mainly used in metal working fluids, sealants, as flame-retardants in rubbers and textiles, in leather processing and in paints and coatings. Their persistence, bioaccumulation, potential for long-ranged environmental transport and toxicity imply that they may have harmful environmental effects at a global level. A global regulation of SCCP has been in place since the end of 2019 through the Stockholm Convention. In 2020, a proposal was made by the UK to list MCCP as a persistent organic pollutant in Annex A, B or C to the Stockholm Convention.

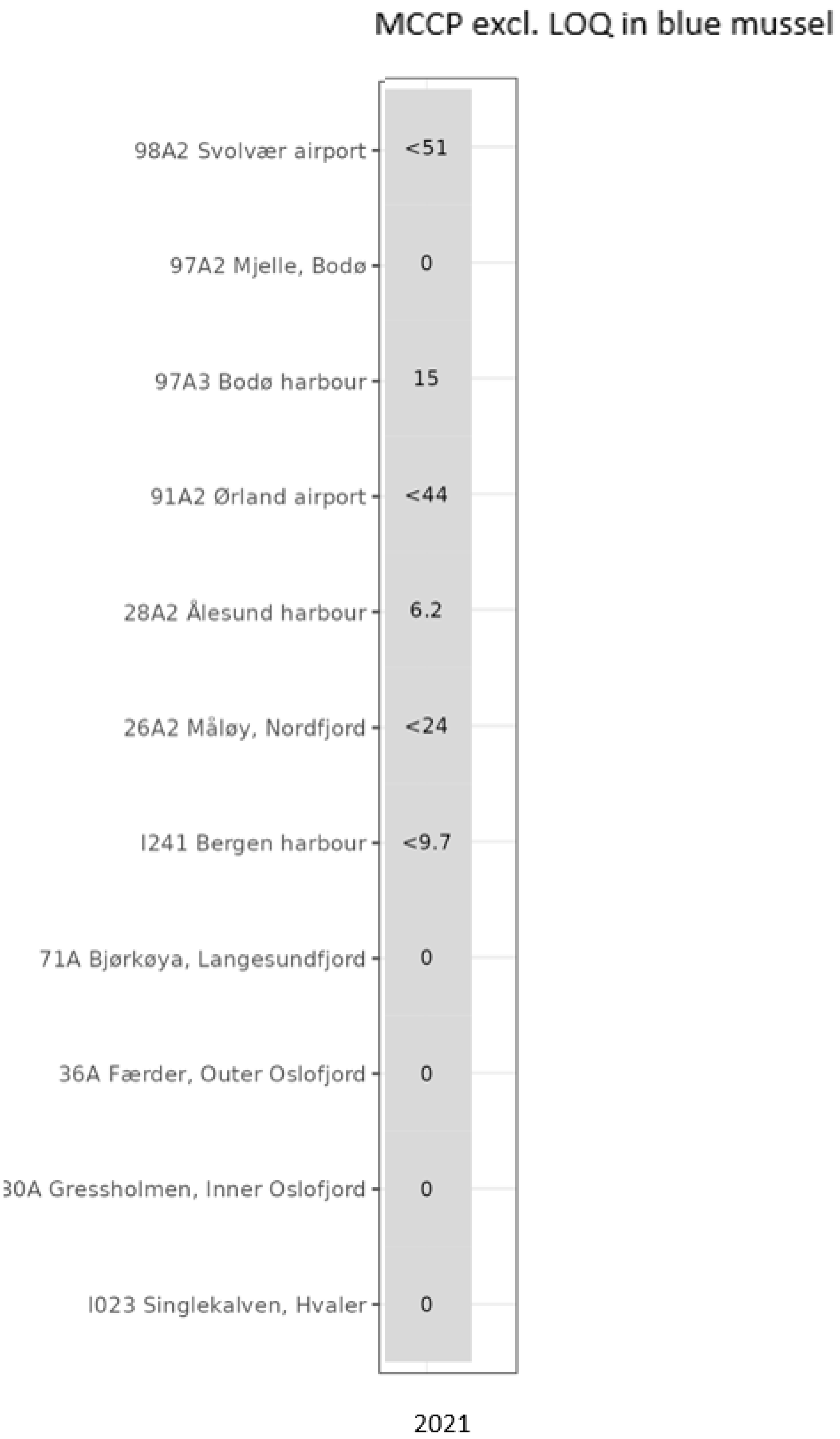
Short-chain chlorinated paraffins (SCCPs) are a mixture of chlorinated hydrocarbons with a chain length of 10 to 13 carbon atoms, and a chlorine content of 40-70%. Medium-chain chlorinated paraffins (MCCPs) are a mixture of chlorinated hydrocarbons with a chain length of 14 to 17 carbon atoms, and the chlorine content range of 40-60%. EQS for SCCPs in biota is 6000 µg/kg ww. EQS for MCCPs in biota is 170 µg/kg ww. Chlorinated paraffins were analysed in samples of cod liver from 14 stations, blue mussel from 13 stations and in samples of blood and eggs of eider from one station (**Table 1**).

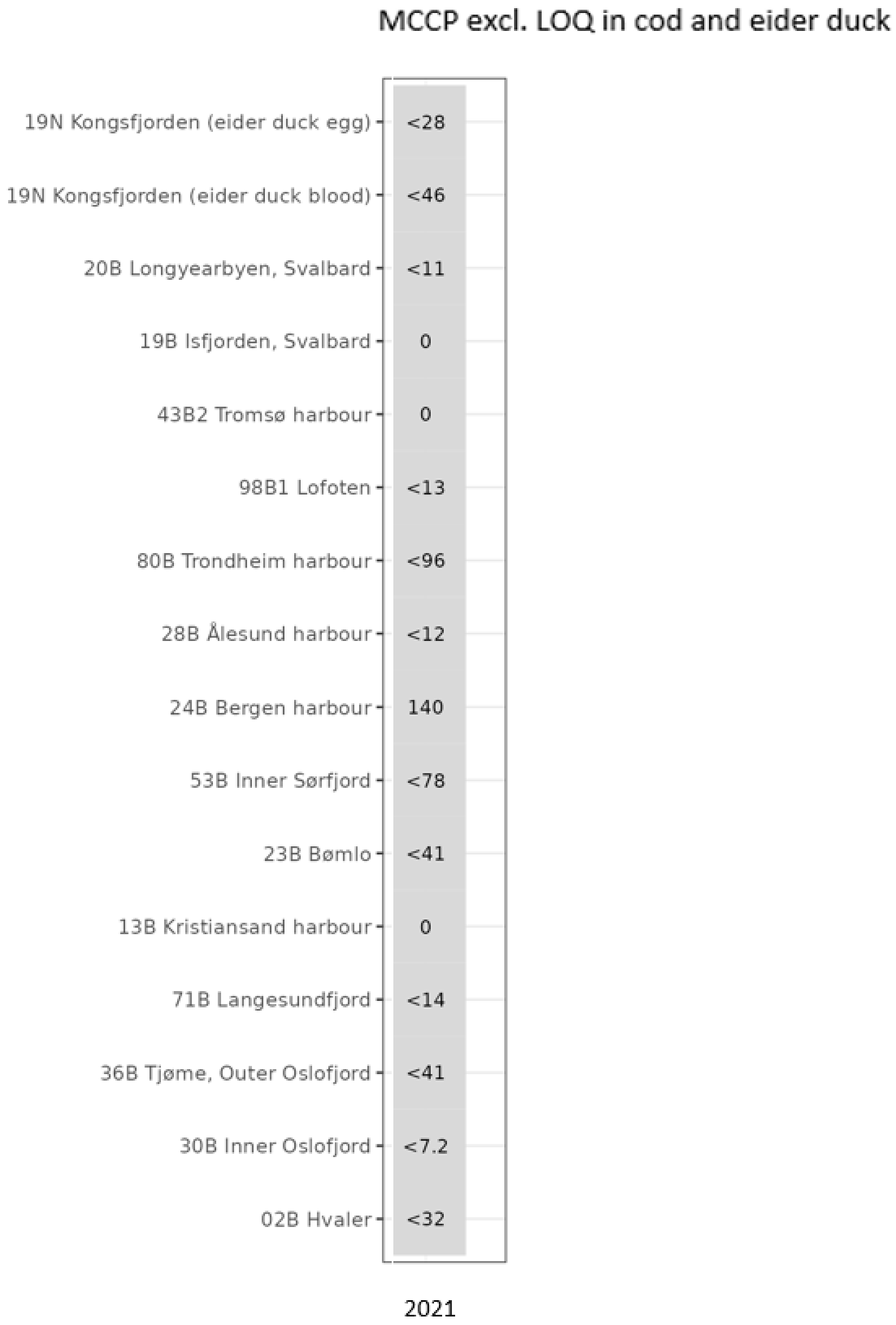
EQS

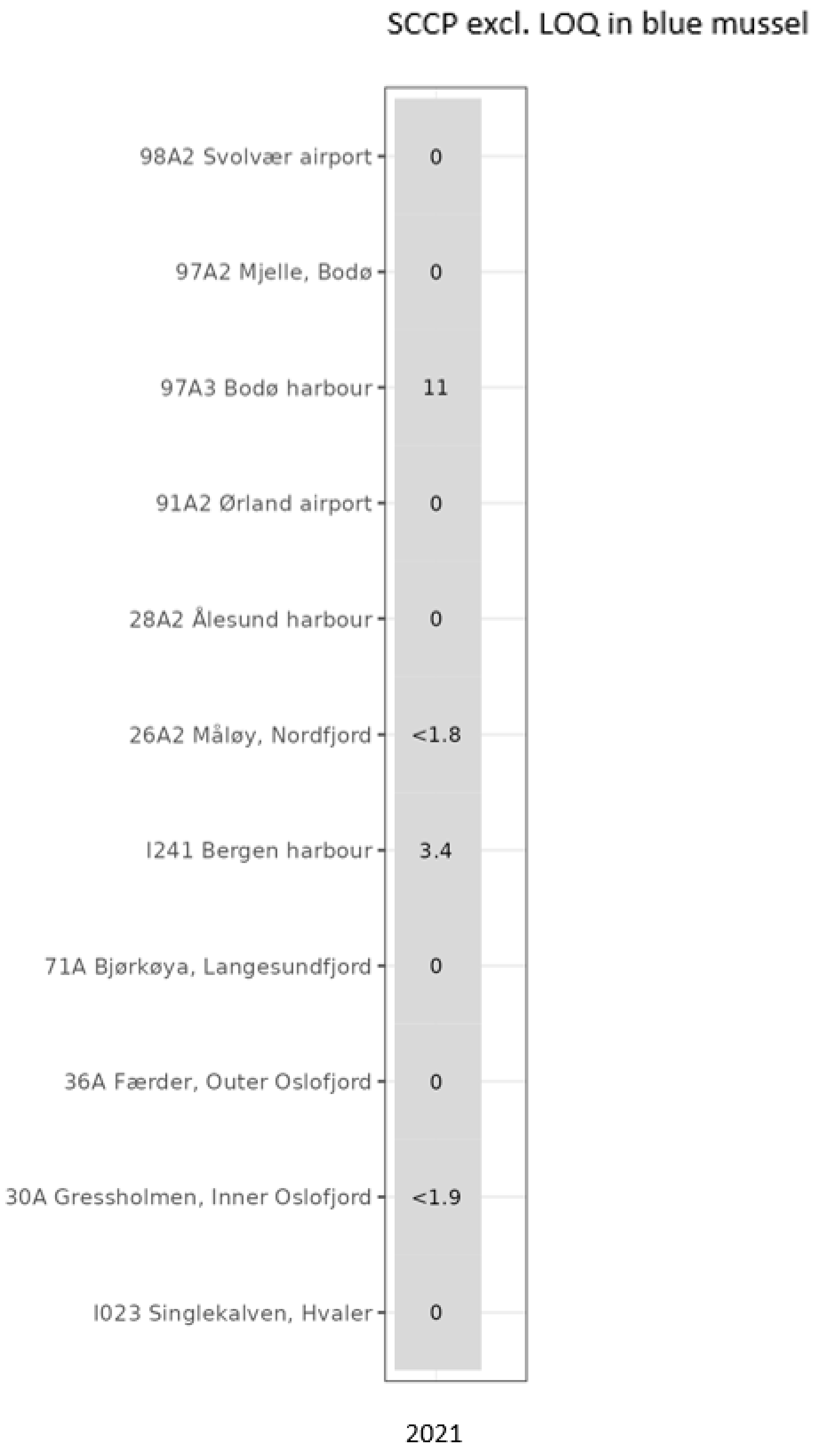
There were no median exceedances of EQSs of SCCPs (6 000 µg/kg ww, **Table 3**) and MCCPs (170 µg/kg ww) in samples of cod, blue mussel, or eider (**Figure 82**). SCCPs and MCCPs were found in low concentrations in liver of cod from Bergen harbour (24B), with median concentration of 140 µg/kg ww for MCCPs and 89 µg/kg ww for SCCPs. A few individual cod exceeded EQS for MCCP at two stations (**Figure 83**), while no cod exceeded EQS for SCCP (**Figure 83**).

SCCPs and MCCPs were quantified in blue mussel from a few stations, all concentrations were low.

There were no median concentrations above LOQ for SCCPs and MCCPs in eggs and blood of eider from Svalbard.


MCCP excl. LOQ in blue mussel
98A2 Svolvær airport
<51
97A2 Mjelle, Bodø
0
97A3 Bodø harbour
15
91A2 Ørland airport
<44
28A2 Ålesund harbour
6.2
26A2 Måløy, Nordfjord
<24
I241 Bergen harbour
<9.7
71A Bjørkøya, Langesundfjord
0
36A Færder, Outer Oslofjord
0
30A Gressholmen, Inner Oslofjord
0
I023 Singlekalven, Hvaler
0
2021


MCCP excl. LOQ in cod and eider duck
19N Kongsfjorden (eider duck egg)
<28
19N Kongsfjorden (eider duck blood)
<46
20B Longyearbyen, Svalbard
<11
19B Isfjorden, Svalbard
0
43B2 Tromsø harbour
0
98B1 Lofoten
<13
80B Trondheim harbour
<96
28B Ålesund harbour
<12
24B Bergen harbour
140
53B Inner Sørfjord
<78
23B Bømlo
<41
13B Kristiansand harbour
0
71B Langesundfjord
<14
36B Tjøme, Outer Oslofjord
<41
30B Inner Oslofjord
<7.2
02B Hvaler
<32
2021


SCCP excl. LOQ in blue mussel
98A2 Svolvær airport
0
97A2 Mjelle, Bodø
0
97A3 Bodø harbour
11
91A2 Ørland airport
0
28A2 Ålesund harbour
0
26A2 Måløy, Nordfjord
<1.8
I241 Bergen harbour
3.4
71A Bjørkøya, Langesundfjord
0
36A Færder, Outer Oslofjord
0
30A Gressholmen, Inner Oslofjord
<1.9
I023 Singlekalven, Hvaler
0
2021

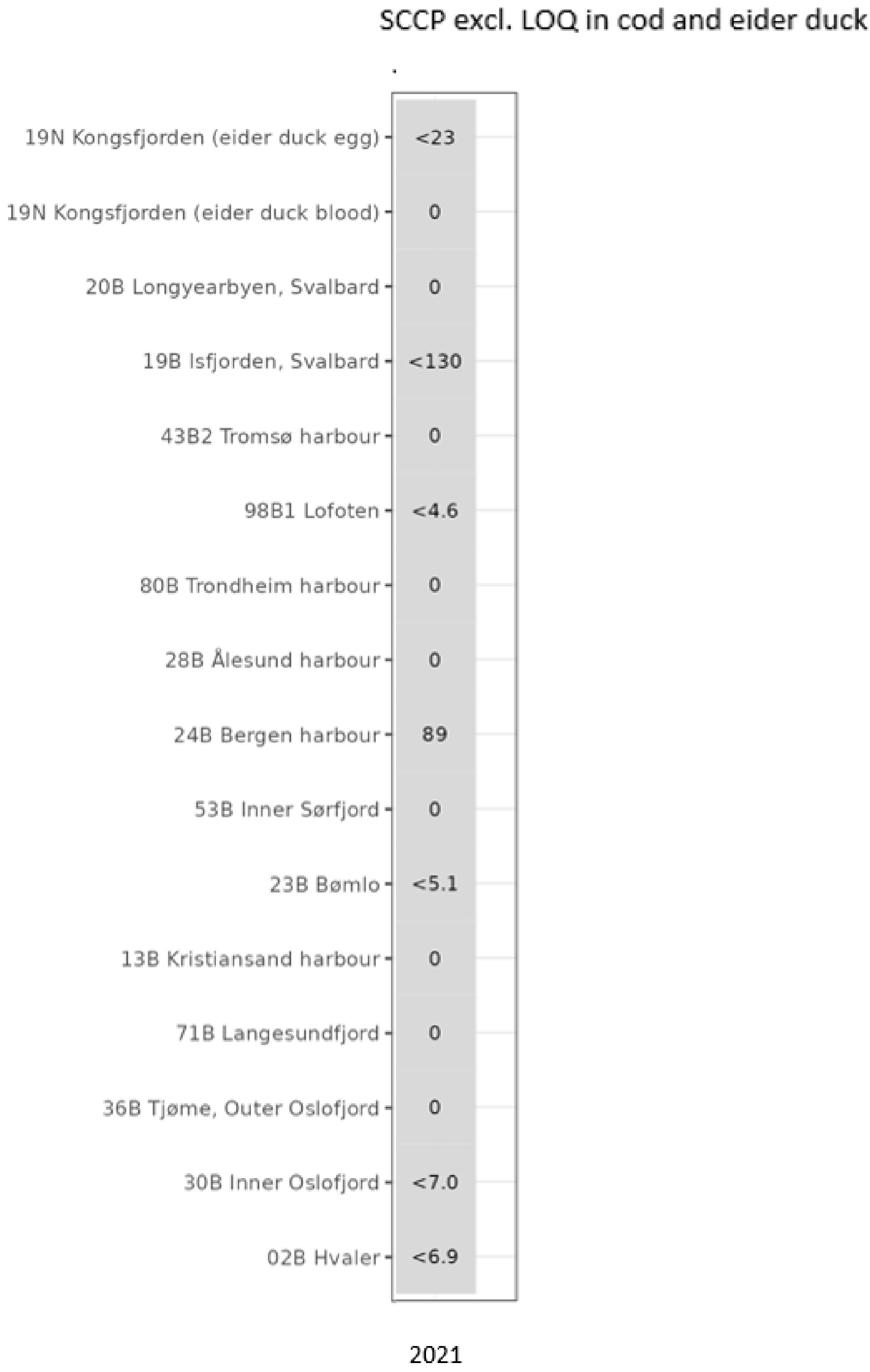
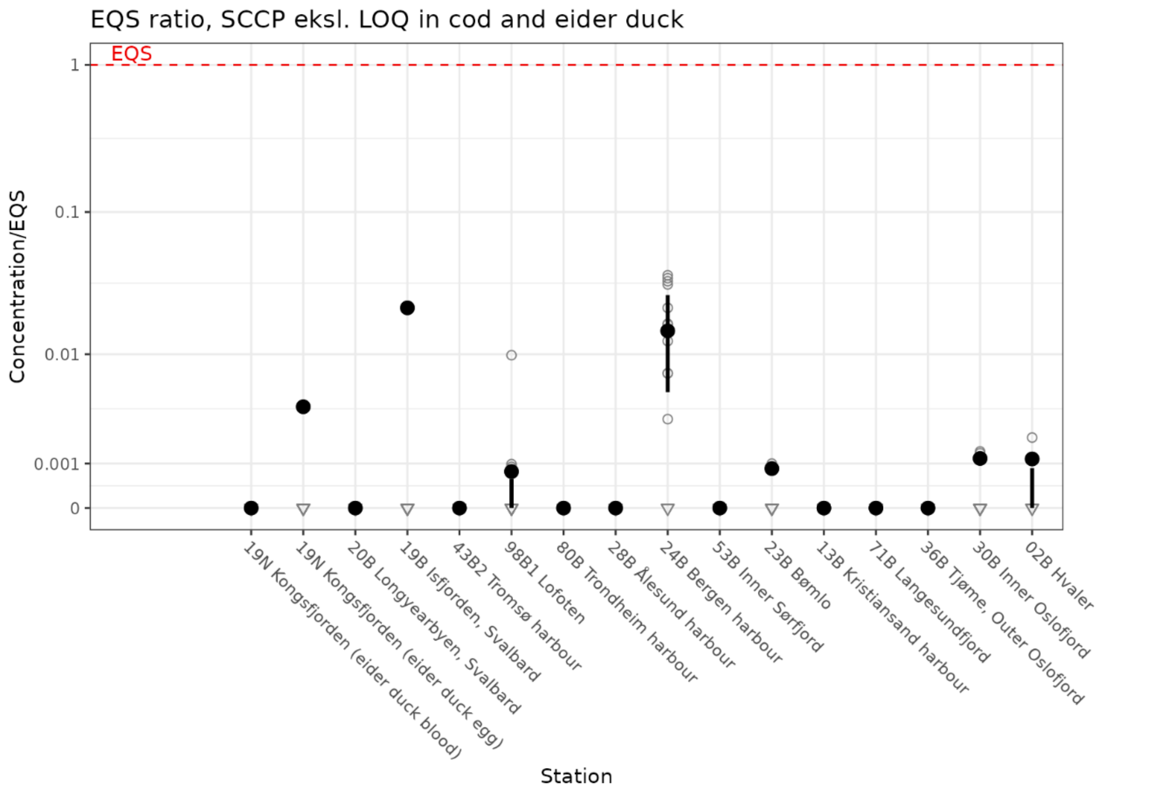
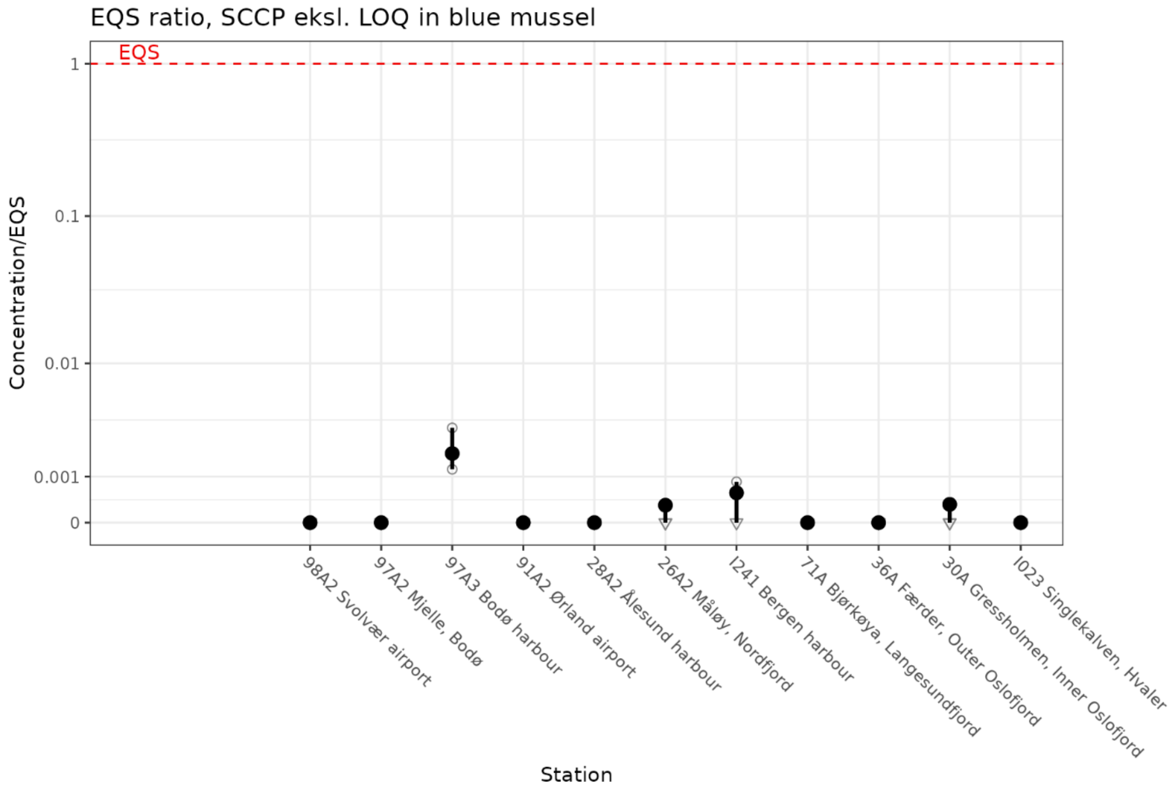


Figure 82. Heatmaps of MCCP and SCCP in blue mussel and cod/eider. For full explanation of figure see example Figure 23. NB: In these heatmaps, only data from 2021 are shown. For these data, the median concentrations are set to 0 when all samples for a station were below LOQ. When there were one or more samples above LOQ, the median concentrations were set to < “the lowest of the quantified values”. This also applies to **Figure 83**.


EQS ratio, SCCP eksl. LOQ in cod and eider duck
EQS
1
0.1
0.01
0.001
0
Concentration/EQS
19N Kongsfjorden (eider duck blood)
19N Kongsfjorden (eider duck egg)
20B Longyearbyen, Svalbard
19B Isfjorden, Svalbard
43B2 Tromsø harbour
98B1 Lofoten
80B Trondheim harbour
28B Ålesund harbour
24B Bergen harbour
53B Inner Sørfjord
23B Bømlo
13B Kristiansand harbour
71B Langesundfjord
36B Tjøme, Outer Oslofjord
30B Inner Oslofjord
02B Hvaler
Station


EQS ratio, SCCP eksl. LOQ in blue mussel
EQS
1
0.1
0.01
0.001
0
Concentration/EQS
98A2 Svolvær airport
97A2 Mjelle, Bodø
97A3 Bodø harbour
91A2 Ørland airport
28A2 Ålesund harbour
26A2 Måløy, Nordfjord
I241 Bergen harbour
71A Bjørkøya, Langesundfjord
36A Færder, Outer Oslofjord
30A Gressholmen, Inner Oslofjord
I023 Singlekalven, Hvaler
Station

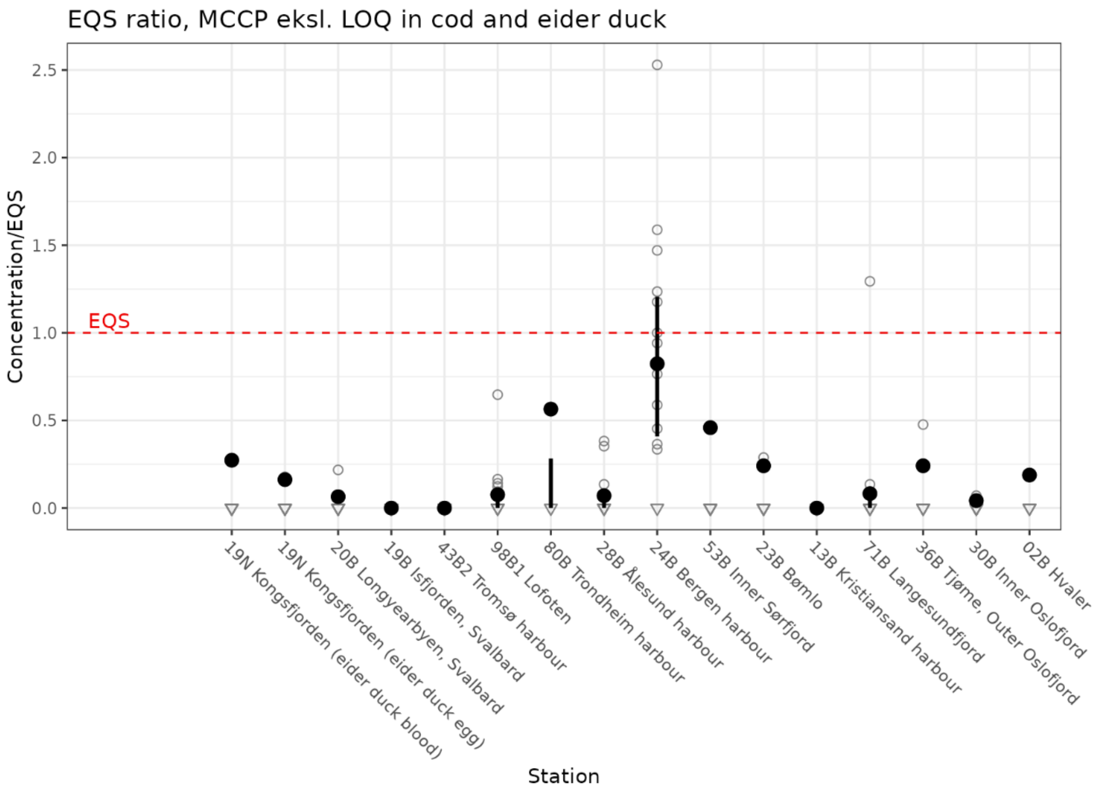


Figure 83. Ratio (concentrations/EQS) for SCCP and MCCP in cod/eider and blue mussel. Note semi-log scale in the SCCP figures. For full explanation of figure see example **Figure 24**. NB: The medians presented in these figures are medians of detected MCCP/SCCP only, excluding samples which were not quantified above LOQ. These medians therefore differ from other medians presented in other corresponding figures.

3.2.2.7 Hexabromocyclododecanes (HBCD)

HBCD is a persistent organic pollutant; it is toxic, persistent, bioaccumulates and undergo long-range environmental transport. This substance is flame retardant and has been used in high-strength polystyrene and in textiles. HBCD is one of the substances identified as priority hazardous substances (EU 2013) and was globally regulated under the Stockholm Convention in 2013. HBCD was analysed in liver of cod from 14 stations, in blue mussel from 13 stations and in blood and eggs of eider from one station.

EQS

The EQS for HBCD in biota is 167 µg/kg ww (**Table 3**). All concentrations in cod liver, blue mussel, and eider were below EQS.

Short-term trends

There were significant decreasing short-term trends for HBCD in cod from 10 stations: Hvaler (02B), Inner Oslofjord (30B), Kristiansand harbour (13B), Tjøme in Outer Oslofjord (36B), Langesundfjord (71B), Bømlo (23B), Inner Sørfjord (53B), Ålesund harbour (28B), Trondheim harbour (80B), and Tromsø harbour (43B2). Trends for two selected stations are shown in **Figure 84**. There were decreasing short-term trends with 22.2% and 18.8% annual decrease for HBCD in cod from the Kristiansand harbour (13B) and from the Langesundfjord (71B). There were significant decreasing short-term trends for HBCD in cod from 10 stations: Hvaler (02B), Inner Oslofjord (30B), Kristiansand harbour (13B), Tjøme in Outer Oslofjord (36B), Langesundfjord (71B), Bømlo (23B), Inner Sørfjord (53B), Ålesund harbour (28B), Trondheim harbour (80B), and Tromsø harbour (43B2). Trends for two selected stations are shown in **Figure 84**. There was also a significant decreasing short-term trend for HBCD in blue mussel from Bergen harbour (I241).

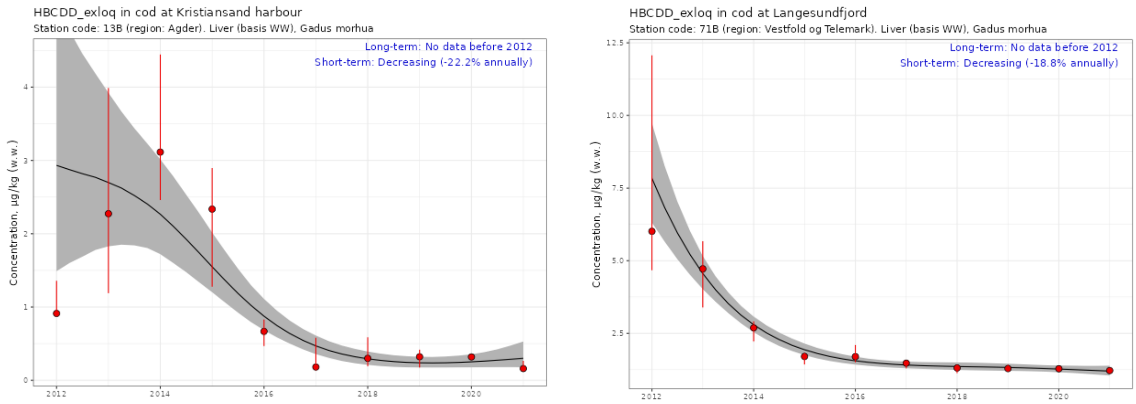


Figure 84. Time trends for HBCD in cod liver from Kristiansand harbour (13B) and Langesundfjord (71B). For full explanation of figure see example **Figure 25**.

There was also a significant decreasing short-term trend for HBCD in blue mussel from Bergen harbour (I241). HBCD was not detected in blood of eider from Svalbard but was detected in eggs of eider. The concentration of HBCD in eggs of eider was lower in 2021 than in the previous four years.

3.2.2.8 Hexachlorobenzene (HCB)

HCB was for many years used as a fungicide, and was also used in the production of rubber, aluminium, dyes, and in wood preservation. HCB is formed as a by-product during the manufacture of other chemicals (mainly solvents) and pesticides. It is an animal carcinogen and is classified as a probable human carcinogen. After its introduction as a fungicide in 1945, for crop seeds, this toxic chemical was found in all types of food. HCB is very toxic to aquatic organisms and is very persistent. HCB is included in the Convention on Long-range Transboundary Air Pollution (LRTAP) and the Stockholm Convention and has been banned globally since 2004. In 2021 HCB was analysed in liver of cod from one station, in blue mussel from two stations and in blood and eggs of eider from one station.

EQS

EQS for HCB is 10 µg/kg ww (**Table 3**). The concentrations of HCB were low, no concentrations exceeded the EQS. The median concentration of HCB in cod liver from the Inner Sørfjord (53B) was 3.8 µg/kg ww, and blue mussel from Kvalnes (56A) and Utne (64A) both had HCB concentrations below LOQ. The concentrations of HCB in blood and eggs of eider from Svalbard (19N) were 0.19 and 4.8 µg/kg ww. The concentrations of HCB in eider were lower than in the previous four years.

Long-term trends

There was a significant decreasing long-term trend for HCB in cod liver from the Inner Sørfjord (53B), with a 3.0% annual decrease (**Figure 85**). The concentrations of HCB have been low since 1993, with concentrations mostly below PROREF and the EQS.

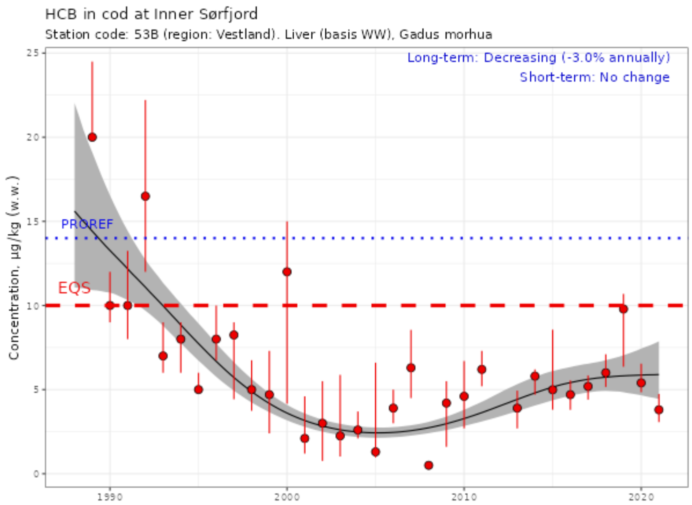


Figure 85. Time trends for HCB in cod liver from the Inner Sørfjord (53B). For full explanation of figure see example **Figure 25**.

3.2.2.9 Dichlorodiphenyldichloroethylene (DDE)

DDT (dichloro-diphenyl-trichloroethane) is the first modern synthetic pesticide developed in the 1940s. DDE is a chemical compound formed by the loss of hydrogen chloride (dehydrohalogenation) from DDT, and DDE is one of the more common breakdown products. The compounds are used for insect -and weed control. Production and use of DDT is prohibited regionally- and globally through the Convention on Long-range Transboundary Air Pollution (LRTAP) and the Stockholm Convention, but use of DDT in disease vector control is still permitted and occurs in several countries (countries in Africa, South America, and India). In Norway, the use of DDT was restricted in 1969 and the last approved use of DDT was discontinued in 1988. However, DDT from landfills, agriculture, forestry, and orchards can still be a problem and the possibility of some long-range transport cannot be excluded. In the present study DDE (p,'p-DDE, dichlorodiphenyldichloroethylene) has been used as a proxy for the priority substance DDT and was analysed in cod liver from one station and in blue mussel from two stations.

EQS

EQS for total DDT is 610 µg/kg ww (**Table 3**), but for the present study we apply the same limit to DDE in biota. Applying this EQS for blue mussel and cod liver, no concentrations exceeded the EQS.

Long-term trends

There is a significant decreasing long-term trend for DDE in cod liver from the Inner Sørfjord (53B) (4.4% annual decrease) (**Figure 86**). The concentrations have been lower than the EQS since 1997. A significant increasing long-term trend was found for DDE in blue mussel from Kvalnes (56A), with 2.6% annual increase (**Figure 86**). Highest concentrations were found in 2005 and 2013, with 66.0 µg/kg ww and 51.0 µg/kg ww. Since 2013 there has been a decrease in concentration of DDE, but not significant.

Short-term trends

There is also a significant decreasing short-term trend for DDE in cod liver from the Inner Sørfjord (53B), with a 1.3% annual decrease (**Figure 86**). The median concentration of DDE was 49.5 µg/kg ww in 2021, whereas in 2020 the median concentration of DDE was 367.0 µg/kg ww. For the two blue mussel stations there are no significant short-term trends. Both blue mussel stations had lower concentrations of DDE in 2021 than in 2020.

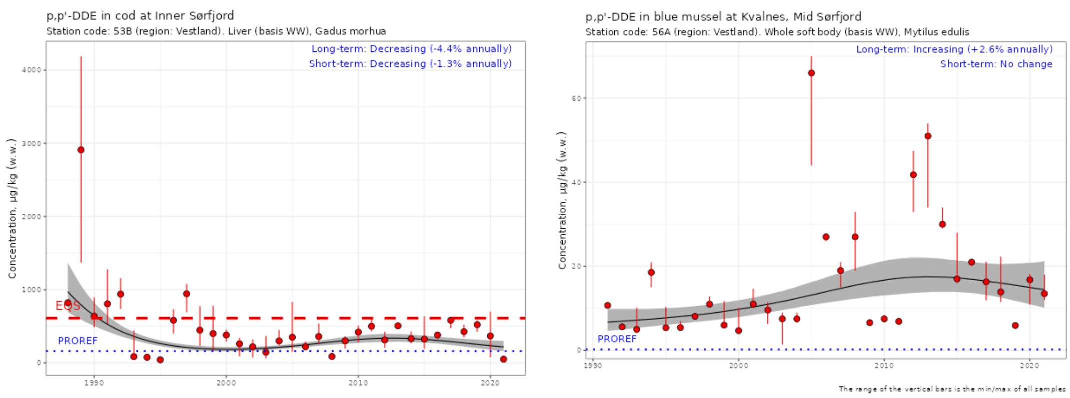


Figure 86. Time trends for p,p'DDE in cod liver from the Inner Sørfjord (53B) and in blue mussel from Kvalnes, Mid Sørfjord (56A). For full explanation of figure see example Figure 25**.**

3.2.3 Biological effect parameters

3.2.3.1 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 (71G Fugløyskjær in Langesund). 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 (< 6.1 µg/kg ww) and common periwinkle (< 1.1 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 (<0.550 µg/kg ww) and common periwinkle (<0.490 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 all stations. There were significant decreasing short-term trends for TBT in dogwhelk at Færder (36G) in the Outer Oslofjord, Risøya (76G) at Risør, Mid Karmsund (227G), Espevær (22G) by Bømlo, and Brashavn (11G) in the Varangerfjord.

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; OSPAR 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 87**) and in the area at Svolvær airport (98G) in Lofoten. Significant decreasing long-term trends were found at Færder (36G) in the Outer Oslofjord (**Figure 87**), Risøya (76G) at Risør, Lastad (131G) at Søgne, Ullerøy (15G) in Farsund, and at Espevær (22G) by Bømlo.

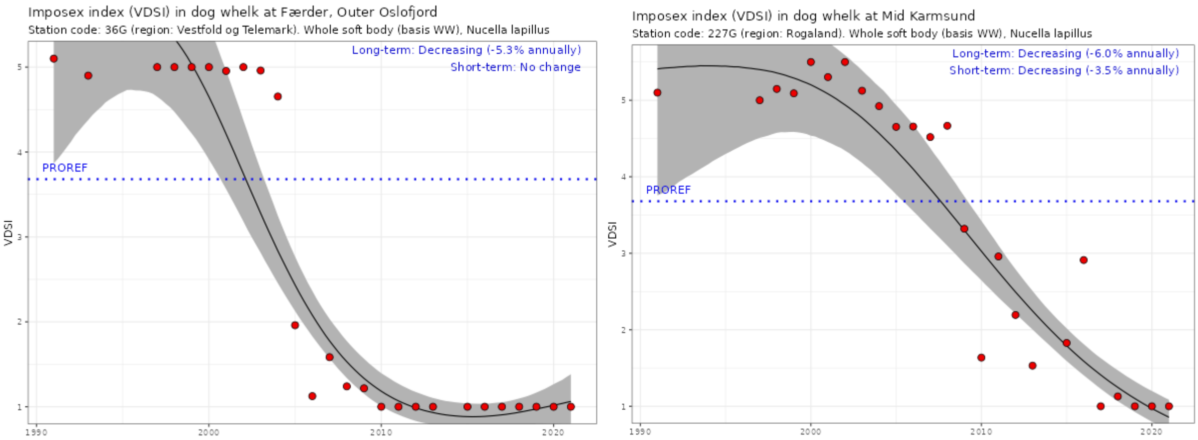


Figure 87. VDSI from 1991 to 2021 for dogwhelk from Færder (36G) in the Outer Oslofjord (left) and in the Mid Karmsund (227G) (right). For full explanation of figure see example **Figure 25**.

Selected time trends

Two time trends for VDSI in dogwhelk are shown in **Figure 87**. VDSI in dogwhelk at Færder (36G) showed decreasing long-term trend (5.3% annually). In the Mid Karmsund (227G), there were both decreasing long- and short-term trends (6.0% and 3.5% annually).

The 2021 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. ISI in common periwinkle is too sensitive for application of BAC and EAC (OSPAR 2013a).

Trends of ISI

The data of ISI in common periwinkle at Fugløyskjær (71G) showed both significant decreasing long- and short-term trends (28.5% and 14.0% annually) (**Figure 88**).

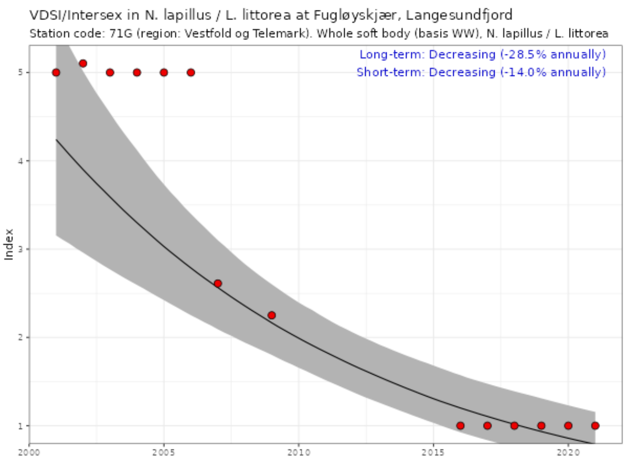


Figure 88. ISI from 2001 to 2021 for common periwinkle from Fugløyskjær in the Langesundfjord (71G). For full explanation of figure see example **Figure 25**.

3.2.3.2 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 used on cod liver samples (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 seen 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 2021 the median (non-normalized [14]) concentration of OH-pyrene metabolites in bile from cod in the Inner Oslofjord (30B) was similar to that in 2020 and resembled the concentrations most recent years. Median OH-pyrene bile concentrations in 2021 was above the ICES/OSPAR assessment criterion (background assessment criteria, BAC) in this area, as well as in fish from the Inner Sørfjord (53B). At Lista by Farsund (15B) and Bømlo (23B, reference station), median OH-pyrene bile concentrations did not exceed the ICES/OSPAR assessment criterion in 2021. Among the four stations, OH-pyrene concentrations were highest in the Inner Oslofjord (30B), followed by the Inner Sørfjord (53B) (Tukey-Kramer HSD).

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 2021 appeared similar as in 2020, and thus most previous years (since 2013). The median activity in the Inner Oslofjord (30B) in 2021 appeared slightly lower than at Bømlo (23B; reference station), however, this was not statistically significant (Tukey-Kramer HSD). Earlier frequent lower activities of ALA-D in cod from the Inner Oslofjord, as well as the Inner Sørfjord (53B) 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 2021.

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 2021, the median EROD activity was higher in the Inner Oslofjord (30B), compared to the reference station at Bømlo (23B) (Tukey-Kramer HSD). Median EROD-activities were below the ICES/OSPAR assessment criterion (background assessment criteria, BAC), at all stations.

[14] Not normalized to absorbance at 380 nm

3.2.4 Analysis of stabile 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 (Post 2002; Layman et al. 2012). 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 = $a + b$ * (Trophic Level)

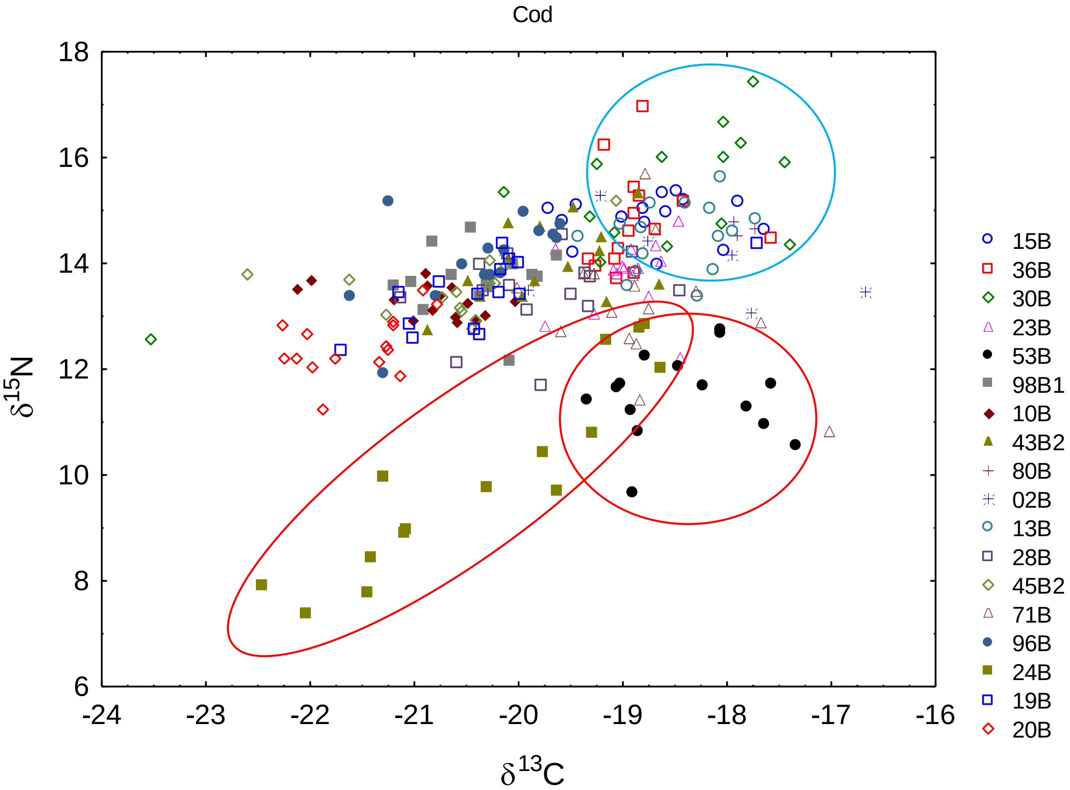
Then:
TMF = 10^{b}

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 2021 generally show the same pattern as observed in previous years i.e., a continual geographical pattern, indicating a spatial trend persistent in time (**Figure 89**).

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 89**). 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 89**) indicating a high baseline.

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.



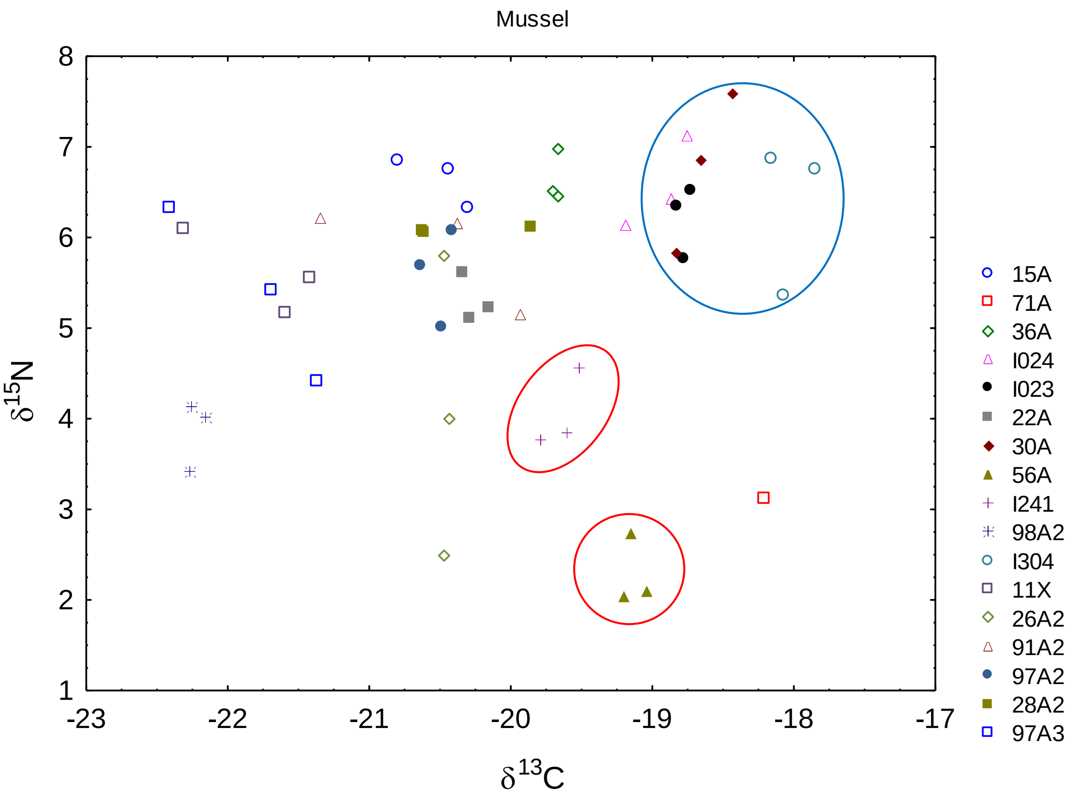


Figure 89. $\delta^{13}C$ plotted against $\delta^{15}N$ for cod and blue mussel. Blue ellipses indicate the position of the samples of cod and blue mussel from the Inner Oslofjord, while red ellipses indicate the position of the samples of cod and blue mussel from the Sørfjord and Bergen harbour.

$\delta^{15}N$ values in eiders from Svalbard (blood and egg) resembled those previously observed (Schøyen et al. 2021). The $\delta^{13}C$ values in the eiders differed between the two matrices (blood and egg; **Figure 90**), likely related to different lipid content, as lipids are $\delta^{13}C$-depleted relative to proteins (Sweeting et al. 2006). Samples were not treated to remove carbonates or lipid prior to stable isotope analysis.

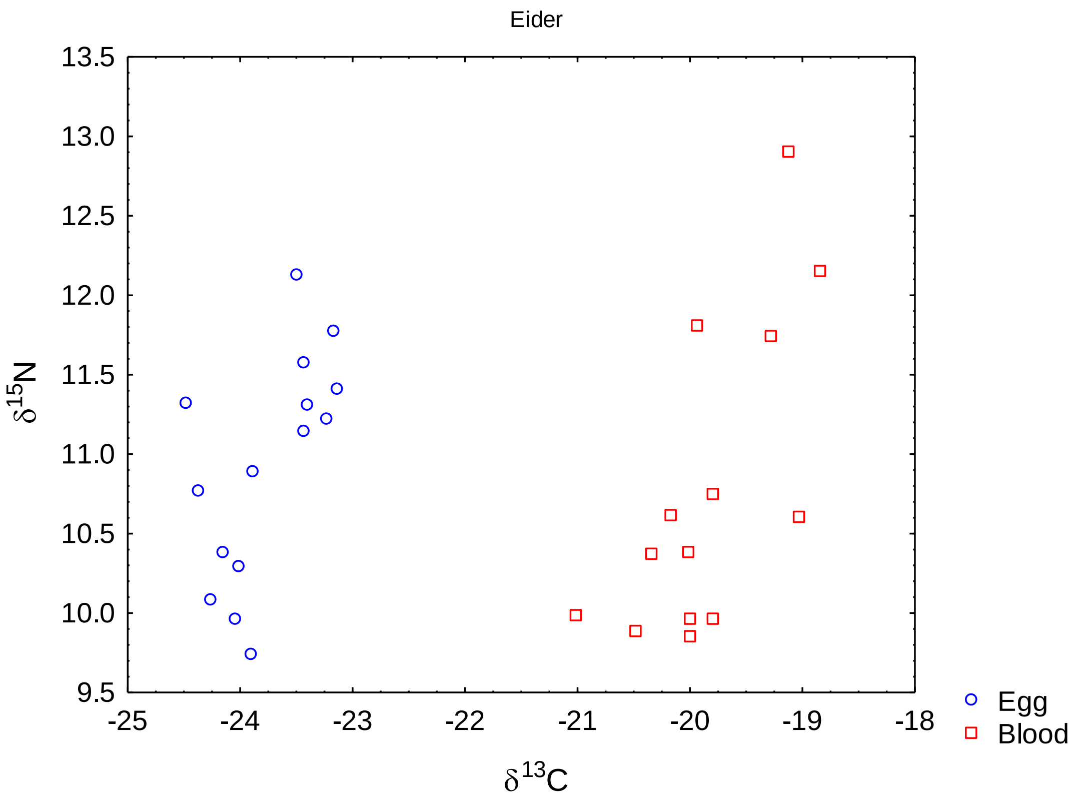


Figure 90. $\delta^{13}C$ plotted against $\delta^{15}N$ in blood (red squares) and egg (blue circles) of eider from Svalbard (19N).

4 Materials and methods appendix

4.1 Sampling and matrices

4.1.1 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.

Samples were collected during 2021 and analysed according to OSPAR guidelines (OSPAR 2021)[15] 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 2021-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. 2021).

4.1.2 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 2**). 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.

The blue mussel samples were collected from 19th August to 1st December 2021. 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 mussel were collected by NIVA, except for some blue mussel stations collected by local contacts.

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

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.

4.1.3 Dogwhelk and common periwinkle

Concentrations and effects of organotin on dogwhelk were investigated at eight stations and one station for common periwinkle (**Table 1**; **Table 2**). 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 given in (Følsvik et al. 1999).

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 7th September to 05th November 2021.

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 described by (Bauer et al. 1995). The ISI ranges from zero (no effect) to four (maximum intersex effect).

4.1.4 Atlantic cod

Atlantic cod was caught from 18 stations (**Table 1**; **Table 2**). 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 18th August to 03rd November 2021. Cod was caught by 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). 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 which confirmed, at least for these samples, that only coastal cod was caught. 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.

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, shell lengths, dry weight percentages) are publicly available but not necessarily used for this report.

4.1.5 Common eider

Contaminants in the common eider were investigated at one station in Kongsfjorden at Svalbard (19N), which the present study considered as a reference station (**Table 1**; **Table 2**). Blood samples were collected from 15 individuals (two subsamples from each) and eggs from 15 other individuals 5th June 2021. All samples are from adult nesting females.

4.2 Analytical procedures and information on quality assurance

The laboratories (NIVA, subcontractors Eurofins 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 2020 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. Uncertain values identified by the analytical laboratory, or the reporting institute are flagged in the database. 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[16].

Summary of quality control results
Standard Reference Materials (SRM) as well as in-house reference materials were analysed regularly (**Table 5**). Apple juice was used as an in-house reference material for the quality control of the determination of metals. The reference material for determination of BDEs, HBCDDs, 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 in 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

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

material, and spiked fish liver was used for quality control of per- and polyfluorinated chemicals (PFAS).

Table 5. Summary of the quality control of results for the 2021 biota samples analysed in 2021-2022. 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 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)	85,7 ± 25,7	20	10	81,2	4,89
BDE47	**2,2,4,4',-Tetrabromodiphenylether**	SB	Internal RM (fish oil)	1590 ± 477	20	10	1646	56,0
BDE100	**2,2',4,4',6-Pentabromodiphenylether**	SB	Internal RM (fish oil)	324 ± 97	20	10	339	13,9
BDE99	**2,2',4,4',5-Pentabromodiphenylether**	SB	Internal RM (fish oil)	248 ± 74	20	10	260	9,85
BDE154	**2,2',4,4',5,6'-Hexabromodiphenylether**	SB	Internal RM (fish oil)	224 ± 67	20	10	254	11,0
BDE153	**2,2',4,4'5,5'-Hexabromodiphenylether**	SB	Internal RM (fish oil)	58,5 ± 17,6	20	10	67,7	7,32
BDE209	**Decabromodiphenylether**	-	-	-	-	-	-	-
BDE49	**2,2',4,5'-tetrabromodiphenyleter**	SB	Internal RM (fish oil)	431 ± 129	20	10	463	16,5
BDE66	**2,3',4,4'-Tetrabromodiphenyleter**	-	-	-	-	-	-	-
BDE119	**2,3',4,4',6-Pentabromodiphenyl ether**	-	-	-	-	-	-	-
CB77	**PCB congener CB77**	-	-	-	-	-	-	-
CB52	**PCB congener CB52**	SB/LI	Internal RM (fish)	444 ± 133	33	12	464	25,6
CB28	**PCB congener CB28**	SB/LI	Internal RM (fish)	269 ± 81	33	12	292	21,7
CB189	**PCB congener CB189**	-	-	-	-	-	-	-
CB180	**PCB congener CB180**	SB/LI	Internal RM (fish)	4590 ± 1377	33	12	4859	322
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)	5289 ± 1587	33	12	5116	308
CB138	**PCB congener CB138**	SB/LI	Internal RM (fish)	3605 ± 1082	33	12	3903	239
CB126	**PCB congener CB126**	-	-	-	-	-	-	-
CB123	**PCB congener CB123**	-	-	-	-	-	-	-
CB118	**PCB congener CB118**	SB/LI	Internal RM (fish)	883 ± 265	33	12	928	51,5
CB114	**PCB congener CB114**	-	-	-	-	-	-	-
CB105	**PCB congener CB105**	-	-	-	-	-	-	-
CB101	**PCB congener CB101**	SB/LI	Internal RM (fish)	1647 ± 494	33	12	1776	322
DDEOP	**o,p'-DDE**	SB/LI	Internal RM (feed)	0,11 ± 0,03	14	10	0,08	0,015
TDEOP	**o,p'-DDD**	SB/LI	Internal RM (feed)	0,27 ± 0,08	14	10	0,23	0,015
DDTOP	**o,p'-DDT**	SB/LI	Internal RM (feed)	0,26 ± 0,08	14	10	0,21	0,034
DDEPP	**p,p'-DDE**	SB/LI	Internal RM (feed)	5,01 ± 1,50	14	10	4,47	0,290

TDEPP	**p,p'-DDD**	SB/LI	Internal RM (feed)	1,73 ± 0,50	14	10	1,57	0,271
DDTPP	**p,p'-DDT**	SB/LI	Internal RM (feed)	0,61 ± 0,20	14	10	0,56	0,038
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)	1,21 ± 0,36	14	11	1,26	0,085
β-HBCDD	**β- Hexabromocyclododecane**	SB	Internal RM (fish oil)	0,08 ± 0,02	14	11	0,07	0,013
γ-HBCDD	**γ- Hexabromocyclododecane**	SB	Internal RM (fish oil)	0,32 ± 0,10	14	11	0,38	0,038
BGHIP	**Benzo[ghi]perylene**	-	-	-	-	-	-	-
ICDP	**Indeno[1,2,3-cd]pyrene**	-	-	-	-	-	-	-
BBJF	**Benzo[b+j]fluoranthene**	SB	Internal RM (fish oil)	513 ± 154	6	6	552	73,5
DBA3A	**Dibenzo[ac,ah]anthracene**	-	-	-	-	-	-	-
BKF	**Benzo[k]fluoranthene**	-	-	-	-	-	-	-
ACNLE	**Acenaphthylene**	SB	Internal RM (fish oil)	1210 ± 363	6	6	1232	239
ANT	**Anthracene**	SB	Internal RM (fish oil)	1040 ± 312	6	6	1188	79,6
BAA	**Benzo[a]anthracene**	SB	Internal RM (fish oil)	511 ± 153	6	6	531	109
BAP	**Benzo[a]pyrene**	SB	Internal RM (fish oil)	233 ± 71	6	6	276	26,5
CHR	**Chrysene**	SB	Internal RM (fish oil)	502 ± 151	6	6	598	34,7
FLU	**Fluoranthene**	SB	Internal RM (fish oil)	3230 ± 969	6	6	3629	323
FLE	**Fluorene**	SB	Internal RM (fish oil)	4490 ± 1347	6	6	4873	377
NAP	**Naphthalene**	-	-	-	-	-	-	-
PA	**Phenanthrene**	SB	Internal RM (fish oil)	9110 ± 2733	6	6	9696	409
PYR	**Pyrene**	SB	Internal RM (fish oil)	2080 ± 624	6	6	2240	309
ACNE	**Acenaphthene**	SB	Internal RM (fish oil)	2140 ± 642	6	6	2072	219
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)	1,5 ± 0,5	5	6	-	0,083
DBT	**Dibutyltin (DBT)**	-	-	-	-	-	-	-
TBT	**Tributyltin (TBT)**	SB	ZRM 81 (mussel)	2,2 ± 0,7	6	5	-	0,034
TPhT	**Triphenyltin (TPhT)**	SB	ZRM 81 (mussel)	1,4 ± 0,4	6	6	1,40	0,038
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	-	-	-	-	-	-	-
	Trans-Nonachlor	SB	Internal RM (feed)	1,39 ± 0,40	21	17	1,35	0,24

* 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 egg and blood samples from common eider (*Somateria mollissima*) and 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).

4.3 QA/QC

Additional to the general quality assurance (QA) done by the individual laboratory all the results from EF and NIVA are transferred into NIVAs laboratory information management system (LIMS). Import of NILU results are now under validation and will be included in this extra quality control from 2023 (2022 data). 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 4.6**) to make this control easier and more efficient. Here trends from the last years will appear and deviating results are marked (example in **Figure 91**). 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 2021 data, four mussel samples were reanalysed for PCB (results confirmed for the stations Ullerøy at Farsund (15A), Vikingneset (65A), Skallnes in the Varangerfjord (10A2), and Brashavn in the Varangerfjord (11X)) and one cod sample were reanalysed for Hg (result was changed). All activity is recorded in NIVAs deviation/control system.

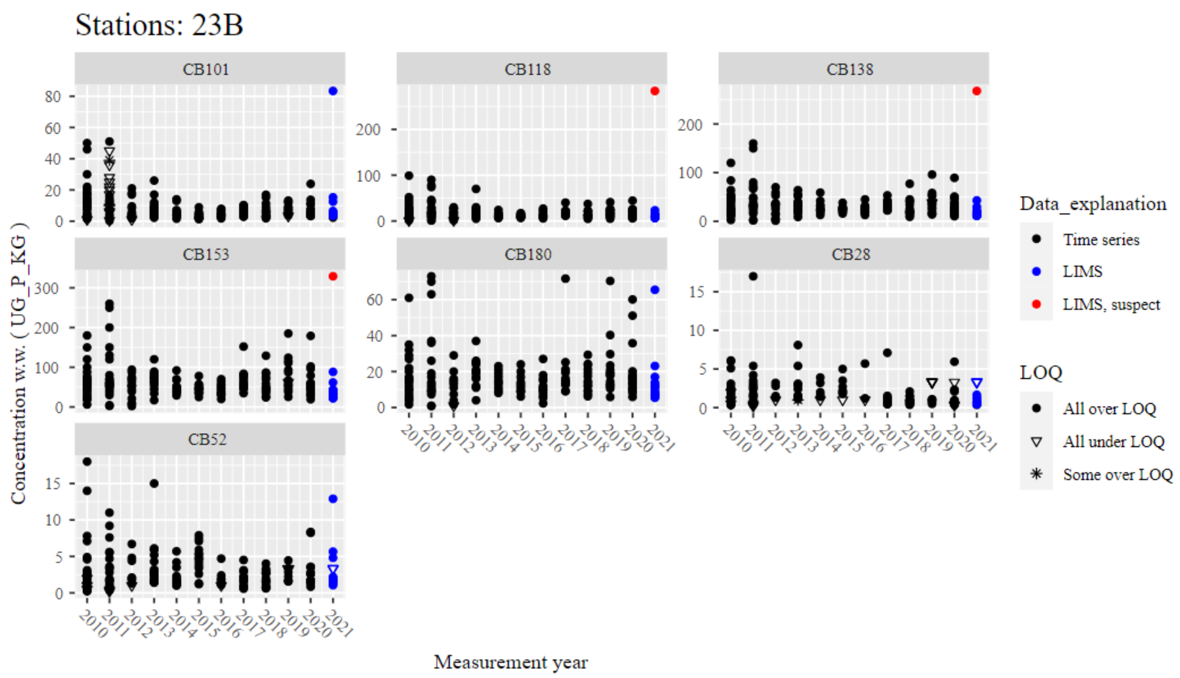


Figure 91. Screenshot of app developed in R of how trend series are presented during QA and made possible for manual assessment. This year's results that are in line with previous years' results appear in blue, while data that are either (a) out of line with other results this year or (b) out of line (suspected outlier) with this year's results are flagged with a red colour.

4.4 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 by (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 Norwegian Environment Agency 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 (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 Hg. To address this issue for this report, and in dialogue with the Norwegian Environment Agency, 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 2021-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.

4.5 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 a given 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 <u>not</u> 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).

4.6 Statistical time trend analysis

The statistical time trend analysis follows the method used in OSPAR for contaminants in biota[17] as closely as possible (there has been changes to the OSPAR methodology every year since 2014[18]). 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 depends on the number of years of data, counting only years with at least one concentration over LOQ[19]:

- 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 in order 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 starts 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.

Using every sample measurement instead of the only annual medians (as in the previous years' analysis) 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

[17] https://dome.ices.dk/ohat/trDocuments/2022/help_methods_biota_contaminants.html
[18] https://dome.ices.dk/ohat/trDocuments/2022/help_methods_changes.html
[19] https://dome.ices.dk/ohat/trDocuments/2022/help_methods_less_thans.html

statistical power; therefore, more time trends are detected than before (see example in **Figure 92**). 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[20].

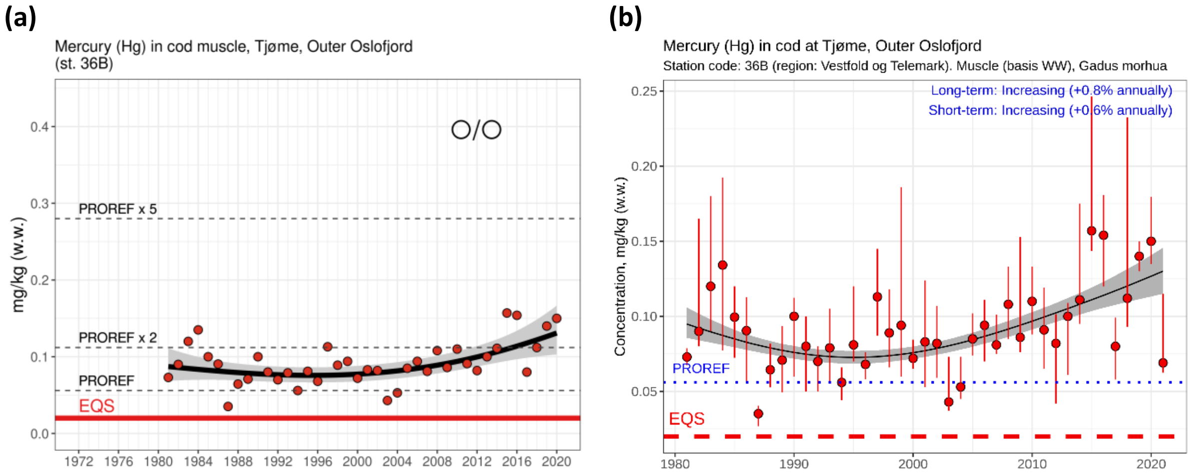


Figure 92. Differences in detection of time trends for mercury in cod muscle at Tjøme (36B). (a) The previous time series approach, using only median values. Neither short- or long-term time trends were detected (P > 0.18). (b) The updated statistical method utilising all data measurements (lines shows 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).

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[21] 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).

4.7 Other statistical analyses

JMP[22] statistical software (version 16.2.0) was used for data treatment after initial treatment in R. Mosaic plots, heatmaps and tables used in extended summary data were produced using JMP.

[20] https://github.com/DagHjermann/leftcensored
[21] https://www.r-project.org/
[22] https://www.jmp.com/

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Supplementary data

A total of 622 datapoints, i.e. combinations of contaminant × station × tissue) have been assessed for exceedances of PROREF for contaminants not selected for presentation in 2021. The contaminants are listed in **Table S1**.

Table S1. List of contaminants *not selected in 2021* for which a PROREF exist. The PROREFs are given in µg/kg (ng/g ww).

	PROREF for contaminants (µg/kg ww)				
	Contaminant	**Blue mussel**	**Cod**	**Dogwhelk**	**Common periwinkle**
Metals	Tin (Sn)	0.3	0.3		
PFAS	perfluorobutane sulfonate (PFBS)		8		
	perfluorononanoic acid (PFNA)		5		
PBDEs	PBDE congener -28 (BDE28)		1.4		
	PBDE congener -49 (BDE49)		3.95		
	PBDE congener -66 (BDE66)		0.59		
	PBDE congener -71 (BDE71)		0.4		
	PBDE congener -77 (BDE77)		1.69		
	PBDE congener -85 (BDE85)		1.72		
	PBDE congener -126 (BDE126)	0.05	0.1		
	PBDE congener -138 (BDE138)		0.3		
	PBDE congener -154 (BDE154)	0.05	1.5		
	PBDE congener -183 (BDE183)	0.3	0.6		
	PBDE congener -196 (BDE196)	0.3	1		
	PBDE congener -209 (BDE209)	1.29	2		
PAHs	Acenaphthene (ACNE)	0.8			
	Acenaphthylene ACNLE	1			
	Anthracene (AN)T	0.8			
	Benzo[b+j]fluoranthene (BBJF)	6.24			
	Benzo[ghi]perylene (BGHIP)	2.07			
	Benzo[k]fluoranthene (BKF)	1.5			
	Dibenz[a,c/a,h]anthracene (DBA3A)	0.5			
	Fluorene (FLE)	1.6			
	Indeno[1,2,3-cd]pyrene (ICDP)	1.73			
	Naphthalene (NAP)	17.3			
	Phenanthrene (PA)	2.28			
PCBs	PCB congener 28 (CB28)	0.12	8		
	PCB congener 52 (CB52)	0.2	16		
	PCB congener 101 (CB101)	0.2	32.3		
	PCB congener 180 (CB180)	0.1	45.8		
HBCDs	β-hexabromocyclododecane (HBCDB)	0.02	0.4		
	γ-hexabromocyclododecane (HBCDG)	0.03	0.89		
DDTs	p,p'-DDE (a DDT metabolite)	0.224	160.		
	p,p'-DDD (TDEPP)	0.1	32		
Pesticides	α HCH = alpha HCH (HCHA)		8		
	Lindane, γ HCH = gamma hexachlorocyclohexane (HCHG)		11		
TBT-related compounds	Dibutyltin (DBT)				1.964
	Dioctyltin (DOT)			1.2	
	Monobutyltin (MBT)				1.344
	Monooctyltin (MOT)			1.2	
	Tributyltin (TBT)			23.54	
	Tricyclohexyl-stannylium (TCHT)			2.33	
	Triphenyltin (TPhT)			1.65	
	Tetrabutyltin (TTBT)			1.0149	
BEM	ethoxyresorufin-O-deethylase (EROD)		192.		
Biomarkers	Vas Deferens Sequence Index (VDSI)			3.68	

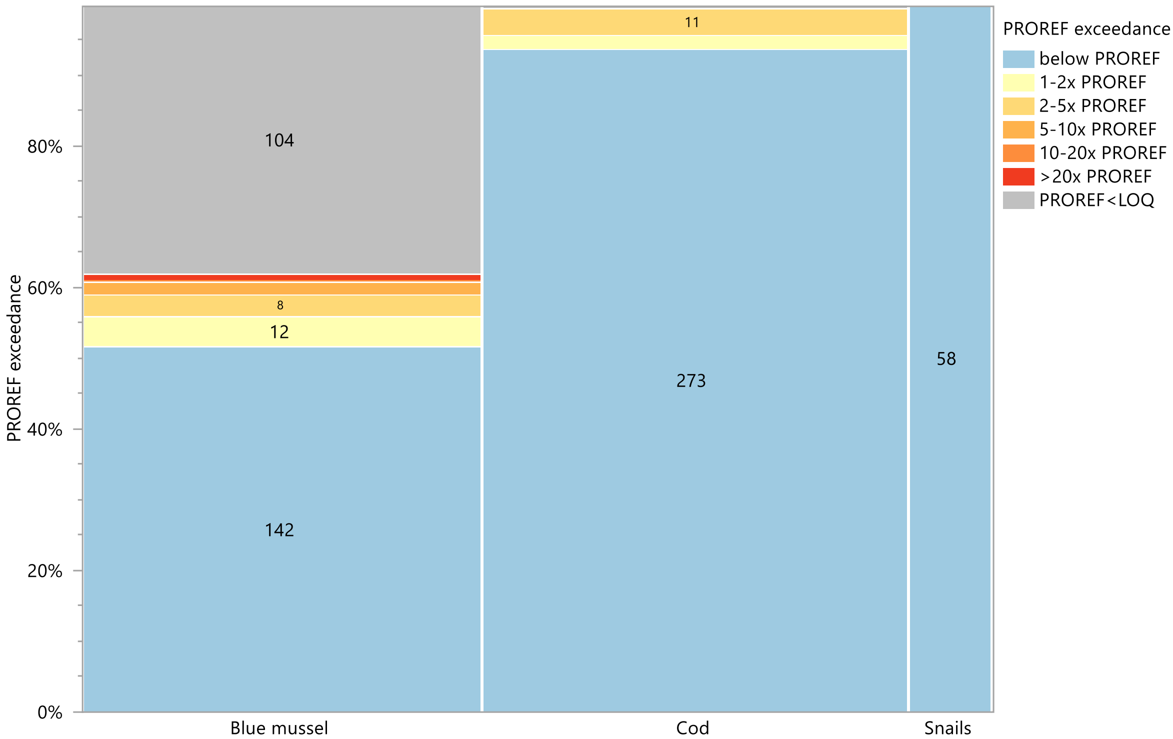

Figure S1. Exceedances of PROREF for contaminants *not selected in 2021* in a mosaic plot. The cells are labelled by the number of stations and parameters. The exceedances are considered by the median for each station and species. The colours represent below or above exceedance of PROFEF (darker yellow to red), or that the PROREF was below LOQ, and therefore could not be classified.

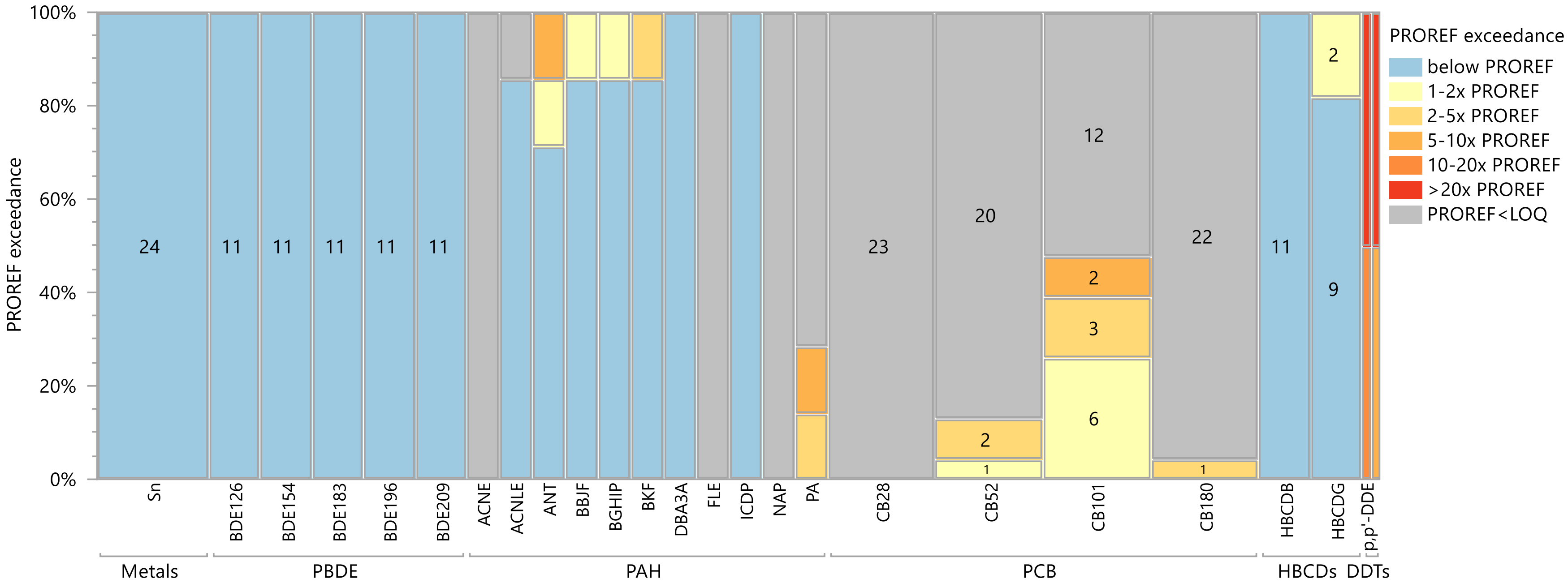

Figure S2. Exceedances of PROREF for contaminants *not selected in 2021* in **blue mussel** by contaminant and group of contaminants. The cells are labelled by the number of stations sampled. The exceedances are considered by the median for each station. The colours represent below or above exceedance of PROFEF (darker yellow to red), or that the PROREF was below LOQ, and therefore could not be classified.

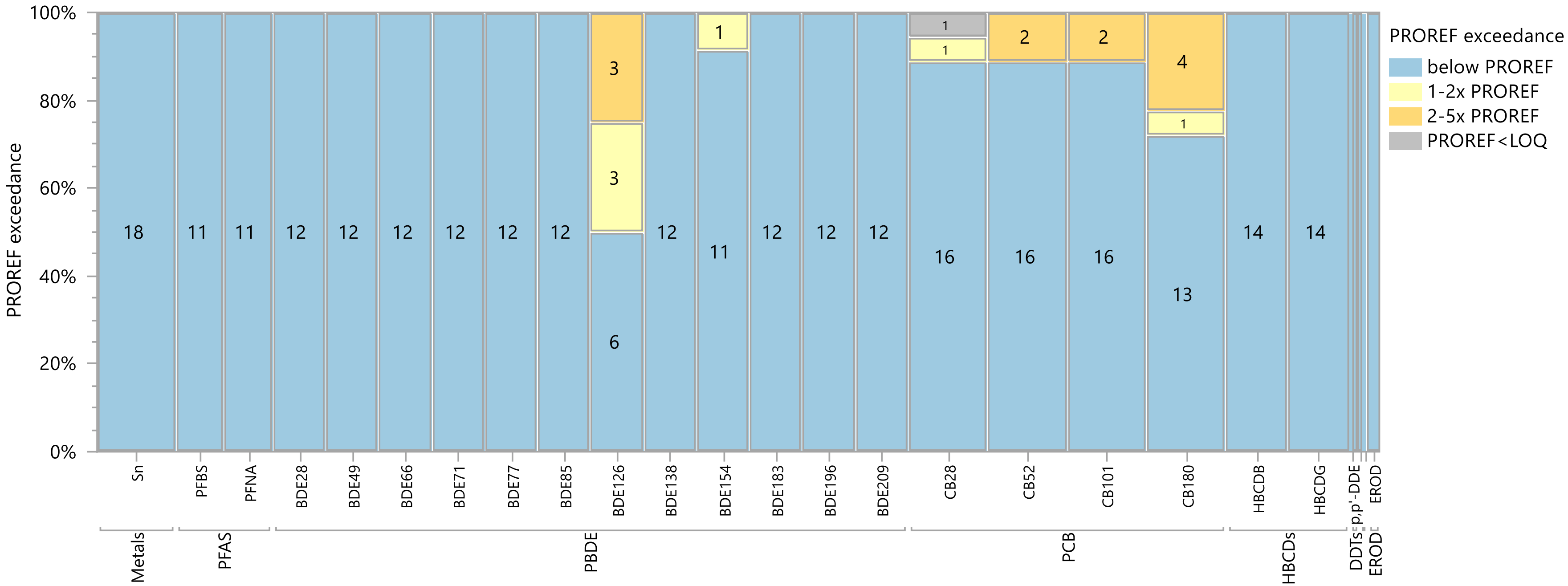

Figure S3. Exceedances of PROREF for contaminants *not selected in 2021* in **cod** by parameter and group of contaminants. The cells are labelled by the number of stations sampled. The exceedances are considered by the median for each station. The colours represent below or above exceedance of PROFEF (darker yellow to red), or that the PROREF was below LOQ, and therefore could not be classified.

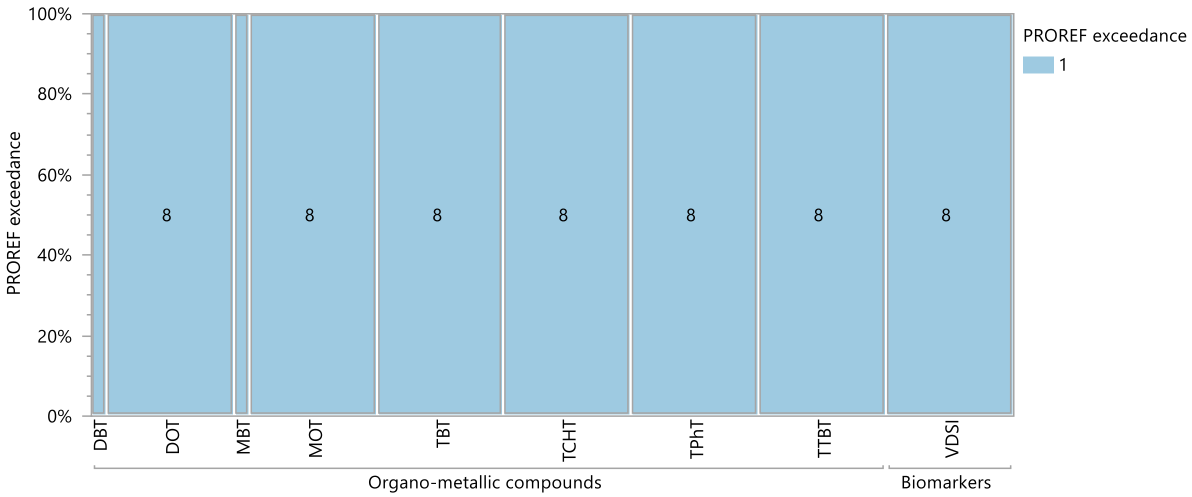

Figure S4. Exceedances of PROREF for contaminants *not selected in 2021* in **snails** by parameter and group of parameters. The cells are labelled by the number of stations sampled. The exceedances are considered by the median for each station. The colours represent below or above exceedance of PROFEF (darker yellow to red), or that the PROREF was below LOQ, and therefore could not be classified.

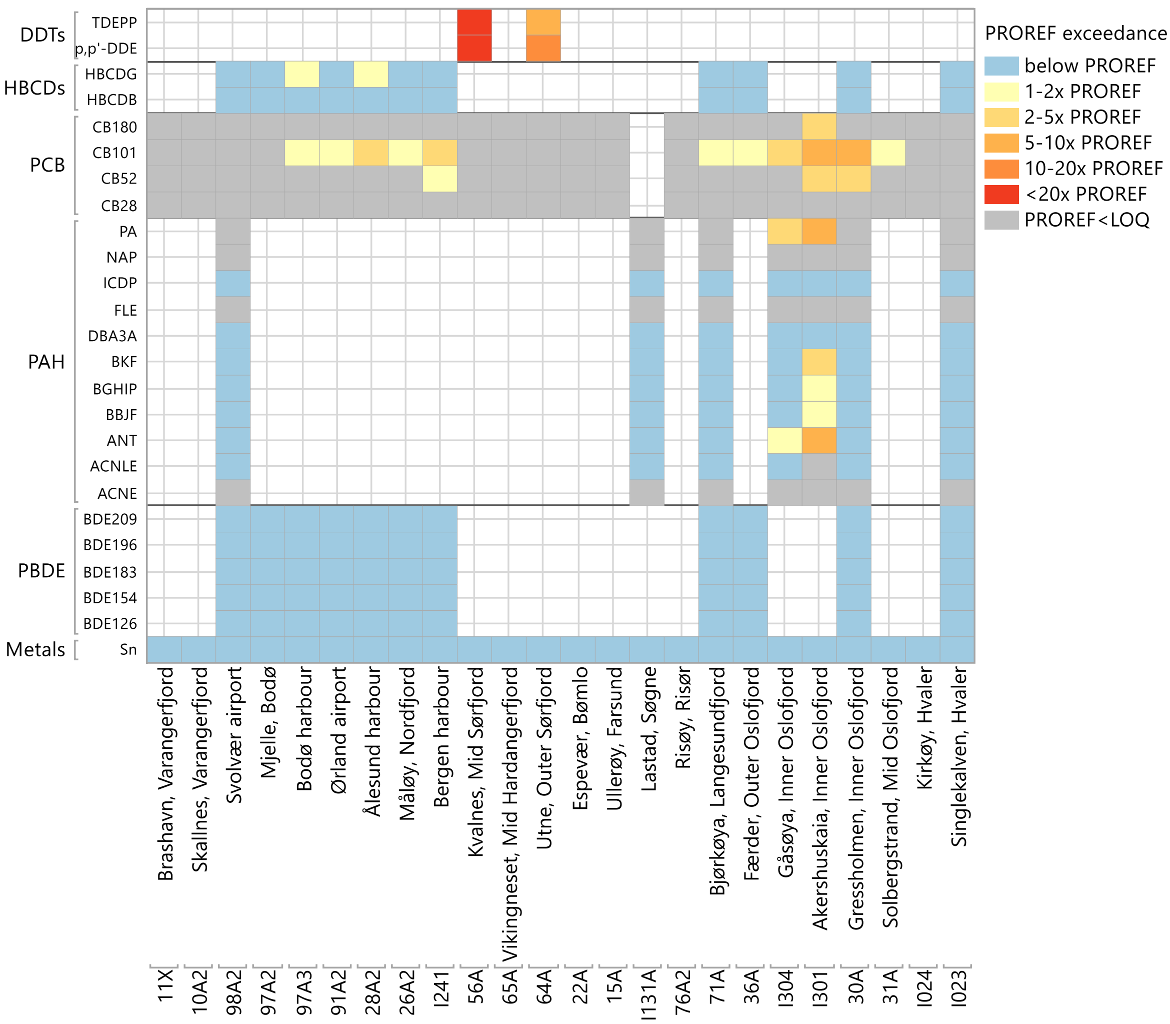

Figure S5. Heatmap of exceedances of PROREF in **mussel** for contaminants *not selected* in 2021. The colours represent below or above exceedance of PROREF. Empty "cells" mean that the contaminant was not analysed for at the indicated station. Grey lines show the midpoint of each station and contaminant, and darker lines have been inserted between contaminant groups.

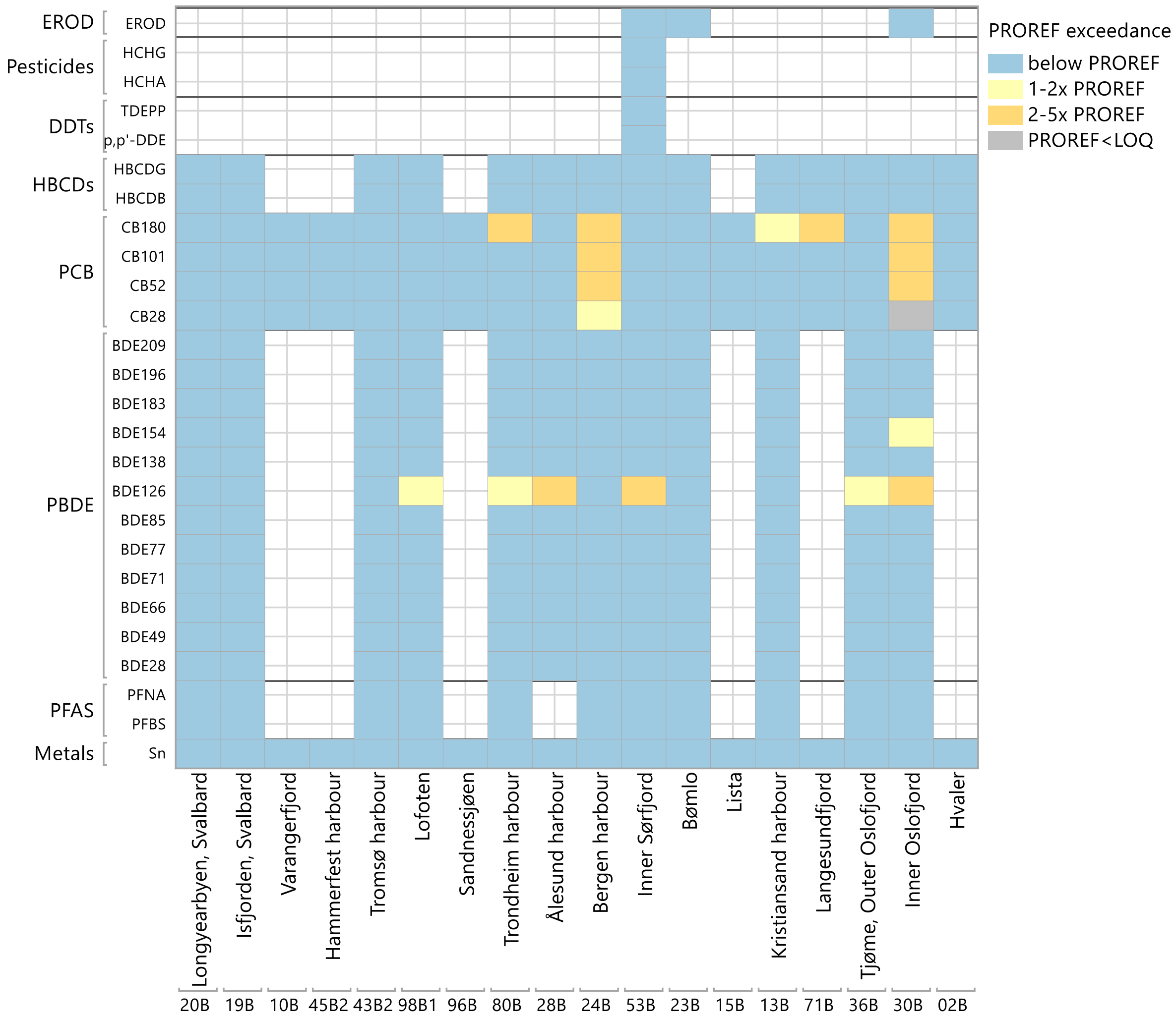

Figure S6. Heatmap of exceedances of PROREF in **cod** for contaminants *not selected* in 2021. The colours represent below or above exceedance of PROREF. Empty "cells" mean that the contaminant was not analysed for at the indicated station. Grey lines show the midpoint of each station and contaminant, and darker lines have been inserted between contaminant groups.

A total of 1412 time trends (combinations of contaminant × station × tissue) have been estimated for contaminants not selected for presentation in 2021. The contaminants are listed in **Table S1**.

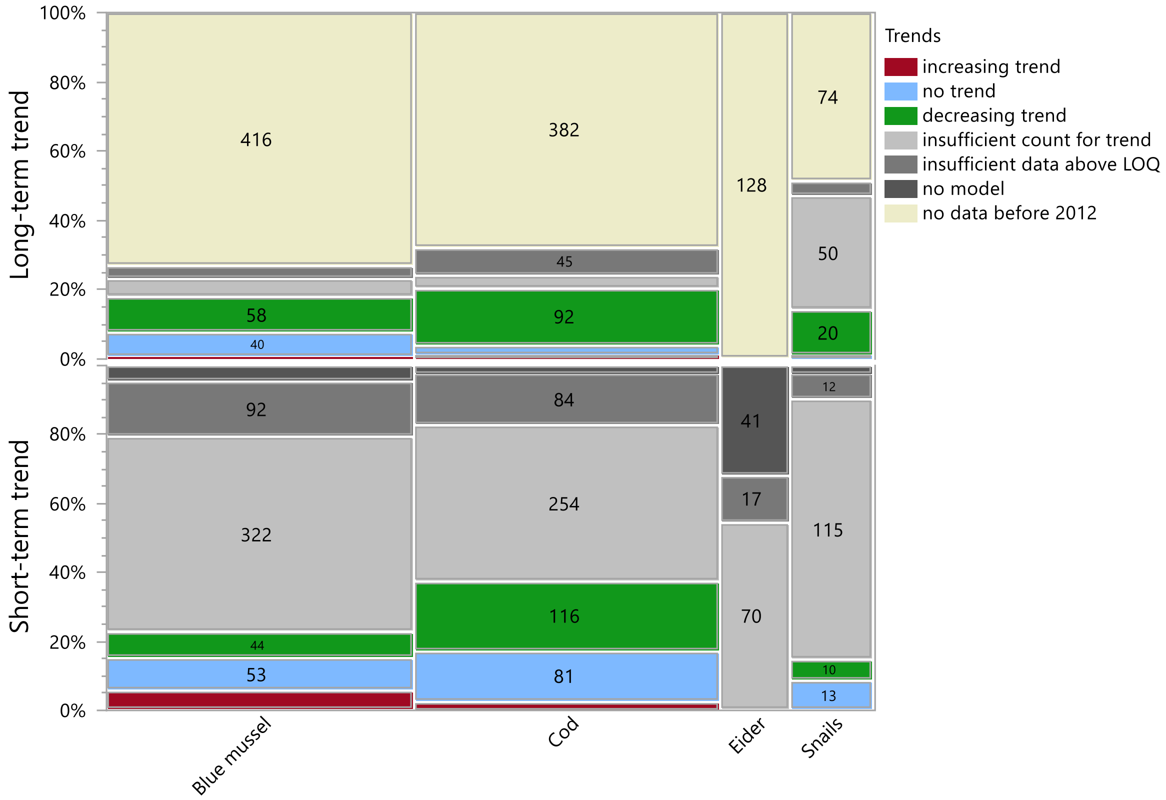

Figure S7. Mosaic plot of time trends for blue mussel, cod eider for contaminants *not selected* in 2021. Upper panel shows long-term trends, while lower panel shows short-term trends. The number of stations/species/tissues are indicated in the respective cells.

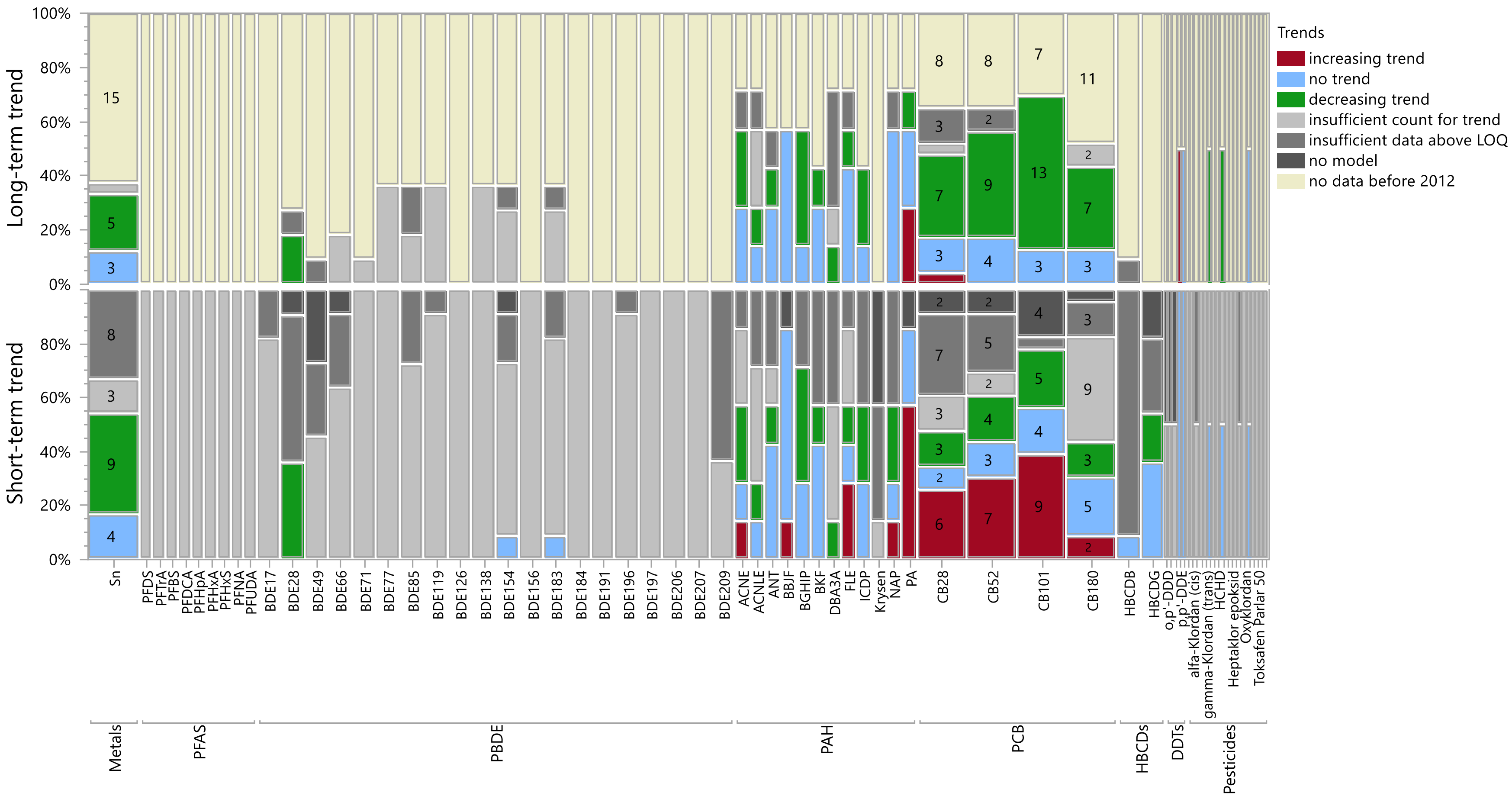

Figure S8. Time trends for **blue mussel** for contaminants not selected in 2021. Upper panel shows long-term trends, while lower panel shows short-term trends. The number of stations is indicated in the respective cells.

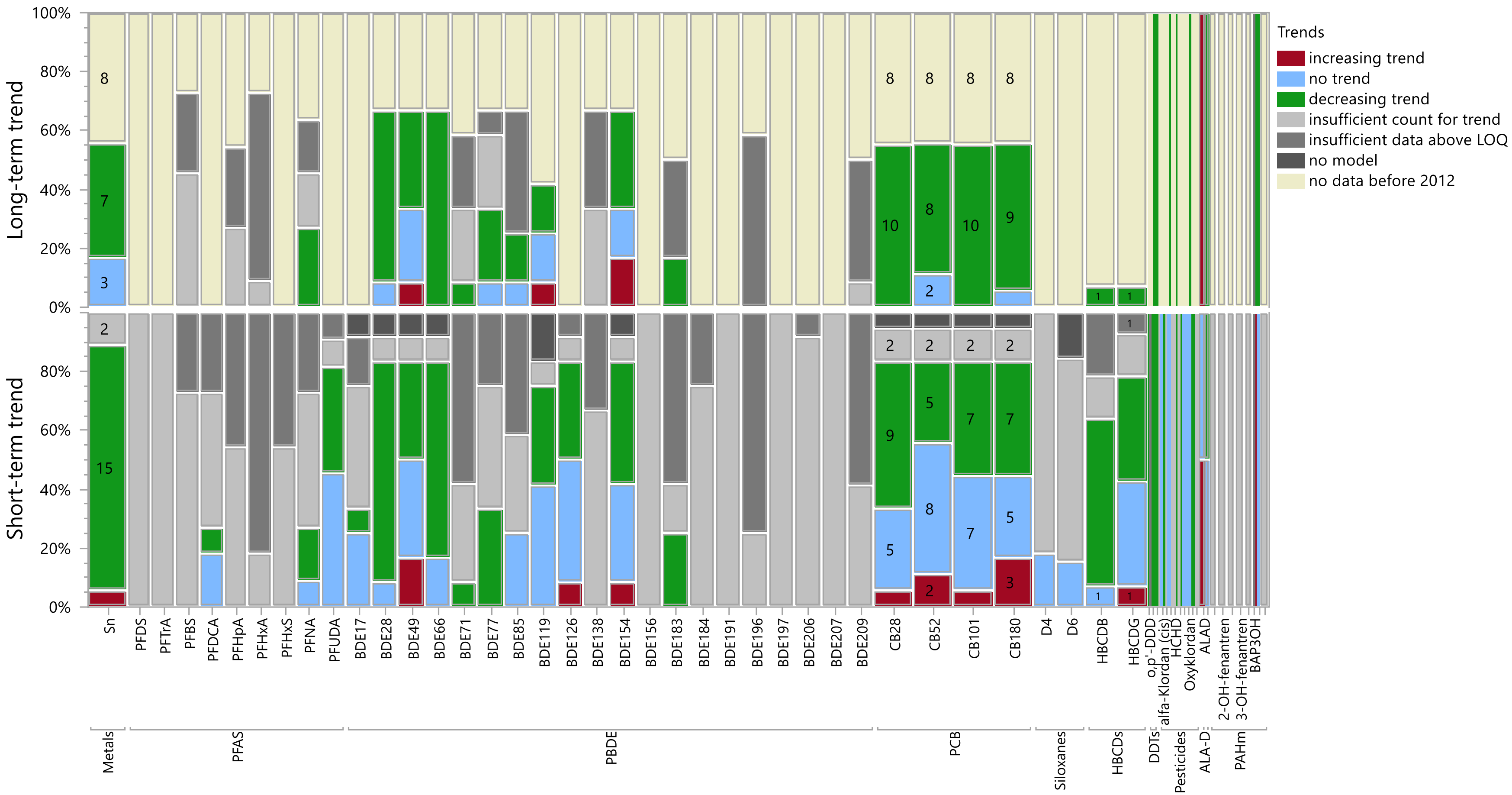

Figure S9. Time trends for **cod** for contaminants *not selected* in 2021. Upper panel shows long-term trends, while lower panel shows short-term trends. The number of stations is indicated in the respective cells.

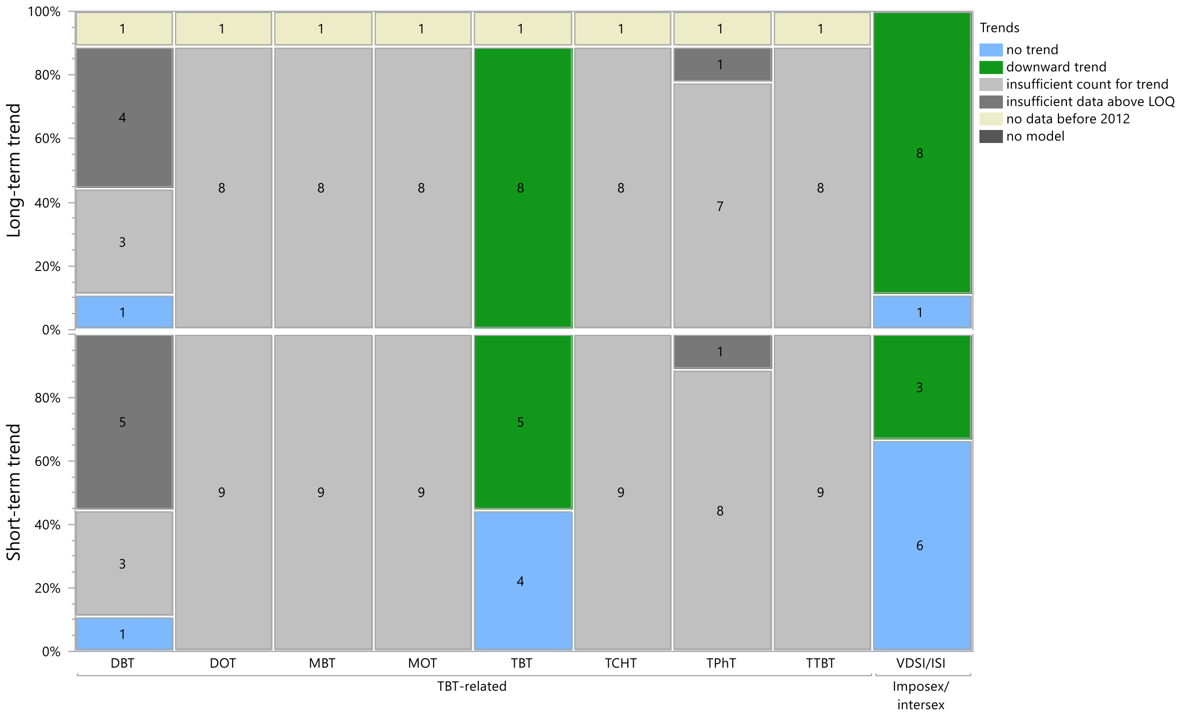

Figure S10. Time trends in **snails** for contaminants *not selected* in 2021. Upper panel shows long-term trends, while lower panel shows short-term trends. The number of stations is indicated in the respective cells.

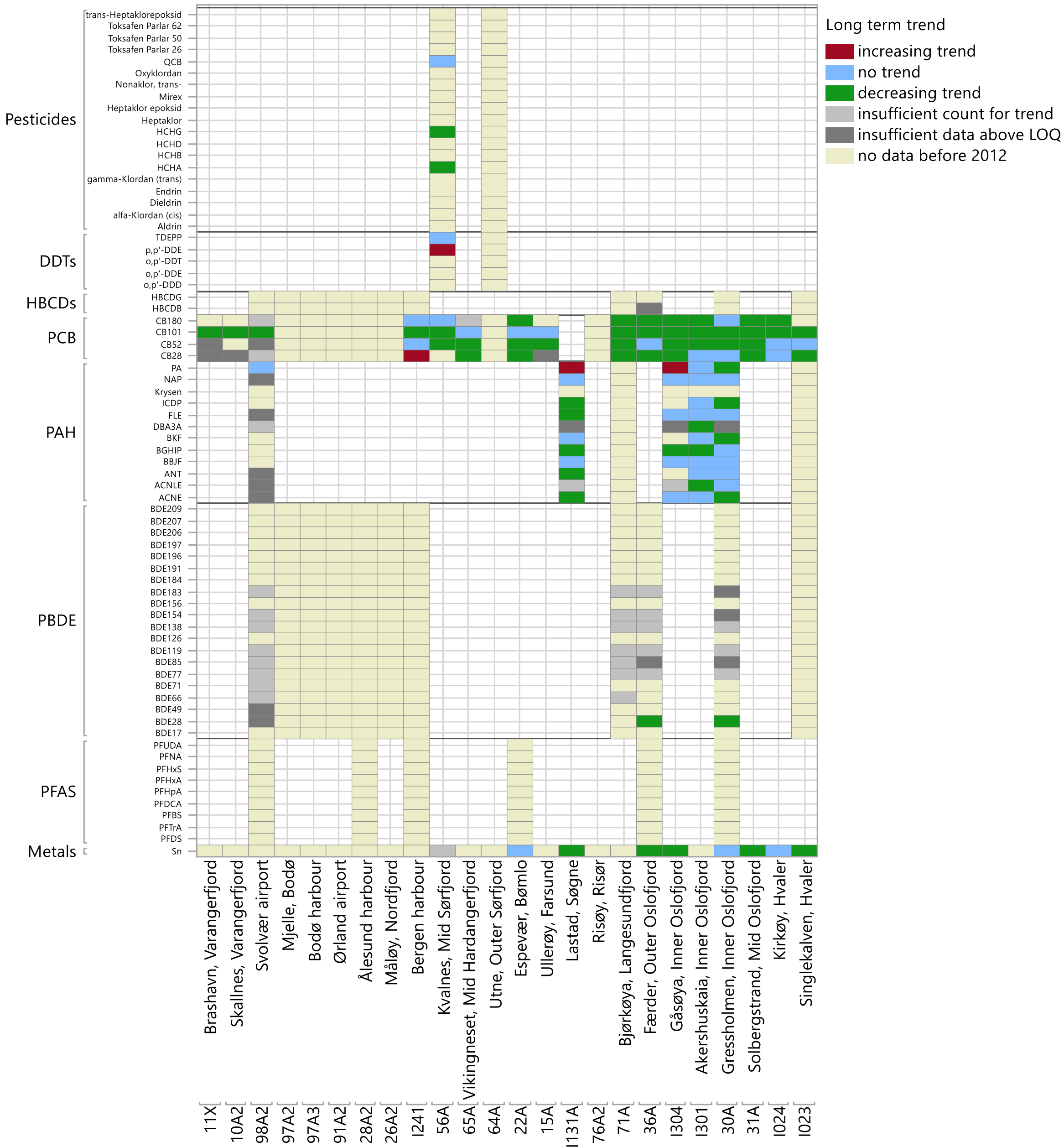

Figure S11. Heatmap for long-term time trends **blue mussel** for contaminants not selected in 2021. The colours represent time trends observed at stations. Empty "cells" mean that the contaminant was not analysed for at the indicated station. Grey lines show the midpoint of each station and contaminant, and darker lines have been inserted between contaminant groups.

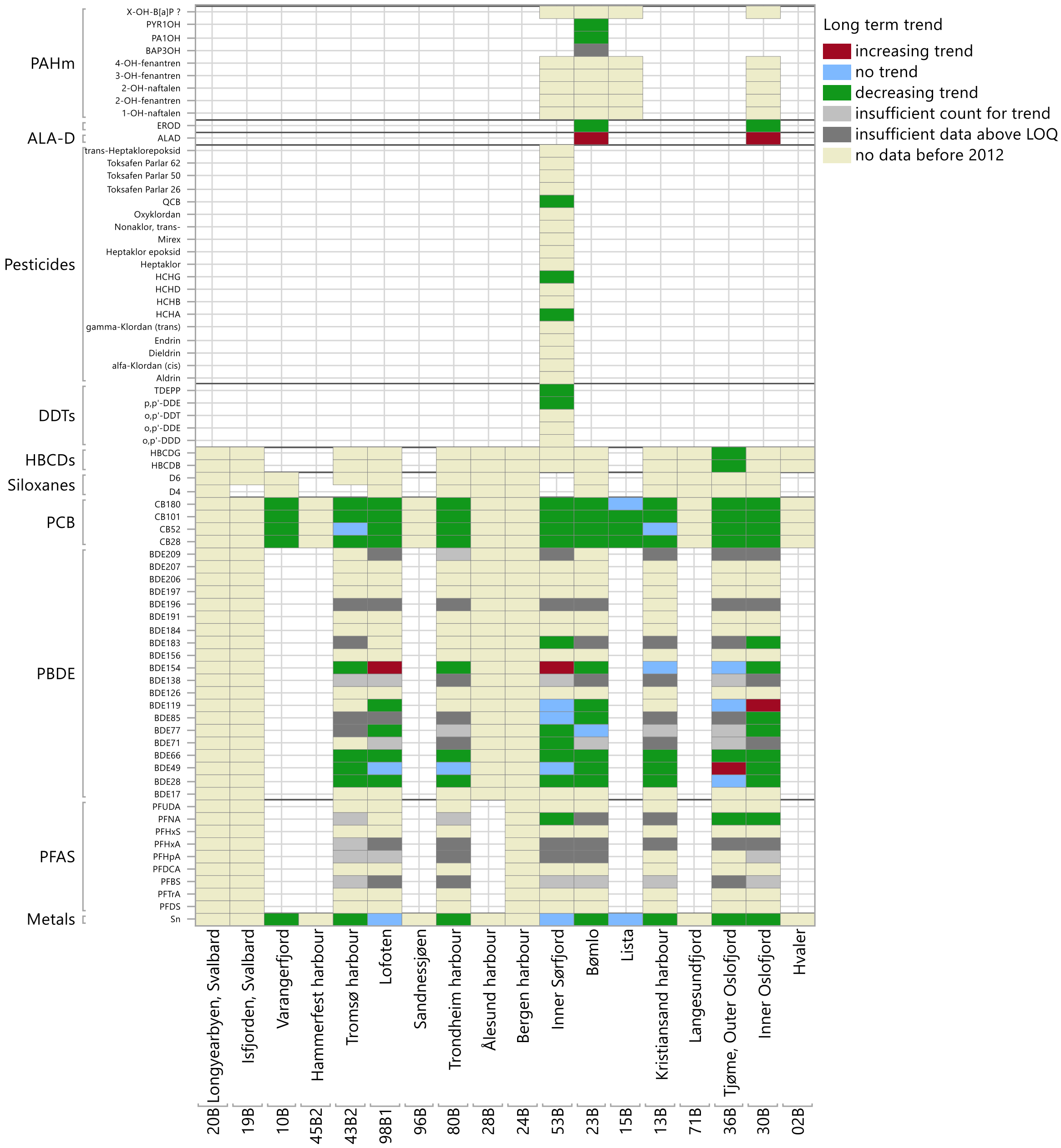

Figure S12. Heatmap for long-term time trends in **cod** for contaminants *not selected* in 2021. The colours represent time trends observed at stations. Empty “cells” mean that the contaminant was not analysed for at the indicated station. Grey lines show the midpoint of each station and contaminant, and darker lines have been inserted between contaminant groups.

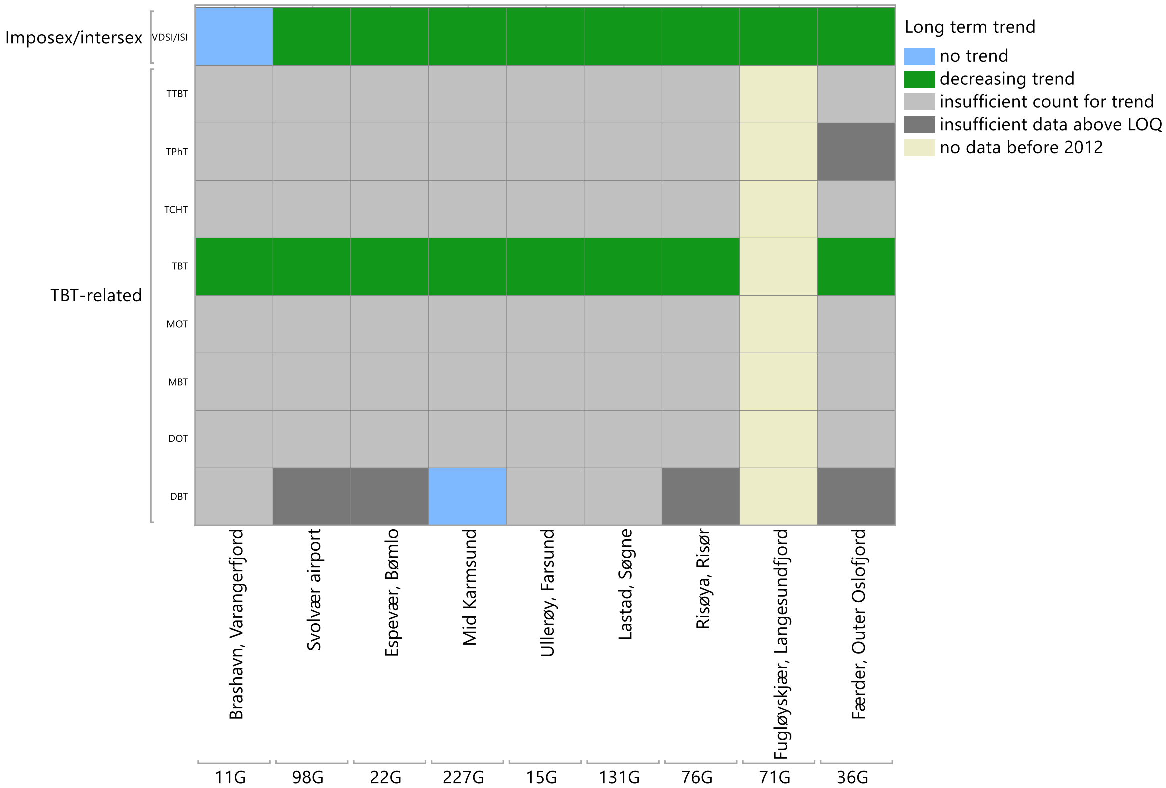

Figure S13. Heatmap for long-term time trends in **snails** for contaminants *not selected* in 2021. The colours represent time trends observed at stations. Empty "cells" mean that the contaminant was not analysed for at the indicated station. Grey lines show the midpoint of each station and contaminant, and darker lines have been inserted between contaminant groups.

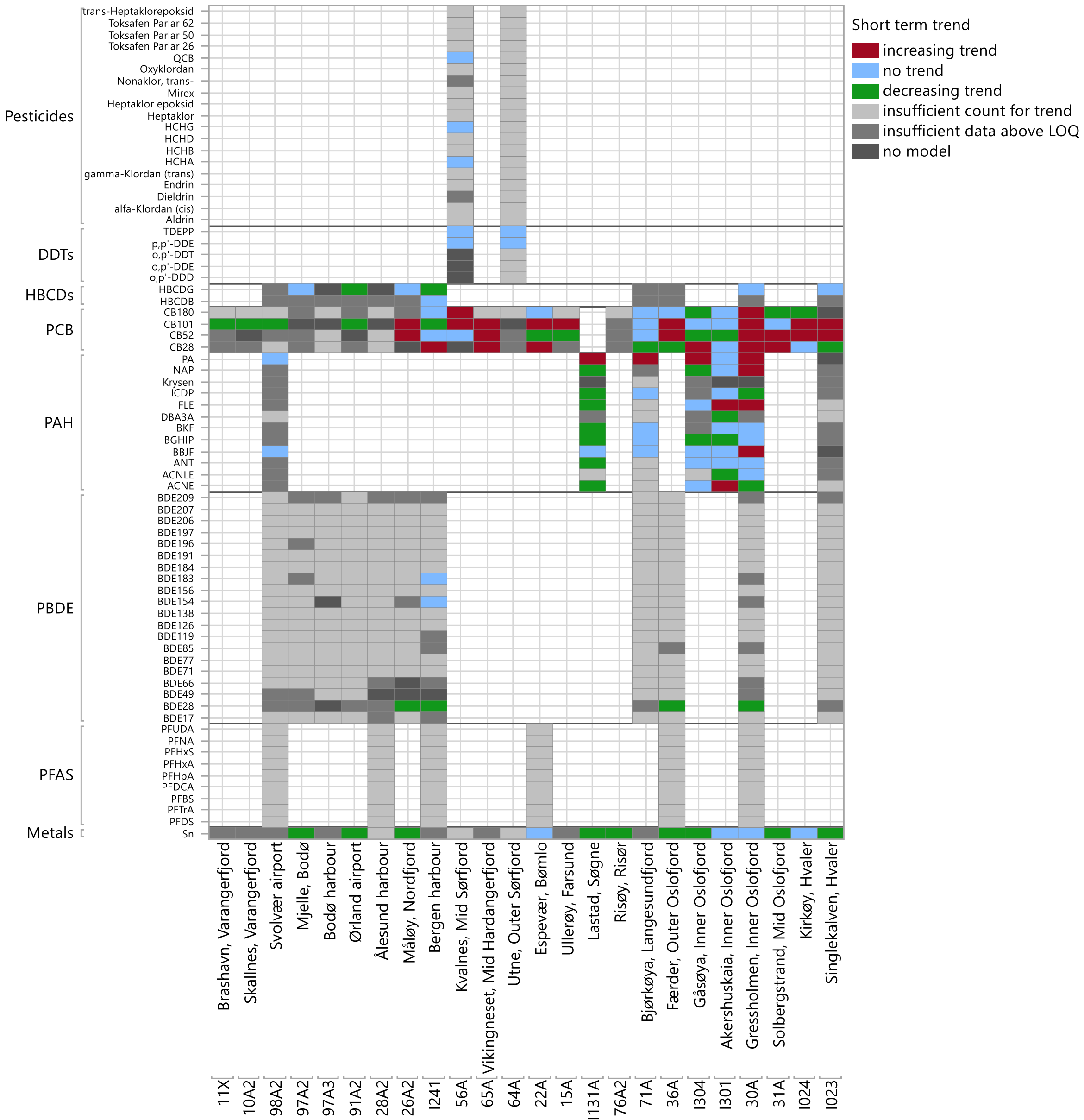

Figure S14. Heatmap for short-term time trends **blue mussel** for contaminants *not selected* in 2021. The colours represent time trends observed at stations. Empty "cells" mean that the contaminant was not analysed for at the indicated station. Grey lines show the midpoint of each station and contaminant, and darker lines have been inserted between contaminant groups.

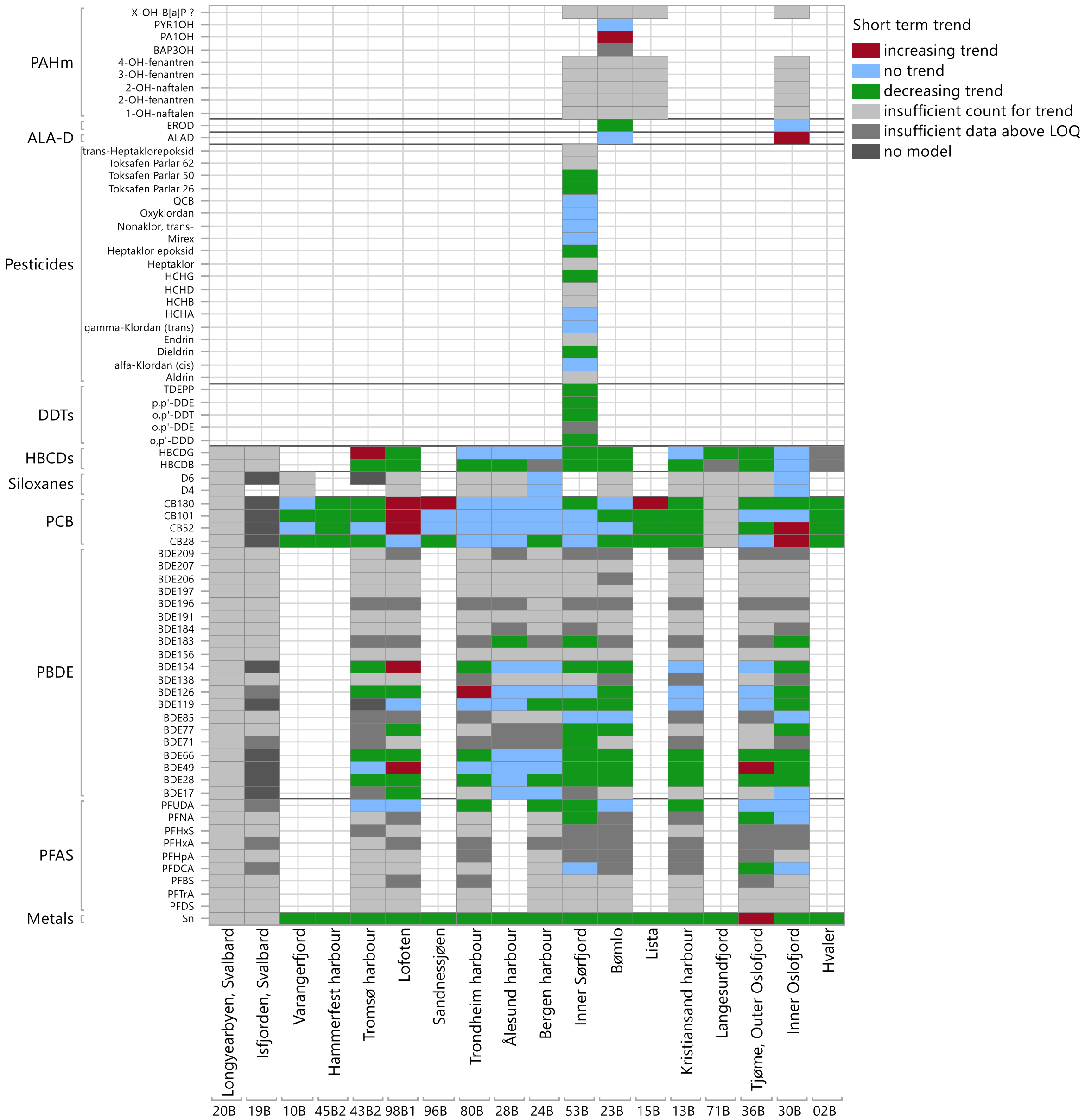

Figure S15. Heatmap for short-term time trends **cod** for contaminants *not selected* in 2021. The colours represent time trends observed at stations. Empty "cells" mean that the contaminant was not analysed for at the indicated station. Grey lines show the midpoint of each station and contaminant, and darker lines have been inserted between contaminant groups.

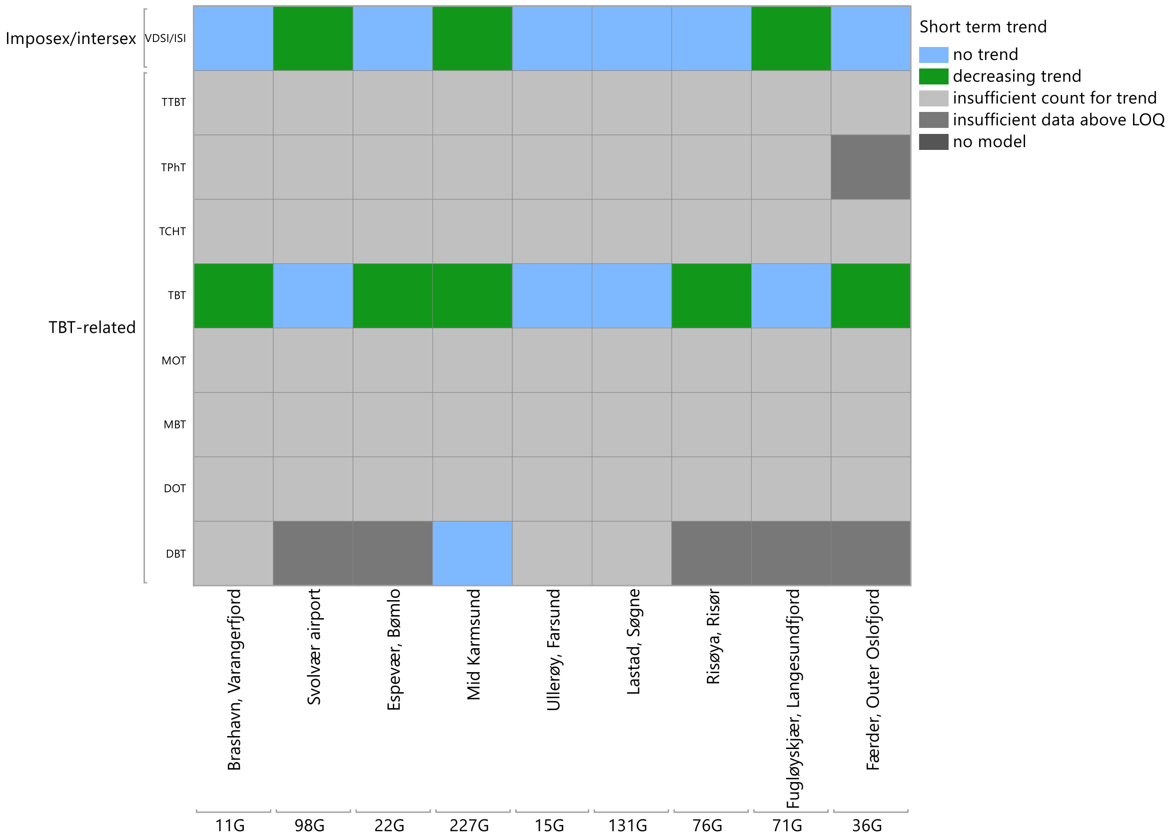

Figure S16. Heatmap for short-term time trends **snails** for contaminants *not selected* in 2021. The colours represent time trends observed at stations. Empty “cells” mean that the contaminant was not analysed for at the indicated station. Grey lines show the midpoint of each station and contaminant, and darker lines have been inserted between contaminant groups.

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