

Co-Occurrence, Bioaccumulation, and Dietary Risk Assessment of Per- and Polyfluoroalkyl Substances and Heavy Metals in Rice

Samir Attia,^{a,b,c} Zhiyuan Hu,^{a,b} Fangjie Qi,^{a,b} Dongling Li,^a Frederic Coulon,^d Xin Song^{a,b,*}

^a State Key Laboratory of Soil and Sustainable Agriculture, Changshu National Agro-Ecosystem Observation and Research Station Institute of Soil Science, Chinese Academy of Sciences, Nanjing 211135, China

^b University of Chinese Academy of Sciences, Beijing 100049, China

^c Water Relations and Field Irrigation Department, National Research Centre, Dokki 12622, Cairo, Egypt

^d School of Water, Energy and Environment, Cranfield University, Cranfield MK43 0AL, United Kingdom

*Corresponding author: Xin Song

Email: xsong@issas.ac.cn

Phone: +86-25-86881543, Fax: +86-25-86881126

Address: No.298 Chuangyou Road, Jiangning District, Nanjing, 211135, China

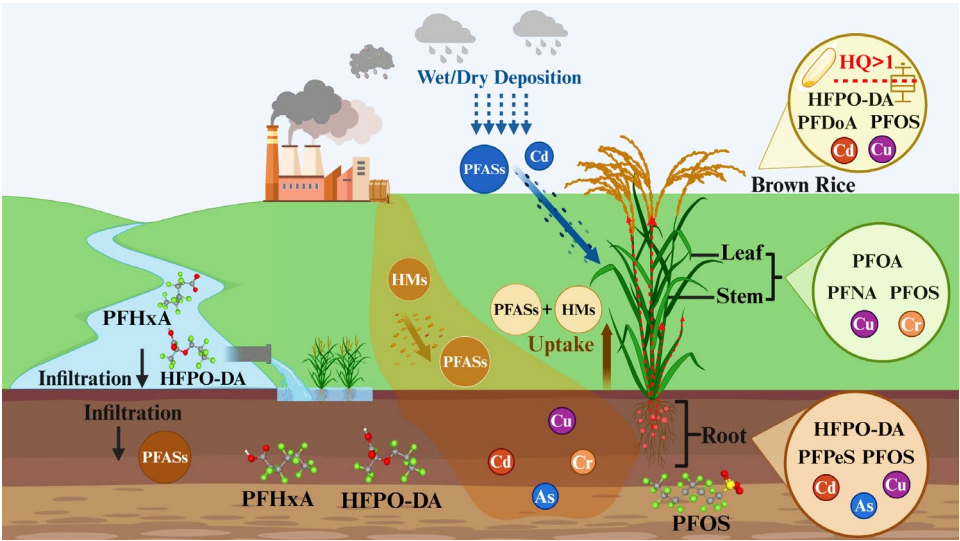
S.A. and Z.H. are contributed equally to this work and should be considered as cofirst authors.

ABSTRACT

The co-occurrence and combined dietary health risks of per- and polyfluoroalkyl substances (PFASs) and heavy metals (HMs) in rice remain poorly understood. This study analyzed 29 PFAS compounds and 4 HMs in rice tissue collected from paddy fields. HFPO-DA was dominant in brown rice (4.84-15.99 ng/g), while PFOA primarily accumulated in leaves and straw. Cd and Cu showed elevated grain concentrations. PFASs exhibited higher bioaccumulation and translocation factors than HMs, especially in aboveground tissues, whereas Cd and As accumulated most in roots. Correlation analyses suggest dual root and foliar uptake contributed to HFPO-DA and Cd grain accumulation. Dietary assessment revealed that HFPO-DA, PFOS, Cd, and Cu in brown rice exceeded safety thresholds across different age groups. Specifically, the cumulative hazard index (HI) reached 10.7 in brown rice for toddlers, twice that of white rice, raising significant health risk concerns. These findings provide critical insights for future food safety monitoring and risk management.

Keywords: PFASs; heavy metals; bioaccumulation; translocation health risk assessment

TOC Graphic



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1. INTRODUCTION

Per- and polyfluoroalkyl substances (PFASs) have been extensively used in industrial applications and commercial products, and their persistence across various environmental media has been well documented.^{1–3} Several studies have linked PFAS exposure to a range of adverse health outcomes, prompting many countries to develop policies and regulatory guidelines to mitigate their impacts.^{4–6} Alongside PFASs, industrial discharges and agricultural activities can release legacy pollutants such as heavy metals (HMs), particularly in regions where fluorinated materials are produced or used (e.g., hexafluoropropylene oxide dimer acid (HFPO-DA), perfluorooctanoic acid (PFOA), and perfluorooctanesulfonic acid (PFOS)).^{7,8} Both PFASs and HMs are now increasingly prevalent in agricultural regions worldwide, including China,^{9,10} the United States,¹¹ and Europe,¹² where their presence in soils, waters, and crops poses risks to ecosystems and human health.^{13–16} The environmental behaviors of individual PFASs and HMs have been explored in limited studies; however, and critical data gaps remain regarding their co-occurrence patterns, bioaccumulation, and combined impacts on plants and human health.^{10,17}

Rice, as a globally important staple, has been shown to be an efficient accumulator of both HMs^{13,18} and PFASs,^{1,19} thereby representing a significant dietary exposure pathway and posing serious risks to human health.^{13,20} Although the relative contributions of root versus foliar uptake remain unclear, recent studies have shown that PFAS uptake occurs mainly via root absorption through anion channels and aquaporins,^{21–24} while the specific transporter genes remain unidentified.²⁴ In contrast, HM uptake and translocation mechanisms, particularly for cadmium

(Cd) and chromium (Cr), are relatively well understood, involving competition with essential nutrient transporters and systemic partitioning processes.^{25–28} Moreover, foliar uptake of airborne Cd has been recognized as an additional pathway in rice paddies near industrial sources.^{29,30} While these mechanistic insights are well established for individual contaminants,^{13,26,31–34} the distribution and coaccumulation of PFASs and HMs in rice, especially in edible grains, remain poorly understood.

Furthermore, while the estimated daily intakes (EDIs) of HMs from rice consumption have been extensively studied, current efforts primarily focused on drinking water quality guidelines, with only limited regulatory standards for PFASs in crops.³⁵ For instance, the European Food Safety Authority (EFSA) established a tolerable intake of 0.63 ng/kg bw/day for the sum of PFOA, PFOS, perfluorononanoic acid (PFNA), and perfluorohexanesulfonic acid (PFHxS) in 2020.³⁶ Similarly, the U.S. Environmental Protection Agency (EPA) proposed a reference dose (RfD) of 30 ng/kg bw/day for HFPO-DA in 2023.³⁷ However, most PFAS congeners found in food matrices remain undefined, and rice-specific EDI benchmarks are currently missing, underscoring the urgent need to evaluate contaminant mobility in rice and dietary exposure risks across populations.

In this study, rice samples, along with soil and irrigation water samples, were collected from rice paddy fields near an industrial cluster to investigate the co-occurrence, bioaccumulation, and dietary risk assessment of PFASs and HMs in different rice tissues. The specific goals are to (1) quantify the concentrations of PFASs and HMs in different rice organs, thereby elucidating their co-occurrence patterns; (2) shed light into the relative contributions of PFASs

and HMs uptake patterns to their concurrent accumulation in rice grains, and (3) evaluate the potential dietary risks associated with the combined presence of PFASs and HMs. By linking contaminant uptake patterns with dietary risk assessment, this study offers critical insights into managing human health exposure to emerging and legacy contaminants through rice consumption.

2. MATERIALS AND METHODS

2.1. Study Area Description

The study area is located in Changshu City, Jiangsu Province, China (31.658308°N lat., 120.765132°E long., Figure 1), a region well known for its equipment, textile, and clothing manufacturing industries.³⁸ Rice paddies are widespread, supported by extensive surficial clay soils that make the area a major rice production zone.^{39,40} The sampling locations were located next to an industrial cluster, which occupies an area approximately 4 km long and nearly 2 km wide. The industrial cluster comprises various companies, such as printing and dyeing, textile, mechanical, electronics firms, metallurgists, optical fiber, and packaging factories. In November 2023, rice, soil, and irrigation water samples were collected from five different rice paddy fields, labelled as ST1-ST5 (Figure 1), within the sampling area. It is worth noting that the Dajin fluorine-chemical industry park, primarily manufacturing polytetrafluoroethylene and polyvinylidene fluoride, is located ~8 km northwest of the sampling area, situated along the Yangtze River.

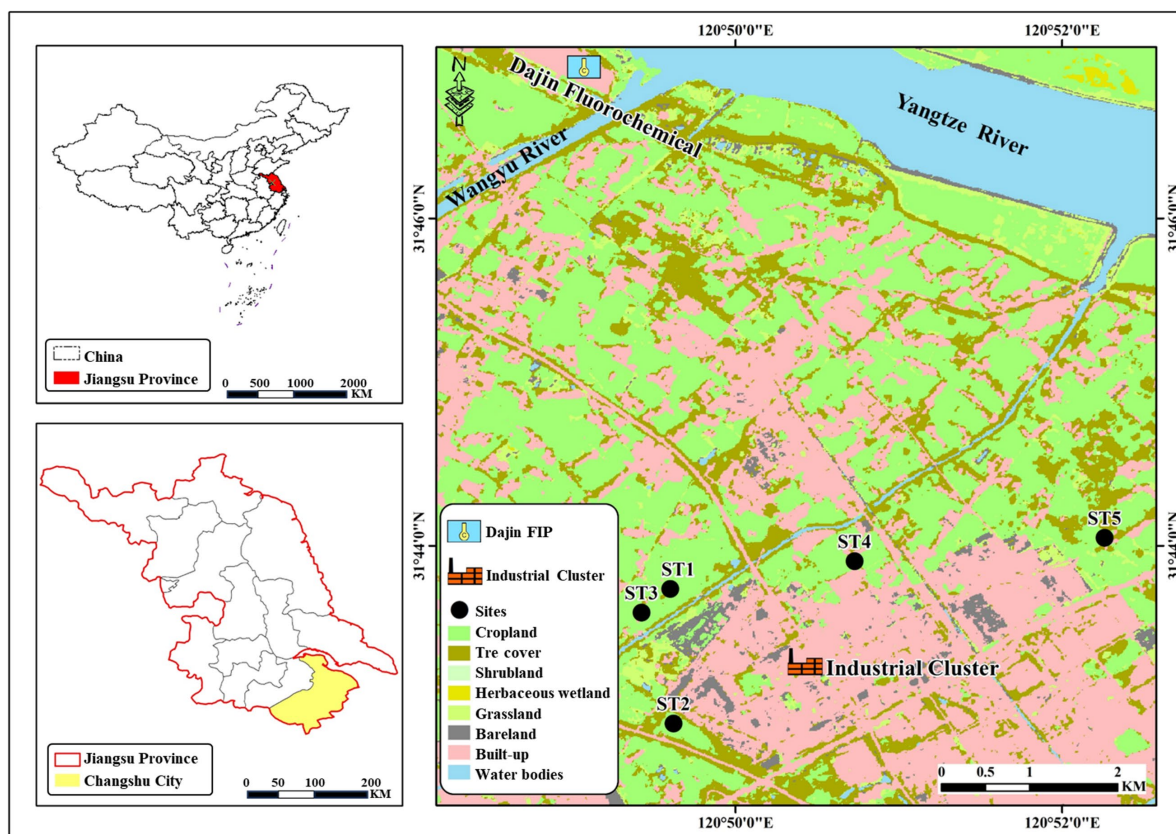


Figure 1. Map of the sampling area in Changshu City, Jiangsu Province, China.

2.2. Sampling Campaign and Collection

During the field sampling, nine rice plant samples and their corresponding nine rhizosphere soil samples (0-20 cm) were collected from the different locations (ST1-ST5) (Figure 1). The number of samples at each location was determined based on the field lot size, with 3 samples from ST1, 2 samples from ST2, and 1 sample each from ST3 through ST5. To ensure representativeness, each sample consisted of rice plants collected from four orientations, resulting in a composite sample. In addition, seven surface water samples were collected from these 5 locations, with 2 samples were collected each at ST1 and ST2 to assess sampling representativeness, while 1 sample was collected at ST3 through ST5 (Figure S1). It is worth noting that the key irrigation period occurs in the months of August through October in the study

area. However, previous studies conducted in the same study area have reported a similar concentration profile across different months, including April, May, and June.^{41,42} All samples were preserved at -4°C and transported on ice boxes for PFASs and HMs analysis following the USEPA 1633 and USEPA 3052 modified methods, respectively.^{43,44} To prevent external contamination, procedural and solvent blanks were included with each sample batch during various stages, including field sampling, transportation, extraction, and analysis.^{45,46} More information for sample collection and replicate analysis verification can be found in Text S1 and Figure S1.

2.3. Standards, Chemicals, and Reagents

The specific 29 target PFAS compound names and their abbreviations, and 13 mass-labeled extracted internal standards (EIS) are provided in Table S2. All PFAS standards were purchased from Wellington Laboratories, Canada, with 99% purity. The sample pretreatment involved using methanol and water of high-performance liquid chromatography (HPLC) grade purchased from Merck Co., Germany, and Tianjin Co., China, respectively. Solid phase extraction was performed using Waters Oasis[®] WAX cartridges (6 cc, 150 mg) from Waters Corp., USA. The extraction cleanup process utilized super clean Envi-Carb powder from Sigma-Aldrich Co., USA.

In the preliminary experiment of this study, analytical analysis was conducted for a total of 8 HMs ions, including Cd, Cr, copper (Cu), arsenic (As), lead (Pb), zinc (Zn), nickel (Ni), and cobalt (Co), among which Cd, Cr, Cu, and As showed notably elevated concentrations and exceeded their relevant guideline levels. Therefore, Cd, Cr, Cu, and As were selected for

subsequent analysis and evaluation due to their ecological risks and frequent detections in soils and rice systems in China.^{18,29,47}

All reagents employed in this study, including ammonium hydroxide solution (NH₄OH), formic acid (CH₂O₂), concentrated acetic acid (CH₃COOH), formic acid (CH₂O₂), hydrofluoric acid (HF), hydrogen peroxide (H₂O₂), nitric acid (HNO₃), and perchloric acid (HClO₄), were used in high-purity grade.

2.4. Sample Preparation

The extraction of 29 target PFASs from different matrices followed the USEPA Method 1633.⁴⁴ For plant samples, tissues (roots, stems, leaves, brown rice, and polished rice) were thoroughly washed with ultrapure water, segregated, lyophilized by frozen machine, and pulverized into a fine powder. A 2.5 g subsample dry weight (dw) was weighed for extraction. The resulting extracts were filtered through a 0.22 µm membrane, transferred to sample vials, and stored at -20 °C until analysis. For soil samples, extraction was performed using wet soil (equivalent to 5.0 g dw) to maintain analyte integrity. The soil extraction followed the identical protocol as described for the plant samples. For irrigation water, 500 mL of each sample was prepared for analysis. Detailed extraction procedures and instrumental parameters are described in Text S2. HMs in all samples were determined following the USEPA Method 3052.⁴³ For plant samples, 0.25 g (dw) of the lyophilized powder was used for digestion. For soil samples, to ensure homogeneity, samples were oven-dried at 60 °C, ground, and passed through a 100-mesh sieve. 0.2 g of the sieved dry soil was then weighed for subsequent microwave-assisted digestion.⁴⁸

For irrigation water, 10 mL of each sample was acidified with 2% HNO₃ (v/v) and filtered through a 0.22 µm membrane prior to analysis. Further details regarding the digestion protocols and quality assurance/quality control (QA/QC) are provided in Text S3.

2.5. Analytical Methods

The 29 target PFAS substances were analyzed using Acquits UPLC coupled to API 5500 triple-quadrupole mass spectrometer (AB SCIEX Inc., Framingham, MA, USA) according to previous studies,^{46,49} with details provided in Table S3. Concentrations of HMs were detected using inductively coupled plasma mass spectrometry (ICP-MS 2000C), with detection limits ranging from 0.1 to 200 ppb.⁵⁰ Water samples were analyzed to determine the pH, electrical conductivity (EC), dissolved oxygen (DO), and total suspended solids (TSS). Soil pH was measured using a 1:2.5 (w/v) soil-to-water suspension, while a 1:5 (w/v) ratio was used for EC and DO detection. Soil moisture content was quantified using a 10 g sample oven-dried at 60 °C, following the outlined method by Radojevic and Bashkin.⁵¹ Detailed results can be found in Table S3.

2.6. Data Analysis

Principal component analysis (PCA) was performed to elucidate multidimensional relationships among PFAS and HM concentrations in irrigation water, soil, and rice (using brown rice as a representative rice tissue), and to identify the dominant pathway for PFAS uptake within rice plant tissues. Bioaccumulation factors (BAFs), defined as the dry-weight-based ratios of chemical concentrations in specific crop organs to those in the corresponding soil, were calculated using (eq. 1). This calculation was performed under the assumption that soil uptake

via plant roots represents the dominant pathway for PFASs accumulation, with atmospheric exchange deemed insignificant.^{31,52,53} Translocation factors (TFs) were quantified as the ratio of chemical concentrations in aboveground organs to those in roots, as shown in (eq. 2).^{31,52,53}

$$BAF = C_{\text{organs}}/C_{\text{soil}} \quad (1)$$

$$TF = C_{\text{aboveground}}/C_{\text{root}} \quad (2)$$

where C_{organs} refers to PFASs concentration (ng/g) and HMs (mg/kg) in individual rice organs, including root, straw, leaves, brown and white rice. $C_{\text{aboveground}}$ refers to PFASs concentration (ng/g) and HMs (mg/kg) in the individual aboveground tissue, including straw, leaves, brown rice, and white rice. C_{soil} and C_{root} are PFASs concentrations (ng/g) and HMs (mg/kg) in the corresponding soil and rice plant roots.

To evaluate dietary exposure, the EDIs for PFASs and HMs via rice consumption were calculated for three distinct age groups: toddlers, children & teenagers, and adults. These calculations were conducted using Eq. (3)¹⁶, which integrates the average concentration of PFAS and HMs among different locations, age-specific body weights, and rice consumption rates to account for physiological and dietary variations across different life stages. Given the elevated vulnerability and the considerable uncertainty in exposure parameters for the youngest population subgroup, a probabilistic risk assessment using Monte Carlo simulation was further conducted specifically for the toddlers. The simulated EDI values were subsequently utilized to compute the hazard quotient (HQ, Eq. (4)) and the total hazard index (HI) for cumulative exposure, as defined by Eq. (5).¹⁵

$$191 \quad \quad \quad EDI = (C_{\text{grains}} \times IR) / BW \quad (3)$$

$$192 \quad \quad \quad HQ = EDI / RfD \quad (4)$$

$$193 \quad \quad \quad HI = HQ_{\text{PFAS}} + HQ_{\text{HM}} \quad (5)$$

194 where C_{grains} refers to the average PFASs concentration (ng/g) and average HMs (mg/kg) in
 195 individual brown and white rice among different locations, IR is the average intake rate (g/day),
 196 BW refers to the average human body weight for each group, and RfD is the reference dose for
 197 PFASs (ng/kg bw/day) and HMs ($\mu\text{g/kg bw/day}$). IR and BW used in this study were 151 g/day
 198 and 17.5 kg for toddlers, 306 g/day and 38.8 kg for teenagers & children, and 242.3 g/day
 199 and 62.9 kg for adults,^{1,16,54} with more details outlined in Table S4. The hazard quotients of
 200 individual PFASs and HMs are denoted as HQ_{PFAS} and HQ_{HM} , respectively.

201 **2.7. Quality Assurance and Quality Control (QA/QC)**

202 To prevent cross-contamination, lab background PFAS contamination was checked, and
 203 laboratory Milli-Q water blank samples were prepared to detect any external contamination
 204 during sample extractions. Blank samples and laboratory control spikes (LCS) were prepared
 205 for the extraction and analytical stages. The limit of detection (LOD) and quantification (LOQ)
 206 were calculated for all PFAS substances. The matrix spike recoveries (MSRs) ranged from 69%
 207 to 121% for water, 73% to 124% for soil, and 58% to 119% for plants, which are considered
 208 acceptable according to the 1633 analysis method limits.⁴⁴ Detailed QA/QC results have been
 209 sorted out in Table S5. For HMs analysis, blank samples and standard reference materials
 210 GBW10049 (GSB-27) for plant and GBW07405a (GSS-5a) for soil were included in every

batch of the analyses. The recovery ratios of standards ranged from 84% to 117% and 81% to 100% for plant and soil, respectively. Detailed results have been sorted out in Table S6.

2.8. Statistical Analysis

A regression analysis was performed to examine the correlation between specific PFAS and HMs concentrations detected in brown rice and their corresponding levels in both leaf and root tissues. The analyses were conducted using Origin 2021 software. The Monte Carlo simulation model was utilized to measure and minimize the uncertainty in evaluating health risks for toddlers. Average daily rice intake, body weight, and specific PFASs and HMs concentrations were input into Oracle Crystal Ball (version 11.1.3.0) software and integrated through a Monte Carlo simulation with 20,000 iterations to measure and minimize the uncertainty in calculations or evaluations of cumulative health risks at a 95% confidence interval.^{14,20}

3. RESULTS AND DISCUSSION

3.1. Co-occurrence and Distribution Patterns of PFASs and HMs in Rice Farmlands

3.1.1. PFASs and HMs Concentrations in Different Rice Organs

The concentrations and distributions of total PFASs (Σ_{29} PFASs) (designated as letters a-e on the x-axis) and 4 HMs ions (designated as letters f-j on the x-axis) in different rice organs at ST1-ST5 were presented in Figure 2. The compositions of PFASs shared a similar profile across different sampling locations, yet the concentrations of Σ_{29} PFASs varied, with the highest total PFASs concentration observed at the sampling location No. 5 (Figure S2). The average concentrations of Σ_{29} PFASs in different rice tissues followed the order: leaf > straw > root >

231 brown rice > white rice (Figure 2a-e and Table S7), with mean values ranging from 4.75 to
232 135.93 ng/g. The observed PFASs distribution pattern among different rice organs is similar to
233 that reported by Yamazaki et al.,²⁰ in Kasaath (*Indica*) rice, where PFASs were more enriched
234 in the straw than in the root, with white rice having the lowest concentration. The PFAS
235 compounds in brown rice and white rice exhibited similar profiles, and the top five PFAS
236 compounds consistently followed the order of HFPO-DA (C6, trade name GenX),
237 perfluorododecanoic acid (PFDoA, C12), perfluorooctanesulfonic acid (PFOS, C8),
238 perfluorononanoic acid (PFNA, C9), and perfluorohexanoic acid (PFHxA, C6). HFPO-DA, an
239 alternative to legacy PFOA, was the most abundant PFAS detected, with concentrations ranging
240 from 4.84 to 15.99 ng/g in brown rice and from 1.60 to 3.82 ng/g in white rice across different
241 locations.

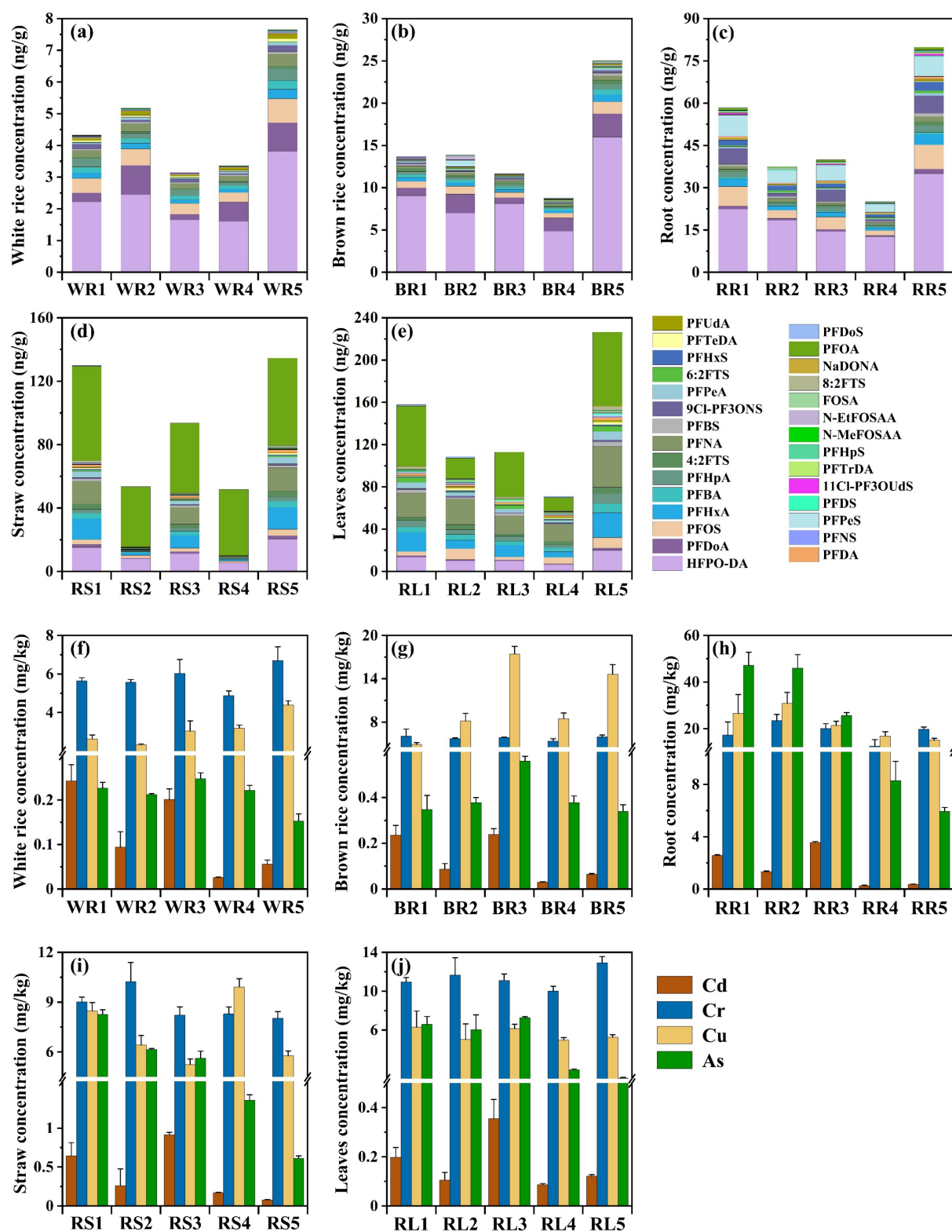


Figure 2. Concentrations and distribution of PFASs (ng/g) in rice organs: (a) white rice, (b) brown rice, (c) roots, (d) straw, and (e) leaves; and HMs (mg/kg) in rice organs: (f) white rice, (g) brown rice, (h) roots, (i) straw, and (j) leaves at five different locations (ST1-ST5).

243 The average HFPO-DA concentrations were 9.00 ng/g in brown rice and 2.35 ng/g in white rice,
244 accounting for 60% and 49% of the total PFASs, respectively (Figure S3a). The other dominant
245 PFAS compounds also showed notable concentrations included PFDoA (1.64 ng/g in brown
246 rice, 0.58 ng/g in white rice), PFOS (0.86 ng/g in brown rice, 0.47 ng/g in white rice), PFNA
247 (0.32 ng/g in brown rice, 0.23 ng/g in white rice), and PFHxA (0.48 ng/g in brown rice, 0.18
248 ng/g in white rice) (Table S7). This finding contrasts with the study by Kim et al.,⁵⁵ which
249 identified PFOA and PFNA as the dominant PFASs in brown rice, likely due to variations in
250 rice cultivars, soil characteristics, and PFAS levels in irrigation water. Notably, in this study,
251 HFPO-DA represented a major proportion of PFASs in rice grains, whereas PFOA was nearly
252 undetectable, underscoring the growing importance of emerging PFAS substitutes and the need
253 for further investigation.

254 In rice roots (Figure 2c), the dominant PFAS compounds followed the order of HFPO-DA
255 (20.58 ng/g on average), perfluoropentanesulfonic acid (PFPeS, 5.49 ng/g), PFOS (4.87 ng/g),
256 chlorohexadecafluoro-3-oxanone-1-sulfonic acid (9Cl-PF3ONS, trade name F-53B, 3.57 ng/g),
257 and PFHxA (2.12 ng/g). Among these, HFPO-DA, with concentrations ranging from 12.70 to
258 35.04 ng/g at all locations, accounted for nearly half of the total PFASs concentration. This high
259 accumulation is likely due to the direct contact between roots and soil, which serves as the
260 primary route of HFPO-DA contamination in the sampled paddy soils. The predominant PFAS
261 compounds in rice straws and leaves followed a similar pattern (Figure 2d-e), in the order of
262 PFOA, PFNA, PFOS, PFHxA, and HFPO-DA, which was significantly different from that
263 observed in rice roots and grains. PFOA exhibited the highest concentrations, measuring 47.68

264 ng/g in straw and 40.48 ng/g in leaves, followed by PFNA with concentrations of 8.11 ng/g in
265 straw and 23.69 ng/g in leaves. This pattern suggested efficient foliar accumulation of these
266 long-chain compounds, consistent with findings reported in a previous study.⁵⁶ PFOS was
267 detected at 2.39 ng/g in straw and 6.53 ng/g in leaves, while PFHxA showed concentrations of
268 7.58 ng/g in straw and 13.29 ng/g in leaves. However, it is worth noting that HFPO-DA,
269 although not one of the dominant compounds, has relatively high concentrations in straw (12.01
270 ng/g) and leaves (11.96 ng/g). Moreover, the concentrations of PFASs varied significantly
271 across different sampling locations, with the highest levels detected in rice from the fifth location
272 and the lowest in rice from the fourth location, reflecting the location-specific contamination
273 profiles.

274 Concentrations and distribution patterns of four HMs in rice tissues revealed substantial
275 variability (Figure 2f-j). The distribution pattern of HMs followed the order of roots > straw and
276 leaves > grains, suggesting a soil origin of HMs, in line with the observations of Dong et al.⁵⁷
277 As shown in Figure 2f and Table S8, the average concentrations of Cd (0.12 mg/kg) and As
278 (0.21 mg/kg) in white rice were relatively low, while Cr (5.76 mg/kg) and Cu (3.12 mg/kg) were
279 the dominant HMs. Conversely, brown rice exhibited similar levels of Cd (0.13 mg/kg) and Cr
280 (5.79 mg/kg) but significantly higher concentrations of As (0.40 mg/kg) and Cu (10.72 mg/kg),
281 indicating a notable decrease in these metals during the polishing process (Figure 2g). Brown
282 rice, along with rice straw and leaves (Figure 2i-j), showed greater HM accumulation,
283 potentially due to leaf stomata distribution and high evaporation rates, which likely contributed
284 to the accumulation of toxic pollutants.³² Rice roots showed the highest concentrations of Cd

(1.61 mg/kg), Cr (18.5 mg/kg), Cu (22.14 mg/kg), and As (26.60 mg/kg), indicating a tendency for roots to retain more HMs than aboveground tissues (Figure 2h). This is consistent with the fact that soil is the primary source of HMs absorbed by plants.^{34,57} The relative concentration percentage (Figure S3b) revealed that the rice roots tend to accumulate and enrich these HMs at higher levels compared to the aboveground parts. The tendency of higher concentrations of HMs to be retained in the roots compared to other tissues, and various pathways of HM translocation from the roots to the grains, have been studied extensively.²⁵ This difference in distribution patterns among rice organs between HMs and PFASs suggested that more uptake pathways are involved in the PFASs uptake, and the translocation mechanisms can be more complex, warranting further in-depth study in well-controlled experimental settings.

3.1.2. PFASs and HMs Concentrations in the Corresponding Soil and Irrigation Water

The Σ_{29} PFASs and 4 HMs concentration profiles in the soil and irrigation water at the 5 sampling locations were presented in Figure 3. In the current study, PFASs and HMs concentrations in corresponding soil and irrigation surface water were analyzed to explore their root uptake and accumulation in different rice organs. The PCA provided valuable insights into the complex environmental dynamics governing the bioaccumulation of PFAS and HMs in brown rice. In the PCA biplots (Figure S4), PC1 and PC2 explained 45.6% and 32.6% of the total variance in PFASs distribution, respectively, suggesting that both soil and irrigation water contributed to PFAS contamination in brown rice. Distinct clustering patterns were observed among individual PFAS congeners, a phenomenon likely driven by the structural heterogeneity inherent to PFAS compounds. Specifically, HFPO-DA, PFOS, and PFHxA showed divergent correlation profiles

306 between their concentrations in brown rice and occurrence in ambient environmental media. For
307 HMs, PC1 of 98.5% of the total variance, indicating that HM accumulation in the grain was
308 almost exclusively attributable to soil-derived inputs, with individual HM ions exhibited distinct
309 uptake and transport pathways within the rice plant. The findings in the current study are
310 consistent with those reported in previous studies.^{46,53,56} The Σ_{29} PFASs in the corresponding soil
311 ranged from 7.41 to 23.49 ng/g (Figure 3a and Table S9). PFOS was the dominant compound
312 in the soil, with concentration ranged (1.26 to 6.39 ng/g) among different locations, reflecting
313 its high affinity for soil particles due to its hydrophobicity and strong binding potential.⁵⁸ Other
314 notable PFASs in soil included PFHxA (0.96 to 3.22 ng/g), PFNA (1.00 to 2.41 ng/g),
315 perfluorodecanoic acid (PFDA, 0.72 to 1.91 ng/g), and perfluoroundecanoic acid (PFUdA, 0.68
316 to 1.81 ng/g), indicating that soils function as the primary reservoir and persistent source of
317 PFAS contamination, significantly influencing long-term bioaccumulation in crops.^{59,60}
318

Figure 3. Concentration profiles of PFASs and HMs in soil and water across different locations (a)
concentration distributions of PFASs in corresponding soil (ng/g), (b) concentration distribution of
PFASs in irrigation water ($\mu\text{g/L}$), and (c) concentration levels of different HMs in soil (mg/kg).

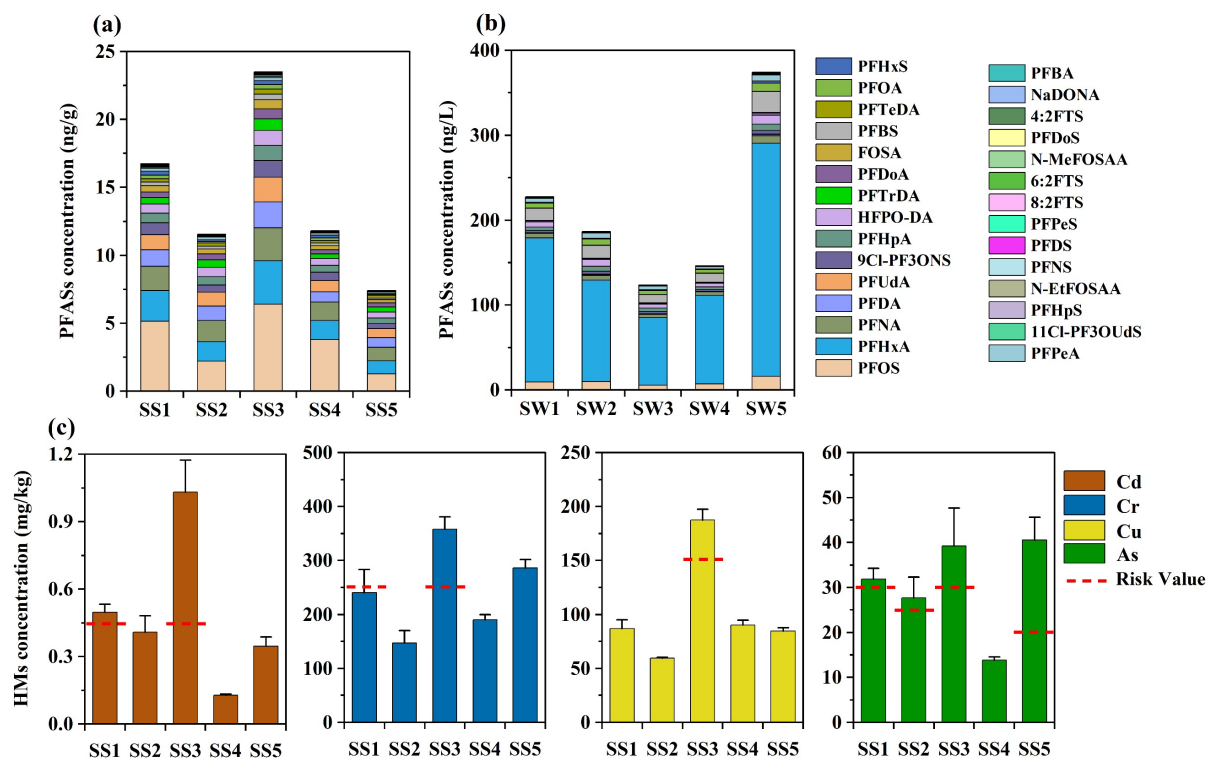


Figure 3. Concentration profiles of PFASs and HMs in soil and water across different locations (a) concentration distributions of PFASs in corresponding soil (ng/g), (b) concentration distribution of PFASs in irrigation water ($\mu\text{g/L}$), and (c) concentration levels of different HMs in soil (mg/kg).

Irrigation water showed significantly high PFAS levels, with an average total concentration of 211.71 ng/L, ranging from 123.46 to 374.20 ng/L (Figure 3b and Table S9). Among the 29 PFASs analyzed, PFHxA exhibited the highest average concentration in irrigation water (149.47 ng/L), making it the predominant compound, followed by perfluorobutanesulfonic acid (PFBS, 14.93 ng/L), PFOS (9.61 ng/L), HFPO-DA (6.62 ng/L), and PFOA (6.33 ng/L). These results were consistent with those of prior studies conducted in China,⁶¹ Japan,⁶² and the United States,⁶³ which also reported elevated PFHxA levels in irrigation water. Legacy PFASs, such as PFOA (C8) and PFOS (C8), were more prevalent in the corresponding soil, while short-chain compounds, including PFHxA (C6) and HFPO-DA (C6), were predominantly detected in

329 irrigation water. This distribution pattern suggested that long-chain PFASs, with their higher
330 molecular weight and hydrophobicity, are more likely to be retained in the soil,^{64,65} whereas
331 short-chain PFASs, characterized by greater water solubility, are more prone to migrate with
332 water flow.^{3,60} This also explained the high concentrations of HFPO-DA detected in rice roots,
333 despite its relatively low levels in the corresponding soil, as it likely migrated from the soil into
334 the roots during the rice growth cycle.⁶⁶

335 It is worth noting that the corresponding irrigation water and rice plants exhibited the highest
336 PFAS levels, while PFAS concentrations in the soil at location 5 (SS5) were the lowest among
337 all locations. In addition, the high levels of PFHxA in irrigation water corresponded to its
338 prominent detections in rice plant tissues, particularly in leaves and straw. This suggested that
339 irrigation water serves as a significant source of PFHxA in roots and that waterborne PFHxA
340 can enter plants through root uptake and subsequent translocation mechanisms.^{59,60} Therefore,
341 these phenomena suggested that irrigation water played an important role in the accumulation
342 of PFASs detected in rice plant, further supporting that both soil and irrigation water contributed
343 to PFAS contamination in brown rice. Moreover, these observed variations might be driven by
344 multiple factors, including differences in physicochemical properties (e.g., soil organic matter
345 and pH), crop-specific translocation mechanisms, and substitutional and functional groups of
346 PFASs.^{16,19,67}

347 Additionally, this study revealed minimal HM levels in irrigation water (Table S10), and all HM
348 concentrations were below the Chinese surface farmland irrigation water quality standards.⁶⁸

349 However, as shown in (Figure 3c, Table S11, and Table S12), the HM concentrations of the soil

significantly exceeded their related soil background values (BVs) in China.¹⁸ Notably, Cd levels in the soil were particularly elevated, with an average concentration of 0.48 ± 0.35 mg/kg across all sampling locations. At location SS3, the maximum Cd concentration reached 1.03 mg/kg, surpassing the risk control threshold of 0.4 mg/kg for contaminated farmland soil as per Chinese standards (15618-2018).⁶⁹ Cd contamination in soil poses a significant threat to rice quality, with the consumption of Cd-contaminated rice being a primary source of human exposure to this toxic metal, thereby posing serious health risks.⁴⁸ The concentrations of Cr, Cu, and As in soil also varied considerably across different locations, with exceptionally high levels of contamination observed at location (SS3). The average concentrations detected were Cr (357.77 mg/kg), Cu (187.49 mg/kg), and As (39.20 mg/kg), indicating substantial soil contamination. These elevated levels explain the high HM concentrations detected in rice roots, particularly compared to the risk screening values in Table S12. Notably, SS3 exhibited the highest concentrations of both PFASs and HMs, highlighting the potential for cocontamination of these pollutants. These findings emphasized that the dominant concentrations detected in rice tissues primarily originate from soil and irrigation water, highlighting the importance of addressing contamination in agricultural areas where PFASs and HMs coexist to ensure food safety and environmental health.^{70,71} Further research is essential to address these challenges and ensure food safety and environmental sustainability.^{24,72}

3.2. Bioaccumulation and Translocation Of PFASs and HMs in Different Rice Tissues

The log BAF values for PFASs and HMs have been illustrated in Figure 4a-c and summarized in Table S13. PFASs and HMs exhibited distinct accumulation patterns across plant tissues, and

a comparative analysis of their tissue distribution, despite their divergent chemical properties, can yield critical insights into their differential translocation potentials. In general, the BAFs of PFASs were significantly higher than those of HMs, exhibiting the trend of leaves > straw > root > brown rice > white rice (Figure 4a). In contrast, the BAFs of HMs followed the order of root > straw/leaves > brown rice > white rice (Figure 4b). The calculated BAFs for PFASs in aerial tissues were notably higher than those for HMs, which are intrinsically linked to their specific translocation mechanisms within the plant. PFASs in rice can be attributed to root uptake from soil and void water,^{55,60} whereas HMs are predominantly derived from soil, as discussed in the above PCA analysis. The highest accumulation of PFASs in leaves suggested that PFASs absorbed by rice roots from soil and irrigation water are efficiently translocated upward. This observation is consistent with conclusions from previous studies that PFASs can be transported via the xylem and preferentially accumulate in leaves due to high transpiration rates and limited phloem remobilization, resulting in higher leaf concentration.^{53,64,73} In addition, it was reported that uptake of PFASs from atmospheric wet/dry depositions can further contribute to PFAS accumulation in leaves,^{60,74,75} warranting future investigations. Additionally, brown rice showed a higher BAF for both PFASs (3.5-fold) and HMs (1.6-fold) compared to white rice. This difference might be attributed to the protective role of the bran layer in white rice, which acts as a barrier to the uptake of PFASs and HMs during grain development.⁵⁵

For PFASs, perfluoroalkyl carboxylic acids (PFCAs) and their alternatives exhibited a trend of decreasing BAF with increasing carbon chain length (Figure 4c), except for PFOA, which showed an exceptionally high BAF in straw (263.32) and leaves (188.63). Notably, HFPO-DA

392 demonstrated significant BAF in roots (30.45) and brown rice (13.16), indicating its efficient
393 translocation from soil to roots and subsequent migration to edible grain tissues.⁷⁶ Interestingly,
394 PFDoA exhibited a higher BAF in brown rice (3.51) compared to other tissues, suggesting that
395 this long-chain compound preferentially accumulates in grains, warranting attention due to
396 potential health risks. In contrast, perfluoroalkyl sulfonic acids (PFSA) and their alternatives
397 showed no clear relationship between BAF and carbon chain length. However, their BAF in
398 roots was generally higher than those of PFCAs, likely due to their hydrophobic nature and
399 larger molecular weights,^{67,77} which facilitate retention in root tissues. Interestingly, PFPeS
400 exhibited an exceptionally high BAF in roots (306.86), followed by perfluorododecanesulfonic
401 acid (PFDoS), perfluorodecanesulfonic acid (PFDS), and perfluorononanesulfonic acid (PFNS).
402 On the other hand, PFAS precursors, such as FOSA, displayed low BAF values, indicating
403 limited accumulation in rice tissues. In contrast, among fluorotelomer sulfonates (FTS), 4:2 FTS
404 exhibited the highest average BAF values, followed by 6:2 FTS and 8:2 FTS, consistent with
405 the findings of previous greenhouse studies conducted by Zhou et al.,⁷⁸ suggesting their high-
406 water solubility promotes efficient accumulation in rice plants.

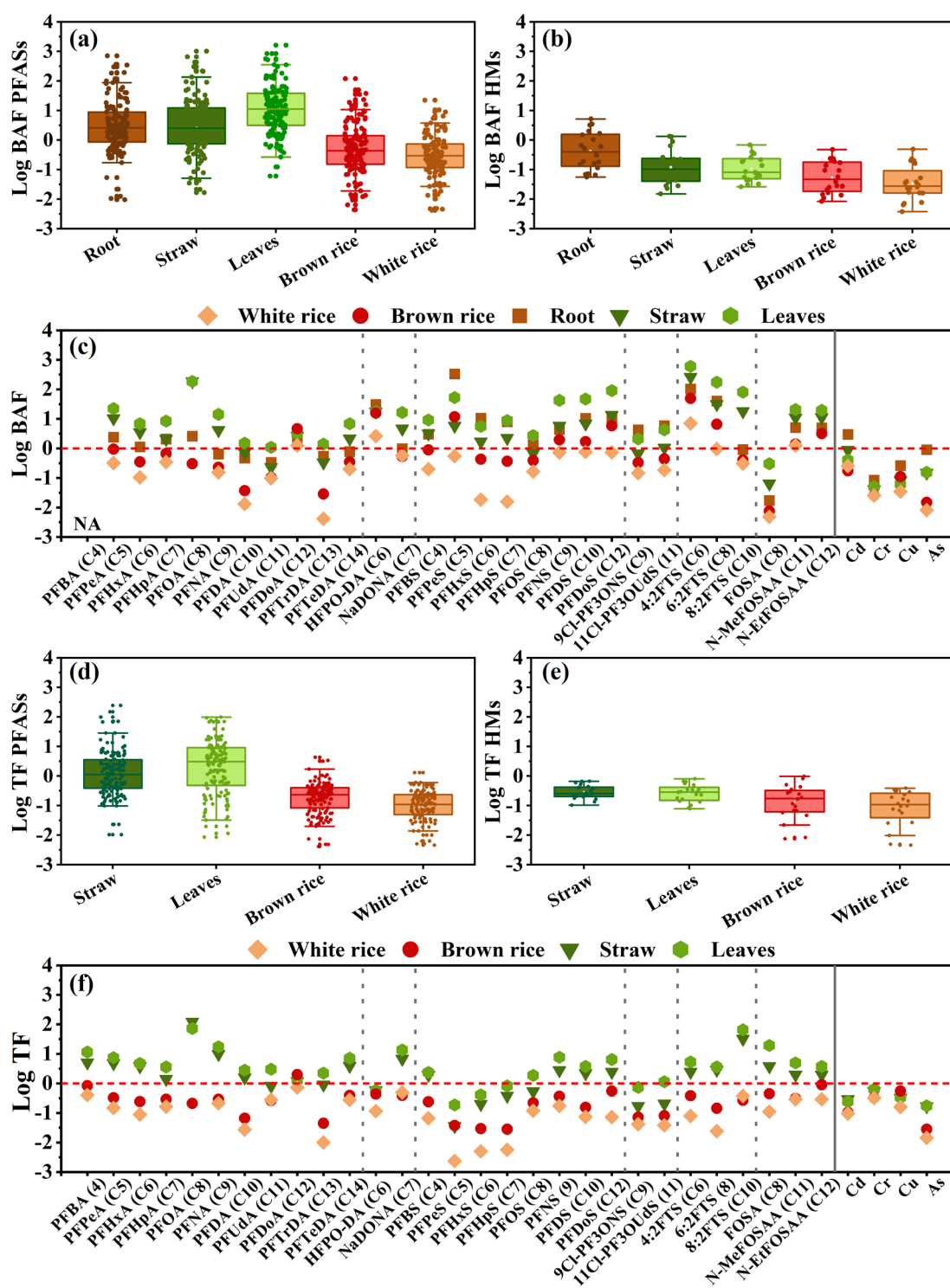


Figure 4. Box plots comparing bioaccumulation factor (Log_{10} BAF) of (a) PFASs and (b) HMs across different rice organs, (c) Log_{10} BAF for individual PFAS compound and HMs, Box plots comparing translocation factor (Log_{10} TF) of (d) PFASs and (e) HMs across aboveground tissues, and (f) Log_{10} TF of individual PFAS compound and HMs from root to aboveground tissues.

408

409 Among the four HMs, the highest BAF was observed in roots, consistent with their primary
410 uptake mechanism from soil (Figure 4b). Cd exhibited significantly higher BAFs across various
411 plant tissues compared to the other three HMs (Figure 4c), indicating its greater tendency to be
412 absorbed and accumulated by rice plants. Furthermore, PFASs may facilitate the uptake of Cd
413 by plant roots, enhancing its translocation to aerial tissues. This finding is supported by
414 mechanistic evidence from Zhao et al.,⁷⁹ who demonstrated that the copresence of PFASs and
415 Cd increased the bioavailability of Cd²⁺ in soil, resulting in elevated Cd accumulation in both
416 roots and shoots of wheat and rapeseed. Moreover, Cr exhibited the lowest BAF, with a
417 maximum value of 0.078, indicating minimal uptake by rice plants. This observation aligns with
418 the findings of Tang et al.,⁸⁰ who reported that a larger proportion of Cr persisted in soil under
419 F-53B cocontamination, suggesting that PFAS may reduce the uptake of Cr by water spinach.
420 The results also indicated a significantly higher BAF for As in the root (0.68) compared to brown
421 rice (0.014) and white rice (0.007), suggesting that the translocation of As within the rice plant
422 is highly restricted, particularly into the rice grains. This limited mobility highlighted the
423 challenge of transporting As from the root to the edible parts of the rice plant.

424 The TF of PFASs and HMs followed a similar trend (Figure 4d, 4e, and Table S14), with straw
425 and leaves showing higher TF. Notably, the TF of PFASs in straw and leaves were significantly
426 higher than those of HMs, while their TF values in white rice and brown rice were in similar
427 ranges. The log TF values for individual PFAS compounds (Figure 4f) indicated that the TFs of
428 perfluoroalkyl carboxylic acids (PFCAs) across rice organs are significantly higher than those

of perfluoroalkyl sulfonic acids (PFASs), which can be attributed to the higher water solubility and smaller molecular weight of carboxyl functional groups,^{81,82} facilitating their upward translocation via the transpiration stream. The TF of PFCAs also exhibited a decreasing trend with increasing carbon chain length, likely influenced by the translocation process. During this process, PFASs must penetrate through the epidermis, cortex, endodermis, and pericycle to reach the xylem, driven by the water potential gradient generated by transpiration. At the endodermis, PFASs must pass through at least one cell membrane, with their solubility in water and lipids (logD_{mw}) determining their ability to cross membranes through passive diffusion, active transport, or facilitated transport.⁸³ Additionally, the Casparian strip acts as an early barrier, promoting selective transport and contributing to the decrease in TF with increasing logK_{ow}, further limiting the translocation of long-chain PFASs.^{60,83} Among these, PFOA exhibited the highest TF values in straw (120.23) and leaves (72.44), likely due to its foliar uptake and subsequent downward translocation through the phloem.⁸⁴

Interestingly, in brown rice, only the TF value of PFDoA exceeded 1 (1.99), indicating that its enrichment capacity in brown rice surpassed that of root uptake (Figure 4f). This suggested that PFDoA has strong translocation potential, consistent with the observation by Che et al.,³ although the specific contribution remains unclear and requires further investigation. Compared to PFASs, HMs exhibited limited translocation within rice plants, with most concentrations in aboveground tissues remaining relatively low (Figure 4e). For instance, Cd showed moderate TF values (Figure 4f), indicating potential accumulation risks in edible plant parts (e.g., grains) for transporting Cd from the roots to the grains during grain maturation.⁴⁸ However, significant

knowledge gaps remained with regard to the precise mechanisms governing the couptake of PFASs and HMs in rice, which warrants future greenhouse studies to isolate synergistic or antagonistic effects. For instance, their interactions may modify the photosystem and metallothionein synthesis, impacting accumulation, translocation, and toxicity.^{79,80}

3.3. Relationship of Specific PFASs and HMs and Their Sources in Rice Grains

Rice primarily absorbs PFASs through two pathways, root uptake and foliar uptake.^{55,84,85} Root uptake of PFASs in rice involves absorption of both dissolved (soil moisture) and sorbed (soil particle-bound) fractions by roots, followed by translocation to aboveground tissues through the transpiration stream, which constitutes the primary contamination routes in paddy systems.^{60,63,86} In addition, it was reported that foliar uptake allows PFASs absorbed by leaves to be redistributed to grains through the phloem, further contributing to their accumulation in rice grains. Compound-specific correlations between brown rice and root/leaf concentrations were shown in Figure 5. According to the significant relationship of HFPO-DA in brown rice with both leaves ($R^2=0.41$, $r=0.64$, and $p < 0.01$) and roots ($R^2=0.35$, $r=0.59$, and $p < 0.001$), it was evident that HFPO-DA in brown rice originated from dual contributions of root uptake and leaf-derived channels (Figure 5c1, c2).^{77,87} Surprisingly, PFDoA, a long-chain perfluoroalkyl carboxylic acid (C12), was detected at the second-highest concentration in white rice (0.58 ng/g) and brown rice (1.64 ng/g), which were significantly higher than those (0.012 ng/g) reported in a previous study conducted by Kim et al.⁵⁵ The detection of PFDoA in various rice tissues suggested that its high concentration in rice grains might result from a combined effect of root uptake and foliar absorption, while the significant correlation between brown rice and roots

471 ($R^2=0.17$, $r=0.40$, and $p < 0.05$) suggested a contribution from root uptake, although the low R^2
472 value implied that grain accumulation was also subject to other complex physiological
473 constraints (Figure 5b1-b2). However, limited studies have been conducted on PFDoA,
474 highlighting the need for further investigation into its behavior, sources, and translocation
475 mechanisms in rice plants.^{75,77}

476 For other legacy PFASs, PFOS, the third most abundant compound detected in brown rice, was
477 found at higher concentrations in roots (4.87 ng/g), suggesting that its presence in brown rice
478 may primarily be influenced by root uptake. Although the correlation was statistically
479 significant ($R^2=0.10$, $r=0.31$, and $p < 0.001$) (Figure 5d1, d2), the low coefficient of R^2 indicated
480 that root-to-grain transfer only partially explained the grain loading, with other factors, such as
481 specific protein binding or restricted phloem mobility, likely exerting significant influence.
482 Interestingly, PFOA was nearly undetectable in brown and white rice despite being dominant in
483 rice shoots and leaves (12.98-70.26 ng/g), which led to weak and non-significant correlations
484 with root or leaf levels ($p > 0.05$) (Figure 5a1, a2), suggesting limited translocation to grains,
485 likely due to the protective role of the rice husk.⁸⁸ However, the precise mechanisms governing
486 this compartmentalization remained unclear and require systematic investigation.⁸⁹ To mitigate
487 the potential risks to food safety and environmental sustainability, future research should focus
488 on the mechanisms driving PFASs accumulation and explore practical strategies for their
489 reduction in agricultural systems.^{16,20} Significant positive correlations between specific PFASs
490 (HFPO-DA, PFDoA, and PFOS) in brown rice and roots underscore food chain transfer risks,
491 consistent with concerns about dietary PFAS exposure.³⁶

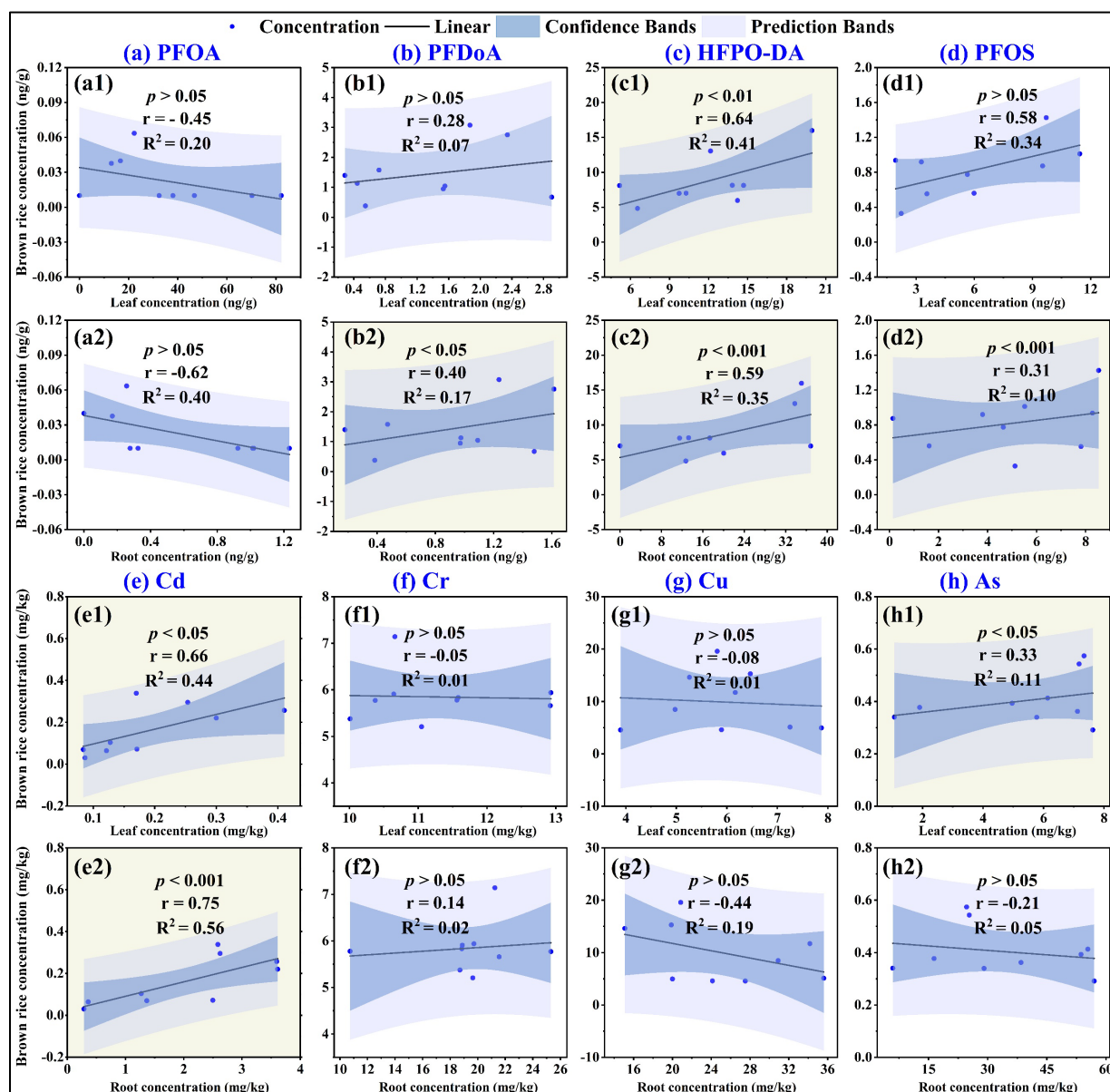


Figure 5. Regression analysis of specific PFAS concentration (ng/g) and HMs (mg/kg) in rice tissues.

(a) PFOA, (b) PFDoA, (c) HFPO-DA, (d) PFOS, (e) Cd, (f) Cr, (g) Cu, and (h) As. Plots (a1-h1) show linear relationships between concentrations in brown rice and leaves, while (a2-h2) display relationships between brown rice and roots. Solid lines represent linear regression fits, with Pearson correlation coefficients (r) and corresponding p -values indicate the strength and statistical significance of the associations. The light olive-yellow shaded plots indicate the statistical significance, with the specific p -value shown in each panel.

While among the different HMs, Cd exhibited strong positive correlations between brown rice and both roots ($R^2=0.56$, $r=0.75$, and $p < 0.001$) and leaves ($R^2=0.44$, $r=0.66$, and $p < 0.05$), indicating that Cd in brown rice originated from a combined contribution of root uptake and foliar absorption (Figure 5e1-e2). Previous studies have reported that foliar absorption can be a significant source of Cd accumulation in rice, contributing 30%-70% to grain Cd levels,^{90,91} which has been attributed to the direct uptake of Cd from atmospheric dry/wet deposition by leaves. Cr exhibited the highest concentration in white rice, whereas Cu accumulated to higher levels in brown rice. Alongside Cd, both metals showed relatively high levels in roots, yet no significant correlations were observed among them. In addition, no significant correlation was observed between total Cr and Cu concentrations in brown rice and those in either roots or leaves (Figure 5f-5g). This pattern suggested that strong regulatory processes in vegetative tissues, together with internal redistribution mechanisms, may mask the apparent contributions of roots or leaves to grain accumulation.⁹² Thus, an absence of correlation did not necessarily indicate a lack of translocation, but rather reflected the complexity of metal transport and partitioning within rice plants.

As was predominantly concentrated in roots, with relatively low levels in rice grains. The significant correlation observed between brown rice and leaves ($R^2=0.11$, $r=0.33$, and $p < 0.05$) (Figure 5h1), and the non-significant correlation with roots (Figure 5h2), suggested that As translocation within rice plants is restricted, the significant but weak correlation between brown rice and leaves suggested that foliar-derived As potentially contributes to grain loading. It has been reported that the limited translocation can be further supported by the critical role of

reducing As(V) to As(III), which exhibits higher affinity for thiol groups and enhanced mobility within plant cells.⁹³ While this conversion facilitates As(III) translocation through the xylem to aboveground tissues and subsequent accumulation in leaves, it simultaneously promotes vacuolar sequestration in roots through phytochelatin complexation, thereby restricting overall As mobility.⁹³

3.4. Health Risk Assessment for Selected PFAS Compounds and HMs

The uptake and enrichment of PFASs and HMs in brown and white rice across different locations were presented in Figure 6, respectively. The calculated EDIs were compared with the existing RfD thresholds to assess the exceedance levels of specific PFASs and HMs in brown and white rice, as established by the USEPA,^{37,94} Minnesota Department of Health (MDH),^{95–98} and Texas Commission on Environmental Quality (TCEQ),⁹⁹ for various compounds, which were outlined in Tables S15 and S16. RfDs chosen were the most conservative based on previous studies,^{14,15} to reflect the significant concentration detected and highlight potential risks. For HFPO-DA, the most recent RfD available was adopted, which has been recently updated by the USEPA.³⁷

The concentrations in both brown and white rice from different locations were logarithmically transformed and compared with the regulatory standards. Noticeable differences in concentration between brown and white rice across all age groups were observed, with toddlers appearing to be the most vulnerable, as their estimated dietary exposure thresholds (red dashed lines) were consistently lower across all compounds. Notably, HFPO-DA concentrations in brown rice exceeded the exposure thresholds for toddlers and children, raising concerns about

potential health risks for younger populations. In addition, PFDoA, PFHxA, and PFHpA showed relatively high levels, with extensive exceedances in brown rice. These exceedances in brown rice may raise concerns for the continuous campaign promoting the inclusion of brown rice, instead of white rice, for a healthier diet.¹⁰⁰

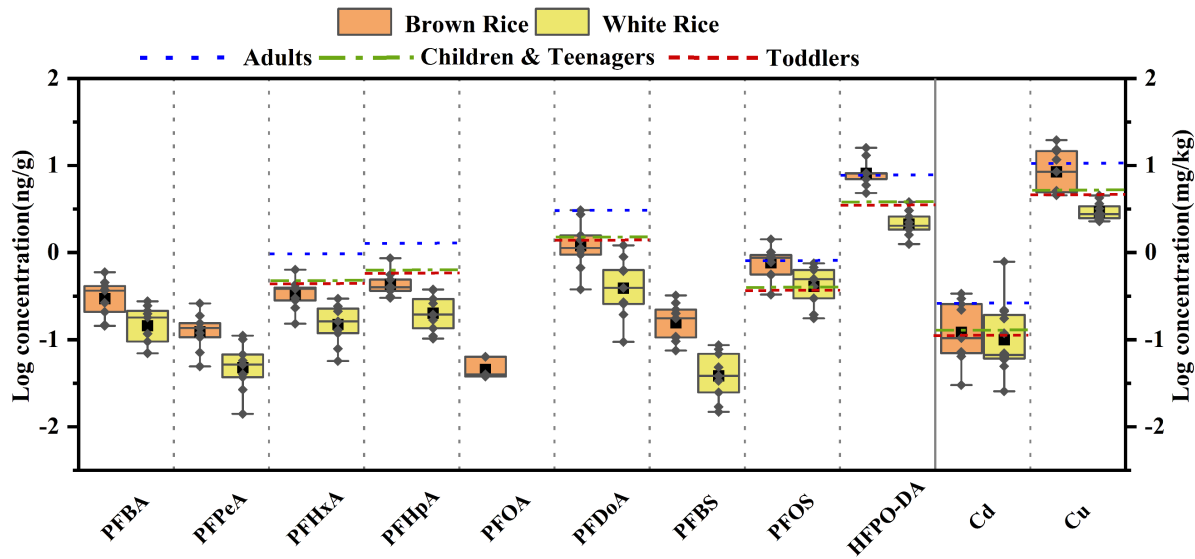


Figure 6. Comparison of specific PFAS and HM concentrations across different age groups in brown and white rice. Box plots showing the log-transformed concentrations of PFAS and HMs in brown rice and white rice. Boxes indicate interquartile ranges of different locations, horizontal lines inside boxes represent medians, and whiskers show data variability outside the upper and lower quartiles.

In comparison, in white rice, PFOS was the only compound exceeding the exposure threshold, suggesting a preferred accumulation of PFOS, raising a major concern about PFOS exposure due to the broad consumption of white rice. Cd, the most well-recognized HM in rice, exhibited levels below the median threshold levels, albeit concentrations in brown rice were higher than those in white rice. Cu also showed similar findings, exceeding the risks for toddlers, children and teenagers in brown rice. These results indicated that the daily consumption of rice was

546 linked to concerning amounts of specific PFAS compounds, including HFPO-DA, PFDoA,
547 PFOS, PFHxA, and PFHpA, on top of potential hazards to HMs, such as Cu and Cd. It should
548 be noted that As and Cr were excluded from the EDI calculations, as only their total
549 concentrations were measured, but their toxicity is highly species-dependent (e.g., As(III) vs.
550 As(V), Cr(VI) vs. Cr(III)).

551 Additionally, the results showed that compared to age groups of children & teenagers and adults,
552 toddlers have the highest EDI for the chemicals evaluated (Fig. 6), suggesting that toddlers face
553 a higher potential health risk, underscoring the importance of conducting targeted risk
554 assessments for this vulnerable age group. The uncertainty quantification of the EDIs of HFPO-
555 DA, PFDoA, PFHxA, and PFOS for toddlers were presented in Figure S5 and Table S17, as
556 well as HMs including Cu and Cd in Figure S6 and Table S18, providing a comprehensive
557 assessment of potential risks and total HI. In brown rice, the 50th percentile EDIs for HFPO-DA,
558 PFDoA, PFOS, and PFHxA were 77.63, 14.14, 7.44, and 4.16 ng/kg bw/day. For HMs, the 50th
559 percentile EDIs for Cd and Cu were 1.13 and 92.45 µg/kg bw/day, respectively. All values
560 exceeded the HQ threshold (HQ>1). In contrast, for white rice, the 50th percentile EDIs for
561 HFPO-DA (20.26 ng/kg bw/day), PFDoA (5.00 ng/kg bw/day), and Cu (26.88 µg/kg bw/day)
562 remained below the threshold (HQ<1). However, it is noteworthy that the 50th percentile of
563 PFOS (4.08 ng/kg bw/day) and Cd (1.07 µg/kg bw/day) exceeded the HQ threshold value,
564 highlighting their potential health concerns.^{15,20}

565 The total HI for HFPO-DA, PFOS, PFDoA, PFHxA, Cd, and Cu at the 50th percentile of
566 exposure level for toddlers, reached 10.7 in brown rice, twice of that measured in white rice.

567 The cumulative HI surpassed the safety threshold of 1, indicative of potential additive toxic
568 effects. The presence of PFASs and HMs in rice-based foods suggested that toddlers may
569 experience chronic exposure to these hazardous substances. Consequently, long-term
570 consumption of such rice-based foods by toddlers may have elevated health risks, particularly
571 in regions where rice is a staple food. These findings provided critical insights for cumulative
572 risk assessments and stressed the need for stringent monitoring and regulation of contaminant
573 levels in crops to ensure sustainable agricultural practices and safeguarded human health
574 associated with PFAS and HM exposure across different age groups and exposure scenarios,
575 especially through a rice-dominated diet.^{1,14}

576 This study provided a comprehensive assessment of the co-occurrence, bioaccumulation, and
577 uptake routes of PFASs and HMs in rice tissues, alongside an evaluation of the potential dietary
578 health risks. Six PFASs, including HFPO-DA, PFDoA, PFHxA, PFOA, PFOS, and PFNA,
579 dominated rice accumulation, with total PFAS concentrations ranging from 4.75 to 135.93 ng/g
580 across tissues. Brown rice accumulated significantly higher levels of PFASs (14.57 ng/g) than
581 white rice (4.75 ng/g), with long-chain PFDoA showing preferential grain accumulation. For
582 HMs, roots were the primary accumulation sites, though brown rice also contained elevated
583 levels of Cd, Cr, and Cu. Contaminant-specific bioaccumulation factors revealed that PFASs
584 such as 4:2 FTS, 6:2 FTS, and PFOA preferentially accumulated in leaves and straw, whereas
585 Cd and As primarily concentrated in roots. Despite differences in initial uptake pathways, both
586 PFASs and HMs exhibited similar systemic translocation patterns, favoring aerial tissues.
587 Analysis of concentration relationships among rice organs indicated that a single absorption

588 route dominated most contaminants; however, the dual uptake pathways, via root absorption
589 and foliar deposition, drive accumulation of HFPO-DA and Cd in edible grains, explaining their
590 elevated accumulation in brown rice. Notably, PFOS, HFPO-DA, Cd, and Cu in brown rice
591 exceeded existing edible safety standards, increasing the potential dietary exposure risks
592 associated with rice consumption consumed by both local rural and urban residents, either
593 through self-consumption or commercial distribution, whoever consumes this rice, and
594 highlighting the need for stringent cumulative risk assessments that consider both PFASs and
595 HMs.

596 While our study provided valuable insights into contaminants coaccumulation and translocation
597 in rice and highlighted the environmental and food safety risks posed by the cocontamination of
598 PFASs and HMs in rice systems, several limitations remain. In particular, the contribution of
599 atmospheric deposition to PFAS accumulation under field conditions remains unquantified, with
600 its potential atmospheric contributions should be considered. Moreover, health risk assessments
601 for Cr and As were constrained by the lack of speciation data for their more toxic forms. Future
602 research should aim to elucidate the mechanisms governing coupled uptake and systemic
603 distribution of PFASs and HMs, assess deposition processes, and determine metal speciation in
604 rice. Such insights are critical for developing strategies (such as implementing crop management
605 (e.g., intercropping) and cultivating low-accumulator varieties) to mitigate contamination and
606 reduce dietary health risks associated with rice consumption.

AUTHORSHIP CONTRIBUTION STATEMENT

Samir Attia: writing-review and editing, writing-original draft, methodology, investigation, formal analysis, data curation, visualizations, and conceptualization. **Zhiyuan Hu:** writing-review and editing, data curation, visualization, and conceptualization. **Fangjie Qi:** investigation and conceptualization. **Dongling Li:** investigation. **Frederic Coulon:** writing-review and editing. **Xin Song:** writing-review and editing, data curation, supervision, methodology, funding acquisition, resources, and conceptualization.

DECLARATION OF COMPETING INTERESTS

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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SUPPORTING INFORMATION

List Item The Supporting Information includes detailed descriptions of sampling collection and pre-treatment, PFAS extraction, and heavy metal digestion procedures (Texts S1–S3). Supplementary figures (Figures S1–S6) present sampling location distributions, concentrations and profiles of PFASs and heavy metals in different rice tissues, relative abundances, and Monte

Carlo-based human health risk assessments for toddlers. Supplementary tables (Tables S1–S18) provide sampling locations, irrigation water and soil properties, analytical quality assurance and quality control parameters, concentrations of PFASs and heavy metals in rice and environmental media, bioaccumulation and translocation factors, estimated daily intakes across age groups, and probabilistic dietary risk assessments (PDF)

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Attia, Samir; Hu, Zhiyuan; Qi, Fangjie; Li, Dongling; Coulon, Frederic ...

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