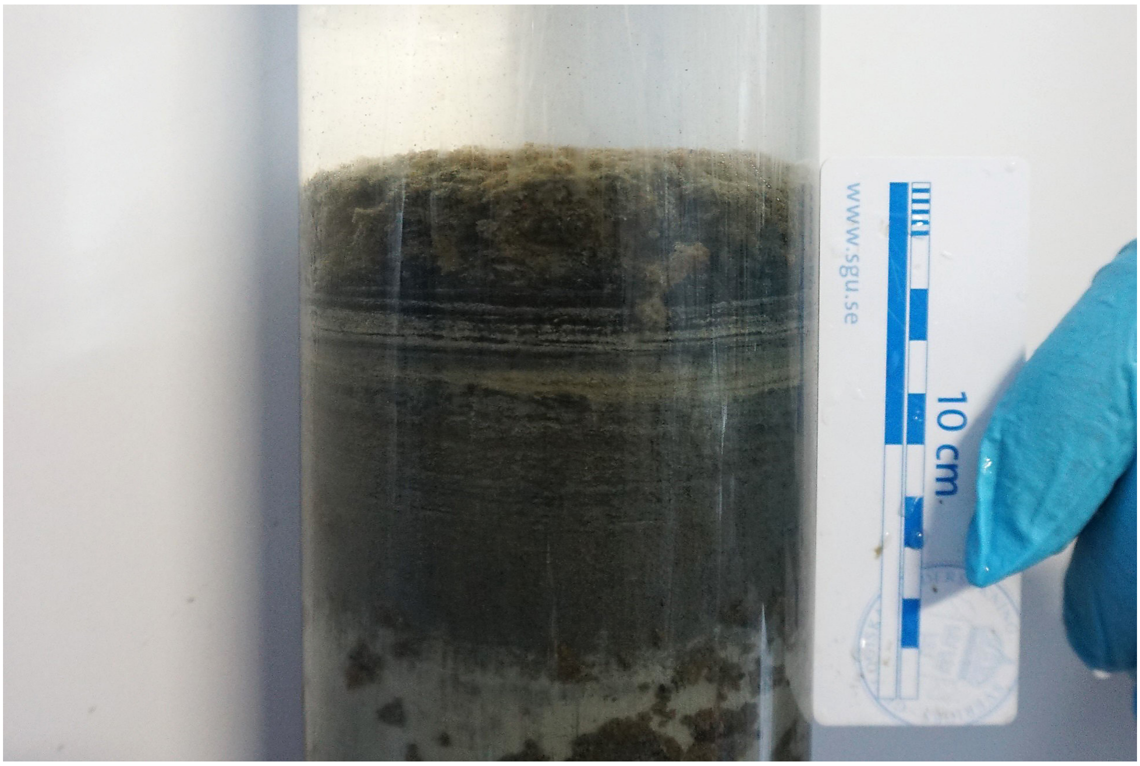
Results from the national environmental monitoring programme

Contaminants in Swedish offshore sediments 2003–2021

Sarah Josefsson

May 2022

SGU-rapport 2022:08
Diarie-nr: 35-1370/2021



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SWEDISH ENVIRONMENTAL PROTECTION AGENCY

NATIONAL ENVIRONMENTAL MONITORING COMMISSIONED BY THE SWEDISH EPA

Swedish Agency for Marine and Water Management

Changes implemented April 21, 2023
Page 51: Second paragraph replaced with updated information.
Page 53: Figure 21 and 22 replaced with updated information.

Front page photo: Sediment core from station SE-9
Photographer: Sarah Josefsson/Olof Larsson

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SWEDISH ENVIRONMENTAL
PROTECTION AGENCY

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ENVIRONMENTAL
MONITORING
COMMISSIONED BY
THE SWEDISH EPA

FILE NO.	NV-08924-19
CONTRACT NO.	213-21-008
PROGRAMME AREA	Coast and sea
SUBPROGRAMME	Contaminants in Swedish offshore sediments

Contaminants in Swedish offshore sediments 2003–2021

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Report title and subtitle Contaminants in Swedish offshore sediments 2003–2021. Results from the national environmental monitoring programme.	**Purchaser** Swedish Environmental Protection Agency, Environmental Monitoring Unit SE-106 48 Stockholm, Sweden **Funding** National environmental monitoring

Keywords for location (specify in Swedish)
Baltic Sea, North Sea, Bothnian Bay, Bothnian Sea, Gulf of Bothnia, Baltic Proper, Kattegat, Skagerrak

Keywords for subject (specify in Swedish)
Sediment, organic contaminants, POPs, elements, metals, sediment accumulation rate

Period in which underlying data were collected
2003–2021

Summary

The environmental monitoring of contaminants in offshore sediments includes organic contaminants and elements in surface sediments at 16 stations located in deeper sea areas around Sweden. The stations are placed far from the coast to reflect general pollution patterns in the environment. So far, sampling has been done four times with intervals of 5–6 years. This report summarizes the results and evaluates geographical trends.

Levels of arsenic and cobalt are highest in the Gulf of Bothnia, while levels of cadmium, copper and zinc are highest in the Baltic Proper. Cadmium levels are so high that the environmental quality standard (EQS) is exceeded. These patterns depend on redox conditions, levels in bedrock/moraine in the catchment, and point sources. Levels of lead and mercury are elevated at specific stations, indicating a greater influence from point sources. Overall, metal levels are elevated compared to reference values that represent pre-industrial levels.

The greatest load of organic contaminants occurs in the Baltic Proper. For most contaminants, for example PAHs, PCBs, organophosphorus compounds, TBT, and many organochlorine pesticides, the highest levels can be observed in this sea area. TOC levels are also high here. Surprisingly, levels of PFASs are markedly higher in the Gulf of Bothnia compared to other sea areas. Levels of chlorinated paraffins, measured in sediment cores from three stations, were also elevated in the Gulf of Bothnia. Levels of TBT have decreased since the start of the monitoring programme, but the EQS is still exceeded at many stations. PAHs also occur at levels that sometimes exceed thresholds. Cybutryne, which is now banned in the EU, has decreased in levels since 2008 and 2014.

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ABSTRACT

This report contains a summary and analysis of data from the environmental monitoring of Swedish offshore sediments. The purpose of the Swedish monitoring and trend programme for pollutants in sediments (SSTMP) is to investigate the load and long-term trends of environmental pollution in offshore sediments within Swedish territorial waters and economic zone. The programme includes sampling and analysis of surface sediments at 16 stations in sea basins off the coast of Sweden. Sampling has taken place four times with intervals of 5–6 years: 2003, 2008, 2014 and 2020/2021. The analyses encompass a variety of substances, including metals, organic contaminants, organic carbon and stable isotopes of carbon and nitrogen.

Metals and other elements

Metals occur naturally in the environment, but levels have increased markedly since the industrial revolution. Levels in sediment have a natural geographical variation dependant on different types of bedrock and moraine within the catchment, but levels also depend on the presence or lack of oxygen in the surface sediments, i.e. redox conditions, as these strongly affect the binding of redox-sensitive metals to the sediment. Due to these factors – anthropogenic impact, local or regional geological variation, and redox conditions combined with sediment properties – levels of metals in sediments vary between the sea area.

Sediments in the Gulf of Bothnia, i.e., the Bothnian Bay and Bothnian Sea, are characterized by elevated levels of arsenic. The levels are probably affected by high levels in the bedrock within the catchment and by one or more point sources. Other elements with high levels in Gulf of Bothnia surface sediments are cobalt, barium, bismuth and manganese.

In the Baltic Proper, cadmium, copper and zinc are found at elevated levels. These metals form weakly soluble sulphides under reducing conditions in sediment, and their precipitation as sulphides is probably a contributing factor to the high levels in sediment in the Baltic Proper, where anoxic conditions prevail in deeper areas. Cadmium levels are also elevated in the Bothnian Bay, where the sediment is oxygenated, and the levels cannot be explained by precipitation as sulphides. Other elements with high levels in Baltic Proper sediments are, for example, antimony, selenium and sulphur, while levels of, for example, aluminium, chromium and lithium are low compared to other monitoring stations.

Levels of mercury and lead are elevated at specific stations rather than in specific sea areas indicating that levels are affected by point sources. Levels are highest at stations SE-1 in the southern Bothnian Bay and SE-12 in the Arkona Basin in the southern Baltic Sea. Lead levels are also high at station SE-11 in the Bornholm Basin. The fact that these toxic metals occur at clearly elevated levels in sediment at specific stations points to the importance of addressing point sources to reduce levels in the Baltic Sea environment, in addition to global efforts to reduce diffuse pollution and long-range atmospheric transport. In comparison with stations in the Baltic Sea, levels of most metals are low in Kattegat and Skagerrak, the North Sea areas.

Cadmium and lead have environmental quality standards (EQS) for sediments within the Water Framework Directive (WFD). These EQS are also proposed as indicator thresholds for the third Helsinki Commission (HELCOM) holistic assessment of the Baltic Sea environment, HOLAS III. The EQS for cadmium was exceeded at some stations in the Baltic Proper during the most recent monitoring in 2020/2021, while the EQS for lead was not exceeded. For copper, an EQS is available in Sweden but it was never exceeded for the sediment sampled in 2020/2021. Copper is proposed to be included as an indicator for sediment in HOLAS III, but with a different proposed threshold than the Swedish EQS. Comparing the proposed threshold for HOLAS III with the measured levels in sediment in 2020/2021, the threshold is exceeded at all monitoring stations except one.

Organic contaminants

Organic contaminants are predominantly of anthropogenic origin, although a few can also be formed in small amounts in natural processes, for example PCDD/Fs and PAHs in forest fires. Some substances can be transported long distances in the atmosphere, while other substances are mainly emitted to the Baltic Sea via the aquatic environment.

Most of the investigated organic contaminants occur at the highest levels in the Baltic Proper, including the southern parts of the Baltic Sea. This is the case for, for example, $\sum PAH_{16}$, HBCDD, organophosphorus compounds, PCBs, TBT, cybutryne, ∑HCH, chlordanes, chlorinated aliphatic hydrocarbons, phthalates, octyl- and nonylphenols, BTEX, and DDE and DDD which are degradation products of DDT. Kattegat and Skagerrak generally have the lowest levels of contaminants, although the levels of some substance groups, for example PAHs, are elevated here compared to the Gulf of Bothnia. The generally higher contaminant levels in the Baltic Sea are expected due to the semi-enclosed nature and relatively high population density within its catchment.

Many organic contaminants sorb to organic carbon, and because the anoxic stations in the Baltic Proper have high TOC levels it is difficult to determine whether the high contaminant levels in sediment are caused by sorption to TOC or by a high contaminant load to the sea area. TOC levels in sediment are affected by redox conditions, with a slower degradation rate in anoxic conditions. The relation between levels of contaminants and TOC in the water column is therefore not necessarily reflected in the sediment.

The levels of some substances, for example PCBs and the organochlorine pesticides, are elevated relative to TOC levels in the southern Baltic Sea, in particular at station SE-12. The levels of many substances, for example PAHs and degradation products of DDT, in the Gulf of Bothnia are however generally lower than what the correlation with TOC predicts.

PFASs occur at surprisingly high levels in the northern part of the Baltic Sea, with on average 14 times higher PFAS levels at Bothnian Bay and Bothnian Sea stations compared to stations in other sea areas. PFASs were included in the monitoring programme in 2014 and showed very elevated levels in the Gulf of Bothnia. This seemed unreasonable as levels of organic contaminants in sediment are generally lower in the Gulf of Bothnia than in the Baltic Proper, and studies of PFASs in water and biota point to lower concentrations in the northern than in the southern parts of the Baltic Sea. Monitoring in 2020/2021 however continued to show markedly higher levels in Gulf of Bothnia sediments. Further investigations are warranted to elucidate the reason behind this.

Chlorinated paraffins also occur at high levels in Gulf of Bothnia sediments. In 2020/2021, sediment cores from three of the monitoring stations were analysed by researchers from Stockholm University. Levels were substantially higher at station SE-1 in the Bothnian Bay compared to SE-5 in the Baltic Proper and SE-13 in the North Sea. The reason for the high levels at SE-1 has been suggested to be emissions from e-waste facilities. More studies are however needed for source identification and to elucidate, for example, the difference in proportion of the different chlorinated paraffins between coastal and offshore sediments.

Levels of the biocide TBT have fallen since the start of the monitoring programme in 2003 and are now below the laboratory reporting limit at some stations. The EQS is however still exceeded at many stations, not only in the Baltic Proper but also in the Bothnian Bay and the North Sea. Another biocide now banned within the EU, cybutryne, also seems to decrease in levels over time and now only occurs above its so-called indicative value in sediment at one station in the Baltic Proper.

PAHs sometimes occur at levels that exceed environmental thresholds. The EQS for anthracene is exceeded at two stations in the North Sea in 2020/2021, while the EQS for fluoranthene has never been exceeded within the monitoring programme for offshore sediments. Of the four PAHs for which there are indicative values, levels in 2020/2021 exceeded values primarily at stations in the North Sea and the Southern Baltic Sea. These are thus the most problematic areas in terms of potentially toxic PAH levels in sediments.

PBDEs are a group of substances that often greatly exceed their EQS in biota in the aquatic environment. There is no EQS for sediments but the proposed indicator threshold for sediment for HOLAS III is not exceeded at any of the stations. The levels of PBDE in sediment are similar between the sea areas indicating that atmospheric deposition or other diffuse sources are likely to be important sources.

SAMMANFATTNING

Denna rapport innehåller en sammanfattning och analys av data från den nationella miljöövervakningen av föroreningar i svenska havssediment. Syftet med *Swedish sediment trend and monitoring programme* (SSTMP) är att undersöka belastning och trender av miljöföroreningar i utsjösediment inom svenskt territorialvatten och ekonomisk zon. I programmet ingår provtagning och analys av ytsediment på 16 stationer på djupa bottnar runt Sverige. Provtagning har hittills skett fyra gånger med 5–6 års intervall: 2003, 2008, 2014 och 2020/2021. Analyserna omfattar en mängd olika ämnen, inklusive metaller, organiska föroreningar, organiskt kol och stabila isotoper av kol och kväve.

Metaller och andra grundämnen

Metaller förekommer naturligt i miljön men halterna har ökat markant sedan den industriella revolutionen. Metallhalterna i sediment har en naturlig geografisk variation. Detta beror på förekomsten av metallerna i berggrund och morän inom avrinningsområdet, men också på syreförhållandena i ytsedimenten, det vill säga redoxförhållanden, eftersom dessa påverkar bindningen av redoxkänsliga metaller till sedimentet. På grund av dessa faktorer – antropogen inverkan, lokal eller regional geologisk variation och redoxförhållanden i kombination med sedimentegenskaper – varierar halterna av metaller i sediment mellan havsområdena.

Sediment i Bottniska viken, det vill säga Bottenviken och Bottenhavet, kännetecknas av förhöjda halter av arsenik. Detta beror sannolikt på höga halter i berggrunden inom avrinningsområdet och på utsläpp från en eller flera punktkällor. Andra grundämnen med höga halter i ytsediment i Bottniska viken är kobolt, barium, vismut och mangan.

I Egentliga Östersjön förekommer kadmium, koppar och zink i förhöjda halter. Dessa metaller bildar svårlösliga sulfider under reducerande förhållanden i sediment. Att de så att säga faller ut som sulfider är sannolikt en bidragande orsak till de höga halterna i ytsediment i Egentliga Östersjön, där syrefria förhållanden råder på djupare bottnar. Kadmiumhalterna är också förhöjda i Bottenviken, där sedimentet emellertid är syresatt och halterna därför inte borde bero på utfällning som sulfider. Andra grundämnen som förekommer i höga halter i Egentliga Östersjöns syrefria sediment är till exempel antimon, selen och svavel, medan halterna av till exempel aluminium, krom och litium är låga jämfört med andra stationer.

Halterna av kvicksilver och bly är förhöjda på specifika stationer snarare än i specifika havsområden, vilket tyder på att nivåerna påverkas av punktkällor. Halterna är högst på stationerna SE-1 i södra Bottenviken och SE-12 i Arkonabassängen i södra Östersjön. Även på SE-11 i Bornholmsbassängen är blyhalterna förhöjda. Att dessa giftiga metaller förekommer i förhöjda nivåer i sediment på specifika stationer pekar på vikten av att ta itu med punktkällor för att kunna minska halterna i Östersjöns miljö, utöver globala ansträngningar för att minska diffusa källor och långväga lufttransport. I jämförelse med stationerna i Östersjön är halterna av de flesta metaller låga i Kattegatt och Skagerrak på svenska västkusten.

För kadmium och bly finns gränsvärden för sediment inom EU:s ramdirektiv för vatten (WFD). Dessa gränsvärden föreslås också användas som så kallade indikatortrösklar för HELCOM:s tredje holistiska bedömning av Östersjön, HOLAS III. Värdet för kadmium överskreds på några övervakningsstationer i Egentliga Östersjön under den senaste provtagningen 2020/2021, medan värdet för bly inte överskreds. För koppar finns en bedömningsgrund som gäller i Sverige men den överskreds aldrig vid provtagningen 2020/2021. Koppar föreslås ingå som indikator för sediment i HOLAS III, men med en annan föreslagen tröskel än den svenska bedömningsgrunden. Om man jämför det föreslagna tröskelvärdet för HOLAS III med de uppmätta halterna i sediment 2020/2021 överskrids tröskeln vid alla övervakningsstationer utom en.

Organiska föroreningar

Organiska föroreningar har till övervägande del antropogent ursprung även om ett fåtal också kan bildas i små mängder i naturliga processer, till exempel PCDD/F och PAH vid skogsbränder. Vissa ämnen kan transporteras långa sträckor i atmosfären medan andra ämnen huvudsakligen släpps ut till Östersjön via vattenmiljön.

De flesta av de undersökta organiska föroreningarna förekommer i högst nivåer i Egentliga Östersjön, inklusive de södra delarna av Östersjön. Detta är fallet för till exempel $\sum PAH_{16}$, HBCDD, organofosfater, PCB, TBT, cybutryn, ∑HCH, klordaner, klorerade alifatiska kolväten, ftalater, oktyl- och nonylfenoler, BTEX samt DDE och DDD vilka är nedbrytningsprodukter av DDT. Kattegatt och Skagerrak har generellt de lägsta föroreningshalterna även om en del ämnen, till exempel PAH, har förhöjda halter här jämfört med Bottniska viken. Att det är högre halter av föroreningar i Östersjön än på västkusten är förväntat på grund av den begränsade vattenomsättningen och den relativt höga befolkningstätheten inom avrinningsområdet.

Många organiska föroreningar binder till organiskt kol. Eftersom de syrefattiga (anoxiska) stationerna i Egentliga Östersjön har höga TOC-halter är det svårt att avgöra om de höga föroreningshalterna här beror på bindning till TOC eller på en hög föroreningsbelastning. TOC-halterna påverkas av redoxförhållandena i sediment, med en långsammare nedbrytningshastighet under anoxiska förhållanden. Förhållandet mellan halter av föroreningar och TOC i vattenmassan återspeglas därför inte nödvändigtvis i sedimentet.

Halterna av vissa ämnen, till exempel PCB och klorerade bekämpningsmedel, är förhöjda i förhållande till TOC-halterna i södra Östersjön, särskilt vid station SE-12. Halterna i Bottenviken är däremot generellt lägre än vad korrelationen till TOC förutspår, till exempel för PAH och nedbrytningsprodukterna av DDT.

PFAS förekommer i förvånansvärt höga halter i norra delen av Östersjön, med i genomsnitt 14 gånger högre PFAS-halter i Bottenviken och Bottenhavet jämfört med andra havsområden. PFAS ingick i övervakningsprogrammet 2014 och visade även då kraftigt förhöjda halter i Bottniska viken. Detta verkade orimligt eftersom halterna av organiska föroreningar i sediment generellt är lägre i Bottniska viken än i Egentliga Östersjön, och studier av PFAS i vatten och biota pekar på lägre halter i norra än i södra Östersjön. Övervakningen 2020/2021 visade dock på fortsatt klart högre halter i sediment i Bottniska viken. Ytterligare undersökningar behövs för att förstå orsaken till de höga halterna här.

Klorparaffiner förekommer också i höga halter i Bottenviken. Under 2020/2021 analyserades sedimentkärnor från tre av övervakningsprogrammets stationer av forskare på Stockholms universitet. Halterna var betydligt högre på station SE-1 i Bottenviken jämfört med SE-5 i Egentliga Östersjön och SE-13 i Kattegatt. De höga halterna vid SE-1 har föreslagits bero på utsläpp från e-avfallsanläggningar. Fler studier behövs dock för att kunna identifiera källor och för att förstå till exempel skillnaden i förhållandet mellan de olika klorparaffinerna mellan kust- och utsjösediment.

Halterna av biociden TBT har sjunkit sedan övervakningsprogrammet startade 2003 och ligger nu under rapporteringsgränsen på en del stationer. Gränsvärdet för sediment överskrids dock fortfarande på många stationer, inte bara i egentliga Östersjön utan även i Bottenviken och i Nordsjön. En annan biocid som nu är förbjuden inom EU, cybutryn, tycks också minska i halter och förekommer nu över sitt så kallade indikativa värde i sediment endast vid en station i Egentliga Östersjön.

PAH förekommer ibland i halter som överskrider gränsvärden. Gränsvärdet för antracen överskrids i sediment på två stationer i Nordsjön 2020/2021, medan gränsvärdet för fluoranten aldrig har överskridits inom övervakningsprogrammet. För de fyra PAH som det finns så kallade indikativa värden för översteg halterna 2020/2021 dessa värden främst vid stationer i Nordsjön och södra Östersjön. Dessa är alltså de mest problematiska områdena vad gäller potentiellt giftiga halter av PAH i sediment.

PBDE är en grupp ämnen som ofta kraftigt överskrider sitt gränsvärde i biota i den akvatiska miljön. Det finns inga svenska bedömningsgrunder för sediment men den föreslagna indikatortröskeln för HOLAS III överskrids inte på någon av stationerna. Nivåerna av PBDE i sediment är likartade mellan havsområdena, vilket pekar på att atmosfäriskt nedfall eller andra diffusa källor sannolikt är viktiga källor.

INTRODUCTION

The bottom of seas, lakes and watercourses are made up of sediments that form an important and dynamic part of the aquatic environment. Sediments in accumulation areas, where the flow rate of water is low enough that fine-grained material settles, constitute potential sinks for contaminants. Many environmental contaminants, both organic substances and metals, bind to particles in the aquatic environment and then settle with the particles to the bottom. New sediment layers are continuously formed while old layers are largely buried and preserved. The sediments thus function as an archive of the levels of pollutants in the environment.

This report contains a summary and analysis of data from the environmental monitoring of Swedish offshore sediments. Data has been collected within the national Swedish monitoring and trend programme for pollutants in sediments (SSTMP), financed by the Swedish Environmental Protection Agency (SwEPA). In 2020/2021, additional funding was provided by the Swedish Agency for Marine and Water Management (SwAM). Sampling and reporting are performed by the Geological Survey of Sweden (SGU). The purpose of the programme is to investigate the load and long-term trends of environmental pollution in offshore sediments within Swedish territorial waters and economic zone. The programme includes sampling and analysis of surface sediments at 16 stations in sea basins off the coast of Sweden. Sampling has so far taken place on four occasions: 2003, 2008, 2014 and 2020/2021. The analyses encompass a variety of substances, including metals, organic contaminants, organic carbon and stable isotopes of carbon and nitrogen.

Results from the first three monitoring years were presented in English in SGU-report 2016:04, *Chemical contamination in offshore sediments 2003–2014*, where the results are reported for each station (Apler & Josefsson 2016). Results from the first three monitoring years have also been presented in a report in Swedish where results are reported for each contaminant or contaminant group (Josefsson & Apler 2019). This report builds on the 2019 report, but with the addition of new substances as well as results from 2020/2021. The purpose of this report is to account for the geographical variation of pollution levels and, if possible, time trends. The latter is not always possible because some substances have not been analysed all years, and analytical methods have varied. More information about the SSTMP programme, sampling and analyses can be found in SGU-report 2016:04. Chemical data can be accessed on SGUs website (www.sgu.se) and is reported to ICES DOME for further use internationally within, for example, OSPAR and HELCOM.

SAMPLING AND ANALYSES

Stations and sampling

The environmental monitoring of contaminants in Swedish offshore sediments consists of 16 stations situated in deep accumulation areas, both in the Baltic Sea (13 stations) and on the west coast of Sweden (3 stations; Fig. 1, Table 1). The top layer of sediment in accumulation areas contains the particles and contaminants that have deposited during the last years. In the environmental monitoring programme, the top 1 centimetre of the sediment is sampled. Because the sediment accumulation rate differs between stations, this depth corresponds to different time frames, for example around 6 years per centimetre at the stations in the northernmost sea basin, the Bothnian Bay, and around 2 years per centimetre at the stations in the Bothnian Sea (see Table 2). Since stations are situated in offshore areas, they are assumed to be unaffected by specific point sources. Instead, they should reflect the general environmental situation in the sea basin. Large point sources can however also affect contaminant levels in offshore areas, not least in semi-enclosed areas such as the Baltic Sea.

The stations are located in the various Swedish sea areas, see Table 1. Station SE-4 in the Sea of Åland is considered as part of the Bothnian Sea. The Baltic Proper, which encompasses a large geographical area, has been divided into the Baltic Proper and the Southern Baltic. The stations SE-11 and SE-12 thus belong in the Southern Baltic. The stations on the Swedish west coast, SE-13 in Kattegat, and SE-15 and SE-16 in Skagerrak, are grouped together as North Sea stations although Kattegat hydrographically is not part of the North Sea.

The monitoring surveys have taken place in 2003, 2008, 2014 and 2020/2021. Out of the samples taken in 2020, samples for organic contaminants from stations SE-5 to SE-16 had been kept in malfunctioning freezers and were exposed to temperatures >0ºC for several weeks. Stations SE-6 to SE-16 were thus revisited in 2021 to sample organic contaminants, while it was not possible to revisit SE-5.

The stations consist of a circle with a radius of 50 m. Sampling takes place at seven locations during each sampling: in the centre and at six randomly selected locations within the circle. The surface sediment (0–1 cm) is sampled with a GEMAX sediment corer. Before 2014, a Gemini sampler was used. At stations with low sediment accumulation rate and a thin layer of more unconsolidated, water-rich sediment it is more difficult to get intact surface samples. For example, results from station SE-9, which is the station with the lowest sediment accumulation rate, often display a higher uncertainty as shown by relative standard deviations (see figures for organic carbon and elements).

For elements, all seven samples from each station are analysed. The first monitoring survey in 2003 is an exception as only one sample per station was analysed for elements. For the analyses of organic contaminants, samples from the seven locations are pooled into one sample per station. The reasons for doing this are the high analytical costs and the large amount of sediment required for the analysis of organic contaminants.

Before sampling, the amount of dissolved oxygen in the water above the sediment surface is measured using an oxygen sensor/CTD instrument. This also measures oxygen, conductivity, temperature and depth throughout the water column. Photographs are taken of the sediment cores after they are retrieved to the ship deck. Sediment surface photographs from the stations are available from previous surveys except for SE-8, which was located too deep for the camera equipment. In 2020, sediment was sampled for the Swedish Radiation Safety Authority for monitoring of radioactive substances and for Gothenburg University for analysis of microlitter. These results are not included in this report.

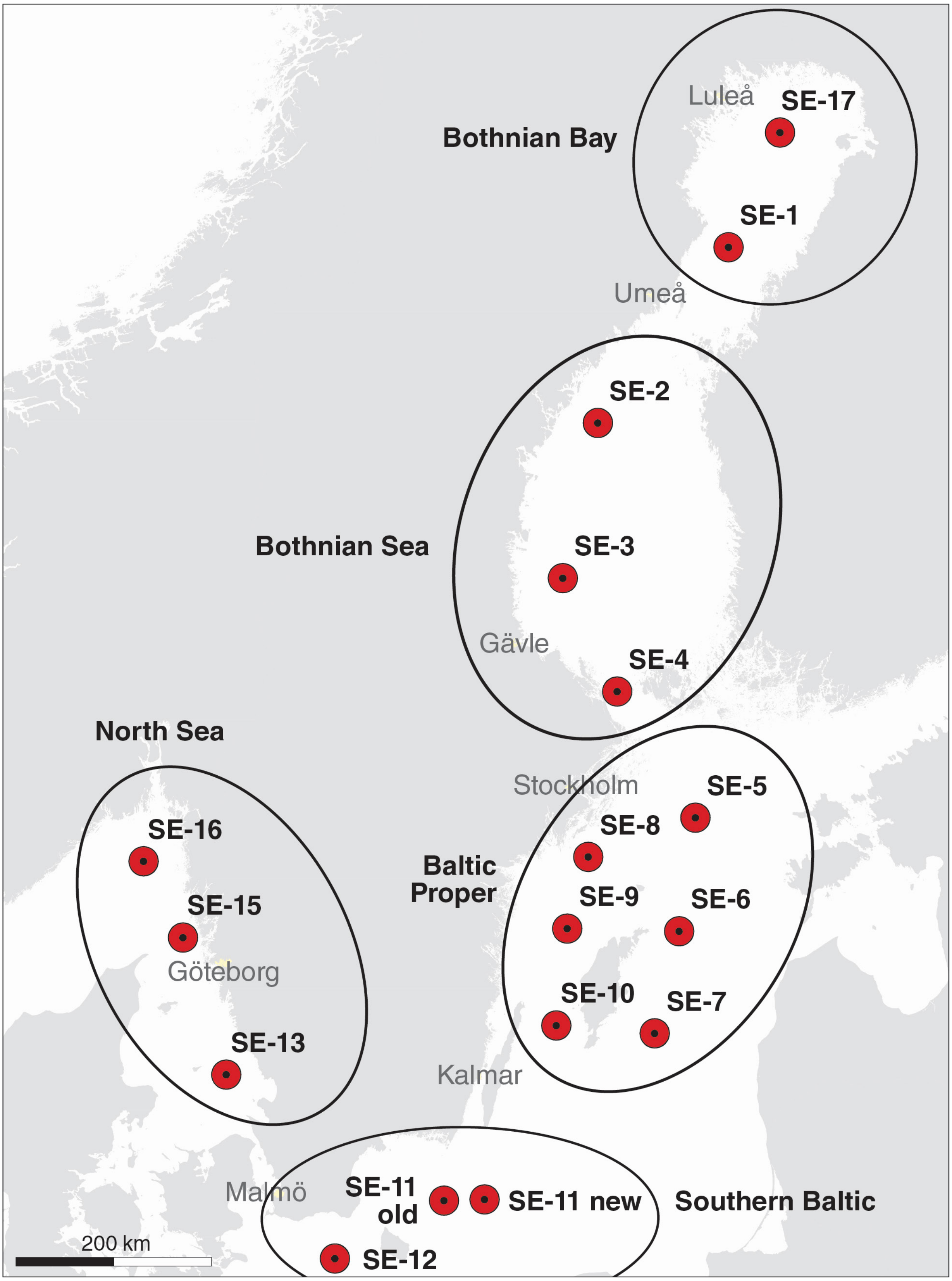

Figure 1. The national monitoring of offshore sediments takes place at 16 stations around the coast of Sweden. Station SE-11 was moved between 2008 and 2014 due to the risk of disturbance from construction of the gas pipeline Nord Stream. It is still situated in the same sea basin, the Bornholm basin. The stations are circled to show which sea area they are considered to belong to; see Table 1 for more information. The map is modified from Apler & Josefsson 2016.

Table 1. The 16 stations are located in deeper accumulation areas around Sweden. Station SE-11 was moved between 2008 and 2014 due to the risk of disturbance from construction of the gas pipeline Nord Stream. The table is modified from Table 1 in Apler & Josefsson 2016.

Station	Sea basin	Sea area	N SWEREF 99TM	E SWEREF 99TM	Depth (m)
SE-17	N Bothnian Bay	Bothnian Bay	7255864	892219	87
SE-1	S Bothnian Bay	Bothnian Bay	7137579	839033	113
SE-2	Härnösand Deep	Bothnian Sea	6956651	704410	200
SE-3	S Bothnian Sea	Bothnian Sea	6796706	668233	79
SE-4	Åland Deep	Bothnian Sea	6679643	724318	230
SE-5	NE Gotska Sandön	Baltic Proper	6549635	804899	175
SE-6	Fårö Deep	Baltic Proper	6432633	788137	195
SE-7	SE Gotland Basin	Baltic Proper	6327222	763118	173
SE-8	Landsort Deep	Baltic Proper	6509084	694601	403
SE-9	Norrköping Deep	Baltic Proper	6435238	672991	178
SE-10	Karlsö Deep	Baltic Proper	6335390	661492	111
SE-11 old	N Bornholm Basin	Southern Baltic	6155024	545712	75
SE-11 new	N Bornholm Basin	Southern Baltic	6156130	587700	70
SE-12	Arkona Basin	Southern Baltic	6095142	433358	47
SE-13	S Rödebank (Kattegat)	North Sea	6284719	321401	47
SE-15	Deep Trench (Skagerrak)	North Sea	6426119	276793	94
SE-16	E Skagerrak	North Sea	6504578	236241	197

Contaminants and analyses

The substances that have been analysed during the four different monitoring years have varied slightly. This report presents most of the organic pollutants but only nine analysed elements in detail. These are the elements for which there are so-called reference values, representing pre-industrial levels, available from the SwEPA (1999). An overview of other elements is given using multivariate statistics.

Organic pollutants were analysed at Analytica AB (today ALS Scandinavia AB) in 2003 and at the Swedish Environmental Research Institute (IVL) from 2008 and onwards. Analyses were performed with different methods for some substances in 2003 compared to the later years. In general, the organic contaminants are extracted from the sediment with Soxhlet or other extraction techniques and final determination is performed by gas chromatography (GC) combined with an electron capture detector (ECD) or mass spectrometry (MS), or high resolution liquid chromatography (HPLC) combined with a fluorescence detector, MS or other techniques. Information about analytical methods for each substance is available in the SGU database.

Elements were analysed at the same laboratory all four monitoring years (ALS Scandinavia AB, which in 2003 and 2008 was Analytica AB). For analysis of arsenic (As), cadmium (Cd) and mercury (Hg), the sample was leached with 7 M nitric acid (HNO_3) which corresponds to a partial digestion. For cobalt (Co), copper (Cu), lead (Pb) and zinc (Zn), a total digestion of the sediment with HNO_3/perchloric acid ($HClO_4$)/hydrofluoric acid (HF) was done in 2003 and 2008, while leaching with 7 M HNO_3 was used in 2014 and 2020. For analysis of chromium (Cr) and nickel (Ni), the sample was fused with lithium borate ($LiBO_2$) followed by acid digestion with HNO_3 or an acid mix. For nickel, however, the method was changed in 2014 to leaching with 7 M

HNO_3, a method used also in 2020. Final determination was performed by plasma emission spectrometry (ICP-AES) or plasma mass spectrometry (ICP-MS).

Total organic carbon (TOC), total carbon (TC) and total nitrogen (TN) were analysed all four monitoring years: 2003 at MikroKemi in Uppsala, 2008 at the Department of Geosciences at the University of Gothenburg, and 2014 and 2020 at the Swedish Meteorological and Hydrological Institute (SMHI). The sediment accumulation rate (SAR) was investigated by the Swedish Radiation Safety Authority by analysing Cs-137 and Pb-210 (Olszewski *et al.* 2018). Black carbon was analysed in samples from the 2020 monitoring at UC Davis Stable Isotope Facility after sample preparation at the Norwegian Geotechnical Institute. The procedure was adapted from Gustafsson *et al.* (2001) and consisted of burning at 375°C for 16 h to remove amorphous organic matter, followed by addition of hydrochloric acid to remove inorganic carbon. Remaining carbon is considered black carbon.

Reporting limits and calculations

A reporting limit is the lowest concentration or amount of a substance that the laboratory can report. The reporting limit can be either the limit of detection (LOD) or the limit of quantification (LOQ). The LOD is the lowest concentration or amount of a substance that can be detected, i.e., differentiated from zero. The LOQ is the lowest concentration or amount of a substance that can be reliably measured, i.e., quantified. The LOQ is therefore higher than the LOD, and values that are above the LOD but below the LOQ are more uncertain. Throughout the monitoring years, the laboratories are sometimes only reporting values higher than the LOQ, sometimes also values higher than LOD. For this reason, the term reporting limit is used throughout the report unless the more specific terms can be used.

The term concentration usually applies to an amount per unit volume. Because the results for sediment contaminants are given per unit weight, such as μg/kg dry weight of sediment, the term level is used instead throughout the report.

For metals, average values per station ($n = 7$, but $n = 5$ for SE-5 2008 and $n = 6$ for SE-6 in 2020) were calculated and are displayed in the figures in this report, with error bars corresponding to 1 standard deviation (SD). If replicates had values below the reporting limit, a value corresponding to half the reporting limit was used for that replicate when calculating the average. If all replicate values for a station were below the reporting limit, however, the average was set to 0.

When sum parameters have been calculated for organic pollutants, for example $\sum PCB_7$, so-called lower bound concentration has been used. This means that for the substances that are included in the sum parameter but are below the reporting limit, the content of the substance has been set to zero. The reason for using this approach was the large differences in reporting limits for organic contaminants over the years, which was related to changes in laboratories and methods.

Environmental quality standards and reference values

To assess whether the measured contaminant levels are high enough to risk causing ecological effects, primarily for sediment-living organisms, environmental quality standards (EQS) for priority substances and so-called river-basin specific pollutants (RBSPs) within the Water Framework Directive (WFD) have been used. The EQS for priority substances are EU-wide while EQS for RBSPs are set nationally. The EQS for Sweden are published by the Swedish Agency for Marine and Water Management (SwAM 2019). Most EQS are for water and biota, but there are EQS for sediment for the metals cadmium, copper and lead, and for the organic pollutants anthracene, fluoranthene and tributyltin (TBT). The EQS within the WFD are not strictly applicable to offshore sea areas but have been adopted also within the Marine Strategy Framework Directive (MSFD; Descriptor 8; SwAM 2012).

Comparisons were made between the measured level at each station and the EQS for each substance according to national guidance (SwAM 2016). An assessment of exceedance can therefore differ from assessments made on the basis of partly the same data within HELCOM and OSPAR, where other assessment areas are used and occasionally also other threshold values.

When EQS based on ecotoxicological studies on sediment-living organisms (SwAM 2019) are missing, so-called indicative values from SwAM (2018b) have been used. These are often based on ecotoxicological studies on pelagic organisms, i.e., organisms living in the water column, and have then been recalculated for sediment. As there are large uncertainties in the differences in toxicity between pelagic and sediment-living organisms as well as in the parameters used for the recalculation, the indicative values are more uncertain. A contaminant level below the indicative value thus does not mean that there is no risk of ecological effects.

The levels of metals were evaluated in relation to reference values that are considered to represent pre-industrial conditions (SwEPA 1999). It is however important to note that there is only one reference value for the whole Swedish coast and sea area, even though the metal levels show natural geographical variations due to, for example, differences in the content of metals in the bedrock.

In addition to these EU/national assessment criteria and reference values, results from the monitoring have been compared to the proposed indicator threshold values for HELCOMs upcoming third holistic assessment of the Baltic Sea (HOLAS III). This was done for all stations, including the two northernmost stations on the west coast of Sweden, which are located in Skagerrak and thus within the OSPAR rather than the HELCOM area.

SEDIMENT CHARACTERISTICS

Redox conditions

Stations differ markedly in redox conditions (Table 2, Fig. 2). During sampling, the dissolved oxygen content of the water above the sediment was measured and the sediment surface was observed with an underwater camera and when samples had been recovered to the deck of the ship. At stations in the Baltic Proper (SE-5 to SE-10) the water was anoxic or hypoxic, i.e., oxygen-free or with a lack of oxygen, as shown by the content of dissolved oxygen being 0 at most stations in 2014 and <0.1 mL/L in 2020. The sediment surface shows clear signs of oxygen deficiency, and these sediments are usually called dead bottoms due to the absence of macroscopic organisms. The oxygen content at SE-11 in the Southern Baltic was also relatively low, 0.7 and 1.2 mL/L in 2014 and 2020, and the sediment surface is on the border between being oxic and anoxic (see Table 2). At other stations the sediment surface is oxic and the oxygen level in the water above the sediment is between 4.2 and 8.1 ml/L and is highest in the Gulf of Bothnia and the North Sea. Information on salinity and temperature in the water column above the sediment at each station can be found in Apler & Josefsson (2016).

An increasing sea floor area of the Baltic Sea has become anoxic or hypoxic during the 20th century and onwards, mainly due to the increased eutrophication due to the supply of nutrients from land (Carstensen *et al.* 2014). These dead bottoms occur in the Baltic Proper around Gotland and east of Bornholm, but the distribution varies over time due to, for example, temporary inflows of oxygen-rich seawater to the Baltic Sea.

Sediment accumulation rate

The sediment accumulation rate (SAR) differs greatly between the stations. High rates of around 1 cm or more per year were measured at SE-16 in the North Sea and SE-5, SE-6 and SE-8 in the northernmost part of the Baltic Proper (Table 2) based on samples taken in 2014. The lowest accumulation rates, below 0.1 cm per year, occurred at stations SE-9, SE-10 and SE-7, all of which are located in the Baltic Proper. The fact that the SAR differs means that the time required to form a one-centimetre layer of sediment particles is not the same for all stations. The low rate at SE-9, SE-10 and SE-7 means that an average one-centimetre layer in the sediment core corresponds to 22, 21 and 13 years, respectively. The sediment at these stations should therefore react more slowly to changes in the contaminant levels of the particles that settle than is the case at stations with high SAR. Note that this is the average SAR in the sediment core, and that the surface layer is generally less dense than the underlying layers that have become more compacted. That means that the sampled surface layer of sediment (0–1 cm) has sedimented for a shorter time than what an average one-centimetre layer in the core has. Stations with oxic conditions and associated macrofauna are also subject to continuous mixing of the sediment surface layer due to animal activity, so-called bioturbation.

Particle size distribution

In 2020, samples were taken to verify that the sediment was fine-grained. To collect enough material, the top 5 cm of sediment was sampled. This can differ in, for example, organic matter content from the top 1 cm that is usually sampled, but the particle size after removal of organic matter should be the same assuming that the conditions affecting sedimentation, e.g. bottom current, have not changed. Analyses confirmed that all samples were fine-grained, that is, with particles <63 μm in diameter. The percentage of particles below 63 μm varied between 91% for SE-1 and 100% for SE-2.

Table 2. Sediment accumulation rates (SARs), the number of years of sedimentation an average centimetre of the sediment corresponds to, the levels of dissolved oxygen in the bottom water, and if the sediment surface was oxic (Ox) or reduced (Red) at the 16 monitoring stations. SARs (± 1 standard deviation) were measured by the Swedish Radiation Safety Authority using samples collected in 2014 and more information about the methods can be found in Olszewski *et al.* (2018). At station SE-10 and SE-16 it was difficult to determine the SAR using ^{137}Cs, and no SAR could be determined at SE-15 due to the core being mixed down to at least 25 cm or due to a very high SAR.

Station	Sea area	SAR, ^{210}Pb (cm/year)	SAR, ^{137}Cs (cm/year)	Years per cm	O_2 (mL/L)[1]	Sediment surface
SE-17	Bothnian Bay	0.18 ± 0.04	0.18 ± 0.05	5.6	8.1, 7.4	Ox
SE-1	Bothnian Bay	0.16 ± 0.01	0.14 ± 0.05	6.3	7.2, 7.2	Ox
SE-2	Bothnian Sea	0.67 ± 0.12	0.66 ± 0.09	1.5	4.2, 3.8	Ox
SE-3	Bothnian Sea	0.37 ± 0.04	0.39 ± 0.09	2.7	4.9, 3.8	Ox
SE-4	Bothnian Sea	0.42 ± 0.05	0.39 ± 0.09	2.4	5.4, 4.4	Ox
SE-5	Baltic Proper	1.03 ± 0.37	0.93 ± 0.09	1.0	0, <0.1	Red
SE-6	Baltic Proper	0.88 ± 0.09	0.75 ± 0.09	1.1	0, <0.1	Red
SE-7	Baltic Proper	0.08 ± 0.01	0.02 ± 0.05	13	0, <0.1	Red
SE-8	Baltic Proper	1.03 ± 0.42	1.02 ± 0.09	1.0	0, <0.1	Red
SE-9	Baltic Proper	0.045 ± 0.003	0.02 ± 0.05	22	0.1, <0.1	Red
SE-10	Baltic Proper	0.047 ± 0.002		21	0, <0.1	Red
SE-11	Southern Baltic	0.36 ± 0.09	0.27 ± 0.04	2.8	0.7, 1.2	Ox/Red[2]
SE-12	Southern Baltic	0.57 ± 0.25	0.56 ± 0.09	1.8	4.4, 4.0	Ox
SE-13	North Sea	0.62 ± 0.15	0.66 ± 0.09	1.6	6.2, 4.4	Ox
SE-15	North Sea				7.4, 4.5	Ox
SE-16	North Sea	1.49 ± 0.95	>1	0.7	7.5, 5.1	Ox

[1] Oxygen levels in the bottom water determined in 2014 and 2020. The oxygen level at station SE-8 in 2014 could not be determined because it was not possible to use the equipment at this depth, but it was assumed to be 0 mL/L.

[2] In 2014 this station had reduced sediment. In 2020, all seven locations sampled at the station had oxic surface sediment except for possibly one location. Benthic fauna (most likely spionid polychaetes) was observed, which indicates oxic conditions. When sampling was repeated in 2021 for organic contaminants, all seven locations had reduced surface sediments and the sulphur-oxidising bacteria *Beggiatoa* could be observed.

TOC

The levels of organic carbon are substantially higher in sediments at stations in the Baltic Proper compared to other sea areas (Figure 2). This is probably due to the reducing conditions prevailing here, which leads to a less efficient degradation of the organic matter than in oxidising conditions (Kristensen *et al.* 1995). The Baltic Proper has a large primary production (Jonsson & Carman 1994), giving a higher sedimentation of organic matter which in turn contributes to creating reducing conditions. The TOC levels are however similar at all six stations in the Baltic Proper even though SARs differ substantially, with only slightly lower TOC levels at stations SE-7, SE-9 and SE-10 which have very low SARs compared to the more northern stations with high SARs (SE-5, SE-6 and SE-8; Table 2). The Southern Baltic is the area with the second highest levels after the Baltic Proper. Levels in the Bothnian Bay are higher than in the Bothnian Sea, which may be related to the large riverine input, with associated organic matter, to the Bothnian Bay. TOC levels are lowest at the North Sea stations, where the higher species diversity leads to a more efficient organic matter utilization.

Because many organic contaminants, and to some extent also elements, bind to organic material, the levels of contaminants can be correlated with the levels of organic carbon. Such a correlation has been demonstrated for organic contaminants in soil in areas without point sources, i.e., where the contaminants have been transported through atmospheric transport and deposition (Meijer *et* *al.* 2003). The contaminant levels can thus be normalised to the levels of organic carbon in sediment to achieve a clearer picture of the geographical load of contaminants. A normalisation, however, gives a misleading picture of the contaminant load to different areas if the degradation of organic carbon in the sediments occurs at different speed at each station, which is likely due to the reducing conditions prevailing at some stations. The contaminant levels are therefore given per kg dry weight (DW) of the sediment in this report instead of being normalised to the content of organic carbon. The correlation between contaminants levels and TOC has however been evaluated for some substances to further examine geographical differences, for example if some sea areas have higher or lower contaminant levels than would be the case if TOC was the only controlling parameter for the levels in the sediment.

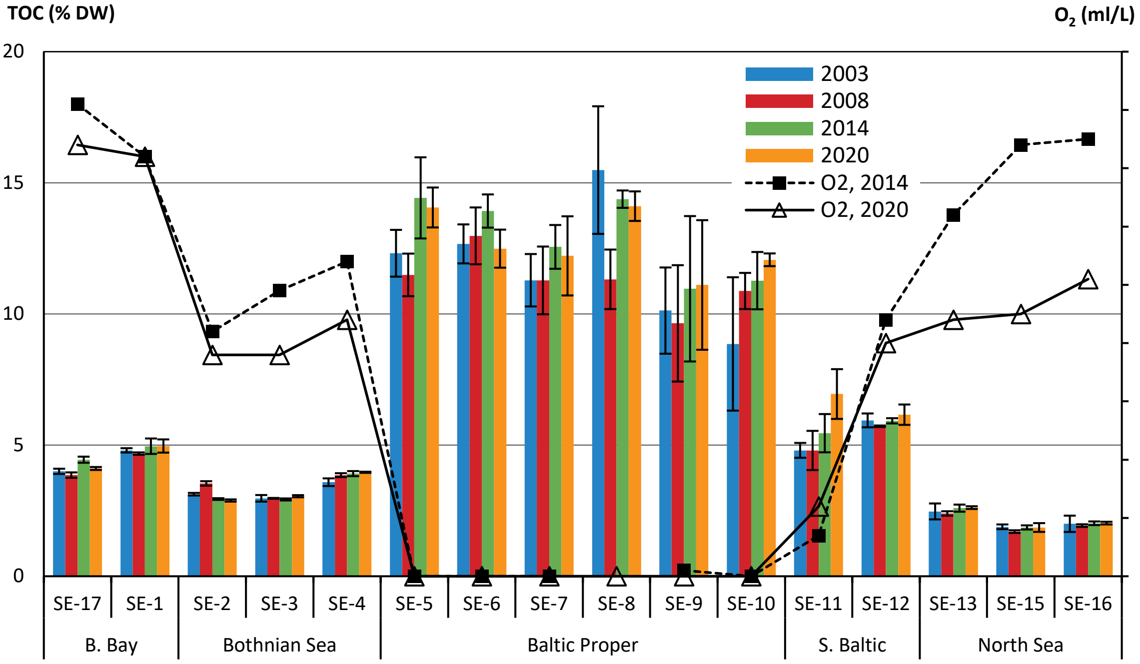

Figure 2. Total organic carbon (TOC, % DW) at the 16 stations during the four monitoring years, and the oxygen levels measured in the bottom water at the time of sampling in 2014 and 2020. The error bars denote ± 1 standard deviation (*n* = 7, except for SE-6 in 2020 when *n* = 6). It is apparent that some stations have larger uncertainties, primarily SE-9 but also the other stations in the Baltic Proper and SE-11.

Black carbon

For the 2020 monitoring, more detailed analyses of the sediment and organic matter characteristics were made. This included, in addition to particle size distribution, analyses of black carbon and stable isotopes.

Black carbon (BC), also known as soot, is formed during incomplete combustion of fossil fuels or biomass. It forms part of the total organic carbon but is more resistant to degradation than the amorphous part of the organic carbon, for example dead algae that has sedimented to the sea floor. Black carbon is a stronger sorbent of contaminants, primarily planar contaminants such as PAHs and PCDD/Fs but also other contaminants, than amorphous organic matter (Accardi-Dey & Gschwend 2002, Jonker & Koelmans 2002).

The levels of BC were highest at the Baltic Proper stations, constituting 1–2% of the sediment on a dry weight basis (Figure 3). BC-levels above 1% were also common in the Southern Baltic and the North Sea, while levels were lower in the Bothnian Bay and even lower in the Bothnian Sea. The percentage of the TOC that consisted of BC was 15–20% in the Bothnian Bay and Sea. In the Baltic Proper it was lower, down to 10%. An explanation might be that there is a larger primary production, and thus a larger sedimentation of amorphous organic matter, in this sea area. In the North Sea, a surprisingly large part of the TOC consisted of BC, for example more than 50% at SE-15. Levels of BC here are similar to the Baltic Proper; however, levels of TOC are substantially lower (see Fig. 2). The reason for the large percentage of BC in the sediment TOC might thus be a more efficient degradation of the amorphous organic matter compared to the situation in the anoxic Baltic Proper sediments.

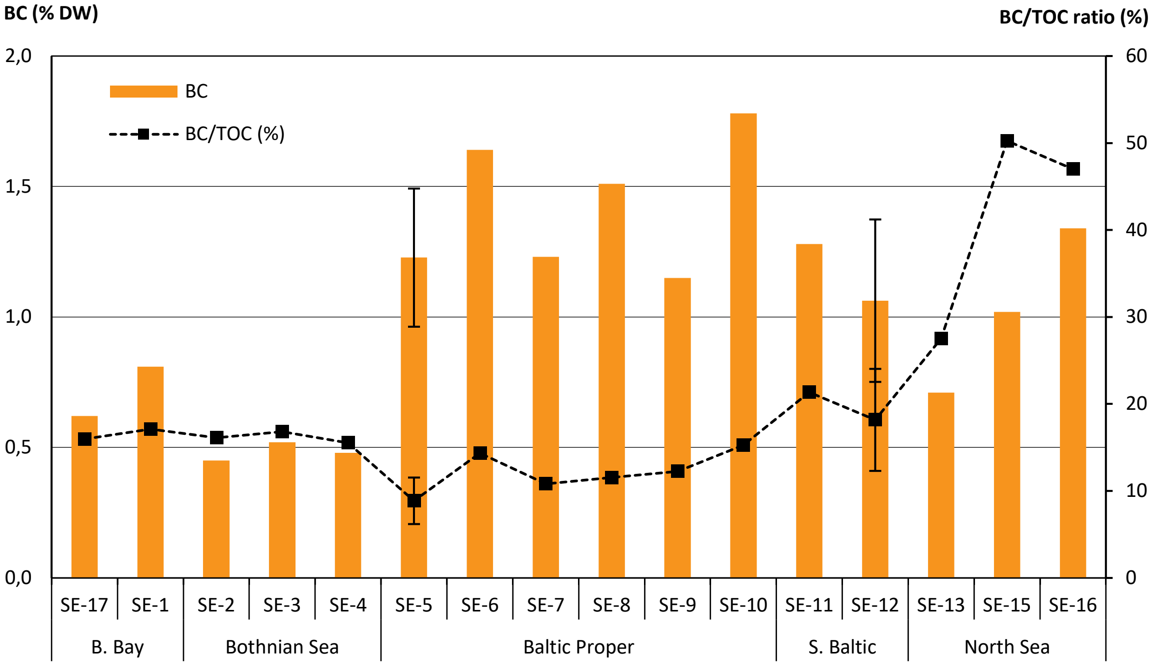

Figure 3. Black carbon (BC, % DW) and the ratio between black carbon and total organic carbon (BC/TOC ratio, %) at the 16 stations in 2020. For most stations, the sample was pooled from the seven replicate samples at each station (*n* = 7, but *n* = 6 at SE-6 and *n* = 4 at SE-10 due to lack of sample), but at SE-5 and SE-12, four of the individual replicates were analysed. The values for these stations are thus an average ± 1 standard deviation.

Carbon and nitrogen stable isotopes

Stable isotopes of carbon and nitrogen were analysed in sediment samples from 2020 by preparation at the Department of Environmental Science at Stockholm University and analysis at UC Davis Stable Isotope Facility, California, USA. Stable isotope results are illustrated as ratios between ^{13}C and ^{12}C in the sample (compared to a standard), δ^{13}C, and between ^{15}N and ^{14}N, δ^{15}N. The δ^{13}C and δ^{15}N provides information on the origin of the organic matter in the sediment as well as biogeochemical processes. In general, ratios are lower in organic matter originating from a terrestrial environment compared to a marine environment (e.g., Hedges *et al.* 1997, Middelburg & Nieuwenhuize 1998, Rolff & Elmgren 2000, Voss *et al.* 2000) as terrestrial organic matter has incorporated C and N predominantly from the forms N_2 and CO_2, which are depleted in ^{13}C and ^{15}N. Marine organic matter, on the other hand, has predominantly incorporated C and N from the forms nitrate and bicarbonate, which are enriched in ^{13}C and ^{15}N, giving higher ratios (Voss *et al.* 2000 and sources therein).

Increasing gradients from the more terrestrially impacted Bothnian Bay sediment samples to the marine North Sea can be seen in Figures 4 and 5. For δ^{13}C, values are around -26‰ in the Bothnian Bay sediments and increase to around -23‰ in the North Sea sediments. For δ^{15}N, values are around 4‰ in the Bothnian Bay and increasing to 6–8‰ in the North Sea. The ratios are however not only impacted by organic material of terrestrial or marine origin. The δ^{15}N can, for example, be affected by manure and sewage rich in ^{15}N or synthetic fertilizers depleted in ^{15}N (Heaton 1986). In Figure 5, it is obvious that the sediment organic matter at the Baltic Proper stations has a very low δ^{15}N, 2–3‰. This has been observed also in other studies (e.g., Struck *et al.* 2000, Voss *et al.* 2000, Remeikaite-Nikiene *et al.* 2016) and it indicates that the organic matter in part derives from cyanobacterial blooms. During these blooms, the plankton incorporates nitrogen in the form of N_2, which has a low δ^{15}N of around 0‰.

The deviating δ^{13}C values for some Baltic Proper and Southern Baltic stations, such as the high values for SE-6 and SE-8 and the low values for SE-9 and SE-11, are more difficult to explain (Fig. 4). The δ^{13}C and δ^{15}N can be impacted by degradation of the organic matter, and the impact can differ depending on if the degradation is occurring in anoxic or oxic sediments (Lehmann *et al.* 2002). Deviating ratios could possibly be attributed to differences in microbial geochemical processes that can influence carbon isotope fractionation, i.e., differences in methane and sulphur cycling (Whiticar 1999). Differences in nitrogen cycling processes driven by microbial communities may also partly explain differences in δ^{15}N between stations in the Baltic Proper (i.e. anammox and denitrification). Only oxygen or depth conditions do not however explain differences in δ^{13}C as the stations in the Baltic Proper are all anoxic. The deepest stations in the Baltic Proper are SE-8 (403 m) and SE-6 (195 m), which are the stations with the highest δ^{13}C; on the other hand, SE-9 is also rather deep (178 m; Table 2).

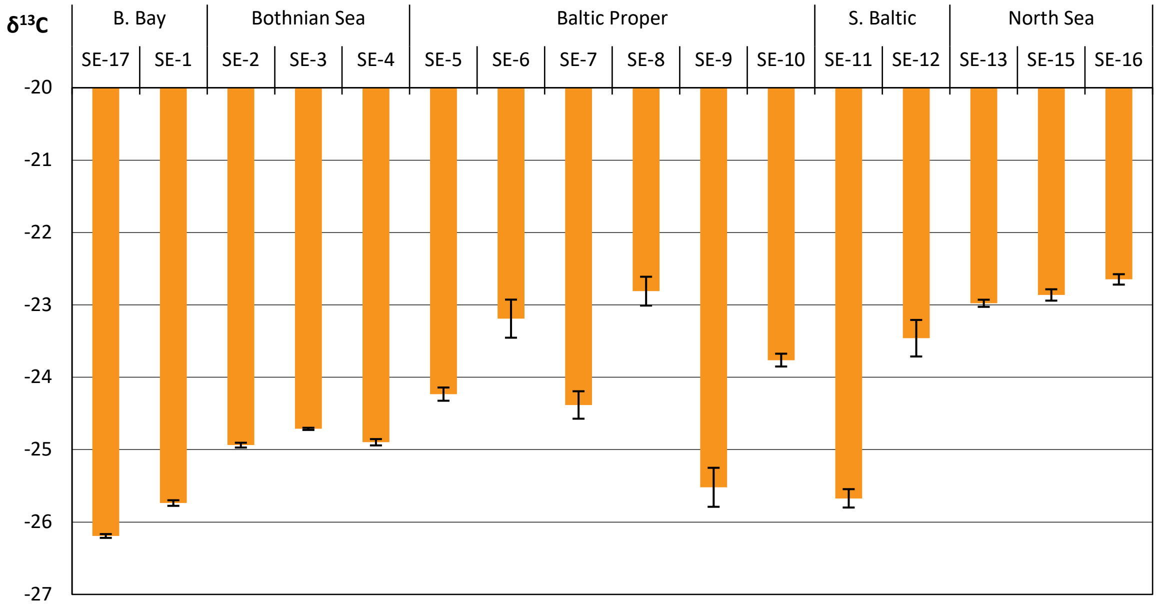

Figure 4. δ^{13}C (‰) at the 16 monitoring stations in 2020. Four of the replicates sampled at each station were analysed ($n = 4$) and the values are averages ± 1 standard deviation.

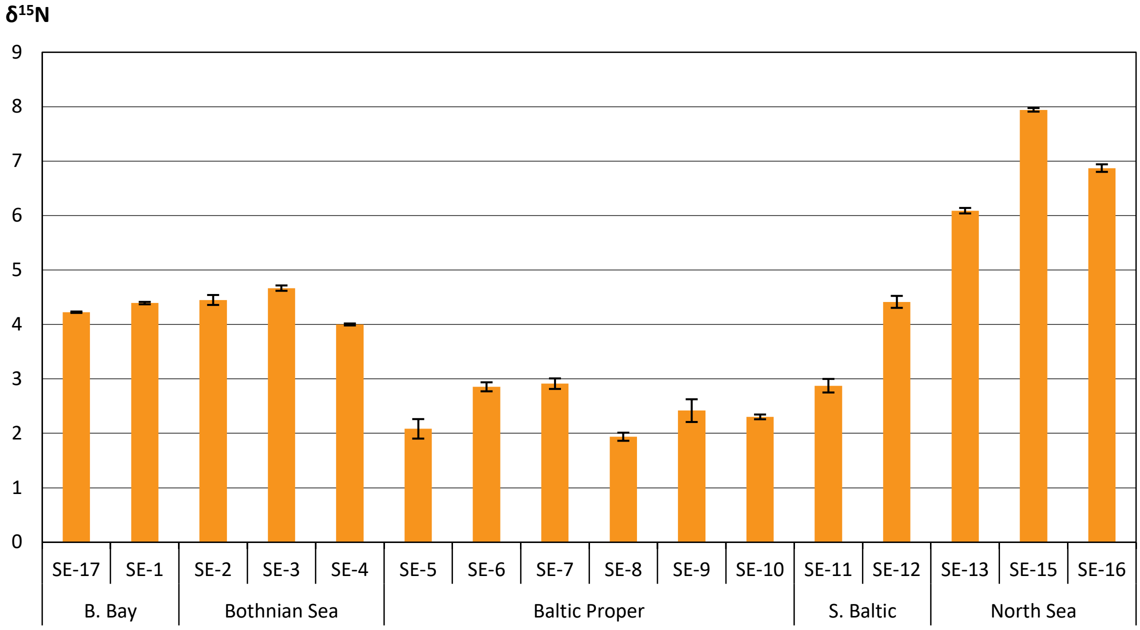

Figure 5. *δ*^{15}N (‰) at the 16 monitoring stations in 2020. Four of the replicates sampled at each station were analysed (*n* = 4) and the values are averages ± 1 standard deviation.

METALS AND OTHER ELEMENTS

Metals and other elements occur naturally in the environment. The levels in sediments are affected by the levels in bedrock and moraine within the catchments. There may therefore be large natural variations in levels between different sea areas and stations. Levels of metals and other elements in the environment have however increased due to human (anthropogenic) activities.

Many elements are redox-sensitive which affects their levels in surface sediment. Arsenic, for example, can be enriched in the surface layer of sediment as it can migrate upwards through reduced sediment and precipitate with oxides or hydroxides in the surface sediment if the oxygen supply there is sufficient (Farmer 1991). Cobalt can also be associated with oxides and hydroxides in oxygenated sediment, while it is more mobile and can be released when the sediments are anoxic (Swanner *et al.* 2014). Cadmium, copper, nickel, lead and zinc, on the other hand, occur as weakly soluble sulphides under reducing conditions (Di Toro *et al.* 1990, Farmer 1991, Ankley *et al.* 1996, Chapman *et al.* 1998), which can affect concentrations in the sediments in the Baltic Sea (Borg and Jonsson 1996). Measurements have shown that sulphur levels in the sediment are higher in the Baltic Proper, where anoxic conditions prevail, than in other sea areas (Apler & Josefsson 2016). Other factors than redox conditions can also affect the metal levels in the sediment, e.g., some metals associate with organic material in water and sediment (Nissenbaum & Swaine 1976, Mantoura *et al.* 1978, Borg & Jonsson 1996). The composition of the sediment and the degree of oxygenation thus determine the distribution of metals and in what form they occur.

In this report, nine elements are presented more in detail. These are the elements that have EQS for sediments or for which there are reference values published by SwEPA (1999). At the end of the chapter, other analysed elements are presented more briefly using multivariate statistics. A comparison between different extraction methods performed in 2020 is also presented.

Arsenic

Arsenic is a toxic metalloid that has been used, among other things, as a wood preservative, pesticide and in alloys. Large discharges have taken place from smelters and incineration plants, but the use of arsenic is nowadays strongly limited, and the unintentional emissions have been reduced.

Levels of arsenic are highest in sediment in the Bothnian Bay, followed by the Bothnian Sea (Fig. 6). The station with the highest levels is always SE-1 in the southern Bothnian Bay. Unlike the four northernmost stations, station SE-4 in the Åland Deep has levels that are not elevated and resemble remaining stations. All stations however display arsenic levels that exceed the national reference value of 10 mg/kg DW, which represent national pre-industrial levels (SwEPA 1999). The only exception is station SE-5 where the average value is slightly below the reference value in 2014. There is no consistent increase or decrease over time of arsenic levels (Fig. 6).

Arsenic occurs in elevated levels in bedrock and moraine in many parts of Sweden, for example in the Skelleftefält (Andersson *et al.* 2014), which is a contributing factor to the high levels in the Bothnian Bay and Bothnian Sea. Another reason for elevated levels in these areas can be emissions from point sources. The metal smelter at Rönnskär has historically released large amounts of metals and the highest levels of arsenic were measured at station SE-1, the station closest to this smelter. A previous sediment survey around the Rönnskär smelter has demonstrated that it was a source of arsenic (Cato & Sellén 2004). During a previous study of metals in Baltic Sea sediments (Borg & Jonsson 1996), the highest levels of arsenic occurred in the Bothnian Bay, which was explained by the high historical emissions from smelters to air and water.

As previously mentioned, the geochemical conditions in the sediments in the Bothnian Bay and Bothnian Sea (anoxic sediments with an oxic surface) can contribute to an enrichment of arsenic in the surface layer of sediment as the substance precipitates with oxides or hydroxides at the oxygenated sediment surface (Farmer 1991). The bottom water is however well oxygenated even at stations in the North Sea and partly in the Southern Baltic, so the high levels in the Gulf of Bothnia cannot be explained by this. Rather, the high levels here are due to the naturally elevated levels of arsenic in bedrock and moraine combined with one or more significant point sources from metal industry in Sweden and Finland.

Arsenic is among the river basin-specific pollutants (RBSP) listed in Sweden according to the WFD (SwAM 2019); however, there is no EQS specified for sediment. Arsenic has not been an indicator within the HELCOM second holistic assessment (HOLAS II) of the environmental status of the Baltic Sea (HELCOM 2018b) and is also not planned to be included in the upcoming HOLAS III.

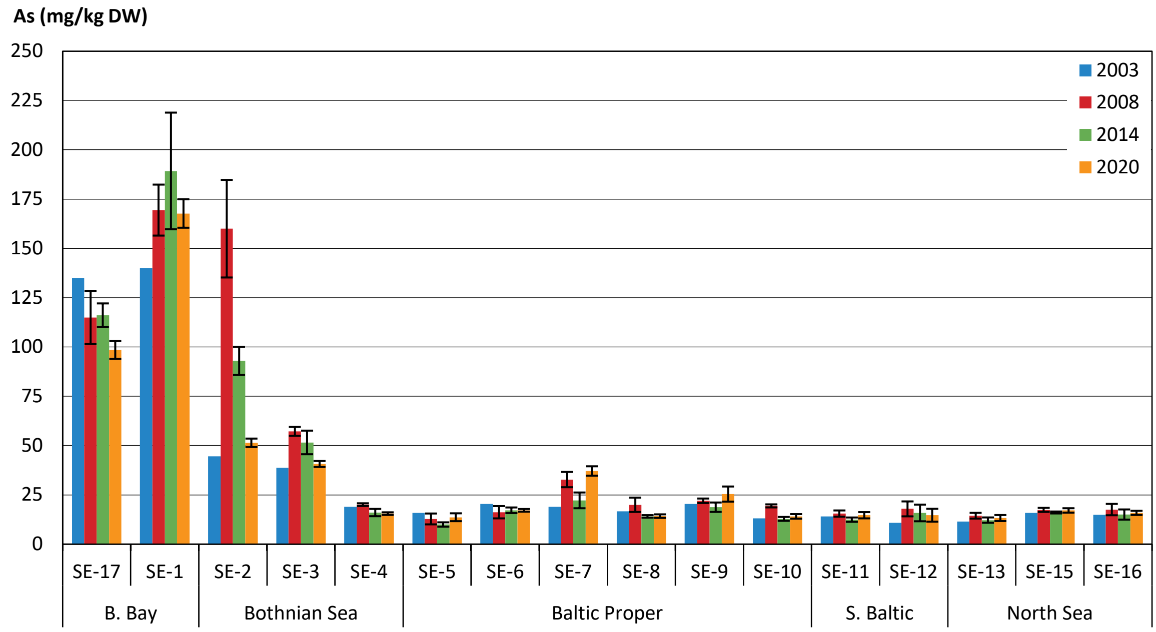

Figure 6. Levels of arsenic (As) at the 16 stations during the four monitoring years. For 2008, 2014 and 2020 the average value ± 1 standard deviation is displayed; *n* = 7 except for SE-5 in 2014 when *n* = 5 and SE-6 in 2020 when *n* = 6.

Cadmium

Cadmium has been used in many different products and processes, and occurs as a contaminant in phosphate fertilizers and zinc products. More recently, its use has been limited and today cadmium is mainly used in batteries, paints and electronics (Andersson *et al.* 2012). The largest anthropogenic cadmium sources in Sweden are believed to be the combustion of fossil fuels, the use of phosphate fertilizers and metal production (Andersson *et al.* 2012). Air emissions have decreased considerably since the early 1990s, partly due to better processes and purification in the metal industry, however a major source of emissions to air is now the combustion of biomass (SwEPA 2019b).

Large amounts of cadmium can also be released from land to the aquatic environment from so-called acid sulphate soils (Andersson *et al.* 2012), which are formed when sulphide-containing soils are oxidised. Acid sulphate soils occur in areas that have previously been seabed but have risen above the coastline due to land uplift. Large areas of acid sulphate soils occur in northern Finland and Sweden, and calculations in Finland show that significantly larger amounts of cadmium, about 5 times more, are emitted to the aquatic environment from acid sulphate soils than from industrial emissions (Sundström *et al.* 2002).

Cadmium levels are clearly highest in the Baltic Proper, followed by the Bothnian Bay, while levels are lowest in the North Sea (Fig. 7). Cadmium is a priority hazardous substance in the WFD and the EQS for sediment is 2.3 mg/kg DW (SwAM 2019), which is also the indicator threshold proposed for HOLAS III. This value is exceeded at several stations in the Baltic Proper. This shows that the levels are high enough to pose a risk to sediment-living organisms in large areas of the Swedish seabed. At these stations, however, macroscopic life generally does not occur due to lack of oxygen caused by eutrophication. The high cadmium levels in sediment in the Baltic Proper have also been observed in a previous sediment study (Borg & Jonsson 1996) and corresponds to results from the national environmental monitoring programme for contaminant in marine biota, where concentrations are generally higher in the Baltic Proper than in the northern parts of the Baltic Sea or in the North Sea (Soerensen & Faxneld 2020). The national reference value for sediments of 0.2 mg/kg DW, which represents pre-industrial concentrations (SwEPA 1999), is exceeded at all stations in the Baltic Sea but never at the three stations in the North Sea.

One explanation for the clearly elevated cadmium levels in the Baltic Proper compared to other sea areas is the redox conditions in the sediment. In the Baltic Proper, reducing conditions prevail in surface sediments and bottom water (Table 2), and it is well known that cadmium forms weakly soluble sulphides in reduced sediments (Di Toro *et al.* 1990, Ankley *et al.* 1996). Reducing conditions in the sediment in the Baltic Proper can thus be a contributing factor to the high cadmium levels here (Borg & Jonsson 1996). The stations in the Bothnian Bay also have elevated cadmium levels despite oxidizing conditions in the sediment and oxic bottom water. This might be linked to the large historical emissions from metal working sources in the area, such as the Rönnskär smelter, although the smelter is not as significant a source of cadmium to sediment compared to arsenic (Cato & Sellén 2004). Another potential source of cadmium are acid sulphate soils, as mentioned previously.

Because the levels of organic carbon are high at the stations with reducing conditions (Fig. 2), the cadmium levels show a positive and significant linear relationship with the levels of TOC ($r^2 = 0.79$, $p < 0.0001$, $n = 64$). This trend also occurs when Baltic Proper stations are excluded from the relationship, but with lower degree of correlation ($r^2 = 0.39$, $p < 0.0001$, $n = 40$). This indicates that cadmium is associated with organic matter in the aquatic environment. At the two stations in the Bothnian Bay, the levels of cadmium are elevated in relation to levels based on the correlation against TOC, which as mentioned may be due to influence from point sources.

There is no consistent increase or decrease in the levels of cadmium over time (Fig. 7). Within the environmental monitoring programme for marine biota, there has been no general trend for concentrations in fish during the last ten years, while there has been a decreasing trend in mussels and bird eggs (Soerensen & Faxneld 2020).

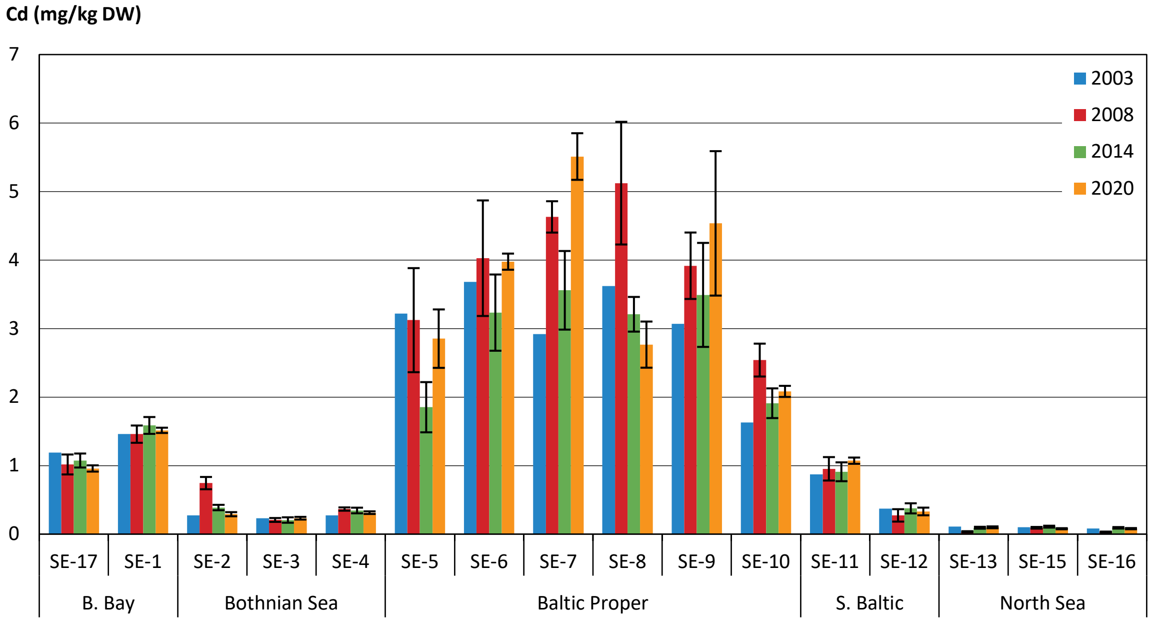

Figure 7. Levels of cadmium (Cd) at the 16 stations during the four monitoring years. For 2008, 2014 and 2020 the average value ± 1 standard deviation is displayed; *n* = 7 except for SE-5 in 2014 when *n* = 5 and SE-6 in 2020 when *n* = 6.

Cobalt

Cobalt is used, for example, in alloys to increase strength and in different types of batteries. Due to an increased market for electric vehicles the use of cobalt is expected to increase.

Levels of cobalt vary less between sea areas than arsenic and cadmium do, but higher levels are observed in sediments of the Bothnian Bay (Fig. 8). Levels are also elevated in the Bothnian Sea. The highest levels always occur at station SE-1 in the southern Bothnian Bay, consistent with arsenic. Unlike arsenic, however, the smelter at Rönnskär is not thought to be a large source of cobalt to sediment (Cato & Sellén 2004). One explanation for the elevated levels may be the high levels of cobalt in bedrock and moraine in northern Lapland and to the south along the mountain range between Sweden and Norway (Andersson *et al.* 2014). Another contributing factor to elevated levels in the Bothnian Bay, and to some extent also in the Bothnian Sea, may be that cobalt, like arsenic, is more soluble under reducing conditions than under oxidizing conditions, and is then enriched by binding to oxides and hydroxides of iron or manganese in oxidized surface sediment (Swanner *et al.* 2014). There is no consistent increase or decrease over time in cobalt levels.

There is no EQS for cobalt in sediments, but the reference value that is assumed to represent pre-industrial levels is 12 mg/kg DW (for chemical analysis according to the Swedish standard method that was used in 2014 and 2020) or 14 mg/kg DW (for the so-called total analysis method that was used in 2003 and 2008; SwEPA 1999). These values were exceeded at six of 16 stations in 2003 and 2008 and at eight of 16 stations in 2014 and 2020.

Cobalt was not included as an indicator in the second holistic assessment of the environmental status of the Baltic Sea (HELCOM 2018b) and is not proposed to be included as an indicator in HOLAS III.

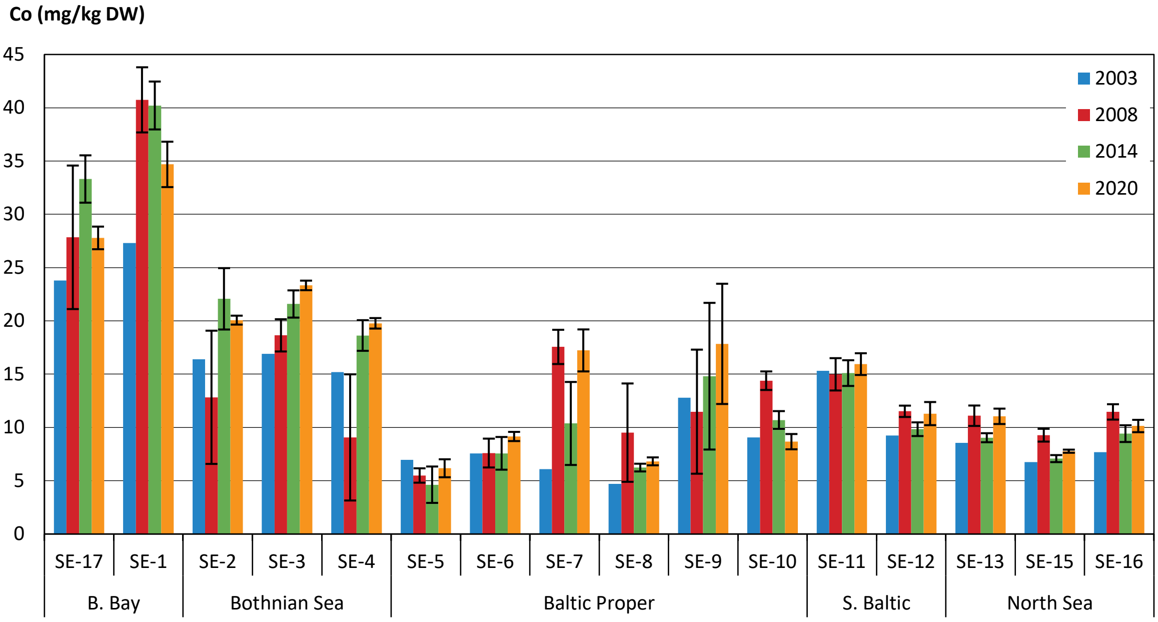

Figure 8. Levels of cobalt (Co) at the 16 stations during the four monitoring years. For 2008, 2014 and 2020 the average value ± 1 standard deviation is displayed; *n* = 7 except for SE-5 in 2014 when *n* = 5 and SE-6 in 2020 when *n* = 6.

Chromium

Chromium is used in large quantities in, for example, stainless steel production, but also for surface treatment of other metals. It is also used in the leather industry for tanning and in paints (SwEPA 2019a).

Chromium levels in sediments are at the same level, around 70–100 mg/kg DW, in all sea areas except the Baltic Proper, which has substantially lower levels (Fig. 9). The chromium levels here vary between 20 and 70 mg/kg DW. The chromium levels thus have a geographical pattern that differs from the other eight elements examined in detail in this report. It is possible that lower chromium levels in sediments in the Baltic Proper are linked to reducing conditions prevailing here. More detailed studies are needed to determine what the cause is, but chromium, unlike cadmium, copper, lead and zinc, does not form weakly soluble sulphides under reducing conditions (Farmer 1991, Guo *et al.* 1997, Berry *et al.* 2004). Like other examined elements, no consistent trends in chromium levels between years are observed (Fig. 9).

Chromium occurs in two different forms in sediments depending on redox conditions: in oxidizing conditions soluble hexavalent chromium is present, which changes to less soluble trivalent chromium under reducing conditions. (Guo *et al.* 1997, Berry *et al.* 2004). At the stations in the Baltic Proper, chromium should thus be bound to the sediment (Berry *et al.* 2004), but it is also possible that the trivalent chromium forms more easily soluble complexes with organic material (Guo *et al.* 1997). In herring sampled in the environmental monitoring of marine biota in the Baltic Sea and the North Sea, chromium levels appear similar in the different sea areas, i.e., levels do not reflect the geographical pattern of levels in sediments (Soerensen & Faxneld 2020).

Chromium is listed as an RBSP for Sweden according to the WFD but there is no EQS for sediments. The reference value that is assumed to represent pre-industrial levels is 80 mg/kg DW (according to the total analysis method, which was used for all years; SwEPA 1999). While this value was never exceeded in the Baltic Proper it was exceeded during at least one monitoring year at all other stations (Fig. 9). Chromium was not included as an indicator in the second holistic assessment of the Baltic Sea environmental status (HELCOM 2018b) and is not proposed as an indicator in HOLAS III.

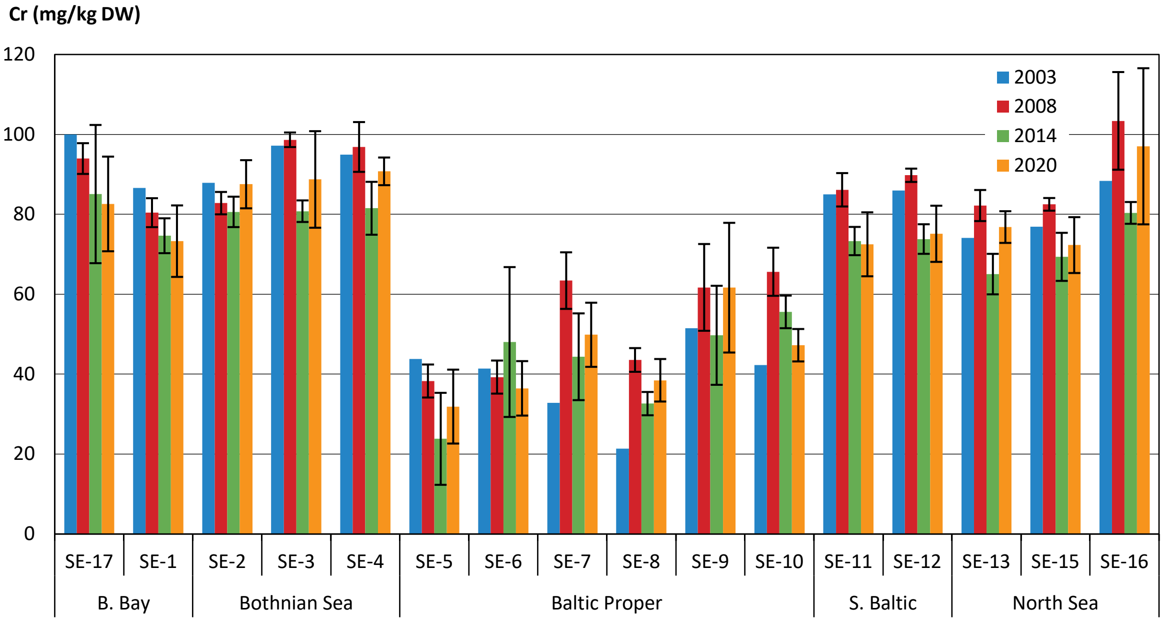

Figure 9. Levels of chromium (Cr) at the 16 stations during the four monitoring years. For 2008, 2014 and 2020 the average value ± 1 standard deviation is displayed; *n* = 7 except for SE-5 in 2014 when *n* = 5 and SE-6 in 2020 when *n* = 6.

Copper

Copper is used for several different purposes, including in power lines and drinking water pipes, but also in pesticides such as in antifouling paints for ship hulls. The emissions of copper into the air come mainly from traffic as the metal is present on brake pads in cars and is released during braking (SwEPA 2019c). Previously, there were also relatively large emissions from the metal industry, but these have decreased due to better treatment techniques.

Copper levels in sediments have a similar geographical distribution to cadmium, with markedly higher levels in the Baltic Proper than in the other sea areas (Fig. 10). This can be explained by copper forming weakly soluble sulphides in reduced sediment (Di Toro *et al.* 1990, Ankley *et al.* 1996). As with cadmium, copper levels are lowest in the North Sea and second lowest in the Bothnian Sea. Unlike cadmium, however, levels are not higher in the Bothnian Bay compared to the Southern Baltic. Concentrations of copper in herring sampled within the environmental monitoring of marine biota were also similar for all sea areas (2010–2019; Soerensen & Faxneld 2020), unlike the pattern in sediments.

Copper levels show a positive and significant linear relationship to TOC levels in sediments ($r^2 = 0.77$, $p < 0.0001$, $n = 64$). This may, as mentioned in connection with cadmium, be related to the reducing conditions in the sediments in the Baltic Proper coinciding with high TOC levels but may also be because copper tends to bind to organic material (Nissenbaum & Swaine 1976, Mantoura *et al.* 1978). This is supported by a positive and significant linear relationship with TOC even when the Baltic Proper stations are excluded ($r^2 = 0.66$, $p < 0.0001$, $n = 40$). No sea areas have consistently elevated copper levels in relation to levels based on the relationship with TOC as opposed to cadmium where the levels were elevated in relation to TOC levels in the Bothnian Bay. Although station SE-1 has consistently elevated copper levels, which may be due to the influence of the Rönnskär smelter (Cato & Sellén 2004) or other metal industries, SE-17 does not have elevated levels of copper in relation to its TOC levels.

There is no consistent trend in copper levels between the years (Fig. 10). At many stations, particularly in the Baltic Proper, the highest levels occurred in 2008. The extraction method was changed between 2008 and 2014 to leaching with 7 M HNO_3, but in both 2003 and 2008 total digestion was used. The elevated levels in 2008 compared to other monitoring years should therefore not be related to the analytical method. Within the environmental monitoring of marine biota in Sweden, no trends or only small increases in copper concentrations have been seen in biota during the last ten years (Soerensen & Faxneld 2020).

Copper is an RBSP in Sweden within the WFD and has an EQS for sediment of 56 mg/kg DW assuming a TOC content of 5% (SwAM 2019). Before comparison to the EQS, the natural background level is subtracted from the measured level. If the national reference value is used as the background content for all stations, an approximation as there is a geographical variation that has not been considered in the reference value, the EQS is exceeded at station SE-8 in 2008 (72 mg/kg DW after background subtraction and normalisation to 5% TOC), but not at other stations or during other years. The national reference value for copper is 15 mg/kg DW (SwEPA 1999) and it is exceeded at all stations except the three stations in the North Sea in 2003.

Copper was not included in the second holistic assessment of the Baltic Sea environmental status (HELCOM 2018b) but is proposed as an indicator for HOLAS III with a threshold of 30 mg/kg DW for a sediment with a TOC content of 5%, however no correction for background levels is made. This threshold is exceeded at most stations during all years – for the 2020 monitoring, only SE-5 has copper levels that do not exceed the indicator threshold.

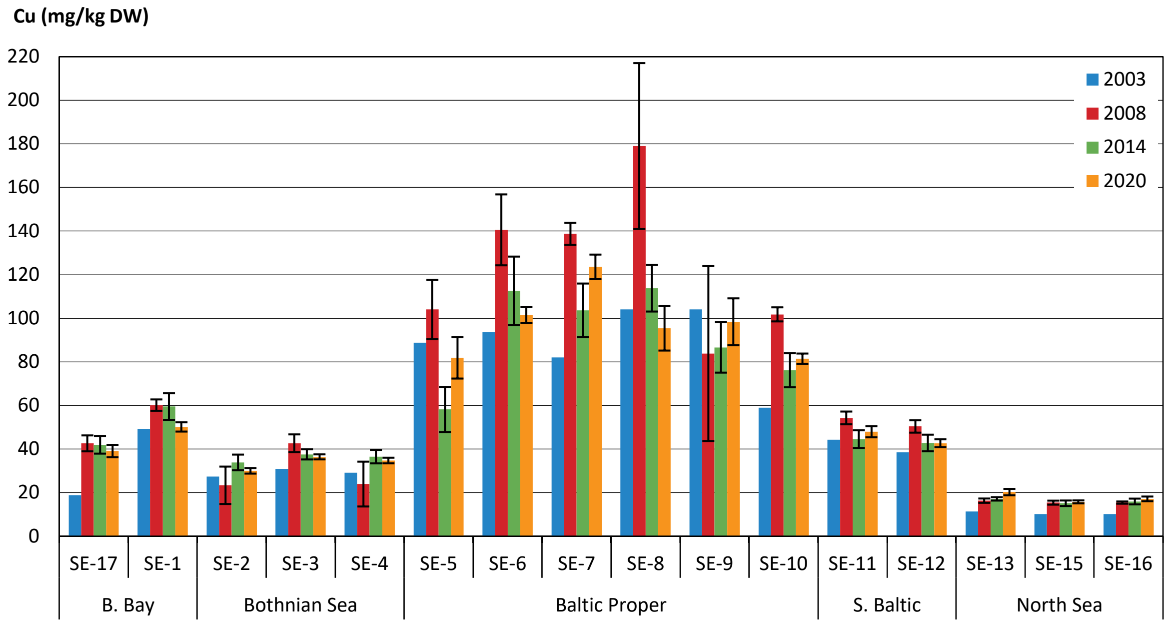

Figure 10. Levels of copper (Cu) at the 16 stations during the four monitoring years. For 2008, 2014 and 2020 the average value ± 1 standard deviation is displayed; *n* = 7 except for SE-5 in 2014 when *n* = 5 and SE-6 in 2020 when *n* = 6.

Mercury

Mercury has been used for several purposes including in thermometers, amalgam fillings or as fungicides, and is now mainly used in low-energy lamps. Due to its toxicity, primarily in its organic forms, the use has been severely restricted and emissions from, among others, metal industry and waste incineration have been significantly reduced.

The highest levels of mercury in sediments occur in the Bothnian Bay, and two stations have substantially higher levels than other stations (Fig. 11). The station with the highest levels is SE-1 in the southern Bothnian Bay, followed by station SE-12 in the Arkona Basin in the Southern Baltic. Sediments at the remaining stations have similar mercury levels, although North Sea stations tend to have medium-high and relatively stable levels.

The fact that levels of mercury are elevated at specific stations rather than in different sea areas indicates an influence from point sources rather than large-scale processes. The station with the highest levels during all monitoring years is SE-1 in the Bothnian Bay. Higher levels of mercury in the Bothnian Bay have also been observed in previous sediment surveys in the Baltic Sea and have been explained by proximity to smelters (Borg & Jonsson 1996). It is known that the Rönnskär smelter has emitted large amounts of mercury, contributing to increased levels of the metal in sediments (Cato & Sellén 2004). This may explain the elevated level of mercury at SE-1. The cause of elevated levels in the sediment at station SE-12 is not known. In the Swedish environmental monitoring of marine biota, concentrations were highest in the north and decreased to the south (Soerensen & Faxneld 2020). This geographical pattern thus only partly corresponds to the pattern seen in sediment.

The mercury levels are negatively but significantly correlated with the TOC content ($r^2 = 0.12$, $p = 0.004$, $n = 64$). This negative correlation is difficult to explain because mercury tends to bind to organic material (Andersson *et al.* 2014, Mantoura *et al.* 1978). The variation is however large and the correlation is mainly attributed to lower levels of mercury and higher levels of TOC in the Baltic Proper. If Baltic Proper stations are excluded, the relationship becomes positive although mainly driven by higher levels at SE-1 and SE-12 ($r^2 = 0.21$, $p = 0.003$, $n = 40$).

No consistent trends in the mercury levels are observed between the four monitoring years. Comparing only the two most recent monitoring years, 2014 and 2020, levels tend to be slightly lower in 2020 for the northernmost stations, but for the southern stations both decreasing and increasing levels compared to 2014 can be observed. Time trends for mercury in environmental monitoring of marine biota show few significant trends for the last decade, although when looking at data for specific basins, a decreasing trend can be seen for the Bothnian Bay and a tendency to an increase at the West Coast of Sweden (2010–2019; Soerensen & Faxneld 2020. The decreasing trend in the Bothnian Bay thus corresponds to what is seen for sediment.

Mercury is a priority hazardous substance in the WFD and frequently exceeds its EQS for biota, but there is no EQS for mercury in sediments. The national reference value, which is assumed to represent pre-industrial levels, is 0.04 mg/kg DW (SwEPA 1999). This value is exceeded at virtually all stations and for all years, except for two stations in the Baltic Proper in 2008, one station in 2014, and two stations in 2020 (Fig. 11).

As previously mentioned, mercury often exceeds its EQS in biota in Swedish coastal and marine areas (SwAM 2015, SwAM 2018a). In the second holistic assessment of the Baltic Sea environmental status, mercury was one of the substances that most markedly exceeded the threshold values in biota and contributed to reduced environmental status (HELCOM 2018b). Mercury is planned to be included as an indicator for HOLAS III; however, without a threshold value for sediment.

High levels of mercury in relation to threshold values mean that a good environmental status is not believed to be achievable in the foreseeable future (SwAM 2018a). Long-distance air transport is an important source of mercury to the Baltic Sea region, and work at the international level has been identified as important (SwAM 2015). The large difference in levels in sediments at different stations however shows that point sources can have a significant impact. It is therefore important to continue to reduce emissions from point sources, for example highly contaminated areas, to further reduce mercury levels in the aquatic environment.

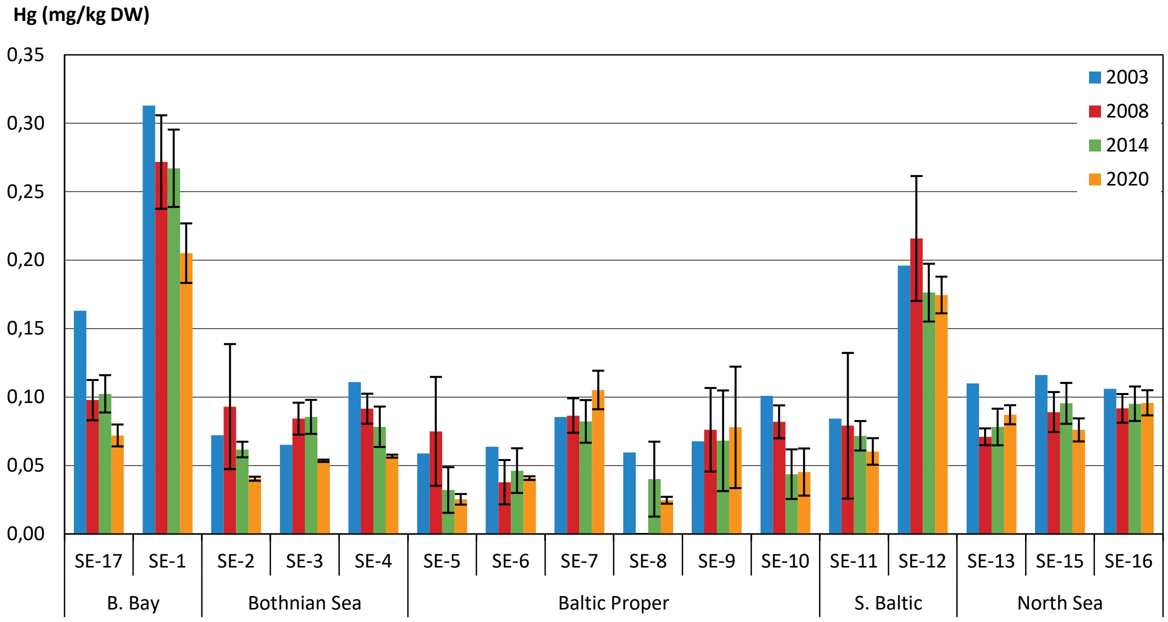

Figure 11. Levels of mercury (Hg) at the 16 stations during the four monitoring years. For 2008, 2014 and 2020 the average value ± 1 standard deviation is displayed; *n* = 7 except for SE-5 in 2014 when *n* = 5 and SE-6 in 2020 when *n* = 6. The laboratory reporting limit for mercury is close to the level occurring in sediment. Levels were below the reporting limit in one or more replicates at stations SE-5, SE-6, SE-9 and SE-11 in 2008 and at SE-5, SE-6, SE-8, SE-9 and SE-10 in 2014. No level could be reported for station SE-8 in 2008 as all seven replicates had mercury levels below the limit of reporting of 0.05–0.08 mg/kg TS. In 2020 a low-level analytical method was used for samples with low levels of mercury, and no replicates were thus below the reporting limit.

Nickel

Nickel is used for a variety of purposes including as a coating agent to protect against corrosion. It mainly occurs in stainless steel but is also found in batteries and catalysts.

Nickel levels in sediment differ relatively little between the different stations and sea areas (Fig. 12). The stations with the highest levels are SE-7 and SE-9 in the Baltic Proper and SE-1 in the Bothnian Bay, but overall, differences between sea areas are low.

Like copper, lead, cadmium and zinc, nickel forms sulphides in reduced sediment. Nickel sulphide is however more readily soluble than sulphides of the other metals (Di Toro *et al.* 1990, Ankley *et al.* 1996), which may explain why nickel does not follow the same geographical pattern as cadmium and copper, i.e. elevated levels in sediments in the Baltic Proper. An earlier study showed that the levels of nickel were higher in sediment in the Baltic Proper than in the Bothnian Sea, and were also correlated with sediment content of organic material (Borg & Jonsson 1996). A positive and significant linear relationship between levels of nickel and levels of TOC in the sediment was also observed in this study, but the degree of correlation was very low ($r^2 = 0.07$, $p = 0.04$, $n = 59$).

Levels of nickel in sediment were higher in 2014 and 2020 compared to earlier years for many, but not all stations. This might be caused by a change in the analytical extraction method between 2008 and 2014, although leaching with 7 M HNO_3 is not expected to give higher yields than the method used in 2003 and 2008 (melting with $LiBO_2$ followed by leaching). In the Swedish environmental monitoring of marine biota, no general trend over time could be seen for nickel over the last decade although there were indications of decreasing concentrations of nickel in some species (2010–2019; Soerensen & Faxneld 2020).

Nickel is a priority substance in the WFD but there is no EQS for sediment. It was not included as an indicator in the second holistic assessment of the environmental status of the Baltic Sea (HELCOM 2018b) and is not proposed to be included as an indicator in HOLAS III. The reference value assumed to represent pre-industrial values is 30 mg/kg DW for chemical analysis according to the Swedish standard method that was used in 2014 and 2020, or 33 mg/kg DW for the total analysis method that was used in 2003 and 2008 (SwEPA 1999). The measured levels did not exceed the reference value at any station in 2003, but in 2008 levels exceeded this value at six stations, in 2014 at twelve stations and in 2020 at thirteen stations (Fig. 12).

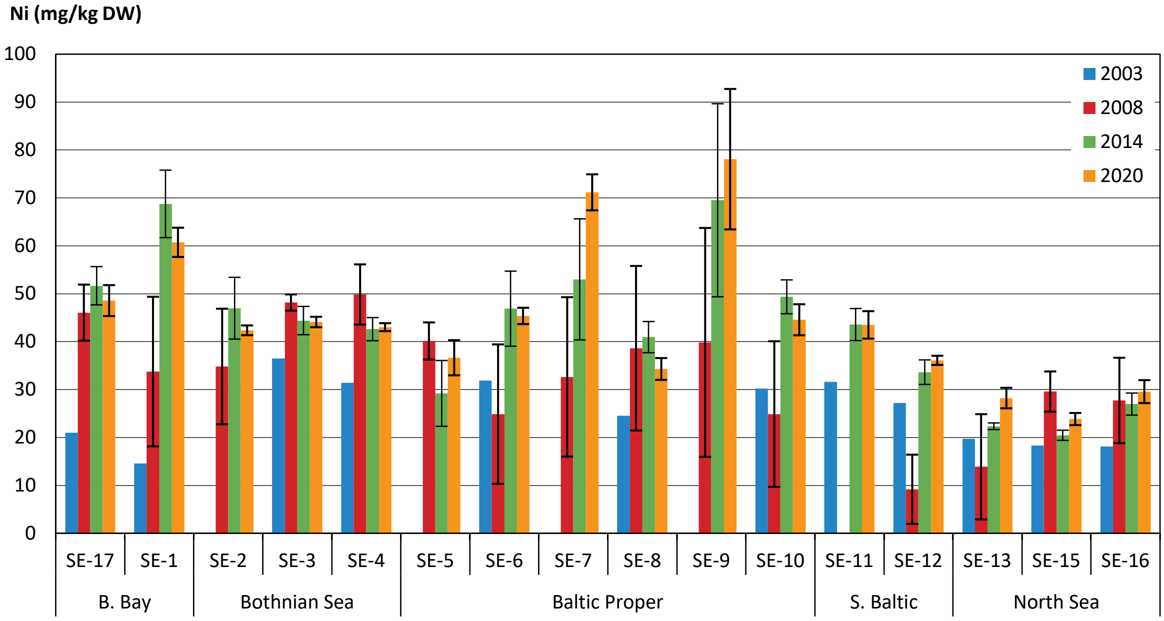

Figure 12. Levels of nickel (Ni) at the 16 stations during the four monitoring years. For 2008, 2014 and 2020 the average value ± 1 standard deviation is displayed; *n* = 7 except for SE-5 in 2014 when *n* = 5 and SE-6 in 2020 when *n* = 6. In 2003 nickel levels at four of the stations were below the reporting limit of the single sample that was analysed for each station, and in 2008 the levels in all seven samples of SE-11 were below the reporting limit (10 mg/kg DW). At stations SE-6 – SE-10 and SE-12 – SE-13, the levels of nickel were below the reporting limit in one or more of the replicates from each station in 2008.

Lead

Lead is a heavy metal that nowadays is used mainly in car batteries and other accumulators, but also in for example ammunition. The ban on the use of lead in petrol caused a large reduction in emissions from the transport sector since the early 1990s. Total emissions in Sweden have also been greatly reduced and the largest sources are now industrial emissions from the metal industry and combustion of fossil fuels.

Levels of lead in sediment are clearly elevated at stations SE-12 in the Southern Baltic and SE-1 in the Bothnian Bay, followed by SE-11 in the Southern Baltic (Fig. 13). Overall, levels of lead tend to be highest in the southern part of the Baltic Sea. The geographical pattern of lead is similar to mercury which also occurs in high levels at stations SE-1 and SE-12 (Fig. 11), probably caused by an influence from point sources. An earlier study by Borg and Jonsson (1996) also showed a correlation between lead and mercury in the Baltic Sea. The difference is that lead also occurs at elevated levels at station SE-11, and that the levels at SE-12 are generally slightly higher than at SE-1. Elevated levels at SE-1 may be caused by regional metal industry, such as a nearby smelter known to be a source of lead and other elements to sediment (Cato & Sellén 2004). The cause of elevated levels of lead at stations SE-12 and SE-11 is however unknown, as is the cause of elevated levels of mercury at station SE-12.

Concentrations of lead in marine biota sampled within the environmental monitoring programme are generally higher in the Southern Baltic than in the northern parts of the Baltic Sea or on the Swedish West Coast, and there are also indications of increasing concentrations in the Southern Baltic (2010–2019; Soerensen & Faxneld 2020). High levels of lead in the Southern Baltic are thus apparent in both sediment and biota, however the high levels in sediment at SE-1 are not reflected in the biota data from the Bothnian Bay.

Like cadmium, copper, nickel and zinc, lead forms weakly soluble sulphides in reducing sediments (Di Toro *et al.* 1990, Ankley *et al.* 1996), and it is therefore surprising that lead, unlike these metals, occurs in lower levels in the Baltic Proper sediments than in other sea areas. One reason for this is probably due to a larger influence from point sources on the levels of lead. Levels in sediments display a significant negative linear relationship with TOC but with a very low degree of correlation ($r^2 = 0.10$, $p = 0.009$, $n = 64$). The correlation is mainly driven by lower levels of lead and higher levels of TOC in the Baltic Proper, which was also the case for mercury. If the Baltic Proper stations are excluded, the correlation becomes positive ($r^2 = 0.55$, $p < 0.0001$, $n = 40$).

There are no consistent trends in lead levels in sediments between the four monitoring years. In environmental monitoring of marine biota, there is no clear overall trend for lead over the last decade although there is a tendency to decreasing trends in many species (2010–2019; Soerensen & Faxneld 2020).

Lead is a priority substance according to the WFD and the EQS for marine sediments is 120 mg/kg DW (SwAM 2019). This is also the proposed threshold value for lead in sediment as an indicator in HOLAS III. This value is not exceeded at any station. Lead was included as an indicator in the second holistic assessment of the Baltic Sea and often exceeded the threshold values for biota and sediment contributing to a reduced environmental status, although not to the same extent as mercury (HELCOM 2018b).

The national reference value for lead, representing pre-industrial levels in sediments, is 25 mg/kg DW (for chemical analysis according to the Swedish standard method that was used in 2014 and 2020) or 31 mg/kg TS (for the total analysis method that was used in 2003 and 2008; SwEPA 1999). This value has been exceeded at seven stations or more every monitoring year.

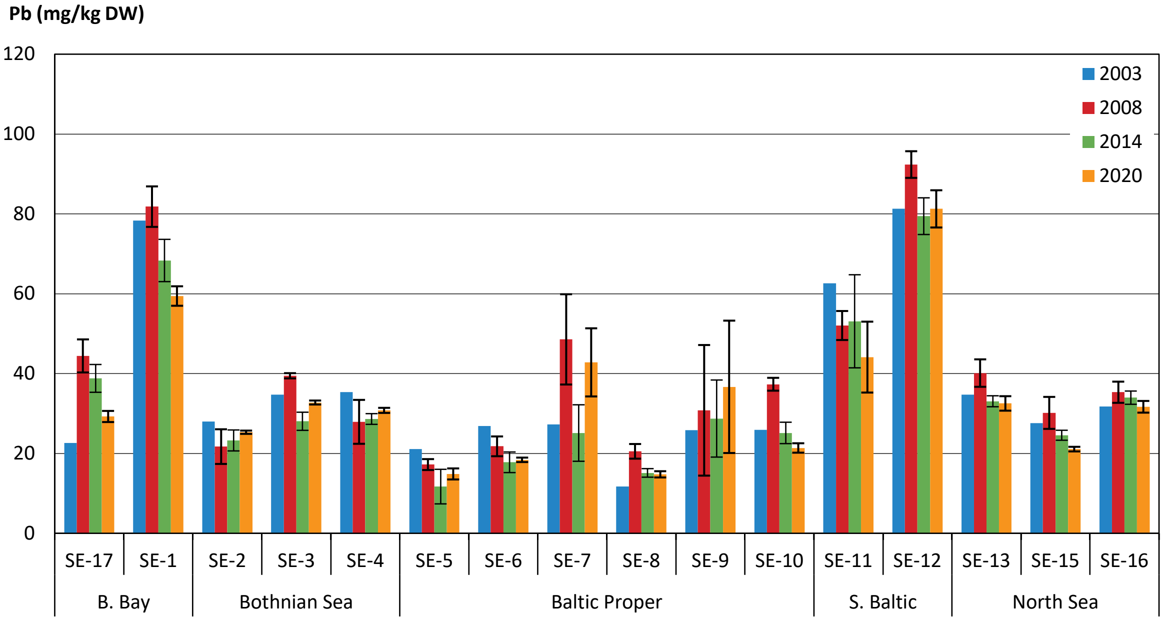

Figure 13. Levels of lead (Pb) at the 16 stations during the four monitoring years. For 2008, 2014 and 2020 the average value ± 1 standard deviation is displayed; *n* = 7 except for SE-5 in 2014 when *n* = 5 and SE-6 in 2020 when *n* = 6.

Zinc

Zinc is a commonly used metal, for example in the metal industry for galvanizing, for brass, and for a variety of other purposes such as in paints and tires.

The highest zinc levels in sediments occur in the Baltic Proper, followed by the Bothnian Bay (Fig. 14). Levels in the Bothnian Sea and the Southern Baltic are similar, while they are clearly lower in the North Sea. Zinc thus has a similar geographical distribution as cadmium and copper, with the highest levels in the Baltic Proper. Similar to these substances, zinc forms weakly soluble sulphides in reducing sediments (Di Toro *et al.* 1990, Ankley *et al.* 1996), which may explain the geographical pattern. Of the stations not located in the Baltic Proper, SE-1 in the southern Bothnian Bay always has the highest levels, which may be related to the proximity of a large smelter (Cato & Sellén 2004). Results from environmental monitoring of marine biota shows similar zinc concentrations in herring in the sea basins around Sweden, i.e., no large geographical variation (2017-2019; Soerensen & Faxneld 2020).

Like cadmium and copper, the levels of zinc in sediment have a significant positive correlation with the TOC levels, but the degree of correlation is lower for zinc ($r^2 = 0.39$, $p < 0.0001$, $n = 64$). As already mentioned, the correlation may be related to the fact that stations with high TOC content also have reducing conditions and hence formation of sulphides, however there is still a significant and positive correlation even when the anoxic Baltic Proper stations are excluded ($r^2 = 0.35$, $p < 0.0001$, $n = 40$).

There is no consistent trend in zinc levels over the four sampling years. The levels are highest in 2008 but the reason for this is unclear because the same analytical extraction method has been used for all years. Higher zinc levels in 2008 are particularly marked at stations in the Baltic Proper, which is also the case for cadmium and copper.

Zinc is listed as an RBSP in Sweden according to the WFD, but there is no EQS for sediment. The substance was not included in the second holistic assessment of the Baltic Sea (HELCOM 2018b) and is not proposed as an indicator in HOLAS III. The national reference value, which is assumed to represent pre-industrial levels, is 85 mg/kg DW and is exceeded at all stations during all years, except SE-15 and SE-16 during some years (Fig. 14).

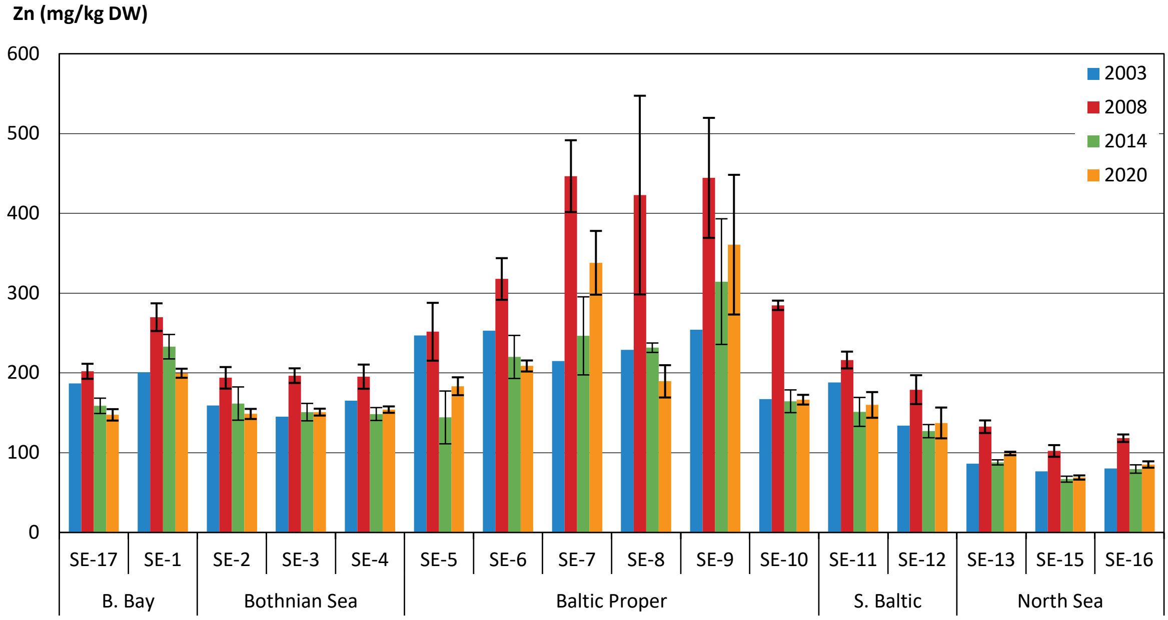

Figure 14. Levels of zinc (Zn) at the 16 stations during the four monitoring years. For 2008, 2014 and 2020 the average value ± 1 standard deviation is displayed; *n* = 7 except for SE-5 in 2014 when *n* = 5 and SE-6 in 2020 when *n* = 6.

Other elements

To show the distribution of the elements that are not displayed in separate diagrams, a principal component analysis was performed (Fig. 15 and 16). All analysed elements were included except for some rare earth elements (REEs) and seven elements only measured in 2020 at levels below the LOQ: Ir, Pd, Pt, Re, Rh, Ru and Te (the last >LOQ for one sample). Some of the REEs were excluded to facilitate the visual interpretation of the PCA. REEs encompass 14 lanthanides (excluding the unstable promethium), scandium and yttrium. A correlation analysis showed that these 16 elements were very well-correlated with each other, except for Lu. Thus, only the light REEs La and Eu, and the heavy REEs Er, Lu, Sc and Y were included in the PCA. The three parameters TOC, loss-on-ignition (LOI) and O_2 were included as supplementary variables, i.e., not included in the calculation of principal components but present in the loading plot to facilitate interpretation of the results.

Most of the variation (50%) can be explained by the first principal component. This shows the difference between the anoxic Baltic Proper stations with high levels of Cd, Cu and Zn to the left in the diagram. These stations also display high levels of Mo, Na, Sb, Se, S, Tl and U, as well as TOC and LOI. Conversely, they have low levels of elements to the right in the loading plot, such as the REEs, Al, Cr, Ti, Li etc., compared to other stations.

The second principal component, displayed in the vertical direction, explains 15% of the variation in the data and mainly shows the difference between stations in the other basins than the Baltic Proper. In the score plot, stations in the Bothnian Bay and the Bothnian Sea are located in the upper half of the plot, stations in the Southern Baltic slightly below, and stations in the North Sea at the bottom part of the plot. The North Sea stations rarely have high levels of elements, but Ca, Hf and Zr occur at their highest levels at these stations. The element B, only analysed in 2020, also occurs at high levels in the North Sea; however, the highest levels of boron can be found at the two stations in the Southern Baltic. At the opposite end of the loading plot, in the top right, are the elements occurring at their highest levels at stations in the Bothnian Bay: As and Co (see also Fig. 6 and 8), Ba, Bi and Mn. P and Ni tend to have rather even levels at all stations, but with slightly lower levels at the North Sea stations. Finally, some elements occur in a pattern with the highest levels in the Bothnian Sea. This is most pronounced for Cs and Li but also for, for example, Al.

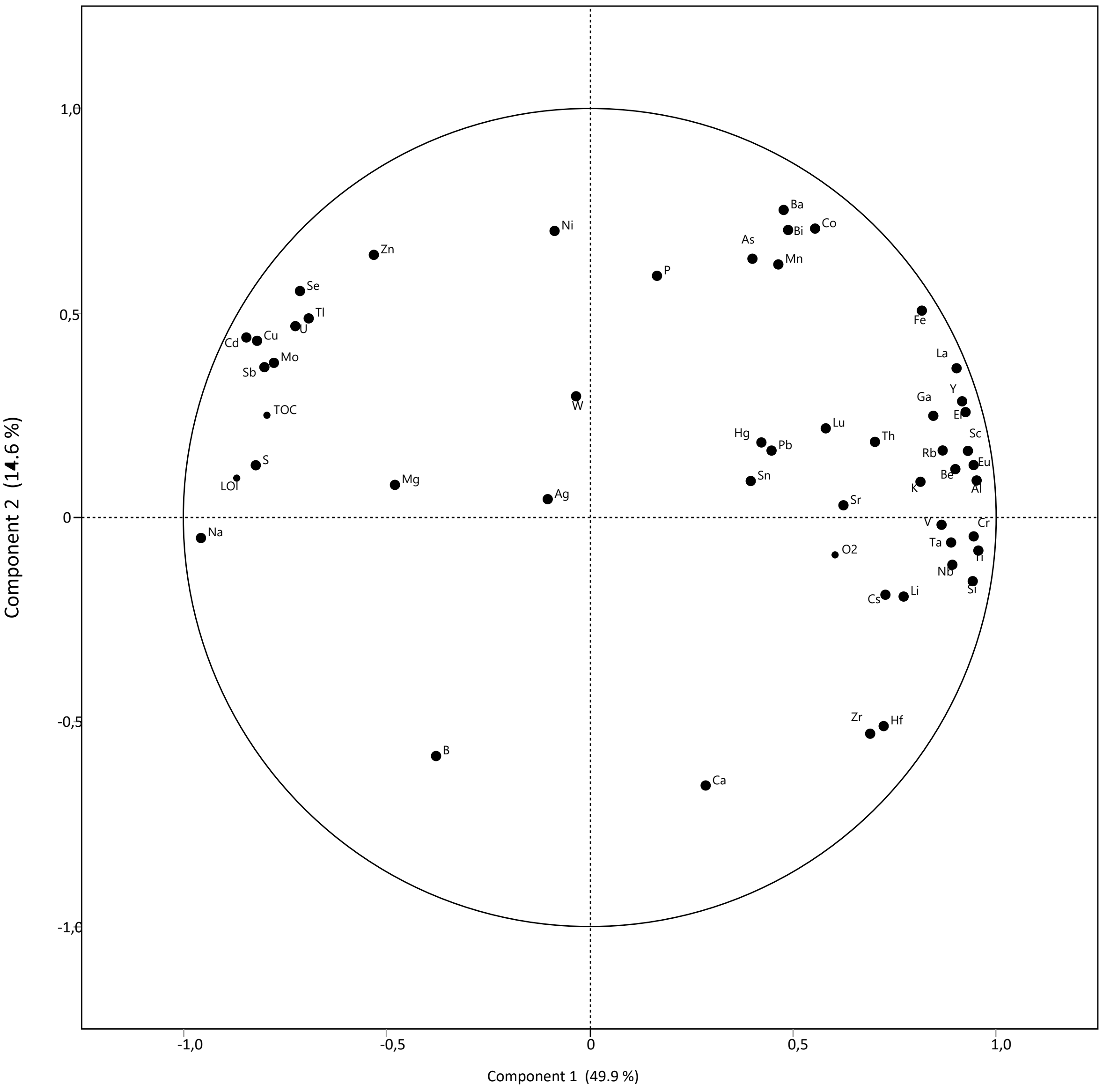

Figure 15. Loading plot from a principal component analysis (PCA) showing which variables (elements) that affect the components. The plot also illustrates the co-variation among the variables. TOC, loss-on-ignition (LOI) and O_2 were included as supplementary variables, i.e., not included in the calculation of principal components.

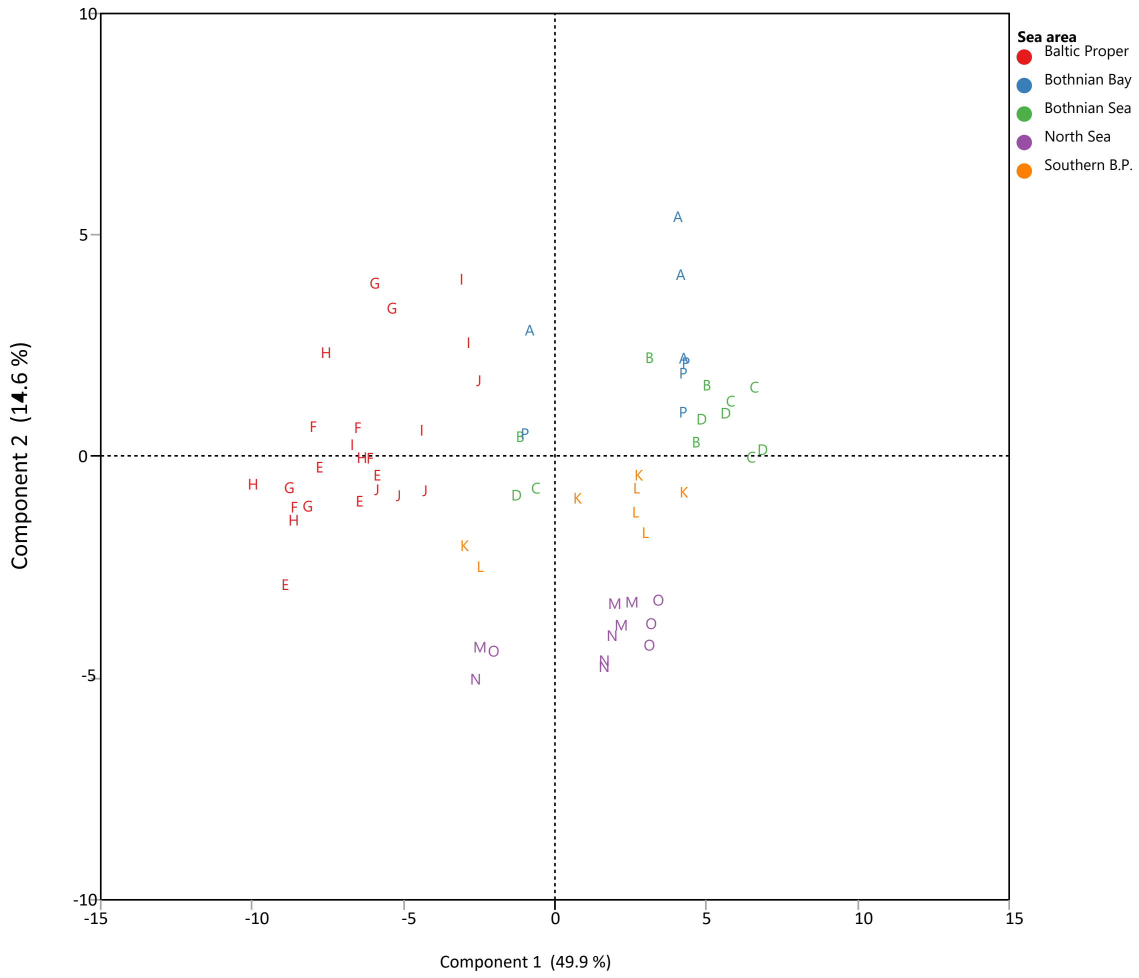

Figure 16. Score plot from a principal component analysis (PCA) showing how the 16 monitoring stations for each year relate to the two first principal components, and to each other. Colours show the different sea areas, while the letter corresponds to a station: A = SE-1, B = SE-2 etc. Because each station has been monitored four times, the letters occur four times. Usually, the data for 2020 is deviating from the three other years due to more elements having been analysed in the monitoring.

Comparison of extraction methods for elements

During the 2020 sampling, a comparison of extraction methods was performed. The purpose was to determine if it is possible to change methods for future environmental monitoring surveys. The comparison was carried out by analysing a pooled sample from each station using an alternative extraction method available at the laboratory (either $LiBO_2$-melt or total dissolution with an acid mix of HNO_3/HCl/HF) and comparing the results to the average for each station analysed using the usual methods in the national monitoring of offshore sediments. The usual methods are $LiBO_2$-melt, leaching using aqua regia or leaching using 7 M HNO_3. For some elements, $LiBO_2$-melt was thus used in both the usual and the alternative extraction method. In the alternative method, both $LiBO_2$-melt and total dissolution with HNO_3/HCl/HF was used to extract the samples, and the laboratory then choose the optimal method for the specific sample. For that reason, for some elements only the HNO_3/HCl/HF-mix was reported, for some only the $LiBO_2$-melt, and for some a mix, i.e., different methods for different stations for the same substance (Table 3).

The results show, unsurprisingly, that for elements usually extracted with $LiBO_2$-melt, the use of alternative extraction methods in the form of $LiBO_2$-melt or HNO_3/HCl/HF-mix was satisfactory (Table 3). For four elements (Be, K, Nb, Zr), the results with the alternative extractions seemed to overestimate the levels. Three of these mainly had the acid mix as the alternative method. Beryllium showed the most deviation from the standard method, both in terms of average recovery and a high standard deviation, which can be explained with environmental levels being relatively close to the reporting limit (occurring at levels from below the reporting limit of 0.5 mg/kg DW to 4.0 mg/kg DW).

For elements usually extracted with 7 M HNO_3, which is a milder acid than the HNO_3/HCl/HF-mix used for total dissolution of minerals, Hg and Pb had substantially higher recoveries using the stronger method. During the usual analysis of the 2020 samples, an additional low-level method was used for Hg, which meant that all samples had levels above the reporting limit. This was not the case for the total dissolution method – some samples were below the reporting limit even though levels were higher due to the stronger dissolution of the sample matrix. Cadmium, which is one of the substances usually extracted using 7 M HNO_3, displayed lower levels (88%) and a larger uncertainty when samples were extracted using the total dissolution method. The lower values were unexpected and could possibly be explained by complexing with chloride, as HCl is included in the acid mix (G. Morris, personal communication, April 20, 2022).

For the substances usually extracted with aqua regia, higher values were generally achieved using the HNO_3/HCl/HF-mix, which is not surprising. The recovery was particularly high for Sn, which may be due to stronger complexing with chloride in the usual aqua regia extraction (G. Morris, personal communication, April 20, 2022). Silver had an average recovery very close to 100%, i.e., the levels using total dissolution were similar to aqua regia; however, the uncertainty was very large. This is even when excluding the three samples that had levels below the reporting limit using the alternative method with total dissolution. The alternative method is thus not satisfactory in comparison to the usual method, even though the average recovery is good.

In summary, changing the extraction methods to $LiBO_2$ or total dissolution would yield higher values for some substances, which is to be expected. Surprisingly, cadmium tended to have lower levels using total dissolution, possibly due to complexing. Before changing methods, however, the relevance of measuring elements that are forming part of a mineral and not being readily available for uptake by biota should be considered.

Table 3. Comparison of extraction methods for elements. The recovery was calculated as the value using the alternative method divided by the average value (*n* = 7, except for SE-6 where *n* = 6) for each station using the usual method. This resulted in 16 recoveries for each substance. An average recovery and a standard deviation were then calculated for each substance. Substances with average recoveries using the alternative method that was within 10% of the usual methods are marked with green, within 10–20% are marked with yellow, and >20% are marked with red. For the alternative method, only samples with values above reporting limits were included. For comparison, loss on ignition (LOI) was also analysed for all samples, and the average recovery ± 1 SD was 99 ± 5%.

Element	Average recovery (%)	SD	No. of samples acid mix/$LiBO_2$
Usual method: Aqua regia; alternative method: HNO_3/HCl/HF-mix			
Ag	98	30	16/0
Li	120	11	16/0
Mo	111	21	16/0
Sb	107	15	16/0
Sn	142	11	16/0
Usual method: 7 M HNO_3; alternative method: HNO_3/HCl/HF-mix[a]			
As	108	7	16/0
Cd	88	13	16/0
Co[a]	115	8	5/11
Cu	105	7	16/0
Ni	111	9	16/0
Hg[b]	135 (168)	31 (38)	16/0
Pb	130	11	16/0
S	104	4	16/0
Zn	106	4	16/0
Usual method: $LiBO_2$; alternative method: $LiBO_2$-melt and HNO_3/HCl/HF-mix			
Al	100	6	8/8
Ba	101	5	5/11
Be	120	27	11/5
Ca	108	9	0/16
Cr	108	11	1/15
Fe	94	6	1/15
K	113	12	15/1
Mg	109	8	3/13
Mn	109	13	4/12
Na	103	5	9/7
Nb	112	7	10/6
P	101	9	5/11
Sc	107	8	11/5
Si	97	7	0/16
Sr	101	5	6/10
Ti	104	8	5/11
V	109	9	3/13
W	109	13	9/7
Y	101	5	5/11
Zr	116	11	0/16

[a] For Co, mainly $LiBO_2$-melt was used as the alternative extraction method

[b] For Hg, samples from 2020 were also analysed with a special low-level method (using AFS). The comparison to this method is given within parentheses and thus shows an even larger exceedance using the alternative method.

ORGANIC CONTAMINANTS

PAHs

Polycyclic aromatic hydrocarbons (PAHs) are formed during incomplete combustion of organic material. They can be formed by, for example, forest fires but they also have an anthropogenic origin especially from the combustion or release of fossil fuels. High levels occur in tar, mainly coal tar and related products such as creosote, which were previously used extensively. More recently, the use of scrubbers to reduce the atmospheric emissions of sulphur oxides from ships has led to more emissions of PAHs and other contaminants such as heavy metals directly to surface water (Hermansson *et al.* 2021).

The sixteen PAHs constituting the so-called EPA-PAHs have been analysed during all four monitoring years, except for acenaphthylene which was not analysed in 2008. Levels of the sum of the sixteen EPA-PAHs, $\sum PAH_{16}$, are lower in the Bothnian Bay and Bothnian Sea than in the Baltic Proper and the Southern Baltic (Fig. 17). The levels show a positive and significant linear relationship with TOC levels in the sediment, which would be expected, but the degree of correlation is very low ($r^2 = 0.06$, $p = 0.047$, n = 64). Sea areas that have deviating levels compared to the correlation with TOC are the Southern Baltic, which have elevated levels, and the Bothnian Bay and the Bothnian Sea, which tend to have lower levels. The highest levels were measured at station SE-12 in the Arkona Basin in the Southern Baltic for all four years, showing that there are more significant PAH sources at this station than at other stations. It is possible that this is related to more intensive shipping at this station (see www.helcom.fi), but SE-12 also has high levels of organic substances not linked to shipping, such as PCDD/Fs and PCBs, and of elements such as mercury and lead. Studies have pointed to the importance of the river Oder as a source of PAHs for the Arkona Basin (Schulz & Emeis 2000, Witt *et al.* 2001).

The PAH distribution pattern, i.e., how much the different PAHs contribute to $\sum PAH_{16}$, is similar between sea areas but with a larger fraction of low-molecular weight PAHs at stations SE-15 and SE-16 in the North Sea (Fig. 18). PAHs with three carbon rings or less (naphthalene to anthracene) and PAHs with four rings (fluoranthene to chrysene) constitute a larger proportion at these stations, while PAHs with six rings (benzo[g,h,i]perylene and indeno[1,2,3-c,d]pyrene constitute a smaller proportion. A larger fraction of low-molecular PAHs could indicate a more recent release of PAHs because the low-molecular PAHs, which overall have shorter half-lives, have not had time to degrade. It could also indicate different sources of PAHs at these stations compared to the Baltic Sea stations.

PAH levels decreased between 2003 and 2008 at most stations (Fig. 17), but this decrease is probably mainly due to a change in analytical laboratory and method rather than actual reductions in levels in the environment. The deposition of benzo[a]pyrene in the Baltic Sea region has been modelled and shows decreasing deposition in the early 1990s, which has since stabilized and no longer decreases (1990–2014; HELCOM 2018a). Comparing levels in sediment between the two later monitoring years, levels are similar or increase in the Bothnian Bay, Bothnian Sea and North Sea, and decrease in the Southern Baltic (Fig. 17). In the Baltic Proper, the stations SE-5, SE-6 and SE-8 display decreasing levels between 2014 and 2020/2021. These are stations with fast sediment accumulation rates (SARs; around 1 year to form an average one-centimetre layer of sediment, see Table 2), thus responding faster to changes in environmental levels. Stations SE-7, SE-9 and SE-10, which are stations with low SARs (13–22 years to form an average one-centimetre layer of sediment), display similar or increasing levels between the years. PAH levels in blue mussels (*Mytilus edulis*) have been measured at a station in the Baltic Sea and two stations in the North Sea within the national environmental monitoring of marine biota, and there is no overall trend for PAHs for the last decade (2010–2019; Soerensen & Faxneld 2020). At the Baltic Sea station there was a general decrease while at the North Sea stations increasing or non-significant trends occurred.

Eight PAHs are priority or priority hazardous substances according to the WFD: naphthalene, anthracene, fluoranthene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, benzo[g,h,i]perylene and indeno[1,2,3-c,d]pyrene. There are EQS for sediment for two PAHs: anthracene (24 μg/kg DW) and fluoranthene (2000 μg/kg DW; SwAM 2019). Both values apply for a TOC content of 5% in the sediment. Fluoranthene levels do not exceed the EQS at any station for any year, while anthracene levels exceed the EQS at the three stations in the North Sea in 2003 and at the two northernmost stations in the North Sea in 2020/2021. In 2008 and 2014, no station exceeds the EQS. Both substances are also proposed indicators for HOLAS III with sediment threshold values set at 24 and 3500 μg/kg DW for anthracene and fluoranthene, respectively, for sediment with 5% TOC. This is thus the same as the EQS for anthracene, while the proposed fluoranthene threshold is higher. Anthracene therefore exceeds the proposed sediment threshold value for HOLAS III at some stations, while fluoranthene never exceeds the threshold at the 16 monitoring stations. Benzo[a]pyrene is also a proposed indicator for HOLAS III, but with a threshold value only for biota. In the second holistic assessment, when these three PAHs were also included as indicators, anthracene was the substance most often present at levels that exceed sediment thresholds, primarily in Öresund and the southern parts of the Baltic Sea (HELCOM 2018b).

Four PAHs (naphthalene, benzo[a]pyrene, benzo[b]fluoranthene and benzo[k]fluoranthene) have less certain indicative values for sediment published by SwAM (2018b; see section on *Environmental quality standards and reference values*). These values are never exceeded for naphthalene but for several stations each year for the other three substances, most frequently for benzo[b]fluoranthene. In 2020/2021, the indicative value for benzo[b]fluoranthene was exceeded at all stations in the North Sea, Southern Baltic, Bothnian Sea, and in addition at station SE-9. The value for benzo[k]-fluoranthene was exceeded at the three stations in the North Sea and at station SE-12 in the Southern Baltic, while the value for benzo[a]pyrene was exceeded at all three stations in the North Sea.

In summary, the North Sea and the Southern Baltic are the most problematic areas in terms of potentially toxic levels of PAHs. Given that EQS and indicative values for PAHs are exceeded even in offshore sediments and that the atmospheric deposition no longer decreases according to modelling studies, further measures will probably need to be taken to lower the levels in the environment.

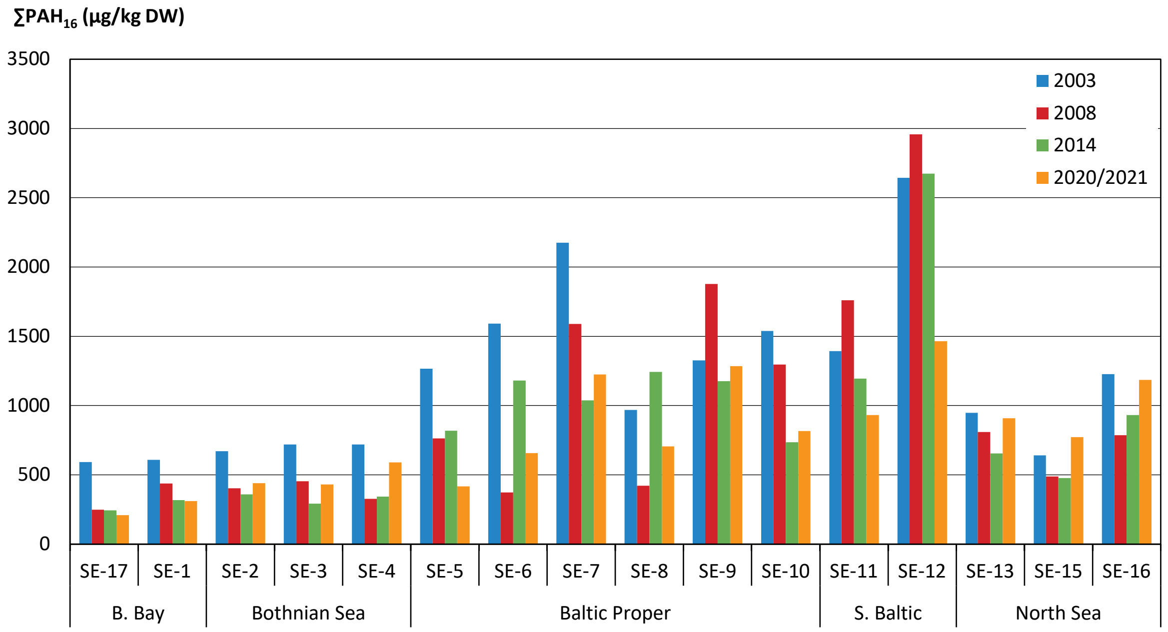

Figure 17. Levels of $\sum PAH_{16}$ during the four monitoring years at the 16 stations. Note that the value for station SE-5 in 2020/2021 is less certain as the sample had been exposed to temperatures above 0°C for an extended time. In 2008, acenaphthylene was not included in the analyses, but the average difference between $\sum PAH_{15}$ and $\sum PAH_{16}$ for the other three monitoring years at each station was used to correct the 2008 values for the lack of acenaphthylene.

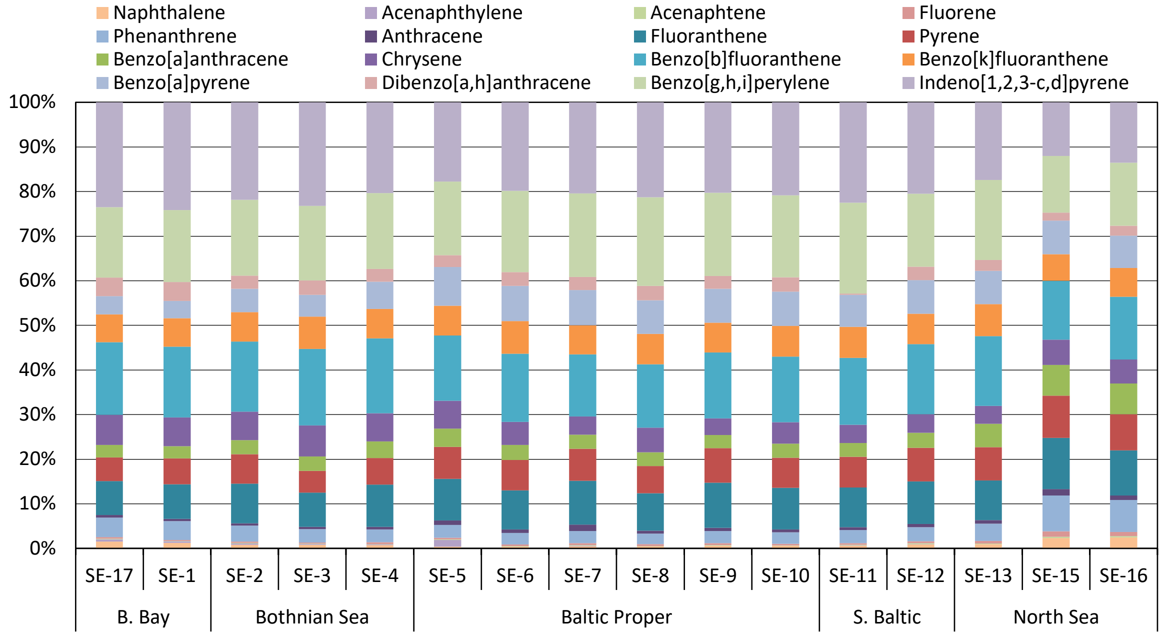

Figure 18. The distribution of the substances constituting $\sum PAH_{16}$ at the 16 monitoring stations in 2020/2021.

PFASs

Per- and polyfluoroalkyl substances (PFASs) are widely used due to their oil- and water-repellent properties and stability, for example against high temperatures. They are unfortunately also very mobile, persistent and bioaccumulating in the environment, and some PFASs are toxic. So far, the substances PFOS and PFOA have been listed in the Stockholm Convention on Persistent Organic Pollutants (POPs) as substances whose use should be eliminated or restricted, but many other PFAS substances are believed to have toxic properties and some are considered for inclusion in the Stockholm Convention.

PFASs were included in the monitoring programme in 2014 and analyses included 11 substances: the four perfluorosulphonic acids (PFSAs) PFBS, PFHxS, PFOS, PFDS; the six perfluorocarboxylic acids (PFCAs) PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA; and PFOSA. These are not the exact same eleven PFASs that are relevant in present ground and surface water legislation[1]. In 2020/2021, PFBA, PFPeA and 6:2 FTSA were also included in the analyses, but to be able to compare results between years, only the initial PFASs analysed are summed as ∑PFAS. Of the analysed substances, the levels of PFOSA and 6:2 FTSA are always below reporting limits: 0.3 and 0.05 µg/kg DW for PFOSA in 2014 and 2020/2021, respectively, and 0.05–0.08 µg/kg DW for 6:2 FTSA in 2020/2021. Some of the other substances were below reporting limits in 2014 but could be measured in 2020/2021 due to lower reporting limits.

The levels of PFASs are substantially higher in Bothnian Bay and Bothnian Sea sediments than in other sea areas (Fig. 19). Compared to average levels for stations in other sea areas in 2020/2021 (1.5 µg/kg DW), levels are 8–17 times higher (on average 14) at Gulf of Bothnia stations. The dominant substances at these stations are PFNA, PFOA and PFOS, while dominant substances in other sea areas are PFUnDA and PFOS (Fig. 20). Comparing average levels of each compound at Gulf of Bothnia stations to average levels in other sea areas (averages only including samples with detected levels), levels are particularly elevated for PFNA (42 times higher), PFOA (30 times higher), PFHpA (16 times higher), PFOS and PFDA (both 10 times higher). Levels of PFHxS are on average 2 times higher in the Gulf of Bothnia compared to other sea areas, while levels of PFUnDA, PFPeA, PFDS and PFBS are more similar. Levels of PFBA and PFHxA could not be compared due to all stations outside the Gulf of Bothnia having levels below the detection limit. The stations in the Bothnian Bay and the station in the Åland Deep (SE-4) had the highest levels while remaining Bothnian Sea stations had slightly lower levels both monitoring years.

The clearly elevated levels in the Gulf of Bothnia are surprising, as the northern parts of the Baltic Sea tend to have lower levels of organic contaminants. Furthermore, studies of PFASs in surface waters of the Baltic Sea and the North Sea instead point to generally lower concentrations in the Gulf of Bothnia than in other sea areas (Nguyen *et al.* 2017, Kirchgeorg *et al.* 2010). Studies on herring have pointed to lower levels in the northern parts of the Baltic Sea (Faxneld *et al.* 2016), and studies in watercourses in Sweden indicate generally lower levels in watercourses in the northern parts of Sweden (Ahrens *et al.* 2014, Ahrens *et al.* 2018). On the other hand, extensive metal as well as paper industries are present along the coast of the Gulf of Bothnia and are large potential sources of various PFASs. Data from the monitoring programme for contaminants in marine biota for the last decade shows that concentrations of PFUnDA tend to be highest in herring from the Bothnian Bay, followed by the Baltic Proper (southern and northern), the Bothnian Sea and the North Sea (2010–2019; Soerensen & Faxneld 2020). Concentrations of PFOS, on the other hand, were higher in herring from the Baltic Proper and lower in herring from the Gulf of Bothnia or the North Sea. The results are thus generally contradictory to the pattern seen in sediments. Although the concentration of PFUnDa was highest in herring from

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[1] The PFASs included in the $\sum$PFAS$_{11}$ of relevance for ground and surface water legislation are PFBS, PFHxS, PFOS, PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA and 6:2 FTSA.

the Bothnian Bay, this substance does not show elevated levels in sediments in the Gulf of Bothnia, while PFOS did.

Comparisons with other studies are complicated, as the number of analysed PFASs and reporting limits vary, but are valuable to put the levels at the monitoring stations into perspective. In a study along the German Baltic and North Sea coasts, ∑PFAS levels (16 detected PFASs) ranged from 0.018 to 2.6 µg/kg DW (Joerss *et al.* 2019). Levels in the Baltic Sea and Skagerrak/Kattegat areas were 0.036–1.6 and 0.20–1.3 µg/kg DW, respectively, which resembles levels at the monitoring stations in the Baltic Proper, Southern Baltic and North Sea, where the range of ∑PFAS are 0.41–2.4 µg/kg DW in 2020/2021. ∑PFAS levels at the Gulf of Bothnia monitoring stations are however substantially higher, 12–26 µg/kg DW. The dominant compounds in the German study varied depending on sea area, but PFOS tended to be the most dominant, followed by PFBA and longer-chained PFCAs. This differs somewhat from the Gulf of Bothnia samples, where the dominant substances were PFOS, PFOA and PFNA, but not PFBA.

∑PFAS levels at the less contaminated monitoring stations are also similar to or slightly higher than levels in offshore sediment in the Arctic and Bering Seas (0.06–1.73 µg/kg DW, dominant compounds being PFOS, PFNA and PFBS; Lin *et al.* 2020), along the coast of South Korea (0.045–1.13, dominant compounds being PFOS, PFOA, PFUnDA and PFTrDA; Lee *et al.* 2020) or along the coast of Spain (only PFOA and PFOS above detection limits with maximum levels of 0.27 and 0.17 µg/kg DW; Leon *et al.* 2020). Corresponding ranges of PFOA and PFOS are <0.05–0.42 and 0.23–1.1 µg/kg DW, respectively, at the monitoring stations in the Baltic Proper, Southern Baltic and North Sea, and 3.0–6.8 and 3.3–7.5 µg/kg DW in the Gulf of Bothnia.

Studies of coastal sediment in China showed levels of ∑PFAS of 0.70–4.1 µg/kg DW in the Bohai Sea (Zhao *et al.* 2020), 1.5–2.7 µg/kg DW in the Dalian Bay (Ding *et al.* 2018), and 7.5–84 µg/kg DW in the South China Sea (Wang *et al.* 2019). The highest values in the later study are higher than levels at monitoring stations in the Gulf of Bothnia and were found in regions with industrial release. Levels at Gulf of Bothnia monitoring stations are also similar or lower than ∑PFAS levels found in a Swedish lake and pond contaminated from aqueous film forming foams (AFFFs) used in fire-fighting (<1–76 µg/kg DW in sediment cores, dominant substances PFOS and PFHxS; Mussabek *et al.* 2019) or in waterbodies in New Jersey, USA (<LOD–30.9 µg/kg DW, highest levels in areas affected by AFFFs (Goodrow *et al.* 2020), or the Great Lakes (mean concentrations of ∑PFAS 16–19 µg/kg DW in the three investigated lakes; Codling *et al.* 2018). Gulf of Bothnia levels are however clearly lower than the very high levels found in two urban US watersheds (∑PFAS around 300 µg/kg DW; Bai & Son 2021), a semi-enclosed, industrialised Korean bay (∑PFAS 6.6–821 µg/kg DW; Shen *et al.* 2018) or close to a factory producing paper products in Norway (mean levels of PFCAs and PFSAs 160 and 250 µg/kg DW, respectively, compared to a range of 9–20 µg/kg DW for PFCAs and 4–8 µg/kg DW for PFSAs in the Gulf of Bothnia; Langberg *et al.* 2020). Corresponding mean levels in Lake Tyrifjorden, downstream of the Norwegian factory, are 6.2 and 4.4 µg/kg DW, which is generally lower than in the Gulf of Bothnia sediments. The clearly predominant PFAS compounds in the Norwegian study, however, were sulfonamides and fluorotelomer sulfonic acids (Langberg *et al.* 2020). In the environmental monitoring, the analysed compounds PFOSA and 6:2 FTSA are representatives of these PFAS groups but were always below detection limits.

Since PFASs have only been analysed for two years in the environmental monitoring of offshore sediments, time trends cannot be determined. Higher values in 2020/2021 compared to 2014 at the four northernmost stations are however worrying considering that levels in other sea areas are relatively stable. Environmental monitoring of marine biota shows that concentrations of PFASs are generally decreasing, or not showing a trend over time (2010–2019; Soerensen & Faxneld 2020). At some stations in the Bothnian Sea and Baltic Proper however, concentrations of PFOA and PFNA are increasing in herring.

In sediment, there is a negative linear relationship between ∑PFAS and TOC ($r^2 = 0.15$, $p = 0.032$, $n = 32$). The degree of correlation is however very low, and when the stations in the Gulf of Bothnia are removed the correlation instead becomes positive with a higher degree of correlation ($r^2 = 0.51$, $p = 0.0002$, $n = 22$). There is also a positive linear relationship between ∑PFAS and TOC when looking only at the Gulf of Bothnia stations ($r^2 = 0.45$, $p = 0.032$, $n = 10$).

Additional PFAS analyses were performed in 2020/2021. The usual analyses only target a limited set of PFASs. PFASs are however a large group of thousands of substances. To get a better picture of the other, usually not measured, PFASs a method called total oxidisable precursor (TOP) assay can be used. In this method, so-called precursors are oxidized to form perfluorocarboxylic acids (PFCAs) before analysis (Houtz & Sedlak 2012). TOP analyses were performed for the five stations in the Gulf of Bothnia and the three stations in the North Sea. Levels of TOP-PFCAs, i.e., the sum of PFCAs after precursors have been converted to PFCAs, are similar in the Gulf of Bothnia and the North Sea samples, 9–18 and 13–17 µg/kg DW, respectively (Fig. 21). PFCA levels before the TOP analysis, on the other hand, are a lot higher in the Gulf of Bothnia, up to 19.6 µg/kg DW, than in the North Sea, up to 0.5 µg/kg DW. The levels of PFCAs in the Gulf of Bothnia sediments are thus slightly lower after the TOP treatment, which might be due to analytical uncertainties, as levels should not be lower after a TOP assay than before. In the North Sea samples, only a small percentage of the TOP-PFCAs can be explained by PFCAs measured in the sample before the TOP assay, on average 2%. Corresponding values for PFOS (levels before compared to levels after) are 96–127% for Gulf of Bothnia and 49–102% for North Sea samples. The distribution pattern of the different PFCAs after the TOP assay also differs between sea areas (Fig. 22). In the Gulf of Bothnia, the pattern is similar to before the TOP, but with an increased proportion of PFHxA and PFBA. In the North Sea samples, PFHxA and PFDA, followed by PFOA, were the dominant PFCAs after TOP assay, while PFUnDA and PFDA were dominant before TOP. The differences between the sea areas should be followed up during future monitoring.

PFOS is a priority hazardous substance in the WFD while $\sum PFAS_{11}$ is an RBSP in Sweden, but there is no EQS for sediment (SwAM 2019). PFOS is one of the substances that most frequently exceeds its EQS and contributes to Swedish water bodies not reaching good environmental status (Vattenmyndigheterna 2018). PFOS is a proposed indicator for HOLAS III, with threshold values for biota and water. It was also an indicator substance in the second holistic assessment of Baltic Sea, where it sometimes exceeded the threshold value for water (HELCOM 2018b). The current draft EQS dossier on PFASs includes 24 compounds and the EQS directive is planned to be revised in 2023.

Although there is no EQS for PFASs valid for Swedish sediments, there is an EQS of 0.23 µg/kg DW for PFOS for coastal sediments in Norway, assuming a TOC content of 1%, and a similar EQS for PFOA of 71 µg/kg DW (Miljødirektoratet 2016). There are also some values suggested in the scientific literature, such as preliminary sediment quality guidelines (SQG) for PFOS in (freshwater) sediment at 22.1 µg/kg DW to protect benthic invertebrates from direct poisoning and 0.19 µg/kg DW to protect wildlife and human health from secondary poisoning (Casado-Martinez *et al.* 2021; both values for a sediment with 5% TOC). A comparison of these threshold values to measured levels of PFOS at the monitoring stations showed that the Norwegian EQS was exceeded at all stations in the Gulf of Bothnia while the suggested SQG for protection from secondary poisoning was exceeded at all stations except for three stations in the Baltic Proper. The suggested SQG for PFOS for benthic invertebrates and the Norwegian EQS for PFOA were not exceeded at any station.

To summarize, levels of PFASs are surprisingly high in sediment in the Gulf of Bothnia. After the 2014 sampling, sediment results were regarded with caution as they could have been caused by contamination or seasonal differences, but as the same results are seen during the 2020/2021 sampling further investigations of the high levels in this part of the Baltic Sea are warranted.

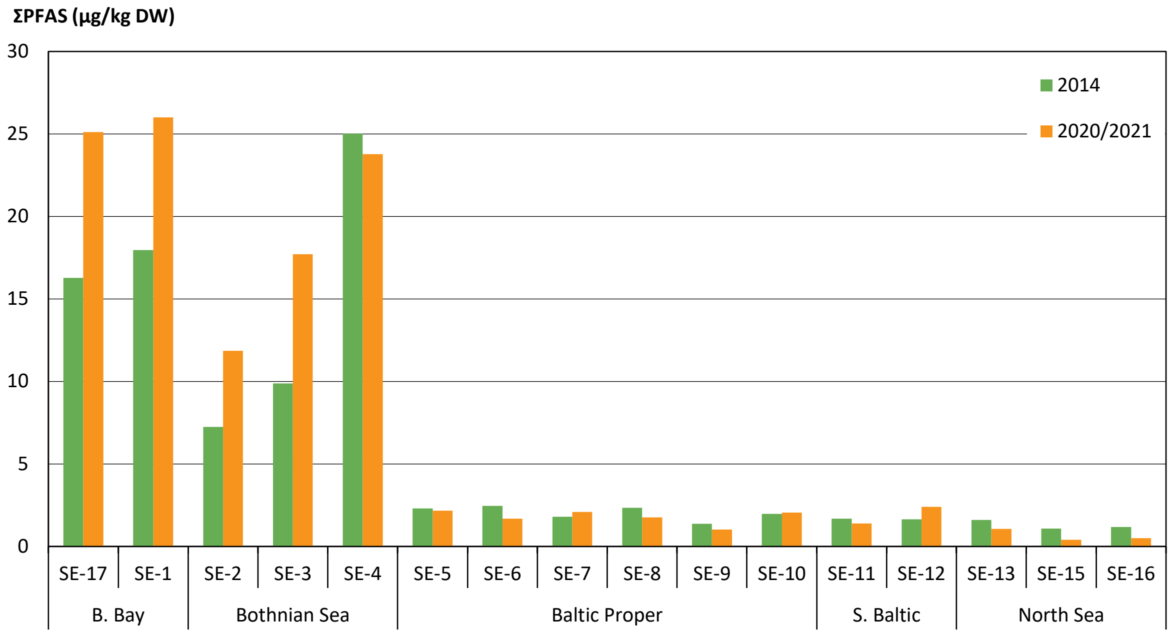

Figure 19. Levels of ∑PFAS at the 16 stations during the two last monitoring years. Note that the value for station SE-5 in 2020/2021 is less certain as the sample had been exposed to temperatures above 0°C for an extended time; however, PFASs are considered as persistent substances. The ∑PFAS in this figure includes PFBS, PFHxS, PFOS, PFDS, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA and PFOSA.

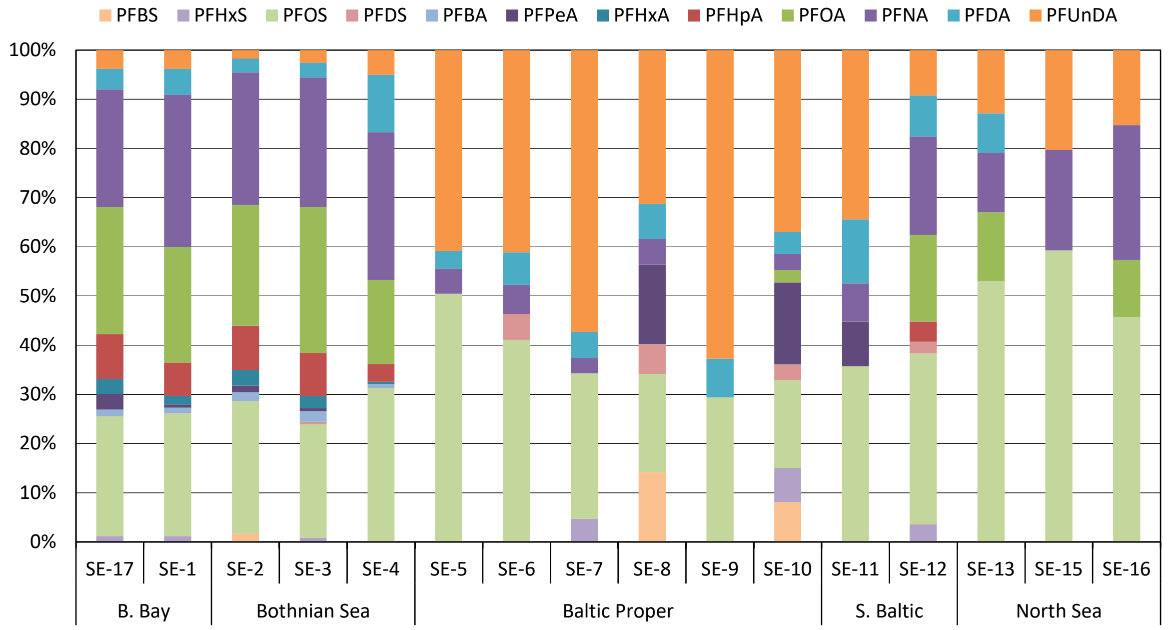

Figure 20. The distribution of the substances constituting PFASs at the 16 monitoring stations in 2020/2021. All substances with at least one detected value have been included, even though some substances (PFBA, PFPeA) are not included in the sum in Figure 19 as they were not analysed in 2014.

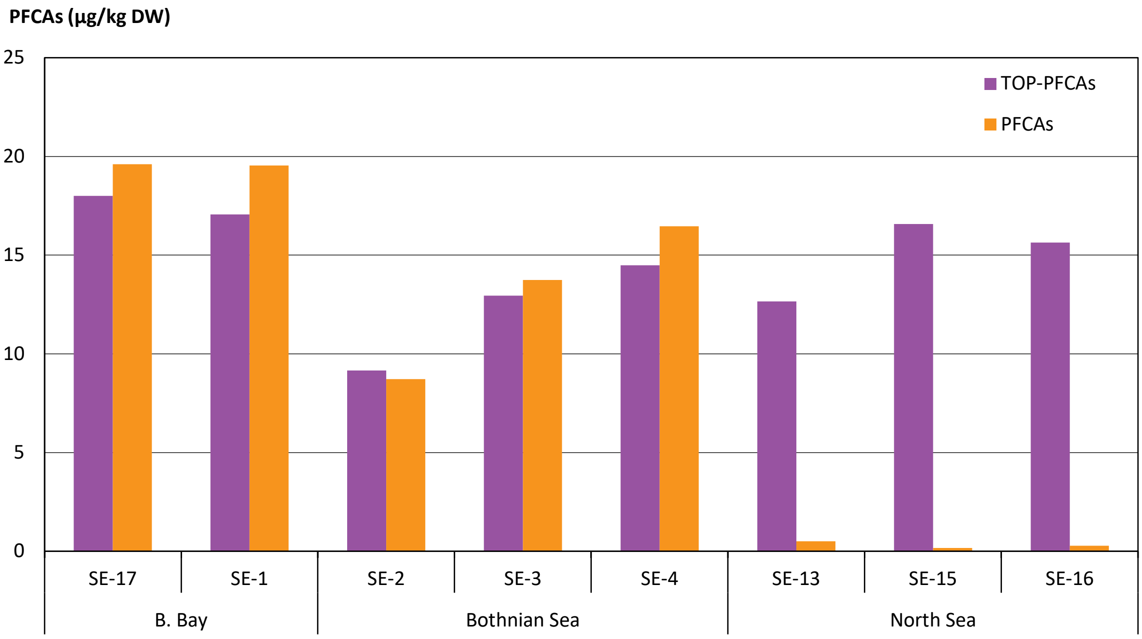

Figure 21. Levels of PFCAs at the 8 stations for which TOP analysis of PFASs was performed in 2020/2021. The figure shows the levels of PFCAs in samples that has not undergone TOP analysis ('PFCAs') and the levels in samples that have undergone TOP analysis ('TOP-PFCAs'). The PFCAs in this figure include PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA and PFUnDA.

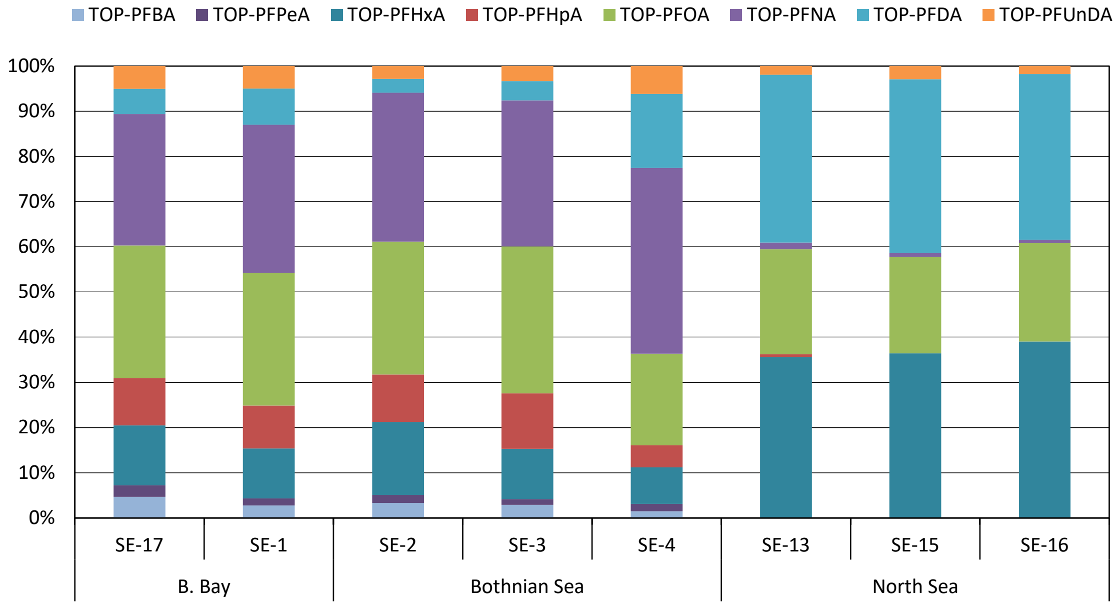

Figure 22. The distribution of the PFCA substances in samples from 8 monitoring stations in 2020/2021 after TOP analysis.

Brominated flame retardants

Polybrominated diphenyl ethers (PBDEs) are a group of 209 substances that have been used as flame retardants in various products. Three different mixtures of PBDEs that differ in degree of bromination were usually produced: pentaBDE, octaBDE and decaBDE. The PBDEs included in penta- and octaBDE are listed as POPs according to the Stockholm Convention since 2009. DecaBDE was included in the Stockholm Convention in 2017 but there are exemptions that allow for the production and use of the substance, for example as an additive in plastic products. Mixtures are dominated by different PBDEs. The dominant congeners in pentaBDE are PBDE 47 and 99, but also PBDE 100, 153 and 154 (La Guardia *et al.* 2006). In octaBDE, PBDE 183, 197, 207 and 209 are the most common, and decaBDE consists almost exclusively of PBDE 209, although there may be large variations in the composition between different products (La Guardia *et al.* 2006).

PBDEs were analysed all four monitoring years, but the number of analysed congeners increased from 2003, when only PBDE 47, 99 and 100 were analysed, to 2014 and 2020/2021 when PBDE 28, 47, 66, 85, 99, 100, 153, 154, 183 and 209 were analysed. Of these, PBDE 183 has occurred at levels above the reporting limit only at one station in 2020/2021 (0.11 µg/kg DW, station SE-16). To be able to compare between areas and years, a sum parameter, ∑PBDE, has been calculated as a sum of PBDE 47, 85, 99, 100, 153, and 154. These six substances have been analysed from 2008 onwards. These are not the same six PBDEs that are included as a priority hazardous substance in the WFD – PBDE 28 is included in the EQS instead of PBDE 85 – and to facilitate comparisons another sum has been calculated for the last two monitoring years when PBDE 28 was analysed, $\sum PBDE_6$. PBDE 209 has not been included in ∑PBDE because the levels of this substance have been highly variable. In 2008, the levels were high and varied from below the reporting limit of 0.5 to more than 19 µg/kg DW, while all other PBDEs had maximum levels of 0.30 µg/kg DW this year. In 2014, PBDE 209 was below the reporting limit of 0.3 µg/kg DW at all stations. In 2020/2021, levels of PBDE 209 were above the detection limit at all stations and varied from 0.074 (above LOD but below LOQ) to 1.6 µg/kg DW.

The levels of ∑PBDE (including PBDE 85 instead of PBDE 28) were relatively even at all stations in 2008, apart from a lower level at station SE-3 in the Bothnian Sea and an elevated level at SE-16 in the North Sea (Fig. 23). In 2014, the levels varied more between the stations and the levels were noticeably higher than in 2008 at some stations. During the last monitoring year, in 2020/2021, levels had decreased compared to 2014 at all stations. The time series is too short, however, for any definite conclusions on decreasing levels in sediments. According to the monitoring programme for contaminants in marine biota, concentrations of PBDEs are decreasing during the last decade with a few exceptions for specific stations and substances (2010-2019; Soerensen & Faxneld 2020).

Due to large variation in levels within different sea areas and over the years, no sea area shows consistently higher levels than other sea areas. Looking at the $\sum PBDE_6$ that includes PBDE 28 instead of 85, there are also no areas with clearly higher or lower levels (Fig. 24). $\sum PBDE_6$ is proposed to be included as an indicator for sediment for HOLAS III, with a threshold value of 310 µg/kg DW for sediment with 5% TOC, aimed to protect the benthic community, as opposed to the corresponding threshold for biota that is aimed to protect human health. Measured levels are far below this threshold value for sediment at all 16 monitoring stations during all years. During the last holistic assessment, PBDE greatly exceeded the indicator threshold value for biota in all monitored sea areas (HELCOM 2018b). Together with mercury, PBDE is the substance that most frequently exceeds the threshold values in biota and causes poor chemical status in surface water in Sweden. Even though the trends in biota for PBDE are declining, levels in relation to threshold values are so high that good environmental status will not be achieved in the foreseeable future (SwAM 2018a).

There is no significant correlation between the levels of ∑PBDE or $\sum PBDE_6$ and the levels of TOC in the sediment. Atmospheric deposition is considered to be the most important route for PBDE to reach the Baltic Sea, and a modelling study points to relatively equal levels of PBDE in coastal and offshore areas unlike, for example, nonylphenol that would have higher levels in coastal environments (Andersson *et al.* 2012). This is due to a combination of lower degradation rate and more supply via the atmosphere to the Baltic Sea for PBDE, meaning that the substance can also be transported to offshore areas (Andersson *et al.* 2012).

Of the seven substances that are included in either ∑PBDE or $\sum PBDE_6$, PBDE 85, 47 and 99 dominate in 2020/2021 (Fig. 25). These were also the dominant congeners in 2014 (data not shown); however, PBDE 28 displays a larger fraction in 2020/2021 at some stations. There seem to be a decreasing fraction of PBDE 85 and an increasing fraction of PDBE 47 at the stations when moving from the Bothnian Bay and outwards to the North Sea (Fig. 25), but this pattern could not be seen in 2014 and should be interpreted with caution.

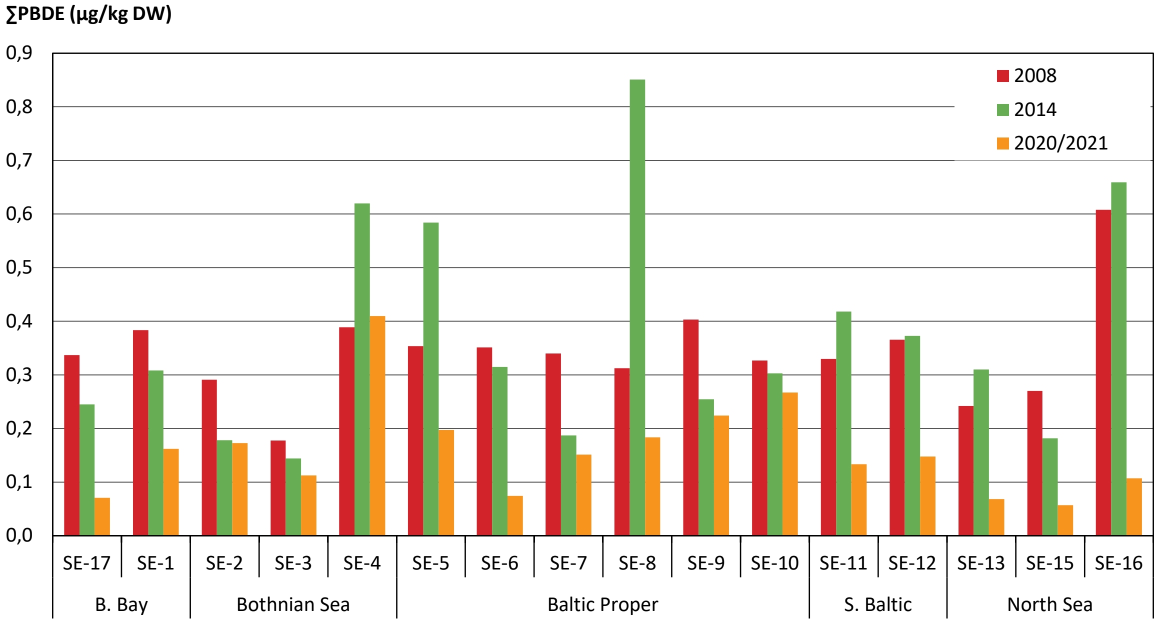

Figure 23. Levels of ∑PBDE at the 16 stations during the three last monitoring years. Note that the value for station SE-5 in 2020/2021 is less certain as the sample had been exposed to temperatures above 0°C for an extended time; however, PBDEs are considered as persistent substances. The ∑PBDE in this figure includes PBDE 47, 85, 99, 100, 153 and 154.

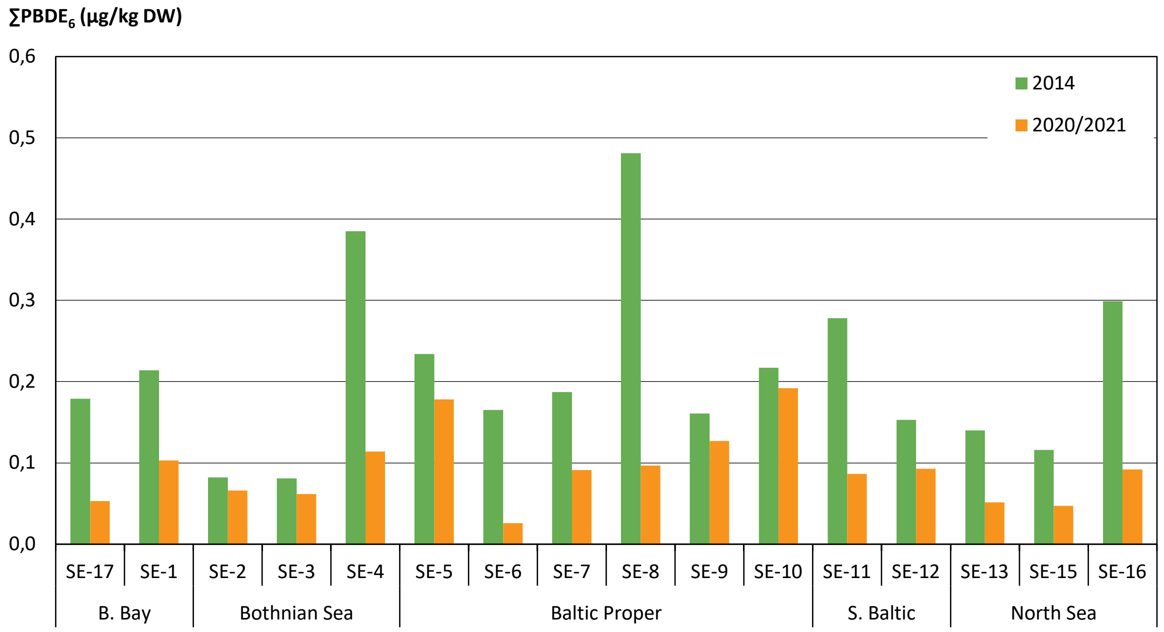

Figure 24. Levels of $\sum PBDE_6$ at the 16 stations during the two last monitoring years. Note that the value for station SE-5 in 2020/2021 is more uncertain as the sample had been exposed to temperatures above 0°C for an extended time; however, PBDEs are considered as persistent substances. The $\sum PBDE_6$ in this figure includes PBDE 28 instead of PBDE 85, so the sum includes PBDE 28, 47, 99, 100, 153 and 154.

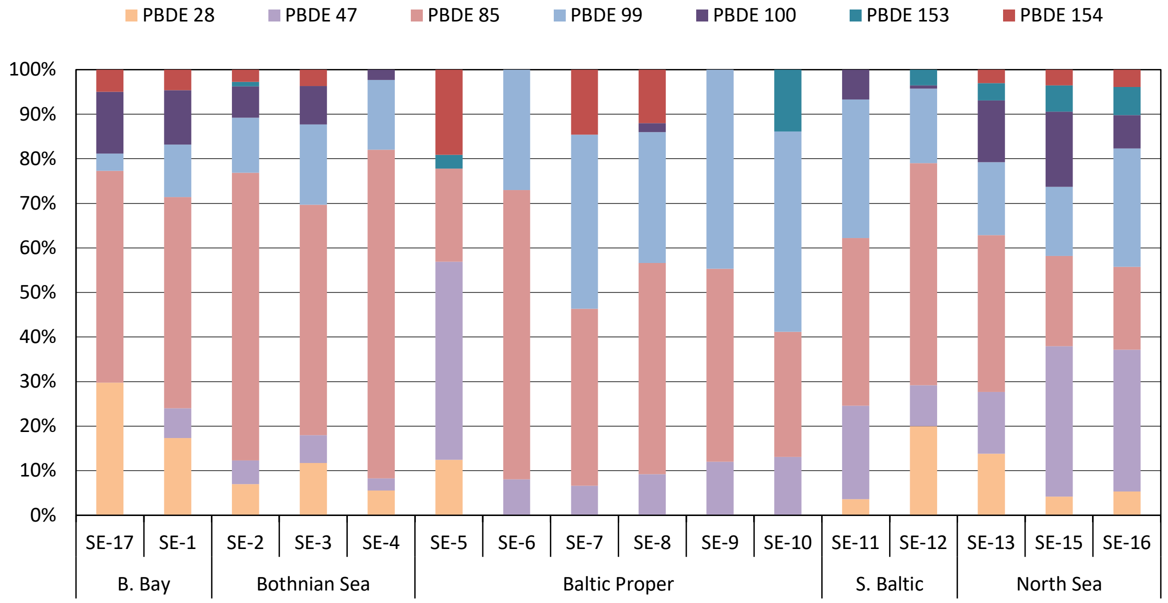

Figure 25. The distribution of PBDE substances at the 16 monitoring stations in 2020/2021. PBDE 209 dominated the pattern in 2020/2021 but was below the detection limit in 2014 and has not been included in the figure. PBDE 183, that could only be measured at levels above reporting limits at SE-16, and PBDE 66, that was always below reporting limits, have also not been included in the figure.

Hexabromocyclododecane (HBCDD) is a flame retardant whose use should be limited according to the Stockholm Convention. HBCDD was analysed in sediments in 2008 and 2014 but levels never exceed the reporting limit (1–5 µg/kg DW). In 2020/2021, the analytical method was changed, and three isomers were analysed: α-, β- and γ-HBCDD. Levels of at least one of the isomers could then be reported from 14 of the 16 monitoring stations, with α-HBCDD being most common and occurring at the highest levels (Fig. 26). Total levels of HBCDD were highest in the Baltic Proper, up to 0.14 µg/kg DW although with large variations between stations. There was no significant correlation between levels of HBCDD and TOC in the sediment.

HBCDD is a priority hazardous substance according to the WFD but has no EQS for sediment. There is however an indicative value from SwAM (2018b) of 170 µg/kg TS for sediment with 5% TOC, which is also the proposed indicator threshold for HBCDD in sediment for HOLAS III. Levels of HBCDD never exceeded this value, even when using the reporting limit as a 'worst-case scenario'.

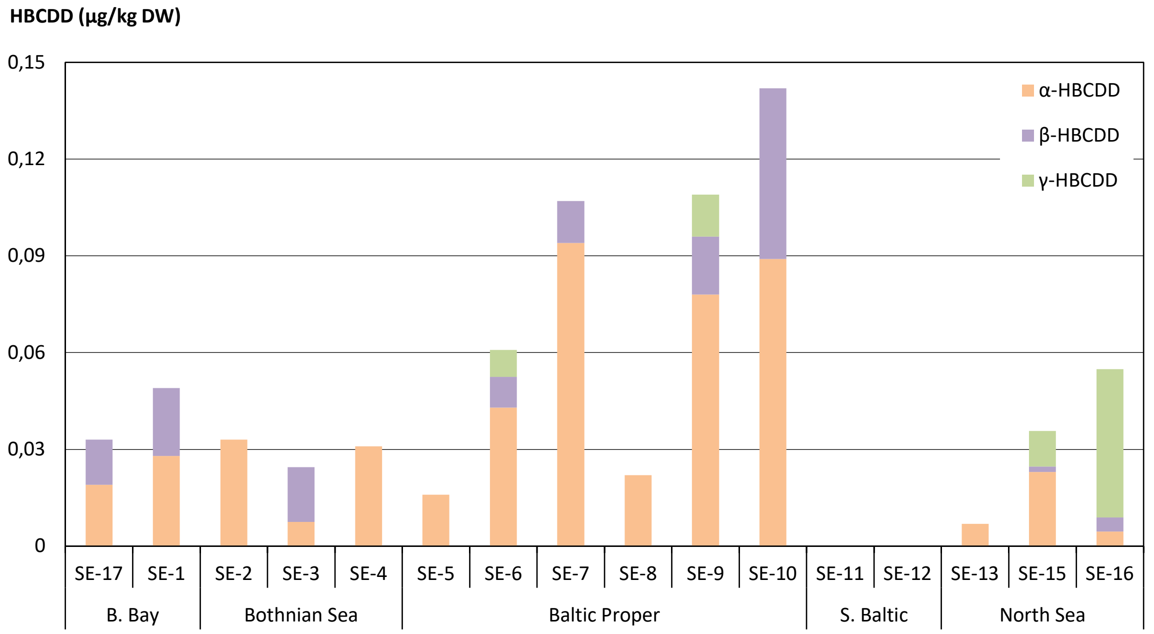

Figure 26. Levels and distribution patterns of the three measured isomers of HBCDD at the 16 stations in 2020/2021. Note that the value for station SE-5 is less certain as the sample had been exposed to temperatures above 0°C for an extended time.

Organophosphorus compounds

For the 2020/2021 monitoring, organophosphorus compounds (OPs) were analysed. These are substances that are used extensively as flame retardants (mainly halogenated OPs) and plasticizers (mainly non-halogenated OPs; Wei *et al.* 2015). They are also used in hydraulic fluids, floor polish etc. Twelve compounds were included in the analysis: tris(ethyl) phosphate (TEP), tris(isobutyl) phosphate (TIBP), tri-*n*-butyl phosphate (TNBP), tris(chloroethyl) phosphate (TCEP), tris(2-chloroisopropyl) phosphate (TCIPP), tris(1,3-dichlorisopropyl) phosphate (TDCIPP), tris(2-butoxyethyl) phosphate (TBOEP), tris(phenyl) phosphate (TPHP), 2-ethylhexyl diphenyl phosphate (EHDPP), tris(2-ethylhexyl) phosphate (TEHP), tri-o-cresyl phosphate (TOCP) and tri-cresyl phosphate (TCP; as a mixture of congeners including TOCP). TBOEP and TOCP were always at levels below their reporting limits of 9.6 and 0.48 μg/kg DW, respectively.

The levels of $\sum$OPs were between 4.6 and 42 μg/kg DW and were highest at stations SE-12 and SE-5 (Fig. 27). The sample from SE-5 had been exposed to temperatures above 0°C for several weeks and it is possible that it had become contaminated with OPs from the sample jar, although it was glass with Teflon-lined lid. The results from this station should therefore be interpreted with caution. Overall, levels were highest in the Southern Baltic, followed by the Baltic Proper. Levels in the Bothnian Sea and Bay were lower and clearly decrease to the north. The lowest levels were found on the two northernmost stations in the North Sea. This may indicate that atmospheric deposition is an important route as the deposition of organic contaminants is generally higher in the south than the north. The compound pattern however differs between the Baltic Sea and the North Sea (Fig. 27). In the Baltic Sea, the dominant compounds, that is, the compounds occurring at the highest levels, are tri-cresyl phosphate (TCP) and 2-ethylhexyl diphenyl phosphate (EHDPP). At the North Sea stations, on the other hand, EHDPP never occurs above the reporting limit while TCP only occur at the southernmost station, SE-13. TCIPP is also one of the most common compounds, occurring above reporting limits at all stations except in the Bothnian Bay.

In a screening study of OPs in Swedish watercourses, TDCIPP/TEHP (analysed as a sum) was most frequently detected in the water, followed by TCEP and EHDPP (Gustavsson *et al.* 2016). TCEP and EHDPP are frequently detected also in the environmental monitoring of offshore sediments while TDCIPP and TEHP are less frequently found. One possible explanation for the differences is that Gustavsson *et al.* analysed water and had lower reporting limits for TDCIPP and TEHP, and higher for e.g. TCIPP, than in the present study. Another explanation may be that the organophosphate esters vary greatly in hydrophobicity, expressed as log K_{OW}, which will affect their partitioning between water and sediment. TEHP however has a high log K_{OW} value of 9.49 and should therefore bind to sediments and other particles, and even though TDCIPP has a lower log K_{OW} of 3.8, it should still bind to particles (Wei *et al.* 2015).

OPs have not been measured previously in the Swedish environmental monitoring of offshore sediments, and levels have therefore been compared to studies of mainly marine sediments found in literature. Levels were similar to levels found in the NW Mediterranean Sea (13–49 μg/kg DW, sum of 9 OPs, Schmidt *et al.* 2021), at coastal aquaculture facilities in seven European countries (1.99–58.2 μg/kg DW, sum of 6 OPs, Aznar-Alemany *et al.* 2018), in the Bohai Bay, China (1.42–52.9 μg/kg DW, sum of 14 OPs, Qi *et al.* 2021 and 1.76–49.9 μg/kg DW, sum of 11 OPs, Liao *et al.* 2020), at Hainan Island, China (0.74–60.0 μg/kg DW, sum of 10 OPs, Mo *et al.* 2019) and in an enclosed bay in the Gulf of San Francisco (9.5–33 μg/kg DW, sum of 13 OPs, Sutton *et al.* 2019). In sediment in the Canadian Arctic, levels were generally lower (Sühring *et al.* 2021). The median of the sum of 10 OPs was 8.3 μg/kg DW, although the range was 0.12–57 μg/kg DW with the highest levels found in sediments affected by the large Mackenzie River (Sühring *et al.* 2021). Even lower levels, 0.159–4.7 μg/kg DW, were measured in a transect from the North Pacific to the Arctic Ocean (sum of 7 OPs, Ma *et al.* 2017).

In a study in the NW Mediterranean Sea, the sum of 8 OPs were 4.5–230 µg/kg DW (Alkan *et al.* 2021). The highest levels occurred near large cities, while lower levels occurred in protected areas and further out from the coast. In even more affected environments, levels can be much higher, e.g. 470–2 800 µg/kg DW in a channel discharged from the Chicago Sewage Treatment Plant (sum of 8 OPs, Peverly *et al.* 2015), up to 10 900 µg/kg DW at a sea-based waste facility in Japan or up to 33 800 µg/kg DW close to a car scrap in Norway (see references in Wei *et al.* 2015). These levels far exceed the levels found in Swedish offshore sediments.

There is a significant and positive linear correlation between the levels of ∑OPs and the levels of TOC in the sediment ($r^2 = 0.36, p = 0.069, n = 16$). OPs were only analysed during one monitoring year, and it would be interesting to repeat the analysis during the next monitoring to see if the geographical patterns are consistent and to see how the environmental levels of this compound group is changing over time.

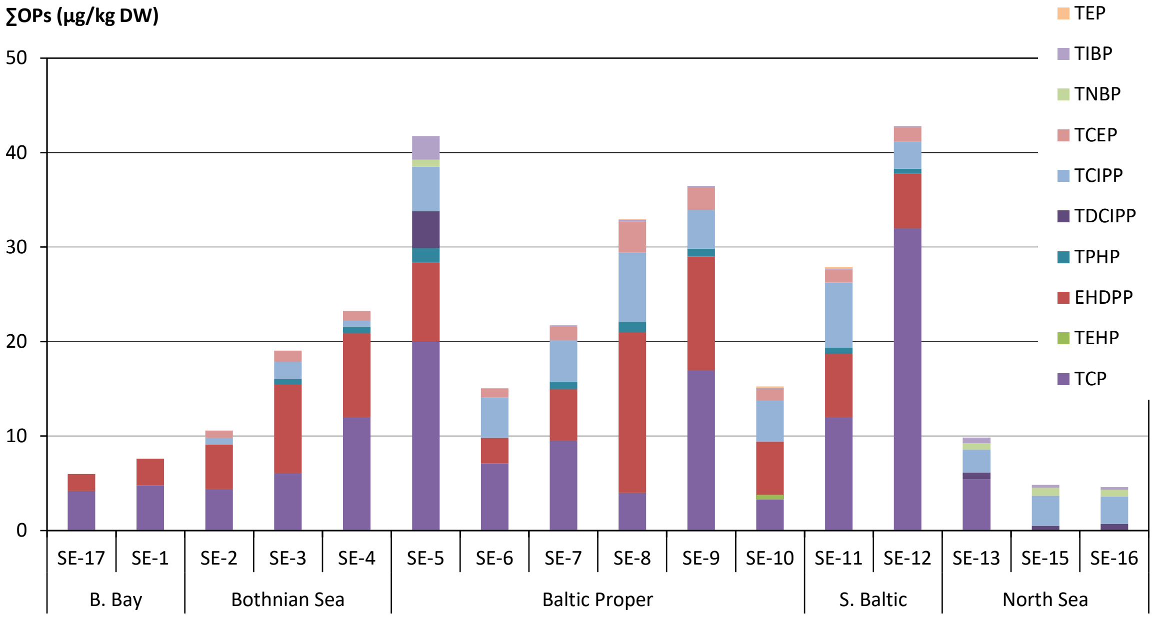

Figure 27. Levels and distribution of the different organophosphorus compounds measured at the 16 stations in 2020/2021. Note that the values for station SE-5 are less certain as the sample had been exposed to temperatures above 0°C for an extended time.

PCBs

Polychlorinated biphenyls (PCBs) are a group of 209 substances (congeners) that have been used, among other things, as plasticizers in sealants and in electrical equipment. PCBs were gradually banned in Sweden, starting in the 1970s, and a total ban on use was introduced in 1995.

PCBs have been analysed during all monitoring years with the seven common PCB congeners 28, 52, 101, 118, 138, 153 and 180 being analysed. These are included in the sum parameter $\sum PCB_7$. In 2014 and 2020/2021, more PCBs were analysed, but these so-called planar PCBs are grouped with PCDD/Fs and reported with this group.

Levels of $\sum PCB_7$ tend to be higher in the Southern Baltic and the Baltic Proper than in the other sea areas, although there are large variations (Fig. 28). The station with the highest levels is SE-12 in the Southern Baltic, except for in 2020/2021 when a higher level was reported for SE-9. The levels of $\sum PCB_7$ display a positive and significant linear correlation with TOC levels in the sediment, but the degree of correlation is relatively low ($r^2 = 0.12, p = 0.0061, n = 64$). All seven congeners, individually, have positive and significant correlations with TOC (when excluding a very high value for PCB 118 in 2003). No sea area consistently deviates from the correlation, but station SE-12 tends to have elevated levels compared with what the correlation with TOC predicts.

According to a model for the Baltic Proper and the Bothnian Sea, atmospheric deposition is believed to be the dominant input route of PCBs to the Baltic Sea (SwEPA 2009). Atmospheric deposition of PCBs has a north – south gradient with the highest deposition in the southern parts of the Baltic Sea region (HELCOM 2018a). The levels of PCBs (118 and 153) in herring sampled within the environmental monitoring of marine biota during the last decade are highest in the Baltic Proper, primarily the Southern Baltic, while levels are lower in the Bothnian Bay and the North Sea (2010–2019; Soerensen & Faxneld 2020), which agrees relatively well with the geographical pattern of PCBs in sediments.

The congener pattern in the sediment, i.e., how much the individual PCBs contribute to $\sum PCB_7$, is similar for all stations, although there are some differences between sea areas (Fig. 29). There seems to be a larger proportion of more highly chlorinated PCBs in the Bothnian Bay. PCBs 153, 138 and 180 make up around 70% of $\sum PCB_7$ at the stations here during all monitoring years (Fig. 29 for 2020/2021), which is more than for other sea areas.

At several of the stations, $\sum PCB_7$ levels in the sediment increase between 2003 and 2008 (Fig. 28), although the difference is not statistically significant. Since a change of laboratory and method took place between these two years, it is likely that the increase is due to this change rather than to an actual increase in levels in the environment. Atmospheric deposition of PCBs has decreased since the early 1990s (HELCOM 2018a; for PCB 153). Levels in sediments in the Baltic Sea also appear to decrease if levels in older (around 1990) and newer (around 2005) samples are compared (SwEPA 2009). In the national environmental monitoring of marine biota, PCB concentrations generally decrease during the last decade, although there are a few stations that deviate possibly due to changes over time in the age and other biological parameters of the sampled herring (2010–2019; Soerensen & Faxneld 2020).

PCBs are one of the original 12 POPs that have been listed in the Stockholm Convention since it entered into force in 2004. According to the convention, the production and use of PCBs must be eliminated and accidental formation minimized. PCBs are also listed as an RBSP in the WFD in Sweden but there is no EQS for sediments. PCBs are proposed as an indicator for HOLAS III, with thresholds for biota, and during the last holistic assessment PCB levels tended to be below the threshold values (HELCOM 2018b).

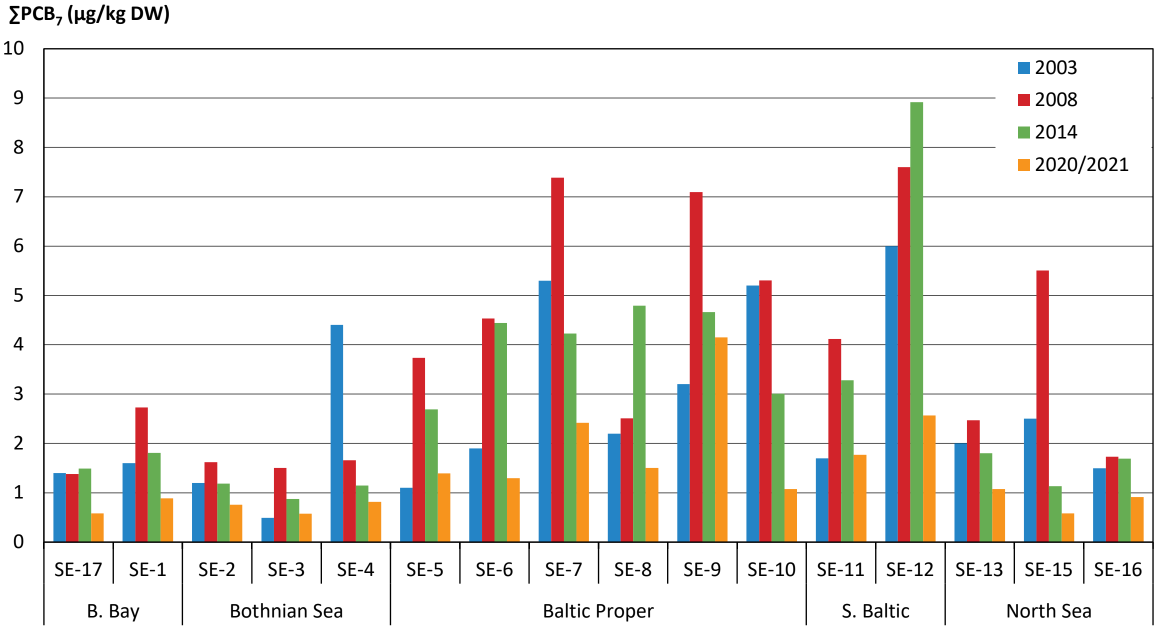

Figure 28. Levels of ∑PCB₇ at the 16 stations during the four monitoring years. Note that the value for station SE-5 in 2020/2021 is less certain as the sample had been exposed to temperatures above 0°C for an extended time; however, PCBs are relatively persistent compounds. The level at station SE-4 in 2003 is uncertain because the congener pattern deviates from other samples due to a very high level of PCB 118 (3.8 µg/kg DW compared to a max of 1.2 for all other stations and years). Such a deviating pattern is abnormal.

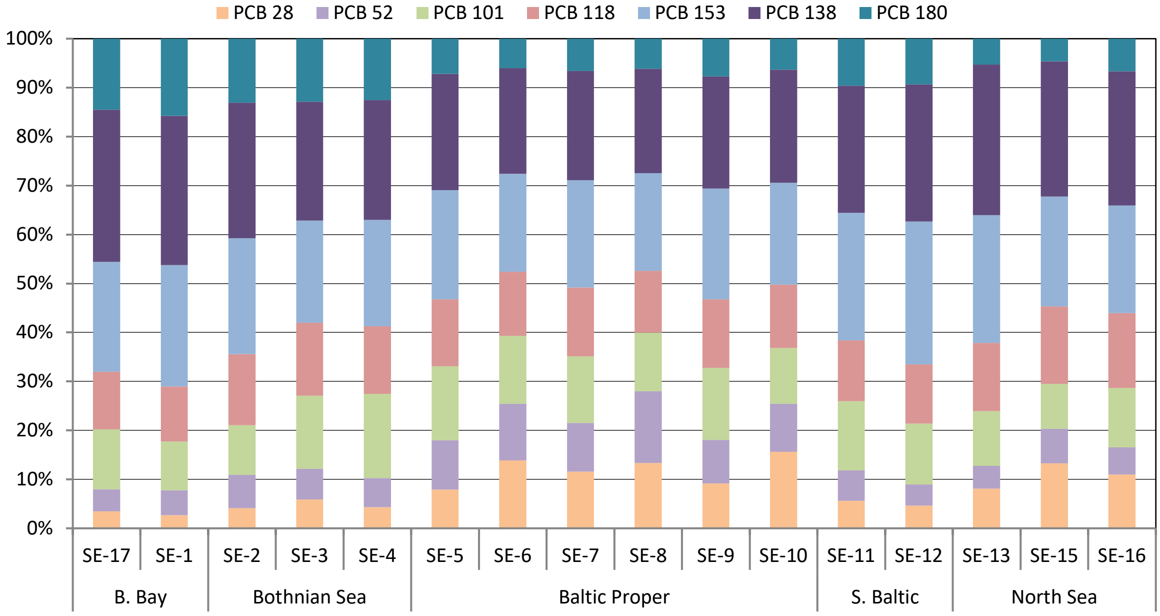

Figure 29. The congener pattern of the seven PCBs constituting ∑PCB₇ at the 16 monitoring stations in 2020/2021. Note that the value for station SE-5 in 2020/2021 is less certain as the sample had been exposed to temperatures above 0°C for an extended time; however, PCBs are relatively persistent compounds.

PCDD/Fs and DL-PCBs

Polychlorinated dibenzo-*p*-dioxins and dibenzofurans, commonly called dioxins and abbreviated as PCDD/Fs, were analysed in 2014 and 2020/2021. This is a group of 210 substances that are inadvertently formed in various processes, including combustion and chlorination processes. The analyses included the seventeen toxic 2,3,7,8-substituted PCDD/Fs, as well as twelve PCBs which like the PCDD/Fs may have a planar structure and therefore have a similar toxic effect.

The levels of PCDD/Fs vary within each sea area and it is not possible to point out a sea area with consistently higher levels (Fig. 30). The highest levels however occur at SE-12 in the Southern Baltic in both 2014 and 2020/2021, and SE-11 had the second highest levels in 2020/2021. In the Baltic Proper, SE-7 and SE-9 had the highest PCDD/F levels in both 2014 and 2020/2021. These two stations have very low sediment accumulation rates, thus having an on average older surface sediment (see Table 2). The third station with a low accumulation rate in the Baltic Proper, SE-10, does not however have elevated levels of PCDD/Fs.

Levels of PCDD/Fs can also be expressed as toxic equivalents (TEQ) where PCDD/Fs that are more toxic, generally those with fewer chlorine atoms, have an increased weight in the calculation of the sum (Fig. 31). It is apparent that SE-12 is less dominant than previously, while stations in the Baltic Proper and Gulf of Bothnia have more comparable levels. The reason is that SE-12, and other stations in the Southern Baltic and North Sea, have a higher fraction of OCDD (Fig. 32). This is a PCDD/F with eight chlorine atoms which is relatively nontoxic in comparison to PCDD/Fs with fewer chlorine atoms. The fraction of OCDD is low (20–30% of $\sum$PCDD/F_{17}) in Gulf of Bothnia sediments but increase gradually throughout the Baltic Sea towards the North Sea, where the fraction of OCDD is 50–60%. In the Gulf of Bothnia, there is instead a higher fraction of PCDFs with many chlorines, i.e., OCDF and one of the HpCDFs, a pattern that was also present in 2014 (data not shown). In Gulf of Bothnia sediments, the PCDFs constituted >50% of the total PCDD/Fs, while PCDDs dominated in remaining sea areas (Fig. 32).

Although dioxins are substances that bind to carbon, there is no significant relationship between the levels of PCDD/Fs and the TOC levels in the sediment, even on a TEQ basis. Atmospheric deposition of dioxins to the Baltic Sea, which is believed to be the dominant input route, has decreased since the 1990s (by 67%, 1990–2014), and is higher in the southern than in the northern parts (SwEPA 2009, HELCOM 2018a). The highest concentrations of PCDD/Fs in herring sampled in the environmental monitoring of marine biota occur in the Bothnian Sea while the concentrations are lowest in the North Sea (2017–2019; Soerensen & Faxneld 2020). This does not match the pattern seen in offshore sediments.

Emissions to air from combustion processes are believed to be the main source of PCDD/Fs to the Baltic Sea, but it is unclear how much fires and emissions from polluted areas account for (Andersson *et al.* 2012). PCDD/Fs can also originate from contaminated areas, for example, the river Kymijoki in Finland, which is polluted from previous chlorophenol production, is a major source of PCDD/Fs to the Baltic Sea. The annual input of PCDD/Fs from Kymijoki has been calculated to correspond to the atmospheric deposition (Andersson *et al.* 2012). Other potential sources are sediments contaminated from chloralkali or bleached paper production, and there are many areas with high levels of PCDD/F along the northern Swedish coast (Norrlin & Josefsson 2017). A modelling study of PCDD/Fs in fish in the Baltic Sea indicates that the largest amount of PCDD/Fs in herring in the Baltic Proper, the Bothnian Sea and Bothnian Bay comes from atmospheric sources, but as these sources decrease due to measures to improve cleaning of combustion processes, historical chlorophenol pollution become more important for PCDD/F levels in herring (Assefa *et al.* 2019).

The levels of PCDD/Fs in sediments in the Baltic Sea have decreased over time with the highest levels occurring in 1985–2002, and a later peak in offshore sediments compared with coastal sediments (Assefa *et al.* 2014). In the environmental monitoring of marine biota, concentrations of PCDD/Fs generally decrease over the last decade except for a few sites in the North Sea (2010–2019; Soerensen & Faxneld 2020). In the monitoring of offshore sediments, levels are higher in 2020/2021 than in 2014, which is unexpected. It could be due to analytical variations but should be followed up during future monitoring campaigns.

PCDD/Fs are among the 12 original POPs listed in the Stockholm Convention, and their unintentional formation must be minimized. PCDD/Fs are priority hazardous substances in the WFD but there is no EQS for sediment. PCDD/Fs are proposed as an indicator for biota for HOLAS III and were an indicator in the second holistic assessment of the Baltic Sea when they generally occurred at concentration below the threshold value (HELCOM 2018b). In coastal water bodies in Sweden, however, PCDD/Fs are one of the substances that most often occur above their threshold value and contribute to reduced environmental status (Vattenmyndigheterna 2018). The levels of PCDD/Fs in fatty fish from the Baltic Sea are still generally too high for the fish to be sold on the EU common market, and the European Food Safety Authority (EFSA 2018) recently reduced the tolerable weekly intake (TWI) of PCDD/Fs seven-fold compared to the previous TWI. It is possible however that the toxic equivalency factors (TEFs) used to calculate TEQs will be adjusted in a planned re-evaluation by WHO, as there are concerns that the toxicity of some planar PCBs is overestimated. Given that PCDD/Fs exceed threshold values, it is likely that further measures will need to be taken to reduce PCDD/F levels in the Baltic Sea environment.

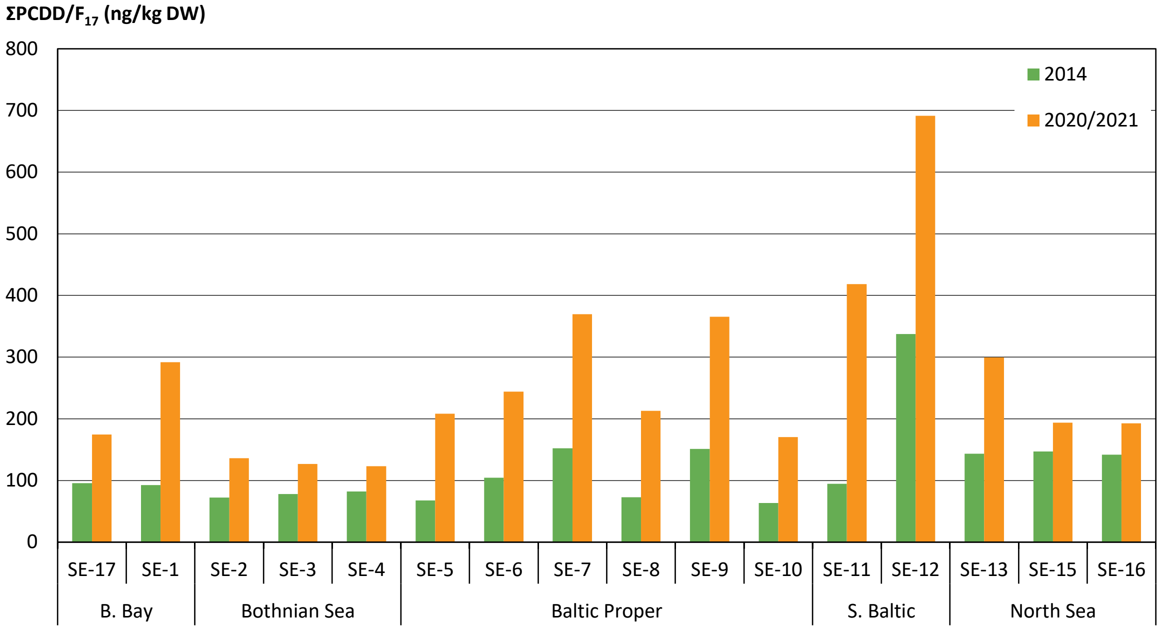

Figure 30. Levels of ∑PCDD/F_{17} at the 16 stations in 2014 and 2020/2021. Note that the value for station SE-5 in 2020/2021 is less certain as the sample had been exposed to temperatures above 0°C for an extended time; however, PCDD/Fs are persistent compounds.

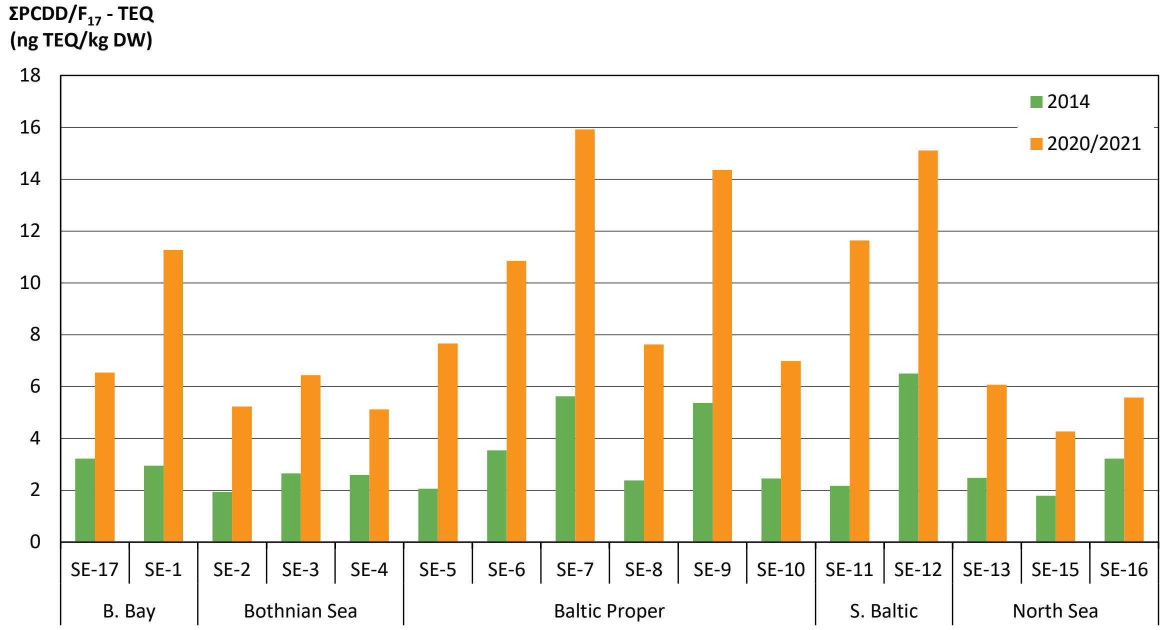

Figure 31. Levels of ∑PCDD/F_{17} at the 16 stations in 2014 and 2020/2021, expressed on a TEQ basis. Note that the value for station SE-5 in 2020/2021 is less certain as the sample had been exposed to temperatures above 0°C for an extended time; however, PCDD/Fs are persistent compounds.

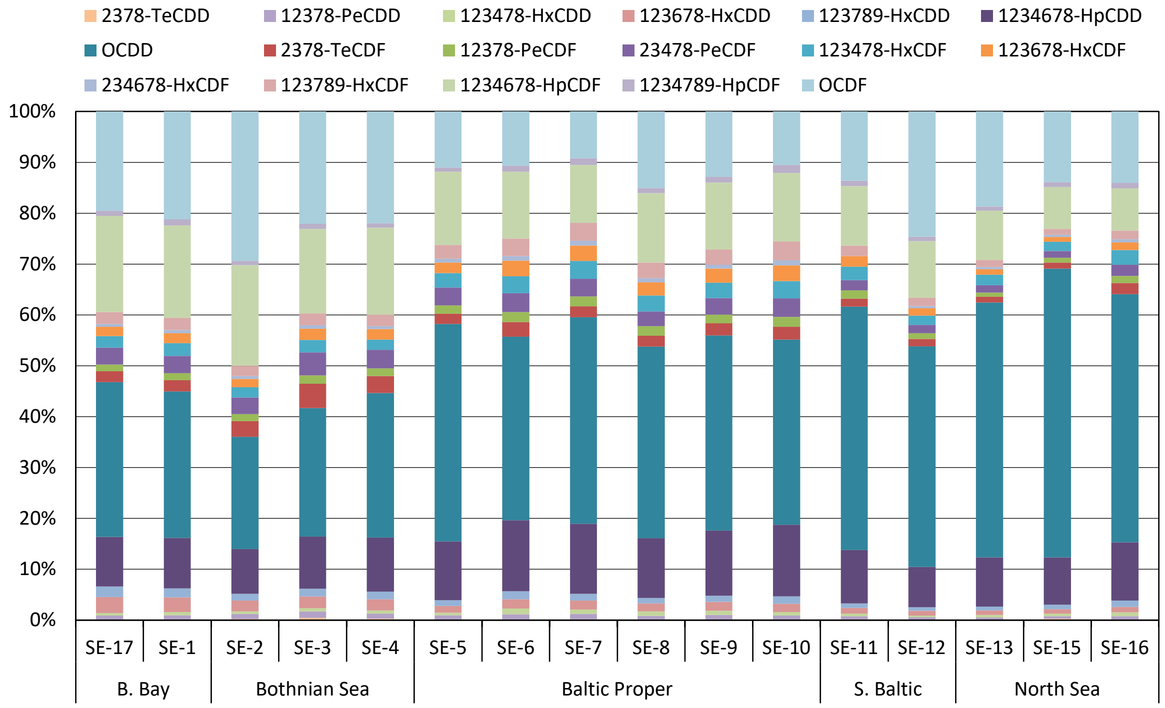

Figure 32. The congener pattern of the seventeen PCDD/Fs constituting ∑PCDD/F_{17} at the 16 monitoring stations in 2020/2021. Note that the value for station SE-5 in 2020/2021 is less certain as the sample had been exposed to temperatures above 0°C for an extended time; however, PCDD/Fs are persistent compounds.

Dioxin-like PCBs, or **DL-PCBs** is a group that includes the congeners PCB 77, 81, 126, 169, 105, 114, 118, 123, 156, 157, 167 and 189. PCB 118 is usually the congener occurring in the highest levels, and it is also included in the sum parameter $\sum PCB_7$. DL-PCBs have a similar geographical pattern to PCDD/Fs, with the highest levels at station SE-12 in the Southern Baltic, and elevated levels at some stations in the Baltic Proper (Fig. 33). In 2020/2021, levels were particularly high at SE-9, which does not match $\sum PCDD/F_{17}$ levels (see Fig. 30). Like PCDD/Fs, the DL-PCBs can be expressed as toxic equivalents, TEQ, and the most toxic congeners are PCBs 126 and 169. SE-9 had high levels of PCB 118 and other congeners that are less toxic, and thus this station does not have the highest levels on a TEQ-basis (Fig. 34). There is no correlation between the levels of ∑DL-PCBs and the levels of TOC in sediments, even when PCB levels are expressed as TEQ, corresponding to what was seen for PCDD/Fs.

The fractions that different PCBs contribute to the ∑DL-PCBs, i.e., the congener pattern, are relatively similar in the different sea areas (Fig. 35). Stations in the Bothnian Bay tend to have a larger fraction of some of the more chlorinated DL-PCBs (PCBs 156, 157, 167 and 189), which was also the case in 2014 (data not shown). Stations SE-6 and SE-7 deviate in their congener pattern as they have very low levels of some of the more chlorinated DL-PCBs. Such a deviating pattern is thought to be related to disturbances in samples during analysis because such a pattern in the distribution does not usually occur in the environment.

DL-PCBs are included in the EQS for dioxins and dioxin-like substances in the WFD, but as mentioned, there is no EQS for sediment.

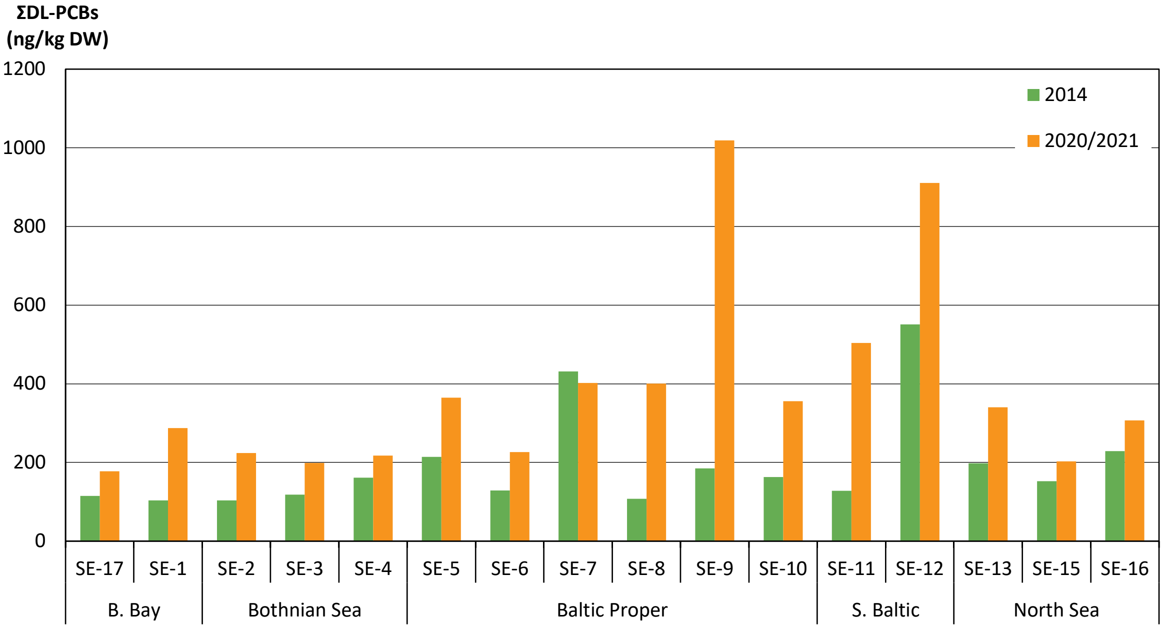

Figure 33. Levels of dioxin-like PCBs (DL-PCBs) at the 16 stations in 2014 and 2020/2021. Note that the value for station SE-5 in 2020/2021 is less certain as the sample had been exposed to temperatures above 0°C for an extended time; however, PCBs are relatively persistent compounds.

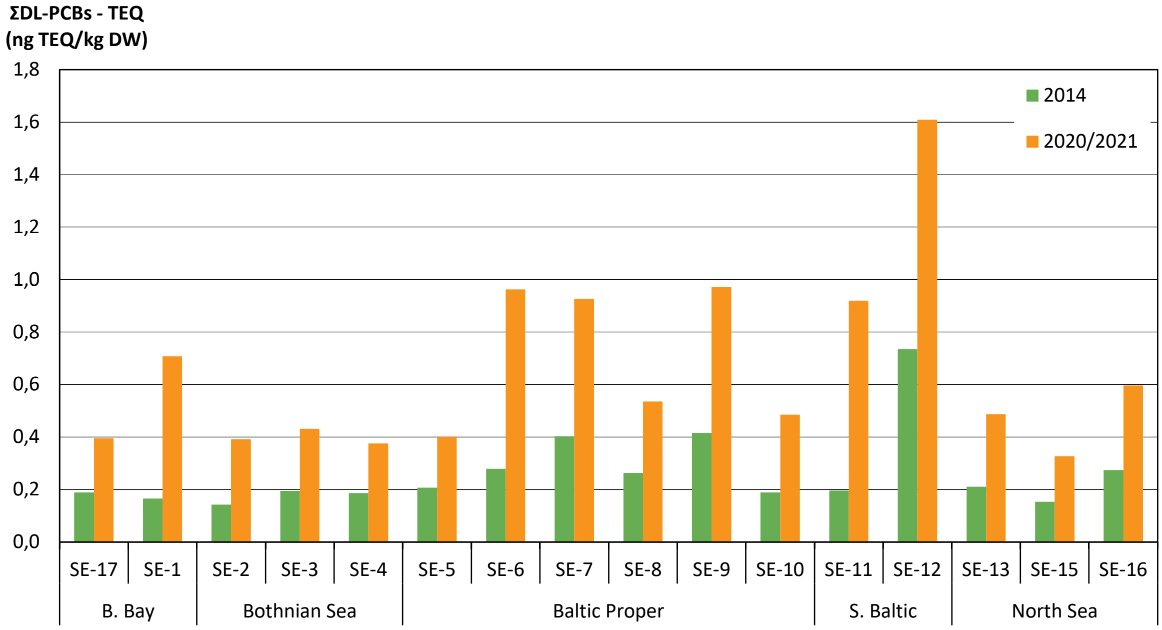

Figure 34. Levels of dioxin-like PCBs (DL-PCBs) at the 16 stations in 2014 and 2020/2021, expressed on a TEQ basis. Note that the value for station SE-5 in 2020/2021 is less certain as the sample had been exposed to temperatures above 0°C for an extended time; however, PCBs are relatively persistent compounds.

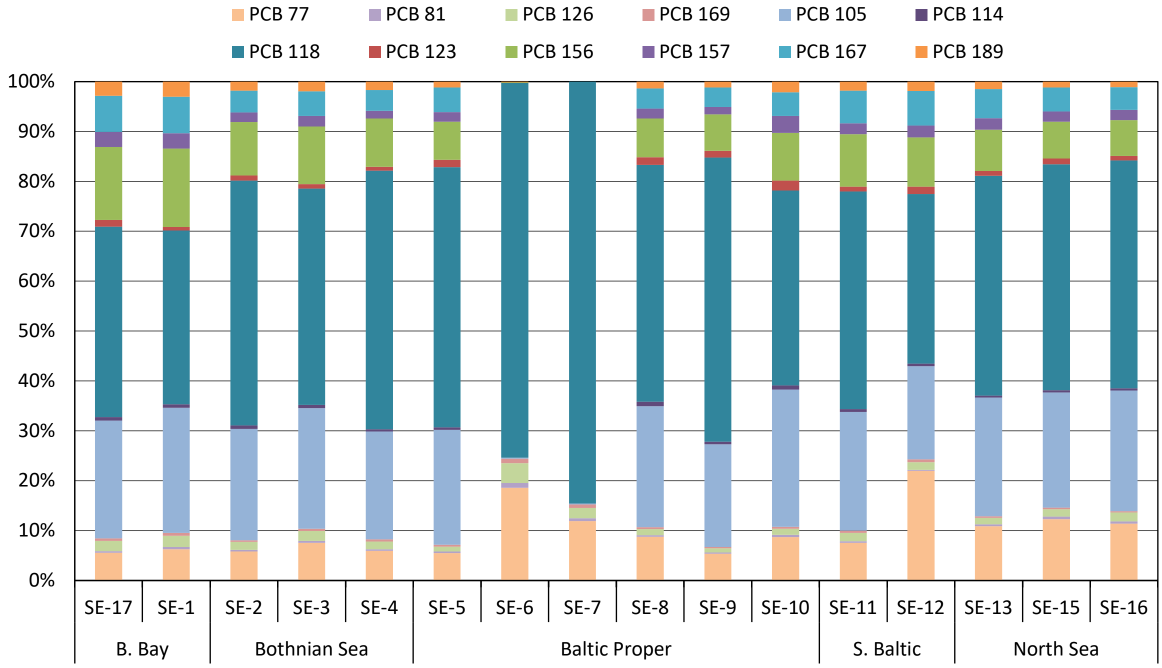

Figure 35. The congener pattern of the ten DL-PCBs at the 16 monitoring stations in 2020/2021. Note that the value for station SE-5 in 2020/2021 is less certain as the sample had been exposed to temperatures above 0°C for an extended time; however, PCBs are relatively persistent compounds.

Chlorinated paraffins

Chlorinated paraffins are used in large quantities as plasticizers and flame retardants in plastics and rubber as well as lubricants in the metal industry. They are divided into short-chain (SCCPs, with a carbon chain of 10–13 carbon atoms), medium-chain (MCCPs, 14–17 carbon atoms in the chain) and long-chain (LCCPs, more than 17 carbons in the chain). There is also a group with 6–9 carbon atoms called very short-chain chlorinated paraffins (vSCCPs). MCCPs were analysed in 2014, and levels were then below the reporting limit of 0.1–0.2 mg/kg DW at all stations. LCCPs and vSCCPs have only been analysed in a separate screening project (see below), not within the ordinary environmental monitoring.

SCCPs were analysed in sediment samples in 2008 and 2014. Levels were below the reporting limit of 0.1–0.2 mg/kg DW at all stations in 2014, but in 2008, levels of 0.03–0.7 mg/kg DW were measured (Fig. 36). Levels were higher in the Baltic Proper than in the Bothnian Bay and the North Sea, while in the Bothnian Sea and the Southern Baltic, intermediate levels were observed. The highest levels of SCCPs were measured at four stations around Gotland, i.e., in the central part of the Baltic Proper. SCCP levels display a positive and significant linear correlation with TOC levels in the sediment ($r^2 = 0.65$, $p = 0.0001$, $n = 16$), which is reasonable because SCCPs are substances with an affinity for organic carbon. Both stations in the Bothnian Bay have SCCP levels that are lower than predicted by the correlation with TOC; otherwise, all sea areas group around the regression line.

SCCPs are a priority hazardous substance and MCCPs an RBSP in Sweden in the WFD (SwAM 2019). There is no EQS for sediment but there is an indicative value for SCCPs of 0.998 mg/kg DW for sediments with 10% TOC, although the value is uncertain and biota levels are more relevant for status assessments (SwAM 2018b). The indicative value is not exceeded at any of the stations, but stations with levels closest to the value are SE-3 (0.76 mg/kg DW after conversion to 10% TOC) and SE-7 (0.59 mg/kg DW after conversion to 10% TOC).

SCCPs have been regulated by the Stockholm Convention since 2017 and are prohibited from being manufactured or placed on the market. They seem to have been replaced by MCCPs and LCCPs, and trends in soil in Switzerland and coastal sediments near presumed sources in Sweden showed declining levels of SCCPs but often increasing levels of MCCPs, resulting in higher levels of MCCPs than SCCPs (Bogdal *et al.* 2017, Yuan *et al.* 2017). Sediment samples from the Stockholm archipelago and Lake Mälaren however indicate that levels of both SCCPs and MCCPs have decreased in recent decades (Länsstyrelsen 2015). This may be due to the use of MCCPs decreasing in Sweden (Andersson *et al.* 2012). Currently, MCCPs are under evaluation for banning production and use in both EU and the Stockholm Convention. Globally, more than 1 million tonnes of chlorinated paraffins are produced every year and future emissions from products containing chlorinated paraffins are likely to be large (Glüge *et al.* 2016).

Due to the large volumes in which the substances have been and still are manufactured and that ecotoxicological studies indicate that MCCPs and LCCPs, like SCCPs, are bioaccumulative (Castro *et al.* 2018), it is important to continue monitoring the substances in offshore sediments. During the sampling in 2020/2021, analyses were not performed for the 16 monitoring stations using the same methods as previously, since reporting limits were high and both SCCPs and MCCPs had levels below the reporting limits in 2014. Instead, a separate screening study of sediment cores was performed, see below.

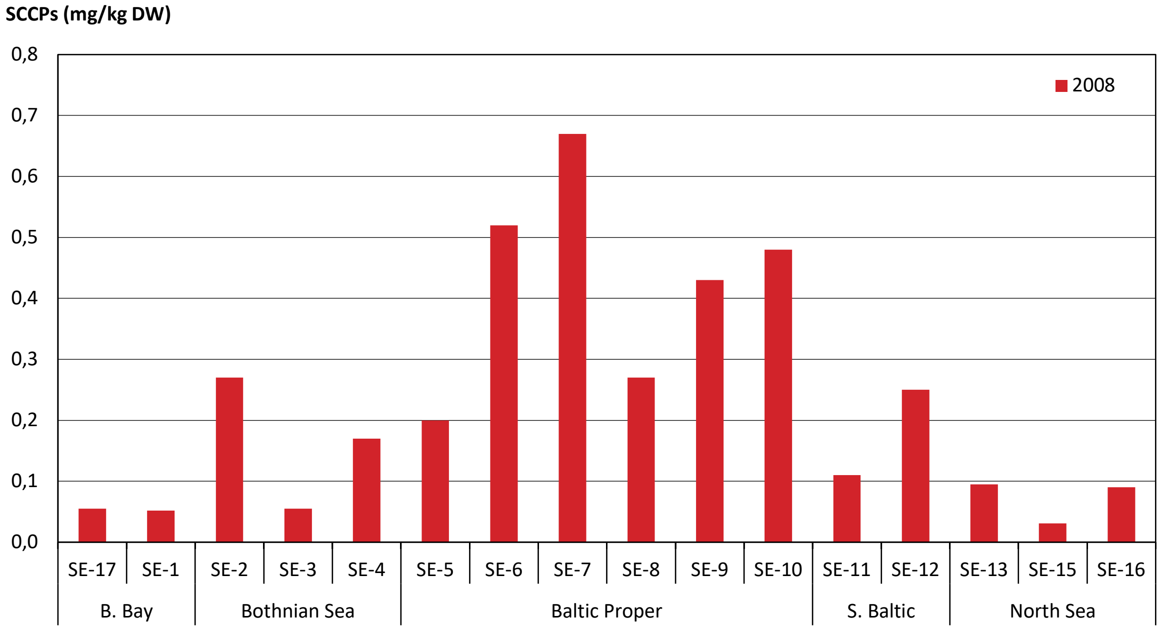

Figure 36. Levels of SCCPs at the 16 stations in 2008.

Chlorinated paraffins in sediment cores

A more detailed study of chlorinated paraffins was conducted by the Department of Environmental Science at Stockholm University on sediment cores collected in 2020 (Yuan *et al.* 2022). Three stations were chosen: SE-1 in the Bothnian Bay, SE-5 in the Baltic Proper, and SE-13 in the North Sea. The cores had been sliced into one-centimetre layers and, to get enough material for the analysis, some layers had to be merged. For the core from SE-5, which had a high water content, 2–5 layers had to be merged, and for the core from SE-1, 2 layers had to be merged in the top layers. For SE-13, no merging was necessary.

Levels of chlorinated paraffins are very elevated at station SE-1 compared to SE-5 and SE-13 (Fig. 37). This is the case particularly for SCCPs, which occur at levels of 150–210 µg/kg DW in the top 10 cm of SE-1 compared to 15–28 µg/kg DW at SE-5 and SE-13, and MCCPs, which occur at levels of 69–110 µg/kg DW in the top 10 cm of SE-1 compared to 7.1–9.6 µg/kg DW at SE-5 and SE-13. Average levels in the top 10 cm of cores are thus 8 and 11 times higher for SCCPs and MCCPs, respectively, at SE-1 compared to SE-5 and SE-13. The levels of vSCCPs are also elevated at SE-1, with 5.5–9.6 µg/kg DW in the top 10 cm compared to 1.6–2.9 µg/kg DW at SE-5 and SE-13, corresponding to on average 3 times higher levels at SE-1. For LCCPs, levels were not elevated at SE-1 compared to the other stations. The high levels at SE-1 were suggested to derive from industrial activities such as a nearby smelter and e-water recycling facility; even higher values were found previously in coastal sediment nearby a metal working factory (Yuan *et al.* 2017, 2022). The high proportion of the now banned SCCPs in offshore sediments (>50% at SE-1) compared to coastal sediments where MCCP is predominant may reflect the content in old, recycled e-waste (Yuan *et al.* 2022). It can also be due to a time lag as it takes time before terrestrial emissions are transported to offshore sediments.

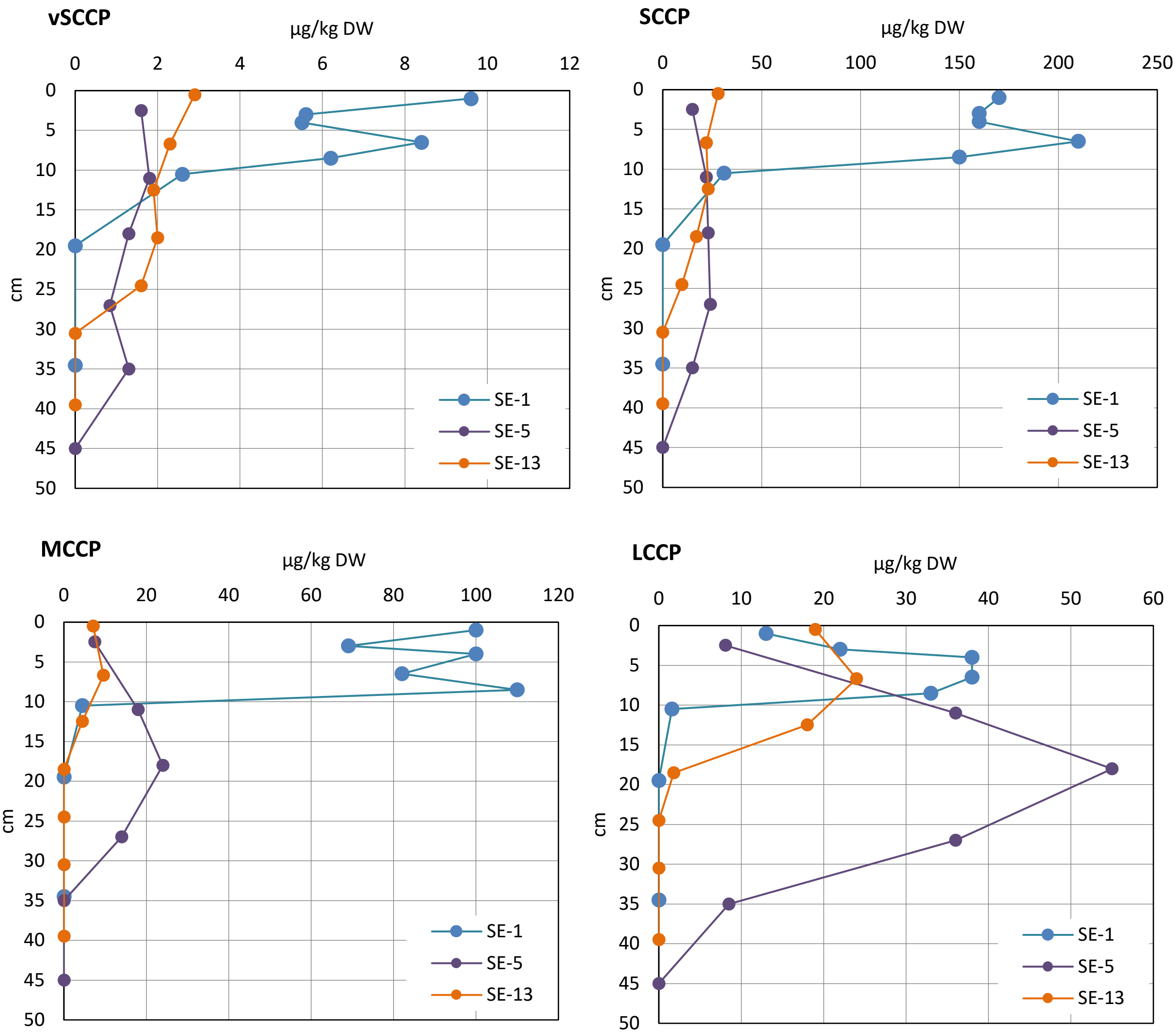

Figure 37. Levels of chlorinated paraffins (vSCCP, SCCP, MCCP and LCCP) on a DW basis in the cores from SE-1, SE-5 and SE-13 in 2020. Values below detection limits were set to 0 in the figure. Detection limits were around 0.7, 6, 4 and 0.2 µg/kg DW for vSCCP, SCCP, MCCP and LCCP, respectively.

The same trends are also apparent when contaminant levels are normalised to the amount of organic carbon in the sediment (Fig. 38). Levels at SE-13 become higher due to the low TOC content in sediment at this station (1.3–2.3%, increasing slightly towards the surface, Fig. 39). TOC levels at SE-5 are higher and increase towards the surface, while TOC levels at SE-1 are in between SE-13 and SE-5 (Fig. 39). When normalised to organic carbon, average levels in the top 10 cm of the sediment core at SE-1 are 8 and 5 times higher, for MCCPs and SCCPs respectively, than at SE-5 and SE-13. This shows that the difference in levels between the stations is not explained by the organic carbon content.

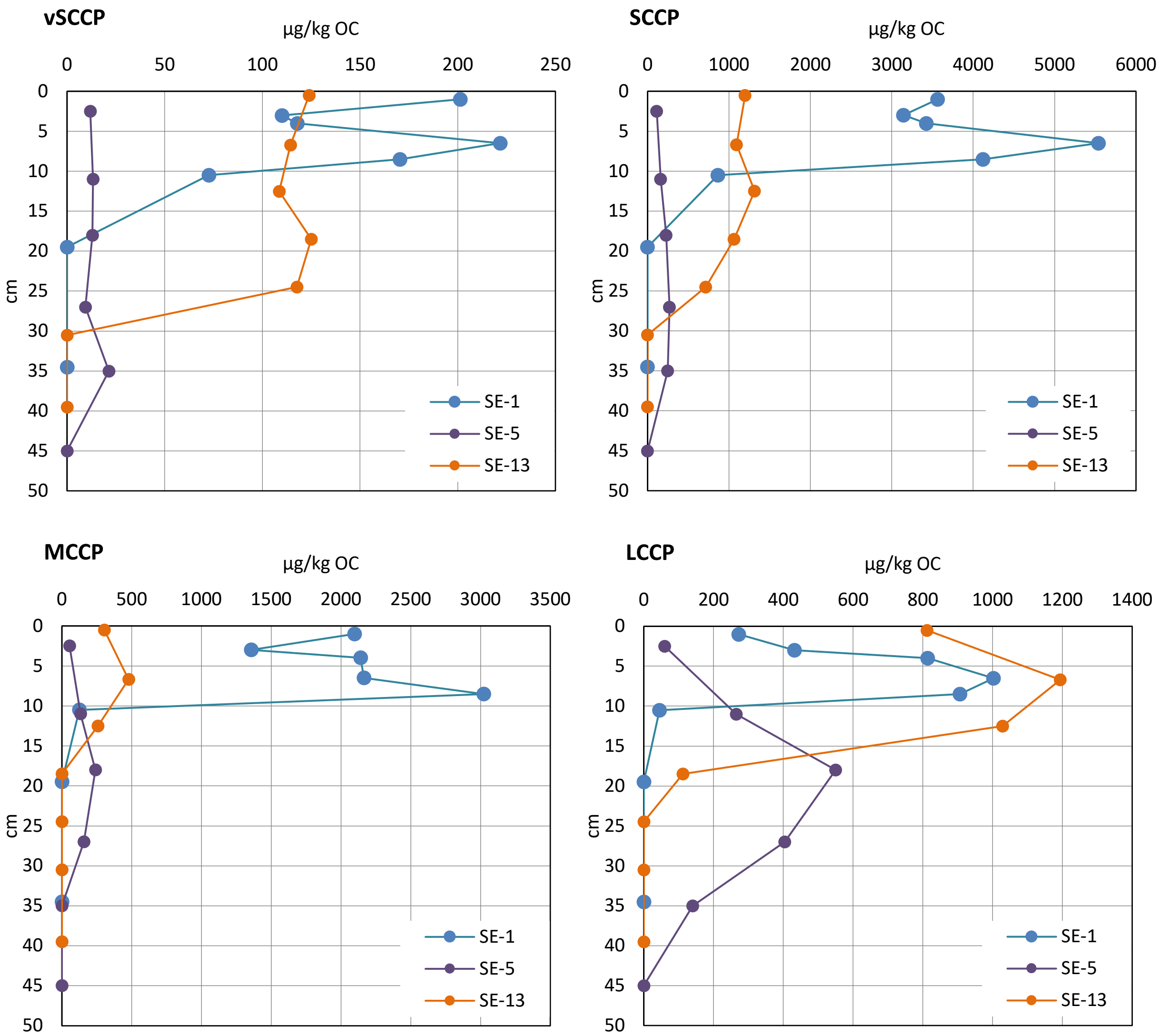

Figure 38. Levels of chlorinated paraffins (vSCCP, SCCP, MCCP and LCCP) on an organic carbon (OC) basis in the cores from SE-1, SE-5 and SE-13 in 2020. Values below detection limits were set to 0 in the figure.

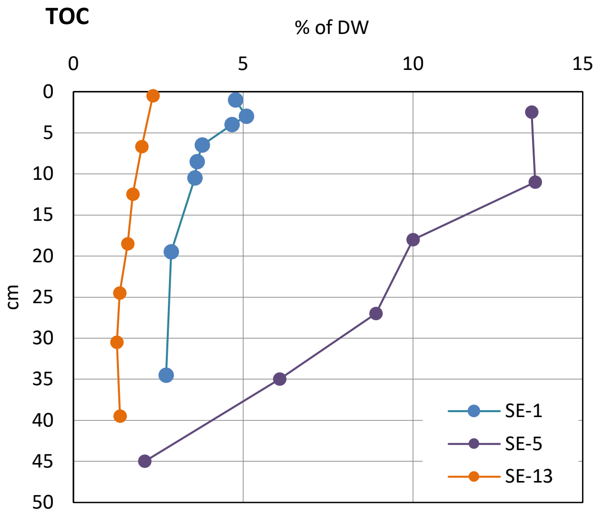

Figure 39. Levels of total organic carbon (TOC) at stations where chlorinated paraffins were analysed in sediment cores.

Time trends at each station (Fig. 40; same trends also on TOC basis) combined with previously determined sediment accumulation rates (Table 2), show the increase in levels to above detection limits at SE-13 started at the sediment depth corresponding to about 1980/1981, slightly later for LCCPs (1989/1991) and vSSCPs (1999/2001). Levels in surface sediments (2018/2020) are somewhat lower for MCCPs and LCCPs than in the layer below (2009/2010) but it is not an obvious decreasing trend. For SCCPs and vSCCPs, levels continue to increase. For station SE-5, the first sample with values above detection limits corresponds to approximately 1985/1987, while values were below detection limits at a sediment depth corresponding to 1975/1977. Levels peak approximately year 2001/2004 for MCCPs and LCCPs, possibly somewhat earlier (1991/1997) for SCCPs. There is a decreasing trend for these three groups while there is no clear trend for vSCCPs. At station SE-1, the first sediment sample with detected levels corresponds to about 1951/1958. There is then a large increase in levels in the sample corresponding to 1964/1970. For LCCPs, values seem to have peaked during 1976–1995 and are now decreasing, while there is no clear decrease for SCCPs and MCCPs. They have instead been at high levels for several decades. For vSCCPs, there is no apparent trend. Considering the increasing trend of SCCPs at SE-13, the consistently high levels of SCCPs and MCCPs at SE-1, and the difference in patterns between offshore and coastal samples (Yuan *et al.* 2022), further studies to identify sources are needed.

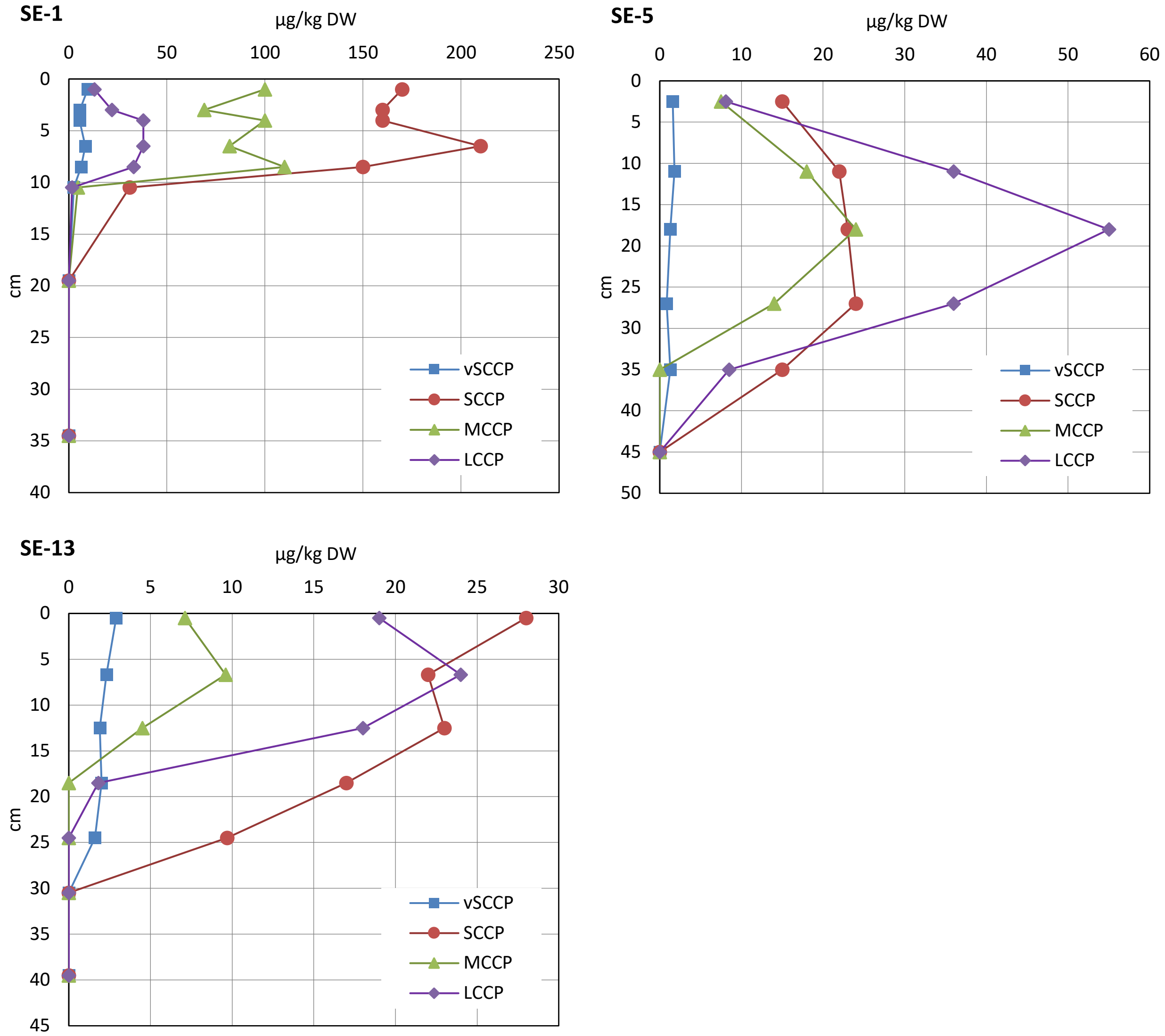

Figure 40. Levels of chlorinated paraffins (vSCCP, SCCP, MCCP and LCCP) at each station.

Organotin compounds and cybutryne

Organotin compounds have mainly been used as biocides, including in antifouling paints for ships. The most well-known among the organotin compounds is the now banned tributyltin (TBT) which, for example, causes endocrine effects (imposex) in sediment-dwelling organisms. In addition to use in antifouling paints, it has been used for wood protection, in soles for shoes and in allergy-friendly pillows (Andersson *et al.* 2012). Triphenyl tin, another compound with three tin atoms, has also been used as a biocide, while mono- and ditin compounds are used in plastic products, for example as stabilizers in PVC. The use of trisubstituted organotin compounds as biocides was banned in the EU in 2006, after the specific use in antifouling paints for ships was banned in 2003 (Andersson *et al.* 2012).

Organotin compounds have been analysed all four monitoring years, except for tetrabutyltin (TeBT) which was not included in 2014. The analyses include, in addition to TBT and TeBT, monobutyltin (MBT), dibutyltin (DBT), monophenyltin (MPhT), diphenyltin (DPhT), triphenyltin (TPhT), monooctyltin (MOT), dioctyltin (DOT) and tricyclohexyltin (TCHT).

Levels of TBT are highest in sediments in the Baltic Proper (Fig. 41). Levels are also high in the Southern Baltic. In the Bothnian Bay and the Bothnian Sea, TBT could only be measured in 2003 and 2020/2021, but not at all stations. The reason for the 're-appearing' levels in 2020/2021 is that reporting limits have been lowered (both detection and quantification limits). In the North Sea, TBT could be measured at levels above the reporting limits in 2003 and 2008, and then again in 2020/2021 after lowered reporting limits. There is a significant positive linear correlation between the levels of TBT and TOC in sediments ($r^2 = 0.24$, $p = 0.0005$, $n = 47$). The degradation products DBT and MBT also display positive and significant correlations with TOC. The correlations remain significant and positive even when excluding Baltic Proper stations for TBT and DBT.

The overall trend over the years is of gradually decreasing levels, most noticeable at the stations in the Baltic Proper, particularly the stations to the east or northeast of Gotland, SE-5 to SE-7. Not all stations have continuously decreasing trends, for example SE-9, SE-11 and SE-12 had somewhat increasing levels between 2008 and 2014, and SE-8 had a higher level in 2020/2021 than in 2014. No comparison to trends in marine biota is possible, as organotin compounds have been below reporting limits in biota during all or almost all the last ten years of the monitoring programme (Soerensen & Faxneld 2020).

TBT is a priority hazardous substance in the WFD and the EQS is 1.6 µg/kg DW for sediment with 5% TOC (SwAM 2019), while the proposed indicator threshold for TBT in sediment for HOLAS III is 1.3 µg/kg DW for a sediment with 5% TOC. In 2014, the EQS was exceeded at all stations with reported levels of TBT, which was all stations in the Baltic Proper and the Southern Baltic. Even though TBT levels have decreased, the EQS and the proposed HOLAS III indicator threshold is still exceeded at seven out of 16 stations in 2020/2021: SE-1 in the Bothnian Bay, SE-7 to SE-9 in the Baltic Proper, SE-11 and SE-12 in the Southern Baltic and SE-13 in the North Sea. To conclude, not all stations in the Baltic Proper exceed the EQS nowadays but stations in other sea areas have been found to still be in exceedance of the EQS thanks to lowered reporting limits from the laboratory.

The ratio between TBT and its degradation products DBT and MBT have changed over the years, with a decrease in the fraction of TBT and an increase in the fraction of degradation products (Fig. 42). This indicates that the TBT in the sediment derive from 'old' releases that have had time to degrade. DBT and MBT are however not only degradation products of TBT but also have their own uses. Levels of DBT and MBT measured in sediments therefore do not necessarily come solely from degradation of TBT. TeBT could be reported in 2020/2021, thanks to a changed sample preparation method. It was then detected at a level of 9.2 µg/kg DW at SE-8, the station with the highest levels of TBT that year. Remaining stations had levels of TeBT below the detection limit of 0.05–0.30 µg/kg DW.

TBT and imposex, i.e., that female snails develop male genitalia due to TBTs endocrine disrupting effects, was an indicator in the second holistic assessment of the Baltic Sea and, second to PBDE, was the parameter that most strongly exceeded its threshold values (HELCOM 2018b). TBT exceeds the EQS in offshore sediments within the monitoring programme, and levels are higher in coastal sediments. The continued risk of dispersal of TBT from contaminated coastal sediments should therefore be addressed.

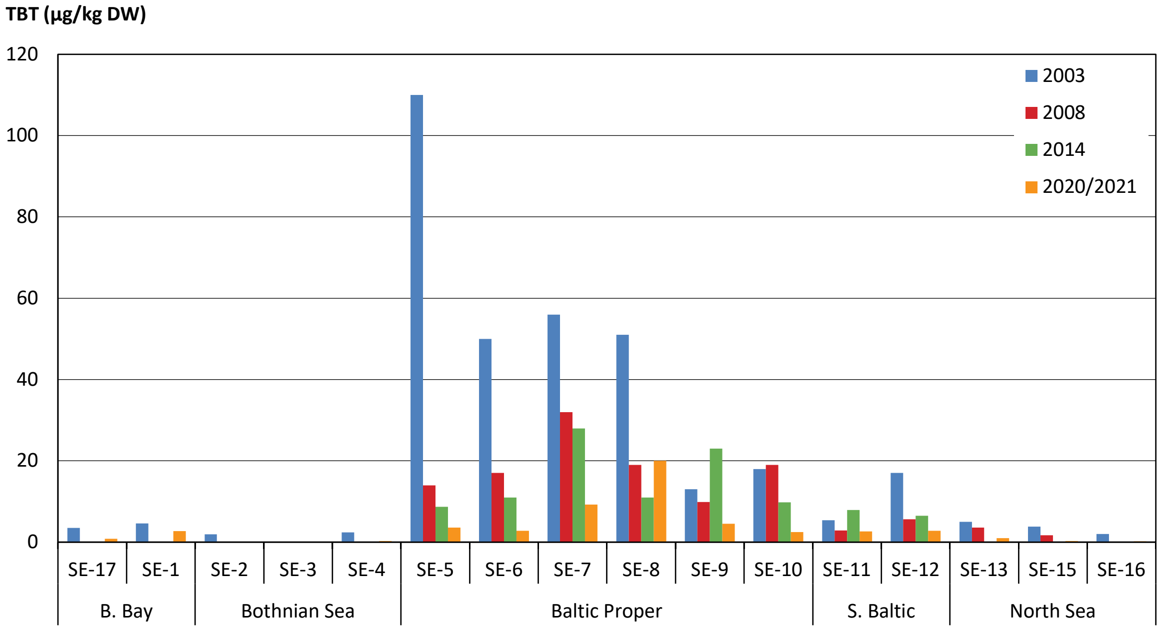

Figure 41. Levels of TBT at the 16 stations during the four monitoring years. Note that the value for station SE-5 in 2020/2021 is less certain as the sample had been exposed to temperatures above 0°C for an extended time.

a)

b)

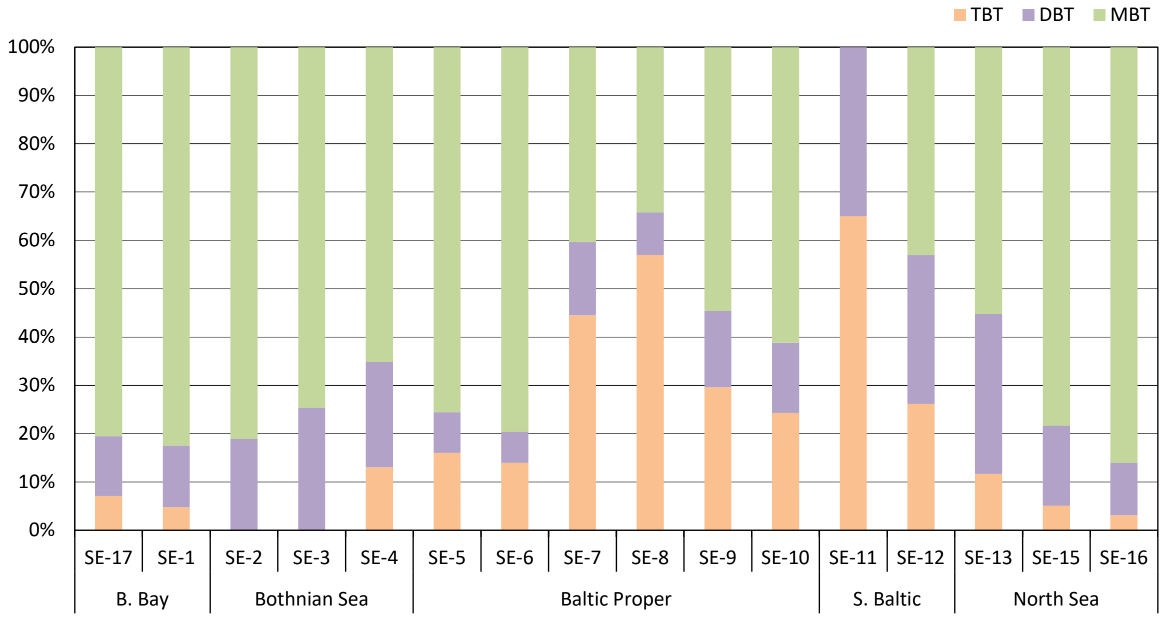

Figure 42. The distribution between TBT and the degradation products DBT and MBT at the 16 monitoring stations in a) 2003 and b) 2020/2021.

The phenyltin compounds MPhT, DPhT and TPhT were analysed all four monitoring years but were all below the reporting limit in 2003, and TPhT also in 2008 and 2014. In 2020/2021, the reporting limits had been lowered to 0.05–0.30 µg/kg DW and the substances were then above the reporting limit at more stations (Fig. 43). It is unclear why levels are so variable over the years but the change of laboratory and method between 2003 and 2008 may have caused the difference between these years, with markedly higher levels of MPhT and DPhT in 2008. There is no clear geographical pattern of the phenyltin compounds, possibly because levels are close to reporting limits, and there are also no correlations between phenyltins and levels of TOC in the sediments.

The octyltin compounds MOT and DOT were below the reporting limit at all stations in 2003, but above reporting limits at several stations during the last three monitoring years (Fig. 44). Levels are low in the Bothnian Bay and Bothnian Sea, but higher and relatively comparable in the remaining sea areas. The highest levels occur in the Baltic Proper in 2020/2021 and in the Southern Baltic in 2014, but at most stations the levels are lower in 2020/2021 than during previous years. No correlation with TOC levels in the sediment could be observed, even when excluding the high value for station SE-7 in 2020/2021.

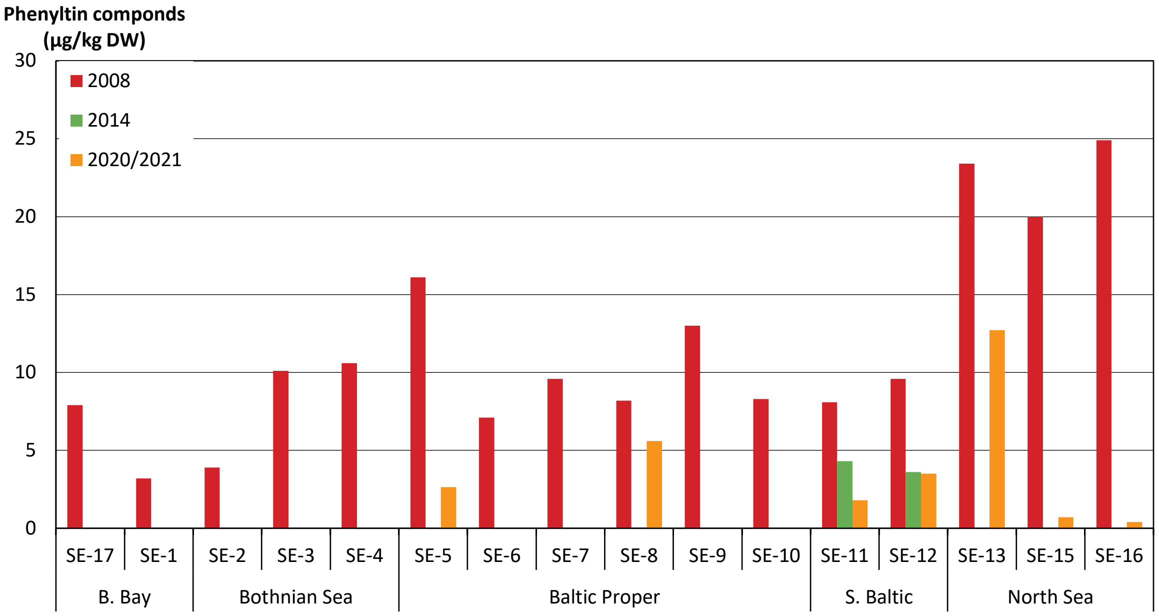

Figure 43. Levels of the sum of phenyltin compounds, i.e., MPhT, DPhT and TPhT, at the 16 stations during the three last monitoring years. All compounds were below the reporting limit of 1 µg/kg DW in 2003. The value for station SE-5 in 2020/2021 is less certain as the sample had been exposed to temperatures above 0°C for an extended time. The high value at SE-13 in 2020/2021 is due to a very high level of DPhT (12 µg/kg DW) and should be considered as more uncertain.

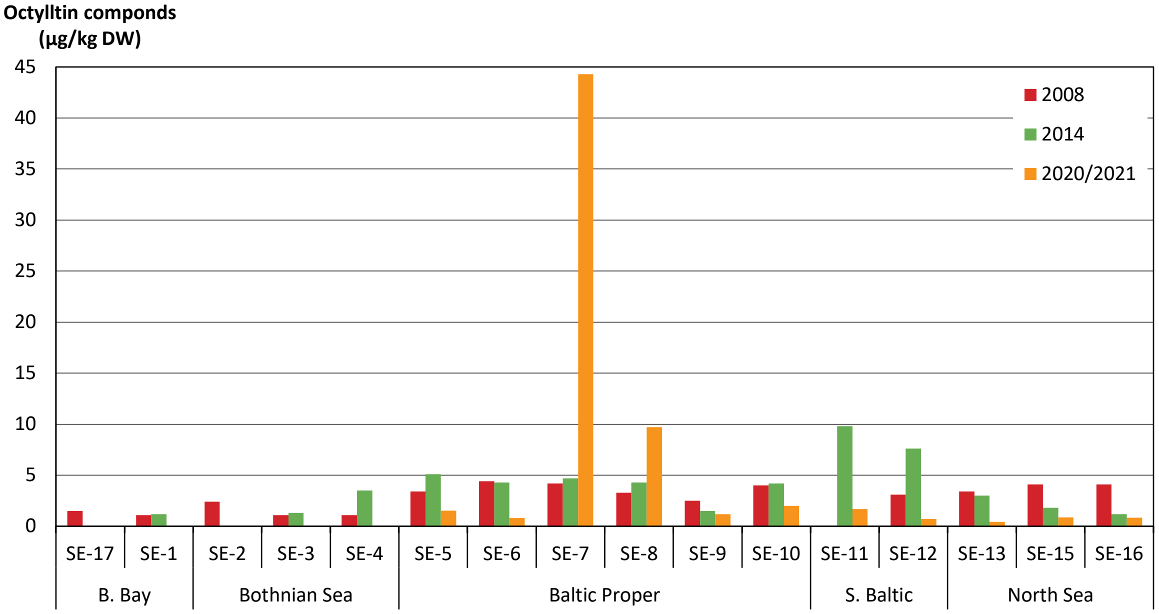

Figure 44. Levels of the sum of octyltin compounds, i.e., MOT and DOT, at the 16 stations during the last three monitoring years. Both compounds were below the reporting limit of 1 µg/kg DW in 2003. The value for station SE-5 in 2020/2021 is less certain as the sample had been exposed to temperatures above 0°C for an extended time. The high value at SE-7 in 2020/2021 is due to a very high level of DOT (44 µg/kg DW) and should be considered as more uncertain.

Cybutryne, also known as irgarol, is an organic compound that has been used in antifouling paints for ships. It was banned for biocidal use in the EU in 2018. Cybutryne has been analysed during the three last monitoring years and occurs in the highest levels in sediments in the Baltic Proper (Fig. 45). At the stations that had measurable levels in both 2008 and 2014, cybutryne levels showed an increase in the Baltic Proper and, to a smaller degree, in the Southern Baltic, while decreasing slightly in the other sea areas. Station SE-8 in the Baltic Proper was an exception with decreasing levels between 2008 and 2014. Levels decreased further in 2020/2021 at most stations.

The three stations in the northern and north-eastern part of the Baltic Proper (SE-5, SE-6 and SE-8) had markedly higher levels than the stations in the south and west (SE-7, SE-9 and SE-10) both in 2008 and 2014, and to some extent also in 2020/2021 (Fig. 45). This is possibly related to the sediment accumulation rates at each station. At stations SE-5, SE-6 and SE-8, the sediment accumulates rapidly and an average centimetre in the sediment corresponds to particles that have sedimented during the past year (see Table 2). At stations SE-7, SE-9 and SE-10, the accumulation is much slower and the time it takes to form an average centimetre is 13–22 years. The high levels at the stations where the particles have sedimented over the past year or so, compared to the stations with lower accumulation rates, indicate that levels have increased in the environment during the last decades. Levels have however dropped between 2014 and 2020/2021, although they are still highest at the three stations with the fastest accumulation rate in the Baltic Proper.

Cybutryne displays a positive and significant linear correlation with TOC levels in sediment ($r^2 = 0.29$, $p = 0.0003$, $n = 41$) when Baltic Proper stations are included. Cybutryne is a priority substance within the WFD and although there is no EQS for sediment, there is an indicative value (SwAM 2018b) of 0.18 µg/kg for sediment with 5% TOC. This value was exceeded at six stations in 2008, at four stations in 2014, and at one station in 2020/2021, SE-5. As cybutryne has been banned in the EU, levels should continue to fall, but the substance should continue to be analysed in the monitoring programme as levels are still close to the indicative value.

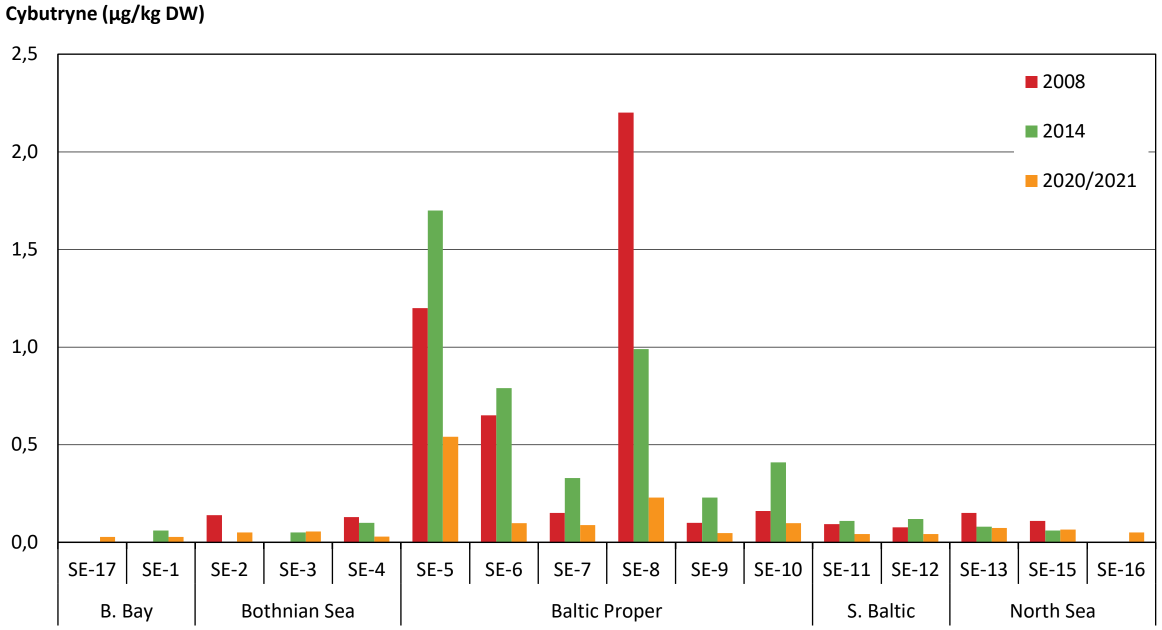

Figure 45. Levels of cybutryne at the 16 stations during the last three monitoring years. Note that the value for station SE-5 in 2020/2021 is less certain as the sample had been exposed to temperatures above 0°C for an extended time.

Organochlorine pesticides

DDT

The insecticide dichlorodiphenyltrichloroethane (DDT) has been prohibited since the 1970s in many countries including Sweden, but is still in use in some parts of the world, primarily to combat malaria. In Sweden, DDT was used in agriculture, forestry (for example to protect plants against pine weevil) and animal husbandry (for example fur farms).

DDT and its degradation products dichlorodiphenylethene (DDE) and dichlorodiphenyldichloroethane (DDD) have been analysed all four monitoring years. The levels of these three substances, that is, *p,p'*-DDT, *p,p'*-DDE och *p,p'*-DDD, are summed as ∑DDT (Fig. 46). The substance *o,p'*-DDT was not analysed in 2003 and 2008 and is therefore not included in the sum. Levels of ∑DDT are clearly higher in sediment in the Baltic Proper and the Southern Baltic than in the other sea areas. In 2020/2021, levels were between 1.7 and 5.9 µg/kg DW in the Baltic Proper and the Southern Baltic, while they were between 0.17 and 0.58 in the other sea basins. Levels of ∑DDT seem to be decreasing over time; only at station SE-9 are levels higher in 2020/2021 than 2014.

Interestingly, the levels of the three different DDT substances have different geographical patterns. Levels of DDE and DDD are in the same range and are highest in the Baltic Proper and Southern Baltic, while levels of DDT are considerably lower than DDE and DDD, but higher in the Bothnian Bay than in other sea areas. A figure showing the distribution between the three substances included in ∑DDT clearly illustrates that the proportion of the original product, DDT, is highest at the stations in the Bothnian Bay where it constitutes around half of the ∑DDT, followed by the Bothnian Sea and the North Sea (Fig. 47). DDT generally degrades to DDD under oxygen-free conditions and to DDE under oxic conditions. A partial explanation for the higher proportion of DDT in, primarily, the Bothnian Bay may be the oxic conditions prevailing in the surface sediment (Table 2). Oxygen conditions should not however be the only factor because the oxygen levels in the bottom water at stations in the North Sea and Bothnian Sea are also quite high. A high proportion of DDT in relation to degradation products is usually interpreted as a recent release, but it is not feasible that this would have occurred only in the Bothnian Bay. The distribution between the ∑DDT substances are also similar between the 2008, 2014 and 2020/2021 monitoring surveys. Other possible explanations could be north – south gradients in the degradation rate of DDT or differences in the atmospheric deposition of DDT and degradation products.

The DDT substance that occurs in the highest concentrations in organisms is generally DDE because DDD can be metabolized into more water-soluble substances that can be excreted from the body (Bernes 1998). Levels of DDT are for example often below the reporting limits in sea eagles, while the levels of DDD only make up a few percent of the sum of DDD and DDE (Helander 2014). Data on DDE in herring sampled within the national environmental monitoring programme for contaminants in marine biota show that levels were generally highest in the Southern Baltic and the Baltic Proper, with lower levels in the northern parts of the Baltic Sea and the North Sea (2017–2019; Soerensen & Faxneld 2020). This is consistent with the geographical distribution of levels in sediments (Fig. 46). Generally downward trends for DDT substances in biota are observed during the last decade (2010-2019; Soerensen & Faxneld 2020).

Levels of ∑DDT display a positive and significant linear correlation with TOC levels in the sediment ($r^2 = 0.28$, $p < 0.0001$, $n = 64$). Since DDT, DDE and DDD are substances that preferably bind to organic carbon, this is not surprising. The southern Baltic Sea tends to have elevated levels while the Bothnian Bay always has lower levels than the correlation with TOC predicts. The Bothnian Sea stations generally have lower levels than the correlation with TOC predicts, while the stations in the Baltic Proper and the North Sea are relatively evenly distributed around the regression line.

DDT is one of the original 12 POPs that were listed in the Stockholm Convention when it entered into force in 2004. The substance and its degradation products are also listed in the WFD but there is no EQS for sediment. DDT and its degradation products were included in the first HELCOM holistic assessment of the Baltic Sea in 2010 but are no longer seen as worrying and were not included in the second holistic assessment, although they should continue to be monitored (HELCOM 2018b). They are not proposed as indicators for HOLAS III.

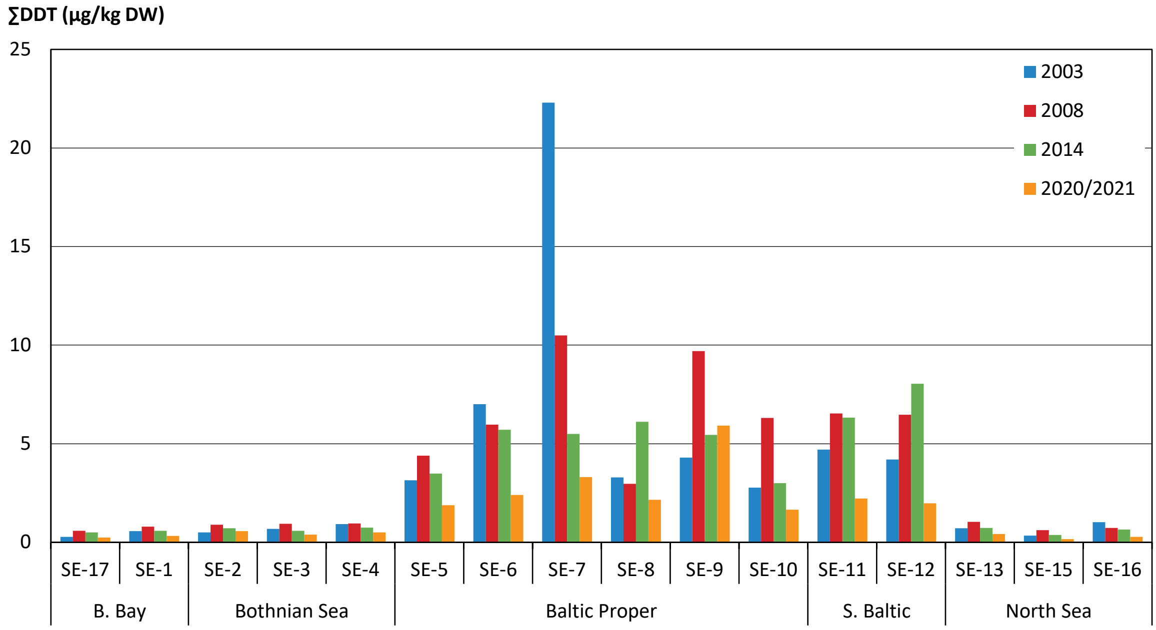

Figure 46. Levels of ∑DDT at the 16 stations during the four monitoring years. Note that the value for SE-5 in 2020/2021 is less certain as the sample had been exposed to temperatures above 0°C for an extended time. The high value at SE-7 in 2003 is mainly due to a high level of *p,p′*-DDD at 17 µg/kg DW.

Figure 47. The distribution of the three substances constituting ∑DDT at the 16 monitoring stations in 2020/2021.

HCHs

Hexachlorocyclohexanes, HCHs, have been used as insecticides mainly in agriculture and forestry. Commercial products, so-called technical HCH, consisted of a mixture of HCH isomers where α-HCH was the most common followed by *g*-HCH (Venier & Hites 2014). The only effective insecticide is however *g*-HCH and over time technical HCH was replaced by a purified product, lindane, where the content of isomers other than *g*-HCH had been minimized. In addition to *a*- and *g*-HCH, the sum of HCH (∑HCH) also includes *b*-HCH. This isomer was present in small quantities in the commercial products but due to its low degradation rate it will form an increasing proportion of ∑HCH in the environment over time. The use of HCH is now banned in most countries, but there is a limited use of lindane against lice and scabies.

HCHs have been analysed all four monitoring years. In 2003, the levels at many stations were below the reporting limits (0.2–0.4 µg/kg DW for *a*-HCH and *b*-HCH, 0.2-1,5 µg/kg DW for *g*-HCH). Due to this and that the analytical laboratory was changed between 2003 and 2008, and the reporting limits were lowered, data from 2003 have been omitted from the data analysis.

The results clearly show that the ∑HCH levels in sediments are higher in the Baltic Proper than in other sea areas (Fig. 48). The Southern Baltic also has higher levels than remaining sea areas. Like ∑DDT, the levels of ∑HCH have a positive and significant linear correlation with TOC levels in the sediment, ($r^2 = 0.32$, p <0.0001, n = 58). Levels of ∑HCHs appear to decrease over time, albeit with some exceptions at some stations (Fig. 48).

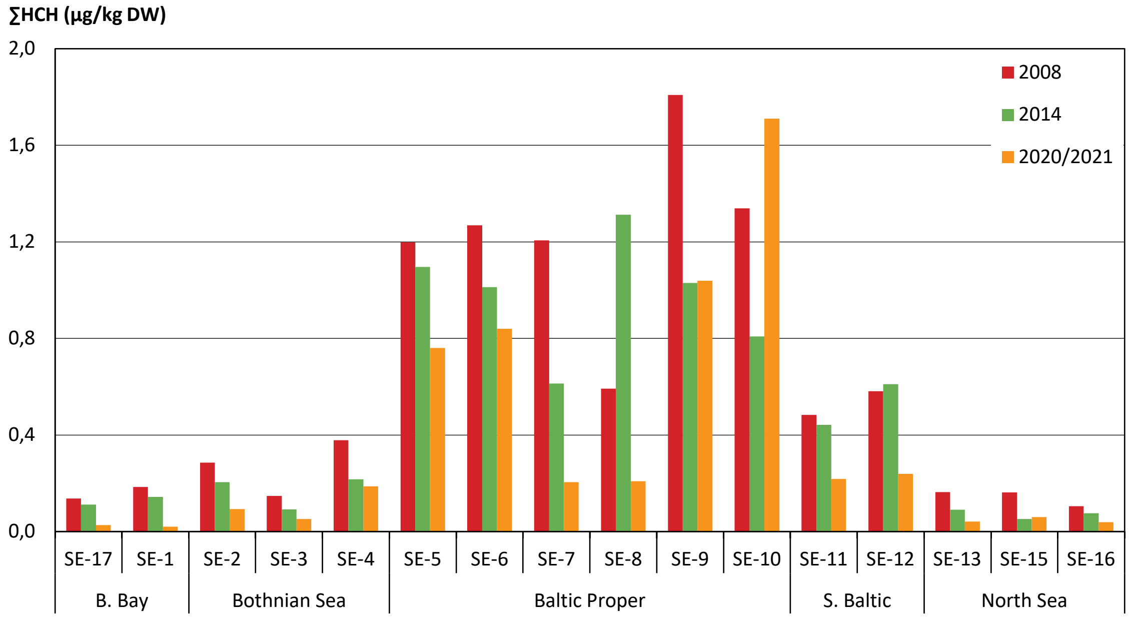

Figure 48. Levels of ∑HCH at the 16 stations during the last three monitoring years. Note that the value for station SE-5 in 2020/2021 is less certain as the sample had been exposed to temperatures above 0°C for an extended time. In 2020/2021, the levels of *a*-HCH are particularly high at SE-5, SE-6, SE-9 and SE-10 (0.38 µg/kg DW or above), while the level of *g*-HCH is particularly high at SE-10 (0.45 µg/kg DW).

There are clear differences between sea areas in the levels of the three substances included in ∑HCH. The levels of *b*-HCH, which is the dominant substance, are higher in the Baltic Proper than in other sea areas, while the levels of *a*-HCH are higher in both the Baltic Proper and the Southern Baltic than in other sea areas. The levels of *g*-HCH, on the other hand, are of the same order of magnitude in all sea areas and occur below reporting limits at some stations in later years. These geographical differences give a pattern with a greater fraction of *b*-HCH in the Baltic Proper, almost equal fractions of *a*-HCH and *b*-HCH in the Southern Baltic and the North Sea, and *g*-HCH being the dominant HCH substance in the Gulf of Bothnia in 2014 (Fig. 49a). This illustrates that the elevated ∑HCH levels in the Baltic Proper are mainly caused by elevated levels of *b*-HCH. Of the various HCH substances, *b*-HCH is the most difficult to degrade (Willett *et al.* 1998), and it is possible that the reducing conditions prevailing at stations in the Baltic Proper (Table 2) make the degradation particularly slow here. In 2020/2021 however, this pattern was not apparent, mainly due to high levels of *a*-HCH at some stations in the Baltic Proper and *g*-HCH being close to or below the reporting limits at some stations (Fig. 49b).

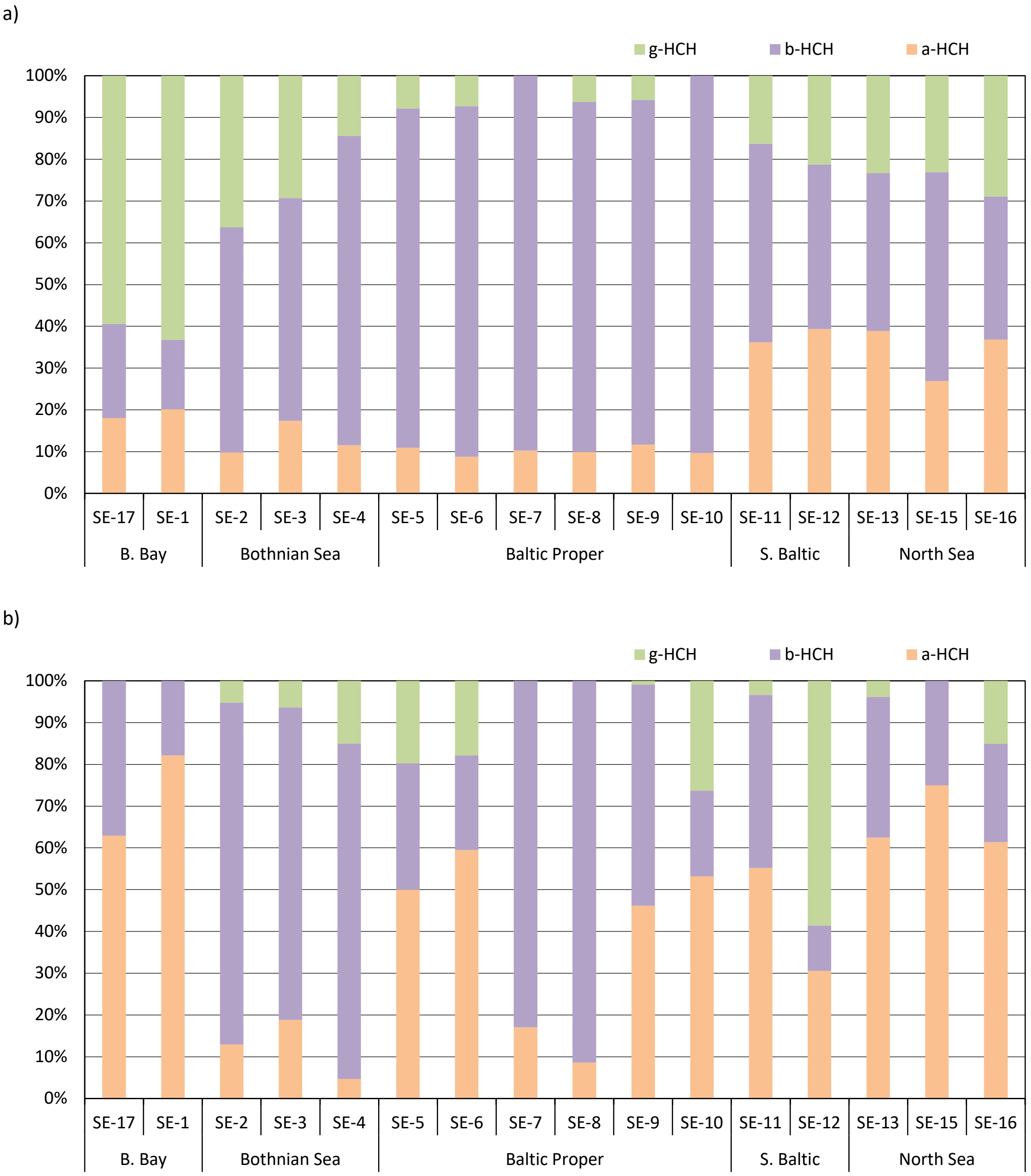

Figure 49. The distribution of the three substances constituting ∑HCH at the 16 monitoring stations in a) 2014 and b) 2020/2021. The distribution pattern for 2008 was similar to 2014.

In the national environmental monitoring of contaminants in marine biota, the levels of *b*-HCH in herring are higher in the Baltic Proper, the southern Baltic and the southern Bothnian Sea than in the Bothnian Bay and the North Sea (2017–2019; Soerensen & Faxneld 2020). The geographical patterns are thus similar but not identical between sediment and biota. A consistent decrease can be seen for *b*-HCH for all types of biota at almost all sites during the last 10 years (2010–2019; Soerensen & Faxneld 2020), and HCH levels thus seem to be decreasing in both biota and sediment.

HCHs were listed in the Stockholm Convention in 2009 and are a priority hazardous substance in the WFD. There is no EQS for HCHs in sediment, but there is an indicative value for marine sediments of 1.1 µg/kg DW, which applies at a TOC content of 10% (SwAM 2018b). The indicative value for ∑HCH was exceeded at stations SE-9 and SE-10 in the Baltic Proper in 2008 and at station SE-10 in 2020/2021, but not at any station in 2014. Like DDTs, HCHs were included in the first HELCOM holistic assessment of the Baltic Sea in 2010 but are no longer seen as worrying even though they should continue to be monitored (HELCOM 2018b).

Chlordanes

Chlordanes have mainly been used in agriculture, but also in gardens and, for example, against termites in buildings. It was banned in Sweden in the 1970s and the quantities used are estimated to be small. Commercial chlordane products could consist of more than a hundred different substances, the most common being α-chlordane (also known as cis-chlordane), *g*-chlordane (also known as trans-chlordane or beta-chlordane) and trans-nonachlor (Dearth & Hites 1991).

Like DDTs and HCHs, chlordanes have been analysed all monitoring years. In 2003, however, the levels of the three substances included in ∑Chlordane (α-chlordane, *g*-chlordane and trans-nonachlor) were often below the reporting limits. Therefore, data from 2003 have not been included. From the results from the other three years, it is clear that the levels of chlordanes have the same geographical pattern as DDTs and HCHs, i.e., with the highest levels in the Baltic Proper and the Southern Baltic (Fig. 50). The chlordane levels increased at some stations between 2008 and 2014 but decreased between 2014 and 2020/2021 at all stations.

Levels of ∑Chlordanes have a positive and significant linear correlation with TOC levels in the sediment ($r^2 = 0.36$, $p < 0.0001$, $n = 54$), like ∑DDT and ∑HCH. There is a tendency towards lower levels than expected based on the correlation with TOC in the Gulf of Bothnia, but the levels of chlordanes are not elevated in the Southern Baltic, at least not for the 2020/2021 monitoring.

There are some differences between the geographical pattern of the three chlordane substances: α-chlordane and trans-nonachlor occur in elevated levels in the Baltic Proper compared to the other sea areas, while *g*-chlordane have elevated levels in both the Baltic Proper and the Southern Baltic, i.e., stations SE-11 and SE-12. That *g*-chlordane is the predominant compound at these two stations is apparent in Fig. 51.

A lower isomer fraction of *g*-chlordane is usually associated with emissions having had time to age and be transported from the source, because there are differences in degradation rate and volatility between *g*- and *a*-chlordane (Bidleman *et al.* 2002, Stern *et al.* 2005). In the commercial chlordane products, the fraction was about 0.5 (Bidleman *et al.* 2002), and that the fraction is higher than this in the Southern Baltic is difficult to explain. One possible explanation could be the influence of other pesticides that are contaminated with *g*-chlordane, e.g., the now banned heptachlor (Bidleman *et al.* 2002).

Chlordanes are one of the original 12 POPs listed in the Stockholm Convention, but they are not priority substances or RBSP in the WFD in Sweden. There are therefore no EQS, indicative values or HOLAS threshold values to compare the measured sediment levels to.

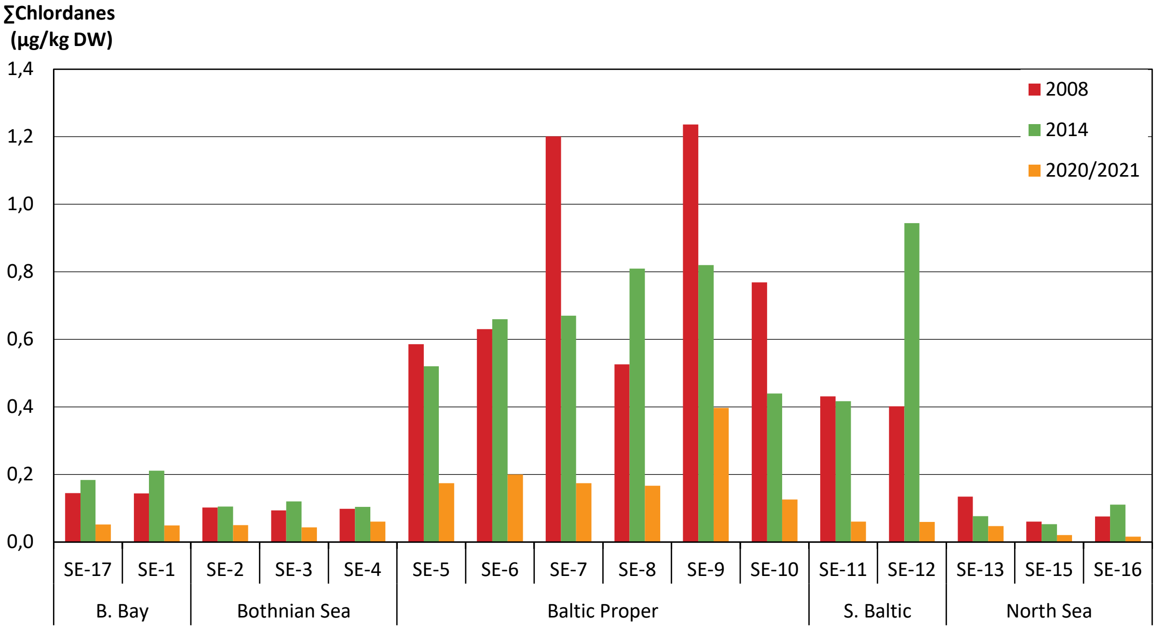

Figure 50. Levels of ∑Chlordanes at the 16 stations during the last three monitoring years. Note that the value for SE-5 in 2020/2021 is less certain as the sample had been exposed to temperatures above 0°C for an extended time.

Figure 51. The distribution of the three substances constituting ∑Chlordanes at the 16 monitoring stations in 2020/2021.

HCB and other chlorobenzenes

Hexachlorobenzene (HCB) has previously been used on a large scale mainly as an insecticide in agriculture. This use has been banned in Sweden since 1980, but the substance is also formed unintentionally in various combustion and chlorination processes. HCB is one of the original 12 POPs in the Stockholm Convention, and intentional global production ceased in 2004 (Wang *et al.* 2010).

HCB has been analysed all four monitoring years and generally occurs in levels above the reporting limit, except for some stations in 2003 (reporting limit 0.2 µg/kg DW). The highest levels of HCB have been measured at stations in the Baltic Proper, up to almost 1 µg/kg DW, but no clear differences in HCB levels between the sea areas can be observed (Fig. 52). The levels of HCB display a positive and significant linear correlation with the levels of TOC in the sediment; however, it has a very low degree of correlation ($r^2 = 0.08$, $p = 0.02$, $n = 59$), and the HCB levels in the sediment are therefore not explained to any great extent by the TOC levels.

HCB is a priority hazardous substance in the WFD and although there is no EQS for sediment, there is an indicative value of 16.9 µg/kg TS which refers to sediment with 10% TOC (SwAM 2018b). This indicative value, normalised to the TOC content in the sediment, was not exceeded at any stations during the four monitoring years. Concentrations of HCB in herring sampled in the monitoring of marine biota are highest in the Baltic Proper (both the southern and northern parts). The lowest concentrations occurred in the North Sea followed by the Bothnian Bay, while concentrations in the Bothnian Sea are higher (2010–2019; Soerensen & Faxneld 2020). There is thus a clearer geographical pattern in marine biota than in sediments, but concentration differences in herring from different sea areas have decreased during the last decades and concentrations are now more similar (Soerensen & Faxneld 2020).

HCB has a high volatility compared to many other persistent organic pollutants and it is thus easily transported via the atmosphere. Atmospheric deposition is believed to be the dominant source of HCB to the Baltic Sea, according to a modelling performed for the Baltic Proper and the Bothnian Sea (SwEPA 2009). This means that the levels are relatively even geographically, although elevated levels can be seen in areas affected by point sources (Josefsson 2018). In 2014, levels increased compared to 2008 at many stations, particularly at stations SE-5, SE-6 and SE-8 in the Baltic Proper which displayed the highest levels in that year. These three stations are, as mentioned, the monitoring stations in the Baltic Sea with the highest sediment accumulation rate, approximately 1 cm per year (Table 2), and they respond more quickly to changes in concentrations in the aquatic environment.

The large increase in levels between 2008 and 2014 at these stations, and a general although lesser increase at most other stations, was worrying. A survey on sediment samples taken between 1986 and 2015 and reported to databases at SGU showed that the levels of HCB in surface sediments previously decreased but increased once more since the beginning of the 2000s (Josefsson 2018). This observation is supported by analyses of sediment cores from the monitoring stations (Sobek *et al.* 2015) and increasing HCB trends in biota in the Baltic Sea (Bignert *et al.* 2016) and in other matrices such as reindeer and air. The reason for the increasing trend was not clear but could be increased primary emissions from unknown sources or increased emissions from secondary sources, i.e., areas previously contaminated with HCB (Josefsson 2018). The latest monitoring in 2020/2021 however revealed decreasing levels compared to 2014 at all stations, and no elevated levels at SE-5, SE-6 and SE-8 compared to other stations (Fig. 52). In the national environmental monitoring of contaminants in marine biota, there is no overall trend for HCB for the last decade (2010–2019, Soerensen & Faxneld 2020).

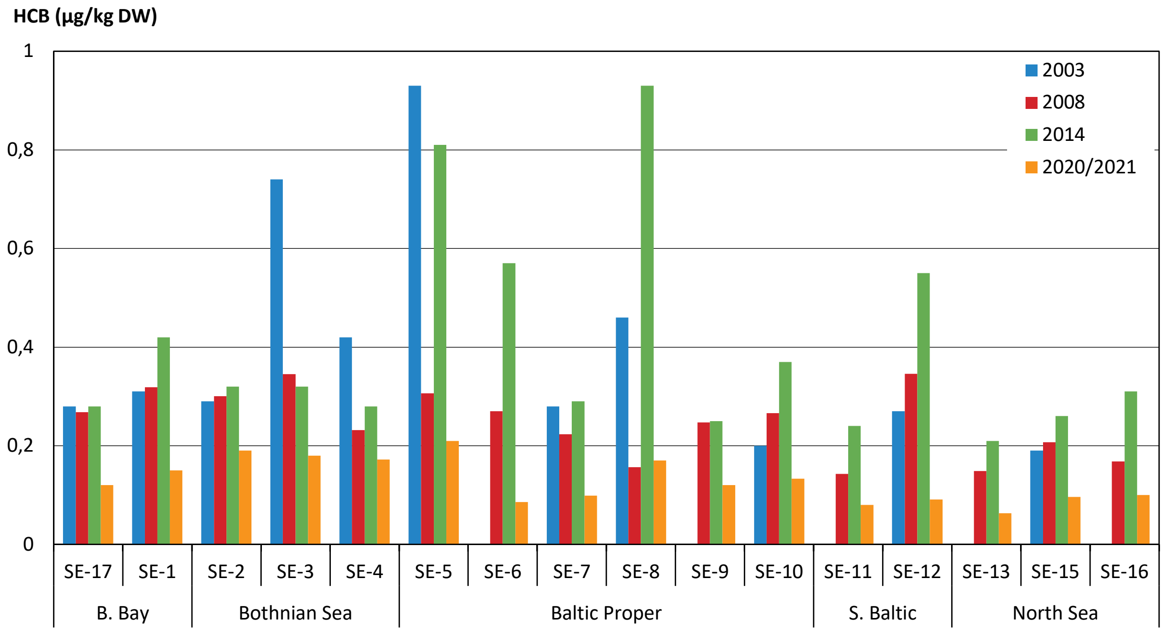

Figure 52. Levels of HCB at the 16 stations during the four monitoring years. Note that the value for station SE-5 in 2020/2021 is less certain as the sample had been exposed to temperatures above 0°C for an extended time.

Pentachlorobenzene was analysed in 2008 and 2014. Levels were below the reporting limit of 1 µg/kg DW in 2008 but could be detected at all stations in 2014 in levels of 0.04–0.27 µg/kg DW. Levels were highest at the same stations as for HCB, and the levels of pentachlorobenzene are well correlated with the HCB levels ($r^2 = 0.79$, $p < 0.0001$, $n = 16$, linear regression on logarithmic data for 2014) and about one third as high. Since HCB and pentachlorobenzene can be formed in the same processes and HCB is degraded to pentachlorobenzene (Adrian & Görisch 2002), the correlation is expected. Pentachlorobenzene has been listed in the Stockholm Convention since 2009, like HCB. It is also a priority hazardous substance in the WFD but lacks an EQS for sediment. There is an indicative value for pentachlorobenzene from SwAM (2018b) at 400 µg/kg DW for sediments with 10% TOC, which is pointed out to be a very uncertain value. It is not exceeded at any of the monitoring stations.

Three **trichlorobenzenes** (1,2,3-, 1,2,4- and 1,3,5-trichlorobenzene) were analysed in 2008 and 2014. Only 1,2,4-trichlorobenzene could be measured in levels above the reporting limit while the other two substances were always below the reporting limits of 0.5 µg/kg DW in 2008 and 0.2 µg/kg DW in 2014. The levels of 1,2,4-trichlorobenzene were below the reporting limit for several stations mainly in the Baltic Proper both years. The highest level (1.4 µg/kg DW) was measured at station SE-12 in 2014, but there were no clear differences between the sea areas. Trichlorobenzenes are priority substances in the WFD but lack EQS or other guidance values for sediment.

Other pesticides

Among the analysed pesticides, some have never been present at levels exceeding the reporting limits. This is the case for a number of pesticides nowadays banned in Sweden and the EU such as **alachlor** (analysed 2003 and 2008, reporting limit 0.2–1 µg/kg DW), **atrazine** (analysed 2003 and 2008, reporting limit 2–10 µg/kg DW), **isoproturon** (analysed 2003 and 2008, reporting limit 2–40 µg/kg DW), **chlorfenvinphos** (analysed 2003 and 2008, reporting limit 1–20 µg/kg DW), **chlorpyrifos** (analysed 2003 and 2008, reporting limit 1–5 µg/kg DW), **pentachlorophenol** (analysed 2003 and 2008, reporting limit 0.5–6 µg/kg DW), and **trifluralin** (analysed 2003 and 2008, reporting limit 0.1–1 µg/kg DW). The pesticide **DCOIT** (dichlorooctylisothiazolinone), that is present in, for example, paints, was analysed in 2008 and all stations had levels below the reporting limit of 0.05–0.1 µg/kg DW.

Different forms of the banned insecticide **endosulfan**, *a*-endosulfan, *b*-endosulfan and endosulfan sulfate, have been analysed in 2008 and 2014 but levels were always below the reporting limit (0.2 µg/kg DW). In 2003, the sum of endosulfan was analysed but was below the reporting limit (0.3–0.8 µg/kg DW) at all stations except SE-12, where the level was 0.3 µg/kg DW, i.e., in the same range as the reporting limit.

Simazine, a banned herbicide, was analysed all monitoring years but levels are generally below the reporting limit (0.5–10 µg/kg DW), except at two stations in 2008 and 2014. The measured levels (0.9–4.6 µg/kg DW) are however in the same range as the reporting limit and no conclusions can be drawn about the geographical patterns or time trend regarding simazine levels.

Diurone is an herbicide that has also been used in antifouling paints for ships, even though no such use has been registered in Sweden according to the Swedish Chemicals Agency's Pesticide Register (www.kemi.se). Diurone has been analysed every year but was always below the reporting limit in 2003 (20–40 µg/kg DW) and 2008 (2 µg/kg DW). In 2014, it was detected at all 16 stations in concentrations from 0.07 to 0.31 µg/kg DW and in 2020/2021 at seven stations, while the rest were below the detection limit of 0.08 µg/kg DW (Fig. 53). As apparent from the figure, there are no clear differences between sea areas or between years.

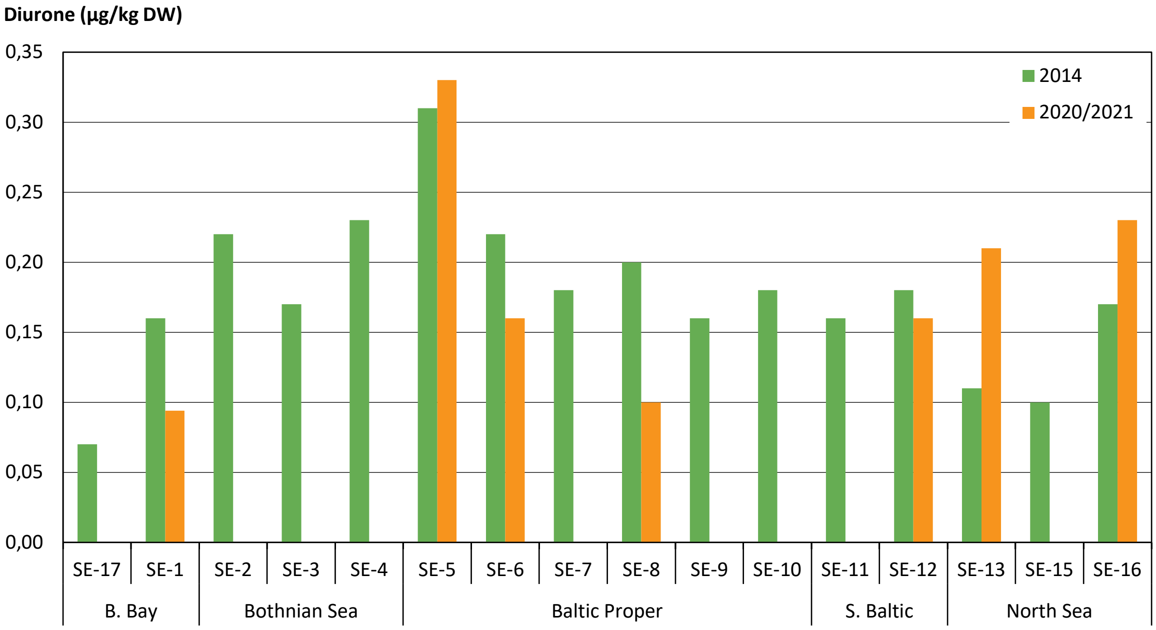

Figure 53. Levels of diurone at the 16 stations during the last two monitoring years. Note that the value for station SE-5 is less certain as the sample had been exposed to temperatures above 0°C for an extended time.

Other chlorinated substances

Chlorinated aliphatic hydrocarbons

Chlorinated aliphatic hydrocarbons (CAHs) have mainly been used as solvents, for example in dry cleaning, and are still used to some extent in Sweden. They were analysed during the 2008 monitoring. The levels of dichloromethane and 1,2-dichloroethane were below the reporting limits of 2.3 and 3.5 µg/kg DW, respectively, at all stations. 1,1,1-trichloroethane (TCA), trichloromethane (chloroform), tetrachloromethane (carbon tetrachloride) and trichloroethene (TCE) could be reported at all stations while tetrachloroethene (PCE) could be reported at 14 of 16 stations (Fig. 54). Trichloromethane was also analysed in 2014 but levels were lower than in 2008 and only above the reporting limit at station SE-1 where the level was 0.20 µg/kg DW.

A comparison between sea areas shows that the highest levels, as a sum of the five substances that could be measured, occurred at stations in the Baltic Proper (Fig. 54). The levels were also well correlated with the levels of TOC in the sediment ($r^2 = 0.88$, $p < 0.0001$, $n = 16$).

Dichloromethane, 1,2-dichloroethane, trichloromethane, tetrachloromethane, TCE and PCE are listed in the WFD, some as priority substances, but there is no EQS for sediment. There is an indicative value for sediment for trichloromethane at 55 µg/kg DW (SwAM 2018b), which is not exceeded at any of the monitoring stations.

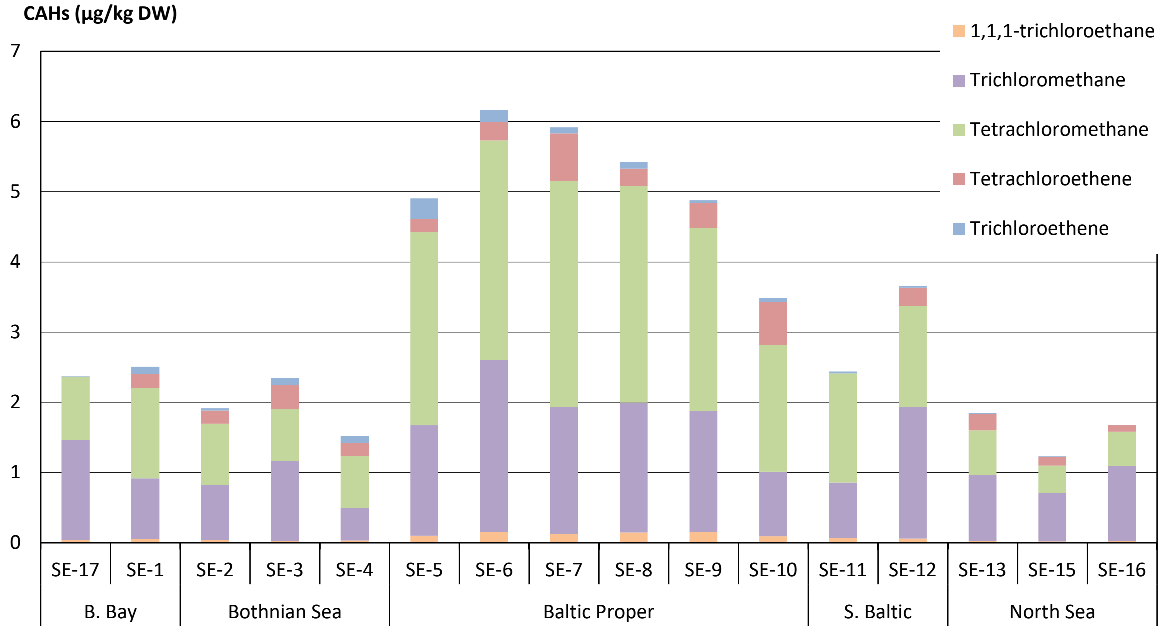

Figure 54. Levels of chlorinated aliphatic hydrocarbons (CAHs) at the 16 stations during the monitoring in 2008.

Hexachlorobutadiene

Hexachlorobutadiene (HCBD) has primarily been used as an industrial chemical, for example as a solvent in the textile industry. The substance was analysed in 2003, 2008 and 2014 but has never been present in levels above the reporting limit (0.2–1.5 µg/kg DW). HCBD is a priority hazardous substance in the WFD but lacks an EQS for sediment. There is an indicative value at 493 µg/kg DW for sediment with a TOC content of 10% (SwAM 2018b), which is never exceeded even when using the reporting limits as a worst-case scenario for sediment levels.

Phthalates

Phthalates are used as plasticizers in plastics and rubber. Some phthalates have been phased out within the EU but may still be present in imported products. In the environmental monitoring of sediments, the phthalates benzyl butyl phthalate (BBP), dibutyl phthalate (DBP), diethyl phthalate (DEP), diisobutyl phthalate (DIBP), diisodecyl phthalate (DIDP) and diisononyl phthalate (DINP) were analysed in 2014 and 2020/2021. Di(ethylhexyl)phtalate (DEHP) was analysed all four monitoring years.

Levels of phthalates are clearly lower in 2020/2021 than in 2014 (Fig. 55). In 2014, the stations in the Baltic Proper have higher phthalate levels than the stations in other areas. In 2020/2021, the stations in the North Sea and particularly SE-5 in the Baltic Proper are instead the stations with the highest levels. The reason for this is possibly sample contamination. SE-5 had been exposed to temperatures above 0°C for several weeks due to malfunctioning freezers. The higher level in this sample is therefore assumed to be due to contamination from the sample jar (glass with teflon-lined lid) or the surrounding environment. Furthermore, the sample from SE-5 was analysed in the same batch as the samples from the North Sea in 2020/2021, samples that also tended to have higher levels. Phthalate levels at SE-5 in 2020/2021 were however lower than in 2014. The differences between stations in 2020/2021 should therefore be interpreted with caution. A consistent pattern for both monitoring years can however be seen in that the levels in the Baltic Proper are higher than the levels in the Bothnian Bay and Bothnian Sea. The dominant phthalates in the samples from 2020/2021 were DBP and DIDP, followed by DEHP (Fig. 56), contrary to the pattern in 2014, when DEHP was the dominant compound.

There is a positive and significant linear correlation between the total phthalate levels and the TOC levels in the sediment ($r^2 = 0.34$, $p = 0.0005$, $n = 32$), caused by the high levels of both phthalates and TOC in the Baltic Proper. If the Baltic Proper stations are excluded, there is a weak positive but no longer significant correlation with TOC.

DEHP has been analysed all four monitoring years, and levels tend to be highest in the Baltic Proper both in 2008 and 2014 (Fig. 57). The highest levels in 2003 occurred in the Baltic Proper but are not shown in the figure due to levels being very high (up to 2800 µg/kg DW at SE-8). The sharp decrease in levels between 2003 and following years is thought to be due to a change of laboratory and method rather than an actual reduction in the environment. In 2020/2021, DEHP levels follow the overall pattern of phthalates, with the highest levels at SE-5 and at the North Sea stations.

DEHP is a priority hazardous substance according to the WFD and although there is no EQS for sediment, there is an indicative value for DEHP of 100 mg/kg DW for sediment with 5% TOC (SwAM 2018b). This value is not exceeded any year at any of the stations. The indicative value is however uncertain and levels in biota are stated to be more relevant for status assessments of the aquatic environment than levels in sediment.

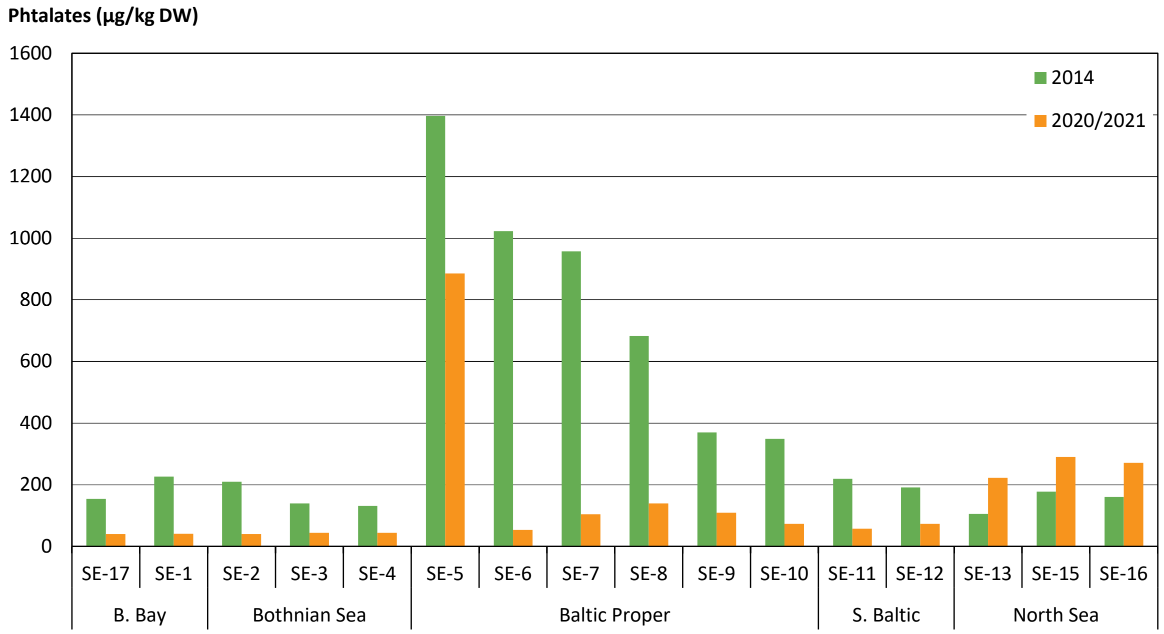

Figure 55. Levels of phthalates at the 16 monitoring stations in 2014 and 2020/2021. Note that the value for SE-5 in 2020/2021 is less certain as the sample had been exposed to temperatures above 0°C for an extended time.

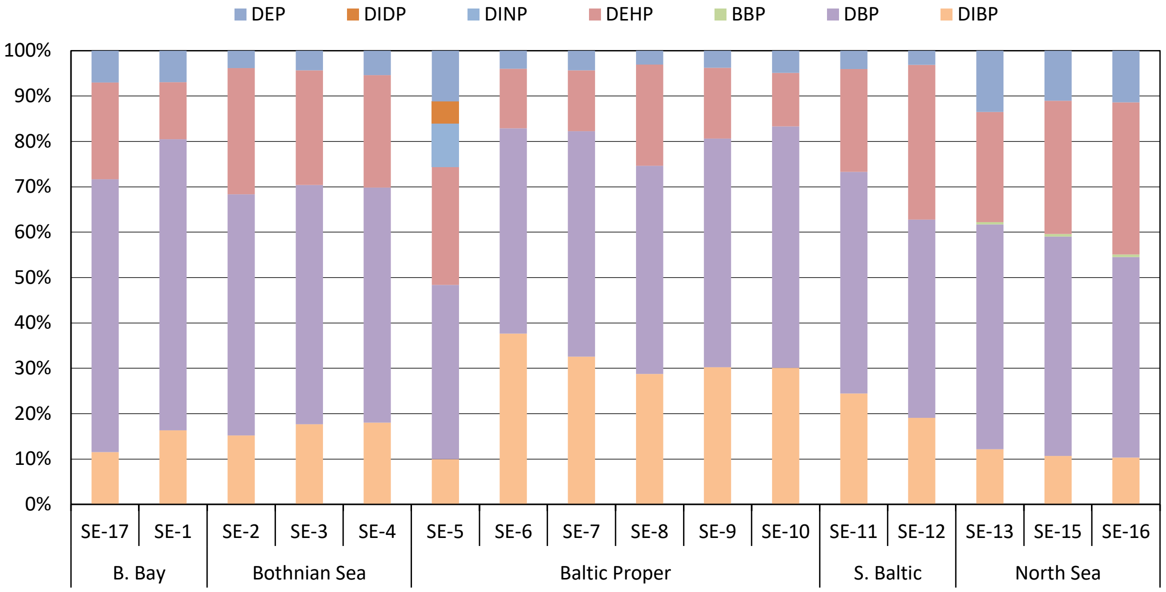

Figure 56. The distribution of the different phthalate substances at the 16 monitoring stations in 2020/2021. Note that the value for SE-5 in 2020/2021 is less certain as the sample had been exposed to temperatures above 0°C for an extended time.

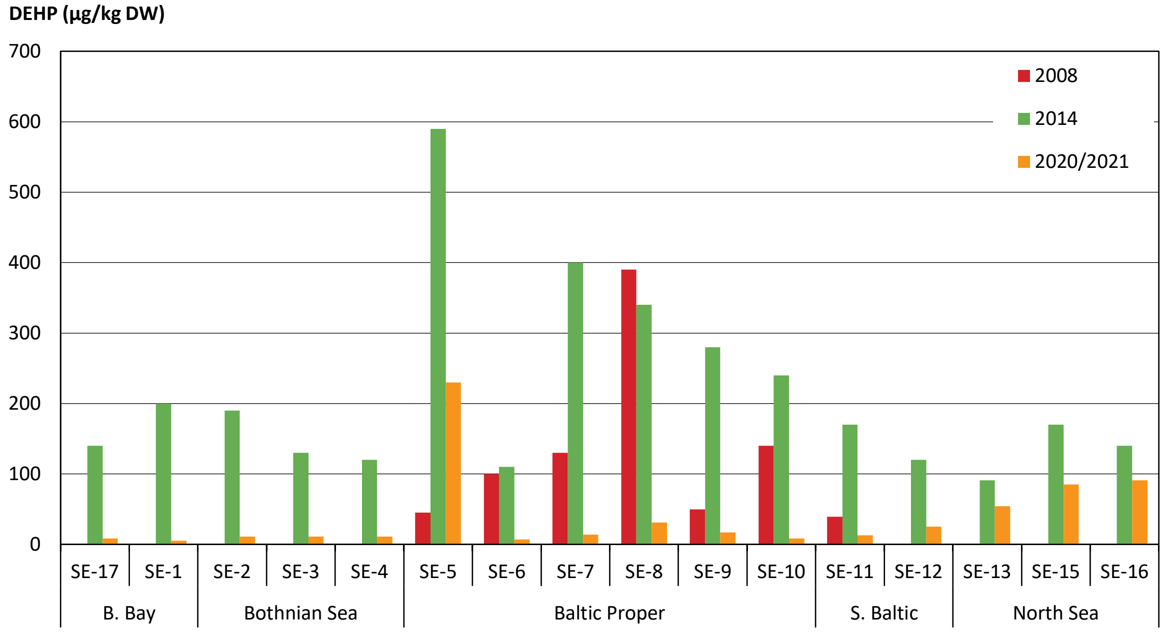

Figure 57. Levels of DEHP at the 16 monitoring stations during the three last monitoring years. DEHP was also measured in 2003 but levels were very high, ≥ 500 µg/kg DW at ten out of 16 stations. Note that the value for SE-5 in 2020/2021 is less certain as the sample had been exposed to temperatures above 0°C for an extended time.

Octyl- and nonylphenols

Octyl- and nonylphenols occur in the environment due to degradation from octyl- and nonylphenol ethoxylates used in cleaning products, but also from other sources such as tire wear and textiles (Ying *et al.* 2002, Andersson & Sörme 2006, Andersson *et al.* 2012). Industrial use, mainly in the leather industry, is considered an important source of nonylphenols and nonylphenol ethoxylates, while emissions due to their presence in products, for example tires, is an important source of emissions for octylphenol and octylphenol ethoxylates (Andersson *et al.* 2012).

The substances were analysed in 2003, 2008 and 2014, but in 2008 only 4-*tert*-octylphenol and 4-*n*-nonylphenol were analysed. The levels of these were then below the reporting limit of 4 and 10–20 µg/kg DW, respectively, at all stations. In 2003 and 2014, 4-*n*-octylphenol and 4-*iso*-nonylphenol, that is, branched nonylphenol, were also analysed. In 2003, all substances were present at levels above the reporting limits at six stations or more, while in 2014, only 4-*iso*-nonylphenol and 4-*tert*-octylphenol had levels above reporting limits.

The highest levels of all four substances occurred at stations in the Baltic Proper, both 2003 and 2014 (Fig. 58). The dominating compound was 4-*iso*-nonylphenol both years, constituting 33–100% of the sum of octyl- and nonylphenols at the different stations in 2003 and 96–100% in 2014. Only trace amounts (up to 1.3 µg/kg DW) of 4-*tert*-octylphenol could be measured in 2014. At SE-17 and SE-16, levels were so low that they were below the reporting limit even for 4-*iso*-nonylphenol in 2014 (Fig. 58).

A screening study of phenolic substances, including nonyl- and octylphenol, in water in ten Swedish watercourses showed no clear differences between different watercourses or any north-south gradient (Ahrens *et al.* 2018). Emissions to water are a clearly larger emission route in the Baltic Sea region than emissions to air, and a modelling study indicates that the levels of nonylphenol should be higher in coastal than in offshore environments due to the emissions occurring from land (not via atmospheric deposition) and that nonylphenol is degraded relatively quickly and therefore does not have time to reach offshore sediments to a large extent (Andersson *et al.* 2012).

Nonylphenol and octylphenol are priority substances according to the WFD, while nonylphenol ethoxylates are listed as RBSP. There are no EQS for sediment but there are indicative values from SwAM (2018b) of 180 and 3.4 µg/kg DW for nonylphenol and octylphenol, respectively, for sediments with 10% TOC. Both values are conversions from EQS in water and in particular the value for octylphenol is stated as uncertain. The levels of 4-*tert*-octylphenol and 4-*n*-octylphenol, and 4-*n*-nonylphenol and 4-*iso*-nonylphenol, respectively, are summed before comparison with the indicative values. In 2003, the indicative value for octylphenol is exceeded at ten of sixteen stations and for nonylphenol at two of sixteen stations. In 2014, neither of the two indicative values were exceeded; the stations closest to exceeding the indicative value are SE-4 and SE-5 for nonylphenol. Due to this, and results of the studies mentioned above, octyl- and nonylphenols were not analysed in 2020/2021.

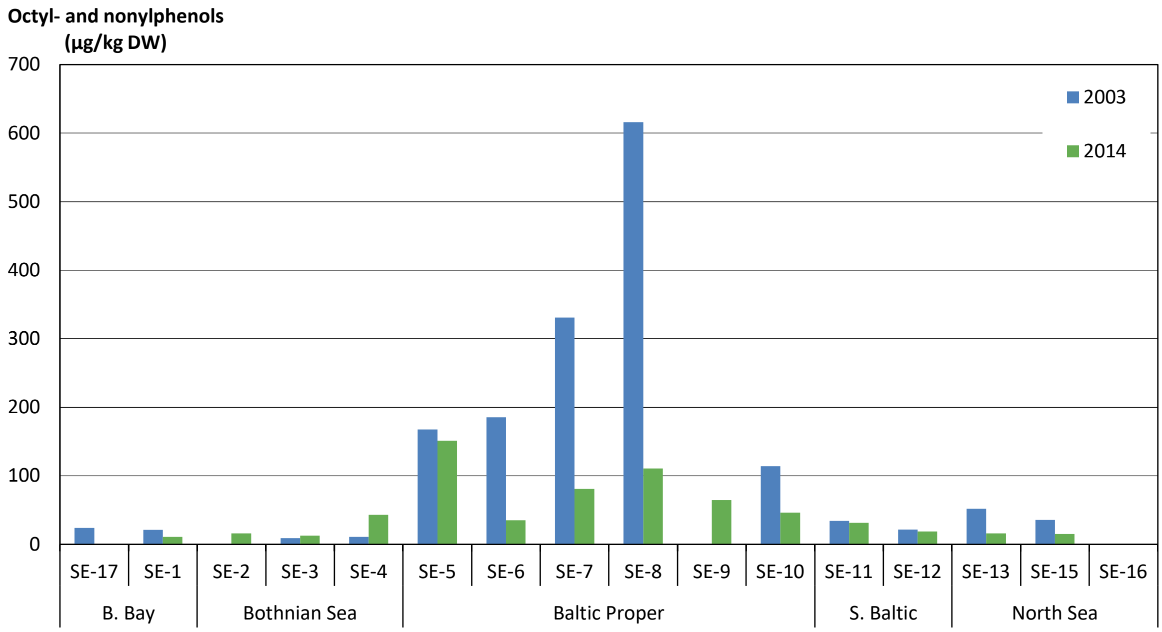

Figure 58. Levels of the sum of 4-*tert*-octylphenol, 4-*n*-octylphenol, 4-*n*-nonylphenol and 4-*iso*-nonylphenol at the 16 monitoring stations in 2003 and 2014.

BTEX and alkyllead compounds

BTEX

Benzene, toluene, ethylbenzene and *meta+para*-**xylene (BTEX)** were analysed in 2008 and the levels were above reporting limits at all stations. The highest levels of the substances occurred at stations in the Baltic Proper (Fig. 59). Levels however varied within the sea area and were higher at the three stations with higher sediment accumulation rates (SE-5, SE-6 and SE-8) than at the three stations with lower sedimentation rate (SE-7, SE-9 and SE-10). This pattern could be interpreted as showing that levels increased in the environment before 2008 leading to higher levels at the stations with a higher sedimentation rate that respond more quickly to changes in environmental levels.

BTEX levels display a positive and significant linear correlation with TOC levels in the sediment ($r^2 = 0.40$, $p = 0.0081$, $n = 16$) and there is a positive but not significant correlation even if the stations in the Baltic Proper are excluded ($r^2 = 0.40$, $p = 0.051$, $n = 10$). Benzene, but not the other BTEX substances, is a priority substance in the WFD, however there is no EQS or indicative values for sediment levels.

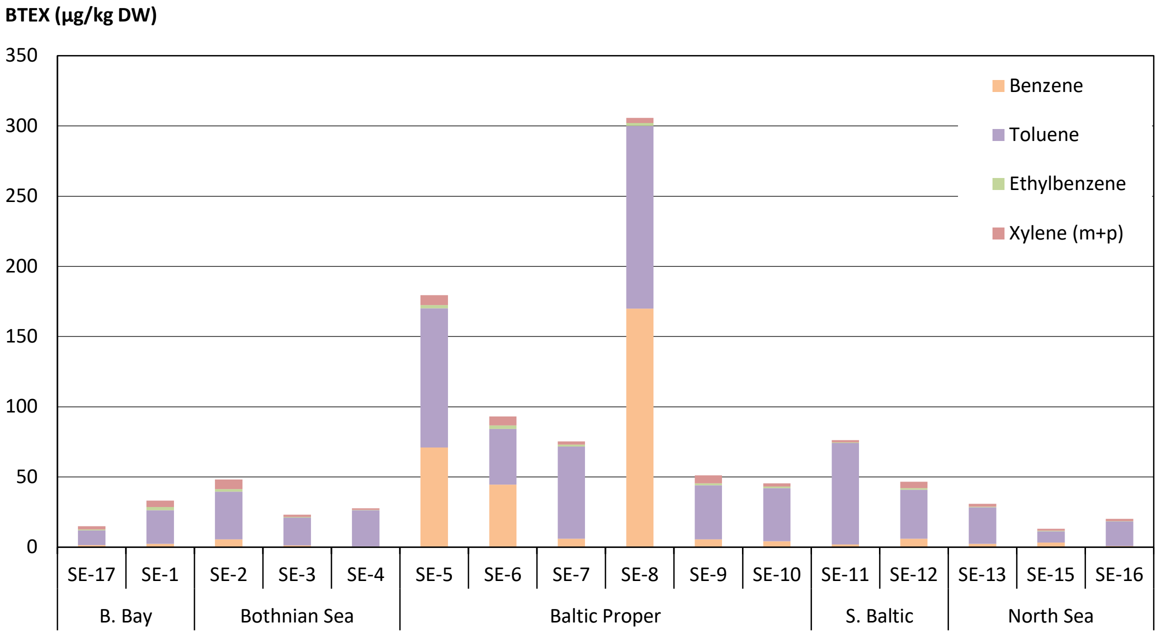

Figure 59. Levels of benzene, toluene, ethylbenzene and *meta+para*-xylene (BTEX) at the 16 monitoring stations in 2008.

Alkyllead compounds

Diethyl, **triethyl**, **tetraethyl** and **tetramethyl lead** were analysed in 2003 but levels were below the reporting limit of 10 mg/kg DW at all stations.

DISCUSSION AND CONCLUSIONS

Metals and other elements

Metals occur naturally in the environment, but levels have increased markedly since the industrial revolution. Levels in sediment have a natural geographical variation, among other things depending on the presence of different types of bedrock within the catchment, but levels also depend on local redox conditions as these strongly affect how the metals bind to the sediment. Due to these factors - anthropogenic impact, local or regional geological variation, and redox conditions combined with sediment properties - the metal contents vary between the sea areas.

The Bothnian Bay and Bothnian Sea sediments are characterized by elevated levels of arsenic and cobalt. The levels of arsenic are probably affected by high levels in the bedrock within the catchment and by one or more point sources. In the Baltic Proper, cadmium, copper and zinc are found at elevated levels. Cadmium, copper and zinc form weakly soluble sulphides under reducing conditions in the sediment, precipitation of which may be a contributing factor to the high levels in Baltic Proper sediments. The levels of these metals show significant positive correlations with TOC levels in the sediment ($r^2 = 0.79$ for Cd, 0.77 for Cu and 0.39 for Zn). This may be because the metals bind to organic material but could also be caused by the correlation between high TOC content and reducing conditions (and hence formation of sulphides) at monitoring stations in the Baltic Proper. In relation to the correlation with TOC, cadmium levels are elevated in the Bothnian Bay while copper and zinc levels are elevated at station SE-1 in the Bothnian Bay, which may be related to the proximity of a smelter point source. Chromium levels, on the other hand, are noticeably lower in the Baltic Proper than in other sea areas. This could be related to the redox conditions in sediment but needs to be investigated further.

The levels of mercury and lead seem to be affected primarily by point sources, as levels are not elevated in specific sea areas but rather at specific stations. Levels are highest at stations SE-1 in the southern Bothnian Bay and SE-12 in the Arkona Basin in the southern Baltic Sea. Lead levels are also high at station SE-11 in the Bornholm Basin. Station SE-1 is probably affected by a point source, as already mentioned, but the reason for high levels in the southern Baltic Sea is unclear. In comparison with the stations in the Baltic Sea, the levels of most metals are low in the North Sea.

In the environmental status classification within the WFD and MSFD, mercury, cadmium, and lead have been the metals that exceed thresholds the most. Mercury has no relevant threshold values for sediment but greatly exceeds its threshold values in biota, and further measures are needed to reduce levels in the environment. Since mercury can be transported long distances in the atmosphere and a large source of mercury to the Baltic Sea is atmospheric deposition, global work to reduce levels is important and is ongoing under the Minamata convention. The fact that mercury occurs at clearly elevated levels in sediment at individual stations however also points to the importance of addressing point sources to reduce levels in the environment.

Cadmium and lead have EQS for sediments which are also proposed as indicator thresholds for HOLAS III. The EQS for cadmium was exceeded at some stations in the Baltic Proper during the most recent monitoring in 2020/2021, while the EQS for lead was not exceeded. For copper, an EQS have been available in Sweden during the last years and copper is proposed to be included as an indicator for sediment in HOLAS III. Comparing the proposed threshold for HOLAS III with the measured levels in sediment in 2020/2021, the threshold is exceeded at all monitoring stations except one in the Baltic Proper.

Organic contaminants

Organic pollutants are predominantly of anthropogenic origin, although a few can also be formed in small amounts in natural processes, for example PCDD/Fs and PAHs in forest fires. Some substances can be transported long distances in the atmosphere, while other substances are mainly emitted to the Baltic Sea via the aquatic environment.

Most of the investigated organic contaminants occur at the highest levels in the Baltic Proper, including the southern parts of the Baltic Sea. This is the case for, for example, $\sum PAH_{16}$, HBCDD, organophosphorus compounds, PCBs, TBT, cybutryne, $\sum DDT$, $\sum HCH$, chlordanes, chlorinated aliphatic hydrocarbons, phthalates, octyl- and nonylphenols, and BTEX. The North Sea generally has the lowest levels of contaminants, although the levels of some substance groups, for example PAHs, are elevated here compared to the Gulf of Bothnia. The generally higher contaminant levels in the Baltic Sea are expected due to the semi-enclosed nature and limited water exchange as well as the relatively high population density within its catchment.

As many organic pollutants tend to sorb to organic carbon, it is often difficult to determine whether the high levels in the Baltic Proper, where stations also have high TOC levels, are caused by this sorption or by a high load of the various substances in this sea area. The levels of some substances, for example PCBs and the organochlorine pesticides, are elevated relative to TOC levels in the southern Baltic Sea, in particular at station SE-12. The levels in the Gulf of Bothnia, on the other hand, is generally lower than what the correlation with TOC predicts, for example for PAH and degradation products of DDT.

One contaminant group with a surprising and unexplainable geographical pattern is the PFASs. These were included in the monitoring programme in 2014 and showed very elevated levels in the Bothnian Bay and Bothnian Sea. This seemed unreasonable as levels of organic contaminants in sediment are generally lower in the Gulf of Bothnia than in the Baltic Proper, and studies of PFASs in water and biota point to lower concentrations in the northern than in the southern parts of the Baltic Sea (Nguyen *et al.* 2017, Kirchgeorg *et al.* 2010, Faxneld *et al.* 2016). It was therefore suspected that the samples had been contaminated, however, the 2020/2021 monitoring showed the same results, with on average 14 times higher PFAS levels at Gulf of Bothnia stations compared to stations in other sea areas. These observations of clearly higher levels in the Gulf of Bothnia warrant further investigations to elucidate the reason behind this.

Chlorinated paraffins also show surprisingly high levels in Gulf of Bothnia sediments. These substances have been analysed in surface sediments within the monitoring programme in 2008 and 2014, with levels above reporting limits only for SCCP in 2008. These results indicated that levels were highest in Baltic Proper sediments. It was decided to not repeat the analyses with the usual analytical methods during the 2020/2021 monitoring as reporting limits were too high. Instead, sediment cores from three stations were investigated by researchers from Stockholm University. These core studies revealed substantially higher levels at station SE-1 in the Bothnian Bay compared to SE-5 in the Baltic Proper and SE-13 in the North Sea. The reason for the high levels at SE-1 has been suggested to be emissions from e-waste facilities. It would be interesting to conduct similar analyses, either in cores or on surface sediments only, for the other monitoring stations and in sediment in the vicinity of SE-1 to see the extent of the area with elevated levels. More studies are also needed to identify sources and the reason for the difference in proportion of the different chlorinated paraffins between coastal and offshore sediments.

TBT levels have fallen since 2003 and are sometimes below the laboratory reporting limit in 2020/2021. The EQS for TBT in sediment is however still exceeded at many stations. During the monitoring in 2014 it was exceeded at all stations in the Baltic Proper and the southern Baltic Sea, but in 2020/2021, not all stations in the Baltic Proper exceed the EQS anymore. Thanks to the improved reporting limits in 2020/2021 however, it became clear that stations in other sea areas, including the Bothnian Bay and the North Sea, also have TBT levels exceeding the EQS. It is however positive to see that levels seems to be decreasing in general and that the proportion of degradation products are increasing, indicating that new inputs have decreased. Another antifouling substance, cybutryne, also seem to decrease in levels over time and now only occurs above its indicative value in sediment at one station, SE-5 in the Baltic Proper. Even though levels of organotin compounds and cybutryne appear to be decreasing, they should continue to be monitored as levels are above threshold values at one or more stations.

PAHs sometimes occur at levels that exceed thresholds. There are EQS or indicative values for sediment for several PAHs. The EQS for anthracene is exceeded at two stations in the North Sea in 2020/2021, while the EQS for fluoranthene has never been exceeded within the monitoring programme for offshore sediments. Of the four PAHs for which there are indicative values, levels in 2020/2021 exceeded the value at all stations in the North Sea, Southern Baltic and Bothnian Sea for benzo[b]fluoranthene, and at all stations in the North Sea for benzo[k]fluoranthene and benzo[a]pyrene. Indicative values were also sometimes exceeded at SE-12 and SE-9. The North Sea and the Southern Baltic are thus the most problematic areas in terms of potentially toxic PAH levels in sediments. The exceedance of threshold values even in offshore sediments illustrates that further measures will probably be needed to decrease the levels in the aquatic environment.

PBDEs are a group of substances that often greatly exceed their EQS in biota in the aquatic environment. There is no EQS for sediments, however the proposed indicator threshold for sediment for HOLAS III is not exceeded at any of the monitoring stations. The levels of PBDE in sediment are generally similar between the sea areas, which indicates that atmospheric deposition or other diffuse sources are important sources for PBDEs. Within the environmental monitoring of offshore sediments, the levels of PBDE should continue to be monitored, and problematic substitute substances should also be included.

For the 2020/2021 monitoring, analyses of organophosphorus compounds were included. These are substances that are used extensively as plasticizers and flame retardants, including as replacements for PBDEs. Like many other organic contaminants, the highest levels of the organophosphorus compounds occurred in sediment in the Baltic Proper. As there are no guidance values for sediment, it is difficult to say if these levels could cause ecological effects. The pattern of the different organophosphorus compounds differed between the Baltic Sea and the North Sea stations, which may indicate different sources. It would be interesting to also include this compound group during the next monitoring year to see if the geographical patterns are consistent and to see how levels change over time.

PCDD/Fs are, like PBDEs, substances that are problematic due to high levels in biota. As for PBDEs, the PCDD/F levels in sediment are not clearly elevated in different sea areas although station SE-12 in the Southern Baltic tend to have high levels of this and many other organic contaminants and metals. The levels of PCDD/Fs are higher in 2020/2021 than in 2014 but is seems unlikely that environmental levels would have increased to this extent, considering that many other studies point to decreasing levels. Due to this, and to PCDD/Fs being highly problematic compounds when it comes to levels in biota, this contaminants group should continue to be analysed in the monitoring programme.

To conclude, some of the investigated compounds exceed their environmental threshold values for sediment. There is however a general lack of threshold values for sediment, even for substances who tend to partition to sediment and for which it would therefore be beneficial to have sediment guidance values. Within the upcoming HOLAS III to evaluate the environmental status of the Baltic Sea, there are proposed indicator thresholds for sediment for the organic contaminants HBCDD, PBDE, anthracene, fluoranthene and TBT. In the monitoring in 2020/2021, these indicator thresholds were exceeded for anthracene and TBT at some stations, but not for HBCDD, PBDE or fluoranthene.

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