Per- and polyfluoroalkyl substances (PFAS) in snow, river and drinking waters from a typical rural area of Shandong, China

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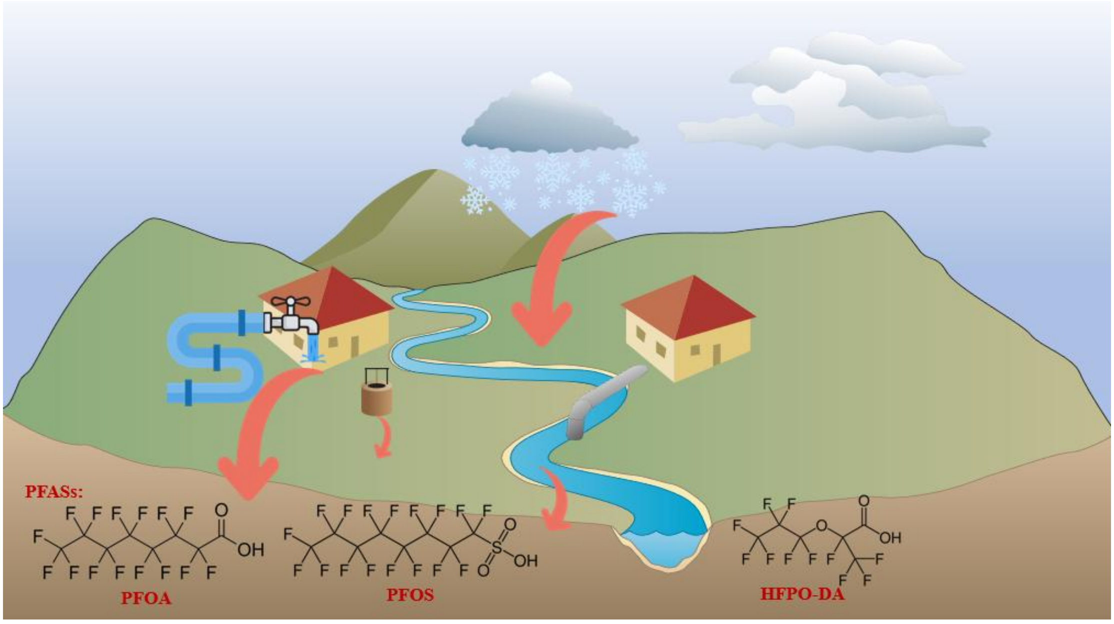
Abstract

Per- and polyfluoroalkyl substances (PFAS) are of global environmental concern because of their persistence and potential threats to ecosystems and human health. In China, most studies have centred on industrial and urban areas, while PFAS contamination in rural regions is not well-understood. To fill this gap, this study explored the occurrence, distribution, and partitioning characteristics of PFAS in snow, river, tap water, and well water in a rural area in Shandong Province, northern China. Among the 72 targeted PFAS, 13 were detected, with total concentrations varying from 4.21 to 453 ng/L. Short-chain PFAS were predominant in the samples, which is in line with their increasing global prevalence. Atmospheric deposition, domestic discharges, and seasonal snowmelt were identified as potential key contributors to PFAS contamination. The sole detection of perfluoropentanoic acid (PFPeA) in tap water indicates the influence of water treatment processes. In well water, the predominance of perfluorobutane sulfonic acid (PFBS), 3:3 fluorotelomer carboxylic acid (3:3FTCA), and perfluorobutanoic acid (PFBA) reflects the mobility and persistence of short-chain PFAS in subsurface environments. These findings show the complexity of PFAS transport and transformation in rural settings and emphasise the need for further research on their sources, environmental behaviours, and potential health risks. Understanding PFAS contamination in rural China is crucial for formulating pollution mitigation strategies and reducing human exposure risks.

Keywords: Fluorinated chemicals; water pollution; snow; well; health

Graphical Abstract



Highlights:

- First comprehensive PFAS study in rural Shandong, China, on snow, river, tap and well water.
- Out of 72 targeted PFAS, 13 were detected (4.21–453 ng/L), with short-chain PFAS dominating.
- Atmospheric deposition, domestic discharges, and snowmelt were important pollution sources.
- Distinct PFAS profiles in water sources: PFPeA in tap water implies treatment influence; short-chain PFAS in well water show subsurface mobility.

1. Introduction

Per- and poly-fluoroalkyl substances (PFAS) are a large number of synthetic chemicals known for their properties of resisting oil and water. There are over 4700 uniquely identified fluorinated organic compounds and this inventory is ever-expanding (OECD 2018; Yi et al. 2023). PFAS are widely used in consumer and industrial items, such as textiles, firefighting and food packaging, covering various fields, including medical, mechanical, electrical, agriculture, automotive, industrial manufacturing and chemical (Buck et al. 2011; Evich et al. 2022; Niu et al. 2024), due to their high chemical stability and ability to decrease aqueous surface tension. PFAS are persistent chemicals that take a long time to break down (US EPA 2016a). In natural environments, precursors are ultimately transferred into stable PFAS through processes like hydrolysis, oxidation, reduction, decarboxylation and hydroxylation (Bruton & Sedlak 2017; Cui et al. 2020; Washington & Jenkins 2015). Multiple studies have shown that PFAS compounds can accumulate in the human body and have negative impacts on various aspects of human health, including the risk of cancer (American Cancer Society 2024), metabolic problems (Dunder et al. 2023), cardiovascular, degenerative diseases (Zhou et al. 2023), immunotoxicity (Bonato et al. 2019; Sunderland et al. 2019), reproductive toxicity (Rickard et al. 2022) and neurotoxicity (Yi et al. 2023). Increasing awareness of their risks led to the Stockholm Convention on Persistent Organic Pollutants (POPs) in 2009 and 2019, which implemented global restrictions on perfluorooctanesulfonic acid (PFOS) and perfluorooctanoic acid (PFOA), respectively (UNEP 2009, 2019).

PFAS are ubiquitous across diverse environmental matrices, such as waters, sediments, soils, and landfill effluents (Banzhaf et al. 2017; Dalahmeh et al. 2018; Houtz et al. 2013). In the Cape Fear River of North Carolina, both legacy and emerging PFAS have been detected in the sources of drinking water (Bai & Son 2021). Snow is an important source of PFAS entering freshwater bodies, both pristine and urban areas (Filipovic et al. 2015; Meyer et al. 2011). Studies of PFAS in snow mainly focus on high-latitude and high-altitude areas such as the Arctic, Antarctic and Alps, indicating that the annual atmospheric deposition is a major source of PFAS and evidence of their long-range transport ability via atmospheric and oceanic circulation (MacInnis et al. 2017; Pickard et al. 2018; Wang et al. 2021; Wang et al. 2019; Wong et al. 2017). Many research efforts have been carried out to explore the presence and behavior of PFAS in surface waters in China, such as Taihu lakes (Yao et al. 2022), Yangtze River and Pearl River (So et al. 2007, Chen et al. 2021a), Dagu Drainage Canal and Haihe River (Li et al. 2011), Bohai Sea, Yellow Sea and East China Sea (Gao et al. 2014; Meng et al. 2021; Zhao et al. 2017). The contamination intensity and dispersion patterns of PFAS in terrestrial-aquatic systems correlate significantly with regional industrial activity and municipal development density. (Wang et al. 2011).

Existing studies have primarily investigated PFAS contamination in principal water bodies within urban-industrial zones (Liu et al. 2022; Wang et al. 2022; Ingold et al. 2023). While limited research has identified PFAS presence in remote areas (Ren et al. 2023; Wang et al. 2013), studies on the distribution of PFAS in a large number of typical rural areas without intensive commercial and industrial activities are still scarce, especially comprehensive investigations of snow, river and drinking water. Rural areas may be the “background areas” for PFAS pollution in areas with intensive human activities. Contamination in rural areas typically comes from domestic and agricultural sources and atmospheric deposition. Moreover, no wastewater treatment plants (WWTPs) are usually set up in very rural areas, so all household wastewater is directly discharged into the surrounding water bodies or applied as fertilizer on cropland, which may lead to PFAS pollution.

Therefore, this study selects a typical rural area in central Shandong to collect snow, surface water and well/tap drinking water to initially understand the occurrence and profile of PFAS. It also helps to identify the potential sources of PFAS transferring to the snow and aquatic environments. The findings of this study can clarify PFAS pollution characteristics in typical rural areas and provide a better understanding of the potential risk of PFAS exposure to humans.

2. Methods and Materials

2.1 Chemical and reagents

This investigation systematically analysed 72 PFAS encompassing a broad range of physicochemical properties. The study covered 12 major subclasses, such as perfluorocarboxylic acids, perfluorosulfonic acids, and fluorotelomer derivatives, with detailed chemical formulas, structures, and physicochemical properties provided in Table S1 and Text S1. All target analytes, along with 19 isotopically labeled internal standards, were obtained from certified suppliers listed in Table S2. Reagents, including HPLC-grade solvents, solid-phase extraction cartridges (Oasis WAX, Chromabond HR-X), and purification materials, were used as specified in the Supplementary Information (Text S2).

2.2 Study region and sampling

This investigation selected a representative rural area in Shandong Province, northern China (Fig. 1). The sampling site is located mainly in Laozhuangzi (LZZ) village with a population of ~ 2500, spreading a living area of ~ 9 km^2, with most human activities are agriculture for growing wheat, maize, peanut, sweet potato, etc with some small poultry farms starting recently. LZZ village is located southeast of Ningying County, Tai'an (city), just north of the hometown of Confucius, Qufu. LZZ village is north of Fengxian Mountain, where the LZZ stream originates. LZZ stream flows north to the Beibi River, which merges into the Chaiwen River (an upstream river to the Dawen River, which flows into Dongping Lake before merging into the Yellow River). No WWTP is available in this village, and all the wastewater (except the toilet waste to the septic tank) is discharged to the ground and eventually drained into the LZZ stream.

Different types of water samples were collected, including snow, stream, and drinking water (tap and well waters). Fresh snow samples were collected via a pre-cleaned stainless-steel basin during snowfall on the 2nd (SNOW-1) and 5th February 2024 (SNOW-2), which were the first and second snowfalls, respectively, in the wintertime in this area. Some of the SNOW-2 samples were boiled (BSNOW) to check if boiling can affect the composition of PFAS (indicating some precursors or transformation). Water samples were collected at the source (RIVER-S, near the Fengxian mountain and far from the village), upstream (RIVER-U, just before the village), middle (RIVER-M) and downstream (RIVER-D, about 1 km away from the village) of the LZZ stream (Fig. 1). The well water samples (WELL-1, WELL-2, WELL-3, used as drinking water) were collected from household wells near the Fengxian Mountain. The tap water samples were collected in a household in LZZ village on two different days (TAP-1: on Feb. 2, 2024; TAP-2: on Feb. 7, 2024) at noon when the tap water was supplied for about 2 h. A tap water (TAP-3) from the town centre Ningyang (suburban, about 40 km west away from LZZ), supplied by a drinking water supplier, and one tap water (TAP-4) from a nearby village (8 km away from LZZ, with different source water), were also collected to compare. All samples were duplicates in 500 mL each in a PET bottle. The latitude and longitude of the river sampling sites are shown in Table S3.

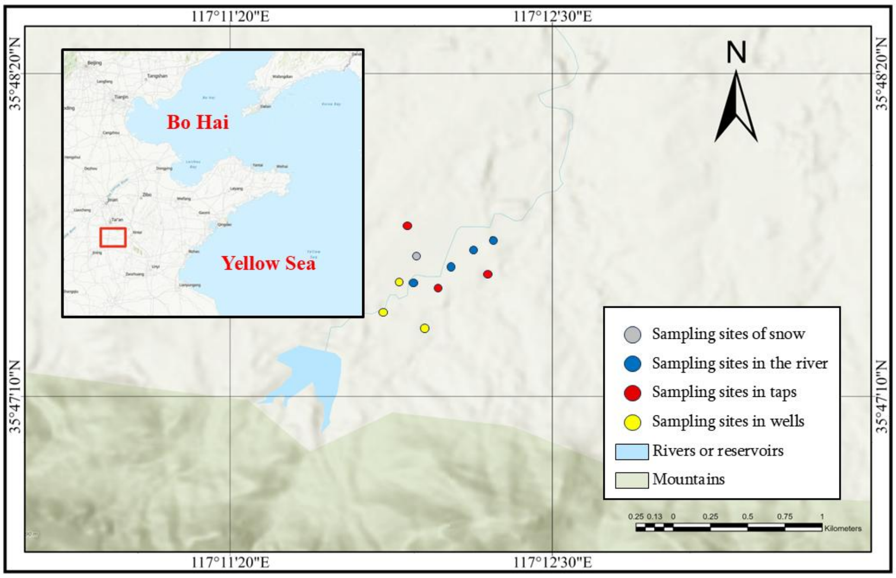


Fig. 1. Diagram of the sampling sites in Laozhuangzi (LZZ) village, Tai'an, Shandong

2.3 Sample pre-treatment and chemical analysis

The extraction of PFAS from water samples was carried out according to our previous methodology (Chen et al. 2021a, Yang et al. 2022). In brief, water samples were first passed through glass fibre filters and extracted by Waters Oasis WAX Cartridges. PFAS were analysed by using a Waters ultra-performance liquid chromatography system coupled with a Waters Xevo TQ-S tandem mass spectrometer (UPLC-MS/MS) through positive and negative electrospray ionisations (depending on the chemical) and operated in multiple reaction monitoring (MRM) modes. To achieve the highest sensitivity, selectivity, and chromatographic resolution for PFAS, the MS parameters were optimized, and the optimized parameters such as retention time, precursor ion, product ion, cone voltage, and collision voltage for each PFAS are listed in Table S4. Detailed information on sample pre-treatment and instrumental analysis are given in Text S3, S4 and S5 of SI.

2.4 Quality assurance/quality control (QA/QC)

QA/QC protocols were rigorously executed across sample collection, chromatographic extraction, and LC-MS/MS quantification phases. The newly bought bottled purified waters (in 500 mL PET bottles) were used as blank samples, while the empty PET bottle was used to collect and store all other samples before analysis. Laboratory controls including procedural blanks and matrix-spiked recoveries, were implemented to monitor potential contamination throughout the workflow. The concentrations of 72 target PFAS compounds were quantified via the internal standard method, using a six-point calibration curve (0.1, 0.5, 1, 2, 5, 20, 50 ng/mL) prepared in a mixture of methanol and water (1:1). The determination coefficients (R^2) of the calibration curves were all greater than 0.98 for 69 PFAS, with 0.5-0.6 for PFHxPA, PFOPA and PFDPA (Table S5). The limitation of detection (LOD) and limitation of quantification (LOQ) were calculated from the lowest point of the calibration curve via 3 and 8 times of the signal to noise ratio, respectively; the LODs and LOQs are in the range of 0.01–0.2, and 0.03–0.6, respectively. Systematic quality control involved inserting procedural blanks and spiked samples at 9-sample intervals to monitor carry-over effects, environmental interference, and method reliability. No PFAS were detected (> LODs) in the blanks. The recoveries were 50%–150%.

2.5 Human health risk assessment

This investigation assessed health implications of PFAS exposure through drinking water consumption across three age cohorts (toddlers, adolescents and adults). Exposure dose estimates (EDI) were calculated according to equation (1) and subsequently compared to established reference doses (R*f*D) (Zhou et al. 2021; US EPA 2016b, 2016c).

$$\mathrm{EDI} = \frac{C \times D_w}{BW} \qquad (1)$$

The concentration of individual PFAS in tap water samples (ng/L) is denoted as C, while D_w represents the daily tap water consumption rate, and BW refers to the average body weight. For toddlers, adolescents, and adults, the daily tap water consumption rates are estimated at 0.21 L, 0.62 L, and 1.35 L, respectively (Qi et al. 2016; US EPA 2011). Meanwhile, the average body weights for these three groups in China are 19 kg, 53 kg, and 63 kg, respectively (Zhou et al. 2021; Guo and Kannan 2011; Liao et al. 2012)..

2.6 Data analysis

PFAS concentrations were reported as mean ± standard deviation (SD). A Spearman rank correlation analysis was performed using RStudio (R version 4.4.1, RStudio Team, 2020) on Windows to assess PFAS correlations in the samples and generate a PFAS correlation plot. Except for this R-based correlation analysis, Excel 2021 (Microsoft Corporation, Redmond, WA, USA) was used for other data statistical analyses. The sampling point map was created with ArcGIS Pro 3.1.0 (ESRI, CA, USA), and the remaining figures were drawn using Origin 2020 (Origin Lab Corp, MA, USA). The threshold for statistical significance was established at $\alpha = 0.05$.

3. Results and discussion

3.1 PFAS occurrence in general

We detected 13 out of 72 targeted PFAS (PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, HFPO-DA, HFPO-TA, 3:3FTCA, PFBS, PFOS, 6:2Cl-PFESA, and 8:2Cl-PFESA) across water and snow samples (Fig. 2). The cumulative concentrations detected PFAS (ΣPFAS) varied between 4.21–453 ng/L, peaking in fresh snow (SNOW-1 and SNOW-2). Boiled snow (BSNOW) had lower ΣPFAS, likely due to volatile PFAS loss or transformation during boiling. PFAS composition varied by sample type, reflecting diverse sources and treatment effects. Short-chain PFAS ($C \leqslant 6$) dominated, with 3:3FTCA most abundant in snow, followed by PFBS, HFPO-DA and PFOA. PFPeA was only found in tap water, suggesting sources specific to treated water systems, such as chemical degradation during water treatment and leaching from water supply infrastructure (see Section 3.4 for discussion). Chlorinated PFESAs, although present at lower concentrations, were consistently detected, highlighting their persistence and potential environmental risks. Notably, Li et al. (2024) reported high levels of several PFAS (PFOA, PFOS, PFNA, PFBS, PFHpA, PFBA, PFHxA, PFPeA) in Shandong. All these substances were also detected in our rural Shandong samples, indicating high regional PFAS background values and validating their widespread presence. These findings provide a comprehensive overview of PFAS distribution across various environmental and water matrices, emphasizing the dominance of short-chain PFAS and briefly indicating that different treatment processes—such as boiling, water treatment, and distribution system influences—can alter PFAS composition through volatilization, chemical transformation, or leaching (Bamdad et al. 2022; Chen et al. 2021b; Feng et al. 2021). The detailed discussion of these treatment effects is presented in later sections. Table S6 presents a listing of PFAS data from snow, river, tap, and well water samples reported in other literatures, enabling a direct comparison with the findings of our study.

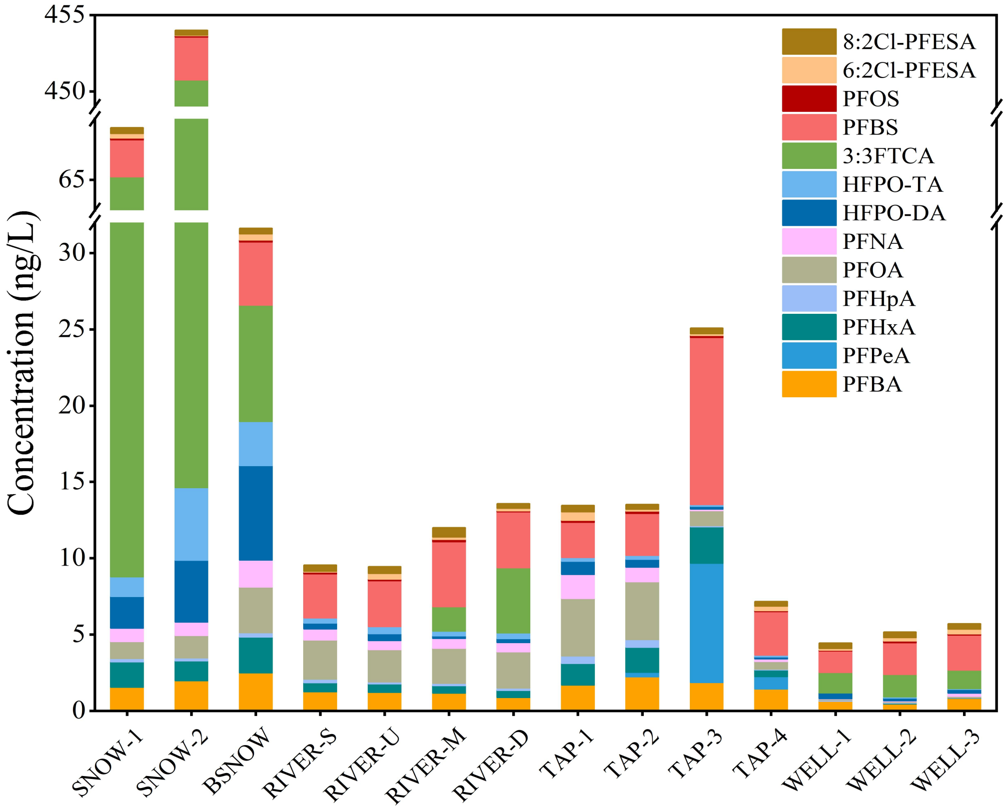


Fig. 2. Levels and pattern of PFAS in water and snow samples from a rural area, North China. BSNOW: boiled snow samples.

3.2 PFAS in snow

Overall, 12 of the 72 targeted PFAS were identified in the snow samples, with concentrations exceeding their respective limits of detection (LODs). This included 8 legacy compounds and 4 alternative PFAS (HFPO-DA, HFPO-TA, 6:2Cl-PFESA, and 8:2Cl-PFESA), as shown in Figure 3. The PFAS composition varied significantly among sample types, likely due to differences in atmospheric deposition and transformation mechanisms (Fig. 3b). Short-chain PFAS ($C \leq 6$) dominated in all samples, with 3:3FTCA being the most abundant, contributing about 90% to ΣPFAS in SNOW-1 and SNOW-2, followed by HFPO-DA and PFBS, which are widely used in industries as substitutes for legacy PFAS like PFOS. Their dominance can be ascribed to their higher volatility and greater potential for long-range atmospheric transport compared to long-chain ones (Persaud et al. 2024). The notably high abundance of 3:3FTCA in snow samples may result from multiple factors. Although structurally distinct from traditional PFCAs, 3:3FTCA could form via atmospheric degradation of volatile fluorotelomer precursors, similar to known PFCA pathways (Ellis et al. 2004). However, its unique structure means specific transformation mechanisms remain unclear. Besides atmospheric formation, local or regional sources such as industrial emissions or contaminated waters may contribute directly. Additionally, the physicochemical properties of snow, including cold temperatures and efficient scavenging capacity, could enhance the deposition and accumulation of 3:3FTCA relative to other PFAS. Snowfall events may act as an effective sink, concentrating certain PFAS species through wet deposition processes. This could explain the dominance of 3:3FTCA in snow samples compared to other matrices where dilution or degradation may occur more readily (MacInnis et al. 2019). Conversely, Cl-PFESAs (6:2Cl-PFESA and 8:2Cl-PFESA) and legacy PFAS such as PFOS and PFOA contributed less than 0.4% to Σ PFAS, indicating limited atmospheric transport or deposition at the study site. The relatively low levels of legacy PFAS compared to alternatives like HFPO-DA and HFPO-TA reflect an ongoing shift in industrial practices toward short-chain and alternative PFAS, consistent with regulatory restrictions on long-chain PFAS due to their persistence and bioaccumulation potential (Hoke et al. 2015; Hopkins et al. 2018). Nevertheless, the detection of Cl-PFESAs and legacy PFOS, even at low concentrations, underscores their environmental persistence and long-range transport potential (Wang et al. 2015).

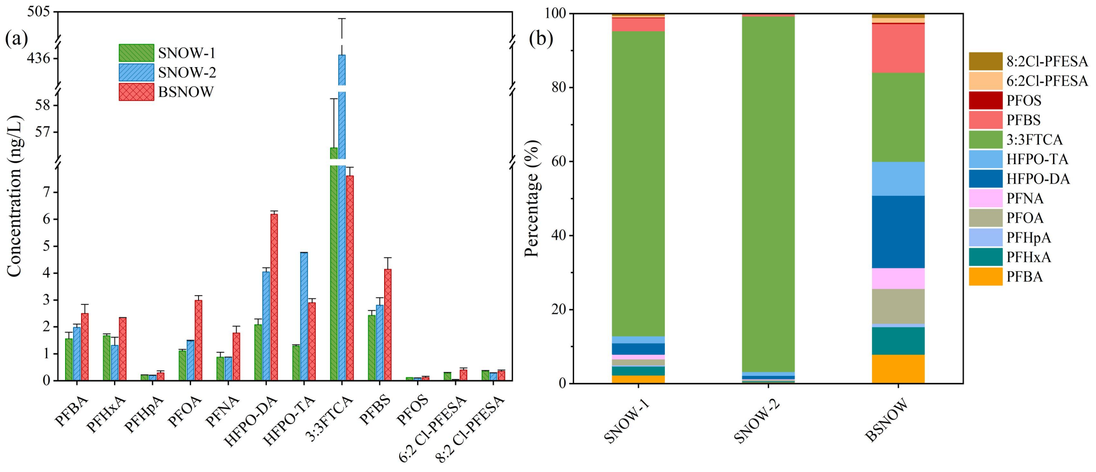


Fig. 3. Levels (a) and patterns (b) of PFAS in snow samples. BSNOW: boiled snow samples.

Boiled snow samples (BSNOW) exhibited a ΣPFAS concentration of approximately 30 ng/L, significantly lower than in fresh snow samples. This reduction indicates the potential volatilization or transformation of certain PFAS during the boiling process. Short-chain PFAS, such as 3:3FTCA and PFBA, likely volatilised due to their higher vapor pressures and lower boiling points (Bamdad et al. 2022), while thermally stable compounds like HFPO-DA and PFBS increased in relative proportions. Notably, HFPO-TA showed a marked decrease, suggesting possible thermal degradation or transformation into unmeasured byproducts. These findings highlight the need for further investigations into the thermal stability and transformation pathways of alternative PFAS.

The ΣPFAS concentrations in fresh snow samples were two to three orders of magnitude higher than those reported in rural regions such as Shenyang (0.63 ng/L) and Zhangjiakou (0.25 ng/L) (Wang et al. 2021). Compared to urban areas such as Hong Kong, Slingerland, and Toulouse, the concentrations in this study were similar to those in Kawaguchi (Kwok et al. 2010). The high ΣPFAS concentrations, particularly of 3:3FTCA, HFPO-DA, and PFBS, suggest significant atmospheric transport of PFAS to the study site. Given the prevailing northern winds in Shandong during winter, these PFAS are likely influenced by emissions from heavily industrialised regions such as Beijing, Tianjin, and Hebei Province, known for fluoropolymer manufacturing, chemical production, and textile industries (Cai et al. 2012; Young et al. 2007). It should be noted that Shandong also has relevant industrial activities that may contribute to local PFAS inputs. While the PFAS profiles (relative compositions) in SNOW-1 and SNOW-2 were generally consistent, indicating similar atmospheric sources and transport mechanisms, the total PFAS concentration in SNOW-2 (454 ng/L) was significantly higher than in SNOW-1 (68.4 ng/L). This disparity, including the markedly elevated 3:3FTCA concentration in SNOW-2, may reflect variability in industrial emissions or meteorological conditions, such as changes in wind direction or precipitation intensity, between the two snowfall events. The higher levels of HFPO-DA and PFBS in SNOW-2 further support the hypothesis of intensified emissions or enhanced deposition during the second event.

3.3 PFAS in river waters

A total of 12 PFAS, including 8 legacy PFAS and 4 alternatives (HFPO-DA, HFPO-TA, 6:2Cl-PFESA, and 8:2Cl-PFESA), were detected in surface water from LZZ Village, as shown in Figure 4. ΣPFAS ranged from 9.46 to 13.6 ng/L, with the highest concentration observed downstream (RIVER-D). Short-chain PFAS ($C \leq 6$) dominated across all sites and no PFAS with a chain length longer than 11 carbons were detected. The concentration of 3:3FTCA increased from the middle reaches (RIVER-M) to the downstream (RIVER-D). This increase can be attributed to the waste (water/solid) discharge from the village including the road snow. Its high volatility, mobility, and low hydrophobicity facilitate transport via snowmelt and surface runoff, underscoring the seasonal significance of snowmelt in delivering PFAS to aquatic systems (Padioleau et al. 2024). The PFAS composition remained relatively stable along the stream, with short-chain PFAS maintaining dominance. Slightly elevated ΣPFAS concentrations in the middle reaches likely stemmed from localized inputs of household wastewater and PFAS-containing products (Feng et al. 2021; Zushi and Masunaga 2009). Previous research has reported similar patterns of PFBS and PFBA dominance in rural and remote regions globally (Clara et al. 2009; Chen et al. 2016; Ren et al. 2023), which suggests the increasing environmental prevalence of short-chain PFAS as substitutes for long-chain compounds. Our finding of short-chain PFAS dominance aligns well with these studies. Li et al. (2024) offered an explanation for this trend, stating that the lower technical performance of short-chain PFAS may lead to a greater use of PFBA to achieve the performance comparable to long-chain PFAS like PFOA and PFOS.

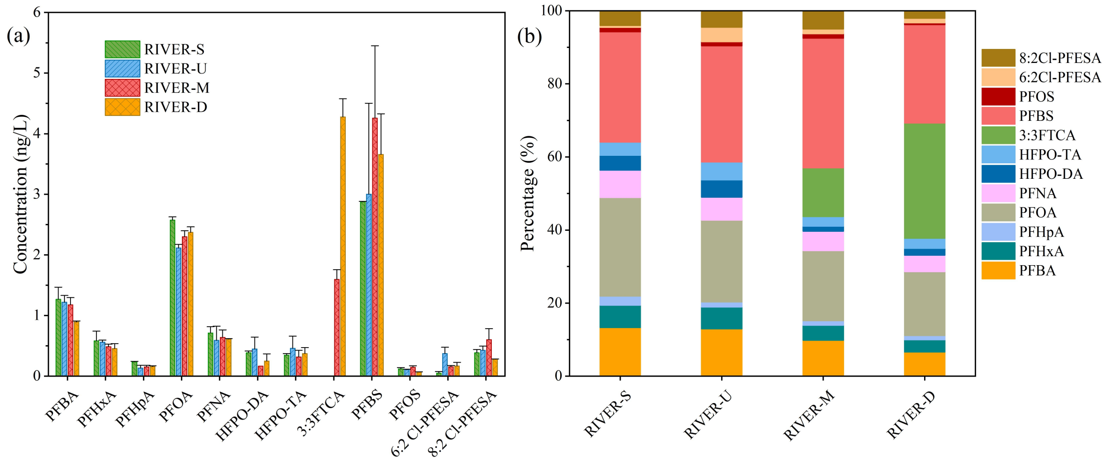


Fig. 4. Levels (a) and patterns (b) of PFAS in river samples from a rural area, North China.

The detection of HFPO-DA and HFPO-TA, even at low levels, signals the emergence of alternative PFAS in rural water systems. HFPO-DA concentrations were slightly higher than HFPO-TA, consistent with its potential formation as an oxidation product (Bao et al. 2019). Although these concentrations were significantly lower than those in industrialised rivers like the Xiaoqing River (Wang et al. 2022), their presence in rural streams emphasises the potential for long-range atmospheric transport of these pollutants. Taken together, the observed spatial patterns—such as the increasing 3:3FTCA concentration downstream and the presence of emerging PFAS in a rural setting with limited local industrial sources—along with the known meteorological conditions favoring pollutant transport from industrialized northern regions, provide indirect but consistent evidence supporting long-range atmospheric transport of PFAS to the study area. Compared to global studies, ΣPFAS concentrations in the LZZ stream (9.54–13.6 ng/L) were similar to those in Lake Victoria, Uganda, and rural rivers in the Tibetan Plateau, but much lower than urban rivers affected by intensive industrial activities (Dalahmeh et al. 2018; Ren et al. 2023; Li et al. 2011). According to Boettger et al. (2025), current targeted methods may miss over half of the PFAS in water, and emerging PFAS often appear in groups. In our study, despite detecting some short-chain or emerging

PFAS, it is highly likely that many other emerging PFAS were overlooked. The grouped co-occurrence of emerging PFAS further complicates the detection process. This suggests that relying solely on targeted analysis may not provide a comprehensive understanding of PFAS pollution. Therefore, future research should emphasise non-targeted analysis to capture the full spectrum of PFAS, especially emerging ones, in different environmental media. This approach will enhance our understanding of the environmental behaviours, sources, and potential risks of PFAS in rural areas like Laozhuangzi Village.

3.4 PFAS in tap waters

A total of 12 PFAS, including 7 legacy PFAS and 5 alternatives (PFPeA, HFPO-DA, HFPO-TA, 6:2Cl-PFESA, and 8:2Cl-PFESA), were detected in tap water, as shown in Figure 5. The ∑PFAS concentrations ranged from 7.16 to 25.1 ng/L, with TAP-3 exhibiting the highest level, nearly twice that of TAP-1 and TAP-2, and over three times that of TAP-4. PFBS was the most abundant compound, followed by PFOA, PFPeA, PFBA and PFHxA, while other PFAS, including PFHpA, PFOS, HFPO-TA, and Cl-PFESAs, contributed less than 3%. PFPeA was exclusively detected in tap water samples, with concentrations ranging from 0.298 ng/L in rural village water to 7.82 ng/L in the municipal water supply (TAP-3). Its presence in treated water but absence in other water sources (e.g., surface water or well water) suggests its origin is closely tied to water treatment and distribution systems. Studies have shown that PFPeA can leach from materials used in water supply infrastructure, such as pipes, coatings, and storage tanks, or result from chemical degradation during the treatment process (Chen et al. 2021b; Feng et al. 2021). Furthermore, the elevated PFPeA levels in TAP-3, a centralised municipal water system, may be attributed to longer distribution networks and higher water treatment intensities compared to rural village systems (TAP-1, TAP-2, and TAP-4). This trend aligns with findings in other regions where PFPeA concentrations increased after water treatment due to its chemical stability and resistance to conventional removal methods (Chen et al. 2021b).

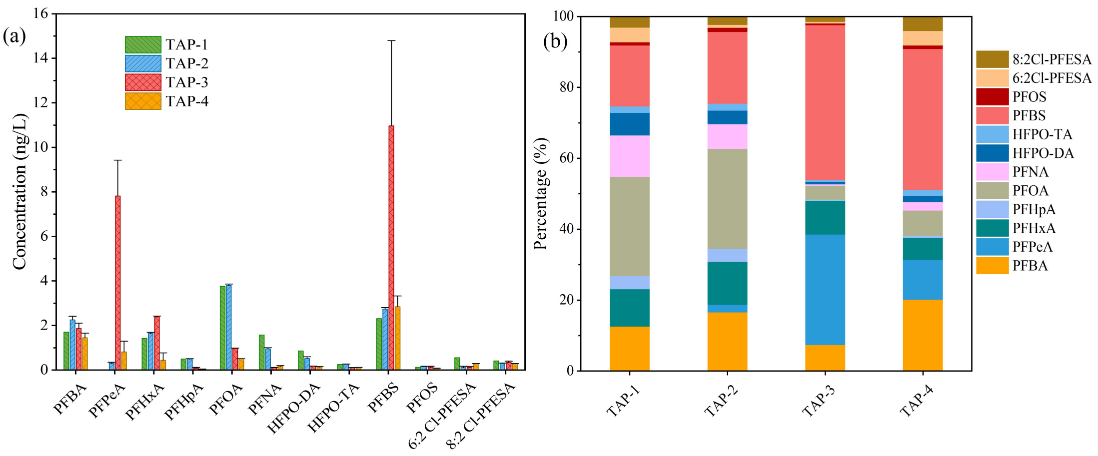


Fig. 5. Levels (a) and patterns (b) of PFAS in tap water samples from a rural area, North China.

The dominance of PFBS, PFBA, and PFPeA in tap water reflects the increasing use of short-chain PFAS as replacements for long-chain compounds, owing to their lower bioaccumulation potential. However, their persistence in water systems poses challenges for removal, as conventional treatment processes such as coagulation, filtration, and chlorination show limited efficiency in reducing short-chain PFAS (Sheng et al. 2018; Chen et al. 2021b; Xin et al. 2019). The significantly higher Σ PFAS in TAP-3 compared to other samples suggests localized differences in raw water quality or treatment practices. The reservoir supplying LZZ village may contribute PFAS contamination to the municipal system, which is compounded by additional leaching from infrastructure during water transport. Compared to other studies, the ΣPFAS levels in LZZ tap water (8.26 – 25.0 ng/L) are moderate. For instance, Huantai, Shandong Province, reported PFOA concentrations as high as 31 ng/L, while PFPeA was a minor contributor (Table S6) (Feng et al. 2021; Xie et al. 2021; Li et al. 2019). In contrast, the relatively high PFPeA levels in LZZ tap water align with urban studies where extensive treatment and distribution networks increase exposure to infrastructure-related PFAS (Chen et al. 2021b). The detection of alternative PFAS, such as Cl-PFESA and HFPO-DA, at low levels is consistent with their limited usage and environmental persistence (Xie et al. 2021).

3.5 PFAS in well water

The Σ PFAS concentrations in well water samples were the lowest among all sample types, ranging from 4.43 to 5.70 ng/L (Fig. 6). The PFAS composition was consistent across all three wells, with 3:3FTCA, PFBS and PFBA as the primary contributors, while legacy PFAS (e.g., PFOS and PFOA) were detected only at trace levels. The predominance of short-chain PFAS reflects their higher mobility in groundwater systems due to weaker sorption to soil organic matter and aquifer sediments. These compounds are more likely to percolate into groundwater compared to long-chain PFAS, which tend to adsorb onto soil particles or organic matter through hydrophobic interactions and electrostatic forces (Li et al. 2022; Zhou et al. 2021; Borthakur et al. 2021). The relatively higher proportions of Cl-PFESAs and HFPO-DA suggest contributions from modern PFAS sources, potentially linked to regional agricultural practices and household product usage, while the low levels of legacy PFAS align with the limited industrial influence in this area.

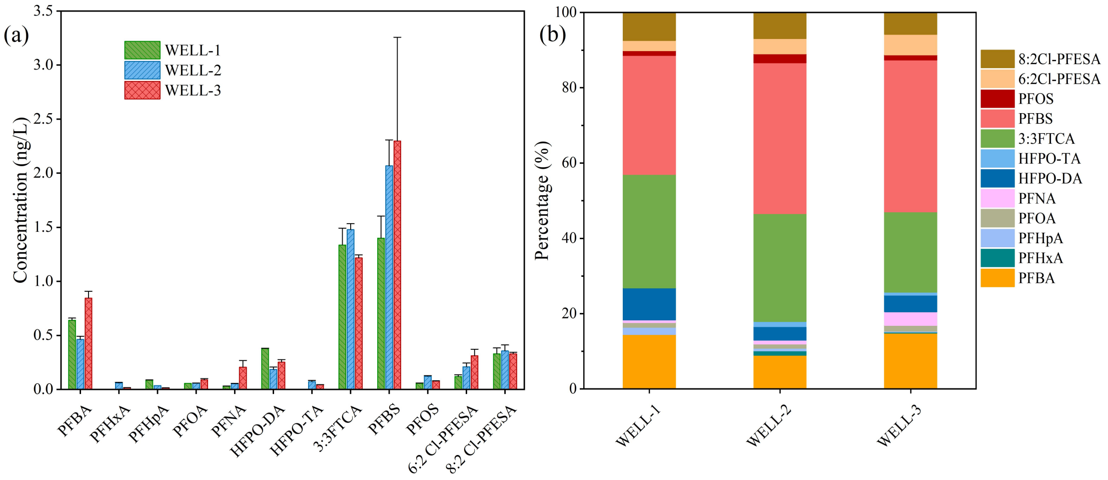


Fig. 6. Levels (a) and patterns (b) of PFAS in well water samples.

The low ΣPFAS concentrations in well water likely result from the isolation of the deep aquifer from surface contamination. Studies have shown that PFAS concentrations decrease with depth due to reduced interaction with surface pollutants and attenuation mechanisms like adsorption and filtration through soil layers (Li et al. 2022; Zhou et al. 2021; Xingchun et al. 2023). Soil organic matter and humic substances in the region likely enhance the retention of long-chain PFAS, while short-chain PFAS remain more mobile, explaining their dominance in the well water samples (Chen et al. 2016; Xu et al. 2021). Seasonal snowmelt could also contribute PFAS to shallow soils, which may gradually infiltrate into the aquifer, but this effect is likely limited due to the depth of the water table and the natural filtration capacity of the soil matrix (Zhou et al. 2021).

PFAS profiles in LZZ well waters are consistent with findings from other rural and agricultural groundwater systems. For example, PFBS and PFBA are frequently reported as dominant PFAS in rural groundwater in northern and eastern China, including Liaocheng, Tai'an, and Beijing suburbs, where Σ PFAS concentrations typically range from a few ng/L to over 30 ng/L depending on proximity to pollution sources like landfills or sewage reservoirs (Table S6) (Chen et al. 2016; Li et al. 2022; Xingchun et al. 2023). Globally, similar PFAS profiles have been observed in rural areas, such as the Maozhou River Basin in China and rural Australia, where PFBS and PFBA were also dominant (Li et al. 2020; Hepburn et al. 2019; Xu et al. 2021). These studies suggest that the widespread occurrence of short-chain PFAS in groundwater reflects their persistence and mobility, as well as the growing application of these compounds to substitute long-chain PFAS.

3.6 PFAS correlation analysis

An important question was to understand the correlations between PFAS concentrations in LZZ village. Fig. 7 shows how relationships among the concentrations of all individual PFAS were explored across four different samples at LZZ village with Spearman's correlation analysis. Positive correlations (blue circles) indicate that analyte concentrations change together, suggesting shared sources or similar environmental behaviours. Negative correlations (red circles) show that one analyte is present while the other is absent. Strong blue correlations among multiple analytes may reflect common origins, such as single-source emissions or degradation of shared precursors. Conversely, white (no correlation) or red circles suggest mixed or diverse sources (Pike et al. 2021).

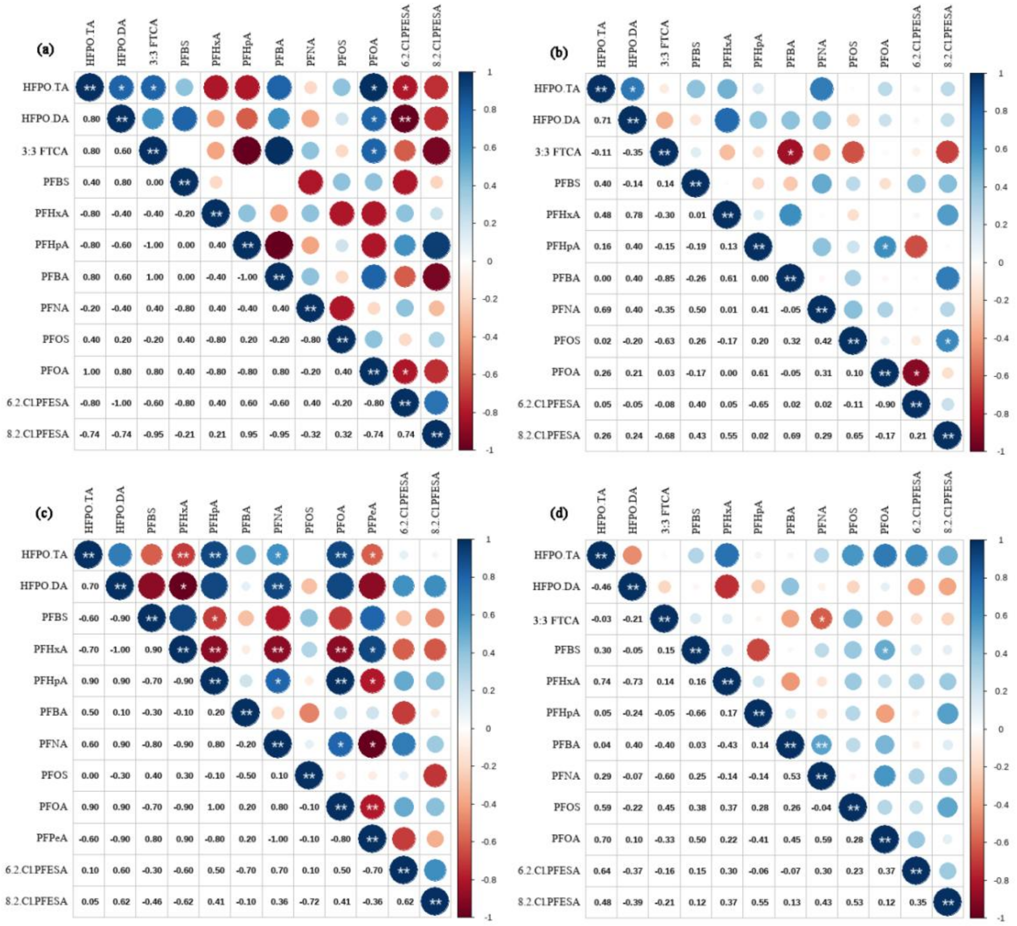


Fig. 7. Correlation plots of PFAS in snow (a), river (b), tap water (c), and well water (d) in LZZ village. Circle size reflects correlation strength, with larger circles indicating stronger links. Blue denotes positive correlations, red indicates negative correlations, and color intensity signifies the magnitude of the relationship. "*" indicates significant correlation ($p < 0.05$), and "**" indicates highly significant correlation ($p < 0.01$).

PFAS contamination in environmental matrices reflects complex interactions between source inputs, environmental transport, and transformation processes. Snow samples in LZZ village exhibited distinct correlation patterns, reflecting contributions from atmospheric deposition and potentially long-range transport. Unlike previous studies in rural (Pike et al. 2021), urban (Kwok et al. 2010), and remote areas, where PFOA exhibited a strong positive correlation with PFNA, these PFAS showed weak negative correlations in LZZ snow. Instead, PFOA exhibited significant positive correlations with HFPO-TA and HFPO-DA, suggesting a shared source. Both HFPO-TA and HFPO-DA are replacement PFAS introduced after regulatory restrictions on PFOA, and their co-occurrence indicates a shift in industrial emissions toward newer compounds (Feng et al. 2021). Conversely, 6:2Cl-PFESA demonstrated strong negative correlations with PFOA and its substitutes, suggesting distinct sources. 6:2Cl-PFESA is often linked to specific industrial applications and may have been introduced into the atmosphere through localised emissions rather than long-range transport. Weak or insignificant correlations among PFSAs (e.g., PFOS, PFBS, and 6:2Cl-PFESA) further indicate independent sources or differing environmental behaviours. PFOS and PFBS, as legacy PFAS, may reflect historical emissions deposited into the environment over time. Their presence in snow likely results from both ongoing low-level emissions and the remobilisation of previously deposited residues. These findings illustrate the complexity of snow as a matrix that captures PFAS from diverse sources, including both recent and historical inputs.

Tap water samples exhibited a high number of significant correlations, many of which were negative. Unlike snow, where PFAS relationships primarily reflect atmospheric sources, the correlations in tap water are likely influenced by disturbances during water treatment and distribution (Dvorakova et al. 2023; Ingold et al. 2023; Li et al. 2019). For example, PFOA, PFNA, and PFHpA showed significant positive correlations in tap water. This is consistent with previous findings in other regions, such as the Czech Republic, where similar correlations were observed in tap water, likely due to common industrial or environmental sources (Dvorakova et al. 2023). The positive correlations among these PFAS suggest they may originate from shared contamination sources, such as industrial discharges, or reflect similar environmental behaviours during water treatment and distribution. However, significant negative correlations were observed between PFHxA and PFHpA, suggesting that these compounds may originate from distinct sources or undergo differential removal during treatment processes. PFHxA is often associated with industrial inputs, while PFHpA may be linked to the degradation of longer-chain precursors, such as 6:2FTOH (Garnett et al. 2022). Furthermore, no significant correlations were observed between PFOS and PFBS, deviating from findings in German tap water (Ingold et al. 2023), likely due to differences in PFAS usage and water treatment technologies.

Correlations in river and well water samples were generally weaker and less consistent than those in snow and tap water, reflecting their exposure to mixed sources and localised inputs. For example, in river water (Fig. 7b), PFOA and PFNA showed weak positive correlations, aligning with findings in TP rivers (Chen et al. 2023; Ren et al. 2023). However, the significant correlation between PFOA and PFHpA suggests contributions from shared sources, such as upstream discharges or the degradation of precursors. In contrast, correlations between PFSAs (e.g., PFOS, PFBS) were insignificant, highlighting the influence of heterogeneous inputs, including domestic wastewater and agricultural runoff. Few significant correlations were all positive in the well samples from LZZ (Fig. 3d). Previous studies on groundwater measurements have investigated comparable relationships (Shen et al. 2016), including the Loess Plateau (northwestern China) (Zhou, et al. 2021), North Carolina (Kotlarz et al. 2024), and the TSR (Li et al. 2022).

Across all matrices, HFPO-TA consistently showed significant positive correlations with PFOA, except in river water. This suggests that HFPO-TA, a replacement for PFOA, shares similar industrial sources or degradation pathways (Chen et al. 2020). HFPO-DA, another PFOA substitute, exhibited weaker correlations, possibly due to differences in its environmental transport or reactivity (Feng et al. 2021). PFOA and PFNA, known degradation products of 8:2FTUCA, were positively correlated in river and tap water but showed weaker or negative correlations in snow and well water (Ellis et al. 2004). This pattern suggests that PFAS degradation processes, such as the oxidation of 8:2FTOH, contribute to their concentrations in surface and treated water systems. However, the absence of detectable precursors like 8:2FTUCA and 8:2FTOH in snow and groundwater implies additional sources or pathways. Direct quantification of these precursors is needed to confirm their role in PFAS dynamics. The distinct behaviours of PFAS in different matrices underscore the influence of both environmental processes and human interventions. Snow integrates atmospheric deposition from local and distant sources, while tap water reflects the impact of water treatment and distribution. River water captures mixed inputs from upstream and local sources, and well water reflects historical contamination patterns shaped by soil leaching and groundwater flow.

3.7 Health risk assessment

In the present study, the concentrations of PFOA and PFOS ranged from 0.028 and 0.052 to 3.80 and 0.154 ng/L, significantly below the Chinese drinking water quality standard published in 2022, which specified PFOA at 80 ng/L and PFOS at 40 ng/L, indicating the lower risk due to these PFAS. Currently, the EU's Drinking Water Directive states that the aggregate level of 20 prevalent PFAS shall not go beyond 100 ng/L, which proves that the studied areas are at a low health risk (European Environment Agency 2024). Although there are no statutory standards for PFAS in drinking water in England and Wales, a more stringent standards implemented in the UK indicate low-risk PFAS levels are 10 ng/L and medium risk are 10-100 ng/L (Royal Society of Chemistry 2023). In this situation, tap water in studied areas is medium risk. In addition, the US EPA issued the final National Primary Drinking Water Regulation (NPDWR) regarding six PFAS in 2024, establishing legally binding concentration limits called Maximum Contaminant Levels (MCLs) (US EPA 2021a, 2021b). The MCL for PFOA, PFOS, PFHxS, PFNA, and HFPO-DA are 4.0, 4.0, 10, 10, and 10 ng/L, respectively. PFAS mixtures with at least two or more of PFHxS, PFNA, HFPO-DA, and PFBS evaluated via a Hazard Index MCL (1 unitless). Though the NPDWR is restricting, public water systems have had a few years to implement solutions to reduce PFAS.

The US EPA has established an oral noncancer reference dose (R*f*D) of 20 ng/kg/day for both PFOA and PFOS (US EPA 2016b, 2016c). Table S7 provides a summary of the EDI calculations for different age groups in LZZ village and Ningyang Town centre. The EDI of PFOA and PFOS in tap water varied by location and age group. For LZZ village, maximum PFOA EDI reached 0.030–0.058 ng/kg bw/day (toddlers-adults), exceeding Ningyang Town centre (0.011–0.022 ng/kg bw/day). While the PFOS EDI in LZZ (0.0013–0.0025 ng/kg bw/day) was comparable to Ningyang (0.0014–0.0027 ng/kg bw/day). Notably, despite lower individual PFOA levels, ΣPFAS concentrations in Ningyang Town centre were twice those in LZZ. This discrepancy highlights that the suburban environment may accumulate diverse PFAS mixtures, leading to higher cumulative exposure risks even when regulated compounds (e.g., PFOA/PFOS) are controlled. EDI of PFOA and PFOS in LZZ village well waters are both 1 to 2 orders of magnitude lower than that in tap water, indicating that well water is safer in terms of these PFAS exposure, consistent with previous findings (Zhou et al. 2021). In brief, PFAS in the studied area is at acceptable levels according to the present regulations of China and the EU. However, it is a medium risk according to the current UK PFAS standard, which should be addressed in the future. Therefore, more research is needed to study how these emerging PFAS spread, break down, and impact ecosystems and human health.

4. Conclusion

This research investigated the presence, spatial spread, and distribution features of 72 PFAS in snow, river, tap water, and well water from a rural area in Shandong Province, northern China. The findings demonstrate that PFAS are widespread in the environment, with ΣPFAS concentrations ranging from 4.21 to 453 ng/L across all samples. Snow samples, dominated by 3:3FTCA and HFPO-DA, highlight the significant role of atmospheric deposition in introducing PFAS into rural ecosystems. The presence of PFAS in river and tap water samples suggests combined contributions from industrial and domestic discharges, as well as seasonal snowmelt. The detection of PFPeA exclusively in tap water underscores the influence of treated water systems on PFAS profiles. In well water, the dominance of PFBS, 3:3FTCA and PFBA reflects the mobility and persistence of short-chain PFAS in subsurface environments, shaped by soil and aquifer filtration processes. Notably, the estimated daily intake of PFOA and PFOS in well water was 1 to 2 orders of magnitude lower than in tap water, indicating safer groundwater sources in terms of these PFAS exposure in the study area. These results highlight the complexity of PFAS transport and transformation in rural environments and highlight the need for further research into their sources, behaviours, and potential health risks, particularly in underrepresented rural regions of China.

Supporting Information

The relevant text, tables, and figures are in the supplementary file.

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Statements and Declarations

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Competing Interests

The authors have no relevant financial or non-financial interests to disclose.

Author Contributions

CEC and YH contributed to the study conception and design. Material preparation, data collection and analysis were performed by XYL, ZXZ and JXZ. The first draft of the manuscript was written by XYL and ZXZ, and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

Data Availability

The raw datasets generated during and/or analysed during the current study are available from the corresponding author on reasonable request.