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Source tracking of per- and polyfluoroalkyl substances (PFAS) from perch in Kvädöfjärden

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Summary
Elevated PFAS concentrations have consistently been found in perch from the Kvädöfjärden monitoring station (Original station) since monitoring of this chemical group started in 2016. In this project we 1) investigated the temporal variability of PFAS in Kvädöfjärden by retrospective analysis of perch from 1981 until 2015, 2) investigated the spatial variability of PFAS concentrations in perch from ten locations collected in the Kvädöfjärden region in 2023 and 3) mapped various harmful activities in the surrounding area of Kvädöfjärden.

The results show that PFOS and several PFCAs has continuously increased from the 1980s. We further see a change in the PFAS homologue fingerprint between 2005 and 2010 that suggests a change from a more marine influence to the influence of a local source.

All perch from the spatial survey have a fractional pattern of the PFAS homologues similar to the Original station, but the PFAS concentrations show high spatial variability. The Original station has the highest PFAS concentrations but elevated concentrations were also seen in perch in Licknevarpfjärden, north of the Original station. The ratio of PFUnDA/PFDA suggest that firefighting foam is not the source of the increasing PFAS concentrations. Moreover, we could not identify any harmful activities in the Kvädöfjärden region that potentially could affect the PFAS concentrations. We are therefore not able to identify the exact source of the elevated PFAS concentrations.

To track a possible source area, we suggest perch samples are collected between the Original station and Licknevarpfjärden. In addition, we suggest that water samples could be collected in the region as a supplement.

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Summary

Kvädöfjärden, situated in the southern Baltic Proper, is included in the National Monitoring Programme for Contaminants in Marine Biota. The area is regarded as pristine, with no known pollution sources and acts as a reference location. However, high PFAS (especially PFOS) concentrations have consistently been found in perch from Kvädöfjärden since monitoring of this group of chemicals started in 2016.

The aim with this study was to 1) investigate the temporal variability of PFAS in Kvädöfjärden by retrospective analysis of perch to explore the onset of a change in the concentration pattern 2) investigate the spatial variability of PFAS concentrations in the Kvädöfjärden region to explore the potential location of a PFAS source and 3) map harmful activities in the surrounding area by using the county boards database over hazardous substances (EBH-stödet) in order to investigate if any of these might be the cause of the elevated PFAS concentrations.

In 2023 perch was collected at ten locations geographically distributed within the Kvädöfjärden region. In addition, perch from the station included in the Monitoring Programme (Original station), which were stored in the Environmental Specimen Bank at the Swedish Museum of Natural History, were used for a retrospective analysis of PFAS, prolonging the short time series between 2016-2022 to cover 1981-2022. 15 target PFAS homologues were analysed in all samples (liver tissue).

The examination of the temporal variability in perch from the Original station in Kvädöfjärden shows that the concentrations of PFOS and several PFCAs, has continuously increased since the 1980s. Using temporal trends and PCA analyses, we find a change in the fingerprint of PFAS homologues between 2005 and 2010. The analyses suggest a change from a more marine influence to the influence of a local source that also drives the increasing concentrations.

All perch in the Kvädöfjärden region from the spatial survey have fractional patterns of PFAS homologues similar to that of the Original station. However, concentrations of PFAS in perch show high spatial variability even on a small scale. The ratio between PFUnDA/PFDA suggest that firefighting foam (AFFF) are not the source of the increasing PFAS concentrations but the ratio does not allow us to say more about the possible source. There is only one water measurement within the Kvädöfjärden region, which does not suggest highly elevated PFOS concentrations compared to recent offshore measurements in the area. The Original station has the highest perch PFAS concentrations in the region but also concentrations in perch in Licknevarpfjärden are elevated (although not as high), which could indicate that the source(s) should be found in the area north of the Original station and south of Licknevarpfjärden where we have no samples from.

Moreover, we could not identify any harmful activities in the Kvädöfjärden region that potentially could affect the PFAS concentrations.

To track a possible source area, we suggest perch samples are collected between the Original station and Licknevarpfjärden. In addition, we suggest that water samples could be collected in the region as a supplement.

Introduction

Per- and polyfluoralkyl substances (PFAS) are a group of manmade, highly fluorinated chemicals containing compounds that are persistent, bioaccumulative and toxic [*Giesy and Kannan*, 2001; *Lau et al.*, 2007] and are found everywhere in the environment [*Cousins et al.*, 2022; *Haukås et al.*, 2007; *Houde et al.*, 2011]. PFAS has been used in e.g. stain and water repellent coatings in textiles, in aqueous film forming foam (AFFF), food packages, and pesticides. Today, there are more than 4700 substances of PFAS, some of which have been produced since the 1950s [*OECD*, 2018]. Perfluorooctanoic sulfonic acid (PFOS) is the PFAS found in the highest concentrations in the environment. The main producer of PFAS, 3M, started to voluntarily phase out their PFAS production in 2000-2002 and in 2009 PFOS was included in the Stockholm convention [*SC*, 2024].

The Swedish National Monitoring Programmes for Contaminants in Marine and Freshwater Biota focus on establishing the concentrations of a suite of contaminant groups at background/reference sites throughout Swedish marine and freshwater environments. Perch is the cornerstone of the freshwater programme but two coastal marine sites are also included in the marine programme as a way of comparing the contaminant concentrations between the freshwater and marine environment in the same species.

Kvädöfjärden is situated in Östergötland County in the Baltic Proper. Perch for contaminant monitoring has been collected in the area since 1981 [*Svahn et al.*, 2023]. While some contaminant groups have been analyzed in perch from Kvädöfjärden since 1981 within the Monitoring Programme for Contaminants in Marine Biota (hereafter referred to as the marine programme), PFAS was analysed for the first time in 2016. During evaluations of the marine and freshwater programmes in 2022-2023 it was discovered that PFOS concentrations in perch from this station were elevated compared to perch from other stations in the monitoring programme [*Faxneld and Soerensen*, 2023b; *Soerensen and Faxneld*, 2023]. The concentrations at Kvädöfjärden were in the same range as those in perch from Lake Fysingen, which has been contaminated by AFFF from fire-training sites at Arlanda airport [*Ahrens et al.*, 2015] and in some years the concentrations have been close to the PFOS EQS in biota of 9.1 ng/g muscle ww. Kvädöfjärden is regarded as pristine, it is included in the Natura 2000 network [*Svahn et al.*, 2023] and there are no known human activities in the area. The elevated PFAS concentrations therefore suggest an unknown source in the area.

The aim with this study was to 1) investigate the temporal variability of PFAS in Kvädöfjärden by retrospective analysis of perch to explore the onset of the elevated concentrations 2) investigate the spatial variability of PFAS concentrations in Kvädöfjärden to explore the potential location of a PFAS source and 3) map harmful activities in the surrounding area to investigate if any of these might be the cause of the elevated PFAS concentrations.

Material and methods

Collection of fish for spatial analysis

During the summer 2023, perch were collected at ten stations geographically distributed in the greater Kvädöfjärden region (including Kvädöfjärden, Trollholmsfjärden, and Licknevarpfjärden) by the Swedish University of Agricultural Sciences (SLU), during their ordinary fish population monitoring (Table 1, Figure 1). At each station, 10 fish were collected, placed individually in polyethylene bags and then put on ice. The fish were transported frozen to the Swedish Museum of Natural History (SMNH). In addition, the "Original" station, which is the monitoring station used for collection of perch within the ordinary monitoring program, were sampled as usual (data 2016-2022; Table 1; Figure 1).

Retrospective material from the Environmental Specimen Bank

Perch from the Original station, stored in the Environmental Specimen Bank (ESB) at SMNH, was used for a retrospective analysis of PFAS from this area. Material from the years 1981, 1990, 2000, 2005, 2010 and 2015 were used (the coordinates used within the marine programme; Table 1, Figure 9). This was added to the 2016-2022 PFAS monitoring data from the Original station.

Sample preparation

Before sample preparation, fish length, weight, and sex were recorded. Age was determined using otoliths. The preparation was performed according to *Stjärnkvist et al.* [2024]. A homogenate sample from each station or time period was prepared with 10 fish in each homogenate. The homogenates consisted of a mix of females and males.

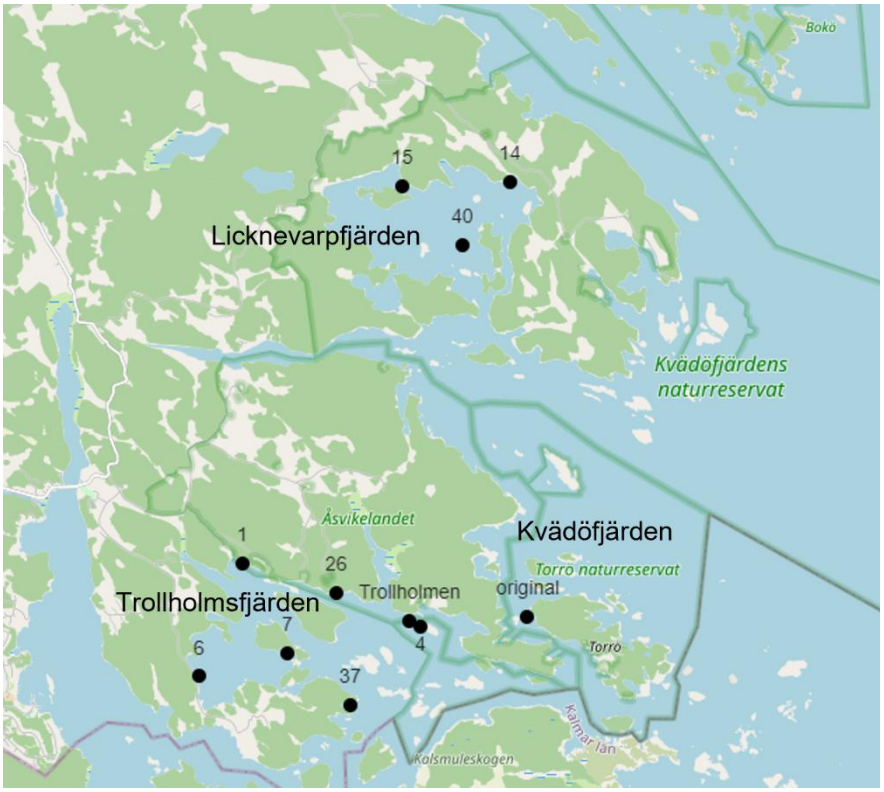


Figure 1. Sampling stations for the spatial analysis in the greater Kvädöfjärden region (here defined as Kvädöfjärden, Trollholmsfjärden, and Licknevarpfjärden). The Original station is the station used for collection of perch within the ordinary marine monitoring program.

Table 1. Station names and coordinates for collection of perch in the greater Kvädöfjärden region in 2023. "Original" is the ordinary monitoring station in Kvädöfjärden. We divide the stations into three sub-areas based on the water exchange in the area (Figure 2). Station 6 and station 1 are located in Häxvassen and Saffjärden respectively but for simplicity they will throughout the report be grouped in the sub-area Trollholmsfjärden.

Area	Sub-area	Station name	N	E
Kvädöfjärden	Kvädöfjärden	Original	58° 00,800	16° 46,573
Kvädöfjärden	Trollholmsfjärden	Trollholmen	58°00,775	16° 44,834
Kvädöfjärden	Trollholmsfjärden	Station 26	58° 00,990	16° 43,760
Kvädöfjärden	Trollholmsfjärden	Station 4	58° 00,730	16° 45,000
Kvädöfjärden	Trollholmsfjärden	Station 37	58° 00,120	16° 43,980
Kvädöfjärden	Trollholmsfjärden	Station 7	58° 00,520	16° 43,040
Kvädöfjärden	Trollholmsfjärden	Station 6	58°00,340	16° 41,750
Kvädöfjärden	Trollholmsfjärden	Station 1	58°01,220	16° 42,390
Kvädöfjärden	Licknevarpfjärden	Station 14	58° 04,180	16° 46,330
Kvädöfjärden	Licknevarpfjärden	Station 15	58° 04,150	16° 44,740
Kvädöfjärden	Licknevarpfjärden	Station 40	58° 03,690	16° 45,610

Analytical method

Samples from 2016-2019 were analysed at the Department of Environmental Science, Stockholm University (SU). Since 2020 all PFAS analysis are conducted by the Department of Aquatic Sciences and Assessment at SLU and the retrospective analysis in this study, i.e. from 1981-2015 have been analysed at SLU. In brief, the analyses involved a sample aliquot of homogenized tissue spiked with mass-labelled internal standards, solvent extraction, and instrumental analysis by ultraperformance liquid chromatography-tandem mass spectrometry (UPLC-MS/MS). The analytical methods are described in more detail in *Ammar et al.* [2024b]. Abbreviations used are according to *Buck et al.* [2011]. The target analytes in this study were PFBS, PFHxS, PFOS, PFDS, FOSA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTrDA, PFTeDA, and PFPeDA. An intercalibration study between the two labs were done before the lab switch, where it was shown that the limit of detection (LOQ) differed for some of the homologues, and it was almost always higher at the SLU lab [*Faxneld et al.*, 2022].

Data sources

The PFAS data generated in this study, is available at the national data host, the Geological Survey of Sweden (SGU). In addition to this data and the PFAS monitoring data from the Original station (2016-2022), other data has also been used in order to get a better understanding of concentrations and patterns of homologues (see Table 2 for data and references).

Table 2. Data used in this project. The table shows the type of data, time period, species and data source. SI = stable isotopes.

Data	Time period	Area	Species	Source
PFAS	2013-2014	Kvädöfjärden (Kaggebofjärden and EROD station)	Perch	Unpubl data NRM (Formas project: ChemToFish)
PFAS	2016	Valdemarsviken	Perch	SGU (regional screening monitoring) https://www.sgu.se/produkter-och-tjanster/nationella-datavardskap/datavardskap-for-miljogifter/rapporterad-data-till-datavardskap-for-miljogifter/
PFAS	2016-2022	Holmöarna	Perch	NRM marine monitoring data
PFAS	2019-2021	Lake Skärgölen, Lake Fysingen	Perch	NRM freshwater monitoring data
PFAS	2016-2022	Kvädöfjärden	Eelpout	NRM marine monitoring data

SI	1998-2015	Kvädöfjärden	Perch	Unpubl data NRM (Formas project: ChemToFish)
SI	1988-2015	Kvädöfjärden	Blue mussels	[*Karlson and Faxneld*, 2021]
SI	2016-2022	Kvädöfjärden	Blue mussels	NRM marine monitoring data [*Ammar et al.*, 2024a]
PFAS	2023	Kvädöfjärden and surrounding area	Water	Data provided by Felicia Stagnefeldt, County board of Östergötland
Hazardous activities				EBH database https://www.lansstyrelsen.se/stockholm/miljo-och-vatten/fororenade-omraden/kartor-over-fororenade-omraden.html

Statistical treatment

LOQ

For evaluating time trends of PFAS, concentrations below limit of quantification (LOQ) were substituted with LOQ/√2. For comparing fractions of PFAS, values below LOQ were replaced with zero. Homologues where more than 50% were below LOQ were removed from all further analyses.

Time trend analysis

To investigate time trend analysis, log linear regressions (for various PFAS) and linear regressions (for biological data) covering the whole time series were done. The slope of the log-linear regression represents the yearly average percent increase or decrease of the time series.

Spatial analysis

For the spatial analysis of PFAS, all stations in the greater Kvädöfjärden region were from 2023 except for the following: the Original monitoring station in Kvädöfjärden (a mean for 2020-2022 was used), and Kaggebofjärden and EROD station (a mean for 2013-2014 was used). In addition, for some of the other stations used in the spatial analysis, means were used as well; the monitoring station in Holmöarna (a mean for 2020-2022) and for the two freshwater monitoring stations, Lake Fysingen and Lake Skärgölen (a mean for 2019-2021 was used). Moreover, for eelpout data from the monitoring stations in Kvädöfjärden and Fladen a mean for 2020-2022 was used.

Trophic position

Stabile isotopes have been analysed retrospectively in perch at the Original station in 1998-2015 (Table 2). These perch are not the same individuals as those that went for PFAS analysis but they are from the same area and the same size of perch.

In order to calculate the trophic position (TP) in perch and eelpout from Kvädöfjärden, the equation proposed by *Post* [2002] with a diet-consumer discrimination ($\Delta^{15}N_{Bulk}$) factor of 3.4 ‰ (Equation 1) was used. As primary consumer, blue mussels were used and they were assigned a TP of 2.

$$TP_{Bulk} = (\delta^{15}N_{Perch} - \delta^{15}N_{base})/\Delta^{15}N_{Bulk} + TP_{base} \quad \text{Equation 1}$$

PCA analysis

Principal Component Analysis (PCA) was applied to PFAS profiles (i.e. concentration data converted to a percentage of ∑PFAS) to study geographical and temporal differences in PFAS patterns.

Harmful activities

A spatial analysis of hazardous activities in the area around Kvädöfjärden were done using data from the EBH database (the County boards database over hazardous activities [EBH-stödet], Table 2). Activities that can cause elevated PFAS concentrations were chosen and plotted on a map in order to investigate if any of these were likely to affect the area of the Original monitoring station.

Water circulation in the greater Kvädöfjärden region

Figure 2 and Table 3 shows how the water flows between the different parts of the greater Kvädöfjärden region [*Länsstyrelsen*, 2024]. As seen in Table 3 the largest water exchange for the greater Kvädöfjärden region is with the Outer coastal waters and a hydrodynamic model by *Bryhn et al.* [2017] supports that it is mainly the Outer coastal water that influences the greater Kvädöfjärden region. This indicates that a source influencing the stations in the greater Kvädöfjärden region is either a point source influencing the area directly or is from Outer coastal water rather than coming from one of the nearby sub-areas.

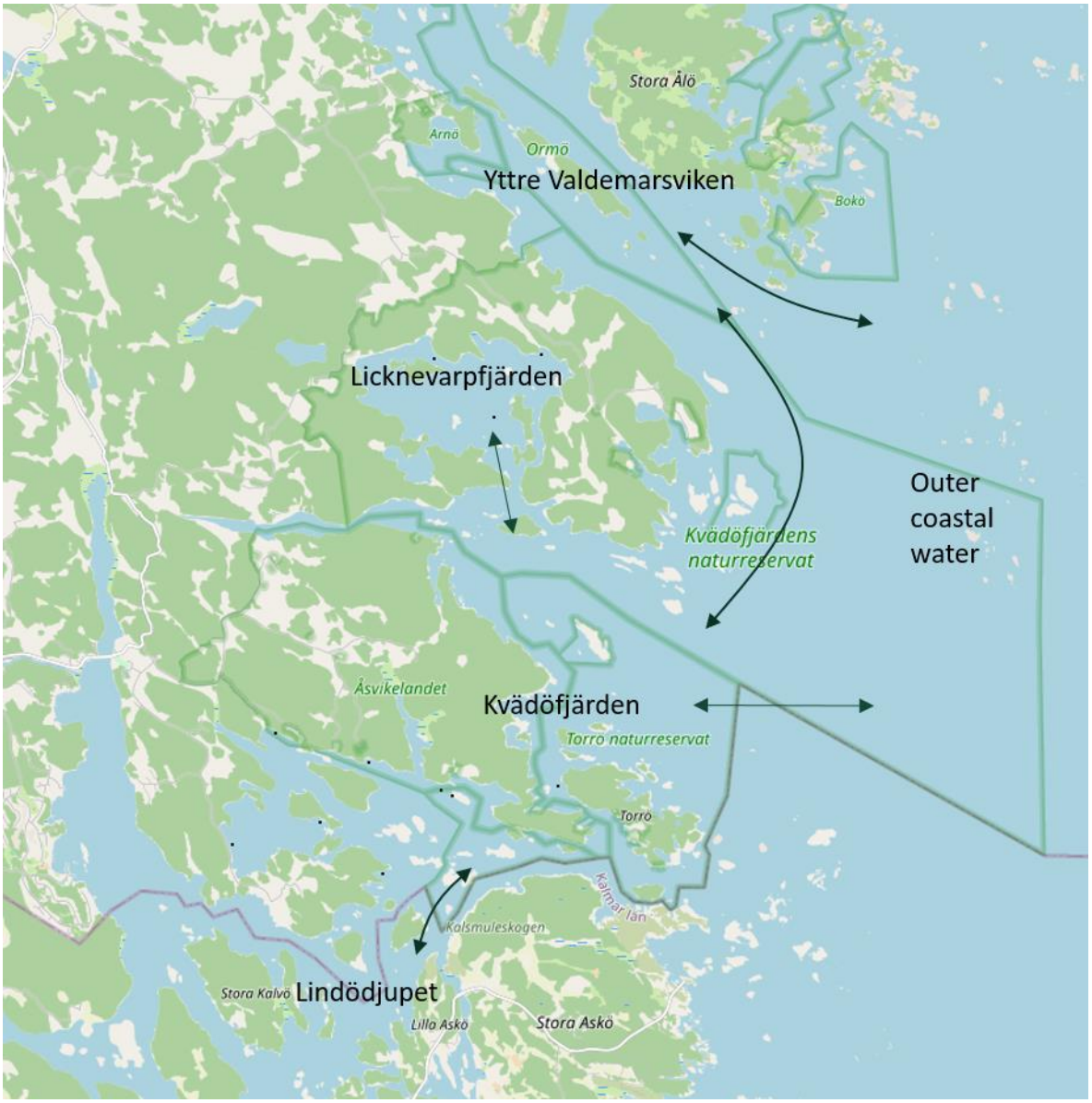


Figure 2. The water exchange in the Kvädöfjärden region between different sea areas. The arrows show how the water flows between the sea areas and outer areas. All samples in the 2023 spatial survey are from the Kvädöfjärden and Licknevarp sub-areas.

Table 3. The water flows (m^3/s) in the straits between the different sub-areas in the Kvädöfjärden region. Data taken from *Länsstyrelsen* [2024].

Strait	Flow in (m^3/s)	Flow out (m^3/s)
Kvädöfjärden – Yttre Valdemarsviken	152	151
Kvädöfjärden – Licknevarpfjärden	20.4	20.4
Kvädöfjärden – Outer coastal waters	1558	1562
Kvädöfjärden – Lindödjupet	46	49

Results and discussion

Temporal trends

Biological variables and stable isotopes

Table 4 shows the mean age, length and weight of perch at the Original station during the different time periods. Over the years, perch has been between 3 and 4 years old, with an exception in 1990 where the mean age was 5.9 years. The mean length and weight of perch has been 16.1-21.1 cm and 47.4-123 g respectively. Figure 3 and Table 5 shows the linear trends in age, length and weight of perch over time, from 1981 to 2022. Perch length has increased with 0.4% per year and weight with 1.5% per year while perch age has decreased with 0.6% per year. The perch has thus gotten larger over the period for the same age, indicating a faster growth.

Table 4. Mean age, total length (cm) and total weight (g) and the standard deviation (sd) in perch at the Original monitoring station from 1981-2022, n=10 fish at each time period between 1981-2015, n=20 fish from each time period between 2016-2022.

year	Age	sd	Total length (cm)	sd	Total weight (g)	sd
1981	3.2	0.42	16.1	1.37	47.5	12.3
1990	5.9	0.33	17.1	0.70	59.6	7.00
2000	3.8	0.63	17.5	0.99	78.8	10.0
2005	3.0	0.00	16.7	0.54	61.8	4.61
2010	3.8	0.79	17.7	1.16	76.2	16.1
2015	3.3	0.48	18.0	0.91	74.7	12.4
2016	3.1	0.22	18.7	1.12	82.6	12.5
2017	4.0	0.32	21.1	0.63	123	14.5
2018	3.1	0.22	19.1	1.24	89.2	19.2
2019	2.2	0.41	18.6	0.98	85.4	14.0
2020	3.1	0.31	18.0	0.76	75.3	13.1
2021	4.2	0.52	19.3	0.58	91.8	7.53
2022	3.6	1.15	18.8	1.40	92.3	22.2

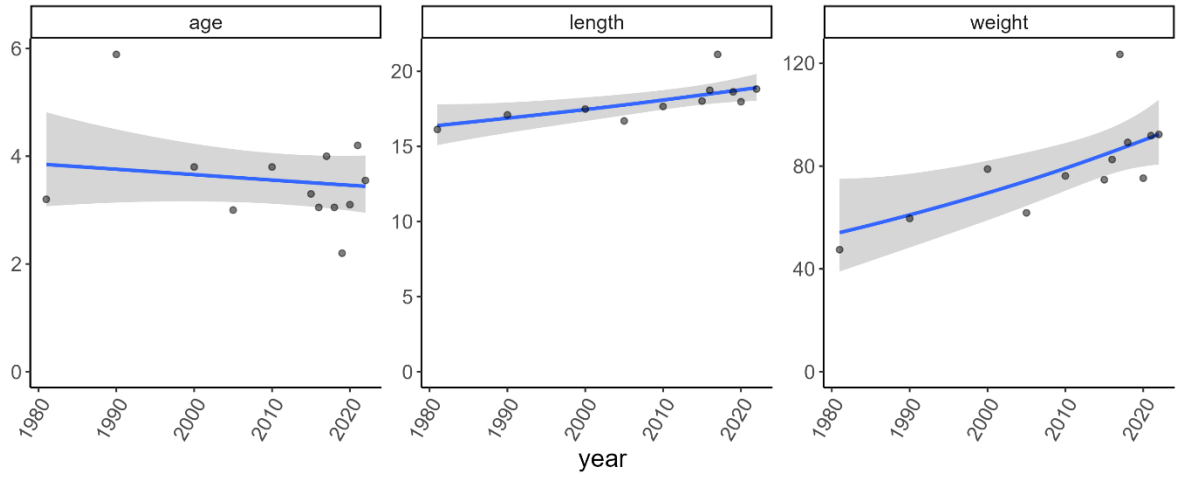


Figure 3. Linear regression for age, length (cm), and weight (g) in perch at the Original station between 1981-2022.

Table 5. Time trends for age, length and weight in perch at the Original station, 1981-2022.

Biological data	trend	conf_lower	conf_upper	p value
age	-0.006	-0.009	-0.003	<**0.001**
length	0.004	0.003	0.005	<**0.001**
weight	0.015	0.012	0.018	<**0.001**

Figure 4 shows that the trophic position (TP) of perch from the Original station has increased with 3% (CI 1.9:3.9) from 1998 to 2021 ($p<0.001$). The TP has increased from approximately 2.6 during the end of the 1990s to 3.1-3.4 in the middle of 2010s, indicating that perch has changed its diet over time. This trend in TP follows the trends in length and weight of the fishes. Perch within the monitoring programme are collected in a certain length span (16-19 cm). This is the length where perch goes through an ontogenetic diet shift as it grows, with a gradual transfer from feeding mainly on smaller invertebrates to feeding more on fish [*Estlander et al.*, 2010]. The increase in TP observed could be a result of the perch eating more small fish today than earlier when they were slightly smaller in size.

Even though PFAS biomagnifies in the food web [*Haukås et al.*, 2007; *Miranda et al.*, 2023] and perch collected within the monitoring programmes is slightly larger, and therefore likely to eat more fish now compared to earlier, this is unlikely to explain the temporal increase in PFAS concentrations at the Original station. Firstly, we do not observe an increase in concentrations of other bioaccumulating contaminants in Kvädöfjärden [*Soerensen et al.*, 2024b]. Secondly, *Waldetoft et al.* [2024] did not find any bioaccumulation of PFAS in perch with increased weight in a coastal area 200 km north of Kvädöfjärden. However, we cannot completely rule out an impact on the PFAS concentration from the change in trophic position.

In addition, $\Delta^{13}C$ has decreased with 13% (CI -19:8) (i.e. become more negative) from 1998-2021 ($p<0.001$) (Figure 5). The $\Delta^{13}C$ is often used to identify food sources, i.e. benthic or pelagic [*France*, 1995; *Post*, 2002]. The decrease in $\Delta^{13}C$ in Kvädöfjärden, going from approximately -15 to -18, does not indicate any drastic shift in food sources – marine benthic food source range from around -12 to -20 [*France*, 1995]. The $\Delta^{13}C$ in perch from Kvädöfjärden during the last years are in the same range as both that of eelpout from Kvädöfjärden and perch from Holmöarna, but it is less negative compared to herring from any of the basins in the Baltic Sea [*Soerensen and Faxneld*, 2020].

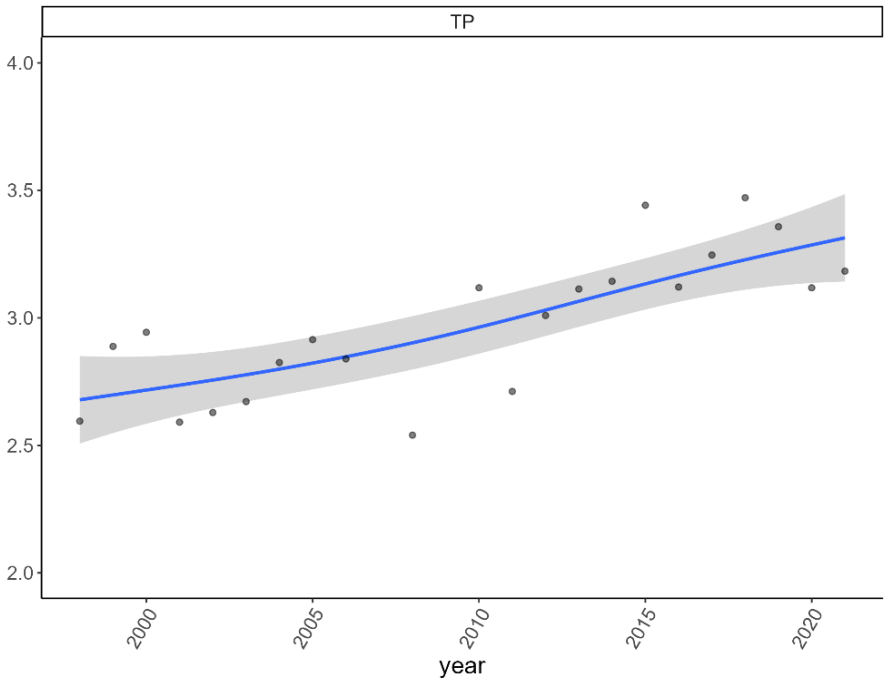


Figure 4. Trophic position (TP) in perch from the Original station, 1998-2021

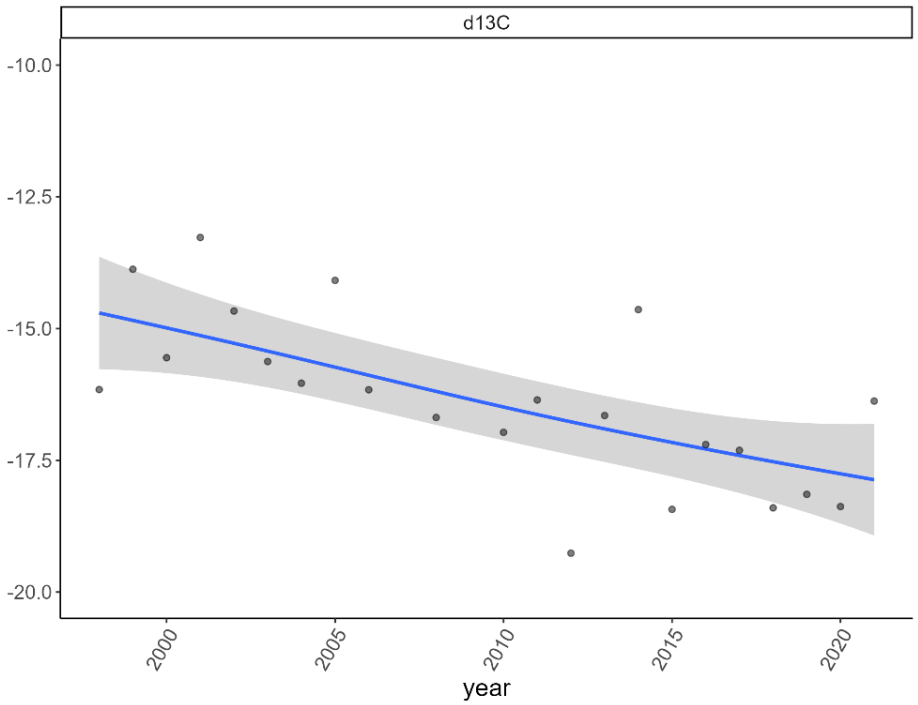


Figure 5. $\Delta^{13}C$ in perch from the Original station, 1998-2021.

PFAS time series

Figure 6 shows temporal trends from 1981 to 2022 of selected PFAS at the Original station. The highest concentrations of PFOS and the three carboxylic acids (PFNA, PFDA and PFUnDA) are seen at the end of the time series, while FOSA peaked in 2000.

Concentration of PFOS and PFCAs show upward trends (4-13% increase per year) since 1981 ($p<0.05$, Table 6), while FOSA shows a downward trend (6%). The pre 2000 upward trends at the Original station are similar to other biota time series from the Baltic Sea, however, increasing trends of PFOS and several PFCAs up to present day are not. Both for marine and freshwater fish as well as eggs from guillemot and white tailed sea eagle in Sweden concentrations generally show stagnant or decreasing concentrations since 2000-2010 [*Faxneld and Soerensen*, 2023a; *Haque et al.*, 2023; *Soerensen et al.*, 2024a; *Soerensen et al.*, 2024b]. The only compounds that show a similar trend to that of other Swedish studies are the FOSA decrease from the end of the 1990s and the PFNA increase, which is seen for some herring stations [*Soerensen et al.*, 2024a]. The only other time series that show a similar pattern as in the Original station for multiple homologues, i.e. increase in PFOS and PFCAs concentrations and decrease in FOSA, is cod from south of Gotland [*Soerensen et al.*, 2024a]. We have previously suggested that the deteriorating condition of the cod could explain this trend [*Casini et al.*, 2016; *Soerensen et al.*, 2024a] and suggest the similarities between the two time series have different drivers. However, the decrease in FOSA seen in perch in the Original station indicates that the influence from this (and likely other) pre-cursors are decreasing, suggesting that the continued increase in PFOS and PFCAs are from another source than in the pre-2000 years.

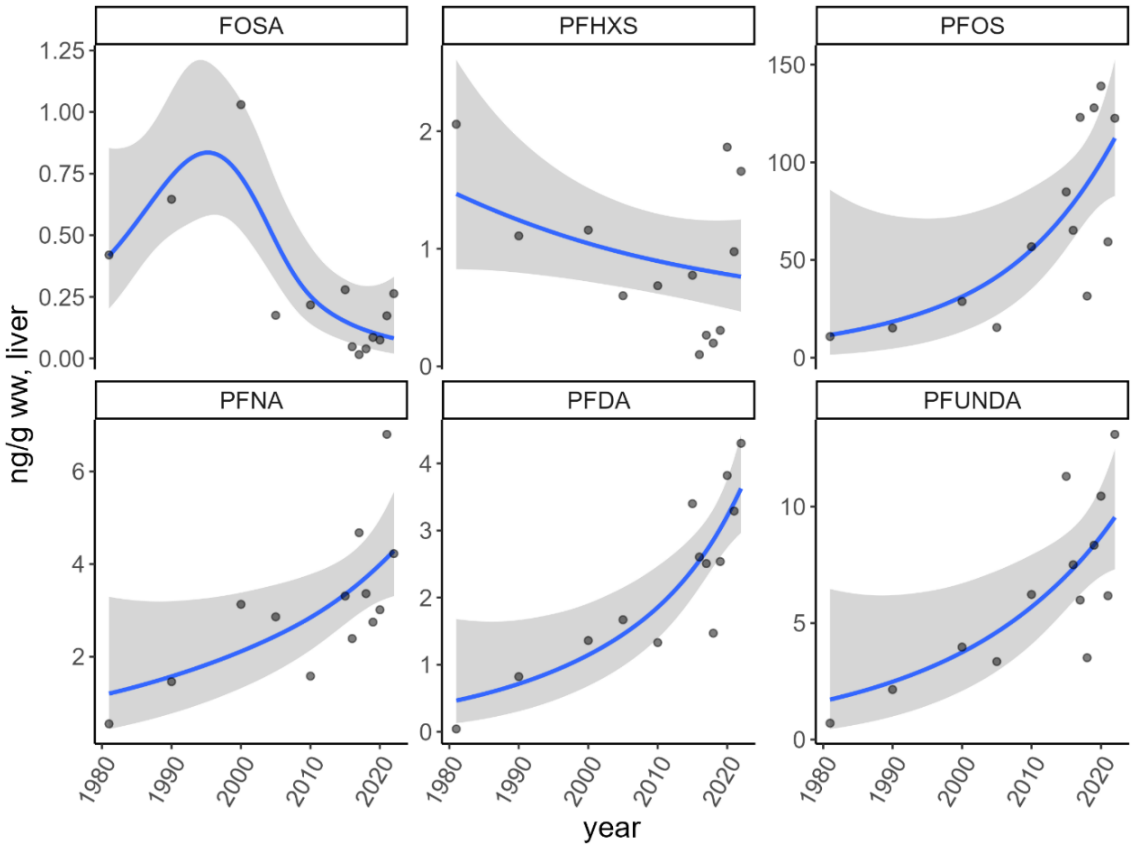


Figure 6. Log linear regressions for various PFAS in perch from the Original station 1981-2022. The grey area indicates the 95% confidence interval.

Table 6. Log linear regressions for various homologues in perch from the Original station, 1981-2022.

Homologues	Trend	conf_lower	conf_upper	p value
FOSA	-0.056	-0.095	-0.015	**0.011**
PFHXS	-0.024	-0.066	0.019	0.251
PFOS	0.062	0.039	0.085	**0.000**
PFNA	0.040	0.022	0.058	**0.000**
PFDA	0.082	0.058	0.107	**0.000**
PFUNDA	0.055	0.037	0.073	**0.000**
PFDODA	0.040	0.021	0.059	**0.000**
PFTRDA	0.128	0.062	0.198	**0.001**
PFTEDA	0.065	0.031	0.100	**0.001**
PFPEDA	0.044	-0.021	0.113	0.177

Figure 7 and Table 7 shows fractions of PFAS homologues in perch from the Original station over time. In 1981 only five homologues were detected, FOSA, PFHxS, PFOS, PFNA and PFDoDA.

PFHxS constituted 15% of total PFAS in 1981 but that fraction decreased over time and during the 2020s only about 1% was PFHxS. PFOS constituted 77% of total PFAS in 1981. This fraction decreased over time to 62% in 2005 but from 2010 and onwards it has been between 75-88%.

We compare the temporal evolution of the PFOS to PFAS fraction of perch in the Original station to that of the two closest long term marine time series for herring. In the northern Baltic Proper (Landsort, station north of Kvädöfjärden) the fraction of PFOS was stable, around 60-70% between 1980 and 2019 and in the southern Baltic Proper (Utlängan, station south of Kvädöfjärden) it decreased from 80% in the 1980s to 58% in the 2000s where it stabilized [*Soerensen et al.*, 2024a]. Similar to herring, data on perch from other areas of the Baltic Sea also suggest a continuous decrease of the PFOS fraction over time. For example, *Berger et al.* [2009] found that perch collected in the northern Baltic Proper (Öregrundsgrepen) in 2001 had a PFOS fraction of 70% while the PFOS fraction at Holmöarna was 70% in the 2010s (Table S 1) and *Junttila et al.* [2019] found that perch collected on the Finnish coastal side of the Baltic Sea in 2014-2016 had a PFOS fraction of 50%. Thus, we expect the PFOS fraction in the Baltic Sea biota to decrease over time due to the phase out and ban of PFOS both at offshore and coastal sites. The increase of the PFOS fraction for perch at the Original station thus indicate an abnormal pattern specific to the area.

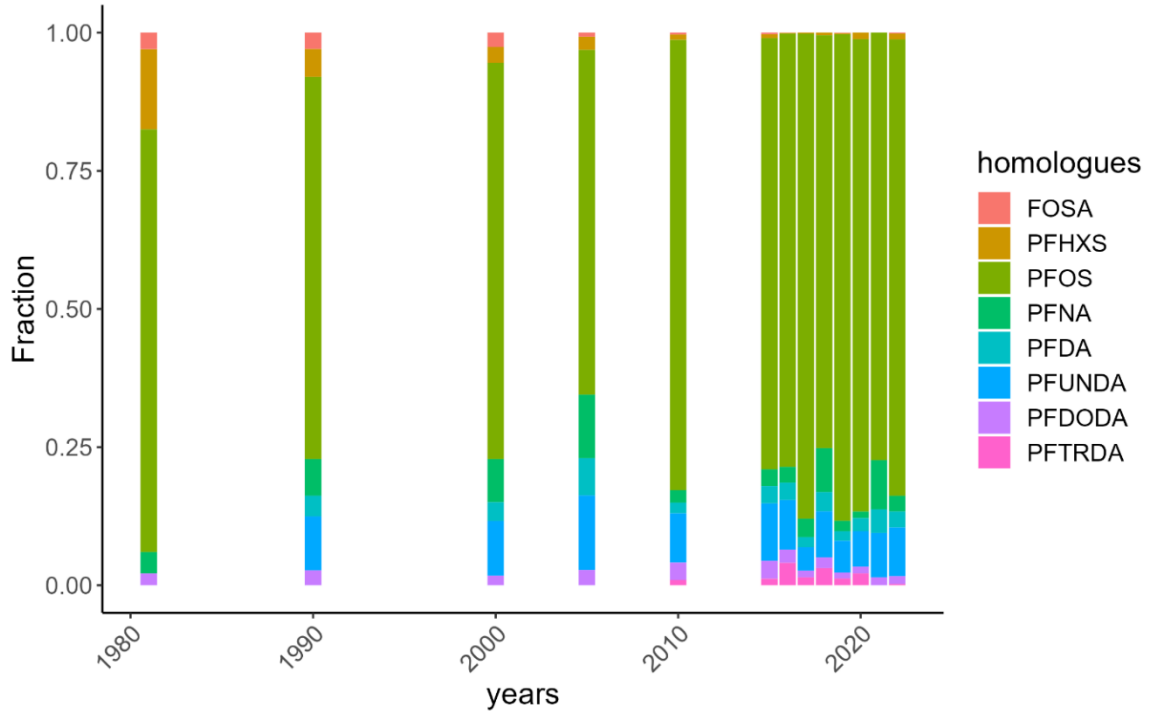


Figure 7. Fraction of various PFAS in perch from the Original station from the different time periods, 1981-2022.

Table 7. Fractions of various homologues for the different time periods in perch from the Original station. Observations below LOQ has been set to zero when calculating the fractions.

Year	FOSA	PFHXS	PFOS	PFNA	PFDA	PFUNDA	PFDODA	PFTRDA	PFUnDA/PFDA
1981	0.030	0.145	0.765	0.039	0.000	0.000	0.022	0.000	
1990	0.029	0.050	0.692	0.066	0.037	0.098	0.027	0.000	2.6
2000	0.026	0.029	0.717	0.078	0.034	0.099	0.018	0.000	2.9
2005	0.007	0.024	0.624	0.115	0.067	0.135	0.028	0.000	2.0
2010	0.003	0.010	0.815	0.023	0.019	0.089	0.031	0.010	4.7
2015	0.003	0.007	0.780	0.030	0.031	0.104	0.033	0.012	3.3
2016	0.000	0.001	0.784	0.029	0.031	0.090	0.024	0.041	2.9
2017	0.000	0.002	0.878	0.033	0.018	0.043	0.012	0.014	2.4
2018	0.000	0.005	0.747	0.080	0.035	0.083	0.020	0.031	2.4
2019	0.001	0.002	0.880	0.019	0.017	0.057	0.011	0.012	3.3
2020	0.000	0.011	0.855	0.012	0.023	0.064	0.012	0.022	2.7
2021	0.000	0.000	0.774	0.089	0.043	0.081	0.014	0.000	1.9
2022	0.001	0.011	0.825	0.028	0.029	0.088	0.014	0.002	3.0

Langberg et al. [2022] suggested the use of odd-even ratios as a mean to determine the influence of local versus atmospheric sources. Due to concentrations being close to LOQ for several of the PFCA homologues, we only used the PFUnDA/PFDA ratio. Here we found that for all the historic observations the pair was >1 (1.9-4.7) (Table 7) indicating that firefighting foam (AFFF) were not at any point a major source in the area (see Table 9 and discussion in the section on Spatial).

Spatial variability

Biological variables

Table 8 shows the mean age, total weight and total length of the fish at the stations within the greater Kvädöfjärden region. The age of perch ranges between 3 and 3.5 years (3.2±0.1 y^{-1}) and total length between 18.3 to 20.3 cm (19.3±0.6 cm). The range of these variables is small and the Original station is represented by the averages with 3.3 years and 19.1 cm. While the total weight has a larger range (76.9-107 g), perch from the Original station (91.4 g) still represents the mean weight of 90.2±9.8 g. Neither *Waldetoft et al.* [2024] nor *Åkerblom et al.* [2017] found evidence of higher ∑PFAS concentrations in larger compared to smaller perch, indicating that the larger range in weight of our perch are not likely to affect the PFAS concentrations.

Table 8. Mean age, total length (cm) and total weight (g) and the standard deviation (sd) in perch at the stations in the greater Kvädöfjärden region, n=10 fish from each station, except for the Original station, where n= 138. For the Original station the average between the years 2016-2022 has been used.

Stations	year	Age	sd	Total length (cm)	sd	Total weight (g)	sd
Original	2016-2022	3.3	0.45	19.1	0.96	91.4	14.7
Trollholmen	2023	3.3	0.67	19.8	1.53	93.8	25.4
Station 26	2023	3.1	0.32	19.5	0.60	91.4	10.7
Station 4	2023	3.3	0.48	19.1	0.55	86.3	9.42
Station 37	2023	3.1	0.32	19.3	0.95	90.5	15.0
Station 7	2023	3.2	0.63	18.6	0.84	78.1	11.2
Station 6	2023	3	0.00	18.4	0.71	76.9	9.90
Station 1	2023	3.5	0.53	18.3	0.78	79.4	11.1
Station 14	2023	3.2	0.42	19.8	0.92	93.7	17.8
Station 15	2023	3.1	0.32	19.8	1.37	103	21.0
Station 40	2023	3.2	0.42	20.3	1.37	108	22.4

PFAS spatial variability

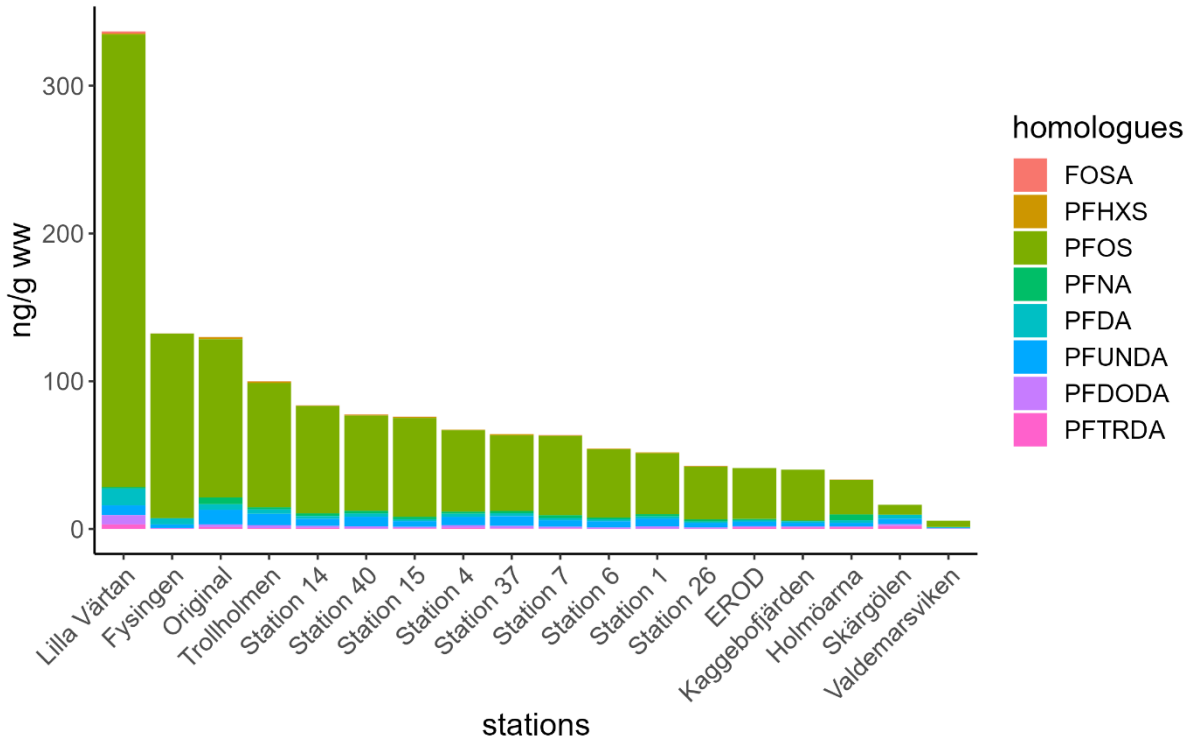
Figure 8a show concentrations of detected PFAS at the various stations including contaminated and reference (monitoring) sites. As expected, PFOS is found in highest concentration at all stations. PFAS from Lilla Värtan, a harbour in Stockholm, and Lake Fysingen, exposed to AFFF from Arlanda airport [*Ahrens et al.*, 2015], have been included to show the difference in concentration between polluted areas and Kvädöfjärden. Perch from Lilla Värtan has the highest PFAS concentration (337 ng/g ww liver), while perch from Lake Fysingen (132 ng/g ww liver) has concentrations similar to the Original station (130 ng/g ww liver). Figure 8a also shows that the concentrations of PFAS at two other monitoring sites (Holmöarna and Lake Skärgölen) are much lower, 36 and 16 ng/g ww (liver) respectively, compared to any of the stations in the greater Kvädöfjärden region. These concentrations are also more in line with PFAS concentrations in herring and cod from other monitoring stations in the Baltic Sea, ranging from 10-20 ng/g ww liver [*Soerensen et al.*, 2024a]. These results confirm that the concentrations of PFAS are elevated in the whole Kvädöfjärden region compared to the expected Swedish freshwater and marine background concentration but also that the Original station has the highest concentrations measured in the greater Kvädöfjärden region.

In Figure 8b and Table S 1 the fractions of the different PFAS are shown. For all stations in the greater Kvädöfjärden region, PFOS constituted 79-88% of total PFAS while PFOS at Holmöarna constituted 70% and at Valdemarsviken 75%. In addition, Fysingen, which is known to be polluted from Arlanda airport, and Lilla Värtan in the Stockholm harbour, which is also polluted by PFAS [*Solna*, 2021], had higher fractions of PFOS, 94 and 91%, respectively, than other stations. In

contrast, Skärgölen which is an uncontaminated lake from the freshwater monitoring programme, has a lower fraction of PFOS, 41%. The high PFOS fraction in the Kvädöfjärden region support other evidence that there is a point source in the area.

At all stations in Kvädöfjärden, as well as Valdemarsviken and Holmöarna, PFUnDA had the second highest concentration, constituting between 5-10% of the total PFAS. PFUnDA also had the second highest homologue concentrations at Lake Skärgölen, constituting 23% of total PFAS. The next highest fractions were for PFNA, PFDA and PFDoDA.

To get a sense of the importance of local versus atmospheric sources, we looked at the odd-even ratios of PFCAs. Due to concentrations for several of the PFCA homologues being close to the LOQ, we only use the PFUnDA/PFDA ratio. The PFUnDA/PFDA ratio for the spatial data in the greater Kvädöfjärden region was >1 (2.5-3.5) (Table 9) but with high PFOS concentrations. *Langberg et al.* [2022] suggested that a PFUnDA/PFDA ratio <1 suggest a local source of firefighting foam (AFFF). This is consistent with our findings of Fysingen and Lilla Värtan having ratios of 0.6 (Table 9). While we cannot determine what source the PFUnDA/PFDA ratio larger than one indicate, this result does allow us to exclude AFFF as the source of the elevated PFAS concentrations in the Kvädöfjärden region.



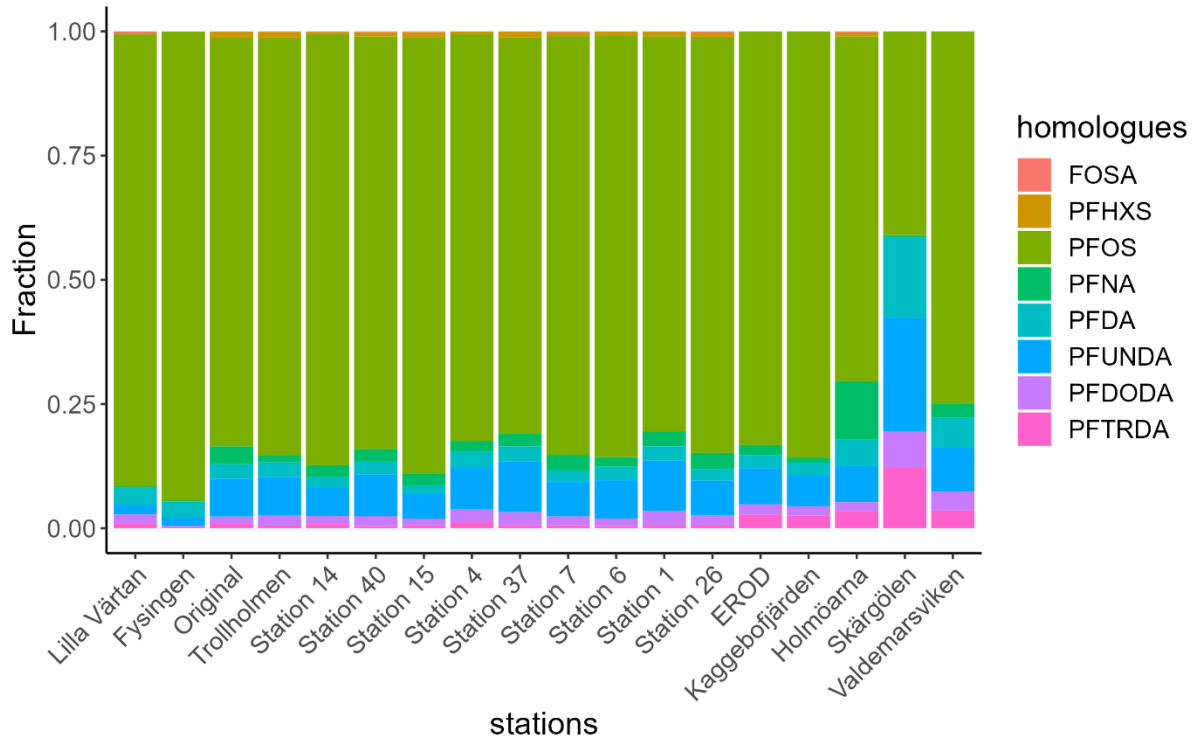


Figure 8. PFAS concentrations (ng/g ww liver) and fractions in perch from the greater Kvädöfjärden region (collected in 2023) and other spatial stations. "Original" is the original monitoring station in Kvädöfjärden, data mean 2020-2022. Holmöarna, monitoring station in Bothnian Bay, data mean 2020-2022. Valdemarsviken, data from 2016. Kaggebofjärden and EROD are two stations that were analysed 2013-2014, hence 9-10 years earlier in the Kvädöfjärden region (see Figure 9 for locations). Fysingen and Skärgölen (two freshwater monitoring stations, data mean 2019-2021).

Table 9. Concentrations (ng/g ww liver) and fractions of PFOS and the PFUnDA/PFDA ratio in perch from the Kvädöfjärden region and the other spatial stations.

Site	Region/water type	Sub-area within Kvädöfjärden region	PFOS (ng/g ww liver)	PFOS (%)	PFUnDA/PFDA
Original	Kvädöfjärden	Kvädöfjärden	107	0.835	2.6
Trollholmen	Kvädöfjärden	Trollholmsfjärden	84	0.840	2.5
Station 26	Kvädöfjärden	Trollholmsfjärden	36	0.838	2.9
Station 4	Kvädöfjärden	Trollholmsfjärden	55	0.818	2.6
Station 37	Kvädöfjärden	Trollholmsfjärden	51	0.797	3.3
Station 7	Kvädöfjärden	Trollholmsfjärden	54	0.843	3.0
Station 6	Kvädöfjärden	Trollholmsfjärden	46	0.848	2.9
Station 1	Kvädöfjärden	Trollholmsfjärden	41	0.794	3.5
Station 14	Kvädöfjärden	Licknevarpfjärden	73	0.867	2.8
Station 15	Kvädöfjärden	Licknevarpfjärden	67	0.878	3.2
Station 40	Kvädöfjärden	Licknevarpfjärden	64	0.830	3.1
Kaggebofjärden	Kvädöfjärden	Kaggebofjärden	34	0.858	2.5
EROD	Kvädöfjärden	Kvädöfjärden	34	0.833	2.7
Valdemarsviken	River		4	0.750	1.5
Holmöarna	Marine		23	0.701	1.4
Lilla Värtan	Marine		306	0.910	0.6
Skärgölen	Lake		7	0.410	1.4
Fysingen	Lake		125	0.944	0.6

Figure 9 and Table 9 presents the spatial pattern of PFOS concentrations in the greater Kvädöfjärden region. The concentration of PFOS ranged from 36 to 107 ng/g ww (liver). A simple visual analysis of the spatial observations does not reveal where a potential PFOS source comes from. The Original station has the highest concentration of all examined stations in Kvädöfjärden (107 ng/g ww PFOS) and the two closest stations (Trollholmen and station 4) have lower PFOS concentrations (20-50%). These two stations, that are geographically close together, also highlight the variability even within a close geographical range as station 4 has 35% lower PFOS concentration than Trollholmen. It seems unlikely that Trollholmsfjärden (the area west of the Original station) is the source area. In Licknevarpfjärden (stations 14, 15, 40), we also see elevated concentrations although these are 30-40% below those measured at the Original station, suggesting that this area is not the source. As no perch were collected in the water between the Original station and Licknevarpfjärden (stations 14, 15, 40) we cannot determine if there could be a potential source in this area that could influence both Trollholmsfjärden and Licknevarpfjärden.

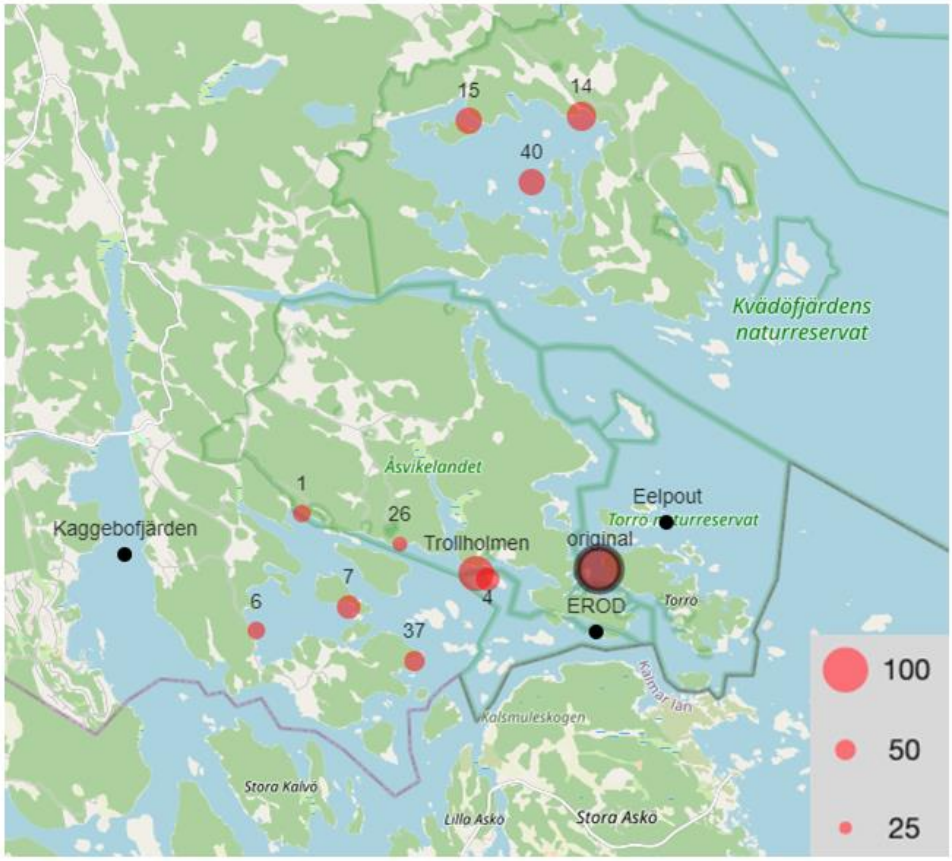


Figure 9. PFOS concentrations in perch (red circles, ng/g ww liver) in the greater Kvädöfjärden region. Black dots indicate three other stations in Kvädöfjärden with historic observations; Kaggebofjärden and EROD station (perch) and Eelpout (eelpout monitoring station). The numbers on the map is the station names. "Original" is the ordinary monitoring station in Kvädöfjärden (mean PFOS concentration 2020-2022).

Figure 10 shows PFOS in surface waters in Kvädöfjärden and the surrounding area. The PFOS sea surface concentrations are between 0.2 and 0.4 ng/L and all concentrations are therefore above the PFOS EQS for sea water of 0.13 ng/L [*HVMFS*, 2019:25]. Within the Kvädöfjärden region, we see some spatial variability in water concentrations, as is also seen in perch from the area. However, there are no measurements in Trollholmsfjärden or close to the Original station, which would allow us to directly compare the spatial pattern of water with that of perch. *Nguyen et al.* [2017] investigated PFOS concentrations in different areas in offshore waters in the Baltic Sea and Kattegat and found concentrations in the range of 0.1 up to 2.5 ng/L. For the three offshore measurements closest to Kvädöfjärden (S11-S13), they found concentrations that ranged between 0.5 and 2.0 ng/L. For Licknevarpsfärden, where we find elevated PFOS concentrations in perch similar to other stations in Kvädöfjärden (Figure 9) the PFOS concentration in water is 0.3 ng/L. This is smaller than for the offshore measurements and thus does not indicate elevated PFOS water concentrations in this area. However, more water samples from the region are needed in order to do a proper evaluation of the spatial variability of PFOS in the water column and its correlation to that in perch.

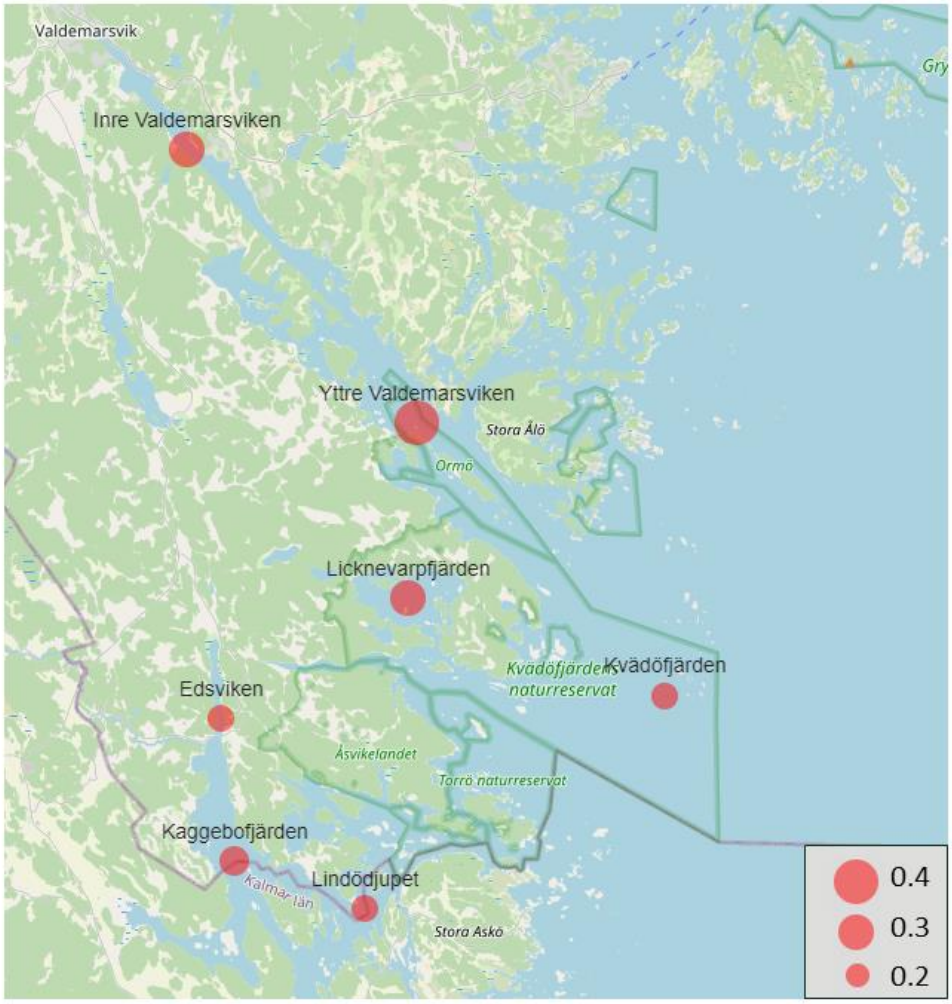


Figure 10. Map with concentration of PFOS (ng/L) in surface water samples from the area around Kvädöfjärden. Data is from 2023.

Comparison perch and eelpout in Kvädöfjärden

Eelpout is collected in Kvädöfjärden as part of the marine monitoring programme. Eelpout is collected on the north eastern side of Torrö while the perch Original station is located on the western side just a few km away (Figure 9).

Figure 11 shows the PFAS concentration and fractions in eelpout. The mean concentration of PFAS in eelpout in Kvädöfjärden is 17 ng/g ww while in perch it is 130 ng/g ww and PFOS/PFCAs makes up 68%/28% of PFAS (Table S 2, Table S 3), which is much lower than in perch (84%/16%, Table 9). A difference in PFAS concentrations is expected between the two species due to the different feeding preferences and TP. We found that perch (mean 3.2) has a TP approximately 0.5 trophic level higher than eelpout (mean 2.8). This is related to their different food preferences.

Eelpout mainly eats gastropods, chironomids, crustaceans and eggs and fry of fishes [*Froese and Pauly*, 2024] while perch eats different benthic invertebrates but also some small fish [*Froese and Pauly*, 2024].

We compare the concentrations to the only other eelpout station within the marine programme (data 2020-2022) located at the Swedish West Coast (Fjällbacka; time trends for all PFAS between 2016-2022 in eelpout is found in [*Soerensen et al.*, 2024b]). The concentrations of PFAS (11 ng/g ww liver) and PFOS (8 ng/g ww liver) as well as the fractions of PFOS and PFCAs are similar in eelpout from these two areas (Figure 11). PFAS in eelpout from the station in Kvädöfjärden therefore does not seem to be elevated in the same way that perch is.

While it seems unlikely that the difference in TP is driving a concentration difference between eelpout and perch in Kvädöfjärden of this magnitude, the different types of food they prey on could be exposed to different levels of PFAS. Further, the difference in location, although minor, could play a possible role, with the Eelpout station located further towards the outer coastal waters, likely being less exposed to any coastal point source.

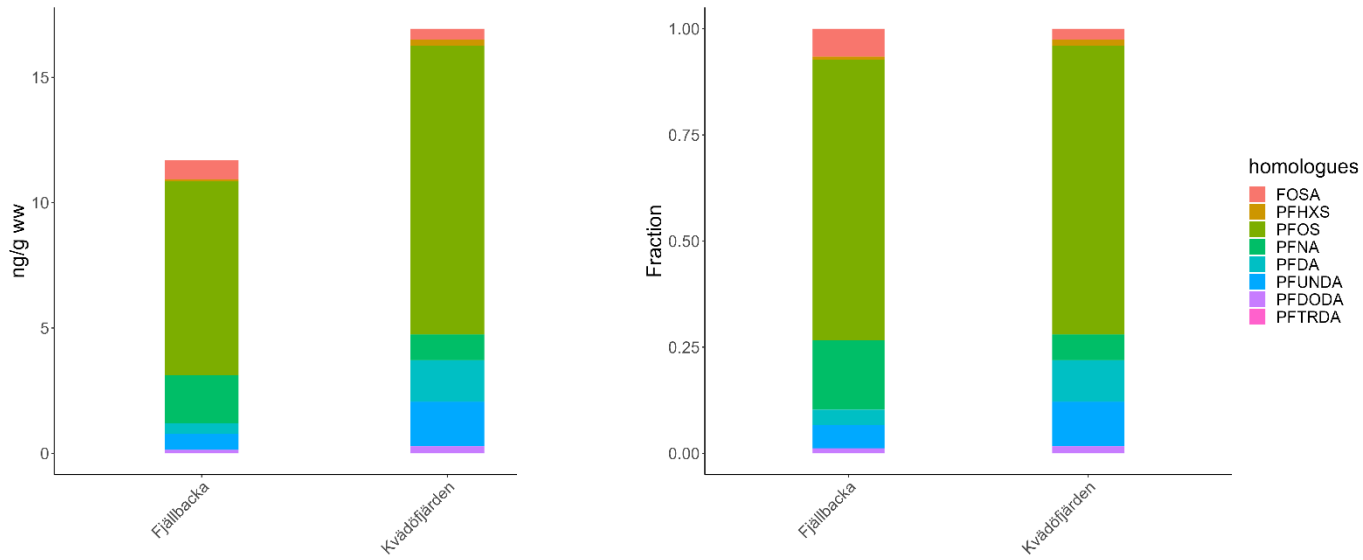


Figure 11. PFAS homologues in eelpout liver (mean 2020-2022) fractions and concentrations (ng/g ww) in Kvädöfjärden and Fjällbacka (a monitoring station on the Swedish west coast).

Harmful activities in the area

We investigate if there are any local activities in the surrounding area including direct releases into the Kvädöfjärden region that might be able to cause the high PFOS concentrations. We use the EBH database (the County boards database over hazardous activities[1]) to extract information on sources that could cause elevated PFAS concentrations. Figure 12 shows the selected activities. The harbour areas are far away from Kvädöfjärden and it seems unlikely that they will have an influence (Figure 12). For other activities (fire-fighting training sites, waste landfills, industrialized areas, sewage treatment plants and engineering industry using halogenated compounds) the activities are found further north, connected to the watersheds around Valdemarsvik. These activities could possibly explain that Yttre Valdemarsvik show the highest surface water concentrations of PFOS in the area (Figure 10). While PFOS from Yttre Valdemarsvik could in theory be transported into Kvädöfjärden (10% of the inflow compared to "Outer coastal waters"; Figure 2) this scenario seems unlikely. PFAS concentrations in perch from this area are needed to explore this possibility further.

[1] https://www.lansstyrelsen.se/stockholm/miljo-och-vatten/fororenade-omraden/kartor-over-fororenade-omraden.html

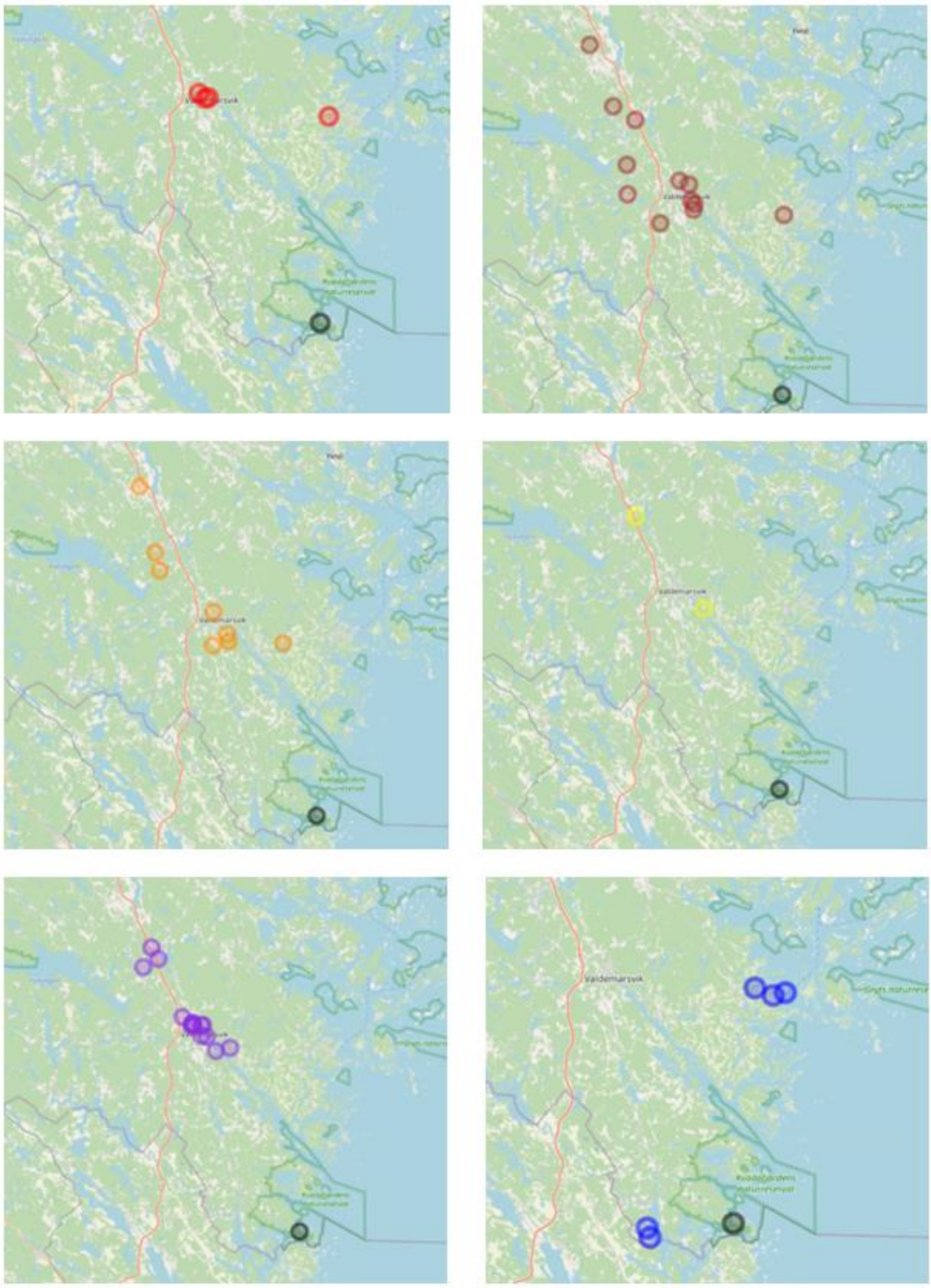


Figure 12. Harmful activities in the Kvädöfjärden region a) Fire-fighting training sites (red) b) Waste landfills (reddish brown) c) Industrialized areas (orange) d) Sewage treatment plants (yellow) e) Engineering industry using halogenated compounds (purple) f) Harbours (blue). The black circle indicates the location of the Original station.

PCA analysis (temporal and spatial)

Figure 13 shows a PCA plot that includes all spatial and temporal observations. It is based on the five homologues that contribute the most to the ∑PFAS (Table 1, Table 9). Together PC1 and PC2 explains 96.51% of the variation. The PC1 represents the PFOS concentrations and PC2 the PFCAs with increasing PFNA (C_9) with lower values and increasing PFDA, PFUnDA and PFDoDA (C_{10}-C_{12}) with higher values. All observations from the spatial survey in Kvädöfjärden in 2023 are located close to each other. In this space, also the observation from Kaggebofjärden and Valdemarsvik (river) are located. The two contaminated sites are placed to the left of this group, as they are explained by their high PFOS fractions. The reference lake is separated from all other stations, having a PFAS pattern with a higher fraction of the C_{10}-C_{12} PFCAs, which is also seen in Figure 8b. For the purpose of the PCA, the historic observations from the Original station has been split into two periods; 1981-2005 and 2010-2022. While the 2010-2022 observations mostly fall within the space of the spatial survey, the earlier observations have a lower value on the PC2 indicating a different fingerprint with a larger impact from PFNA. These observations are located close to the marine reference site. This indicate that something happened to change the PFAS fingerprint in the perch between 2005 and 2010. The fact that the older observations are located in the same space as the marine site could indicate that there, during this period, was a shift from a primary marine influence to an influence from some more local source.

Two of the recent years from the Original station (2018 and 2021) are located close to the 1981-2005 observations. These two years also stand out by having lower PFAS concentrations than other recent years – they are visibly lower on Figure 6 than other recent years for both PFOS (27 and 59 ng/g ww respectively) and PFCA (e.g. PFUnDA; 3.5 and 6.2 ng/g ww respectively) compared to the average during the 2010-2022 period of 92±40 ng/g ww and 8±3 ng/g ww. The lower PFAS concentration and more marine homologue fingerprint indicates that the perch caught during these two years have a higher proportional influence from the open ocean.

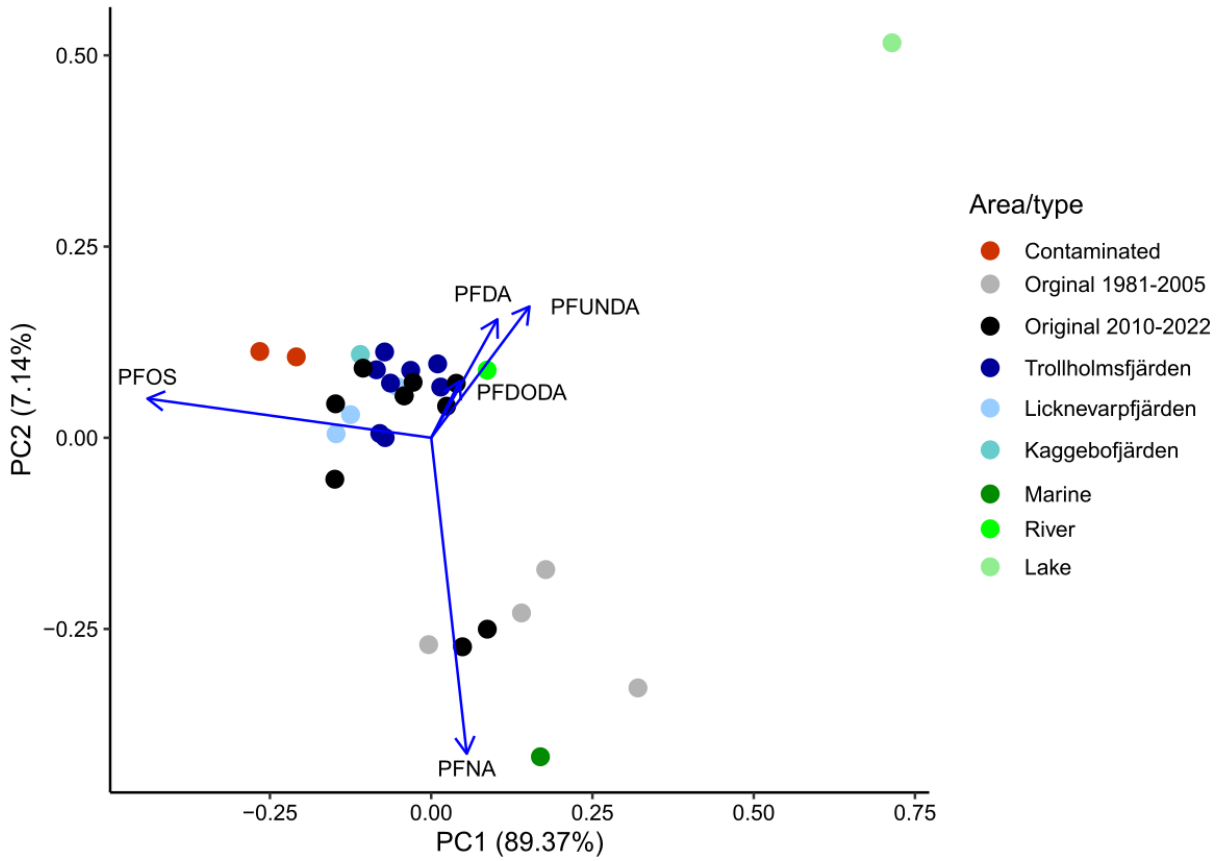


Figure 13. PCA plot with temporal and spatial PFAS fractions in perch. Contaminated=Fysingen and Lilla Värtan, Original old =1981-2005, Original=2010-2022, Trollholmsfjärden= stations nr 1, 6, 7, 26, 37, 4, and Trollholmen, Licknevarpfjärden=station nr 14, 15, 40, Marine=Holmöarna, River=Valdemarsvik, Lake=Skärgölen.

Discussion summary

We had three aims with this report. To 1) investigate the temporal variability of PFAS in Kvädöfjärden to explore the onset of a change in the concentration pattern 2) investigate the spatial variability of PFAS concentrations in Kvädöfjärden to explore the potential location of a PFAS source and 3) map harmful activities in the surrounding area.

Temporal variability: The examination of the temporal variability in perch from Kvädöfjärden shows that the concentrations of PFOS and several PFCAs, has continuously increased since the 1980s. This is opposite to other time series from the Baltic Sea, where concentration decreases or are stagnant after 2000-2010. The trophic position for perch at the Original station has increased

over time. However, we find it unlikely that this is driving the increase in PFAS concentrations as other contaminants with a high biomagnification potential measured in the same fish, do not show a similar increase. We find a change between 2005 and 2010 for the PFAS fingerprint, which indicate that a new source driving the increasing concentrations was introduced during that time. The PFAS fingerprint also suggest that the main source of PFAS in the region changed from a marine signal to that of a local source.

Spatial variability: All perch in Kvädöfjärden from the spatial survey are located in the same space on the PCA and have similar PFAS fractional patterns. However, concentrations of PFAS in perch show high spatial variability even on a small geographical scale, e.g. Trollholmen and station 4 which are very close to each other. The PFUnDA/PFDA ratio suggest that AFFF are not the source of the increasing PFAS concentrations but the ratio does not allow us to say more about possible sources. Opposite to perch, eelpout collected close to the Original station is not elevated but we do not have enough data to explore what could drive this in more depth. There is only one water measurement within the Kvädöfjärden region, and this does not suggest highly elevated PFOS concentrations compared to recent offshore measurements. The fact that the Original station has the highest perch PFAS concentrations in the region and that also perch in Licknevarpfjärden are elevated (although not as high), could indicate that the source(s) should be found in the area north of the Original station and south of Licknevarpfjärden. The water circulation in the area could support an exchange to both these water bodies from the Kvädöfjärden sub-area water body, which in turn could be influenced both from coast and from water exchange with Valdemarsviken and the offshore marine water (Figure 2). However, more measurements across the Kvädöfjärden sub-area are needed to explore this further.

Harmful activities: It was not possible to find any harmful activities in the Kvädöfjärden region that potentially could affect the PFOS concentrations. All investigated activities are located further north towards Valdemarsvik. The somewhat elevated concentrations of PFOS in surface water which was seen at Yttre Valdemarsvik can possibly be explained by these activities. PFOS from this site can in theory be transported into Kvädöfjärden, but since there is no data in perch from this area or the area between Yttre Valdemarsvik and the Original station it is hard to know if the sea surface concentration can have an influence in perch in the Kvädöfjärden region.

Conclusion

In this study, we can conclude that PFOS and PFCA concentrations have continuously increased since 1980s in perch in Kvädöfjärden. The fingerprint in perch at all sampled locations in the greater Kvädöfjärden region are similar but they differ from the older perch at the Original station, indicating that a new source has appeared between 2005-2010. The PFUnDA/PFDA ratio in perch suggest that AFFF is not the source of the increasing concentrations.

In order to determine the source area, we suggest to analyse perch caught between the Original station, with the highest PFAS concentration, and Licknevarpfjärden (with elevated concentrations) where observations are lacking. In addition, no water samples have been taken in this area or close to the Original station. Additional water samples would also be a help for identifying a local source.

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Supporting information

Table S 1. Fractions of various homologues in perch at the different stations in the greater Kvädöfjärden region. "Original" is the original monitoring station in Kvädöfjärden, data mean 2020-2022. Holmöarna, monitoring station in Bothnian Bay, data mean 2020-2022. Valdemarsviken, data from 2016 taken from SGU. The EROD station and Kaggebofjärden (data 2013-2014), and Skärgölen and Fysingen (two freshwater monitoring stations, data mean 2019-2021). B-PFOS shows the fraction of this isomer in relation to PFOS.

Site	Area	FOSA	PFHXS	PFOS	b-PFOS (fraction of PFOS)	PFNA	PFDA	PFUNDA	PFDODA	PFTRDA
Original	Kvädöfjärden	0.000	0.012	0.823	0.07	0.036	0.029	0.076	0.013	0.010
Trollholmen	Kvädöfjärden	0.001	0.011	0.840	0.08	0.015	0.030	0.076	0.020	0.006
Station 26	Kvädöfjärden	0.003	0.008	0.838	0.07	0.032	0.024	0.070	0.019	0.007
Station 4	Kvädöfjärden	0.000	0.006	0.818	0.08	0.022	0.032	0.084	0.025	0.013
Station 37	Kvädöfjärden	0.000	0.013	0.797	0.07	0.025	0.031	0.102	0.026	0.007
Station 7	Kvädöfjärden	0.002	0.007	0.843	0.07	0.031	0.023	0.070	0.018	0.006
Station 6	Kvädöfjärden	0.000	0.008	0.848	0.08	0.019	0.027	0.077	0.015	0.004
Station 1	Kvädöfjärden	0.000	0.010	0.794	0.06	0.030	0.029	0.101	0.029	0.005
Station 14	Kvädöfjärden	0.001	0.005	0.867	0.07	0.023	0.021	0.058	0.016	0.009
Station 15	Kvädöfjärden	0.001	0.010	0.878	0.06	0.024	0.016	0.051	0.013	0.006
Station 40	Kvädöfjärden	0.002	0.009	0.830	0.06	0.025	0.027	0.084	0.020	0.004
Kaggebofjärden	Kvädöfjärden	0.000	0.000	0.858	0.10	0.010	0.025	0.062	0.019	0.025
EROD	Kvädöfjärden	0.000	0.000	0.833	0.11	0.020	0.027	0.073	0.021	0.028
Valdemarsviken	Marine	0.000	0.000	0.750	-	0.028	0.060	0.088	0.038	0.036
Holmöarna	Marine	0.003	0.008	0.694	0.07	0.117	0.053	0.074	0.017	0.035
Lilla Värtan	Marine	0.004	0.002	0.910	0.13	0.003	0.034	0.020	0.019	0.009
Skärgölen	Freshwater	0.000	0.000	0.410	0.14	0.004	0.163	0.228	0.072	0.124
Fysingen	Freshwater	0.000	0.000	0.944	0.10	0.001	0.031	0.018	0.003	0.002

Table S 2. Concentration (ng/g ww liver) of detected PFAS in eelpout in Kvädöfjärden and Fjällbacka.

Homologue	Kvädöfjärden	Fjällbacka
FOSA	0.426	0.767
PFHXS	0.245	0.089
PFOS	11.52	7.732
PFNA	1.020	1.913
PFDA	1.666	0.419
PFUNDA	1.777	0.643
PFDODA	0.281	0.141

Table S 3. Fractions of detected PFAS in eelpout in Kvädöfjärden and Fjällbacka.

Homologue	Kvädöfjärden	Fjällbacka
FOSA	0.0252	0.0655
PFHXS	0.0144	0.0076
PFOS	0.6803	0.6606
PFNA	0.0602	0.1635
PFDA	0.0984	0.0358
PFUNDA	0.1049	0.0549
PFDODA	0.0166	0.0121