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Per- and Polyfluoroalkyl Substances in Water (2008–2022) and Fish (2015–2022) in The Netherlands: Spatiotemporal Trends, Fingerprints, Mass Discharges, Sources, and Bioaccumulation Factors

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Abstract: Per- and polyfluoroalkyl substances (PFAS) are persistent, bioaccumulative, and toxic synthetic chemicals of concern, which have been detected in nearly all environmental compartments. The present study provides a data analysis on PFAS concentrations in the Dutch inland and coastal national waters and fish sampled from 2008 to 2022 and 2015 to 2022, respectively. Although the fish database is relatively small, the water database is unique because of its temporal dimension. It appears that PFAS are omnipresent in Dutch water and fish, with relatively small spatial differences in absolute and relative concentrations (fingerprints) and few obvious temporal trends. Only perfluorooctanoic acid and perfluorooctanesulfonic acid (PFOS) aqueous concentrations in the rivers Rhine and Scheldt have substantially decreased since 2012. Still, PFOS concentrations exceed the European water quality standards at all and fish standards at many locations. Masses of PFAS entering the country and the North Sea are roughly 3.5 tonnes/year. Generally, the data suggest that most PFAS enter the Dutch aquatic environment predominantly through diffuse sources, yet several major point sources of specific PFAS were identified using fingerprints and monthly concentration profiles as identification tools. Finally, combining concentrations in fish and water, 265 bioaccumulation factors were derived, showing no statistically significant differences between freshwater and marine fish. Overall, the analysis provides new insights into PFAS bioaccumulation and spatiotemporal trends, mass discharges, and sources in The Netherlands. *Environ Toxicol Chem* 2024;43:965–975. © 2024 The Authors. *Environmental Toxicology and Chemistry* published by Wiley Periodicals LLC on behalf of SETAC.

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INTRODUCTION

Per- and polyfluoroalkyl substances (PFAS) are human-made chemicals, belonging to a group of thousands of individual compounds (Buck et al., 2011; Organisation for Economic Co-operation and Development, 2021) of which some have been produced since the 1940s (Prevedouros et al., 2006). Research during recent years has revealed that PFAS are very persistent, bioaccumulative, and toxic for humans and aquatic and terrestrial life (Ankley et al., 2021; Fenton et al., 2021). They are produced on a very large scale and have applications in numerous industrial branches and consumer products (Glüge et al., 2020). Consequently, environmental emissions of PFAS are very large, although exact numbers are lacking (Glüge et al., 2020). The extensive emissions, combined with the fact that PFAS either are very persistent or break down into very persistent degradation products (Liu & Mejia Avendaño, 2013; Prevedouros et al., 2006; Xiao, 2022), cause the chemicals to be ubiquitous in the environment (Evich et al., 2022). The aquatic environment plays a central role in the transport and fate of PFAS following their emissions. Many PFAS discharges take place in rivers through sewage water effluents from industry and household sources (Kurwadkar et al., 2022; Prevedouros et al., 2006). From there, the contaminants are transported to oceans, which act as global sinks for PFAS (Kurwadkar et al., 2022; Muir & Miaz, 2021; Prevedouros et al., 2006), predominantly

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owing to the chemicals' relatively high aqueous solubilities (CONCAWE, 2016). At the same time, the aquatic environment acts as a source of PFAS for sediments, aquatic organisms, and air through the formation of sea spray aerosols (Sha et al., 2022).

Because of their high aqueous solubilities and persistence, modeling PFAS concentrations in surface waters has been quite successful (Muir & Miaz, 2021). Continued refinement and validation of fate models are, however, hampered by the limited availability of recent descriptive data on, for example, mass discharges, point sources, and trends in concentrations in aquatic systems. Although information about aqueous PFAS concentrations on different spatial scales is available to some extent (Kurwadkar et al., 2022; Pan et al., 2018), temporal trends are very scarce (Land et al., 2018; Muir & Miaz, 2021); and recent mass discharges for large parts of the world are lacking (Muir & Miaz, 2021). Temporal trends and mass discharges are crucial for understanding PFAS transport and fate but also for assessing the effectiveness of chemical management actions on local, national, continental, and global scales (Muir & Miaz, 2021; Point et al., 2021). In addition, the mechanistic understanding of PFAS fate is underdeveloped because detailed knowledge about PFAS sorption to sediments and accumulation in aquatic organisms is still lacking. Bioaccumulation of PFAS does not equal the well-defined partition process occurring for "simple" nonionic hydrophobic chemicals, which primarily accumulate in lipids (Ankley et al., 2021). The number of available bioaccumulation factors (BAFs) for most PFAS in aquatic species is limited (Burkhard, 2021), which further impedes the development of improved understanding of the process and leaves simple questions yet unanswered, such as whether bioaccumulation of PFAS in marine and freshwater species is similar (Burkhard, 2021). Actually, in their recent critical review, Ankley et al. (2021) stated that "understanding bioaccumulation is arguably the most significant exposure challenge relative to PFAS."

The executive agency of the Dutch Ministry of Infrastructure and Water Management (Rijkswaterstaat; hereafter referred to as RWS) has monitored PFAS in the Dutch national waters since 2008. In addition, concentrations in fish (liver) have been measured as of 2015. Consequently, RWS has access to an extensive PFAS database, which includes concentrations in rivers, lakes, and coastal waters (https://waterinfo.rws.nl), and thereby provides a unique opportunity to address some of the knowledge and data gaps discussed above. The present study aimed to analyze the RWS PFAS database in terms of spatial trends, compliance testing with European and national standards, short- and long-term temporal trends, compound profiles, and mass discharges in the catchments of three important European rivers, that is, the Rhine, Meuse, and Scheldt. Furthermore, concentrations in water and fish were combined to derive in situ freshwater and marine BAFs. Although PFAS long-term trend analyses have been published for different biotic compartments (Land et al., 2018), to the best of the author's knowledge, the present database on aqueous concentrations is unparalleled in its temporal dimension (Land et al., 2018; Muir & Miaz, 2021). The results of the data analysis provide novel insights into several aspects of the occurrence and distribution of PFAS in the aquatic environment.

MATERIALS AND METHODS

PFAS

The number of PFAS monitored by RWS in water has evolved since 2008: Initially only two compounds (perfluorooctanoic acid [PFOA] and perfluorooctanesulfonic acid [PFOS]) were analyzed, but in 2022, 31 PFAS were quantified. These included 11 perfluorocarboxylic acids (PFCAs), nine perfluorosulfonic acids, and 11 other PFAS, including some telomers, perfluorosulfonamidoacetic acids, and polyfluorosulfonic acids. From the list of 31 compounds, 14 were selected for the present data analysis. These included the more "classical" PFAS, as well as some "emerging" ones. Those for which aqueous concentrations generally (i.e., at most locations) were below reporting limits were excluded, and the sum of the concentrations of the selected 14 PFAS comprised >95% of the total PFAS concentration quantified in the water samples. Two of the selected PFAS concerned isomers, that is, the linear and branched isomers of PFOS, which were summed for the sake of data analysis and compliance testing with European standards (European Commission, 2011, 2013). As such, the water data analysis was performed for 13 compounds, that is, perfluorobutanoic acid (PFBA), perfluoropentanoic acid (PFPeA), perfluorohexanoic acid (PFHxA), perfluoroheptanoic acid (PFHpA), PFOA, perfluorononanoic acid (PFNA), linear perfluorobutanesulfonic acid (PFBS), linear perfluoropentanesulfonic acid (PFPeS), linear perfluorohexanesulfonic acid (PFHxS), PFOS, 2,3,3,3-tetrafluoro-2-(heptafluoropropoxy)propanoic acid (GenX, also referred to as HFPO-DA), 2-(perfluorohexyl)ethane-1-sulfonic acid (6:2 FTS), and *N*-ethyl-perfluorooctane sulfonamidoacetic acid (EtFOSAA).

As of 2015, on behalf of RWS, fish (livers) were analyzed by the laboratory of Wageningen Marine Research (IJmuiden, The Netherlands) for the presence of 16 PFAS: PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFuDA), perfluorododecanoic acid (PFdoDA), perfluorotridecanoic acid (PFtDA), perfluorotetradecanoic acid (PFteDA), PFBS, PFHxS, perfluoroheptanesulfonic acid, PFOS, and perfluorodecanesulfonic acid. Out of these 16 compounds, PFBA, PFPeA, PFHxA, PFHpA, and PFBS were not detected, implying concentration data in fish (livers) for a maximum number of 11 PFAS.

Sampling locations

The RWS PFAS database contains data for approximately 60 water sampling locations, in either the freshwater or the marine environment. Considering this large number and the fact that for some of the locations aqueous concentrations did not concern recent data, a selection of 17 inland and coastal locations was made for in-depth analysis in the present study. These locations were selected on the basis of their geographical position and the availability of at least 4 years of recent data. Four of the selected locations are located in the Meuse catchment, six in the Rhine catchment, three in the Scheldt catchment, and four along the North Sea coast (marine locations). Figure 1A shows the locations on a map of The Netherlands. Sampling location names (abbreviations) are those officially used by RWS, which were adopted in the present study. Explanation of the abbreviated names is provided in Supporting Information, Table S1. Data availability for these locations varied between 4 (2019–2022) and 15 (2008–2022) years. Annual sampling frequency at the locations varied between one and 16; an overview is presented in Supporting Information, Table S2.

Concentrations of PFAS above detection limits in fish were available for six to 11 locations sampled between 2015 and 2022. Locations sampled in the different years partly overlap, such that data were available for in total 16 different locations, of which six are located in the marine and 10 in the freshwater environment (see Figure 1B). Samples concerned roach (*Rutilus rutilus*; whole fish) at freshwater locations and flounder (*Platichthys flesus*; whole fish or liver) or plaice (*Pleuronectes platessa*; liver) at marine locations (see Supporting Information, Table S3, for an overview of samples).

Sampling methodology, sample extraction, and chemical analyses of water and fish samples are described in the Supporting Information.

Data analysis

Data for the selected water sampling location–PFAS combinations (i.e., 14 PFAS × 17 locations = 238 data subsets) were extracted from the crude database. For each subset, annual average concentrations (nanograms per liter) with accompanying standard deviations were calculated for each individual PFAS, which subsequently were analyzed in terms of spatiotemporal trends, compound profiles, and mass discharges in the different catchments. Averaged concentrations (arithmetic means) were calculated to be able to directly compare the results to environmental quality standards (EQSs; given as annual averages) and because calculating median values for years with a limited sampling frequency (e.g., 2; see Supporting Information, Table S2) is not possible. Flow rates required for estimating mass discharges were obtained from the RWS water database (https://waterinfo.rws.nl).

Concentrations of PFAS above reporting limits in fish (liver) were averaged per year (if applicable) and presented as annual average (micrograms per kilogram wet wt) with standard deviation per individual compound and per location. These concentrations were analyzed in terms of spatiotemporal trends and compound profiles. In addition, BAFs were calculated by dividing the concentration in whole fish or fish liver by the annual average concentration in water from the same sampling location. If the latter was not available, aqueous concentrations from a nearby location in the same water body were used in the calculations.

RESULTS AND DISCUSSION

Spatial trends

The calculated annual average concentrations of the selected PFAS in water ($^{AA}C_w$; n ~ 2050) are graphically presented

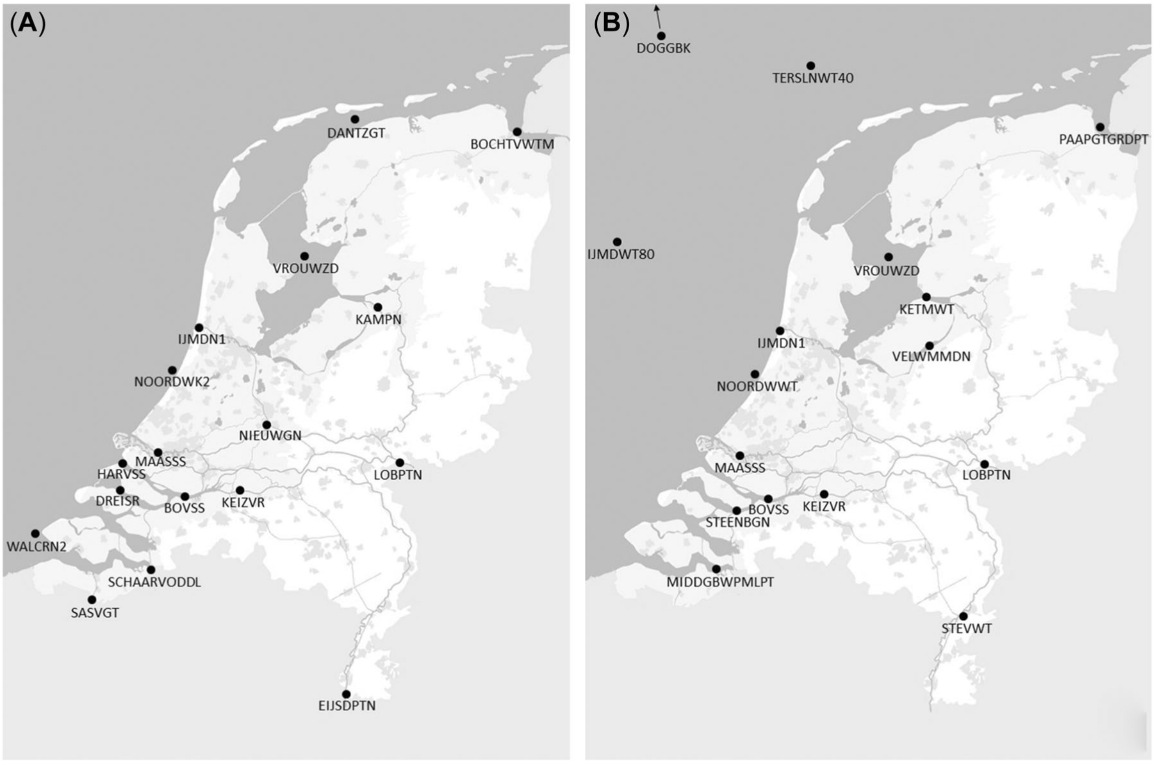

FIGURE 1: Geographical position of the per- and polyfluoroalkyl substances sampling locations selected for data analysis in the present study for water (**A**) and fish (**B**). The arrow next to location DOGGBK in (**B**) indicates that this location is (far) outside the borders of the map. Sampling location abbreviations are explained in Supporting Information, Table S1.

in Supporting Information, Figure S1 (grouped by chemical). These data first of all demonstrate that PFAS are present at all selected locations. Some of the compounds have been detected in the nanograms per liter range since 2008. The observed concentrations are different for the different compounds, with differences up to a factor of 10,000. Generally, PFBA and PFBS occur at the highest concentrations (up to ~400 ng/L), whereas EtFOSAA has the lowest $^{AA}C_w$s (generally <1 ng/L). Concentrations on the same order of magnitude have been detected in surface waters from developing countries (Baabish et al., 2021), Europe, North and South America, Asia, and Africa (Kurwadkar et al., 2022; Muir & Miaz, 2021; Pan et al., 2018), although concentrations of several PFAS (primarily PFOS and PFOA) at certain locations in Asian, European, and US rivers have been found to be much higher (Kurwadkar et al., 2022). This once more indicates that PFAS are globally distributed (Evich et al., 2022; Pan et al., 2018) and most probably well mixed in the global aquatic environment. Likely because of the latter, differences in $^{AA}C_w$s for a specific compound detected at different locations in The Netherlands are relatively small. This is illustrated in Supporting Information, Table S4, in which the most recent $^{AA}C_w$s (2022) are listed, and in Supporting Information, Table S5, in which median and percentile data of the overall selected data set are presented. Still, concentrations in coastal waters generally are lower than those in inland water systems (dilution principle), and two hotspot locations exist: SASVGT and SCHAARVODDL, locations receiving input from Belgium through the river Scheldt. Here, 2022 $^{AA}C_w$s are up to seven times as high as at the other inland locations. For locations in the latter group, $^{AA}C_w$s for a specific PFAS only differ by a factor of 1.5 to 2.8, excluding GenX, 6:2 FTS, and EtFOSAA, regardless of the exact geographical position of the location. In Supporting Information, Figure S2A, these relatively small spatial differences are visualized for PFOS as an important representative. In contrast, $^{AA}C_w$s for in particular GenX vary considerably across the country. Generally, concentrations are very low (<1 ng/L), but at certain locations, concentrations are substantially higher. For this reason, five additional locations were included in the GenX data analysis (see Supporting Information, Table S4 and Figure S1). The variability in GenX concentrations is explained by the fact that, in contrast to the other selected PFAS, this chemical has more specific, local sources (further discussed in the section *Sources and means for source identification*).

Concentrations of PFAS detected since 2015 in fish are presented in Supporting Information, Figure S3. As compared with the water compartment, the number of PFAS detected is lower, and concentrations of several (short-chain) PFAS are often below detection limits. However, for the rest a similar picture emerges: The lowest concentrations are found in organisms caught at distant offshore locations, concentrations are higher at inland locations, and the highest levels occur in flounder from the Scheldt (see Supporting Information, Table S6). In the latter case, in particular PFOS concentrations are very high (up to 190 µg/kg in 2015), although it should be noted that even higher concentrations have been detected in fish from certain hotspots, such as airports (Ahrens et al., 2015; Kwadijk et al., 2014; Langberg et al., 2022). The spatial differences in PFOS concentrations in fish caught in (the data-rich year) 2021 at locations from across the country are visualized in Supporting Information, Figure S2B. Overall, the current fish database demonstrates PFAS concentrations on the same order of magnitude (low to medium micrograms per kilogram wet wt range) as those previously detected in Dutch fish (Zafeiraki et al., 2019) and fish from, for example, the Laurentian Great Lakes (Point et al., 2021), European lakes (Valsecchi et al., 2021), and Belgium (Byns et al., 2022; Teunen et al., 2021).

Fingerprints

Another way of presenting PFAS concentrations is by plotting the concentrations of all compounds per location in a single graph. This is done in Supporting Information, Figure S4, by plotting all compounds separately, instead of in stacked columns as is done in most other studies (Byns et al., 2022; Langberg et al., 2022; Point et al., 2021; Remucal, 2019; Schultes et al., 2020; Teunen et al., 2021; Valsecchi et al., 2021; Zafeiraki et al., 2019), because stacked bar charts are somewhat difficult to interpret. Supporting Information, Figure S4, presents a location-specific PFAS profile or "fingerprint" for all locations, based on the 2022 aqueous concentrations. Although differences do exist, fingerprints for the different locations are rather similar, showing a clear general pattern, which is illustrated in Figure 2A: Of all PFCAs, PFBA generally occurs in the highest concentration. For PFCAs with longer carbon chains, concentrations in water gradually decrease up to PFNA, with the exception of PFOA, for which the concentration always peaks. For the perfluorosulfonic acids (PFSAs), concentrations in water are generally the highest for PFBS and always the lowest for PFPeS. Going from PFPeS to PFOS, concentrations gradually increase. Finally, for the other three PFAS (not belonging to a common group), aqueous concentrations are generally the highest for 6:2 FTS and the lowest for EtFOSAA. Although the PFCA pattern may suggest a "hydrophobicity effect" (hydrophobicity of the compounds increases with increasing carbon chain length, thereby lowering the aqueous solubility and increasing sorption potential toward sediment or suspended matter), such an effect explains neither the PFOA peak nor the pattern observed for the PFSAs. Also, aqueous concentrations (nanograms per liter range) are all well below these compounds' solubilities (milligrams to grams per liter range; CONCAWE, 2016). Therefore, most likely the fingerprints are primarily determined by emission patterns instead: Higher emissions will cause higher aqueous concentrations. In addition, the PFOA peak and the relatively high concentrations of PFOS may partly be explained by degradation of certain other PFAS ("precursors") into these persistent C_8 degradation products (Liu & Mejia Avendaño, 2013; Prevedouros et al., 2006; Xiao, 2022), next to the fact that they are (still) being released themselves.

Despite the general similarities, specific differences in fingerprints for different locations are observed. For instance, the concentrations of the first three PFCAs in the Meuse are similar,

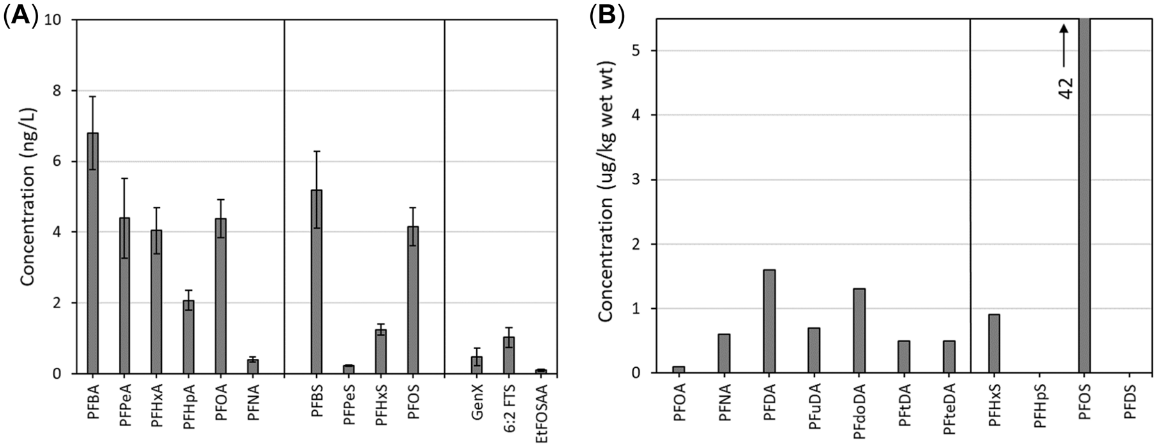

FIGURE 2: Concentrations of a series of per- and polyfluoroalkyl substances (PFAS) in water (**A**; 2022 data) and flounder (**B**; 2021 data) from location IJmuiden (IJMDN1). The relative distribution of the concentration bars provides a PFAS profile or "fingerprint." The fingerprints shown here represent the generally observed pattern. Vertical lines separate different PFAS groups. The bar for perfluorooctanesulfonic acid (PFOS) in (**B**) is cut off for visual reasons (actual concentration indicated). Error bars in (**A**) represent standard deviations ($n = 13$). Data in (**B**) are based on a single measurement ($n = 1$). PFBA = perfluorobutanoic acid; PFPeA = perfluoropentanoic acid; PFHxA = perfluorohexanoic acid; PFHpA = perfluoroheptanoic acid; PFOA = perfluorooctanoic acid; PFNA = perfluorononanoic acid; PFBS = linear perfluorobutanesulfonic acid; PFPeS = linear perfluoropentanesulfonic acid; PFHxS = linear perfluorohexanesulfonic acid; PFOS = perfluorooctanesulfonic acid; GenX = 2,3,3,3-tetrafluoro-2-(heptafluoropropoxy)propanoic acid; 6:2 FTS = 2-(perfluorohexyl)ethane-1-sulfonic acid; EtFOSAA = *N*-ethyl-perfluorooctane sulfonamidoacetic acid; PFDA = perfluorodecanoic acid; PFuDA = perfluoroundecanoic acid; PFdoDA = perfluorododecanoic acid; PFtDA = perfluorotridecanoic acid; PFteDA = perfluorotetradecanoic acid; PFHpS = perfluoroheptanesulfonic acid; PFDS = perfluorodecanesulfonic acid.

and specific deviations exist for the two hotspot locations. Based on such deviations, fingerprints may help identifying sources, as will be discussed in the section *Sources and means for source identification*.

Fingerprints of PFAS for fish are presented in Supporting Information, Figure S5 (for the most data-rich year 2021). This Figure shows that also for fish a general pattern is visible, as illustrated in Figure 2B, with the highest concentration always occurring for PFOS. The second highest PFSA concentration always concerns PFHxS, and the highest PFCA level is generally observed for PFDA. Concentrations of PFCAs with longer carbon chains than PFDA generally decrease with increasing length, although in one-third of the cases the concentration of PFdoDA peaks and is similar to that of PFDA. Interestingly, fingerprints for whole flounder and flounder liver are slightly different, with PFtDA being enriched in the latter matrix. Concentrations of PFAS in liver are often higher than those in whole fish or fillet (Valsecchi et al., 2021) and are so in the present data (Supporting Information, Figure S3). Probably, this is due to binding to specific proteins. However, the reason for PFtDA enrichment in the present liver samples is unknown; both favored protein binding and specific biotransformation of precursors to PFtDA may be involved. The overall fingerprint for fish will therefore be determined by additional factors on top of those driving the "aqueous fingerprint," including degradation or metabolization of precursors (Ellis et al., 2004; Zhang et al., 2021). In fact, this may perhaps explain the sometimes observed relatively lower concentrations of PFCAs with even-numbered (*n*), as compared with the longer ($n + 1$) odd-numbered, carbon chain lengths in Supporting Information, Figure S5 (Ellis et al., 2004). Primarily, however, fish fingerprints will be driven by the compounds' affinities for water, on the one hand, and fish (constituents), on the other hand, that is, by partition processes, as will be discussed in the section *BAFs*. Although it has been proposed to use fish fingerprints for source identification (Langberg et al., 2022), it should be noted that fingerprints for the different Dutch locations are very similar and that too few "standard" fingerprints for the many different specific PFAS sources are available to perform successful source apportionment on the present data. Instead, the water data were used for an investigation of (point) sources (see section *Sources and means for source identification*).

Temporal trends

Supporting Information, Figure S1, shows ${}^{AA}C_w$ values for up to 15 sampling years and thus can be used to investigate if any long-term temporal trends exist. To the best of the author's knowledge, no other such long-term data sets are available in the scientific literature, and earlier temporal trend analyses of aqueous concentrations concerned only 2- to 4-year sampling periods (Land et al., 2018) or pooling and averaging data from multiple studies conducted in different years (Muir & Miaz, 2021). In Supporting Information, Table S7, a summary of the most important temporal trends observed for the selected PFAS is presented. Generally, only few clear and/or recent trends are visible. Note that this conclusion is based on visual interpretation of the ${}^{AA}C_w$ time series. Statistically significant declining or increasing trends (i.e., changes in concentrations with a couple of percentages per year) may perhaps be identified when performing statistical trend analyses; however, such analyses deliberately were not conducted because the results will be dependent on the data used (${}^{AA}C_w$ vs. monthly measurements) and the statistical model applied (type and linear vs. nonlinear; Land et al., 2018). Also, in the author's opinion (considering the data variability caused by, e.g., seasonal fluctuations and analytical uncertainty), the value and applicability of any such trends are very limited. However, for PFOA and PFOS, clear and substantial long-term temporal trends are visible (see Figure 3). Most notably, since 2012 PFOA concentrations have gradually decreased by a factor of approximately 4 in the river Scheldt and by a factor of approximately 2 in the river Rhine. Similarly, since 2008 PFOS concentrations have decreased by a factor of approximately 3 (location LOBPTN) to approximately 6 (location IJMDN1) in the river Rhine and by a factor of approximately 2 in the river Scheldt. Although the current PFOA concentrations in the river Meuse are approximately half the concentrations detected in 2011 to 2014, a gradual decrease as seen in the other rivers is not observed. Concentrations of PFOS in the Meuse actually do not show any long-term temporal trend at all. The decrease in PFOS concentrations in the Rhine and Scheldt is probably the result of the restrictions in production and use for this chemical, which went into force in 2006, and its placement on the Stockholm Convention Annex B list in 2009. From 2012 on, PFOA was replaced with (GenX) alternatives by the fluoropolymer industry, which coincides with the onset of the decrease observed for this chemical in the same rivers. Since July 2020, a REACH (Registration, Evaluation, Authorisation and Restriction of Chemicals) restriction applies to PFOA.

The observation that the concentrations of both compounds more or less stabilized 6 to 7 years ago, at least in the Rhine and Meuse, may be due to the following: (i) several (essential) applications of PFOS are still permitted; (ii) both compounds can be released during the production of other PFAS, during use or disposal of products still containing PFOS and PFOA (Ahrens & Bundschuh, 2014), and from historically contaminated environmental compartments (e.g., soils and sediments); and (iii) as mentioned before, both chemicals can be formed during the degradation of PFAS precursors (Liu & Mejia Avendaño, 2013; Prevedouros et al., 2006; Xiao, 2022).

In addition to long-term trends, the database was used to study short-term trends by focusing on monthly measurements and constructing scatter plots, which actually provide insight into the underlying variability of the $^{AA}C_w$ values. Examples of monthly concentration profiles and short-term trends are presented in Supporting Information, Figure S6, for two PFAS. Generally, a seasonal fluctuation pattern is observed, with PFAS concentrations varying by a factor of approximately 1.5 to 4. Most probably, this pattern is determined by the water level or discharge flow because concentrations are generally the lowest in December/January to March/April, when river water discharges are the highest (data not shown). For the coastal locations a similar pattern is (therefore) not (as clearly) observed. Deviations from the general pattern will be discussed in the section *Sources and means for source identification*.

Despite the fact that PFAS have been analyzed in Dutch fish since 2015, generally too few data points are available per location to observe temporal trends for fish. Data for 6 years at

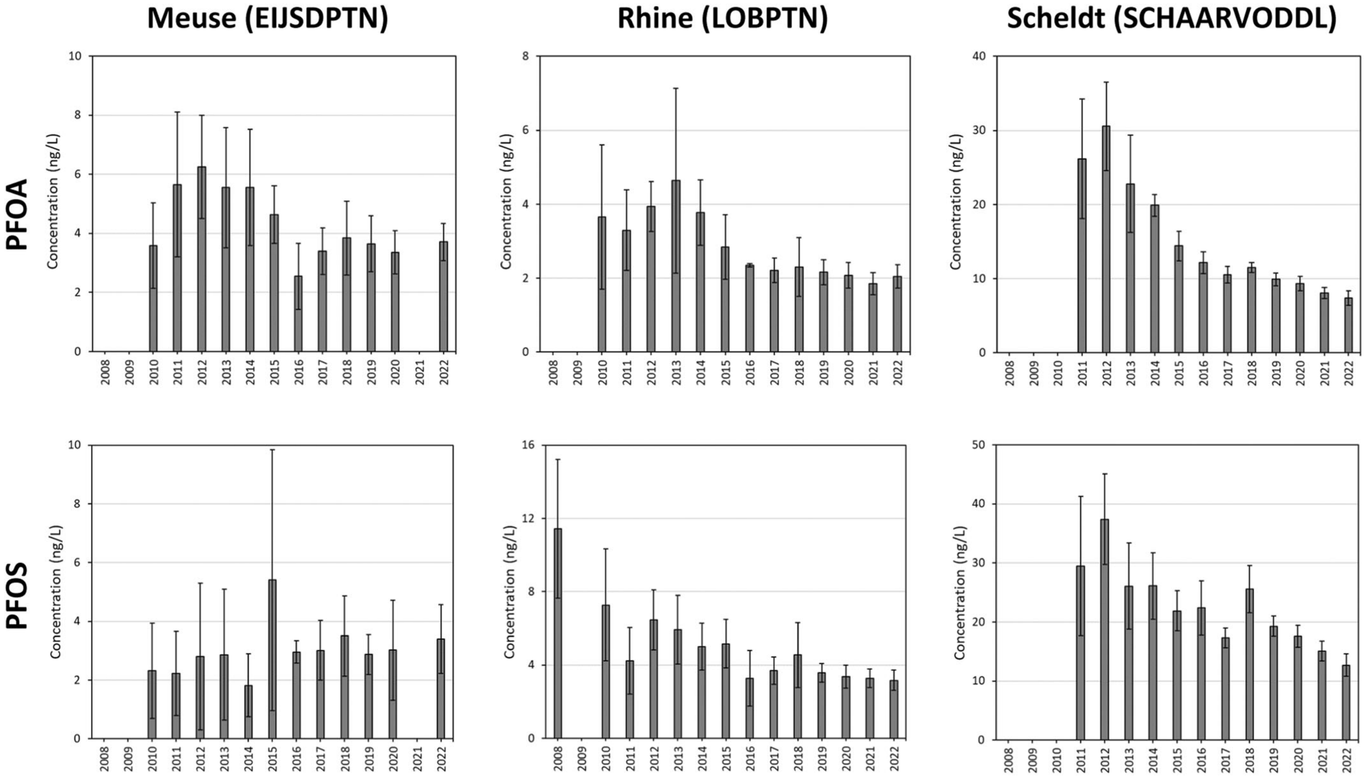

FIGURE 3: Annual average concentrations of perfluorooctanoic acid and perfluorooctanesulfonic acid in the rivers Meuse (location EIJSDPTN), Rhine (location LOBPTN), and Scheldt (location SCHAARVODDL) between 2008 and 2022. The absence of a bar indicates the absence of data for the year/compound in question. Error bars represent standard deviations ($n = 2–16$; see Supporting Information, Table S2). PFOA = perfluorooctanoic acid; PFOS = perfluorooctanesulfonic acid.

most are available for certain locations. In these cases, the data do not suggest any temporal trends. This generally agrees with the lack of clear trends observed in fish in other reports, at least from 2000 on (Land et al., 2018; MacGillivray, 2021; Remucal, 2019). Yet, one exception exists in the current database: At location MIDDGBWPMLPT (Scheldt), PFDA, PFuDA, PFHxS, and PFOS concentrations in flounder liver have decreased by a factor of approximately 2 (PFOS) to 3 (other three compounds) between 2015 and 2021 (Supporting Information, Figures S3 and S7). It should be noted though that concentrations increased again in 2022, except for PFDA. Interestingly, the decreasing trends are more pronounced than those observed in aqueous concentrations (if any; see Supporting Information, Figure S1 and Table S7). The reason for this discrepancy is unclear.

Mass discharges

To estimate the PFAS masses carried by the large European rivers flowing through The Netherlands and entering the North Sea, $^{AA}C_w$ values were multiplied by the annual average river flow rates. This was done by using the 2008 to 2021 data for locations LOBPTN (Rhine), EIJSDPTN (Meuse), and SCHAARVODDL (Scheldt), thus focusing on masses entering the country. Estimating PFAS masses that leave the country and enter the North Sea is challenging because reliable flow rates of all water systems connecting to the North Sea are not available. Hence, calculating accurate mass balances (country in vs. out) and thereby the Dutch addition to the mass discharges is not possible. Mass discharges for 2022 could not be estimated because flow rates for this year were not available at the time of data analysis. Also, 2021 mass discharges for location EIJSDPTN could not be calculated because 2021 concentrations were not available. All results are graphically presented in Supporting Information, Figure S8; the 2020 and 2021 mass discharges are presented numerically in Supporting Information, Table S8.

The data in Supporting Information, Table S8, demonstrate that in total more than 3 and 3.5 tonnes of the selected 13 PFAS entered The Netherlands in 2020 and 2021, respectively. These masses will also be the minimum masses that flowed into the North Sea in these years, which is close to the yearly PFAS discharge estimated for 2015 to 2019 by Muir & Miaz (2021). In 2020, approximately 60% of this total mass was carried by the river Rhine, 34% by the Scheldt, and only 6% by the Meuse. Note, however, that approximately one-third of the total mass was represented by PFBA, of which the largest part was transported by the Scheldt. It should also be noted that the PFAS mass carried by the Scheldt almost immediately leaves the country and enters the North Sea, whereas PFAS in the rivers Rhine and Meuse travel throughout a large part of the country. Approximately one-third of the total PFAS mass enters the North Sea at Rotterdam (location MAASSS; data and calculations not shown).

Because the graphs in Supporting Information, Figure S8, show data for multiple years, they also allow for identifying temporal trends in mass discharges. These trends are, however, similar to the trends observed for the $^{AA}C_w$ values (Supporting Information, Figure S1) because mass discharges were calculated by multiplying $^{AA}C_w$ values by flow rates, which are rather constant throughout the years. Obviously, the same applies to mass discharge–based fingerprints. Summing the yearly masses demonstrates that since the start of the PFAS measurements in 2008, at least 43 tonnes of the selected 13 PFAS were carried by the three rivers, with an averaged load of approximately 3.5 tonnes per year. Although these discharges are quite substantial, it should be noted that the estimated total mass discharge by Chinese rivers amounts to (much) more than 40 tonnes per year (Muir & Miaz, 2021; Pan et al., 2018).

Sources and means for source identification

Taken all together, the above suggests that PFAS sources in Dutch national waters predominantly are diffuse. After all, (i) $^{AA}C_w$s at different inland locations are similar (Supporting Information, Table S4), (ii) $^{AA}C_w$s are relatively stable over the years (exempting the cases discussed), (iii) short-term (yearly) variation is limited, and (iv) fingerprints for different locations are similar. The major PFAS load is carried by the large rivers from abroad and enters the country at the borders. However, because concentrations and fingerprints downstream in the different catchments do not change considerably and water is added to the rivers through rainfall and wastewater discharges, Dutch PFAS sources seem to be primarily diffuse as well and similar to foreign sources. Still, the database can potentially be used to trace point sources, in fact based on the relatively stable concentrations and patterns noted. High standard deviations for $^{AA}C_w$s (corresponding to more than a factor of 2–3 caused by seasonal fluctuations) may be a first indication for incidental discharges and thus specific point sources. Focusing on monthly concentrations (plotted over the course of, e.g., 2 years) may subsequently reveal any outliers or considerable deviations from the "standard" seasonal fluctuation pattern, which would add to the suggestion of the presence of a specific source. This also applies to deviations from the "standard" fingerprint within a specific catchment. Applying these "tools" to the current database suggests the existence of several point sources, of which three will be discussed below. The most obvious case relates to GenX. Until 2020, this chemical was not detected in Rhine water entering the country but was found downstream in the west and middle of the country in the vicinity of Rotterdam. Previous reports already noted that GenX was emitted to air and water during the production of fluoropolymers by a Chemours (formerly Dupont) factory in the city of Dordrecht (Gebbink et al., 2017). Concentrations at locations close to this factory (Supporting Information, Figure S9) are obviously the highest and most variable but have decreased substantially from 2017 on, after the company announced it would reduce emissions 99% by 2020. The database demonstrates that a 99% reduction was not fully achieved, but, more remarkable, the 2021 $^{AA}C_w$ data are elevated and highly variable. Monthly data of the two closest locations (BRIENOD and MAASSS) suggest GenX discharges in the autumn of 2021, based on peak concentrations of up to 50 ng/L (Supporting Information, Figure S10). Using the available flow rates at MAASSS, the estimated cumulative GenX mass being discharged between September 2021 and April 2022 would be approximately 120 kg, whereas the factory had a license for discharging 7 kg/year.

A second point source example concerns location SASVGT, which receives Scheldt water from Belgium but has a clearly deviating 2021 fingerprint (not shown). Concentrations of PFOS are highly variable, on average being almost four times as high as PFBS concentrations, instead of being similar or 1.5 to 2 times lower (cf. the "standard" fingerprint; Figure 2). Likewise, the variable 2021 $^{AA}C_w$s of PFHxS are similar to those of PFBS, instead of being many times lower. Also, PFPeS concentrations are relatively variable. In addition, the monthly concentration profiles of these three chemicals demonstrate concentration peaks in the spring of 2021, actually at the time that concentration dips are observed for other PFAS (Supporting Information, Figure S11). Furthermore, elevated concentration are seen for PFBS and GenX in the autumn of 2020 and 2021, respectively. Altogether, these data suggest a local source(s), which, considering the specific compounds, may be one or more waste-processing companies in Belgium (Supporting Information, Figure S9).

Third, $^{AA}C_w$s and the fingerprint at SCHAARVODDL (Scheldt) also point to local PFAS sources. Most obviously, PFBA concentrations at this location are very high, both absolutely and relatively (Supporting Information, Figure S4). A possible, major source of this compound and several other PFAS at this location is Indaver, another large waste-processing company in the harbor of Antwerp, as suggested by very high PFBA levels in wastewater of the company (https://www.vmm.be/data/waterkwaliteit) and monthly concentration profiles of several PFAS (e.g., PFBA, PFPeA, PFHpA, and GenX) at SCHAARVODDL being highly similar to those at the harbor dock receiving the company's wastewater (Supporting Information, Figure S12). From this dock, water is discharged into the Scheldt just downstream of location SCHAARVODDL (Supporting Information, Figure S9). For the other PFAS, concentrations in the Scheldt are generally higher than in the harbor dock, pointing to additional sources outside of the dock (i.e., upstream in the Scheldt), such as the 3M factory in Zwijndrecht. Note that the above suggests that waste-processing companies, processing waste streams from PFAS-producing companies, can be substantial PFAS sources themselves.

Compliance testing

Despite the existence of several thousand PFAS, so far EQSs have only been derived for a handful of these compounds (Ankley et al., 2021). In The Netherlands, EQSs for the maximum tolerable annual average concentration in water (AA-EQS) are available for PFOS (sum of linear and branched isomers; 0.65 ng/L in freshwater and 0.13 ng/L in seawater), PFOA (48 ng/L), and GenX (118 ng/L; advisory limit value only, not legally binding). In Supporting Information, Figure S13, the 2021 and 2022 $^{AA}C_w$s of these compounds at the selected locations are compared with their AA-EQSs. For PFOS, $^{AA}C_w$s exceed the standards at all locations, with an average factor of 5 for the inland locations, 7 for the coastal locations, and up to 23 and 34 at locations SCHAARVODDL and SASVGT in 2021, respectively. In contrast, AA-EQSs for PFOA and GenX are exceeded at none of the locations. However, new risk limits have recently been advised by the Dutch National Institute for Public Health and the Environment for a series of 26 PFAS (Rijksinstituut voor Volksgezondheid en Milieu, 2022). At least for the above three PFAS, these limit values are much stricter (i.e., 0.007, 0.3, and 10 ng/L for PFOS, PFOA, and GenX, respectively). To date, the risk limits have not been implemented by the ministry; but for approximately 60% of the included PFAS, concentrations in the inland national waters exceed these new advisory limit values.

For fish, a European EQS only exists for PFOS (EQS_{biota} = 9.1 µg/kg wet wt). In Supporting Information, Figure S14, PFOS concentrations in whole fish are tested against this value: In approximately 80% of the cases, the standard is exceeded. However, this percentage increases to 100 if the concentrations are compared with the recently proposed Dutch risk limit of 77 ng/kg wet weight (Rijksinstituut voor Volksgezondheid en Milieu, 2021). This limit value may seem very strict, but it was derived in response to the 2021 European Food Safety Authority opinion. The European Commission's scientific advisory committee (Scientific Committee on Health, Environmental and Emerging Risks) has recently endorsed the 77 ng/kg wet weight risk limit, which may result in its future adoption and subsequent failure of the majority, if not all, of Dutch fish meeting the EQS.

BAFs

In situ BAFs were calculated for roach, flounder, and flounder liver ($^{AA}C_w$ data are not available for plaice sampling [open-sea] locations). This resulted in 265 individual values, including 184 for marine fish, which are presented in Supporting Information, Table S9. These data add approximately 21% to the overall existing PFAS BAF database, which currently holds 1273 values (Burkhard, 2021). According to the quality criteria presented by Burkhard (2021), the present BAFs would all rank "high," although it should be mentioned that 35% of the values (i.e., the majority of data for PFuDA–PFteDA) are based on measured aqueous concentrations being below the official RWS reporting limits and thus are associated with high uncertainty. Averaged values for the different fish species/samples with standard deviations are presented in Supporting Information, Table S10. Generally, the present BAFs are on the same order of magnitude as other literature values (Burkhard, 2021) and demonstrate several phenomena.

First, BAFs increase with increasing length of the PFAS carbon chain (Figure 4), for both PFCAs and PFSAs (Burkhard, 2021; Kwadijk et al., 2010; Remucal, 2019). However, the increase is neither uniform nor linear: Going from PFOA to PFNA, the increase is much larger than the increase for subsequent CF_2 fragment additions (Burkhard, 2021), and BAFs for PFCAs seem to level off beyond PFuDA (although PFtDA BAFs are according to expectations based on extrapolation of the PFNA–PFuDA data). Any mechanism underlying the first observation is unknown (Burkhard, 2021), but the second may be related to the uncertainty associated with the extremely low aqueous concentrations involved and/or may point to artifacts (i.e., biased data). With an increasing carbon chain length, compound hydrophobicity, and thereby the association strength with suspended matter and dissolved organic carbon, increases. Given the total concentration measurements the current data are based on, freely dissolved concentrations (i.e., the concentrations from which BAFs should be calculated) may increasingly be overestimated as a function of carbon chain length. This typically may cause the leveling-off phenomenon observed in Figure 4 (Jonker & Van der Heijden, 2007). Still, a mechanistic explanation (e.g., specific/preferential metabolism or sorption to biomolecules) cannot be excluded. To investigate any such mechanism, dedicated experiments would be required.

Second, BAFs for freshwater roach are not significantly different (*t* tests, $p > 0.05$) from BAFs for marine flounder (Figure 4A). So far, comparing freshwater and marine species has been challenging because of limited data, and literature BAFs for marine fish were found to be either higher, lower, or equal to freshwater values (Burkhard, 2021). Third, concentrations in flounder liver generally are approximately a factor of two higher than concentrations in whole fish, which translates to an averaged difference of 0.29 log units in BAFs. Yet, because of the data variability, BAFs for liver and whole fish are not significantly different (*t* tests, $p > 0.05$), except for the data for PFteDA, PFHxS, and PFOS (Figure 4B). Finally, note that the current BAFs are all presented on a wet weight basis, which is usually done for PFAS (Burkhard, 2021), because normalizations to dry weight or lipid content generally do not (substantially) reduce data variability for PFAS (Valsecchi et al., 2021). Using the current database (which includes whole fish and fish liver dry weights and lipid contents), these normalizations were performed for the most data-rich compounds, PFOS and PFDA. Relative standard deviations for BAFs actually increased by these steps (results not shown), again supporting the use of wet weight–based values, that is, as long as the exact drivers for PFAS bioaccumulation are unknown. Whether simple normalizations to specific organism fractions such as those performed for nonionic chemicals may be applicable to the diverse chemical group of PFAS is questionable because the drivers for PFAS bioaccumulation are likely diverse as well. After all, PFAS may associate with (specific) proteins, phospholipids, and storage lipids, with the relative contribution of the association with these phases to the overall bioaccumulation potential probably depending on the type and carbon chain length of the PFAS (Sun et al., 2022). In addition, renal elimination may be important in determining bioaccumulation of certain (short carbon chain), but not all, PFAS (Sun et al., 2022).

IMPLICATIONS

The results of the current data analysis demonstrate that PFAS are ubiquitous chemicals in the Dutch national inland and coastal waters. Concentrations in these waters are relatively uniform, which indicates that the chemicals optimally distribute in the aquatic environment. Although worrying from an environmental perspective, this does simplify fate and effect modeling, a questionable benefit strengthened by rather well-defined BAFs for legacy PFAS and the observation that BAFs are similar for freshwater and marine fish.

The fact that fingerprints in the catchments of the rivers Rhine, Meuse, and Scheldt are similar suggests that emission patterns of different European countries are similar. Note that

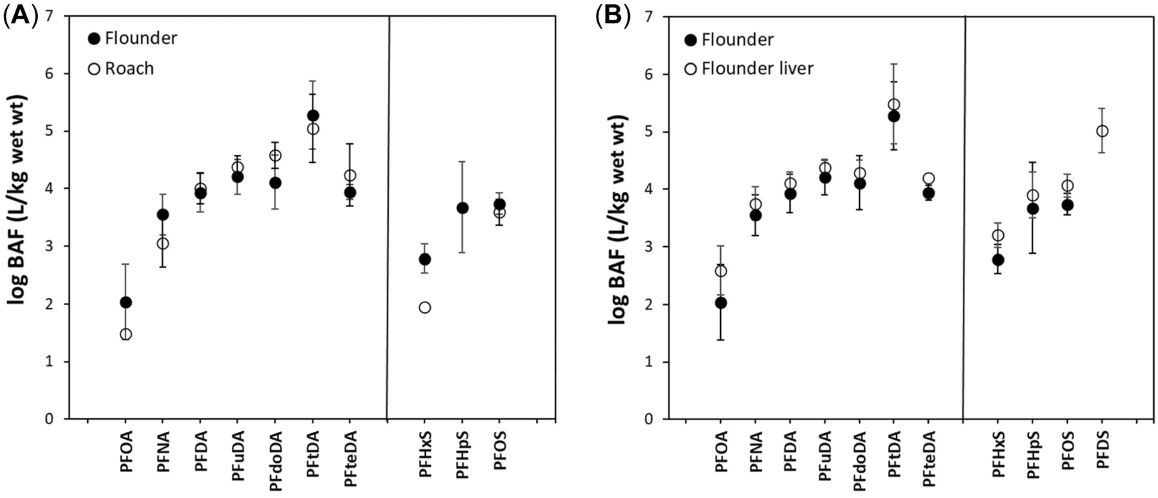

FIGURE 4: Logarithmic bioaccumulation factors (logBAF) for per- and polyfluoroalkyl substances with different carbon chain lengths. (**A**) Comparison of logBAF values for roach and flounder (both whole fish). (**B**) Comparison of logBAF values for flounder (whole fish) and flounder liver. The vertical line in each graph separates perfluorocarboxylic acids from perfluorosulfonic acids. Error bars represent standard deviations (*n* = 2–16; see Supporting Information, Table S10). PFOA = perfluorooctanoic acid; PFNA = perfluorononanoic acid; PFDA = perfluorodecanoic acid; PFuDA = perfluoroundecanoic acid; PFdoDA = perfluorododecanoic acid; PFtDA = perfluorotridecanoic acid; PFteDA = perfluorotetradecanoic acid; PFHxS = linear perfluorohexanesulfonic acid; PFHpS = perfluoroheptanesulfonic acid; PFOS = perfluorooctanesulfonic acid; PFDS = perfluorodecanesulfonic acid.

the Rhine originates in Switzerland and flows through Germany before entering The Netherlands, whereas the Meuse and Scheldt originate in France, flow through Belgium, and then enter The Netherlands. Because fingerprints at locations close to the North Sea are similar to those at locations where the rivers enter The Netherlands, the overall Dutch emission pattern seems to be similar as well. Moreover, the absence of clear and recent time trends implies that PFAS concentrations have not increased substantially over the past years but have not decreased considerably either (excepting the PFOS and PFOA cases discussed). Generally, and particularly since 2016, the concentrations, including those of PFOS and PFOA, have been relatively to very stable. This demonstrates that the effectiveness of (inter)national chemical management actions aiming to ban PFOS and PFOA from the environment has been limited so far. Considering the absence of (recent) time trends and the fact that PFOS ${}^{AA}C_w$s exceed the EQSs at all locations, PFOS concentrations are not expected to meet the standards in a few years from now. This concerns both aqueous concentrations and those in fish, all the more so because standards are likely to be lowered in the (near) future. This has far-reaching implications in terms of the European Water Framework Directive, which dictates that member states should achieve a good chemical water quality status for all natural waters by 2027. Because the present study shows that PFAS concentrations in Dutch national waters are primarily driven by riverine input from abroad, the desired chemical status may only be acquired through international actions.

Supporting Information—The Supporting Information is available on the Wiley Online Library at https://doi.org/10.1002/etc.5846.

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Data Availability Statement—The original monitoring data can be found at https://waterinfo.rws.nl. All results of the data analysis are presented in the Supporting Information. Any other data are available on request from the corresponding author.

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