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Tissue specific uptake and elimination of perfluoroalkyl acids (PFAAs) in adult rainbow trout (*Oncorhynchus mykiss*) after dietary exposure

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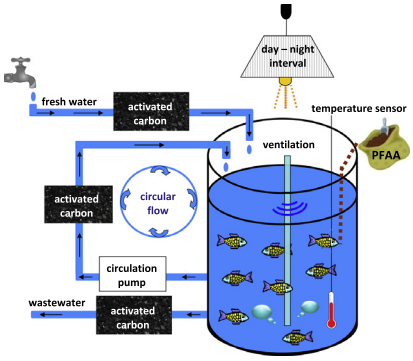
HIGHLIGHTS

- Tissue specific uptake and elimination of PFAAs in adult rainbow trout.
- Dietary exposure via spiked food over a time period of 28 d (accumulation period).
- Accumulation period was followed by a 28 d depuration period (PFAA free food).
- Muscle, liver, kidneys, gills, blood, skin and carcass were examined individually.
- Estimation of tissue specific elimination half-lives for each substance.

GRAPHICAL ABSTRACT



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ABSTRACT

Tissue specific uptake and elimination of perfluoroalkyl acids (PFAAs) were studied in rainbow trout (*Oncorhynchus mykiss*). Adult trout were exposed to perfluorobutane sulfonic acid (PFBS), perfluorohexane sulfonic acid (PFHxS), perfluorooctane sulfonic acid (PFOS), perfluorooctanoic acid (PFOA) and perfluorononanoic acid (PFNA) via food over a time period of 28 d. In the following 28-d depuration period the fish were fed PFAA-free food. At defined sampling times four animals were removed from the experimental tank, euthanized and dissected. Muscle, liver, kidneys, gills, blood, skin and carcass were examined individually. At the end of the accumulation phase between 0.63% (PFOA) and 15.5% (PFOS) of the absolute, applied quantity of PFAAs was recovered in the whole fish. The main target organ was the liver with recovery rates between 0.11% (PFBS) and 4.01% (PFOS) of the total amount of ingested PFAAs. Perfluoroalkyl sulfonic acids were taken up more readily and had longer estimated elimination half-lives than perfluoroalkyl carboxylic acids of the same chain length. The longest estimated elimination half-lives were found to be for PFOS between 8.4 d in muscle tissue and 20.4 d in the liver and for PFNA between 8.2 d in the blood and 11.6 d in the liver.

1. Introduction

PFAAs are ubiquitously found in watery milieus (Gellrich et al., 2013; Kim and Kannan, 2007; Liu et al., 2009), and consequently they are distributed throughout the global water pathways. Thus,

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especially aquatic organisms are exposed to PFAAs. Shorter chain PFAAs exhibit a relatively high aqueous solubility and a relatively weak binding affinity to organic material. The sorption potential of PFAAs rises with an increase in hydrophobic fluorocarbon chains, resulting in an increase in the sediment-water distribution coefficient of 0.5–0.60 log units with each CF2 moiety. The sediment-water distribution coefficient of perfluoroalkyl sulfonic acid is 0.23 log units higher than that of the corresponding perfluoroalkyl carbonic acid (Higgins and Luthy, 2006). Studies have shown that PFAAs tend to bioaccumulate in aquatic organisms, dependent upon their chain length and functional group, according to the previously described sorption potential (Conder et al., 2007; Houde et al., 2011). The toxicokinetics of PFAAs in aquatic organisms are still not well understood. Studies on the uptake and elimination behavior after dietary exposure of PFAAs in aquatic organisms have, to the best of our knowledge, only been undertaken on juvenile rainbow trout (De Silva et al., 2009; Martin et al., 2003a,b). In juvenile trout the elimination of PFAAs after dietary exposure appears to decrease with increasing chain length and perfluoroalkyl sulfonates generally appear to exhibit slower elimination rates than perfluoroalkyl carboxylic acids of comparable chain length (De Silva et al., 2009; Martin et al., 2003b). Martin et al. (2003b) explicitly indicate that these findings should not be extrapolated to larger fish. Therefore the present study represents an evaluation of data in regard to substance and tissue specific uptake and elimination behavior of PFAAs in adult rainbow trout after dietary exposure based on a biomagnification study by Goeritz et al. (2013). The aim of this study is to describe the tissue specific uptake and to estimate the tissue specific PFAA elimination half-lives in adult rainbow for comparison of the data with the PFAA elimination half-lives reported in the literature for juvenile trout (De Silva et al., 2009; Martin et al., 2003b). In addition, the percentages of recovery in the tissues of adult rainbow trout at the end of the accumulation phase will be presented in relationship to the absolute amounts of PFAAs applied. The PFAA concentrations measured in muscle tissue at the end of the accumulation phase will be compared with the levels of PFAAs found in market fish. The potential health risk for consumers resulting from the consumption of PFAA contaminated fish will be considered and discussed in relationship to the tentative tolerable daily intake (TDI) for PFOS and PFOA as suggested by the European Food Safety Authority (EFSA, 2008) Finally, the concentration that would be required to reach 100% of the TDI will be calculated.

2. Methods

The current paper and the paper by Goeritz et al. (2013) are based on the same trout feeding study carried out jointly by the Fraunhofer Institute for Molecular Biology and Applied Ecology (IME) and the Hessian State Laboratory. The fish feeding experiment presented in the manuscript (incl. experimental setup, fish rearing, food spiking, fish feeding and fish sampling) was carried out by Ina Goeritz scientifically and technically supported by Fraunhofer IME and a detailed description of the fish feeding experiment and an evaluation of the data on fish growth performance were already presented by Goeritz et al. (2013). Sample preparation as well as PFAAs analysis of all food, tissue and water samples took place at the Hessian State Laboratory.

2.1. Standards

Analytes in this study include perfluorobutane sulfonic acid (PFBS), perfluorohexane sulfonic acid (PFHxS), perfluorooctane sulfonic acid (PFOS), perfluorooctanoic acid (PFOA) and perfluorononanoic acid (PFNA). The criteria for selection of test substances are described in detail in Goeritz et al. (2013). A table with the full name, abbreviation, chemical purity and supplier of each standard and each mass-labeled internal standard is included in the supplementary material (Table S1). The reagents used in this study are also described in detail in the supplementary material.

2.2. Study design

PFAA spiked fish food (2.6% of the mean live weight of the fish per day as recommended by the fish food manufacturer) was fed throughout the 28-d accumulation phase. From day 28 on and throughout the whole depuration phase non-spiked fish food was fed accordingly. Throughout the whole study, including the accumulation and depuration phase, a total of eight samplings took place (on day 0, 7, 14, 28, 31, 35, 42 and 56) in which four fish were caught, euthanized, dissected and prepared for analysis of PFAAs. The first sample (on day 0) was used to rule out possible background PFAA contamination of fish before commencing the feeding study (blank value control).

2.3. Fish food

Production and preparation of the test food was performed according to Goeritz et al. (2013) and is briefly described in the supplementary material. The amount of food administered depended upon the current mean live weight of the fish present in the experimental tank (see the section "Study design"). For this calculation the mean weight of the fish removed from the tank for sampling was assumed to be representative of the mean weight of the remaining fish in the tank.

2.4. Fish rearing

Fish rearing and control of water quality took place in tanks of the Fraunhofer Institute for Molecular Biology and Applied Ecology (IME, Schmallenberg, Germany). Fish rearing and water quality criteria are described in detail in the supplementary material.

2.5. Sampling

Fish were selected randomly and immediately after removal from the experimental tank weighed and anesthetized in a water bath containing 150 mg L^{-1} ethyl 3-aminobenzoate methanesulfonate (MS 222, Sigma Aldrich) (Goeritz et al., 2013). After removal from the narcosis basin, blood was withdrawn immediately by cardiocentesis and the anesthetized fish were euthanized by cutting the spine. Tissue samples for analysis (muscle, skin, liver, kidneys and gills) were removed and like the blood weighed and then frozen at −18 °C (±1 °C) in Falcon tubes (VWR, Darmstadt, Germany) awaiting further sample preparation. The remaining part of the fish (carcass) was freed of any remaining undigested PFAAs bearing food by flushing the stomach and intestinal tract with demineralized water and removal of the pyloric caeca. The carcass was also weighed and frozen in polypropylene bags. At the beginning of the study (see the section "Study design") whole fish were removed from the tank and the abdomen opened by ventral section, the pyloric caeca were removed and the stomach/intestinal tract emptied and flushed with demineralized water. These whole fish were also packed in polypropylene bags and frozen. The weight of each fish removed from the experimental tank for sampling was recorded. Water samples (100 mL) were collected from the experimental tank at the beginning of the study and subsequently twice weekly in order to rule out PFAA cross-contamination from the water. The water samples were frozen at −18 °C (±1 °C) until they could be analyzed.