



Legacy and emerging per- and polyfluorinated alkyl substances (PFASs) in sediment and edible fish from the Eastern Red Sea[☆]



Aasim M. Ali^{a,*}, Monica Sanden^a, Christopher P. Higgins^b, Sarah E. Hale^c, Walied M. Alarif^d, Sultan S. Al-Lihaibi^d, Erik Magnus Ræder^e, Håkon Austad Langberg^{c,f}, Roland Kallenborn^{g,h}

^a Section of Contaminants and Biohazards, Institute of Marine Research (IMR), P.O. 1870 Nordnes, NO-5817, Bergen, Norway

^b Department of Civil & Environmental Engineering, Colorado School of Mines, 1500 Illinois Street, Golden, CO, 80401, USA

^c Geotechnics and Environment, Norwegian Geotechnical Institute (NGI), N-0855 Oslo, Norway

^d Department of Marine Chemistry, Faculty of Marine Sciences, King Abdulaziz University, PO Box 80207, Jeddah, 21589, Saudi Arabia

^e Faculty of Veterinary Medicine, Norwegian University of Life Sciences (NMBU), 0033, Oslo, Norway

^f Department of Biology, Norwegian University of Science and Technology (NTNU), NO-7491, Trondheim, Norway

^g Faculty of Chemistry, Biotechnology and Food Science (KBM), Norwegian University of Life Sciences (NMBU), P.O. Box 5003, Christian M. Falsen Veg 1, No-1432, Ås, Norway

^h Arctic Technology Department (AT), University Centre in Svalbard (UNIS), P.O. Box 156, Longyearbyen, Svalbard, Norway

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ABSTRACT

Poly- and perfluorinated alkyl substances (PFASs) are ubiquitously detected all around the world. Herein, for the first time, concentrations of 16 selected legacy and emerging PFASs are reported for sediment and edible fish collected from the Saudi Arabian Red Sea. Mean concentrations varied from 0.57 to 2.6 $\mu\text{g kg}^{-1}$ dry weight (dw) in sediment, 3.89–7.63 $\mu\text{g kg}^{-1}$ dw in fish muscle, and 17.9–58.5 $\mu\text{g kg}^{-1}$ dw in fish liver. Wastewater treatment plant effluents represented the main source of these compounds and contributed to the exposure of PFAS to biota. Perfluorooctane sulfonate (PFOS) was the most abundant compound in sediment and fish tissues analysed, comprising between 42 and 99% of the $\sum_{16}\text{PFAS}$. The short chain perfluorobutanoate (PFBA) was the second most dominant compound in sediment and was detected at a maximum concentration of 0.64 $\mu\text{g kg}^{-1}$ dw. PFAS levels and patterns differed between tissues of investigated fish species. Across all fish species, $\sum_{16}\text{PFAS}$ concentrations in liver were significantly higher than in muscle by a factor ranging from 3 to 7 depending on fish species and size. The PFOS replacements fluorotelomer sulfonate (6:2 FTS) and perfluorobutane sulfonate (PFBS) exhibited a bioaccumulation potential in several fish species and 6:2 FTS, was detected at a maximum concentration of $7.1 \pm 3.3 \mu\text{g kg}^{-1}$ dw in a doublespotted queenfish (*Scomberoides lysan*) liver. PFBS was detected at a maximum concentration of 2.65 $\mu\text{g kg}^{-1}$ dw in strong spine silver-biddy (*Gerres longirostris*) liver. The calculated dietary intake of PFOS, perfluorooctanoic acid (PFOA), perfluorononanoic acid (PFNA) and perfluorohexane sulfonic acid (PFHxS) exceeded the safety threshold established by the European Food Safety Authority (EFSA) in 2020 in doublespotted queenfish muscle, indicating a potential health risk to humans consuming this fish in Jeddah, Saudi Arabia.

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Main findings summary

PFASs in sediment and fish from the Saudi Arabian Red Sea were documented. WWTP effluents contributed to the exposure of PFAS

to biota in the coastal environment.

1. Introduction

The Red Sea is a unique marine coastal ecosystem. Owing to its semi-enclosed shape and its location in an arid region, the Red Sea is one of the world's warmest and most saline seawaters. Further, the large coral reef complexes extending over 4000 km along the Red Sea shoreline provide the necessary living habitat for a large

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* Corresponding author.

E-mail address: aasim.ali@hi.no (A.M. Ali).

number of fish species and marine invertebrates (Raitso et al., 2015; Voolstra and Berumen, 2019). The Red Sea is a vital resource for the Kingdom of Saudi Arabia (KSA), providing 90% of the potable water by desalination (Hoteit et al., 2020). In the Saudi Red Sea coast, annual fish production has been estimated at 24,000 metric tons, most of which comes from nearshore traditional fishing (Ministry of Agriculture, 2000–2012). However, the coastal environment of the Red Sea suffers from increased overpopulation and industrialization, and resulting anthropogenic activities pose a significant threat to this region (Al-Mur et al., 2017).

Several recent studies conducted on the Eastern Red Sea revealed elevated concentrations of various contaminants, including heavy metals, legacy organic pollutants, and a wide spectrum of organic contaminants of emerging concern (e.g. arsenic up to 94.2 mg kg⁻¹ in muscle of fish, mercury up to 1.96 mg kg⁻¹ in sediment, sum of 16 polycyclic aromatic hydrocarbons up to 2930 µg kg⁻¹ in sediment, and pharmaceuticals up to a few µg L⁻¹ concentrations in water) (Al-Mur et al., 2019; Ali et al., 2017; Ali et al., 2018; Burger et al., 2014a; El Zokm et al., 2021). However, there are many groups of priority contaminants that have not yet been sufficiently monitored in this region. Per- and polyfluorinated alkyl substances (PFASs) are one such group. PFASs are synthetic organic surfactants that have many industrially relevant properties (e.g. flame-retardant and water-resistant) (Banks et al., 2013; Kissa, 2001) and are unique compared to conventional organic pollutants. For instance, PFASs tend to associate with protein-rich tissues rather than lipids (Jones et al., 2003). As a consequence of their persistent nature, PFASs have reached pristine Arctic environments via long-range atmospheric and oceanic transport. In addition to their environmental stability and bioaccumulation potential, PFASs are not only present in many consumer products but also associated with point source releases. PFASs such as perfluoroalkyl sulfonates (PFSAs) and perfluoroalkyl carboxylates (PFCAs) are now considered ubiquitous contaminants. PFSAs and PFCAs are the final degradation products of many polyfluorinated precursors such as *N*-ethyl perfluorooctane sulfonamidoethanol (*N*-EtFOSE) and fluorotelomer alcohols (FTOHs) (Ellis et al., 2004; Rhoads et al., 2008). Major emission sources of PFASs include landfills, firefighting training facilities using aqueous film-forming foams (AFFF), industrial and municipal sewage effluents, and the production of paper products (Ahrens et al., 2014; Ahrens, 2011; Langberg et al., 2020). Such sources are prevalent in the Saudi coastal area and by monitoring PFASs, it is possible to investigate direct anthropogenic influences on the Red Sea coastal environment.

Several worldwide studies have reported that fish are one of the main dietary source of PFASs to humans (Hö;lzer et al., 2011; Yamaguchi et al., 2013), in addition to drinking water, fruit and egg products (EFSA, 2020). Two of the most studied and detected PFASs are perfluorooctane sulfonate (PFOS) and perfluorooctane carboxylic acid (PFOA) which are regulated under the UN Stockholm Convention. PFOS is listed in the Stockholm Conventions' B-list, meaning that all countries which have signed this treaty are obligated to limit production and use of this chemical, while PFOA is on the A-list, requiring countries to take action to "eliminate the production and use" of the chemical (UNEP, 2019). The European Commission has also included PFOS and its precursors on the list of priority substances that must be monitored in EU water bodies and set an environmental quality standard (EQS) of 9.1 µg kg⁻¹ wet weight for fish (Commission Directive, 2013). In 2020, the European Food Safety Authority (EFSA) reviewed the human health risk related to PFASs and established a group tolerable weekly intake (TWI) of 4.4 ng kg⁻¹ body weight (bw) for PFOA, PFOS, perfluorononanoic acid (PFNA) and perfluorohexane sulfonic acid

(PFHxS) (EFSA, 2020).

The phase-out of PFOS and PFOA have resulted in increased production of shorter chain length PFASs that are expected to be less bioaccumulative, in addition to other new alternative substances to PFOS and PFOA. These alternatives include perfluorobutanesulfonic acid (PFBS), 6:2 fluorotelomer-based substances and perfluoroether sulfonic acids (PFESAs) (Wang et al., 2015b), in addition to perfluoroalkyl ether acids and chlorinated polyfluoroalkyl ether acids (e.g., F-53B, GenX, and ADONA), which are expected to be less persistent (Munoz et al., 2019; Wang et al., 2013b). There may be other new PFASs with an unknown identity (Willach et al., 2016) as they are either proprietary substances or byproducts (Sun et al., 2016). Recent studies have detected such substances in waters at microgram per liter concentrations (Newton et al., 2017; Sun et al., 2016) and these compounds are now considered priority emerging PFASs (Munoz et al., 2019). However, little or no information is available on the environmental properties, fate, transport, exposure, or toxicity of these emerging PFASs (Mulabagal et al., 2018).

A baseline study on the environmental fate of selected PFASs in seawater collected from effluent dominated sites along the Saudi coastal Red Sea has recently been reported (Ali et al., 2021). This study revealed elevated concentrations of some PFASs in seawater affected by effluents from wastewater treatment plants (WWTP). Concentrations of PFASs in sediment and wild edible fish species living in these waters were not determined. To address this, a follow-up study has been carried out to examine the levels of PFASs in sediment and biota from the same coastal region. The distribution of ten perfluoroalkyl carboxylates (PFCA, C_{4,6-14}), three perfluoroalkyl sulfonates (PFSA, C_{4,6,8}), one fluorotelomer sulfonate (FTS, 6:2 FTS), perfluorooctane sulfonamide (FOSA), and four emerging PFASs (ADONA, GenX, 6:2 Cl-PFAES, 8:2 Cl-PFAES) was determined for the first time in sediment and edible fish tissues from the Eastern Red Sea environment off Jeddah, Saudi Arabia. In order to investigate the human health risk posed by the detected concentrations of these PFAS, the weekly intake was calculated and compared to EFSA's TWI. The findings presented here represent important baseline information about contamination of legacy and emerging PFASs in sediment and several edible fish species from the Saudi Arabian coast of the Red Sea.

2. Material and methods

2.1. Target compounds

Detailed information on sample preparation, analytical method, method validation, and quality control is outlined the supplementary information section. The linear isomers of the following target compounds and their isotopically labeled standards were obtained from Wellington Laboratories (Guelph, ON, Canada): ten carboxylates (C_{4,6-14} PFCAs), three sulfonates (C_{4,6,8} PFSAs), six precursors compounds including perfluorooctane sulfonamide (FOSA), 6:2 fluorotelomer sulfonate (6:2 FTS), *N*-ethyl and *N*-methyl perfluorooctane sulfonamidoethanol (*N*-EtFOSE and *N*-MeFOSE), and *N*-ethyl and *N*-methyl perfluorooctane sulfonamide (*N*-EtFOSA and *N*-MeFOSA). Emerging PFASs targeted in the current study include two perfluoroethers (dodecafluoro-3H-4,8-dioxanonoate (ADONA) and 2,2,3,3-Tetrafluoro-3-(trifluoromethoxy) propionic acid (HFPO-DA or GenX), and two chlorinated polyfluorinated ether sulfonates F-53B (6:2 chlorinated polyfluoroalkyl ether sulfonate (6:2 Cl-PFAES) and (8:2 Cl-PFAES). Names, CAS registry numbers, and structures of the substances are presented in Table S1 in the supplementary information. HPLC grade solvents (acetonitrile and methanol), ammonium acetate, and formic acid were purchased from VWR (West Chester, PA, USA).

2.2. Sampling locations and sampling

Jeddah is the second largest city in the Kingdom of Saudi Arabia and in 2015 had a population of 4.1 million people (Murad and Khashoggi, 2020). The city includes more than 450 factories of various products, such as paper and packaging, chemicals and pharmaceuticals, refineries, and petrochemicals industries (Basaham, 2009). In Jeddah, only 20% of the city area is covered by the wastewater collection and treatment network (Al-Farawati, 2010) and receives more than the operational capacity of the existing WWTP. This leads to discharge of raw and partially treated municipal and industrial wastewater in the coastal region of the Red Sea, and this in turn is considered one of the biggest marine environmental problems facing the region (Al-Farawati, 2010).

Sampling sites in the current study were selected based on their proximity to the main WWTP outlets, and were previously identified as being contaminated by various emerging contaminants (Al-Lihaibi et al., 2019; Ali et al., 2017, 2018, 2019, 2021). Samples were collected from these WWTP impacted sites in the Red Sea at the coastal city of Jeddah.

For nearshore sediment samples, three WWTP impacted sites along Jeddah coast (Al-Arbaeen, Al-Shabab, and Al-Khumrah), and one background site (Obhur creek) were selected (see Fig. 1). Al-Arbaeen and Al-Shabab are semi-enclosed coastal lagoons received partially treated sewage at estimated rates of 100,000

$m^3 day^{-1}$ (El Sayed et al., 2015) from Al-Balad and Al-Ruwais WWTP until 2017. In January 2017, the Presidency of Meteorology and Environment (PME) announced the suspension of Al-Balad and Al-Ruwais WWTP effluents (<https://www.spa.gov.sa/1580838>), in view of the Saudi Arabian coastal development agenda. The Al-Khumrah site has been receiving around 300,000 $m^3 day^{-1}$ of treated sewage from Al-Khumrah WWTP (Basaham, 2009). Unlike the two lagoons, the Al-Khumrah coastal site has higher water exchange with the open seawater. Therefore, Al-Arbaeen and Al-Shabab lagoons are susceptible to a greater accumulation of pollutants than Al-Khumrah (Fig. 1). Obhur is a coastal creek located about 35 km north of Jeddah. Although Obhur creek is about 30 km away from the main WWTP effluents, uncontrolled disposal of untreated municipal sewage has been observed along the shores of the creek (Basaham and El-Sayed, 2006; Ghandour et al., 2014). The entire shore of the Obhur creek is now occupied by resorts, hotels, and other recreational facilities such as jetties and marinas, in addition to the campuses of faculty of Maritime Studies and faculty of marine sciences. Owing to this site's location, it was considered a background site representing coastal waters.

A total of 13 nearshore surface sediment samples were sampled as follows: four samples from Al-Arbaeen lagoon (Arb1-4); three samples from Al-Shabab (Shab1-3), four samples from Al-Khumrah (Kum 1-4); two samples from Obhur creek (Obhur1 and 2) (Fig. 1). One to two replicates were analysed for each sediment sample,

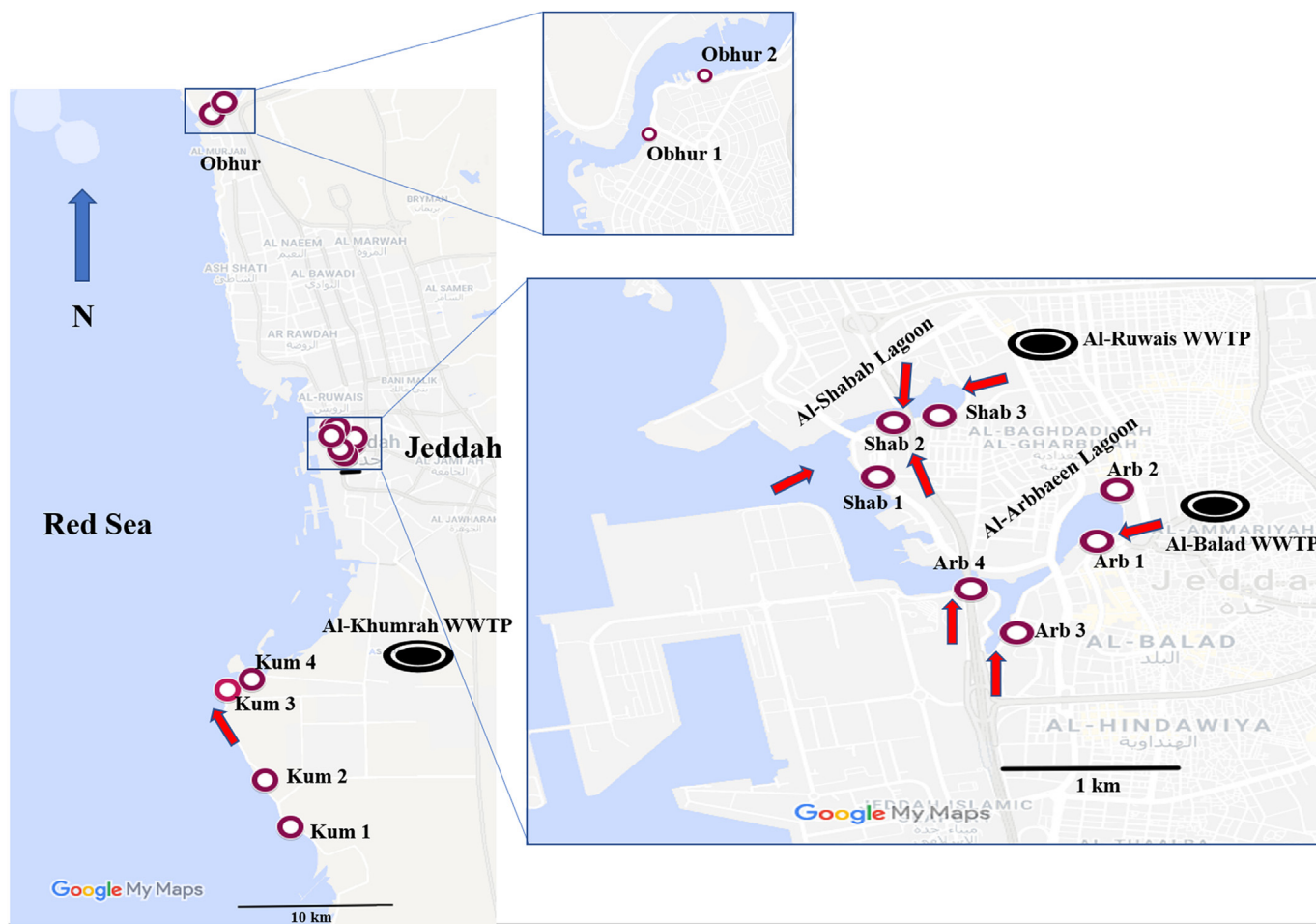


Fig. 1. Map showing the geographical location of the study area (source: google map). Red circles represent the sampling locations. WWTP effluents are indicated by red arrows. Three wastewater treatment plants (WWTP): Al-Khumrah, Al-Balad, and Al-Ruwais are indicated by black circles. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

leading to a total of 21 sediment samples. Approximately 1 kg of sediment was collected from the top 10 cm of sediment at approximately 1 m water depth from each location using a pre-cleaned stainless-steel shovel. The sample was stored in a pre-cleaned glass beaker and covered with aluminum foil. On arrival at the laboratory, sediment samples were freeze-dried.

Fish were sampled from the Al-Arbaeen lagoon site as its waters showed the highest PFAS concentration amongst all sites (Ali et al., 2021). Six seafood fish species were collected: Doublespotted Queenfish (*Scomberoides lysan*, $n = 5$), Marbled Spinefoot (*Siganus rivulatus*, $n = 7$), Bigeye Scad (*Selar crumenophthalmus*, $n = 7$), Strongspine silver-biddy (*Gerres longirostris*, $n = 7$), Bluefin Trevally (*Caranx melampygus*, $n = 3$), and Bonefish (*Albula glossodonta*, $n = 2$). All fish samples were collected in December 2018 by fishermen with hook-and-line supervised by the researchers. More information on fish samples can be found in Table S2. On arrival at the laboratory, the length and weight of all fish individuals were recorded, and from each individual fish, liver and muscle tissues were manually separated and freeze-dried. The freeze-dried fish and sediment samples were shipped for further treatment and analysis.

2.3. Quality assurance and quality control

Detailed information on method validation and quality control is provided in the supplementary information section (Section S2).

To investigate possible contamination during sample preparation, procedural blanks (made of Milli Q water and HPLC grade methanol) were included and processed as real samples. Furthermore, methanol was injected after every 10 samples as a solvent blank to monitor the instrumental contamination and carryover. A delay column (Agilent Eclipse Plus C18, 4.6×50 mm, $3.5 \mu\text{m}$) was installed before the autosampler to minimize potential PFAS contamination resulting from the HPLC tubing (Powley et al., 2008). None of the targeted PFASs were detected in the methanol blanks but some substances were detected in the procedural blanks. For these compounds, the limit of quantification (LOQ) was determined as $C + 3SD$, where C is the mean concentration measured in blank samples and SD is the standard deviation. However, no blank subtraction was made for these compounds. Limit of detection (LOD) and limit of quantification (LOQ) were determined using the compound specific amount corresponding to a signal area resulting in a signal to noise ratio (S/N) of 3 (LOD) or 10 (LOQ), respectively.

2.4. Calculations

The following equation was used to calculate the weekly intake of PFAS (WI) ng/per kg body weight from fish consumption:

$$WI = C_{\text{muscle}} \times M/Wt$$

where C is the concentration of the sum of PFOA, PFNA, PFHxS, and PFOS in muscle ($\mu\text{g kg}^{-1}$ wet weight); M is the weekly intake of consumed seafood (g/week), and Wt is body weight (kg). The value of M used was 259.7 g/week which is based on the seafood intake per week in Jeddah (Burger et al., 2014b), and an average body weight of 70 kg was used (EFSA, 2020). PFAS wet weight basis concentrations were obtained based on the moisture content (%) of each fish specimen (Table S2). The calculated weekly intake of PFAS from fish consumption was compared to the EFSA tolerable weekly intake (TWI) of 4.4 ng per kilogram of body weight per week for PFOA, PFOS, PFNA and PFHxS to evaluate the safety of the consumption of fish. After estimating an overall method uncertainty of 30–40% in compound specific PFAS concentration range in the target fish, the low-bound TWI values were used for the subsequent

discussions as recommended by EFSA (EFSA, 2018a).

2.5. Statistical analysis

Analysis of variance (ANOVA) carried out in Statistica (StatSoft, Tulsa, OK, USA) was used to test the significance of the difference between the means. The associations between PFAS concentration and biological variables (weight and length) of individuals of each fish species were investigated using Pearson's or Spearman's rank correlations. The significance level was set at $p < 0.05$. Concentrations below the LOQ were assigned half the LOQ for statistical analyses.

3. Results and discussion

3.1. Method validation

Method performance (accuracy and precision) was assessed using the matrix-spiked samples. For fish, percent recoveries ranging from $51.4 \pm 0.90\%$ to $90.8 \pm 4.0\%$ and $46.5 \pm 1.7\%$ to $79.2 \pm 4.9\%$ were determined at high and low-spiking levels, respectively. The emerging PFASs (ADONA, 8:2 Cl- PFESA, 6:2 Cl- PFESA, and HFPO-DA) displayed lower recoveries at both fortification levels (31.6 ± 2.6 to $56.7 \pm 4.5\%$), most likely due to the lack of their exactly-matched, isotopically-labeled internal standards (Miaz et al., 2020). These compounds were included in quantification owing to their excellent precision indicated by the relative standard deviations below 5% (Table S3). In light of the lower recoveries, it is possible that their reported concentrations may be underestimated. Similar recoveries were observed in spiked sediment samples. *N*-MeFOSE, *N*-MeFOSA, *N*-EtFOSE, and *N*-EtFOSA showed unacceptable recoveries (<15%), and their concentrations are therefore not reported. Limits of detection and quantitation (LOD and LOQ, respectively) depicted in Table S3 demonstrate the good sensitivity of the applied method. Overall, it can be concluded that the validation data presented here demonstrate good method performance for most target compounds.

3.2. Levels and composition profiles of PFASs in sediment

Concentrations ($\mu\text{g kg}^{-1}$ dry weight; dw) of legacy and emerging PFASs in near shore sediments collected from different sites along the Saudi coast of the Red Sea are presented in Fig. 1 and Table S4. PFASs were detected in all sediment samples collected from the Jeddah coast. The sum concentrations of the 16 quantified PFASs ($\sum_{16}\text{PFAS}$) ranged from 0.57 (Obhur) to $2.66 \mu\text{g kg}^{-1}$ dw (Al-Arbaeen), indicating their widespread presence. $\sum_{16}\text{PFAS}$ concentrations in sediments of the semi-enclosed lagoons Al-Arbaeen and Al-Shabab ($1.63 \pm 0.195 \mu\text{g kg}^{-1}$ dw) were significantly higher (ANOVA, $p = 0.036$) than in sediments from the background site Obhur creek ($0.843 \pm 0.213 \mu\text{g kg}^{-1}$ dw). As previously reported, the local WWTPs are found to be significant PFAS sources (Ali et al., 2021). Given that the marine sediment and biota samples are taken from the same locations, the disposal of WWTP sewage can be confirmed as a significant source of PFASs in this region. For the past decades, these coastal lagoons have received partially treated sewage from two main WWTPs (Al-Balad and Al-Ruwais) that apply sedimentation and secondary treatment (Fig. 1).

Sediment collected from the Al-Khumrah site (Kum1-4) had $\sum_{16}\text{PFAS}$ concentrations of $0.91 \pm 0.12 \mu\text{g kg}^{-1}$ dw. This site is impacted by an active WWTP effluent from the biggest WWTP in Jeddah, Al-Khumrah WWTP, which discharges a tertiary treated wastewater at this site. Therefore, the relatively low concentrations detected in this area compared to the concentrations in sediments from the two lagoons can either be attributed to the dilution effect

due to water exchange with the open sea, or to a more efficient removal from raw sewage during treatment at the Al-Khumrah WWTP.

The reference site Obhur creek is considered as a rural/suburban background-level site for other contaminants (Al-Lihaibi et al., 2019). The lowest average \sum_{16} PFAS concentration was detected at Obhur and can possibly be attributed to discharges from nearby resorts in addition to diffuse pollution. In general, sediment collected from sites closer to the WWTP effluents (Arb 1, 3 and 4 and Shab 2) had higher PFAS concentrations (See Figs. 1 and 2).

Treatment at a WWTP can affect the PFAS profile in the sludge and resultant water as processes such as partitioning to sludge and transformation of precursors occur (Eriksson et al., 2017; Kunacheva et al., 2011). Previously PFCAs have been reported at higher concentrations in WWTP effluent compared to influents, owing to the fact that they can be formed from the degradation of precursors during waste water treatment (Guerra et al., 2014; Schultz et al., 2006). PFOS (sum isomers) was the predominant compound in the WWTP impacted sediment investigated here, accounting for 53–98% of \sum_{16} PFAS in more than 52% of the samples analysed, and was detected at a maximum concentration of $1.80 \mu\text{g kg}^{-1}$ dw in Al-Arbaeen lagoon sediment. This supports the previous study carried out in the same area, where shorter chain PFASs (PFHxA, PFHxS, and 6:2 FTS) were most prevalent in the water phase at these sites (Ali et al., 2021). This can be attributed to the physicochemical properties of PFOS, which preferentially partitions to sediment and is less mobile in water compared to PFHxA, PFHxS, and 6:2 FTS (Guelfo et al., 2013). These data are also consistent with the observation of high salinity of the Red Sea coastal water (average = 40 psu) and the greater impact of salinity on PFOS sorption as compared to other PFASs (due to “salting out”) (Jeon et al., 2011; Wang et al., 2020).

PFBA was the second most dominant compound, detected in sediments from all sites and accounting for 45–60% of the total PFASs in 30% of sediment samples analysed, at concentrations ranging from 0.17 ± 0.12 in Obhur creek sediment to $0.51 \pm 0.04 \mu\text{g kg}^{-1}$ dw in Al-Shabab sediment. PFBA is known to have a sorption potential similar to that of PFOA which has a longer chain length (Guelfo et al., 2013) and this may explain the observation here. The dominance of PFBA in sediments is supported by the increase in the production of short chain PFASs as an alternative to PFOS (Wang et al., 2020). PFBA has previously been detected in meat and meat-processed foods collected from the capital of Saudi Arabia at a maximum concentration of $0.422 \mu\text{g kg}^{-1}$ (Pérez et al.,

2014), and recently, Banjabi et al. (2020) detected PFBA in 99% of the analysed serum samples collected from individuals from Jeddah, Saudi Arabia, at a mean concentration of 1.56 ng mL^{-1} . A similar dominance of PFBA in sediment was also observed in samples collected from different locations (Ahrens et al., 2010; Meng et al., 2014; Yeung et al., 2013) and similar absolute PFBA concentrations were reported in impacted sediment from Bohai Bay, China (Liu et al., 2019). In addition to its direct sources, PFBA may occur in sediments as a result of transformation of precursor compounds. For example, the biotransformation of 6:2 FTOH and 6:2 FTS in sediment has been reported, with PFBA identified as a major transformation product (Hamid et al., 2020; Zhao et al., 2013). Further, degradation of precursors during sewage treatment in WWTP can be an additional source of PFBA (Guerra et al., 2014).

The other short chain PFCA (PFHxA) was detected in all sediment samples from the WWTP impacted semi-enclosed lagoons (Al-Arbaeen and Al-Shabab) at a maximum concentration of $0.06 \mu\text{g kg}^{-1}$ dw. In the sediments from Al-Khumrah and the background site Obhur, PFHxA was detected in one third of samples at the LOQ concentration. The degradation of 6:2 FTS present in these sediments may have contributed to the detection of PFHxA, as PFHxA is known to be among the degradation products of 6:2 FTS (Wang et al., 2011).

Long chain PFCAs (C_8 – C_{14}) were detected in between 43 and 90% of samples at a maximum concentration of $0.04 \mu\text{g kg}^{-1}$ dw for PFUnDA. The predominance of longer-chained PFASs in sediment is attributed to their higher tendency to partition into the sediment (Higgins et al., 2006). Similar PFAS profiles have been observed in WWTP effluents and sludge previously (Campo et al., 2014; Ma and Shih, 2010), supporting the conclusion that WWTP effluents are the primary sources of PFASs in the current study.

6:2 FTS was detected in certain sediment samples at maximum concentrations of $0.22 \mu\text{g kg}^{-1}$ dw (Al-Arbaeen), $0.50 \mu\text{g kg}^{-1}$ dw (Al-Shabab) and $0.97 \mu\text{g kg}^{-1}$ dw (Obhur). 6:2 FTS was predominant in the water phase at the sites investigated here, reported at concentrations up to 325 ng L^{-1} (Ali et al., 2021). 6:2 FTS is likely a transformation product of compounds present in AFFF such as fluorotelomer thioether amido sulfonate (Harding-Marjanovic et al., 2015). 6:2 fluorotelomer sulfonamide alkylbetaine (6:2 FTAB) is the major component of the fluorosurfactant Forafac® which has been used in some AFFF formulations (Hagenaars et al., 2011). 6:2 FTAB is a potential precursor for 6:2 FTS and can be considered as indicative of an AFFF source. Firefighting stations in the vicinity of Al-Shabab and Al-Arbaeen were previously suggested as potential local sources of this compounds (Ali et al., 2021) and the results presented here support this conclusion. The 6:2 FTS detected in sediment from the background site (Obhur) could be attributed to the firefighting training events conducted for students and border guards at the faculty of maritime studies nearby Obhur creek (100–200 m from the sampling site).

Previous studies reporting concentrations of PFAS in marine sediments are limited. The sediment samples investigated in this study had comparable concentration levels of \sum_{16} PFAS (\sum_{16} PFAS = 0.51 – $2.66 \mu\text{g kg}^{-1}$ dw, mean $1.09 \mu\text{g kg}^{-1}$ dw) for worldwide sediments impacted by WWTP (see Table S6). For example, sediment collected from Bohai Bay, China had mean \sum_{17} PFAS of $6.76 \mu\text{g kg}^{-1}$ dw (Liu et al., 2019; Wang et al., 2015a). Similar concentration ranges were reported for sediments from the North and Baltic Seas and Korean coasts (\sum_{16} PFAS = 0.02 – $2.6 \mu\text{g kg}^{-1}$ dw, mean $0.64 \mu\text{g kg}^{-1}$ dw) (Joers et al., 2019; Lee et al., 2020).

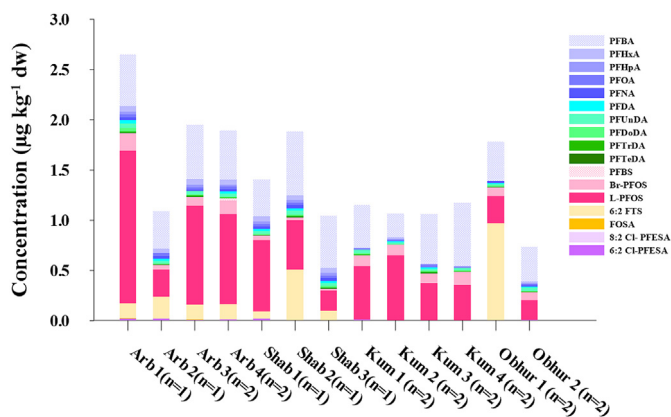


Fig. 2. Concentration ($\mu\text{g kg}^{-1}$ dw) of PFASs in surface sediment samples collected from the Eastern Red Sea Arb:Al-Arbaeen lagoon; Shab: Al-Shabab lagoon; Kum: Al-Khumrah. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

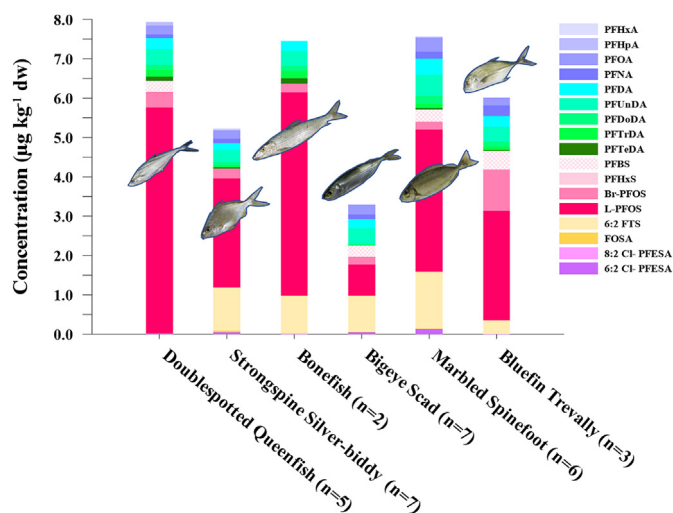


Fig. 3. PFAS concentrations ($\mu\text{g kg}^{-1}$ dw) in fish muscles of fish individuals collected from the Saudi coastal waters. The number of fish individuals are shown in bracket. The fish species in the bars apply to is shown. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

3.3. Levels and composition profiles of PFASs in fish muscles

Fig. 3 and Table S5 show the concentrations of PFASs in muscles of fish individuals collected from effluent-impacted waters on the Saudi Coast of the Red Sea at the Al-Arbaeen coastal lagoon. The six fish species sampled represent the native ecosystem in this coastal seawater. All target legacy PFASs were detected above the LOQ in at least one muscle sample. PFOS and PFUnDA were the most frequently detected substances and were detected in all muscle samples with maximum concentrations of 15.13 and $0.84 \mu\text{g kg}^{-1}$ dw, respectively, indicating their high bioaccumulation potentials. This is in agreement with a recent study where PFOS and PFUnDA were the most dominant compounds detected in muscles of coral reef associated fish species collected from the South China Sea (maximum concentrations of 27.05 and $0.35 \mu\text{g kg}^{-1}$ ww, respectively) (Pan et al., 2018). The fish species investigated in the current study are also considered to be coral reef-associate species, and this might be a reason for the observed profile similarities. PFDA was the second most dominant PFAS detected and was present in 98% of the muscle samples analysed, at concentration up to $0.80 \mu\text{g kg}^{-1}$ dw. The long chain PFASs ($C \geq 8$; PFOS and PFCAs C_{8-14}) were detected in around half of the samples (48% frequency of detection), whereas the short chain PFASs (PFBS and PFCAs C_6 and C_7) were detected in much lower frequencies (less than 25%). Interestingly, 6:2 FTS was detected in muscles of all species except doublespotted queenfish, possibly due to the high concentration observed in the coastal waters of Al-Arbaeen lagoon (maximum concentration of 450 ng L^{-1}) (Ali et al., 2021). Although, the compound was not detected in any muscle of doublespotted queenfish, the maximum average concentration was observed in livers of this species ($7.1 \pm 3.3786 \mu\text{g kg}^{-1}$ dw). This may indicate potential bioaccumulation of this compound in these coral reef-associated fish, despite previous literature showing that this substance has a relatively low bioaccumulation potential (Hoke et al., 2015). 6:2 FTS has been observed to have the potential to form PFCAs *in vivo* (≤ 7 C) (Yang et al., 2014), which may help explain the occurrence of short chain PFCAs in muscles of the fish investigated here.

One-way ANOVA carried out between the species showed that Σ_{16} PFAS significantly differed ($F(5,28) = 8.624$, $p = 0.00005$), muscles of doublespotted queenfish showed significantly higher

levels of PFASs (mean Σ_{16} PFAS = $7.91 \pm 0.72 \mu\text{g kg}^{-1}$ dw) than strongspine silver-biddy and bigeye scad (means of Σ_{16} PFAS = 4.09 ± 0.48 and $2.32 \pm 0.60 \mu\text{g kg}^{-1}$ dw, respectively). Doublespotted queenfish has been reported to have a slightly higher trophic index (4.0 ± 0.67) than bigeye scad (3.8 ± 0.2) (Available online: <http://www.fishbase.in>; accessed on March 07, 2021). Although, results presented here did not confirm these trophic levels indices (Table S2), doublespotted queenfish had the highest individual weights (621–912 g), while bigeye scad had the lowest individual's weights (14–18 g). This may support the fact that doublespotted queenfish has higher trophic level than bigeye scad (Romanuk et al., 2011). In general, higher PFAS concentrations were observed in muscles of fish species with higher trophic indices (doublespotted queenfish and Bluefin Trevally) compared with species having relatively lower trophic levels (bonefish, bigeye scad, and strongspine silver-biddy) (Table S2). However, the potential impact of other factors (e.g. age, protein to lipid ratios, and PFAS depuration rates) cannot be excluded.

Some differences of PFAS profiles in muscle were observed among fish species. On average, PFOS was the predominant compound in muscle samples, accounting for 41–83% of the total PFASs observed. Long chain PFCAs (C_{8-14}) made up to 30% of Σ_{16} PFAS. The composition profile of PFHxA, PFHpA, PFNA, PFDoDA, PFTeDA, PFBS, 8:2 Cl-PFESA and 6:2 Cl-PFESA was similar among the investigated species, while profiles of PFOA, PFDA, PUnDA, PFTrDA, PFOS, and FOSA differed significantly ($p < 0.05$) between species. Muscles of bigeye scad individuals showed the highest contributions of PFOA, PFDA, and PFUnDA, and the lowest PFOS percentages (41%). The highest PFOS percentages (83%) were observed in bonefish and doublespotted queenfish. The differences observed in the composition profiles of certain PFASs between fish species can be partially attributed to the significant differences in trophic level, feeding behavior, and protein content (Babut et al., 2017; de Miranda et al., 2020).

PFAS profiles in fish tissues may not represent only uptake from the surrounding media and diet as precursor biotransformation may play a part (Benskin et al., 2009b). The only PFOS precursor detected here was FOSA (Buck et al., 2011), in muscle samples. FOSA was detected in 92% of muscle samples at levels up to $0.077 \mu\text{g kg}^{-1}$ dw. However, the significant differences in the [FOSA]/[PFOS] ratios observed between species suggested that differences in biotransformation capabilities of polyfluorinated precursors between the species could be another reason for the observed profile of PFOS among fish species. Doublespotted queenfish and bluefin trevally showed the lowest [FOSA]/[PFOS] ratios. These species also had the highest trophic indices (4.0 ± 0.67 and 4.5 ± 0.8 , respectively) and the highest individual sizes. The low ratios for these species could be due to the higher PFOS concentrations in these species or due to their lower FOSA concentrations. The latter is supported by reports of fish at high trophic levels that have higher metabolic capacities to transform FOSA than fish at lower trophic levels. Alternatively, a larger amount of PFOS precursors could be transformed as they are transferred through the food chain, resulting in fish at higher trophic levels being exposed to more PFOS and less FOSA via their diet, as compared to fish at lower trophic levels (Gebbink et al., 2016a).

Biological data (weight and length) of the sampled fish are shown in Table S2. For fish muscles, the Pearson correlation indicated that Σ_{16} PFAS concentrations in muscles of marbled spinefoot were positively correlated with both length ($r^2 = 0.784$, $p = 0.036$) and weight ($r^2 = 0.793$, $p = 0.033$) of fish individuals. The correlation observed here can be attributed to age and the increase of food uptake rates and/or differences in food selection with fish development (Pan et al., 2014). In contrast, significant negative correlations were observed between Σ_{16} PFAS and individual weight ($r^2 = -0.998$, $p = 0.032$), and length ($r^2 = -0.998$,

$p = 0.032$) for bluefin trevally. Likewise, for bigeye scad, significant negative correlations were observed between \sum_{16} PFAS and individual weight ($r_2 = -0.830$, $p = 0.020$) and individual length ($r_2 = -0.847$, $p = 0.016$). For both doublespotted queenfish and strongspine silver-biddy, no significant correlations were observed. For fish liver, there was no significant correlation between fish length or fish weight with \sum_{16} PFAS or PFOS (data not shown). This highlights that the correlation between PFASs and the fish size is species and tissue dependent. Both positive correlations between size and \sum PFAS concentrations have previously been reported for some fish species (e.g. Korean rockfish: *Sebastes schlegelii* and tilapia) (Hung et al., 2020; Pan et al., 2014), as well as negative correlations (Taylor, 2020), as well as studies that did not observe correlations (Akerblom et al., 2017; Gewurtz et al., 2012).

When compared with fish muscles collected worldwide, the PFAS concentrations in muscles in the current study are within the same range for legacy PFASs and of the same order of magnitude as other fish (Table S7). However, PFAS concentrations in fish are influenced by several biological and environmental parameters, which may cause a variation within tissues and among samples, which weakens the comparisons among different monitoring programs (Valsecchi et al., 2020).

3.4. Levels and composition profiles of PFASs in fish liver

PFASs were also analysed in livers of the same specimen (except marbled spinefoot and bigeye scad) collected from the WWTP-impacted lagoon. PFAS concentrations in livers of four fish species are shown in Fig. 4 and Table S6. Ten legacy PFASs and two emerging PFASs were detected above the LOQ in at least one liver sample. The precursor 6:2 FTS was detected in 80% of the liver samples, at a maximum mean concentration of $7.86 \mu\text{g kg}^{-1}$ dw (doublespotted queenfish). On average, mean \sum_{16} PFAS concentrations varied from 17.9 (strongspine silver-biddy) to $72.1 \mu\text{g kg}^{-1}$ dw (doublespotted queenfish). Overall, PFAS concentrations were higher in liver compared to fish muscles, which is consistent with previously reported tissue distribution studies (Martin et al., 2003; Peng et al., 2010; Valsecchi et al., 2020). For doublespotted queenfish, PFNA, PFDA, PFUnDA, PFDoDA, PFTrDA, PFOS, and 8:2 Cl-PFESA, and 6:2 FTS were significantly higher ($p < 0.05$) in liver

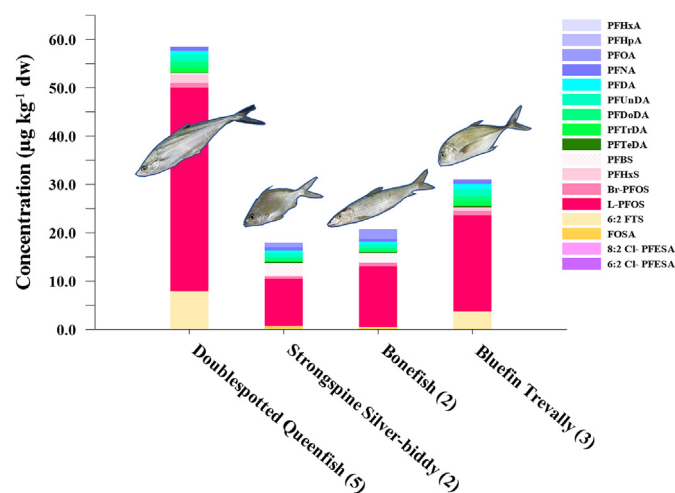


Fig. 4. PFAS mean concentrations ($\mu\text{g kg}^{-1}$ dw) in livers of individual fish collected from Al-Arbaeen lagoon, Saudi coastal waters. The number of fish individuals are shown in bracket. The fish species in the bars apply to is shown. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

samples than muscles, which is consistent with a recent study by Hung et al. (2020). This is likely due to the fact that PFASs bind more strongly to serum proteins as well as proteins in the liver and kidney than to fatty tissues (Jones et al., 2003).

In contrast to muscles, livers showed no statistically significant association between \sum_{16} PFAS and fish weight and fish length, in agreement with a previous study (Arinaitwe et al., 2020). However, similar to muscles, one-way ANOVA carried out between the species showed that \sum_{16} PFAS differed between livers of fish species ($F(3, 6) = 6.2764$, $p = 0.027$). The highest \sum_{16} PFAS was detected in doublespotted queenfish ($57.3 \pm 5.2 \mu\text{g kg}^{-1}$ dw) and the lowest was detected in strongspine silver-biddy ($17.9 \mu\text{g kg}^{-1}$ dw).

To compare the bioaccumulation of PFASs with different fish species collected worldwide, liver-to-muscle concentration ratios based on individual fish were calculated for C_9 : C_{11} PFCA and PFOS, which were detected in all samples at concentrations above the LOQs. High PFOS liver-to-muscle ratios were observed in doublespotted queenfish and bluefin trevally (8.3 and 8.1, respectively) compared to strongspine silver-biddy, and bonefish (4.2 and 2.9, respectively). These PFOS ratios were much lower than the ratio calculated for Chinese sturgeon collected from a freshwater environment of 61.5 (Peng et al., 2010). However, a ratio of 10 was reported for edible fish from the Mediterranean Sea (Nania et al., 2009). This indicates that fish species investigated in the current study have different PFOS tissue distributions with higher PFOS percentages in muscles. Overall, PFOS had the highest liver-to-muscle ratios, indicating its higher accumulation potential in liver compared to PFCA, consistent with several studies (Pan et al., 2014; Shi et al., 2012). The liver-to-muscle ratio for PFNA was only calculated for two species, and the average ratios were found to be 3.1 and 2.6 for bluefin trevally and doublespotted queenfish, respectively. The average liver-to-muscle ratios for PFDA were 4.0, 3.2, and 2.0 for bluefin trevally, doublespotted queenfish, and bonefish, respectively. Mean ratios in the same range were reported for PFDA (2.90–9.77) and PFUnDA (2.62–10.26) in different fish species (Shi et al., 2012). In all fish species investigated, the PFCA ratios increased with PFCA chain length, indicating higher liver accumulation potential for long chain PFCA, supporting previous studies (Martin et al., 2003; Shi et al., 2012).

The concentrations ratio of PFOS to C_9 : C_{11} PFCA in fish (PFOS/ \sum LC-PFCA ratio) of >1 can be used as a proxy for the impact of local PFAS sources related to human activities, while a ratio of <1 indicates an atmospheric PFAS source (Valsecchi et al., 2020). Sediments investigated in the current study had PFOS/ \sum LC-PFCA ratios of 7.0 ± 3.7 , which supports the presence of a local contamination source (wastewater disposal). Similarly, fish had PFOS/ \sum LC-PFCA ratios of 2.7 ± 1.5 and 5.3 ± 2.6 for muscles and livers, respectively, indicating the presence of a direct contamination source.

Given the extensive nature of the coral reefs in the Red Sea and the fact that large volumes of water flow daily on reefs at mean currents of 5 cm/s (Reidenbach et al., 2006), it is plausible that PFASs partition to the reef surfaces. Therefore, reefs may act as PFAS sinks and may represent an additional source of PFASs for the fish species investigated here. Further research into this potential is warranted.

3.5. Isomer profiles of PFOS in sediment and fish

PFOS produced by 3M Co. via electrochemical fluorination (ECF) is known to contain $70 \pm 1.1\%$ linear isomer and $30.0 \pm 0.8\%$ branched isomer (Vyas et al., 2007). There is a difference in the environmental behavior of PFOS isomers (Fang et al., 2014) and this can be used to elucidate PFAS sources (Langberg et al., 2020).

In sediment samples, linear PFOS (L-PFOS) isomers and branched PFOS (Br-PFOS) isomers contributed 84.7 ± 7.8 and

15.3 ± 7.8% of \sum PFOS, respectively. This enrichment of L-PFOS in sediment, could be explained by the difference in the physico-chemical properties of the linear and branched isomers. The linear isomer has been predicted to be more hydrophobic compared to the branched PFOS isomers (Beesoon et al., 2011), which results in a larger potential to partition to sediment. An enrichment of branched isomers in the waters collected from the same locations has previously been observed (Ali et al., 2021), in support of the assertion that sediments are more enriched in linear PFOS as compared to the overlying surface waters. It is worth noting that a significantly higher L-PFOS percentages were found in sediments of Al-Arbaeen (88.6 ± 2.9%) and Al-Shabab (93.1 ± 1.4%) than in sediments of Al-Khumrah (80.2 ± 5.2%) and Obhur (73.3 ± 3.6%) ($p = 0.001$). This trend agrees with the sediment-specific parameters observed by Rasiq et al. (2018). Due to sewage disposal, high percentages of total organic carbon (TOC %) and clay were determined in sediments of Al-Arbaeen (4.9 ± 4.3 and 60.8 ± 32.2, respectively) and Al-Shabab (4.6 ± 2.3, 55.1 ± 27.3, respectively) compared to Obhur sediment (0.28 ± 0.1 and 1.4 ± 2.1, respectively). Hence, the higher L-PFOS ratio in Al-Arbaeen and Al-Shabab is likely due to the higher OC content, as L-PFOS more readily partitions to sediment OC as compared to Br-PFOS (Chen et al., 2012, 2015).

Higher percentages of L-PFOS were observed in fish liver and muscles: 96.4 ± 1.4 and 90 ± 6.5% of \sum PFOS, respectively, in agreement with previous studies (Fang et al., 2014; Houde et al., 2008). A possible mechanism for this linear PFOS isomer enrichment is the selective bioconcentration of L-PFOS isomer and/or the preferential excretion of Br-PFOS (Benskin et al., 2009a; Sharpe et al., 2010). The PFOS isomer profile changed with fish tissue and species. In general, lower Br-PFOS ratios were observed in fish livers compared to muscles (only significant in doublespotted queenfish ($p < 0.03$)). In addition, the PFOS isomer profile significantly differed ($p < 0.001$) between the muscle of fish species. The preferential biotransformation of the branched isomers of PFOS precursors (Benskin, 2009b) could explain this. For instance, the low percentages of Br-PFOS observed in the high trophic doublespotted queenfish (25.0%) fish compared to the bigeye scad (37.3%) can be attributed to the lower contribution from PFOS precursors which is consistent with the discussion above. However, the preferential difference in elimination of Br-PFOS relative to L-PFOS could also influence these ratios (Peng et al., 2014).

3.6. Emerging and replacement PFASs in fish and sediment

PFBS is a potential degradation product of perfluorobutane sulfonyl fluoride (PBSF)-based surfactants, which are considered alternatives for PFOS-related products and have increasing applications (Newsted et al., 2008). PFBS was detected in about 28% of muscle and liver samples, at maximum concentrations of 2.13 and 2.65 $\mu\text{g kg}^{-1}$ dw, respectively. The bioaccumulation of PFBS was species and tissue dependent. PFBS was not detected in any muscle samples of strongspine silver-biddy and bonefish individuals. PFBS was not detected in any liver samples of bluefin trevally individuals, but was detected in livers of three doublespotted queenfish individuals at an average concentration of 0.94 $\mu\text{g kg}^{-1}$ dw, and was detected in livers of all the analysed individuals of strongspine silver-biddy and bonefish at 2.65 and 1.22 $\mu\text{g kg}^{-1}$ dw respectively. Overall, the detection of this short chain compound highlights its persistent and bioaccumulative nature, the increase in its production (and/or its precursors) and use following the phase out of PFOS (Gebbinck et al., 2016b). Data regarding the occurrence of PFBS in biota is limited. Similar concentrations were reported for herring from Sweden (maximum concentration of 0.10 $\mu\text{g kg}^{-1}$ ww), while lower concentrations were also reported for polar bear livers from

Greenland (average concentration of 0.032 ± 0.008 $\mu\text{g kg}^{-1}$) (Gebbinck et al., 2016b; Ullah et al., 2014), and higher concentrations were reported (up to 16.9 $\mu\text{g kg}^{-1}$ ww) for fish from Yadkin-Pee Dee River, US (Penland et al., 2020).

PFBS was not detected in any sediment samples, although PFBS is a predominant PFAS in seawater of the sites investigated here (Ali et al., 2021). This suggests that the PFBS in the water phase is the main source of PFBS to biota, either directly or via the degradation of its precursors. PFBS has been reported to be a major contaminant in WWTP effluents (Ahrens et al., 2009).

In the current study, four emerging PFASs were quantified: ammonium 2,3,3,3-tetrafluoro-2-(heptafluoropropoxy) propanoate (HFPO-DA; Gen-X), 4,8-dioxa-3H-perfluorononanoate (ADONA), potassium9-chlorohexadecafluoro-3-oxanonane-1-sulfonate (6:2 Cl- PFESA), and potassium 11-chloroeicosafuoro-3-oxaundecane-1-sulfonate (8:2 Cl- PFESA). These compounds are among the current alternatives to the long-chain perfluoroalkyl acids (PFOS and PFOA) (Wang et al., 2013b). The two chlorinated polyfluoroalkyl ether sulfonic acids (6:2 Cl- PFESA and 8:2 Cl- PFESA with the former as a major and the later as a minor component) constitute the product commercially called F-53B, which is a PFOS alternative manufactured in China (Kotthoff et al., 2020). F-53B is mainly used in the chrome plating industry as a mist suppressant (Wang et al., 2013a). Due to the introduction of alkyl ether (C-O-C) bonds in the fluoroalkyl chain, these compounds are expected to have lower bioaccumulation potential and lower environmental persistence compared to PFOA and PFOS (Munoz et al., 2019).

8:2 Cl- PFESA and 6:2 Cl- PFESA were detected in fish muscles at trace concentrations (maximum concentration of 0.10 and 0.48 $\mu\text{g kg}^{-1}$ dw, respectively) in 40 and 29% of samples, respectively. In addition, both 8:2 Cl- PFESA and 6:2 Cl- PFESA were detected in 44 and 16% of liver samples, respectively, at maximum concentrations of 0.06 and 0.21 $\mu\text{g kg}^{-1}$ dw, respectively. In sediment samples, 6:2 Cl- PFESA was only detected in samples collected from Al-Arbaeen and Al-Shabab lagoons at a maximum concentration of 0.02 $\mu\text{g kg}^{-1}$ dw. Similar concentrations ranging from <0.002 to 0.013 $\mu\text{g kg}^{-1}$ dw were reported for 6:2 Cl- PFESA in sediments collected from Korean coasts (Lee et al., 2020). ADONA and GenX were not detected in any sediment or fish samples, which is consistent with previous studies (Hassell et al., 2020; Lee et al., 2020). The bioaccumulation of these compounds remains unclear (Munoz et al., 2019).

3.7. Estimated intakes and potential human health risks of PFOS exposure

Concentration thresholds for PFOS and PFOA have become increasingly conservative over recent years (EFSA, 2018b; EFSA, 2020) as exposure to these chemicals is known to lead to adverse health effects (Sunderland et al., 2019). A decreased response of the immune system to vaccinations was found to be one of the most critical human health effects reported by EFSA (EFSA 2020). The most recent risk assessment carried out by EFSA has set a new safety threshold for four PFAS: PFOA, PFOS, PFNA, and PFHxS, that accumulate in the body (EFSA, 2020). A group tolerable weekly intake (TWI) of 4.4 ng per kilogram of body weight per week has been set as the maximum tolerable intake from food. The potential risk associated with the consumption of fish from the study site here was investigated in relation to this EFSA threshold (EFSA, 2020).

PFOA, PFNA, PFHxS, and PFOS were detected in muscles of the edible fish studied here. To determine the risk from these four compounds (\sum_4 PFAS) in fish consumed by humans in the Saudi coastal city Jeddah, information on fish consumption rates

Table 1

Average weekly intake (WI) and its percentage for the sum of four PFASs (PFOS, PFHxS, PFNA and PFOA) of muscles of fish species collected from Jeddah coastal waters assuming mean meal size for each fish species and 70 kg body weight. Values below LOQ were set to 0 for TWI calculations.

Species	WI [ng kg ⁻¹ bw week ⁻¹]	Relative to TWI [%]
Doublespotted queenfish	5.1	115
Strongspine silver-biddy	2.9	66
Bonefish	4.2	96
Bigeye scad	1.3	29
Marbled spinefoot	3.8	87
Bluefin trevally	3.9	88

(including fish species similar to the ones investigated in the current study) for Jeddah was used (Burger et al. (2014)). Calculated weekly intake (WI) of \sum_4 PFAS for fish investigated ranged from 1.3 to 5.1 ng kg⁻¹ bw week⁻¹ (Table 1).

A comparison of the calculated WI values for \sum_4 PFAS with the values from EFSA showed that the intake for doublespotted queenfish exceeded the EFSA threshold from fish consumption. This finding suggests that some of the fish species consumed in Jeddah could represent a potential risk to humans living in Jeddah, Saudi Arabia considering 2.65 fish meals per week (Burger et al., 2014b).

Importantly, the weekly intake calculated here just applies to \sum_4 PFAS from the investigated seafood, while the new EFSA TWI is set for the presence of these substances in all food sources including fruit and egg products, as well as for drinking water. In addition, preparation of the fish (cooking, fileting, backing, etc) was not considered. Earlier studies confirm that cooking appeared to increase PFAS concentrations in fish muscle due to loss of moisture during cooking (Bhavsar et al., 2014; Vassiliadou et al., 2015). However, cooking of fish is not an effective approach to reduce dietary exposure to the proteins associated PFASs, since the PFAS amount, the consumer is finally exposed to, is not changed (Bhavsar et al., 2014).

This preliminary risk assessment revealed that human exposure to PFASs through fish consumption in the coastal city Jeddah may be of concern. Thus, further risk assessment is warranted for protecting local consumers from uncontrolled PFAS exposures.

4. Conclusion

Though the occurrence of PFASs in sediment and in fish has been extensively studied, only sparse information currently exists related to concentrations in Middle East environments. The current study provides baseline information on the levels of these compounds in sediment and fish tissues from the Eastern Red Sea coastal waters off Jeddah. Several legacy PFASs were found to be widely distributed in the Saudi coastal waters of the Red Sea, while emerging PFASs were rarely found. The discharge of partially treated wastewater in the region was identified as the main source of these compounds. Firefighting training was considered an additional source for some PFASs. The contamination profile was mostly dominated by PFOS, but short-chain PFASs (PFBA and PFBS) and 6:2 FTS were frequently detected. Several fish species investigated in the current study have the potential to accumulate 6:2 FTS and PFBS. For the fish species investigated here, the correlation between PFAS concentration and fish size (weight and length) was tissue- and species-dependent. This is also true for the PFAS profile and levels. The dietary intake of PFOA, PFNA, PFHxS, and PFOS for the muscle of doublespotted queenfish exceeded the EFSA safety threshold set for these substances in food, indicating a potential risk to humans living in Jeddah, Saudi Arabia. However, due to the

relatively small sample numbers, further work would be needed to verify these findings. In addition, the trace analytical method used here did not allow the detection and quantification of other relevant polyfluorinated precursors such as FTOHs, FTS and FSA related PFASs. Therefore, further research is recommended to address these limitations. Finally, exploring concentrations of PFAS in direct industrial wastewater influents would provide additional relevant information.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envpol.2021.116935>.

Author contribution statement

Conceptualization, Methodology, Writing – original draft preparation: AMA with support from MS and RK. Fieldwork and sampling planning: WMA and RK, SSA and AMA. Critical Review, writing and editing: MS, CPH, SEH, WMA, SSA, EMR, HAL, and RK. Funding acquisition: MS, SEH and RK.

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