Evaluation and prediction of anthropogenic impacts on long-term multimedia fate and health risks of PFOS and PFOA in the Elbe River Basin

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Abstract

Perfluorooctane sulfonic acid (PFOS) and perfluorooctanoic acid (PFOA) have aroused great concern owing to their widespread occurrence and toxic effects. However, their long-term trends and multimedia fate remain largely unknown. Here, we investigate the spatiotemporal characteristics and periodic oscillations of PFOS and PFOA in the Elbe River between 2010 and 2021. Anthropogenic emission inventories and multimedia fugacity model were developed to analyse their historical and future transport fates and quantify related human risks in each medium for the three age groups. The results show that average PFOS and PFOA concentrations in the Elbe River were 4.08 and 3.41 ng/L, declining at the annual rate of 7.36% and 4.98% during the study period, respectively. Periodic oscillations of their concentrations and mass fluxes were most pronounced at 40–60 and 20–40 months. The multimedia fugacity model revealed that higher concentrations occurred in fish (PFOS: 14.29, PFOA: 0.40 ng/g), while the soil was their dominant sink (PFOS: 179, PFOA: 95 tons). The exchange flux between water and sediment was the dominant pathway in multimedia transportation (397 kg/year). Although PFOS and PFOA concentrations are projected to decrease by 22.41% and 50.08%, respectively, from 2021 to 2050, the hazard quotient of PFOS in fish is a low hazard. This study provides information for the assessment of PFOS and PFOA pollution in global watersheds and the development of related mitigation policies, such as banning fish predation in polluted rivers, to mitigate their risks.

Keywords

PFOS and PFOA; Environmental modelling; Emission inventory; Risk assessment; Long-term prediction; Elbe River Basin

1. INTRODUCTION

Per- and polyfluoroalkyl substances (PFAS) have been identified as emerging contaminants with great importance and have been recognized to be ubiquitous in the environment (Evich et al., 2022). Long-chain perfluoroalkyl acids (PFAAs), particularly perfluorooctane sulfonic acid (PFOS) and perfluorooctanoic acid (PFOA), have been frequently used historically and attracted the focus of considerable research for two decades (Kirkwood-Donelson et al., 2023). PFOS and PFOA have been used in the manufacturing of a wide range of industrial and household materials with nonstick, repellent, and surfactant properties for more than 70 years (Pistocchi and Loos, 2009; Sakurai et al., 2010). The widespread use of PFOS and PFOA has resulted in their ubiquitous occurrence in various environmental media, including the air (Morales-McDevitt et al., 2021), soil (Bolan et al., 2021), water (Crone et al., 2019), and sediment (Meng et al., 2021). PFOS and PFOA have been recognised as highly toxic compounds because of their association with liver diseases, cardiovascular problems, birth defects, and other adverse health effects (Rogers et al., 2021; Schulz et al., 2020). Moreover, the IARC has identified PFOA and PFOS as carcinogenic and possibly carcinogenic, respectively, to humans (Zahm et al., 2024). Consequently, owing to their extensive distribution and toxicity, there is an urgent need to assess and predict the pollution levels of PFOS and PFOA in various environmental media to support the implementation of more effective treatment measures.

The Elbe River Basin in Germany is a dynamic environment with frequent interactions between human activity and nature (Huang et al., 2021). The Elbe River Basin has relatively high PFOS and PFOA concentrations, which may pose a significant threat to the biota and inhabitants (Cui et al., 2009; Zhao et al., 2015). Because PFOS and PFOA are persistent, it is essential to analyse their long-term pollution tendencies in such large watersheds. However, previous studies have focused on the short-term spatiotemporal distribution of their contamination levels because chronic measurements were not available (Joerss et al., 2019; Zhao et al., 2015). Moreover, owing to the potentially toxic effects of PFOS and PFOA on ecosystems and humans,

investigating the characteristics of PFOS and PFOA in the Elbe River and quantifying the risks of long-term exposure could provide an important scientific basis and insight for developing policies for preventing and controlling PFOS and PFOA in global river systems.

Regarding environmental migration, PFOS and PFOA are primarily emitted into water and the atmosphere, with subsequent transport to soil, sediment, and organisms, where they pose toxic exposure risks (Xu et al., 2013). Hence, there is an urgent need to construct a multimedia fugacity model that includes the major compartments to characterise the fate and transport of PFOS and PFOA in the environment. The multimedia fugacity model has been used in many previous regional-scale studies to reveal the multimedia transport of persistent organic pollutants and their future destinations, such as polycyclic aromatic hydrocarbons (Du et al., 2020; Nam et al., 2021; Nie et al., 2024). However, there are comparatively few studies on the multimedia fate of PFAS and associated changes over long-term periods (Chen and Yuan, 2009). Although PFOS and PFOA concentrations were shown to decrease in the Elbe River Basin in 2006, 2007, and 2011 (Ahrens et al., 2009a; Ahrens et al., 2009b; Zhao et al., 2015), the long-term prediction of the trends in PFOS and PFOA in environmental multimedia within the Elbe River Basin has been unexplored. Therefore, simulating the historical and future multimedia fates and risks of PFOS and PFOA can facilitate a comprehensive exploration of the contamination status and guide governments, organisations, and individuals in developing early sustainable development plans to safeguard human and ecosystem health.

To advance strategies for reducing PFOS and PFOA contamination in the Elbe River Basin, historical multimedia fugacity models were constructed in this study to characterise the PFOS and PFOA contaminant levels in the air (air and particulate matter), water (water, fish, and suspension), soil, and sediment. Based on the projections of emissions and environmental parameters, this study simulated the PFOS and PFOA concentrations in multimedia for 2030, 2040, and 2050 using a fugacity model. The major objectives were to: (1) investigate the temporal variation and spatial distribution of PFOS and PFOA concentrations and mass fluxes in the water phase

along the Elbe River from 2010 to 2021; (2) develop a multimedia fugacity model to characterise the levels of PFOS and PFOA in various media in the Elbe River Basin; and (3) predict the concentrations and health risks of PFOS and PFOA in environmental multimedia in the Elbe River Basin for the next three decades. This study provides new perspectives for preventing, mitigating, and controlling PFOS and PFOA pollution in global watersheds and anthropogenically intensive environments. Our findings can contribute to the understanding of the transport fate of PFOS and PFOA in watersheds, particularly the total fluxes between multimedia, which will help governments assess the status of pollution and develop relevant mitigation policies.

2. METHODS AND MATERIALS

2.1 Data source

From January 2010 to December 2021, 908 surface water samples were collected to determine the PFOS and PFOA concentrations in the Elbe River. To implement the European Water Framework Directive (Directive, 2000), the concentrations of various trace substances in the Elbe River were measured to evaluate the health risks of these trace substances in humans. The measurements were obtained from the Elbe Data Information System (FIS) (www.fgg-elbe.de, accessed May 2023). Different stations were located upstream and downstream of the Elbe River in Germany. In this study, a concentration indicator was used to assess the average PFOS and PFOA pollution levels in the Elbe River. Mass flux was used to evaluate the total contamination load, which was obtained as the concentration multiplied by the streamflow (Viticoski et al., 2022). Detailed information regarding PFOS and PFOA is provided in **Table S1**.

According to the guidelines of the Federal Government and the Federal Institute for Hydrology (BfG) (www.bafg.de, accessed October 2023), the water samples were collected and sent to the Betriebsgesellschaft für Umwelt und Landwirtschaft (BfUL), Landesbetrieb für Hochwasserschutz und Wasserwirtschaft Sachsen-Anhalt (LHW), Nds. Landesbetrieb für Wasserwirtschaft, Küsten- und Naturschutz (NLWKN) - Bst. Lüneburg, NLWKN - Bst. Stade, and the Institut für Hygiene und Umwelt, Hamburg

for measuring. The specific sampling, pretreatment, and testing procedures are described in **Part S1**. Wavelet analysis, Mann–Kendall trend detection, and health risk assessment are described in **Part S2-S4**.

2.2 Multimedia fugacity model

The level III fugacity model has been widely applied to depict the dynamics of organic chemicals in multimedia (Li et al., 2020a). It was developed to optimise the characterisation of the transport fate of PFOS and POFA in multimedia for the Elbe River Basin. The environments were categorised into four compartments: air, water, soil, and sediment (SED), with airborne particulate matter (PM), waterborne suspended matter (SS), and fish as the three submedia. The optimised multimedia fugacity model framework and major transmission processes of PFOS and PFOA in each environmental compartment are shown in **Fig. 1**.

The multimedia fugacity model was established by employing the fugacity capacity (Z, mol/m$^3 \cdot$Pa) of the seven bulk compartments and the transport parameter (D, mol/h$\cdot$Pa) between the bulk compartments. The fugacity (f, Pa) was imported to generate the mass balance equation for the main media based on the entry and exit of the compartments (**Table S2**). The concentrations (C, mol/m^3) and mass fluxes (T, mol/h) of PFOS and PFOA were calculated as follows:

$$C_x = Z_x \times f_x \quad (1)$$

$$T_x = D_x \times f_x \quad (2)$$

where x represents the medium in which PFOS and PFOA are located. The codes for air, water, soil, and sediment are 1, 2, 3, and 4, respectively. Detailed definitions and calculations of all process parameters are presented in **Tables S3** and **S4** (Kong et al., 2018b). The fugacity model contained 68 environmental and material parameters from previous studies on the Elbe River, data published by German institutions, and geographical calculations (**Tables S5** and **S6**) (Kong et al., 2018b; Li et al., 2020b; Zhou et al., 2022). The transmission fates of PFOS and PFOA in the Elbe River Basin from 2010 to 2021 were modelled using the above parameters. Furthermore, this study

predicted the multimedia transmission fluxes for 2030, 2040, and 2050 based on the projected emission inventories and environmental parameters predicted using the autoregressive integrated moving average (ARIMA) model (**Table S7**). A detailed description of the proposed methodology is given in **Part S5**.

The Morris method was employed to obtain the parameter sensitivity analysis results by altering the parameters individually to observe their corresponding effects on the selected state variables (Kong et al., 2018b). Relevant parameter ranges were collected to conduct uncertainty analysis using Monte Carlo simulations. The Monte Carlo simulations used 10000 parameter combinations randomly sampled from predefined statistical distributions of the 68 parameters. Detailed descriptions of the sensitivity and uncertainty analyses are provided in **Part S6** and **S7**.

2.3 Emission inventories of PFAS

The historical global releases of PFOS equivalents from consumer use and disposal have been estimated to be 42,000 tons in water and 235 tons in air (Paul et al., 2009). Moreover, sewage plant effluent and sludge are recognised as major sources of PFOS and PFOA in river systems, whereas relatively small amounts of precursor residues volatilise into the atmosphere (Huset et al., 2008; Sinclair and Kannan, 2006). Previous studies estimated the emission density of PFOS from sewage plants to be up to 1000 mg/km^2·d in the eastern coastal region of China in 2010 (Xie et al., 2013). Accordingly, this study calculated the emissions of PFOS and PFOA in the Elbe River Basin based on effluent and sludge produced from sewage plants and the PFOS and PFOA concentrations found therein. Meanwhile, PFOS, PFOA, and their precursor compounds are released into the air during wastewater treatment, and atmospheric emissions must be included in the total emissions. Therefore, the PFOS and PFOA emissions in the Elbe River Basin were calculated as follows (Ahrens et al., 2011; Xie et al., 2013; Zhang et al., 2013):

$$f(Q, M, C_{water}, C_{sludge}) = (Q \times C_{water} \times 10^{-3} + M \times C_{sludge})/A \quad (3)$$

$$\partial = 0.0452 \times f(Q, M, C_{water}, C_{sludge}) \quad (4)$$

$$ED = f(Q, M, C_{water}, C_{sludge}) + \partial \quad (5)$$

where the units of $f(Q, M, C_{water}, C_{sludge})$ are the discharges of PFOS and PFOA in the Elbe River Basin, ∂ is the emissions of PFOS and PFOA and their precursor compounds to air during wastewater treatment, and *ED* is the total emissions (mg/km^2·d). *Q* is the wastewater plant discharge flow per state in Germany (m^3/d), which includes industrial and municipal wastewater. *M* was the dry sludge production of a wastewater plant per state in Germany (t/d). *A* represents the regional area of the wastewater treatment plant service (km^2). C_{water} and C_{sludge} are the PFOS and PFOA concentrations in the terminal effluent (ng/L) and sludge (ng/g dry weight), respectively (**Table S8**).

The effluent discharges and sludge discharges from sewage plants in the Elbe River Basin were obtained from the Statistische Ämter des Bundes und der Länder in Germany (www.statistikportal.de, accessed August 2023). However, the time scale of the data does not cover the entire period of 2010–2021. To solve this problem, we applied an estimation method based on population and gross per capita product to calculate the PFOS and PFOA emissions in the Elbe River Basin for years with missing discharge data. Xie et al. (Xie et al., 2013) and Chen et al. (Chen et al., 2022) applied this method to predict PFOS emissions in eastern China in 2010 and 2013, respectively. The use of consumer goods has been suggested to be one of the main sources of PFOS and PFOA; thus, population density (*PD*) and per capita gross domestic product (*GDP*) are considered important factors in emission density (Pistocchi and Loos, 2009; Xie et al., 2013). The optimisation of the model was based on the cftool function in MATLAB, and the optimised model for the emissions of PFOS and PFOA in water was as follows:

$$f(PD, GDP) = a \times PD^b \times GDP^c + p_1 \times PD^2 + p_2 \times PD \times GDP + p_3 \times GDP^2 \quad (6)$$

where *PD* was the population density of each German state and *GDP* was the gross domestic product per capita of each state (units: km^{-2} and euro, respectively). Moreover, the emission inventories of PFOS and PFOA in the Elbe River Basin in 2030, 2040, and 2050 were predicted by the projected values of *PD* and *GDP* in these years based on f(*PD, GDP*). The historical and projected values of *PD* and *GDP* data were derived

from the Statistische Ämter des Bundes und der Länder in Germany (www.statistikportal.de, accessed August 2023) (**Table S8**). The linear fit of $f(Q, M, C_{water}, C_{sludge})$ and $f(PD, GDP)$ performed well (R^2: PFOS: 0.9862, PFOA: 0.9931) (**Fig. S1**). The detailed emission inventory estimation process is described in **Part S5**.

3. RESULTS

3.1 Spatial distributions of PFOS and PFOA

Fig. 2 shows the average PFOS and PFOA concentrations at the 11 stations along the Elbe River from 2010 to 2021. The mean concentration ranges for PFOS and PFOA detected at individual stations were 0.8–5.9 ng/L and 1.6–6.0 ng/L, respectively (**Table S9**). Geographically, the upstream stations (S1, S2, and S3) had high PFOS concentrations, while the downstream stations (S10 and S11) had low PFOS concentrations. For example, the highest PFOS concentration occurred at S2 (5.9 ng/L), whereas site S11 had the lowest concentration (0.8 ng/L). On the contrary, the PFOA concentrations at the upstream stations (S1: 1.7 ng/L, S3: 1.6 ng/L) were relatively low, whereas the highest average concentration (6.0 ng/L) occurred at site S6. The spatial distributions of PFOS and PFOA concentrations at the stations along the Elbe River during the monitoring period showed different distribution characteristics, which might be related to differences in economic development and the intensity of anthropogenic activities along the river.

3.2 Decreasing trends and seasonal variations of PFOS and PFOA

As shown in **Fig. 3 (a–d),** the annual average concentrations and mass fluxes of PFOS and PFOA in the Elbe River showed declining trends from 2010 to 2021. Notably, the annual average PFOS and PFOA concentrations in the study period were 4.08 and 3.41 ng/L (range: 0.015–41 and 1-25 ng/L, respectively). The mean mass fluxes of PFOS and PFOA between 2010 and 2021 were 1131 and 1851 μg/s (range: 3–14480 and 8–22296 μg/s, respectively) (**Table S10**). Furthermore, the annual average concentrations and mass fluxes of PFOS and PFOA peaked during 2012–2014 and

maintained decreasing trends after 2015. According to the Mann–Kendall trend detection, PFOS and PFOA concentrations and mass fluxes displayed significant ($p < 0.05$) decreasing trends from 2010 to 2021 (slopes of PFOS: -0.0062 and -1.36, and PFOA: -0.0030 and -2.90, respectively) (**Table S11**). The overall decrease in PFOS and PFOA contamination in the Elbe River during this long-term period can be attributed to the continuous implementation of relevant restriction policies.

Fig. 3(e–h) shows violin boxplots of the monthly average concentrations and mass fluxes of PFOS and PFOA. Over this period, the mean PFOS concentrations in the Elbe River in hot seasons (summer: 4.81 ng/L and autumn: 5.28 ng/L) were relatively higher than those in cold seasons (spring: 2.98 ng/L and winter: 3.36 ng/L) (**Table S12**). The seasonal trends were consistent with those proposed in previous studies (Wang et al., 2016; Zhao et al., 2015) and may have been influenced by changes in the flow of the Elbe River. Higher winter snowmelt in Europe leads to higher streamflow, and concentrations may be diluted to lower levels during the cold seasons (**Fig. S2**). Comparatively, the highest mass flux of PFOA in the Elbe River occurred in spring (1458 μg/s), which may be related to the following reasons. Primarily, more pollution enters the aquatic environment due to rain and snow flushing during the cold season. Lower temperatures in the cold season slow the degradation of precursors, leading to the accumulation of pollutants in receiving waters (Kwok et al., 2010; Taniyasu et al., 2013).

Compared to the worldwide range of PFOS and PFOA concentrations in water, the PFOS and PFOA concentration ranges in the Elbe River from 2010 to 2021 were lower than those of the Pra and Kakum Rivers in Ghana (range: 77.2–275 and 1.78–321 ng/L, respectively) (Essumang et al., 2017) and the Haw River in the USA (range: ND–110.8 and 0.7–133.3 ng/L, respectively)(Pétré et al., 2022). The PFOS and PFOA concentration ranges in the Elbe River during this period were higher than those in certain water bodies, such as Gironde estuary in France (range: 0.02–0.07 and 0.4–1.9 ng/L, respectively) (Munoz et al., 2019) and Rhine River in Germany (range: 1.41–6.38 and 0.61–3.44 ng/L, respectively) (**Table S13**) (Möller et al., 2010). Compared with the latest EPA limits for PFOS and PFOA (4 ng/L and 4 ng/L, respectively) (EPA, June 23,

2022), the PFOS concentrations in the Elbe River were higher than the limit. Globally, the PFOS and PFOA concentrations in the Elbe River were relatively high during the study period. In addition, basin-scale emissions of PFAS are an important indicator of pollution. Although the Elbe River has slightly higher PFOS and PFOA concentrations, the emission of PFAS from the Elbe River (94 kg/year) is lower than that from most water bodies worldwide (**Table S14**). Examples include the Rhine River in Germany (2003 kg/year) (Li et al., 2023) and the Cape Fear River in the USA (1476 kg/year) (Pétré et al., 2022). However, the PFAS emissions from the Elbe River were higher than those from the Hun River in China (15 kg/year) (Podder et al., 2021) and the Vantaanjoki River in Finland (5.2 kg/year) (Junttila et al., 2019). Therefore, it is more meaningful to evaluate basin-scale PFAS contamination from the dual perspective of concentration and emission. Consequently, adequate attention and more effective measures are required to mitigate PFOS and PFOA in the Elbe River.

3.3 Periodic oscillation of PFOS and PFOA pollution

Wavelet analysis was used to identify the periodic variations of PFOS and PFOA concentrations and mass fluxes in the Elbe River between 2020 and 2021. The wavelet real part of the concentrations and mass fluxes of PFOS and PFOA are shown in **Fig. 4**. The left axis reflects the time scale of the wavelet variation, and the lower axis consists of 144 months from 2010 to 2021. When the PFOS and PFOA concentrations and mass fluxes tend to increase, the wavelet real value was > 0, and the contour plot showed warm colours, and vice versa. Colour alternation was employed to determine the periodic oscillations of PFOS and PFOA concentrations and mass fluxes. As shown in **Fig. 4(a)**, the periodic oscillations of the PFOS concentration in the Elbe River are 20–40 and 40–60 months. The values of the wavelet modulus were higher on the time scale of 40–60 months, indicating that the periodic oscillations of the PFOS concentrations were more significant on this scale (**Fig. S3**). Meanwhile, the wavelet variance in PFOS concentration intuitively reflects the distribution of oscillations, and the value of wavelet variance for PFOS concentration reached an extreme value at a time scale of 54 months, which reveals the dominance of periodic oscillations at 40–60 months (**Fig.**

S4). Therefore, the PFOS concentrations exhibited the most significant periodic oscillations on a scale of 40–60 months. Furthermore, periodic oscillations in the PFOS mass fluxes were largely consistent with their concentrations. In contrast, the periodic oscillations of the concentration and mass flux of PFOA fluctuated less and were mainly distributed over a timescale of 20–40 months. Therefore, the most significant periodic oscillations for the concentrations and mass fluxes of PFOS and PFOA occurred at timescales of 40–60 months and 20–40 months, respectively.

Moreover, the periodic oscillations may be influenced by chemical industry production, consumption of fluorinated substances, and natural conditions. Consequently, wavelet analysis corroborates that the concentrations and mass fluxes of PFOS and PFOA in the Elbe River from 2010 to 2021 have periodic variations and are probably influenced by multiple factors. For example, the water temperature of the Elbe River from 2010 to 2021 was broadly consistent with periodic variations in PFOS and PFOA, which are the main factors influencing the PFAS concentration in the environment and perhaps the main contributors to the cycle of change (**Fig. S5**) (Podder et al., 2021; Zhao et al., 2013). Moreover, human activities and climatic conditions also contribute to periodic oscillations in PFOS and PFOA (Göckener et al., 2022; Kong et al., 2018a).

3.4 Multimedia simulation and prediction

Based on the emission inventory and environmental parameters, the PFOS and PFOA concentrations in the seven compartments of the Elbe River were simulated using a multimedia fugacity model, as shown in **Fig. 5**. According to previous studies, this criterion of model reliability is that the deviation of the modelled values from the measured values should be < 0.7 log units (Kong et al., 2017). The measured values used in the model validation were obtained primarily from the data portal and references (**Table S15**). The overall average validation showed a positive simulation performance for PFOS and PFOA for each medium in the Elbe River (**Table S15**) (**Fig. 5(a–b)**). For instance, the average modeled PFOS and PFOA concentrations in water were 3.36 ng/L and 2.82 ng/L from 2010 to 2021, respectively, which were comparative to measured

values of PFOS (4.08 ng/L) and PFOA (3.41 ng/L).

Regarding individual media, the highest modeled PFOS concentrations in the Elbe River were found in fish (1.5×10^{4} ng/L), followed by soil (1.17×10^{4} ng/L), sediment (7.98×10^{3} ng/L), suspended matter (3.76×10^{2} ng/L), water (3.36 ng/L), particulate matter (4.29×10^{-4} ng/L), and air (2.55×10^{-8} ng/L) (**Table S16**). In contrast, the media with the three highest modeled PFOA concentrations in the period differed slightly from those of PFOS, which were soil (6.22×10^{3} ng/L), sediment (2.73×10^{3} ng/L), and fish (4.23×10^{2} ng/L) (**Table S17**). This was because the bioconcentration factor (BCF) of PFOS (2990 L/g) was higher than that of PFOA (100 L/g). Regarding the total mass of PFOS and PFOA in each compartment, soil had the highest modelled mass (PFOS: 179 tons, PFOA: 95.2 tons) (**Fig. 5(c–d)**), followed by sediment, water, and fish. This result implies that soil and sediment are the main sinks for the environmental fate of PFOS and PFOA.

Fig. 5(e–f) show a comparison of the average modelled and measured values of PFOS and PFOA in water from 2010 to 2021 and the predictions for 2030, 2040, and 2050. The results indicate that the modelled and measured values of PFOS and PFOA concentrations are generally in acceptable agreement for almost all years except 2021. The lower measured PFOS concentrations in 2021 were likely caused by the lockdown impact of the COVID-19 outbreak. Because the electroplating and fire protection industries have been recognised as major contributors to PFOS in many studies, the COVID-19 outbreak caused some related industries in Germany to shut down, which led to low PFOS concentrations in the environment (Li et al., 2023; Lian et al., 2023).

From 2010 to 2021, the annual average simulated PFOS and PFOA concentrations in water decreased by approximately 1.87% and 2.23% annually, respectively. In contrast, predicted PFOS and PFOA contamination levels decline from 2.82 and 2.23 ng/L in 2021 to 2.19 and 1.11 ng/L in 2050 by 7.45% and 16.74% per decade, respectively. Additionally, the PFOA concentrations are significantly lower than those of PFOS in 2050, mainly due to the lower emissions of PFOA (**Table S7**). Moreover, the decreasing trends in the modelled concentrations and total masses of PFOS and PFOA in air, fish, soil, and sediment in the Elbe River Basin were generally similar to

those in water (**Fig. S6**). Taking the most important sink as an example, the masses of PFOS and PFOA in the soil decreased from 194 to 166 t and 101 to 89 tons from 2010 to 2021, respectively. The predicted total masses of PFOS and PFOA in soil decrease by 5.72% and 7.12% per decade, respectively. The uncertainties in the PFOS and PFOA concentrations in each medium over this period are presented in **Figs. S7** and **S8**.

3.5 Multimedia transport fluxes of PFOS and PFOA

As shown in **Fig. 6**, the transport fluxes of PFOS and PFOA are calculated using the multimedia fugacity model by multiplying the fugacity by the transfer rate coefficients. The total average inputs of PFOS and PFOA in the Elbe River Basin from 2010 to 2021 are 518 kg/year. Explicitly, the total contributions from human discharges and upstream inflows are 415 kg/year and 60.7 kg/year, respectively, which are markedly higher than the total air inflow (41.7 kg/year) (**Fig. 6(a)**). Among the multimedia transport processes, sedimentation had the highest total PFOS and PFOA fluxes (397 kg/year). The total degradation rate in the sediment was 364 kg/year and entered the water with 32.6 kg/year for resuspension. In addition, air-soil interaction fluxes through wet-dry deposition were the most important pathways for PFOS and PFOA to enter the soil. Although the enrichment of PFOS and PFOA for fish was lower than that for most of the other fluxes (0.072 kg/year), their concentrations in fish were relatively high (**Fig. 5(a–b**)). Consequently, the main export pathways for PFOS and PFOA in the Elbe River Basin were sediment degradation (364 kg/year) and outflow from water (84.6 kg/year). The migration patterns observed in this study were consistent with the results of previous studies (Kong et al., 2018a; Sánchez-Soberón et al., 2020). In addition, the total fluxes of PFOS and PFOA in different environmental media decreased from 2010 to 2021 (**Table S18** and **S19**).

To obtain a more intuitive representation of the interaction between the media, the average transport fluxes of PFOS and PFOA from 2010 to 2021 are presented in **Fig. 6(b–c)**. The highest flux of PFOS was from the transport of water and sediment (247 kg/year), followed by emissions into the water and degradation in the sediment (196 kg/year). Meanwhile, the air-soil interaction fluxes of PFOA (63 kg/year) were almost

three times higher than those of PFOS (23 kg/year). Furthermore, PFOS degradation in the sediment was relatively higher than that of PFOA (175 kg/year) during this period, which was due to the higher content of PFOS in the sediment.

Fig. 6(d–e) show the projected multimedia fluxes of PFOS and PFOA in 2050. Although the transport pattern in 2050 is essentially similar to that of previous periods, significant decreases in the fluxes of PFOS and PFOA occurred in the Elbe River. For example, the average annual emissions of PFOS and PFOA are expected to decrease from 176 and 193 kg/year in 2021 to 127 and 89 kg/year in 2050, respectively. Meanwhile, the sedimentation of PFOS and PFOA declines by 61% and 35%, respectively, from the previous period to 2050. The predicted transport fluxes of PFOS and PFOA are shown in **Fig. S9–10**.

3.6 Human health risk assessment

Fig. 7(a) shows the average hazard quotient (HQ) values of PFOS and PFOA in the air, soil, water, and fish in the Elbe River Basin for children, teenagers, and adults from 2010 to 2021. The HQ values associated with PFOS in fish were much higher than those in the remaining media (children: 0.83, teenagers: 0.49, adults: 0.22), followed by water, soil, and air, in decreasing order (**Tables S20–23**). The HQ values of PFOS in fish were higher than 0.1 for the three age groups, indicating that PFOS in fish is a low hazard for humans. Moreover, the risk of PFOS in fish for children was approximately twice that for teenagers and adults. The reason for this phenomenon is that children have weaker immunity, lower body weight, and are exposed to the strongest health risks of PFOS in their early developmental years (Li et al., 2023). The HQ value of PFOS in fish for children in 2013 was 0.95, approaching the threshold criterion of moderate hazard (standard value: 1) (**Table S22**). In contrast, PFOS in air, water, and soil, and PFOA in the four media posed negligible health risks to the three age groups, as these HQ values were < 0.01 for the three age groups. The exposure risk of PFOS was higher than that of PFOA because PFOS has a lower *Rfd*, indicating that it is more hazardous. Therefore, more attention should be given to the health risks associated with PFOS in fish.

Fig. 7(b) shows the decreases in the average annual HQ values for PFOS and PFOA in air, water, fish, and sediment for children, teenagers, and adults between 2010, 2021, and 2050. Specifically, the HQ values of PFOS in multimedia decreased in the following order: fish and water (34.77%), air (20.60%), and soil (19.21%) between 2010, 2021, and 2050. The decline in the HQ values of PFOA in each medium was more dramatic, with fish and water (60.48%), air (26.59%), and soil (24.83%). Thus, the risk of human exposure to PFOS and PFOA in the Elbe River Basin has gradually diminished. However, the potential health risks to humans caused by PFOS in fish have not been fundamentally mitigated. In 2050, the predicted HQ values for PFOS in fish are 0.54, 0.32, and 0.14 for children, teenagers, and adults, respectively. Despite this decline, the human risk of exposure to PFOS in fish remains low. The predicted HQ values are presented in **Fig. S11**.

4. DISCUSSION

4.1 Impacts of anthropogenic activities on PFAS

This study revealed that PFOS and PFOA contamination in the Elbe River decreased by 6.34% and 4.98% annually, respectively, from 2010 to 2021 (**Fig. 4**). The downward trend**s** in PFOS and PFOA are probably related to the European Chemicals Regulation placing PFOS and PFOA on the candidate list of substances of very high concern, leading to the phase-out of PFAS by relevant manufacturers. Although PFOS and PFOA concentrations (0.71 and 2.01 ng/L in 2021, respectively) in the Elbe River Basin continue to decrease over the long-term, they remain higher than the standard (0.65 ng/L) set by the European Water Framework Directive. This may be due to the slow degradation of PFOS and PFOA, resulting in a relatively high stock in the Elbe River. Consequently, to maintain acceptable PFOS and PFOA concentrations, effective measures should be taken to reduce these concentrations and enhance subsequent management of PFOS and PFOA in the Elbe River.

Furthermore, the PFOS and PFOA concentrations in soil and sediment, which are the most important sinks for PFOS and PFOA in the environment, will decrease from

2021 to 2050 (soil: 12.88% and 19.36%; sediment: 22.41% and 50.08%, respectively). The highest PFOS concentration from 2010 to 2021 was found in fish (1.50×10^4 ng/L) (**Fig. 5**). Meanwhile, the risk of PFOS in fish (HQ:0.95) is predicted to remain relatively high until 2050 (HQ: 0.54), which might be related to the higher BCF of fish for PFOS (BCF: 2993 L/g) (Hung et al., 2020; Quinete et al., 2009). The enrichment of PFOS contamination from other media to fish through the transport process also contributed to a slow decline in the risk of PFOS exposure in fish. Therefore, more measures are urgently needed to accelerate the decline of PFOS in river systems to reduce health risks to fish. In addition, because emissions are prominent in the sensitivity analysis, our predictions were emission-based and combined with certain environmental parameters. Future changes in control policies, fluctuations in environmental conditions, and improvements in treatment technologies could mitigate PFOS and PFOA in the environment.

4.2 Appropriate remediation pathways

Because the PFOS and PFOA concentrations in individual media remained relatively high, how to remediate the PFOS and PFOA contamination status in these media requires further discussion. Primarily, emissions were the most important sources of PFOS and PFOA pollution in the Elbe River Basin, accounting for 84% of the total input to the system. The model sensitivity analysis showed that the variation in PFOS and PFOA concentrations in each medium caused by changes in the remaining factors was markedly lower than that of the emissions. For instance, a 10% decrease in emissions caused the PFOS and PFOA concentrations in water to decrease by 8.12% and 8.6%, respectively (**Fig. S12**). Therefore, source control is the most effective measure for managing PFOS and PFOA in the Elbe River Basin.

Despite source control, PFOS and PFOA pollution can be remediated using various approaches, including water treatment, sediment removal, soil remediation, and biological management. Currently, the major methods for treating PFOS and PFOA contamination in water include granular activated carbon (GAC), nanofiltration, and the photo-Fenton process. Furthermore, water treatment with GAC and sediment

removal are the most cost-effective ways to remove PFOS and PFOA from the environment (5.94×10^{3}–1.29×10^{4} and 6.29×10^{3}–1.57×10^{4} dollars/g, respectively), which are considerably lower than those of other governance methods (1.82×10^{4}–6.04×10^{8} dollars/g) (**Table S24**). Sediment removal frequently destroys the benthic environment, and benthic organisms require > 15 years to recover (Waye-Barker et al., 2015). Therefore, sediment removal is unsuitable for large-scale applications and more attention should be paid to finding a balance between protection and destruction (Richir et al., 2021).

Cost is not the only criterion in assessing the merits of treatment measures. From a health risk perspective, even if the cost of biological management (fish) is high (5.15×10^{4}–1.03×10^{5} dollars/g), it is still necessary to catch fish species with high bioconcentration factors of PFOS and PFOA to minimise their potential harm to humans. Meanwhile, the ban on fish predation in the Elbe River during the remediation period will help reduce the intake of PFOS and PFOA (Pickard et al., 2022). From the perspective of environmental stocks, it is necessary to prioritise the mitigation of PFOS and PFOA in soils with the highest total mass. Therefore, it is necessary to comprehensively consider various factors when selecting governance methods to ensure satisfactory results.

5. CONCLUSION

To assess the PFOS and PFOA pollution in the Elbe River Basin, this study constructed the long-term emission inventory, established a multimedia fugacity model to determine their fate in the environment, and projected the exposure to human risks from 2010 to 2050. The main implications are as follows:

(1) Due to the strict phase-out policy, the PFOS and PFOA concentrations in the Elbe River showed a long-term decreasing trend from 2010 to 2021. The periodic oscillations of their concentrations and mass fluxes were most pronounced at 40–60 and 20–40 months.

(2) The multimedia fugacity model indicated that the PFOS and PFOA concentrations were higher in the fish and soil, while the soil was their dominant sink.

Meanwhile, the transport flux between water and sediment was most significant.

(3) The health risk of PFOS in fish from 2010 to 2050 was determined as low hazard, whereas PFOS and PFOA in other media had no adverse effects on human health.

In summary, policymakers need to increase efforts to limit emissions and accelerate the development of alternatives to facilitate governance of PFOS and PFOA. It is also critical to emphasise the implementation of multimedia co-management of fish and soil remediation, and the establishment of a comprehensive monitoring program to address exposure risks.

AUTHOR CONTRIBUTIONS

Ruifei Li and Xu Zhu designed the original draft and conducted the analysis, methodology, and writing. Yu Luo, Yu Li, Zhenyu Wang, and Wenyu Yang contributed to conceptualization and software. Jin Zhang and Hui Li contributed to the reviewing and editing.

ACKNOWLEDGMENTS

The authors would like to thank the River Basin Community Elbe (Flussgebietsgemeinschaft Elbe) for providing the dataset. This work was jointly supported by the National Key Research and Development Plan (2023YFE0115200), the National Natural Science Foundation of China (Grant Nos. 42307470, 42125706, 42077401, 42077156), Shanghai Pujiang Program (23PJ1402900) and the Chenguang Program of Shanghai Education Development Foundation and Shanghai Municipal Education Commission. This manuscript has not been subject to peer review by the above agencies and does not, therefore, reflect the views of the above agencies nor should any official endorsement be inferred.

CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

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