Heavy Metals and PAHs Drive Ecological and Health Risks in Chinese Water Level Fluctuation Zones

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Abstract

Water-level fluctuation zones (WLFZ) adjacent to rivers, lakes and reservoirs are ecologically sensitive areas, regulating water quality and maintaining ecological health. However, research on the pollution characteristics, their spatiotemporal variability and primary sources as well as the pollution-related ecological and health risks in WLFZ soils remains limited. This study developed a comprehensive dataset of soil pollutants in Chinese WLFZs, covering nearly 3,000 sampling locations across 353 sites. The findings revealed that heavy metals and polycyclic aromatic hydrocarbons (PAHs) are the key pollutants, with the Three Gorges Reservoir identified as the primary focus area. Among the heavy metals, Cd exhibited relatively severe contamination, with Monte Carlo simulations indicating a 68.5% probability of moderate or higher contamination levels. The pollution levels of heavy metals decreased in the order: Cd > As > Pb > Cu > Zn > Cr > Hg > Ni. Despite limited non-carcinogenic risks, carcinogenic risks from As and Cd are concerning. Although the non-carcinogenic risks posed by heavy metals were limited, the average carcinogenic risk indices for children due to As (1.42e-05) and Cd (2.19e-06) were relatively high and warrant attention. Some Chinese WLFZs are contaminated with PAHs, with BaP identified as the dominant carcinogenic compound. Pollution in Chinese WLFZs is primarily attributed to coal combustion, industrial and agricultural activities, and navigation. Furthermore, current research on emerging pollutants in WLFZs remains limited, and risk assessments of their impacts on ecosystems are urgently needed. Our results provide the scientific basis for developing management strategies for pollution control in Chinese WLFZs.

Keywords: Water-level fluctuation zone; Soil pollution; Risk assessment; China

1. Introduction

Water-level Fluctuation Zones (WLFZ) are vulnerable and sensitive transitional area between fully aquatic and terrestrial ecosystems (Keller et al., 2021; Liu et al., 2024). They experience periodic submersion and exposure driven by water level fluctuations in adjacent aquatic environments and are widely distributed around rivers, lakes, reservoirs and other aquatic ecosystems (Pei et al., 2018). The WLFZ also constitutes a critical zone where organic matter and nutrients interact, migrate and transform between soils and overlying waters, exerting an important regulatory influence on material cycling and energy flow within the watershed (Bao et al., 2015; Liu et al., 2024; Liu et al., 2025). For instance, the repeated redox shifts characteristic of WLFZ soils enhance the mineralization of organic matter and the release of CO_2 and CH_4, making this zone a major hotspot of greenhouse gas emissions in reservoir ecosystems (Keller et al., 2021; Yang et al., 2020; Zhang et al., 2021). Moreover, hydrological fluctuations involving alternating wetting and drying can influence the speciation and environmental behavior of pollutants (Zhang et al., 2019; Zhang et al., 2022). Under reducing conditions, heavy metals may experience valence changes that increase their mobility and bioavailability, whereas under oxidizing conditions they can be stabilized (Pei et al., 2018; Ye et al., 2021). In addition, organic contaminants such as polycyclic aromatic hydrocarbons (PAHs) may be adsorbed/desorbed under periodic wetting–drying cycles, influencing their distribution between soils and water (Han et al., 2021; Hu et al., 2017; Sun et al., 2020). Furthermore, these zones, depending on environmental conditions, can be regarded as both sources and sinks of pollutants (Zhang et al., 2019). During the non-flood season, pollutants transported from WLFZs to adjacent environments, particularly to lower-lying aquatic ecosystems, are predominantly derived from anthropogenic sources (Pei et al., 2018; Zhang et al., 2019).

These sources may include agricultural activities such as floodplain cultivation, as well as industrial discharges, municipal wastewater, and vehicular emissions (Fig. 1a). Under flood conditions, floodwaters may carry pollutants into WLFZs, where they subsequently accumulate through deposition processes (Hu et al., 2017).

Human population growth and economic development have exerted substantial impacts on WLFZs, affecting both their spatial distribution and the intensity of environmental and ecological pressures they face (Keller et al., 2021; Zhang et al., 2019). For example, growing demands for renewable energy have driven a global increase in the number and size of hydropower reservoirs (Fig. 1b), consequently expanding the extent of WLFZs (Soued et al., 2022). These newly created or modified zones are frequently exposed to intensified anthropogenic disturbances, such as changes in land use, pollutant inputs, and modifications to hydrological regimes, which often collectively exacerbate ecological degradation within WLFZs (Hu et al., 2017; Pei et al., 2018). Notably, China has constructed more than 97,000 dams, including nearly 40% of the world's largest dams, with their reservoirs contributing to a substantial increase in the number and area of WLFZs (Li et al., 2018; Ye et al., 2011). For example, water levels within the Three Gorges Reservoir (TGR), the world's largest hydropower project, fluctuate between 145 and 175 meters above mean sea level in front of the dam (Fig. 1c), creating one of the largest WLFZs in the world (Bao et al., 2015; Gao et al., 2016; Yang et al., 2022). Given the substantial increases in Chinese WLFZs, establishing their soil pollution status is important for developing strategies to protect water quality and maintain ecosystem health across the nation.

Previous studies have shown that due to human activities, the soils in many of China's WLFZs are widely contaminated with heavy metals (Dong et al., 2023; Gao et al., 2016), polycyclic aromatic hydrocarbons (PAHs) (Han et al., 2021; Hu et al., 2017), pesticides (Sun et al., 2013), and microplastics (Zhang et al., 2019). For example, in the TGR, discharges of industrial and domestic wastewater, along with agricultural runoff containing fertilizers and pesticides, are major causes of soil pollution within the reservoir's WLFZ (Fig. 1d). Notably, heavy metal contamination has been frequently reported, with cadmium (Cd), arsenic (As), and lead (Pb) being the most typical heavy metal elements that represent significant threats to human health (Ye et al., 2011; Zhang et al., 2021). For example, a study undertaken in the Wushan and Zigui sections of the TGR's WLFZ identified serious Cd pollution that was linked to high human health and ecological risks given that high-dose Cd exposure is associated with kidney and cardiovascular disease, as well as cancer (Huang et al., 2019a; Pei et al., 2018; Yan et al., 2022). Similarly, severe Cd pollution in WLFZ soil of Danjiangkou Reservoir (Hubei Province, Central China) as a result of industrial and agricultural practices has impacted water quality with implication for the safety of water supplies (Dong et al., 2023). Detection rates of persistent organic pollutants (POPs) including PAHs, pesticides, and polychlorinated biphenyls (PCBs) in the soil of Chinese WLFZs have also been relatively high (Hu et al., 2017; Wu et al., 2016; Yuan et al., 2023). Of these, PAHs have been a primary focus of research due to their carcinogenic, teratogenic, and mutagenic properties, along with their persistence in the environment (He et al., 2019; You et al., 2024). The dominant PAH species in the WLFZs include naphthalene, phenanthrene, and fluoranthene (Han et al., 2021). Although these earlier studies have assessed soil pollution within China's WLFZs, most have largely focused on specific geographical areas or individual pollutants. To the best of our knowledge, a comprehensive analysis of soil pollution across China's WLFZ is still lacking.

Several indices, such as the geo-accumulation index, pollution load index, Nemerow pollution index, and potential ecological risk index, have been developed for assessing pollution levels and have been applied to evaluate soil pollution within WLFZs (Dong et al., 2023; Huang et al., 2023; Zhang et al., 2021). These indices can be employed to provide comprehensive assessments of soil pollution and associated ecological risks, facilitating development of effective management and remediation strategies for WLFZs (Hu et al., 2017; Pei et al., 2018). Despite the utility of these indices, current WLFZ soil pollution risk assessments rely entirely on deterministic risk assessment (DRA) methods, primarily considering the total concentration of pollutants and the most likely exposure parameters (Huang et al., 2023; Huang et al., 2024; Lin et al., 2023). These approaches lead to either underestimation or overestimation of the risks (Huang et al., 2024; Lin et al., 2023). In contrast, probabilistic risk assessment (PRA) applies stochastic methods such as Monte Carlo simulations to characterize uncertainty and variability in input parameters. This enables the estimation of a probabilistic range of potential outcomes and supports a more comprehensive understanding of risk under uncertainty (Guan et al., 2022; Wu et al., 2021).

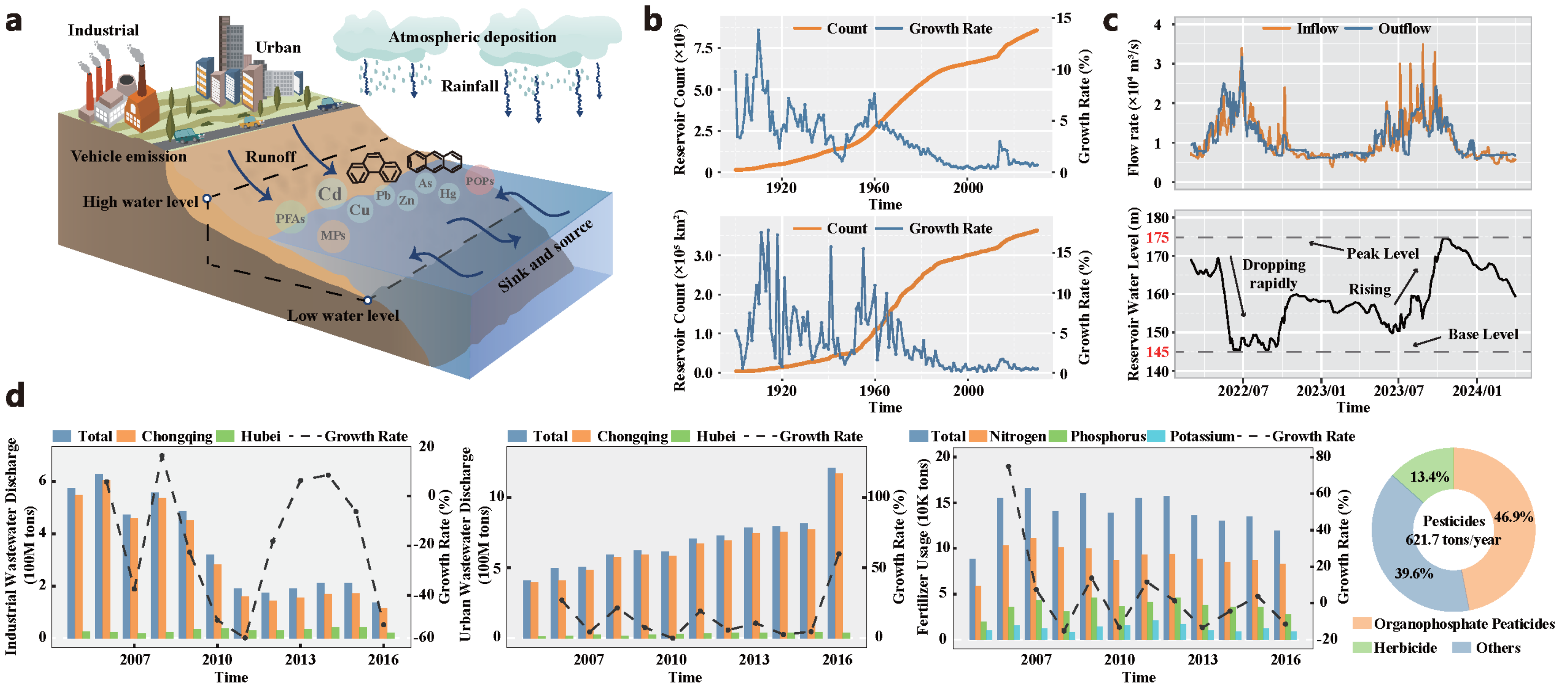


Fig. 1. Schematic diagram of the water-level fluctuation zone (WLFZ) and its potential pollution sources. a. Environmental behavior of typical pollutants in the WLFZ. **b.** The cumulative count and area of reservoirs globally. **c.** Water level fluctuations and inflow/outflow rates in the Three Gorges Reservoir. **d.** The discharge of industrial wastewater, urban wastewater, and the application of fertilizers and pesticides in the Three Gorges Reservoir area (Source: Bulletin on the Ecological and Environmental Monitoring Results of the Three Gorges Project).

Addressing key knowledge gaps, this study aims to: (i) systematically summarize the research on soil pollution in China's WLFZs, identifying the main pollutants of concern and the core geographical areas via a comprehensive literature review; (ii) assess soil pollution and related health risks in Chinese WLFZs via the application of a Monte Carlo analysis within the PRA framework, and propose a prioritized risk management catalogue; and (iii) identify potential pollution sources and key contaminants in China's WLFZs using the positive matrix factorization (PMF) model and quantify health risks associated with each source. The study is the first to provide a comprehensive, nationwide, understanding of soil pollution in China's WLFZs, including evaluating the associated ecological and health risks. It provides a scientific basis for developing targeted management strategies to mitigate environmental contamination and protect both human and ecosystem health.

2. Materials and methods

2.1 Development of a soil pollution database for China's WLFZ

A database of soil pollution in China's WLFZs was established using peer-reviewed papers retrieved from Web of Science Core Collection and the references cited by these papers (search conducted on March 18, 2024). Our search strings combined synonyms with "OR" and main elements with "AND" (Wang et al., 2024). The search terms included the following: TS (Topic Search) = (("drawdown zone" OR "riparian zone" OR "fluctuation zone" OR "fluctuating zone" OR "water level fluctuation zone" OR "hydro-fluctuation belt" OR "littoral zone") AND ("contaminated" OR "polluted" OR "contamination" OR "pollution")). A Perl-based script for "front page filtering" (detailed in Fig. S1) was developed to screen the title, abstract, and keywords of each retrieved paper and to exclude irrelevant records (Fu et al., 2012; Yan et al., 2022). This study employed the Newcastle-Ottawa Scale to assess the quality of each paper (Ofori et al., 2021; Yan et al., 2022), and studies lacking specific sampling location information were excluded, as the absence of such details limits effective comparison with regional soil background values. Additionally, studies focusing on deeper soil layers were also excluded, as surface soils are more representative of recent pollution inputs and have a more direct relevance to human health risk assessment (Table S1, S2) (Hu et al., 2020; Huang et al., 2019b; Yan et al., 2022). As a result, a total of 54 high-quality papers containing information about soil pollution from 353 WLFZ sites within China were identified. In total, data from individual 2,959 sampling locations were reported across these sites. Each pollutant type detected at a site was counted as a separate entry, irrespective of whether multiple pollutants were reported at the same location. Notable sites encompassed both WLFZs associated with reservoirs (e.g., the Three Gorges and Danjiangkou reservoirs) and floodplain-type zones along undammed rivers, reflecting the geographic distribution of China's major WLFZs. The following information was extracted for each paper: (i) publication information (title, keywords, publication year); (ii) sampling locations and types of pollutants; and (iii) the average values of pollutant concentration (Table S3-S4). These pollution data were obtained from tables or extracted from figures using GetData Graph Digitizer software (Eftim et al., 2017). A keyword co-occurrence network was constructed to identify the main pollutants and frequently studied regions across China (Yan et al., 2022).

2.2 Pollution assessment of major pollutants

2.2.1 Index of geo-accumulation (I_{geo})

The index of geo-accumulation (I_{geo}) can be used to estimate contamination from anthropogenic sources by eliminating the influence of background values. It has been widely applied to assess heavy metal contamination in various environments (Barbieri, 2016; Dong et al., 2023; Pei et al., 2018; Yan et al., 2022). I_{geo} for each heavy metal in the soil was calculated at each WLFZ sampling location using equation (1):

$$I_{geo} = \log_2 \frac{C_n}{1.5B_n} \quad (1)$$

where C_n is the concentration of heavy metal element n extracted from the selected papers (Table S3); and B_n is the geochemical background concentration of that element corresponding to the Chinese province in which each sampling site is located, determined by site's geographic coordinates (Table S5). The I_{geo} index was classified into seven categories (0-6), representing the degrees of pollution from practically uncontaminated to extremely contaminated, as detailed in Table S6.

2.2.2 Risk quotient (RQ) of 16 PAHs

The risk quotient (RQ) method, as described in equation (2) (Arias et al., 2023; Cao et al., 2010), was applied to evaluate the ecological risk of 16 individual PAHs in WLFZ soils across China.

$$RQ_{NCS} = \frac{C_{PAH}}{C_{QV}} \quad (2)$$

where C_{PAH} is the exposure concentration of individual PAHs, and C_{QV} is the corresponding quality value. PAH concentrations below the negligible concentrations (NCs) pose negligible risk, while those exceeding the maximum permissible concentrations (MPCs) pose unacceptable risk (Tables S7-S8). The RQ of NC and MPCs are defined using equations (3) and (4), respectively (You et al., 2024).

$$RQ_{NCS} = \frac{C_{PAH}}{C_{QV(NCs)}} \quad (3)$$

$$RQ_{MPCS} = \frac{C_{PAH}}{C_{QV(MPCs)}} \quad (4)$$

The total RQ is the sum of the RQ values for 16 individual PAH compounds, with only RQ values ≥ 1 taken into account, as shown in equations (5)-(7) (Cao et al., 2010):

$$RQ_{\sum PAHs} = \sum_{i=1}^{16} RQ_i \quad (RQ \geqslant 1) \tag{5}$$

$$RQ_{\sum PAHs\,(NCs)} = \sum_{i=1}^{16} RQ_{(NCs)} \quad (RQ_{(NCs)} \geqslant 1) \tag{6}$$

$$RQ_{\sum PAHs\,(MPCs)} = \sum_{i=1}^{16} RQ_{(MPCs)} \quad (RQ_{(MPCs)} \geqslant 1) \tag{7}$$

Based on the computed RQ_ΣPAHs values, each sample site was assigned to a specific risk class, ranging from risk-free to high risk. The classification criteria for these risk levels are summarized in Table S9.

2.2.3 BaP toxic equivalent concentrations (TEQ_{Bap})

TEQ_{Bap} was used to assess the overall contamination levels of 16 PAHs in the soil for each of the WLFZ sampling locations. This method is based on the multiplication of the concentration of each component by its corresponding toxic equivalency factor (TEF) value, followed by summing the toxic equivalency concentrations of 16 PAHs to reflect the total toxicity (Wang et al., 2011). TEQ_{Bap} was calculated following equation (8) (Wang et al., 2011; You et al., 2024):

$$TEQ_{BAP} = \sum C_{PAH} \times TEF_{PAH} \tag{8}$$

where C_{PAH} is the concentration of 16 PAH compounds extracted for each site from the reviewed papers, and TEF_{PAH} is the corresponding toxic equivalent factors (Table S10).

2.3 Health risk assessments of major pollutants

2.3.1 Exposure assessment

The main exposure pathways to contaminated soil within WLFZ sites considered in the human health risk assessment are oral ingestion, inhalation through the mouth and nose, and dermal absorption (i.e. skin contact) (Hemati et al., 2024). Daily exposure doses (ADD) via ingestion ($ADD_{Ingestion}$), inhalation ($ADD_{Inhalation}$), and dermal uptake (ADD_{Dermal}) were estimated using equations (9)-(11):

$$ADD_{Ingestion} = C \times \frac{CF \times IR_{Ingestion} \times EF \times ED}{BW \times AT} \quad (9)$$

$$ADD_{Inhalation} = C \times \frac{IR_{Inhalation} \times EF \times ED}{PEF \times BW \times AT} \quad (10)$$

$$ADD_{Dermal} = C \times \frac{CF \times SA \times AF \times ABS \times EF \times ED}{BW \times AT} \quad (11)$$

where C is the concentration of specific pollutants (heavy metals and PAHs, mg/kg) obtained from the reviewed literature, and the definitions and values of the other parameters are detailed in Table S11. In addition, localized exposure parameters from the China Exposure Factors Handbook (Table S12) were incorporated into the health risk assessment for adult males and females in China, allowing for a more accurate representation of their exposure characteristics.

2.3.2 Non-carcinogenic risk assessment

Non-carcinogenic risk refers to the potential for negative effects on human health following exposure to pollutants through different pathways including ingestion, inhalation, and dermal absorption (Han et al., 2024; Hemati et al., 2024). The consequences of non-carcinogenic risks are associated with harmful health effects, including adverse reactions, physiological dysfunctions, and diseases affecting the neurological and cardiovascular systems (Gao et al., 2016; Huang et al., 2021; You et al., 2024). The hazard quotient (HQ) was used in this study to assess these non-cancer risk. It is defined as the ratio of daily exposure doses (ADD) to the reference concentration (RfC) as defined in equation (12) (Hemati et al., 2024; You et al., 2024):

$$HQ = \sum \left(\frac{ADD_i}{RfC_i}\right) \quad (12)$$

where ADD_i is the daily exposure dose of pathway i, and RfC_i is the reference dose of pollutants in pathway i. The values of RfC_i are shown in Tables S13-S14. The hazard index (HI), calculated by summing the HQ of each pollutant, is used to assess the overall non-carcinogenic risk (Hemati et al., 2024). If the value of the HQ or Hazard HI exceeds 1, the population may be exposed to potential non-carcinogenic risks.

2.3.3 Carcinogenic risk assessment

Given the potential links of many pollutants to cancer, our study included a carcinogenic risk (CR) assessment for each of the WLFZ sampling locations. Assuming no antagonistic or synergistic interactions between pollutants, the overall carcinogenic risk can be determined as the sum of the carcinogenic risks associated with exposure to various carcinogenic pollutants through different pathways (Tepanosyan et al., 2017). The CR was calculated using equation (13) (Hemati et al., 2024):

$$CR = \sum (ADD_i \times SF_i) \tag{13}$$

where SF_i is the cancer slope factor for different pollutants via pathway i, with their values established from the literature and listed in Tables S14.

2.4 Probabilistic risk modeling and sensitivity analysis

The Monte Carlo method was employed for uncertainty analysis by selecting sets of model parameter values based on defined probability distributions for specific exposure factors (e.g. exposure duration, exposure frequency, ingestion rate, and inhalation rate) (Lin et al., 2023; Wu et al., 2021). This, in turn, provided probability distributions for pollutant risks for soils across the WLFZ sites. In addition, a sensitivity analysis using Monte Carlo simulation was employed to examine how a one-unit change in each individual parameter affects the model results, thereby identifying the most hazardous contaminant (Guan et al., 2022; Jafarzadeh et al., 2022). Specifically, 10,000 Monte Carlo simulations were performed with a 95% confidence level using the Oracle Crystal Ball plugin for Microsoft Excel (detailed in Text S1). The probability distribution settings for each parameter in the Monte Carlo simulation are summarized in Table S15. If the CR or TCR exceeds 1E-04, the population faces cancer risk, while values between 1E-04 and 1E-06 are considered within an acceptable range (Lin et al., 2023; Wu et al., 2021).

2.5 Source apportionment model PMF 5.0

Positive Matrix Factorization (PMF) is a source apportionment receptor model endorsed by the US Environmental Protection Agency (EPA) (Niu et al., 2020). It operates under nonnegative constraints, does not require source spectrum, and can incorporate uncertainty estimates, thereby enhancing the practicality of the analytical results (Niu et al., 2020). Within the PMF model the sample concentration data matrix (X) is decomposed into three factor matrices as described in equation (14):

$$X = GF + E \tag{14}$$

where X is the concentration dataset represented as a matrix with dimensions $n \times m$, G is the source contribution ratio matrix ($n \times p$), F is the source component spectral matrix ($p \times m$), and E is the residuals matrix ($n \times m$). Equation (14) can be transformed into equation (15) (Niu et al., 2020): And the objective function (Q) defined by residuals and uncertainties as described in Equation (16).

$$x_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij} \tag{15}$$

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left(\frac{e_{ij}}{u_{ij}}\right)^2 \tag{16}$$

where u_{ij} is uncertainty of element j in sample i, and was calculated using the lowest method detection limit (MDL), the concentration of element (heavy metals and PAHs), and an error fraction (σ). When $x_{ij} \leq MDL$, u_{ij} was calculated using equation (17); otherwise, u_{ij} was obtained using equation (18) (Niu et al., 2020).

$$u_{ij} = \frac{5}{6} \times MDL \tag{17}$$

$$u_{ij} = \sqrt{(\sigma \times x_{ij})^2 + (0.5 \times MDL)^2} \tag{18}$$

In this study, EPA PMF 5.0 was applied to identify potential pollution sources and quantify their contributions to soil contamination in each WLFZ sampling locations. The analysis process, following the EPA PMF 5.0 User Guide (detailed in Text S2)., used concentration data and associated uncertainty data for each pollutant as inputs, with missing values replaced by the indicator (-999) and the number of runs set to 20 (Sarkar et al., 2017).

2.6 Statistical data analysis and visualization

The Kruskal-Wallis one-way ANOVA was applied to assess the concentration differences for the different pollutants in soils of the WLFZ sampling locations (Hemati et al., 2024). The total link strength (TLS) of keywords, calculated as the sum of a keyword's link strengths, was used to identify key research areas and major pollutants, highlighting their overall connectivity and importance (Yan et al., 2022). In addition, consistent with our previous research (Yan et al., 2023a; Yan et al., 2023b), data processing and visualization were predominantly undertaken in R using the packages “readxl” (version 1.4.3), “cowplot” (version 1.1.3), “reshape2” (version 1.4.4), “ggplot2” (version 3.5.1), and “ggspatial” (version 1.1.9).

3. Results and discussion

3.1 Main pollutants and concentrations in soil of China’s WLFZs

This study ultimately obtained soil pollution data from 2,959 sampling locations across 353 sites within Chinese WLFZs. These sites are primarily located in the TGR, Danjiangkou Reservoir, the Yangtze and Lancang rivers, Taihu Lake and Poyang Lake, covering the major distribution of WLFZs in China (Fig. 2a, Tables S2-S4). Among all recorded pollutant entries across the WLFZ sites, heavy metals accounted for 73.2%, PAHs for 17.6%, and the remainder included pesticides, microplastics (MPs), and per- and polyfluoroalkyl substances (PFASs) (Fig. 2a) According to the keyword frequency statistics from the selected papers, heavy metals represented the highest proportion (total link strength (TLS) = 471), with Cd, Hg, Pb, and As being the primary focus of the research reported. Organic pollutants were primarily represented by PAHs, and the TGR emerged as the dominant location featuring within the selected papers (Fig. 2b). In the keyword network, heavy metals also occupied a central position, further highlighting their significance as a research focus for pollution in China's WLFZs.

The dataset revealed that the mean concentrations (mg kg^{-1}) of the detected pollutants were as follows: As (18.00 ± 10.14), Cd (0.80 ± 1.15), Cr (80.04 ± 59.16), Cu (45.53 ± 31.07), Hg (0.06 ± 0.04), Ni (42.41 ± 44.51), Pb (44.47 ± 29.53), Zn (111.21 ± 60.87), and Σ16 PAHs (0.15 ± 0.17) (Fig. S2-S3). Compared with China's Grade II Soil Environmental Quality Standard, the exceedance rate of Cd was 69.7% (Fig. 2c), indicating that it was the most severely polluting heavy metal. With a relatively low mean concentration of 0.80 mg kg^{-1}, Cd exhibited the highest coefficient of variation (1.44) among all of the assessed pollutants, indicating pronounced relative dispersion and substantial spatial variability, potentially driven by point-source pollution or localized geochemical conditions within the WLFZ soils. Among the 16 PAHs detected in WLFZ soils at varying levels, three-ring PAHs (Acpy, Acp, Flu, PA, and Ant) and four-ring PAHs (FL, Pyr. CHR, and BaA) accounted for relatively high proportions, with mean values of 32.1% and 28.8%, respectively. In contrast, six-ring PAHs had the lowest proportion (7.5%; Fig. S3). In general, the concentrations of major pollutants such as Cd, Cr, Cu, and PAHs in WLFZ soils followed a log-normal distribution (Fig. 2, Fig. S2), with a few sampling locations within the study sites showing extremely high concentrations, likely linked to specific, spatially discrete, pollution sources (Hu et al., 2020; Yan et al., 2022). Pollutant concentrations in WLFZ soils showed no significant differences among sampling elevations (Fig. S4), as indicated by the Kruskal–Wallis test ($P > 0.05$). This pattern may be attributed to varying pollutant input mechanisms across elevations (Zhang et al., 2019). Lower-elevation areas are subject to periodic flooding, which enhances pollutant accumulation through sedimentation, whereas higher elevations, although not affected by flooding, may still receive inputs from atmospheric deposition and terrestrial runoff from adjacent uplands (Ye et al., 2011). These processes together may lead to comparable overall pollutant concentrations across different elevations (Ye et al., 2011; Zhang et al., 2019). The type of WLFZs did influence pollutant concentrations across different elevations (Hu et al., 2017). Taking the TGR as an example, sampling locations were categorized into three geomorphological types based on slope characteristics (Bao et al., 2015). The first type included gently sloping areas with gradients less than 15°, typically found at lower elevations and often covered by relatively thick, uniform soil layers, where pollutants such as heavy metals and organic contaminants tend to accumulate, resulting in elevated pollutant concentrations in the soil. Slopes exhibiting a distinct inflection point, where the gradient transitions from gentle to steep, and characterized by pronounced variability in soil thickness along the slope profile constituted the second geomorphological type. Pollutant concentrations in these areas were more variable, reflecting differences in soil depth and runoff patterns. The third type consisted of steep or cliff-like zones with gradients exceeding 45°, usually associated with thin or exposed soil layers (Bao et al., 2015). These areas are susceptible to wind and water erosion, which leads to the transport and redistribution of pollutants, with pollutants more likely to be carried to lower-slope or downstream regions rather than accumulating locally (Bao et al., 2015; Hu et al., 2017).

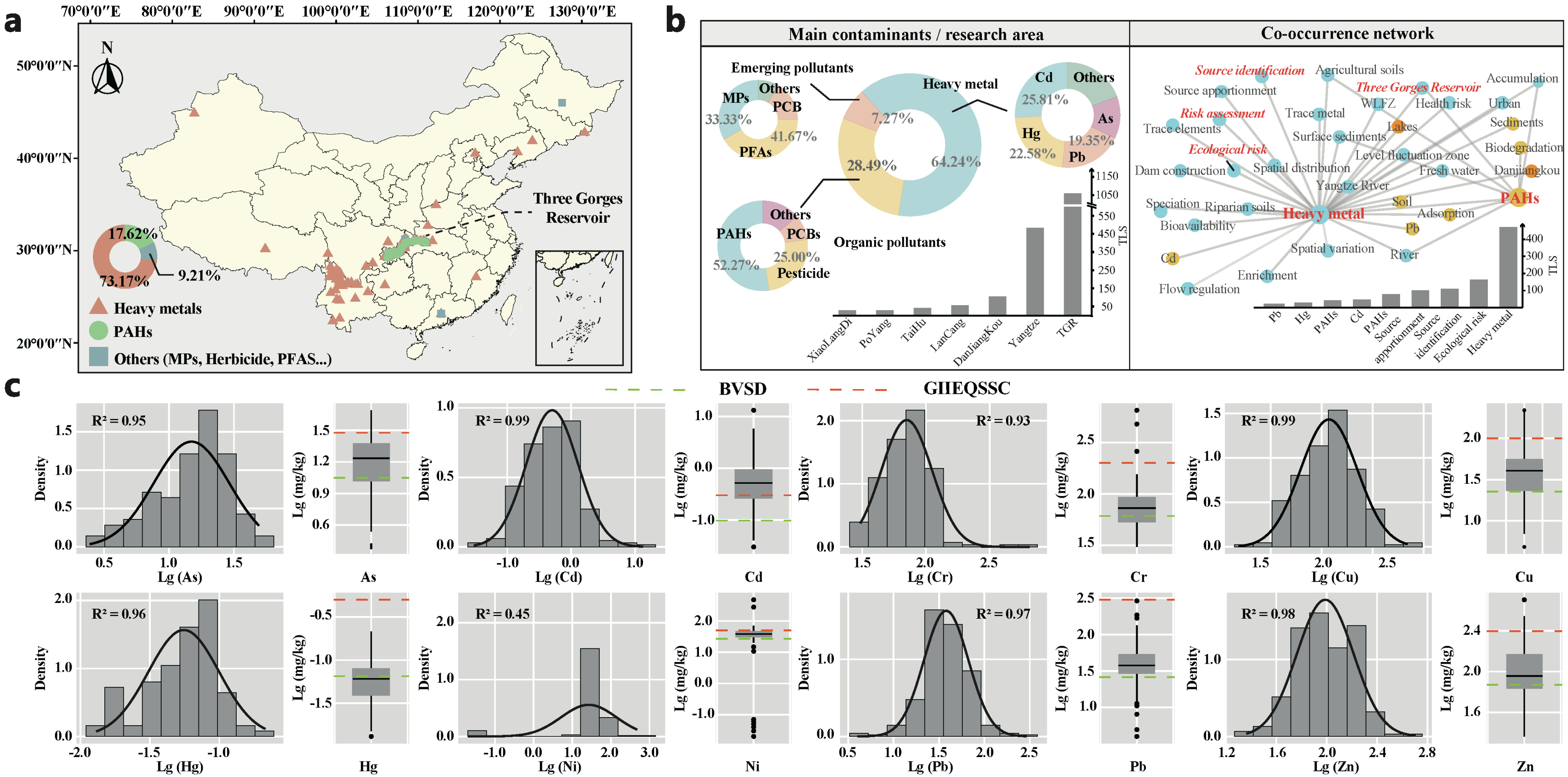


Fig. 2. Primary pollutants and major research sites within existing studies. a. Spatial distribution of primary pollutants reported in WLFZs across China. **b.** Keyword frequency and co-occurrence network for existing studies. **c.** Histograms of log-transformed data and boxplots of soil heavy metal concentrations (BVSC: Background values for soils in China. GIIEQSSC: Grade II environment quality standard for soils in China).

Results also indicated that concentrations of major pollutants (particularly Cd, As, Cr, and Cu) in WLFZ soil samples collected between 2010 and 2015 were significantly higher than those in samples collected before 2010 and after 2015 ($P < 0.05$) (based on data from broadly comparable sites to reduce spatial variability) (Fig. S4). This trend could be attributable to China's strengthened environmental governance, including the launch of the "Water Ten Plan" in April 2015 and the "Soil Ten Plan" in May 2016. These policies were designed to reduce pollutant inputs by enforcing stricter industrial discharge regulations and improving water quality management, with several studies reporting reductions in key pollutants across various environmental media since their implementation (Huang et al., 2019b; Niu et al., 2020).

3.2 Ecological risk of main pollutants in WLFZs

Cd had the highest index of geo-accumulation (I_{geo}), with an average value of 1.69 (moderately contaminated), while Cr and Ni exhibited relatively low average I_{geo} values of -0.70 and -0.88 (practically uncontaminated), respectively (Fig. 3a). Monte Carlo simulation results showed that mean I_{geo} values reduced in the order: Cd > As > Pb > Cu > Zn > Cr > Hg > Ni, with probabilities of moderate or higher contamination levels based on cumulative frequency distribution estimated at Cd (68.5%), As (8.5%), Pb (5.2%), Cu (8.3%), Zn (3.2%), Cr (0.7%), Hg (1.4%), and Ni (8.6%).

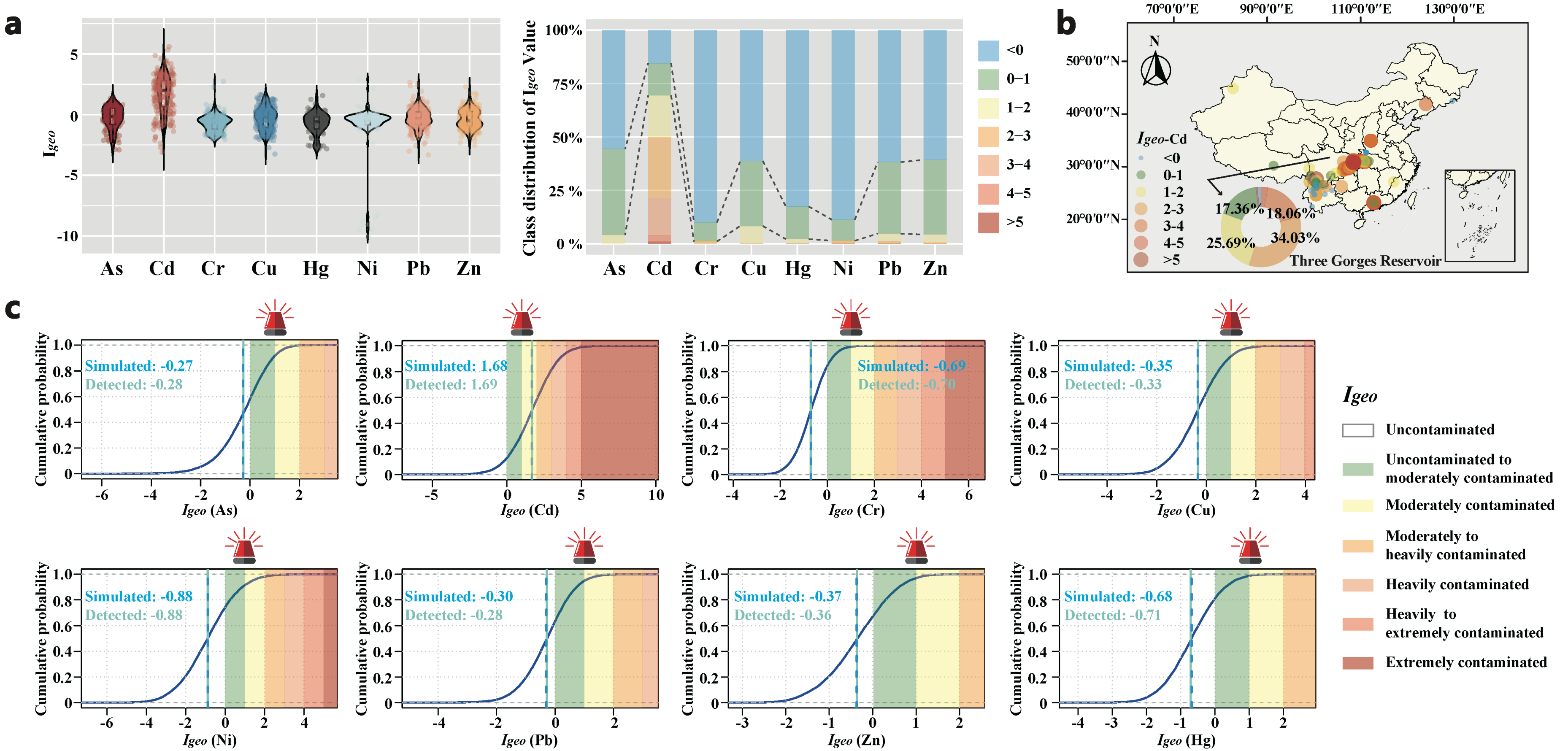
The highest ecological risk level for Cd is consistent with previous findings from rivers, lakes, and sediments in China (Liu et al., 2024). The values of I_{geo} for other heavy metals were mostly classified as "practically uncontaminated" or "uncontaminated to moderately contaminated" and risk levels were lower than soils from industrial and mining areas and within acceptable thresholds (Guan et al., 2022). Low-ring PAHs exhibited higher risk quotients, with Nap (13.47) and Pyr (10.96) having the highest values that were classified as moderate-risk. In contrast, the risk quotients for CHR

(0.08), DBA (0.07), and BghiP (0.07) were classified as risk-free (Fig. 4a). In most sampling locations, 2-ring and 3-ring PAHs concentrations were classed as moderate-risk, while high-ring PAHs were generally classified as risk-free (Fig. 4b). The total TEQ_{BaP} of soil PAHs ranged from 0.09 to 67.05 ng g^{-1}, with a mean value of 12.12 ng g^{-1}, 52.3% of the sampling sites fell within the low to moderate risk category, with 5-ring PAHs (BbF, BaP, BkF, and DBA) contributing most to total TEQ_{BaP} (Fig. 4c).

3.3 Health risks of main pollutants in WLFZs

According to the probability distributions, the hazard quotient (HQ) values for individual heavy metal elements for both adults and children were, in general, below the respective thresholds and so within safe ranges (Fig. 5a). The average HI values for adults and children were 9.66e-02 and 6.38e-01, respectively. With only 9.2% of HI values for children exceeding the critical value of 1, and none for adults. In addition, the analysis incorporating localized exposure parameters demonstrated that adult males generally exhibited higher HI values than females (Fig. S6), which is likely attributable to greater body weight and a larger dermal contact surface area. Overall, the non-carcinogenic risk caused by heavy metals in WLFZ soils was considered limited. Although the overall ecological risk was relatively low, previous research has shown that heavy metals in the soil of WLFZs can be released and redistributed through wet–dry cycles (Bao et al., 2015). As a result, these metals could potentially enter the food chain through adsorption, deposition, or diffusion. If such exposure recurs year after year, it may pose a long-term carcinogenic risk (Hu et al., 2020).

Carcinogenic risk values for the typical heavy metals considered in this study follow the descending order: As > Cd > Cr > Pb > Ni (Fig. 5b). Specifically, the average carcinogenic risk (CR) values for As in adults and children were 6.37e-06 and 1.42e-05, respectively, both exceeding the US EPA threshold of 1e-06 but below 1e-04, indicating that the carcinogenic risk was within an acceptable level. However, reliance on average values alone may obscure localized hotspots.



432
433 **Fig. 3. Geo-accumulation (I_{geo}) indices of heavy metals in soil of China's WLFZs. a.** Class distribution of I_{geo} value for eight heavy metals. **b.**
434 Geographical distribution of I_{geo} index for Cd. The donut chart represents the proportion of different I_{geo} index classes for Cd in the Three Gorges
435 Reservoir. **c.** Cumulative probability distribution of I_{geo} indices of eight heavy metals based on Monte Carlo simulations.

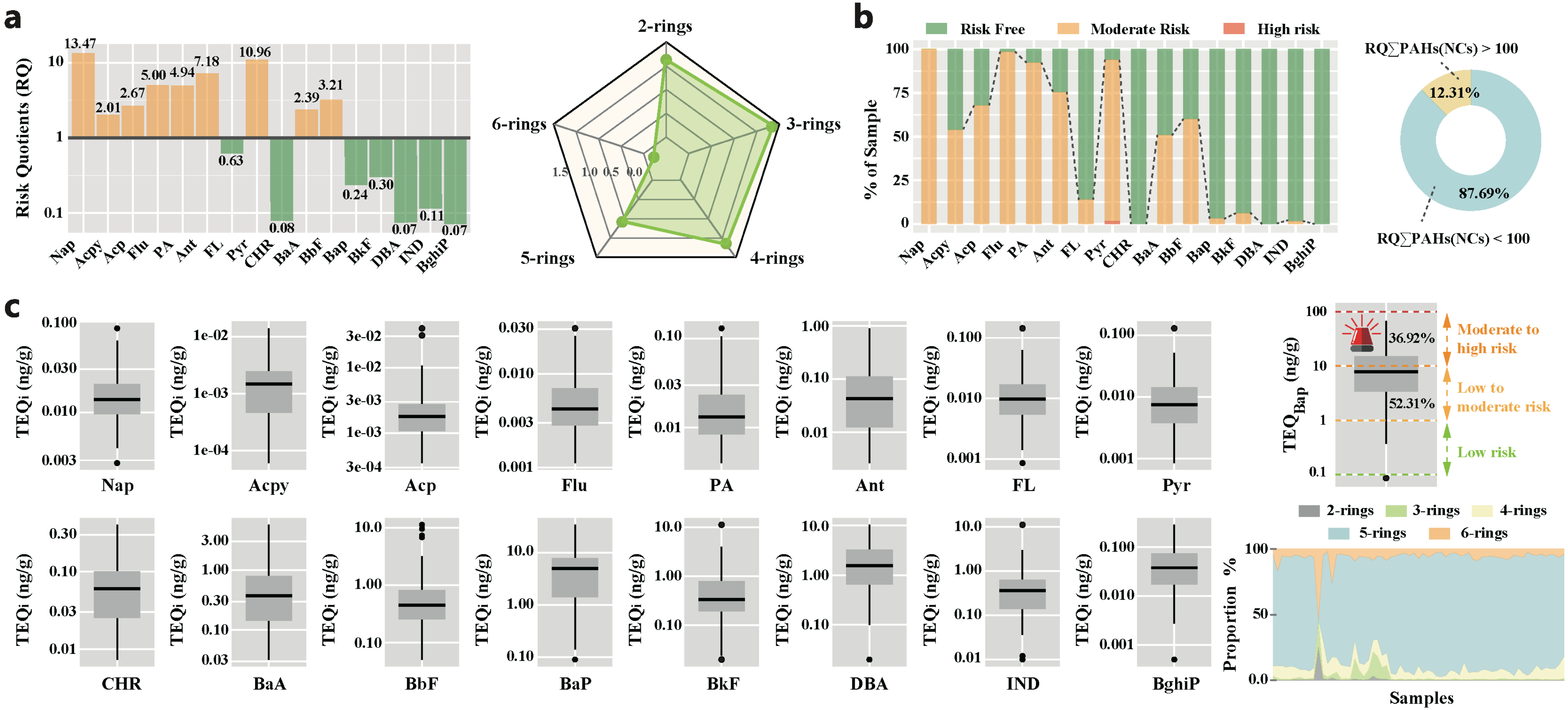


Fig. 4. Ecological risks of PAHs pollution in soil of China's WLFZs. a. Average risk quotients of 16 PAHs and radar chart indicating the log-transformed mean values of PAHs with different ring numbers. **b.** Ecological risk proportions of 16 PAHs in different samples and the ranges and proportions of total risk quotients. **c.** Toxic equivalents of PAHs and proportions of toxic equivalents for PAHs with different ring numbers.

Probabilistic risk analysis indicated that the probabilities of exceeding the threshold were 63.4% for children and 25.1% for adults, suggesting the presence of site-specific contamination sources that warrant further attention (Fig. 5b). Overall, As and Cd were the two heavy metals whose concentrations in WLFZ soils are currently a cause for concern. This finding is consistent with results from earlier studies on agricultural and industrial soils (Guan et al., 2022; Yan et al., 2022). The mobility and bioavailability of Cd and As make them more likely to enter the human body through the food chain, thereby increasing health risks (Huang et al., 2024; Lin et al., 2023). Previous studies suggest that As and Cd pollution in WLFZ soils may originate from fertilizer leaching from agricultural land and from industrial emissions (Ye et al., 2011; Zhang et al., 2021). Compared to adults, children are inherently more sensitive to heavy metal exposure due to their lower body weight and higher ingestion rates (e.g. frequent hand-to-mouth behaviors), which is reflected in the established risk assessment methodology used in this study (Huang et al., 2021; You et al., 2024). Although the overall concentrations of heavy metals in WLFZ soils were generally within acceptable limits, several sampling locations exhibited relatively high concentrations and potential health risks, particularly in the case of Cd and As. These elevated levels may reflect localized inputs from industrial activities, agricultural runoff, or historical pollution legacies, thereby emphasizing the need for site-specific monitoring and tailored remediation strategies (Hu et al., 2017). Therefore, future efforts should prioritize targeted assessment and management in higher-risk WLFZs or catchments with known anthropogenic pressures that are likely to elevate heavy metal pollution.

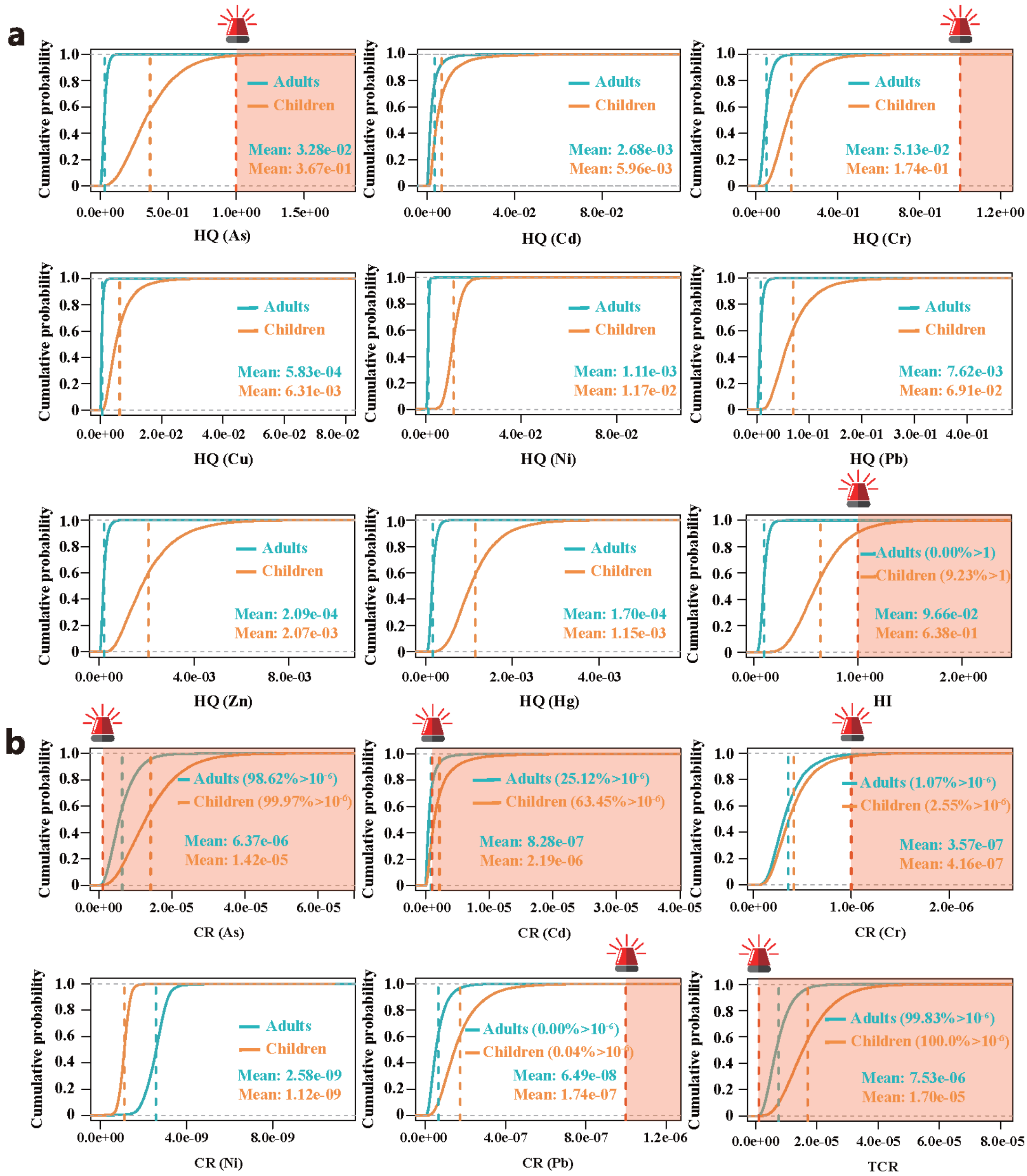


Fig. 5. Probability distribution of hazard quotient (HQ), hazard index (HI), carcinogenic risk (CR), and total carcinogenic risk (TCR) of soil heavy metals in China's WLFZs. a. Probability distribution and average values of HQ for As, Cd, Cr, Cu, Ni, Pb, Zn, Hg and HI for adults and children. **b.** Probability distribution and average values of CR for As, Cd, Cr, Ni, and Pb, and TCR for adults and children. **Notes:** The green and orange dashed lines represent the mean values for adults and children, respectively, while the red dashed vertical lines indicate the hazard quotient threshold (1) and the acceptable carcinogenic risk level (1.0e-06).

Non-carcinogenic risks caused by PAHs were limited in WLFZ soils. The mean HI was 2.24e-05 for adults and 3.00e-05 for children, with soil concentrations of PA, Pyr, and Nap being the main influencing factors (Fig. 6). Ingestion (averaging 70.4%) was the dominant exposure pathway for non-cancer risks, followed by inhalation (averaging 29.5%), for both adults and children. This finding is consistent with previous studies suggesting that, despite PAH contamination in the soil, its impact on non-cancer risk is limited (Wu et al., 2021). However, although PAH contamination in the soil of China's WLFZs has a limited impact on non-carcinogenic health risks, the carcinogenic properties of PAHs still pose a significant health risk with long-term exposure (Han et al., 2021; Hu et al.,2017). The average total carcinogenic risk (TCR) values were 2.93e-07 for adults and 7.91e-08 for children. Based on probabilistic risk analysis using Monte Carlo simulations, the probabilities of exceeding the threshold value of 1e-06 were 3.1% for adults and 0.1% for children, respectively. Although the carcinogenic risk for adults was slightly higher than that for children, the overall risk is well below the acceptable threshold, with dermal contact and ingestion being the primary exposure pathways, while direct inhalation was negligible (Fig. 6). PAHs in WLFZ soils may enter water bodies during the period of seasonal water level rise, primarily through processes such as surface runoff, desorption into floodwaters, and physical erosion of contaminated soils (Hu et al.,2017; Yuan et al., 2023). In addition, the wet–dry cycles characteristic of WLFZs can alter soil physicochemical properties, such as total organic carbon content and particle size distribution, thereby enhancing the mobilization of PAHs (Han et al., 2021). Specifically, variations in particle size distribution can influence the soil's surface area and porosity, thereby affecting the sorption and desorption of PAHs and, consequently, their concentrations. Similarly, the health risk assessment of PAHs indicated that adult males consistently had higher TCR values than females (Fig. S7), with BaP representing a carcinogenic risk of particular concern. Although overall PAH-related health risks in China's WLFZs appear to be limited, probabilistic risk assessment revealed non-negligible exceedance probabilities at specific locations, particularly for vulnerable populations such as children. Therefore, future research should focus on targeted risk-based management strategies aimed at reducing long-term human exposure in high-risk areas.

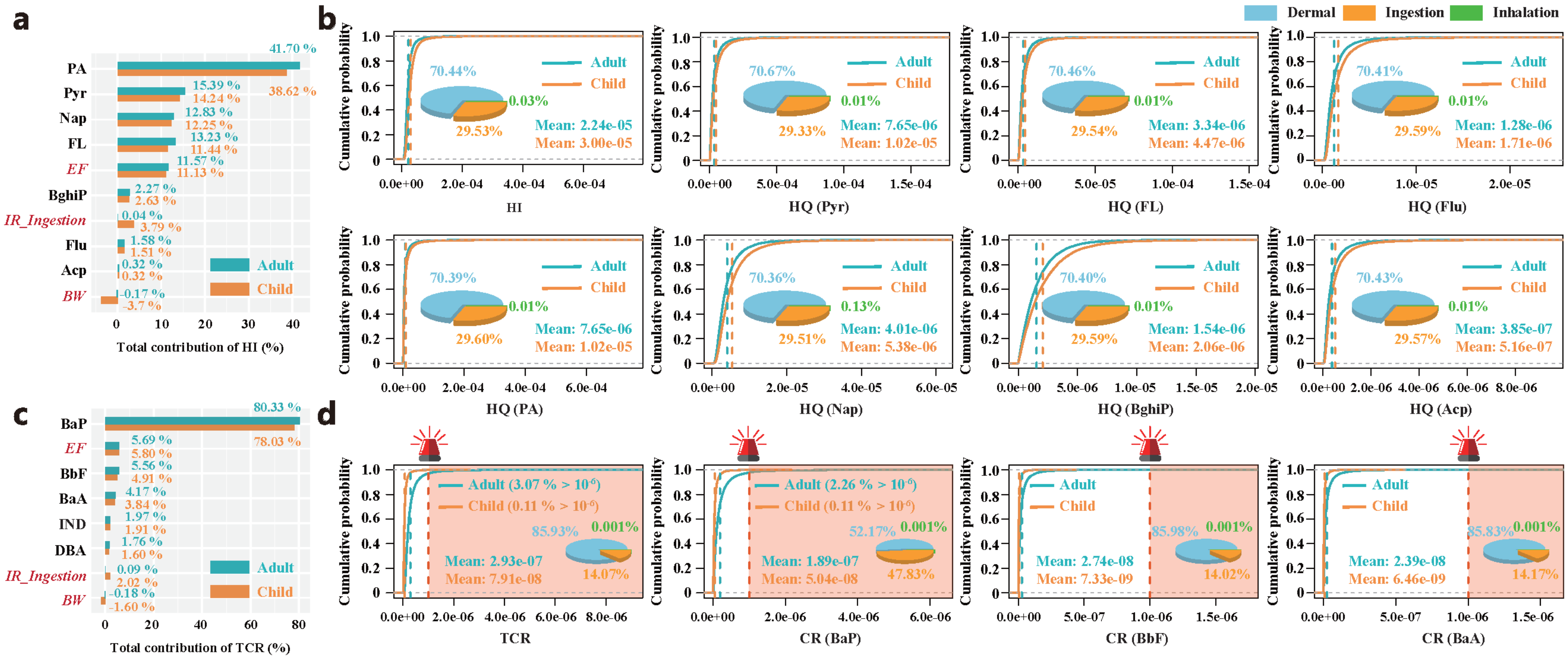


Fig. 6. Probability distribution of hazard quotient (HQ), hazard index (HI), carcinogenic risk (CR), and total carcinogenic risk (TCR) of PAHs in soil of China's WLFZs and risk sensitivity analysis of PAHs. a. Assessment of the sensitivity of relevant parameters to the non-carcinogenic risk of PAHs. **b.** Probability distribution and average values of HI and HQ. **c.** Sensitivity analysis of effective parameters in carcinogenic risk of PAHs. **d.** Probability distribution of TCR and CR. The pie charts represent the proportion of carcinogenic and non-carcinogenic risks associated with different exposure pathways (blue for dermal contact, orange for ingestion, and green for inhalation).

3.4 Source apportionment of main pollutants in the WLFZs

According to the results of the PMF model, the element As, which posed significant carcinogenic risk, was mainly associated with Factor 1 (40.2%), Factor 4 (53.4%), and Factor 6 (6.1%), while the element Cd, which contributed to ecological risk, was predominantly associated with Factor 3 (83.2%) and Factor 4 (10.8%) (Fig. 7). In addition, both Hg (54.2%) and Zn (14.0%) were associated with Factor 1, suggesting that this factor may be related to coal combustion and industrial activities (Cheng et al., 2023). Coal combustion has, for example, been shown to be a significant contributor to As and Hg pollution with studies demonstrating that As and trace amounts of Hg spread to surrounding soil and water bodies through smoke diffusion and particulate matter deposition (Hu et al., 2020; Liang et al., 2017; Pei et al., 2018). For example, previous research has demonstrated that widespread coal combustion in the regions surrounding the TGR contributes to the accumulation of heavy metals in the environment, with the TGR WLFZ serving as a representative area of other sites impacted by similar contamination (Liu et al., 2018). This may account for the elevated concentrations observed in several of the reviewed studies that focused on this region (Liu et al., 2018). The strong association between Cd and Factor 3 suggests that agricultural activities could be a potential source (Zhou et al., 2024). Moreover, Cd contamination in soils is widely recognized as being primarily linked to the long-term use of fertilizers and pesticides, particularly in intensively farmed regions of China as well as other countries (Dong et al., 2023). These source apportionment results are consistent with the environmental characteristics of WLFZs in China, as previous studies have also shown that WLFZs in many river basins are subjected to overlapping pressures from industrial emissions and agricultural activities, which jointly promote the pronounced accumulation and enrichment of pollutants in these areas (Hu et al., 2020; Ye et al., 2011). Importantly, concentrations of heavy metals in the soils of China's WLFZs have exhibited a discernible declining trend in recent years (Fig. S4), which is plausibly attributable to the cumulative impact of successive environmental protection policies such as the "Water Ten Plan" (issued on April 2, 2015), "Soil Ten Plan" (issued on May 31, 2016), and "Environmental Protection Law of the People's Republic of China" (officially implemented from January 1, 2015), which were designed to curb emissions from coal combustion, industrial operations, and agricultural practices (Huang et al., 2024).

Low-ring PAHs that pose ecological risks, including Nap (48.9%), Acp (33.7%), Flu (63.4%), and PA (32.2%), were significantly associated with Factor 1, which was possibly caused by oil spills or the incomplete combustion of gasoline (Fig. 7). Specifically, Nap is a marker of petroleum pollution, and it has been suggested that in some Chinese cases PAHs in WLFZ soils may originate from oil leaks during ship navigation (Chen et al., 2013). Additionally, some chemical enterprises located in areas adjacent to WLFZs may be sources of considerable petroleum-derived pollutants, as previously reported in the TGR region (Hu et al., 2017). High-ring PAHs with significant carcinogenic risk, including BaP (64.5%), BbF (39.3%), and BkF (41.5%), were more significantly associated with Factor 3, which again may be related to combustion processes. BaP is produced during the incomplete combustion of organic matter under high-temperature conditions (Wang et al., 2023). This inference can be evidenced by isomer ratio analysis, which indicated that the combustion of grass, wood, and coal is an important source of PAHs (Fig. S5). Given the highly carcinogenic nature of PAHs, it is essential to enhance environmental regulation and pollution control not only within WLFZs but also across adjacent catchment areas, where primary pollutant sources may be located with pollutants being subsequently transported into the WLFZs.

Moreover, optimizing energy utilization and minimizing incomplete combustion are widely recommended as measures to mitigate high-ring PAH pollution from combustion in general that could also contribute to tackling pollution within WLFZs.

Furthermore, based on the source apportionment results, the management of soil pollution in Chinese WLFZs should be oriented toward region specific and source differentiated control strategies. In regions where industrial emissions and coal combustion are major contributors to soil contamination, it is essential to strengthen the monitoring of external pollutant inputs and to effectively implement emission reduction and permitting requirements for relevant industries, while areas influenced mainly by agricultural inputs should prioritize optimizing fertilizer and pesticide application and improving farmland management practices to reduce pollutant transport (Pei et al., 2018; Ye et al., 2011). The sustained implementation of these measures will play a critical role in reducing long-term pollutant loads in WLFZs and in safeguarding their ecological integrity (Bao et al., 2015).

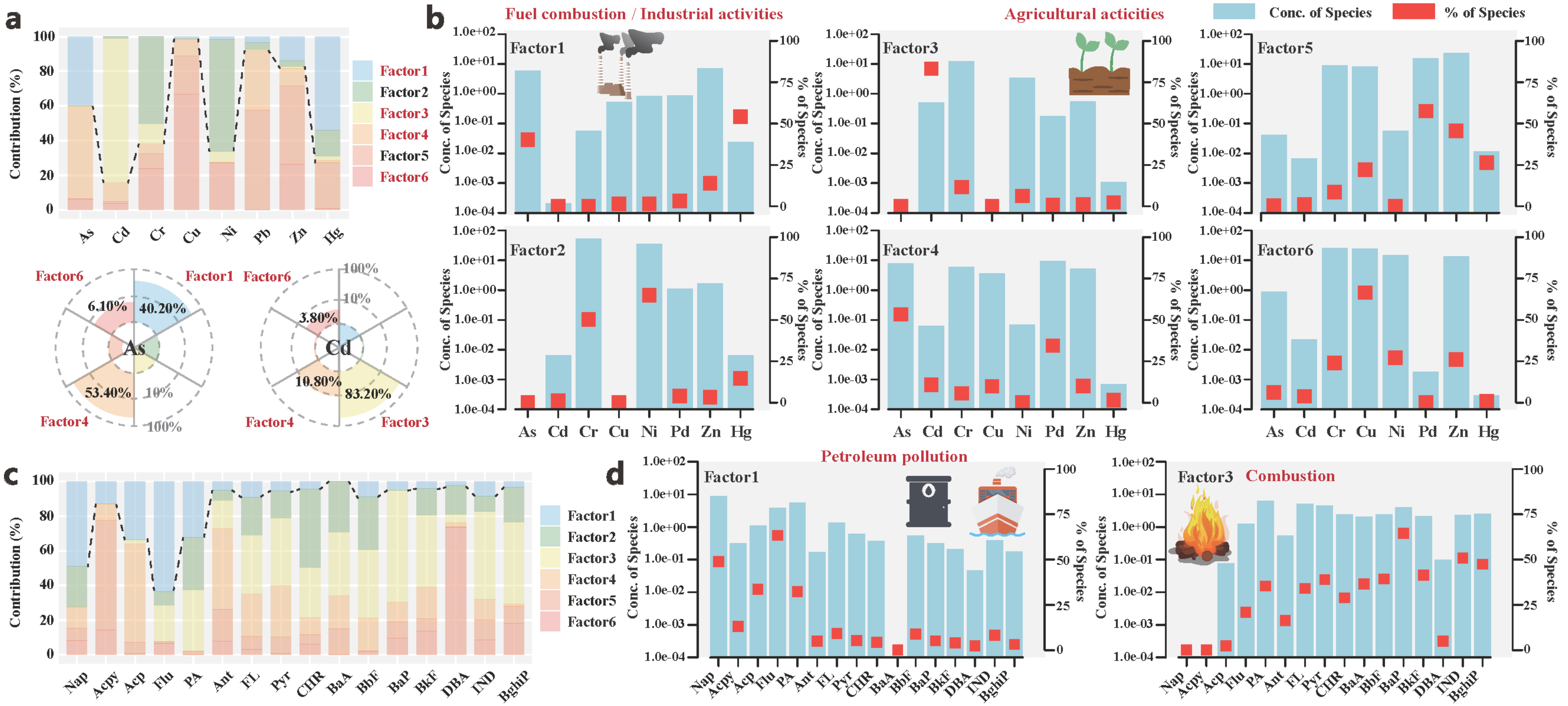


Fig. 7. Source apportionment and contribution analysis of heavy metals and PAHs pollution in soils of China's WLFZ using the positive matrix factorization (PMF) model. a. Proportion of heavy metals in different sources (factors). **b.** Source profiles (concentration and proportion of species) for heavy metals. **c.** Proportion of PAHs in different sources. **d.** Contribution and distribution of two factors (factors 1 and 3) to PAHs.

3.5 Characteristics of emerging pollutants in China's WLFZs

Emerging pollutants are contaminants that have not been traditionally monitored or regulated but may pose significant risks to the environment and human health (Li et al., 2022; Zhang et al., 2019; Zhang et al., 2022). Among them, emerging pollutants of particular concern in Chinese WLFZ soils include MPs, phthalate esters (PAEs) and PFASs (Fig. 2B, Table S16). The sources of emerging pollutants in WLFZ soils are diverse and complex, likely originating from agricultural practices (e.g. mulching and fertilizer application), industrial and municipal wastewater discharges, ship operations, vehicular emissions from road traffic, and atmospheric deposition of airborne contaminants derived from combustion processes and industrial activities (Zhang et al., 2022). Although emerging pollutants share common sources and transport pathways with conventional contaminants, such as emissions from local activities and long-range transport via water and atmospheric currents, their unique environmental behaviors, persistence, and associated ecological risks in WLFZs warrant increased attention (Fig. 8). The concentrations of emerging pollutants in WLFZ soils exhibit notable variations during water level fluctuations, and WLFZs can act as both sources and sinks for these pollutants (Table S16). However, research on the concentrations, environmental forms and transformation behaviors of these emerging pollutants in WLFZ soils remains limited. Existing studies also vary considerably in monitoring targets, analytical methods, spatial coverage and data completeness (Li et al., 2022; Zhang et al., 2019). This restricts robust studies on their ecological and health risks as well as source apportionment. Additionally, the lack of standardized analytical methods to measure such pollutants makes it difficult to compare results from different studies (Zhang et al., 2019). Taken together, these limitations indicate that current research on emerging pollutants in WLFZ soils remains insufficiently comprehensive and requires further development. In addition, targeted and methodologically consistent investigations will be essential for establishing a robust foundation for the risk assessment and source apportionment of emerging pollutants. Moreover, source-specific mitigation measures should be implemented according to the different sources of emerging pollutants, including stricter control of agricultural non-point inputs, improved industrial and municipal wastewater treatment, and stronger regulation of atmospheric emissions to reduce external pollutant loads (Zhang et al., 2022).

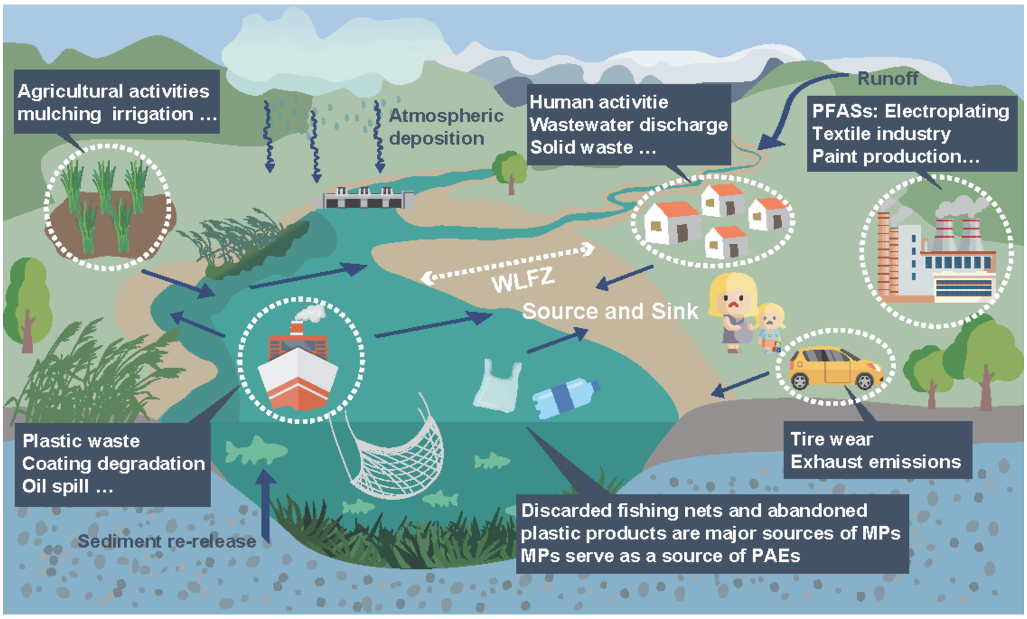


Fig. 8. Schematic diagram of emerging pollutant sources in WLFZ soils

3.6 Implications and limitations

Although this study conducted a detailed analysis of soil pollution in China's WLFZs based on extensive published literature, employing a "front page filtering" script alongside a robust step-by-step review to ensure literature quality and minimize publication bias, certain limitations remain. The data in this study were derived from many published papers, and differences in sampling designs, extraction procedures, and analytical methods across studies (Table S2) may affect the consistency and comparability of the results. In addition, since current Chinese risk screening standards and most previous studies are established on total concentrations (Table S2), this study also relied on total concentrations to estimate human health risks. However, it should be noted that total concentrations do not fully reflect the biologically effective fractions that drive toxic responses, and may overestimate the associated health risks (He et al., 2019; Yan et al., 2022). Nevertheless, we believe that this bias does not compromise the main findings of this study. This work will enhance understanding of soil pollution status and human health risks in China's WLFZ, identify priority pollutants for control, and provide critical information for the management of these fragile and ecologically sensitive areas, serving as a valuable reference for future research and policy-making.

Overall, this study identified multiple pollution hotspots, such as TGR, Lancang river, and Danjiangkou reservoir, along with key contaminants within China's WLFZs, offering a critical foundation for elucidating spatial pollution patterns and associated environmental risks. These findings underscore the need to prioritize high-risk areas in future monitoring and regulatory initiatives and emphasize the importance of formulating targeted, region-specific pollution mitigation strategies to protect ecosystem health and ensure public safety. In addition, it is important to note that current research on the occurrence and environmental behavior of emerging pollutants in WLFZ soils remains limited, which constrains comprehensive ecological and health risk assessments. Future investigations should strengthen efforts in this area to support the development of science-based environmental policies and management frameworks.

4. Conclusions

In this study, a "front page filtering" script and subsequent detailed review of published papers was employed to establish a dataset of soil pollutants in China's WLFZs that included data from 2,959 sampling locations in 353 WLFZs. The dataset included the Three Gorges Reservoir, Danjiangkou Reservoir, the Yangtze, Lancang and Liao rivers, Taihu Lake and Poyang Lake with sampling sites representing the major WLFZ across China. Heavy metals were the main pollutants of concern, with Cd (0.80 ± 1.15 (mg kg^{-1}) being the element responsible for the most severe contamination. Simulations based on the Monte Carlo method indicated that the probability of Cd reaching moderate pollution levels was 68.5%, whereas the equivalent probabilities for As, Pb, Cu and Ni were in the range 5-10%, and those for Zn, Cr and Hg were below 5%. Non-carcinogenic risks associated with heavy metals in WLFZ soils were limited. Notably, the average carcinogenic risk indices for children for As (1.42e-05) and Cd (2.19e-06) were relatively high and should be of concern. Some PAH pollution was identified in some WLFZ soils, with Nap and Pyr having the highest risk quotients, while BaP was the primary carcinogenic pollutant. The main exposure pathways were dermal contact and ingestion, while direct inhalation could be neglected. According to the results of the PMF model, coal combustion, industrial and agricultural activities, and ship navigation are the likely major sources of pollution. Although site-specific longitudinal data were unavailable, a comparative analysis of published studies from broadly similar sampling locations suggests that concentrations of major pollutants in WLFZ soils were generally lower after 2015 compared to the 2010–2015 period, potentially reflecting the effects of strengthened environmental governance measures. Future research should further explore the environmental behavior of emerging pollutants in WLFZs and conduct comprehensive risk assessments of these pollutants for both ecosystems and human health. This study contributes to the understanding of the ecological and human health risks of soil pollutants in China's WLFZs and will provide important guidance for the formulation of future environmental policies for these ecologically sensitive areas.

CRediT authorship contribution statement

Kang Yan: Writing – review & editing, Writing – original draft, Investigation, Methodology. **Weiming Li:** Writing – review & editing, Funding acquisition, Validation, Conceptualization. **Hong Yang:** Writing – review & editing, Validation, Supervision, Methodology. **Ying Xi:** Methodology, Investigation. **Yixian Tan:** Methodology. **Ying Teng:** Writing – review & editing, Supervision. **Haizhen Wang:** Methodology, Investigation. **Julian R. Thompson:** Writing – review & editing.

Declaration of competing interest

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

Data availability

Supporting data are provided in Tables S2–S4 (Supporting Information).

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