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Anthropogenic impacts on polycyclic aromatic hydrocarbons in surface water: Evidence from the COVID-19 lockdown

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KEYWORDS

Polycyclic Aromatic Hydrocarbons; Human Activities; long-term; lockdown; Source
Apportionment; Risk Assessment

ABSTRACT

The lockdown restrictions against coronavirus disease 2019 (COVID-19) have led to unprecedented reductions in global anthropogenic activities. Polycyclic aromatic hydrocarbons (PAHs) are carcinogenic combustion-induced pollutants, but the influence of anthropogenic responses to COVID-19 on PAH contamination remains largely unknown. Here we systematically investigated the spatial and temporal distributions of 16PAH concentrations dissolved in the water phase and absorbed on the suspended particulate matter (SPM) in the Elbe River from 2015 to 2021 and quantified the impacts of lockdown restrictions on PAH pollution in receiving water. Results show that the annual average PAH concentrations in water and SPM were determined as $0.055 \mu\text{g}\cdot\text{L}^{-1}$ and $3.77 \text{ mg}\cdot\text{kg}^{-1}$ from 2015 to 2021, respectively. Pronounced declines in PAH on SPM (up to -18%) were observed during the three lockdowns in Germany from 2020 to 2021. However, dramatic rebounds of anthropogenic activities during the removal of the lockdown led to increases (up to 29%) in $\sum_{16}\text{PAH}$ concentrations compared to the same period in previous years. Through the source apportionment method, vehicle and coal emissions were recognized as the two most predominant sources of PAHs in the river, accounting for 60.9% and 27.9% of the concentrations, respectively. Health risks for three age groups were assessed as potential low risk and decreased by 18% from 1.54×10^{-4} in 2015 to 1.27×10^{-4} in 2019, and rebounded to 1.40×10^{-4} in 2020-2021. The findings of this study highlight the strong consistency between PAH concentrations and anthropogenic intensity, implying that source control from improved cleaner production is an effective pathway for mitigating PAH contamination in the aquatic environment.

1. INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs), a hazardous class of persistent organic compounds with two or more benzene rings, have been a global concern due to their serious risks (Lv et al. 2020, Wang et al. 2020a, Zeichner et al. 2023). These substances are recognized as highly lipophilic, teratogenic, mutagenic, and carcinogenic, and have the potential to cause severe harm to human health and the ecosystem even at low exposure levels (Wang et al. 2021). In 2015, it was estimated that 6.9×10^4 people worldwide could potentially develop lung cancer due to inhalation of PAHs (Li et al. 2022b). Additionally, the accumulation of PAHs in the aquatic system has damaging effects on fish, marine invertebrates, and amphibians (Man et al. 2020). High PAH concentrations in marine ecosystems have been linked to increased mortality rates in coral reef fishes (Johansen et al. 2017). Due to their widespread presence and the associated hazard, certain PAHs are designated as prior contaminants by the European Water Framework Directive and the United States Environmental Protection Agency (US EPA). Furthermore, the International Agency for Research on Cancer classified some PAHs as Group 1 and Group 2B carcinogens (Naudin et al. 2019).

Owing to their steady chemical and physical characteristics, PAHs are ubiquitous environmental contaminants distributed in virtually all environmental media, including air (Chakraborty et al. 2021), soil (Zheng et al. 2023), water (Chen et al. 2023), and sediments (Cao et al. 2024). With regard to environmental fate, surface water serves as a major reservoir for PAHs in different media (Gan et al. 2009). PAHs reach receiving rivers via various paths, such as urban surface runoff, residential and industrial wastewater, and atmospheric deposition (Goswami et al. 2018, Romero et al. 2018). In 2010, anthropogenic activities in Germany were reported to release 191.5 tons of PAHs into the atmosphere (Umweltbundesamt 2016) and were largely responsible for PAH contamination in the aquatic environment (Fuchs et al. 2010). Moreover, suspended particulate matter (SPM) may serve as an essential medium of PAHs in aquatic systems due to their low solubility and high resistance to degradation by anoxic bacteria (Sun et al. 2009). The transport carrier of SPM allows PAHs to be spread over large areas in

waters, thereby increasing the extent of contamination and the risk to human health and the surrounding environment (He et al. 2021). Thus, it is critical to characterize the long-term PAH pollution in water and SPM in order to develop effective source-orientated mitigation strategies and improve risk assessment.

More than 220 countries or regions around the world were affected by the unprecedented international pandemic of coronavirus disease 2019 (COVID-19) (Frey et al. 2024, Pierce et al. 2022). Based on the WHO COVID-19 Dashboard, approximately 199.5 million people have been infected with coronaviruses globally, resulting in more than 4.2 million deaths by August 2021 (Liu et al. 2022). To curb the spread of the new virus, over 170 countries have imposed government-mandated lockdown restrictions, including the closure of international borders, cessation of non-essential industries and businesses, and implementation of online education (Sekiya et al. 2023). The lockdown restrictions not only disrupted the transmission path of the virus but also brought economic activities related to transportation and mobility to a near-complete stagnation in many countries. As a result, the pandemic led to a global recession in 2020 with an economic growth rate of -4.9% (Liu et al. 2022). The sudden reduction in anthropogenic activities improved the environmental quality during the lockdown (He et al. 2020). Many previous studies focused on the influences of COVID-19 on the common atmospheric emissions. For instance, global CO₂ emissions decreased sharply by 8.8% (-1,551 Mt) in the first half of 2020 compared to the same period in 2019 (Liu et al. 2020). The population-weighted concentrations of surface-level fine particulate matter (PM_{2.5}) and nitrogen dioxide (NO₂) were reported to decline by 31% and 60% in the lockdown period of 34 countries (Venter et al. 2020). As the undesired side-products of incomplete combustion, PAHs are one of the most toxic atmospheric pollutants and are mainly released from human-made sources, such as coal combustion (Sofowote et al. 2008, Xu et al. 2020), and vehicular emissions (Zhang et al. 2012, Zheng et al. 2018). However, the influence of lockdown on the PAH pollution remains largely unknown. Besides, Liu et al. (2022) found that river water quality in China was also improved during the lockdown period, with significant

decreases in concentrations of ammonia nitrogen, chemical oxygen demand, and dissolved oxygen. Since receiving water acts as the main sink of PAHs, it is essential to comprehensively assess the impacts of anthropogenic changes during the lockdown period on PAHs in the aquatic system. In addition, the long-term monitoring data in the surface water provides the opportunity to compare the historical PAH concentrations with those in the lockdown and to investigate the changes in the source contributions of pollutants during COVID-19, which can reveal the significant impacts of anthropogenic activities on PAH pollution.

As one of the longest waters in Europe, the Elbe River represents the dynamic environments where nature and anthropogenic activities unfold a complex interaction. The catchment area is inhabited by approximately 18.5 million people, who consume aquatic products from the river and its water for drinking supply and recreational swimming (Zhu et al. 2024). Due to the intense human contact with river water, prevention of water pollution, especially PAH pollution, is of great importance for the protection of human health. Germany also imposed lockdown restrictions several times during the epidemic, leading to a sharp decline in atmospheric pollution (Balamurugan et al. 2022). In addition to the socioeconomic consequences of the global measures to COVID-19, there is an unprecedented opportunity to evaluate the short-term impacts of economic activities contrary to "business as usual" on PAH contamination in receiving waters and their relationship to human health. The major aims of this study were to (1) characterize the spatial and temporal distributions of PAH concentrations in water and SPM in the Elbe River from 2015 to 2021; (2) quantify the impacts of lockdown restrictions on PAH pollution in receiving water; (3) determine the potential sources of PAHs in each year by using source apportionment method; and (4) estimate the long-term human health risks from PAH exposure. The findings presented in this study will contribute to policymakers' insights into the anthropogenic impacts of PAHs in rivers and optimize mitigation strategies on a regional and global scale.

2. METHODS AND MATERIALS

2.1 Data sources

In order to implement the European Water Framework Directive (Directive 2000), several sampling stations were set up along the Elbe River to measure hundreds of contaminants for continuously assessing water quality. From January 2015 to December 2021, 1540 samples in total were collected from different stations to monitor the concentrations of PAHs dissolved in the water and absorbed on the SPM in the Elbe River (**Fig. 1a**). These monitoring stations cover the main water flow pathways and populated areas; therefore, the information is representative of the contamination extents and health hazards of PAHs along the Elbe River in Germany. As shown in **Table S1**, 16 priority PAHs, namely Benzo(*a*)pyrene (BaP), naphthalene (NAP), anthracene (ANT), benzo[*b*]fluoranthene (BbF), phenanthrene (PHE), fluorene (FLU), dibenzo[*a,h*]anthracene (DBA), acenaphthylene (ACY), indeno[1,2,3-*cd*]pyrene (IDP), fluoranthene (FLUH), benzo[*a*]anthracene (BaA), acenaphthene (ACE), chrysene (CHR), pyrene (PYR), benzo[*k*]fluoranthene (BkF), and benzo[*g,h,i*]perylene (BgP), are monitored and obtained from the Elbe Data Information System (FIS) (www.fgg-elbe.de, accessed Oct. 2023). $\Sigma_{16}\text{PAHs}$ is defined as the summation of sixteen individual PAH concentrations.

On the basis of the Elbe River basin community, the frequency of water sample collection is monthly. According to the German DIN 38402-24 standard (A24), SPM (less than 2 mm) samples are primarily taken from the filtration residues of settling containers or by centrifuges. The sample remains stored in the settling tank for about one month, and then it is sampled and analyzed for PAHs on a monthly interval. This ensures that the average load of PAHs at a given location is uniformly detected over a period of time. The samples were delivered to the Staatliche Betriebsgesellschaft für Umwelt und Landwirtschaft (BfUL, the laboratory of the Public Operating Company for Environment and Agriculture) for conducting chemical assessments. Gas chromatography–mass spectrometry (GC–MS) and high-performance liquid chromatography (HPLC) were adopted to perform PAHs analysis following the methods of EPA 610 and German standards DIN 38407-39 (F39), DIN EN ISO 17993

(F18), DIN 38414-21 (S21), and DIN ISO 13877. Details about the sample are presented in **Text S1**.

2.2 Positive Matrix Factorization Model

The Positive Matrix Factorization Model (PMF) receptor model was employed in this study to pinpoint the potential sources of PAH contamination and quantify the contributions of individual sources on the basis of pollution fingerprints (Li et al. 2023, Qi et al. 2020, Yu et al. 2019). Some source apportionment methods, such as diagnostic ratios, principal component analysis with multiple linear regression, and the Unmix receptor model, have limitations in dealing with noisy data and uncertainty weighting, and negative factor loadings are difficult to explain in terms of positively determined parameters (e.g., concentrations) Wu et al. (2019). The advantage of the PMF model over other source apportionment models is that it exploits non-negative constraints and has improved performance in generating source profiles and contributions by weighing individual values of user-supplied uncertainty. The multivariate model decouples the input data matrix into factor profiles (f) and source contributions (g) to address the equation:

$$x_{ij} = \sum_{k=1}^p g_{ik} f_{kj} + e_{ij} \quad \text{Eq. 1}$$

In this chemical mass balance equation, x_{ij} refers to the PAH concentration of species j detected in the i^{th} sample. g_{ik} represents the k^{th} factor's contribution to the i^{th} sample, and f_{kj} refers to the PAH concentration of species j in the source profile of the k^{th} factor. p represents the given number of sources, and e_{ij} refers to the residues of species j detected in the i^{th} sample. To achieve the most appropriate solution, the PMF model minimizes the objective function Q by modifying g_{ik} and f_{kj} for a defined number of factors, as the following equation:

$$Q = \sum_{i=1}^n \sum_{j=1}^m \left(\frac{x_{ij} - \sum_{k=1}^p g_{ik} f_{kj}}{u_{ij}} \right)^2 = \sum_{i=1}^n \sum_{j=1}^m \left(\frac{e_{ij}}{u_{ij}} \right)^2 \quad \text{Eq. 2}$$

where u_{ij} represents the concentration uncertainty of the species j in the i^{th} sample, while m and n refer to the number of species and samples, respectively.

In the PMF model published by the US EPA (version 5.0, www.epa.gov), three methods of error estimation were performed to identify the most optimal number of factors, which are the displacement (DISP), bootstrap-displacement (BS-DISP), and bootstrap (BS) methods. When the following criteria for three methods are met, PMF results could be considered valid, i.e. 1) All mapping factors for the BS method are greater than 80; 2) No swaps were found in the first row ($dQ_{\max} = 4$) and the rate of decrease in Q for the DISP method was less than 1.0%; and 3) The BS-DISP method has a decrease in Q -value of less than 0.5 %. The results of the PMF are highly dependent on the scale of the data, and this study combined 7 years of data from each monitoring station to determine the source of PAH contamination over the study period. Detailed information about the source apportionment can be found in **Text S2**.

2.3 Health risk assessment

The toxicity equivalence factor (TEF) method for human health risk assessment has been commonly utilized to convert all individual PAH concentrations to equivalent concentrations of benzo[a]pyrene (Gbeddy et al. 2020, Zhu et al. 2019). With this approach, each PAH compound's toxicity can be represented corresponding to the toxicity of BaP. The toxic BaP equivalent concentration of all PAHs (C_{TEQ}) is generated as follows:

$$C_{TEQ} = \sum TEF_i \times C_i \quad \text{Eq. 3}$$

where TEF_i refers to the transforming factor of the i^{th} PAH species (**Table S2**), C_i represents the i^{th} PAH concentration ($\text{mg} \cdot \text{cm}^{-3}$).

The incremental lifetime cancer risk (ILCR) assessments have been commonly utilized to evaluate the PAH exposure risks in different environmental media due to the carcinogenic properties of PAHs (Han et al. 2019, Rajasekhar et al. 2018). Since receiving water is available as an important source of drinking supply and daily use,

oral and dermal exposure are regarded as the two exposure routes of the human body. To assess the human uptake of PAHs, the chronic daily intake (CDI) is generated with the following equations (Sarria-Villa et al. 2016b, USEPA 2001):

$$CDI_{dermal} = \frac{DAD \times SA \times ED \times EF \times EV}{BW \times AT} \quad \text{Eq. 4}$$

$$DAD = 2 \times FA \times K_P \times C_{TEQ} \times \sqrt{\frac{6 \times t_{event} \times \tau_{event}}{\pi}} \quad \text{Eq. 5}$$

$$CDI_{oral} = \frac{C_{TEQ} \times CF \times IR \times ED \times EF}{BW \times AT} \quad \text{Eq. 6}$$

where CF refers to the concentration conversion factor ($\text{cm}^3 \cdot \text{L}^{-1}$), C_{TEQ} has the unit of $\text{mg} \cdot \text{cm}^{-3}$, EF refers to the exposure frequency ($\text{d} \cdot \text{y}^{-1}$), SA represents the surface area of skin (cm^2), τ_{event} refers to the lag time per event ($\text{h} \cdot \text{event}^{-1}$), EV refers to the event frequency ($\text{event} \cdot \text{d}^{-1}$), t_{event} represents event duration ($\text{h} \cdot \text{event}^{-1}$), K_P refers to the dermal permeability coefficient, FA represents the fraction of absorbed water, BW represents body weight (kg), ED is the exposure duration (y), AT refers to the average time (d), and IR represents the ingestion rate of water ($\text{L} \cdot \text{d}^{-1}$). Values of BW were derived from the statistical data of Germany (www.gbe-bund.de, accessed on Oct. 2023). ILCR from oral ingestion and dermal contact are generated as:

$$ILCR_{dermal} = CDI_{dermal} \times \frac{SF_O}{ABS_GI} \quad \text{Eq. 7}$$

$$ILCR_{oral} = CDI_{oral} \times SF_O \quad \text{Eq. 8}$$

where ABS_GI refers to the gastrointestinal absorption factor and SF_O represents the slope factor of oral exposure ($\text{mg} \cdot \text{kg}^{-1} \cdot \text{d}^{-1}$), which is derived from the integrated risk information system (IRIS) database (USEPA 1991a, 2008). The health assessment for PAHs in SPM is similar and takes the concentrations of SPM in the water of each station into consideration. Health risk assessments were conducted for adults (21-70 years), teenagers (11-20 years), and children (1-10 years) because of the different physical and

living conditions of different age groups. The parameter values classified into different groups are shown in **Table S3**.

2.4 Uncertainty analysis

It is necessary to perform an uncertainty analysis to quantify the range of health risks from PAHs because the input parameters for the health risk assessment (e.g., *SA*, *ED*, *BW*, *IR*, and toxic BaP equivalent concentrations) are non-constant. The uncertainty ranges of the variables are presented in **Table S3**. In this research, 10^5 combinations of variables were randomly selected for uncertainty analysis using the Latin Hypercube Sampling Monte Carlo Integration method. Mann-Kendall trend test is applied to determine the trend in long-term PAH concentrations in each station (**Text S3**).

3. RESULTS

3.1 Spatiotemporal distributions of PAHs in water and SPM

Fig. 1b-c shows the average $\sum_{16}\text{PAHs}$ in water and SPM measured in different stations along the Elbe River from 2015 to 2021. In the aqueous phase, average $\sum_{16}\text{PAHs}$ concentrations ranged from 0.037 to 0.1 $\mu\text{g}\cdot\text{L}^{-1}$ with less variation between W1-W11 sites (**Fig. 1b**). In contrast, average $\sum_{16}\text{PAHs}$ concentrations absorbed on SPM were found to have a large spatial difference between upstream and downstream (**Table S4**). Explicitly, average upstream concentrations at S1-S7, which ranged from 3.9 to 5.85 $\text{mg}\cdot\text{kg}^{-1}$, were much higher than those detected at downstream of S8-S10 (0.29 - 1.57 $\text{mg}\cdot\text{kg}^{-1}$) (**Fig. 1c**). This phenomenon might be related to the larger loads of SPM in downstream. In the Hamburg harbor at downstream, PAHs were adsorbed on a large number of SPMs on average, leading to a decrease in concentrations (**Fig. S1**).

Fig. 2a-b shows the composition of 16PAHs in water and SPM of the Elbe River from 2015 to 2021, which is classified by 2-6 benzene-member rings. Both PAHs in water and SPM were dominated by 4-rings PAHs, accounting for 37.8% and 47.8%, respectively. Furthermore, the percentage of PAHs with high molecular weight (4-6

rings) in SPM (82.4%) was higher than that in water (68.7%). Since the carcinogenicity of PAHs increases with molecular weight, a higher predominance of high benzene-rings PAHs in SPM may result in a greater potential carcinogenic risk than PAHs in water.

The temporal changes in annual $\sum_{16}\text{PAH}$ concentrations in water and SPM of the Elbe River are presented in **Fig. 2c-d**. The annual average concentrations of $\sum_{16}\text{PAHs}$ in water and SPM were determined as $0.055 \mu\text{g}\cdot\text{L}^{-1}$ and $3.77 \text{ mg}\cdot\text{kg}^{-1}$ from 2015 to 2021, respectively (**Table S5**). Based on the Mann-Kendall trend detection, $\sum_{16}\text{PAH}$ concentrations in two phases of the whole Elbe River remained essentially constant with no significant trend. However, there were downward trends determined in several stations during the period (**Table S6, Figs. S2-3**). For instance, $\sum_{16}\text{PAH}$ concentrations in W7 and S6 decreased from 2015 to 2021 with a slope of 0.0003 and 0.017, respectively (**Fig. 2e-f**). Moreover, at the beginning of the COVID period, there were great declines in the concentrations of $\sum_{16}\text{PAHs}$ in both water and SPM, which could be attributed to seasonal variations in natural factors and a virtual stagnation of anthropogenic activities during the lockdown.

3.2 Impacts of lockdown restrictions on PAH pollution

Fig. 3a shows the monthly comparison of average $\sum_{16}\text{PAH}$ concentrations on the SPM of Elbe River in each station. For most stations, lower PAH concentrations on SPM occurred between April to August (hot season), while higher pollution levels appeared between October to Mar (cold season). Explicitly, average concentrations during cold season ($4.45 \text{ mg}\cdot\text{kg}^{-1}$) were much higher than those at the hot season ($3.81 \text{ mg}\cdot\text{kg}^{-1}$). Similar seasonal changes in PAHs have also been recognized in other rivers (Bayowa and Agbozu 2016, Cetin et al. 2018, Goudarzi et al. 2017). This phenomenon is probably attributed to the increased consumption of fossil fuels (biomass and coal) for residential heating in cold seasons, resulting in more emissions of PAHs into the environment (Kural et al. 2018).

In order to estimate the impacts of the lockdown on PAH pollution in the river system, the gap between the concentrations in the COVID period (2020-2021) and the average values of 2015-2019 is generated in **Fig. 3b**. In March 2020, due to the sudden

outbreak of the epidemic, Germany imposed its first lockdown, which lasted until the end of May. The gap between $\sum_{16}\text{PAH}$ concentrations on SPM in April 2020 and the average values of 2015-2019 was $-0.48 \text{ mg}\cdot\text{kg}^{-1}$. In November 2020, the Delta strain was discovered and began spreading in Germany, which initiated a second round of lockdowns that lasted until April 2021. All PAH concentrations on SPM during the second lockdown (2020.11-2021.4) were lower than those in the same months of previous years. Explicitly, the largest decrease in concentrations occurred in March 2021, which accounted for 18% of the previous value. The third lockdown occurred in November 2021, during which time a decline in concentrations was also observed. However, during the removal of the lockdown, $\sum_{16}\text{PAH}$ concentrations showed a substantial increase compared to the same period in previous years. For instance, PAH concentrations on SPM measured in 2021.7-2021.9 showed an average increase of 23.3% ($0.77 \text{ mg}\cdot\text{kg}^{-1}$). This phenomenon may be due to a sudden increase in anthropogenic activities after the lifting of the lockdown, resulting in significant emissions and a consequent rise in concentrations. $\sum_{16}\text{PAH}$ concentrations varied from previous years during the pandemic, suggesting that anthropogenic activities had a considerable impact on PAH pollution. The impact of the lockdown on PAHs in water is presented in **Fig. S4**.

3.4 Source apportionment of PAHs

Fig. 4 illustrates the percentage source contributions and fractional source concentration of individual PAHs in water and SPM in the Elbe River from 2015 to 2021. In the case of PAHs on the SPM, the PMF model extracted four source contributors (**Fig. 4e**). The first factor contained the largest percentage of NAP (55%), which was indicative of the volatilization of petroleum-related products (Hu et al. 2013). The second contributor was characterized by CHR, ANT, PYR, FLUH, BaA, and PHE. Furthermore, among these PAHs, PHE (59%), FLUH (45%), and PYR (54%) were regarded as the source fingerprint of coal combustion (Harrison et al. 1996, Larsen and Baker 2003, Sofowote et al. 2008). Hence, the second potential source contributor was represented by coal combustion. The third source was predominated by BbF, BaP, BgP,

IDP, and BkF, which were consistent with the characteristics of compounds from the emissions of gasoline-powered engines (Harrison et al. 1996, Ravindra et al. 2008, Simcik et al. 1999). The final source provided high contribution percentages of BaA, CHR, and DBA, which were recognized as diesel tracers (Feng et al. 2019, Lang et al. 2015, Wang et al. 2020b).

The percentages of potential source contribution to PAH in water and SPM of the Elbe River from 2015 to 2021 are presented in **Fig. 4b** and **f**. For PAHs in water, vehicular emissions from gasoline (40.7%) and diesel (25.9%) -powered engines had the largest contributions, followed by coal combustion (21.3%) and volatilization (12.1%). For PAHs on SPM, coal combustion (34.6%) and gasoline-powered engine emissions (34.2%) were determined as the two predominate sources, followed by diesel-powered engine emissions (21.1%) and volatilization of petroleum-related products (10.1%). Thus, the dominant sources of PAHs in the Elbe River can be determined as vehicle and coal combustion emissions. The source apportionment is consistent with the identified potential sources of PAHs in other waters around the world (**Table S7**). The evaluation of the PMF model is presented in **Text S4**, **Fig. S5**, and **Table S8**.

From the perspective of spatial distribution, **Fig. 4c** and **g** show the source contribution to PAHs in water and SPM for each station along the Elbe River. For the stations (W6-W11) at downstream, vehicular emissions occupied the predominated contributions (65% - 94%) to the PAHs in water (**Fig. 4c**). By contrast, the contributions of vehicular emissions for the stations (W1-W5) at the upstream ranged from 25% to 54%. In addition, the contributions of diesel emissions to PAHs on SPM occurred at the downstream stations of S8 (34%) and S9 (32%) were also relatively higher than those at the other stations (9% -25%) (**Fig. 4g**). This heterogeneity of source contribution might be attributed to the spatial difference in anthropogenic activities along the Elbe River. Hamburger, one of the most populous cities in Germany, was located at the downstream of Elbe River. As a result, a large amount of vehicles in the city emitted PAH emissions to the surrounding environments. Furthermore, since Hamburger is the largest seaport in Germany, extensive shipping activities rendered diesel emissions to

be the predominant source of PAHs at the downstream of Elbe River.

From the perspective of temporal variation, **Fig. 4d** and **h** show the percentage contribution of sources to PAHs in water and SPM for the individual years of 2015-2021. Vehicular emissions from gasoline and diesel-powered engines remained the predominant contributions to both PAHs in SPM (46%-69%) and water (61%-71%) during this period. Moreover, in 2020, the vehicular contribution to PAHs in SPM decreased from 51% to 46%, while the contribution of coal combustion increased by 5% (**Fig. 4h**). The shift of source contribution in 2020 might be attributed to the lockdown for COVID-19. The traffic closure during the lockdown period might result in a reduction in vehicular contribution. Since people spend more time at home, the high level of residential activities might lead to an increase in PAHs from coal combustion.

3.5 Health risks from PAH exposure

Fig. 5a shows the average ILCR values of $\sum_{16}$ PAHs in water and SPM of the Elbe River for children, teenagers, and adults from 2015 to 2021 through oral and dermal exposure. Human health risk can be determined as potentially high risk if the risk value is larger than 10^{-3} ; potential moderate risk if the risk value is between 10^{-4} and 10^{-3} ; potential low risk if the risk value is between 10^{-6} and 10^{-4} ; and virtually safety if risk value is lower than 10^{-6} (Keshavarzifard et al. 2018). The main exposure pathway of PAH risk is dermal contact whose ILCR (1.41×10^{-4}) was two orders of magnitude higher than that of oral (2.09×10^{-6}). From the perspective of exposure media, the total cancer risk from SPM (8.13×10^{-5}) was higher than that from water (6.15×10^{-5}), and both of them were identified as potential low-risk. Concerning different population groups, cancer risk for adults was much higher than that for children and teens, which might be attributed to the larger ingestion rate and surface area of adults. Besides, the relatively high ILCR in children may be explained by their lower body weight, which demonstrates that the early development of their bodies makes them more susceptible to cancer risk. The risk pattern generated in this study is in agreement with previous literature (Rajasekhar et al. 2018, Sarria-Villa et al. 2016a, Wu et al. 2011).

Fig. 5b presents the annual total health risks of PAH exposure from water and SPM in the Elbe River from 2015 to 2021. Changes in health risks depend on the trend

in individual 16PAHs with different toxicities. Annual ILCR values of PAHs in water and SPM decreased by 18% from 1.54×10^{-4} in 2015 to 1.27×10^{-4} in 2019, and rebound to 1.40×10^{-4} in 2020-2021. Based on uncertainty analysis, the probability distribution of risk for adults from PAHs on SPM is generated in **Fig. 5c**. Although the 50th percentile risk values were determined as potential low-risk, the realistic risks in extreme conditions might be much higher. For example, the 90th and 99th percentile dermal ILCR values for adults were 5.1×10^{-4} and 1.1×10^{-3} , respectively, thereby suggesting potential moderate to high cancer risks. The uncertainty distributions of other ILCR values are presented in **Fig. S6-7**.

4. DISCUSSION

4.1 PAH characterization in the Elbe River

With the assistance of long-term monitoring along the Elbe River, PAH pollution was characterized by the aspects of level, trend, and potential sources. Through the comparison with other waters worldwide (**Tables S9-10**), the PAH contamination range in the water of the Elbe River was lower than that obtained for the Tonghui River in China (Zhang et al. 2004), Northern Greece (Manoli et al. 2000), and the Mississippi River in the USA (Zhang et al. 2007). In comparison, the concentration range of PAHs on SPM exceeded that measured in Barcelona in Spain (Guitart et al. 2007), Incheon Harbor in Korea (Kim et al. 1999), and York River in the USA (Countway et al. 2003). These findings suggest that the relatively high PAH content of SPM in the Elbe River should be given enhanced priority.

Previous studies have shown a decreasing trend in PAH concentrations between 2001 and 2016 due to the European Water Framework Regulation (Li et al. 2022a). However, in recent years PAH concentrations have remained almost constant between 2015 and 2021. This may be due to the emergence of a bottleneck in the reduction of PAH emissions, which is related to the end-of-pipe control technologies, energy mix, and consumption. Although the amount of energy consumption shows a decreasing trend from 2000 to 2022, oil and coal are predominated in Germany's energy mix,

accounting for 53% of total energy in 2022 (**Fig. S8**). The difficulties in energy transition and the improvement of end-of-pipe control technologies (e.g. residential emission sources) slow down the decline of PAH emissions and concentrations. In addition, this study identified the predominant sources of PAHs in the Elbe River as coal and vehicular emissions through PMF source apportionment. Therefore, promoting and popularizing electric vehicles, improving exhaust emissions from automobiles, and enhancing the utilization of clean energy can effectively reduce the source emissions and PAH pollution in the environment.

4.2 Impacts of anthropogenic activities on PAH contamination

The lockdown during the epidemic could be equivalent to a large-scale socio-environmental experiment, demonstrating the intense impacts of anthropogenic activities on environmental pollutants. Many previous studies have reported significant decreases in atmospheric pollutants during the lockdown around the world. PAHs are one of the by-products of combustion, and the main source of its environmental occurrence is atmospheric emissions from anthropogenic activities. In this study, we validated the remarkable influence of anthropogenic changes in the epidemic on PAH pollution in aquatic systems by characterizing the long-term monitoring of 16PAHs in the Elbe River, Germany. As shown in **Fig. 2e-f**, relatively lower concentrations of $\Sigma_{16}\text{PAHs}$ in both water and SPM were observed during the COVID period. Because seasonal factors can significantly affect variations in concentrations, we adopted the difference between the concentration during the epidemic and the average values of previous years in the same month. As PAHs had high sorption to SPM, changes in PAHs on SPM were more sensitive to the sudden shift in anthropogenic intensities. During each of the three closures in Germany over two years, there was a pronounced downward trend in PAH on the SPM (**Fig. 3b**). However, there was a dramatic increase in human activity after the unsealing, releasing large amounts of PAH emissions. This resulted in the annual average concentration ($3.94 \text{ mg}\cdot\text{kg}^{-1}$) during the epidemic being almost the same as in previous years (**Fig. 2d**). Furthermore, energy consumption in Germany also shows a similar trend to PAH concentrations. For instance, energy from

coal combustion declined dramatically from 2.36×10^6 TJ in 2019 to 1.87×10^6 TJ in 2020 and rebounded to 2.23×10^6 TJ in 2021 (**Fig. S6**). Therefore, this study demonstrated a strong consistency between PAH concentrations and changes in the anthropogenic intensity, implying that source control from improved cleaner production is an effective pathway for mitigating PAH pollution in the aquatic environment.

5. CONCLUSION

On the basis of the continuous long-term monitoring of priority PAHs in the Elbe River, this study quantified the great impacts of lockdown restrictions against COVID-19 on the PAHs contamination in the surface water. The major conclusions include:

(1) The average concentrations of $\sum_{16}\text{PAHs}$ in water and SPM remained essentially constant with $0.055 \mu\text{g}\cdot\text{L}^{-1}$ and $3.77 \text{ mg}\cdot\text{kg}^{-1}$ from 2015 to 2021, respectively. PAH concentrations on SPM at upstream were higher than those at downstream.

(2) For each lockdown in Germany, significant reductions in concentrations of PAHs on SPM were determined from 2020 to 2021. Nevertheless, dramatic rises in $\sum_{16}\text{PAH}$ concentrations were observed during the removal of the lockdown, which might be attributed to the rebound of anthropogenic activities.

(3) The major sources of PAHs were identified as the emissions from vehicle and coal combustion in the Elbe River. Besides, the health risks of PAHs were determined as potential low risk.

In summary, this study provides empirical evidence at a river basin scale for a coupling between reductions in anthropogenic activities and declining PAH concentrations in receiving water. Furthermore, it is essential for worldwide countries to figure out socially and economically sustainable alternatives to fossil fuel consumption in industrial and transportation sectors, as well as cleaner household-used energies to achieve the reductions in PAH pollution we have observed during the response to COVID-19.

AUTHOR CONTRIBUTIONS

Ruifei Li designed the original draft and conducted the analysis, methodology, and writing. Yu Luo, Xu Zhu, Jin Zhang, Zhenyu Wang, and Wenyu Yang contributed to conceptualization and software. Yu Li, and Hui Li contributed to the reviewing and supervision.

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CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

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