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Influence of Source Apportionment of PAHs Occurrence in Aquatic Suspended Particulate Matter at a Typical Post-Industrial City: A Case Study of Freiburger Mulde River

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Abstract: Polycyclic aromatic hydrocarbons (PAHs) have received extensive attention because of their widespread presence in various environmental media and their high environmental toxicity. Thus, figuring out the long-term variances of their occurrence and driving force in the environment is helpful for environmental pollution control. This study investigates the concentration levels, spatial variance, and source apportionment of PAHs in suspended particulate matter of Freiburger Mulde river, Germany. Results show that the concentrations of the 16 priority PAHs suggested by USEPA ($\Sigma_{16}\text{PAHs}$) were in the range of 707.0–17,243.0 $\mu\text{g kg}^{-1}$ with a mean value of $5258.0 \pm 2569.2 \mu\text{g kg}^{-1}$ from 2002 to 2016. The relatively high average concentrations of $\Sigma_{16}\text{PAHs}$ were found in the mid-stream and upstream stations of the given river (7297.5 and 6096.9 $\mu\text{g kg}^{-1}$ in Halsbrucke and Hilbersdorf, respectively). In addition, the annual average concentration of $\Sigma_{16}\text{PAHs}$ showed an obvious decreasing pattern with time. Positive Matrix Factorization (PMF) receptor model identified three potential sources: coke ovens (7.6–23.0%), vehicle emissions (35.9–47.7%), and coal and wood combustion (34.5–47.3%). The source intensity variation and wavelet coherence analysis indicated that the use of clean energy played a key role in reducing PAHs pollution levels in suspended sediments. The risk assessment of ecosystem and human health suggested that the $\Sigma_{16}\text{PAHs}$ in the given area posed a non-negligible threat to aquatic organisms and humans. The data provided herein could assist the subsequent management of PAHs in the aquatic environment.

Keywords: long-term; PAHs; PMF; source identification; ecological and human health risk assessment

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs), which consist of a group of hydrocarbons composed of two or more linear, angular, or clustered benzene rings, have received high attention because they are abundant in the ecological environment and their high carcinogenicity, genotoxicity, teratogenicity, bioaccumulation [1–3]. PAHs enter the environment through various pathways, which might pose a serious threat to environmental organisms. As many as 200 forms of PAHs and their metabolites have been identified, among which $\Sigma_{16}\text{PAHs}$ were regarded as the main contaminants [4,5]. Therefore, an in-depth investigation of the occurrence of PAHs in the environment and its associated driving forces is essential.

Regarding the environmental behaviors, PAHs widely exist in a variety of environmental media, such as the atmosphere [6–8], water [1,7,9], sediments [10–12], soil [13–15], and road dust [16–19]. Research has proven that the aquatic suspended sediment was an important sink and transport medium of PAHs discharged into the environment [1,20,21]. Therefore, investigating concentration levels, spatial distribution, and source apportionments of PAHs in the water system is helpful to mitigate PAH in the aquatic environment.

Changes in the source dynamics of PAHs often lead to spatiotemporal differences in their concentrations and compositional characteristics in the environment. Quantitatively analyzing the changes in the source contribution of PAHs in the environment could help to investigate the impact of human activities on the occurrence of PAHs in the environment. There have been many studies on the source identification of PAHs in river suspended sediments [22–25]. However, quantitative analysis studies on long-term changes in the source driving forces of PAHs in suspended sediments are still lacking.

In terms of source apportionment, the receptor model has been a commonly used source identification method, which identifies the main factors affecting the dataset by extracting the key characteristic information in the environmental dataset, such as principal component analysis (PCA) [16,26,27], chemical mass balance (CMB) [26,28,29], UNMIX [13,16,27], and positive matrix factorization (PMF) [16,30,31]. The PMF model is the most widely used receptor model in recent years [14,32]. It imposes non-negative constraints on the source profile matrix and the source contribution matrix to improve the physical meaning of the model solution [30,33]. In addition, PMF affords users the ability to process input data individually, thus helping reduce the possible adverse effects of data anomalies caused by sampling, experimentation, or other processes on the source apportionment results [34–36]. Compared with other receptor models, the PMF model has higher accuracy and stability, and it can calculate more complete and detailed source apportionment results [12,37,38]. Therefore, the PMF model was employed in this study to conduct the quantitative source identification of PAHs in suspended particulate matter in the aquatic environment.

Accordingly, to characterize the occurrence of PAHs in the aquatic environment, a long-term variation and source shift were investigated. The detailed focuses were to (1) firstly evaluate the concentration level, the spatiotemporal distribution of PAH in the suspended particulate matter; (2) figure out the potential sources and their driving force during the long-term period; and (3) assess the ecological risk and possible human hazards posed by PAHs in the given area.

2. Materials and Methods

2.1. Study Area

The Mulde river (consisting of the Freiburger Mulde, the Zwickauer Mulde, and the Vereinigte Mulde) is one of the most diverse ecosystems in the Saxony state, Germany. In the past, the Mulde river basin has received widespread attention due to its thriving mining activities [39–41]. The Freiburger Mulde river originates near Moldova on the Erzgebirge ridge in the Czech Republic and flows north to Freiberg in Saxony, Germany. The Czech Republic and the state of Saxony experienced relatively rapid urbanization and significant economic growth during 2002–2016, which might lead to more serious PAHs pollution in the river.

The locations of PAH measurement stations in the Freiburger Mulde river are shown in Figure 1. Sampling data for the suspended particulate matters were obtained at 6 stations: Katzenstein (upstream), Hilbersdorf (midstream), Halsbrucke (midstream), Siebenlehn (midstream), Dobeln (midstream), and Erlbn (downstream). Suspended particulate matter samples were obtained mainly using centrifugation or filtration of residues from special sedimentation tanks (mostly monthly mixed samples) at the measuring stations. In this study, 16 PAHs, namely naphthalene (NAP), acenaphthene (ACY), acenaphthene (ACE), fluorene (FLU), phenanthrene (PHE), anthracene (ANT), fluoranthene (FLUH), pyrene (PYR), benzo[a]anthracene (BaA), chrysene (CHR), benzo[b]fluoranthene

(BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), dibenzo[a,h]anthracene (DBA), indeno [1,2,3-cd]pyrene (IDP) and benzo[g,h,i]perylene (BgP), were analyzed. The detailed information on the sampling collection and analysis is provided in Section S1 of Supplementary Materials.

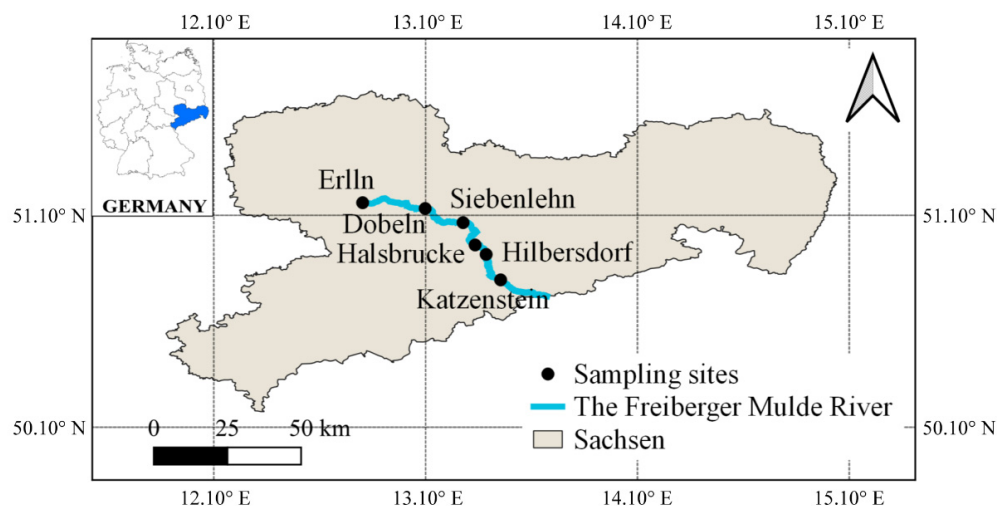


Figure 1. Sampling sites and spatial distribution of PAHs concentrations.

2.2. PMF Receptor Model

PMF is a multivariate analysis model that solves the problem in the weighted least squares sense with non-negative constraints [35]. The data matrix provided by the user would be divided into factor contribution matrix G , factor profiles matrix F , and residual matrix E . It supposes X as the initial data set:

$$X = GF + E \quad (1)$$

The elements e_{ij} of matrix E could be written as:

$$e_{ij} = x_{ij} - \sum_{k=1}^p g_{ik}f_{kj} \quad (2)$$

where e_{ij} is residual content for the j^{th} pollutant determined in the i^{th} sample. x_{ij} is the concentration of j^{th} pollutant determined in the i^{th} sample, g_{ik} is the relative contribution of factor k to sample i , and f_{kj} is the concentration of j^{th} pollutant in factor profile k .

The PMF model aims to reduce the residual matrix Q to a minimum. The function Q with $f_{kj} \geq 0$, $g_{ik} \geq 0$ is expressed as:

$$Q = \sum_{i=1}^n \sum_{j=1}^m \left(\frac{x_{ij} - \sum_{k=1}^p g_{ik}f_{kj}}{u_{ij}} \right)^2 = \sum_{i=1}^n \sum_{j=1}^m \left(\frac{e_{ij}}{u_{ij}} \right)^2 \quad (3)$$

where u_{ij} is the data uncertainty. The missing values in the data matrix are suggested to replace by species median values.

2.3. Wavelet Analysis

Wavelet analysis has been widely used to decompose time series to characterize their significant local power changes [3,42,43]. Wavelet analysis divides the variations in time series into time scales and time locations, which helps researchers to further analyze local changes in time series [44]. A detailed introduction to the wavelet analysis method is provided in Section S2 of Supplementary Materials. Wavelet coherence (WTC) is used to analyze the degree of local correlation of the sequence. In this study, wavelet

coherence was used to analyze the potential correlations of temporal variations between PAHs concentrations in suspended sediments and the anthropogenic emissions-related indicators (e.g., fuel, vehicle emissions, etc.). The WTC coefficient is calculated by the following formula [45]:

$$R^2(a, b) = \frac{S(a^{-1}|W_{xy}(a, b)|^2)}{S(a^{-1}|W_x(a, b)|^2) \cdot S(a^{-1}|W_y(a, b)|^2)} \quad (4)$$

where $0 < R^2(a, b) < 1$ and S is the smoothing operator.

2.4. Ecological Risk Assessment

The sediment ecological toxicity screening guideline for *ERL* and the U.S. National Oceanic and Atmospheric Administration's effect range median (*ERM*) values were used to determine the possible biological effects of PAHs in aquatic sediment [46]. The hazard quotient (*HQ*) for different PAHs was determined as the ratio between the concentration of individual PAHs ($\mu\text{g kg}^{-1}$) and the *ERM* [47,48]. Therefore, the values of *HQ* and *MHQ* were obtained for the total PAHs:

$$HQ_i = C_i / ERM_i \quad (5)$$

$$MHQ_i = \sum HQ_i / n \quad (6)$$

where C_i is the content of species i ($\mu\text{g kg}^{-1}$), ERM_i is the *ERM* value of species i , and n is the number of PAHs. For example, if the value of *MHQ* was less than 0.11, indicating that there was a 9% probability of being toxic to benthic fauna, while the value within the range of 0.11–0.51 indicates a probability of 21%; the value between 0.51 and 1.5 indicates a probability of 49%, and the higher value represents a probability of 76% [49].

2.5. Carcinogenic Risk Assessment

The potential carcinogenic risk was determined based on the value of toxic benzo[a]pyrene equivalent (*TEQ*). To transform the concentration and estimation of benzo[a]pyrene equivalent doses, the *TEFs* used in this study are provided in Table S2 [50–52]. PAHs species i was calculated as follows:

$$BaP_i dose_i = TEF_i \times C_i \quad (7)$$

where C_i is the content of species i ($\mu\text{g kg}^{-1}$), and TEF_i is the toxic factor of species i . The *TEF* values of individual PAHs are listed in Table S2. The total *TEQ* can be calculated by:

$$TEQ = \sum_i BaP_i dose_i \quad (8)$$

3. Results and Discussion

3.1. Distribution Characteristics of PAHs

As shown in Table 1, the general concentrations of Σ_{16} PAHs in six stations during 2002–2016 ranged from 707.0 to 17,243.0 $\mu\text{g kg}^{-1}$, with a mean value of $5258.0 \pm 2569.2 \mu\text{g kg}^{-1}$. The multi-year average concentrations of individual PAHs rank as: FLUH ($922.5 \pm 517.4 \mu\text{g kg}^{-1}$) > PYR ($731.1 \pm 392.6 \mu\text{g kg}^{-1}$) > PHE ($496.6 \pm 300.0 \mu\text{g kg}^{-1}$) > CHR ($445.8 \pm 220.5 \mu\text{g kg}^{-1}$) > BaA ($431.2 \pm 211.0 \mu\text{g kg}^{-1}$) > BbF ($425.9 \pm 206.7 \mu\text{g kg}^{-1}$) > BaP ($409.6 \pm 191.0 \mu\text{g kg}^{-1}$) > IDP ($358.6 \pm 195.5 \mu\text{g kg}^{-1}$) > BkF ($335.5 \pm 177.3 \mu\text{g kg}^{-1}$) > BgP ($286.3 \pm 144.1 \mu\text{g kg}^{-1}$) > ANT ($140.0 \pm 122.2 \mu\text{g kg}^{-1}$) > NAP ($81.2 \pm 83.8 \mu\text{g kg}^{-1}$) > DBA ($80.8 \pm 50.2 \mu\text{g kg}^{-1}$) > FLU ($50.0 \pm 50.1 \mu\text{g kg}^{-1}$) > ACY ($33.1 \pm 19.4 \mu\text{g kg}^{-1}$) > ACE ($29.8 \pm 20.9 \mu\text{g kg}^{-1}$). The value of the coefficient of variation (CV) represents the level of spatiotemporal variation in the concentration of Σ_{16} PAHs. The relatively high values of CV were observed in the concentrations of ACE (2.9) and IDP (2.1), indicating that they might be related to a sudden pollution incident or point

source pollution. Meanwhile, the relatively low one was observed in the concentrations of BbF (0.6) and BkF (0.6), which might be related to emissions from non-point sources.

Table 1. The statistics results of PAHs (in $\mu\text{g kg}^{-1}$ of dry weight).

PAHs	Katzenstein		Halsbrücke		Hilbersdorf		Erlin		Siebenlehn		Dobeln		Sum	
	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD
NAP	37.7	31.3	98.6	60.9	90.7	121.7	93.1	94.8	77.5	43.9	38.3	25.5	81.2	83.8
ACY	20.8	10.4	45.5	18.7	41.2	24.4	26.2	10.8	34.7	16.8	20.4	18.7	33.1	19.4
ACE	19.1	9.0	35.2	21.3	32.6	20.6	26.1	16.9	35.7	26.2	17.7	8.4	29.8	20.9
FLU	23.9	10.3	55.0	34.7	54.2	41.0	49.5	52.0	60.0	68.1	22.9	12.9	50.0	50.1
PHE	319.8	120.6	677.4	393.9	584.5	302.5	385.6	215.1	578.8	292.9	261.0	128.7	496.6	300.0
ANT	82.9	40.6	180.7	94.4	160.1	86.8	99.5	65.2	183.6	192.5	73.0	50.2	140.0	122.2
FLUH	698.0	315.5	1311.7	625.5	1077.0	507.3	622.8	238.0	1117.3	536.6	536.5	229.0	922.5	517.4
PYR	580.4	260.7	1013.5	442.4	871.3	410.4	501.7	189.2	860.0	398.8	443.5	176.6	731.1	392.6
BaA	377.9	169.0	580.0	234.6	487.7	189.6	335.9	130.3	490.4	240.7	260.5	108.8	431.2	211.0
CHR	390.9	190.1	624.8	250.7	507.2	192.0	330.6	131.7	512.5	239.2	264.5	100.3	445.8	220.5
BbF	385.0	167.7	631.3	270.6	485.9	186.0	298.5	100.5	479.6	193.3	276.0	109.9	425.9	206.7
BkF	275.7	147.9	477.8	217.0	390.1	179.2	235.0	91.2	385.7	168.0	216.5	116.0	335.5	177.3
BaP	394.1	170.1	547.0	214.3	476.7	182.2	308.2	111.4	455.5	199.1	258.5	112.1	409.6	191.0
DBA	122.9	87.9	101.3	49.8	101.1	56.6	55.2	25.2	86.3	43.2	43.6	13.5	80.8	50.2
IDP	368.3	170.1	521.3	256.4	412.6	183.7	254.0	91.6	394.5	210.5	230.0	88.4	358.6	195.5
BgP	292.7	150.7	396.5	176.1	324.0	148.1	207.8	75.2	323.3	142.2	185.0	62.1	286.3	144.1
$\Sigma_{16}\text{PAHs}$	4390.1	1893.8	7297.5	2958.3	6096.9	2429.2	3829.6	1429.6	6075.5	2627.5	3147.8	1196.5	5258.0	2569.2

Notes: SD: Standard deviation.

The spatial distribution of the average annual concentration during 2002–2016 of $\Sigma_{16}\text{PAHs}$ at six stations in the Freiburger Mulde river is illustrated in Table 1. A similar average annual concentration was observed in Hilbersdorf, Halsbrücke, and Siebenlehn stations. The highest mean concentration of $\Sigma_{16}\text{PAHs}$ was in Halsbrücke ($7297.5 \mu\text{g kg}^{-1}$), and the second-highest value was located in Hilbersdorf ($6096.9 \mu\text{g kg}^{-1}$). High concentration levels of PAHs suggested that these stations might be strongly influenced by anthropogenic activity. Meanwhile, the lowest mean concentration of $\Sigma_{16}\text{PAHs}$ was observed in Dobeln ($3147.8 \mu\text{g kg}^{-1}$). The higher PAHs concentration levels were mainly distributed in the upstream of the river, which indicated that there were obvious spatial differences in the emission sources of PAHs.

Figure 2 showed the annual changes in the concentrations of $\Sigma_{16}\text{PAHs}$ from 2002 to 2016. The annual average concentration of PAHs decreased from $5457.0 \mu\text{g kg}^{-1}$ in 2002 to $3095.0 \mu\text{g kg}^{-1}$ in 2016. In addition, Mann–Kendall trend analysis was used to explore the long-term trends of $\Sigma_{16}\text{PAHs}$ concentrations from 2002 to 2016. The results suggested that the PAHs pollution level in the river decreased significantly during the period (confidence level = 97.7% and slope = -198.6). In addition, the maximum and minimum annual average concentrations of PAHs appeared in 2009 and 2012, respectively. Two extremely high PAHs concentration levels were observed (up to $57,480.0$ and $55,950.0 \mu\text{g kg}^{-1}$, respectively) monitored at the Hilbersdorf site in 2009, which might be related to a special pollution event at the station.

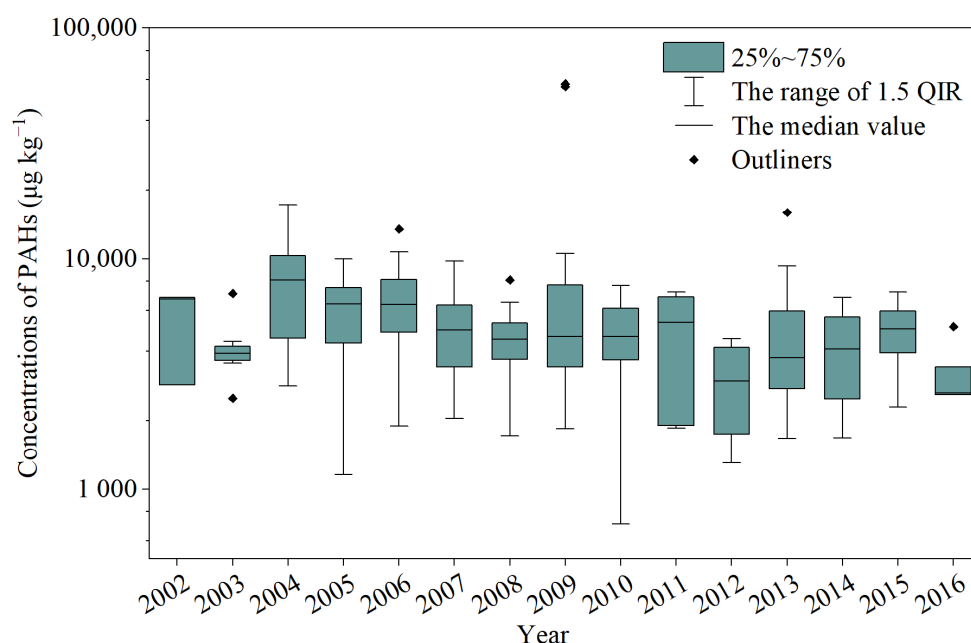


Figure 2. The annual changes in the concentrations of Σ_{16} PAHs from 2002 to 2016.

Compared to research globally, the concentration of PAHs in suspended particulates in the Freiburger Mulde river was comparable to Victoria Harbour (China) [53], Kaohsiung Harbor (China) [54], Pearl River Estuary (China) [55], and Henan Reach of Yellow River (China) [56]; higher than Taranto Gulf (Italy) [57], Incheon Harbor (Korea) [58] and Hun River (China) [23]; and significantly lower than Xiamen Harbor (China) [53], Boston Harbor (USA) [59], Norwegian Harbor (Norway) [60] and Western Harbour (Egypt) [61] (Table S3). Following the general practice for the determination of relative rates of emissions [62], four levels of contamination can be divided: (1) low level: $0.0\text{--}100.0\text{ }\mu\text{g kg}^{-1}$; (2) moderate level: $100.0\text{--}1000.0\text{ }\mu\text{g kg}^{-1}$; (3) high level: $1000.0\text{--}5000.0\text{ }\mu\text{g kg}^{-1}$; and (4) very high level: $>5000.0\text{ }\mu\text{g kg}^{-1}$. Accordingly, the PAH concentration in the suspended particulate matter from the Freiburger Mulde river from 2002 to 2016 was cataloged as high to very high levels, indicating the moderate-high PAHs pollution in the study river.

3.2. Composition Profiles of PAHs

Figure 3a shows the profile of the benzene-member ring of Σ_{16} PAHs in different stations. The maximum proportion of PAHs was found in station Halsbrucke (23.7%), followed by stations Hilbersdorf (19.8%), and Katzenstein (14.2%). In addition, the proportion of PAHs with various ring numbers in Σ_{16} PAHs in different stations is illustrated in Figure 3b. These 16 PAHs were commonly classified into three classes based on the number of benzene rings: low molecular weight (LMW, 2–3 rings) (NAP, ACE, ACY, FLU, ANT, PHE), middle molecular weight (MMW, four rings) (FLUH, PYR, BaA, CHR) and high molecular weight (HMW, 5–6 rings) (BaP, BbF, BkF, BgP, IDP, DBA). The annual average concentration of the MMW-PAHs ranged from 1505.0 to $3530.0\text{ }\mu\text{g kg}^{-1}$; that of the HMW-PAHs ranged from 1209.6 to $2675.2\text{ }\mu\text{g kg}^{-1}$, and that of the LMW-PAHs ranged from 433.2 to $1092.3\text{ }\mu\text{g kg}^{-1}$.

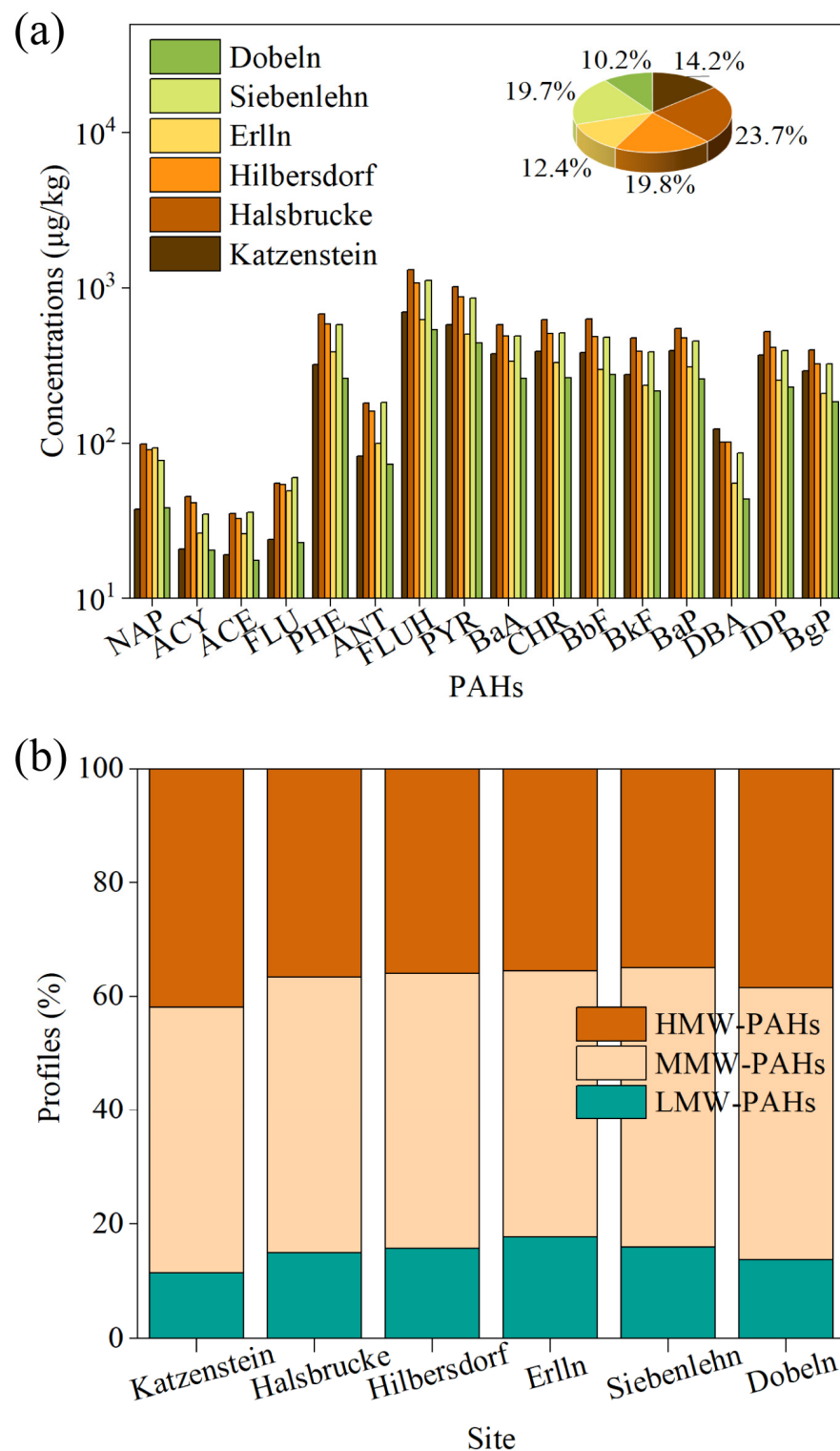


Figure 3. Spatial variance in different stations (a) and composition of PAHs with different ring numbers (b).

Previous studies have reported the composition of PAHs in suspended sediments from other rivers. In developing countries, the content of LMW-PAHs in river water environments is usually higher. For example, LMW-PAHs dominated the suspended sediments in the Pearl River Estuary, followed by MMW-PAHs [63]; MMW-PAHs dominated the suspended sediments of the Subarnarekha River in India, followed by LMW-PAHs [64]. In

developing countries, biomass burning remains an important source of energy [65,66]. The high proportion of LMW-PAHs in these rivers might be associated with higher-intensity biomass burning emissions nearby.

The higher proportions of high molecular weight PAHs might be indicative of anthropogenic influences from higher urbanization. In this study, the MMW-PAHs contributed the highest proportion of Σ_{16} PAHs (46.6–49.1%) in all stations, while the HMW-PAHs contributed the second-highest proportion of Σ_{16} PAHs (35.0–41.9%). Previous studies have shown that the spatial variances of MMW-PAHs and HMW-PAHs concentrations in the environment are highly positively correlated with the densities of industrial activities and traffic emissions, respectively [67,68]. The proportions of MMW-PAHs exhibited fewer spatial differences in the river. Meanwhile, the proportion of HMW-PAHs in the upstream site was significantly higher than that in the downstream site, and the highest proportion of LMW-PAHs appeared in the midstream site. Therefore, the study river might be heavily polluted by emissions from upstream urban traffic and industrial activities. In addition, studies have shown that the Elbe is highly polluted by HMW-PAHs [1]. As one of the important tributaries flowing into the Elbe, the Freiburger Mulde river is one of the important input sources of PAHs with a high ring number. However, the above analysis indicates that the higher content of HMW-PAHs in the river might be attributed to upstream input.

3.3. Source Apportionment Derived by PMF

In this study, the long-term PAHs concentration dataset was divided into three parts: 2002–2006, 2007–2011, and 2012–2016. Quantitative source identification of Σ_{16} PAHs in different periods was performed using the PMF model to investigate the characteristics of the variation in the number and intensity of PAHs sources over the years. Table S4 lists the correlation coefficient (R^2) values between observed and predicted PAHs concentrations of the PMF model [69]. All the values of R^2 were greater than 0.6, which indicated that the PMF model had a high prediction accuracy for the source apportionment of PAHs in the river.

The PMF model results are illustrated in Figure 4. The model identified three potential sources of Σ_{16} PAHs during the study period. Factor 1 counted for 7.6–23.0% of the total PAH concentrations. This factor was dominated by a high portion of NAP and a relatively high portion of FLU. The NAP was the predominant PAHs of the coke oven [70]. Moreover, FLU was the important tracer of PAHs emitted from the coke ovens in Chicago [6]. Thus, this factor was denoted as a coke oven. Factor 2 counted for 35.9–47.7% of the total PAHs concentrations, and it was dominated by BbF, BkF, BaP, BgP, IDP, and DBA. The high loading of BgP was suggested as the traces of vehicular emissions, especially in gasoline engines [6,71]. The high weighting of BbF and BkF represented the emissions from diesel-power vehicles [72]. In addition, the high content of IDP was mostly observed in emissions of the high-way tunnel and vehicle engines [70]. Therefore, factor 2 was denoted as vehicle emissions in this study. Factor 3 counted for 34.5–47.3% of the total PAHs concentrations. This factor was characterized by FLUH, ACE, PYR, and ANT. High concentrations of alkyl PAHs (such as FLUH, ACE, and PYR) were the predominant tracers of coal combustion [6]. The dominance of HHW-PAHs in this factor also indicated that this factor was highly related to industrial coal combustion (high-temperature combustion processes) [73]. Moreover, ANT was the important tracer of wood combustion [72]. Thus, factor 3 was identified as coal and wood combustion.

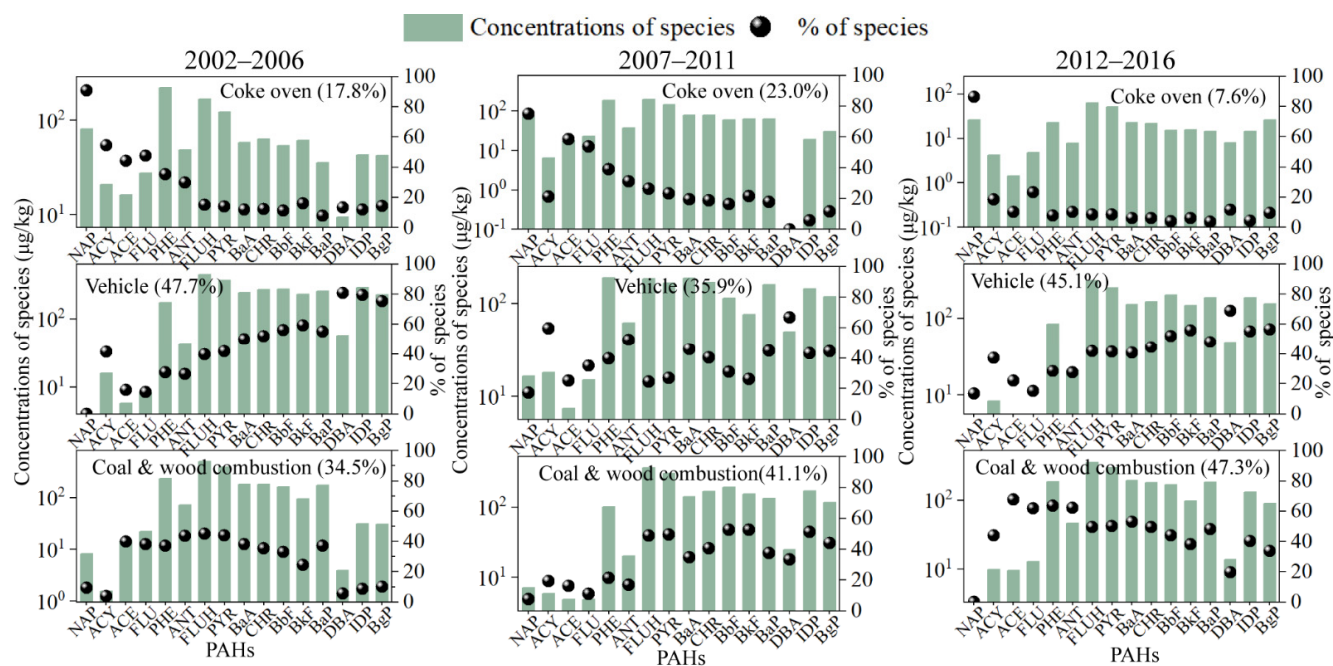


Figure 4. The profiles and contributions of each source by PMF model.

In addition, the results were compared with the source analysis results of PAHs in other rivers. In developing countries, coal and biomass are often the main sources of energy supply. For example, the main sources of PAHs in the seven major river basins in China and the Guanlan River (China) are coal and biomass combustion (source contributions account for 40.0% and 42.8% of the total, respectively) [74,75]. In the more urbanized developed countries, the proportion of traffic emissions is usually higher. The main sources of PAHs in the Heihe and Ashtabula Rivers (Ohio) and Brisbane Rivers (Australia) were characterized by traffic emissions (source contributions accounted for 58.0% and 42.8% of the total, respectively) [76,77]. In this study, the contributions of coal and biomass combustion and traffic emission sources are similar. The unexpectedly high portion of coal and biomass combustion sources might be attributed to the relatively low level of industrialization in the cities along the river.

In addition, the results of displacement (DISP) and bootstrap (BS) were used to evaluate the stability of the PMF model (Table S5). The matching rates of BS runs in all PMF models were over 80%, which indicated that the random errors of the models were sufficiently small to be acceptable [4,34]. In addition, the DISP was mainly developed to analyze the rotational ambiguity of the PMF model results. No swapped factors were observed in the DISP running of all PMF models, and the decrease in Q value was within a reasonable range [4]. Therefore, the above results indicated that the PMF model in this study was sufficiently stable and reliable enough to be accepted.

3.4. Potential Driving Force of PAHs

The quantitative source apportionment results of PMF indicated that the pollution levels of PAHs in urban rivers were mainly affected by anthropogenic activities. The contributions to PAHs concentrations in the Freiburger Mulde river from different sources vary significantly over time (Figure 5). Furthermore, the results of wavelet coherence revealed the detailed effects of various anthropogenic activities on the concentrations of PAHs in suspended sediments (Figure 6). The numerical range for judging the value of wavelet coherence (WC) is between 0 and 1, and the larger value indicates a stronger correlation. Arrows to the left and right represent positive and inverse correlations, respectively.

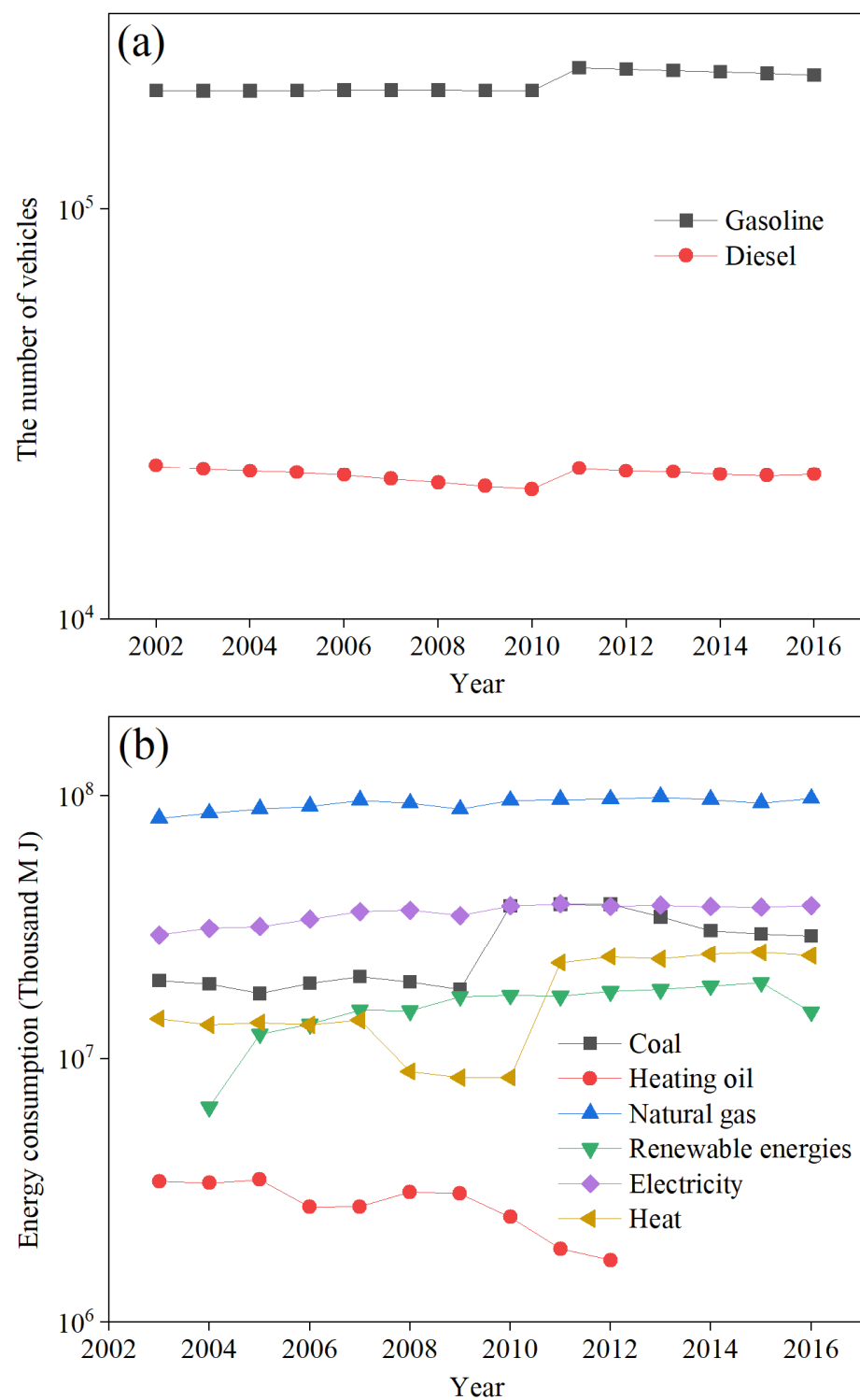


Figure 5. The annual changes in numbers of vehicles (a) and energy consumption (b). Notes: The calculation of changes in energy emissions and the number of vehicles was based on Saxony statistics (www.regionalstatistik.de, accessed 18 March 2022).

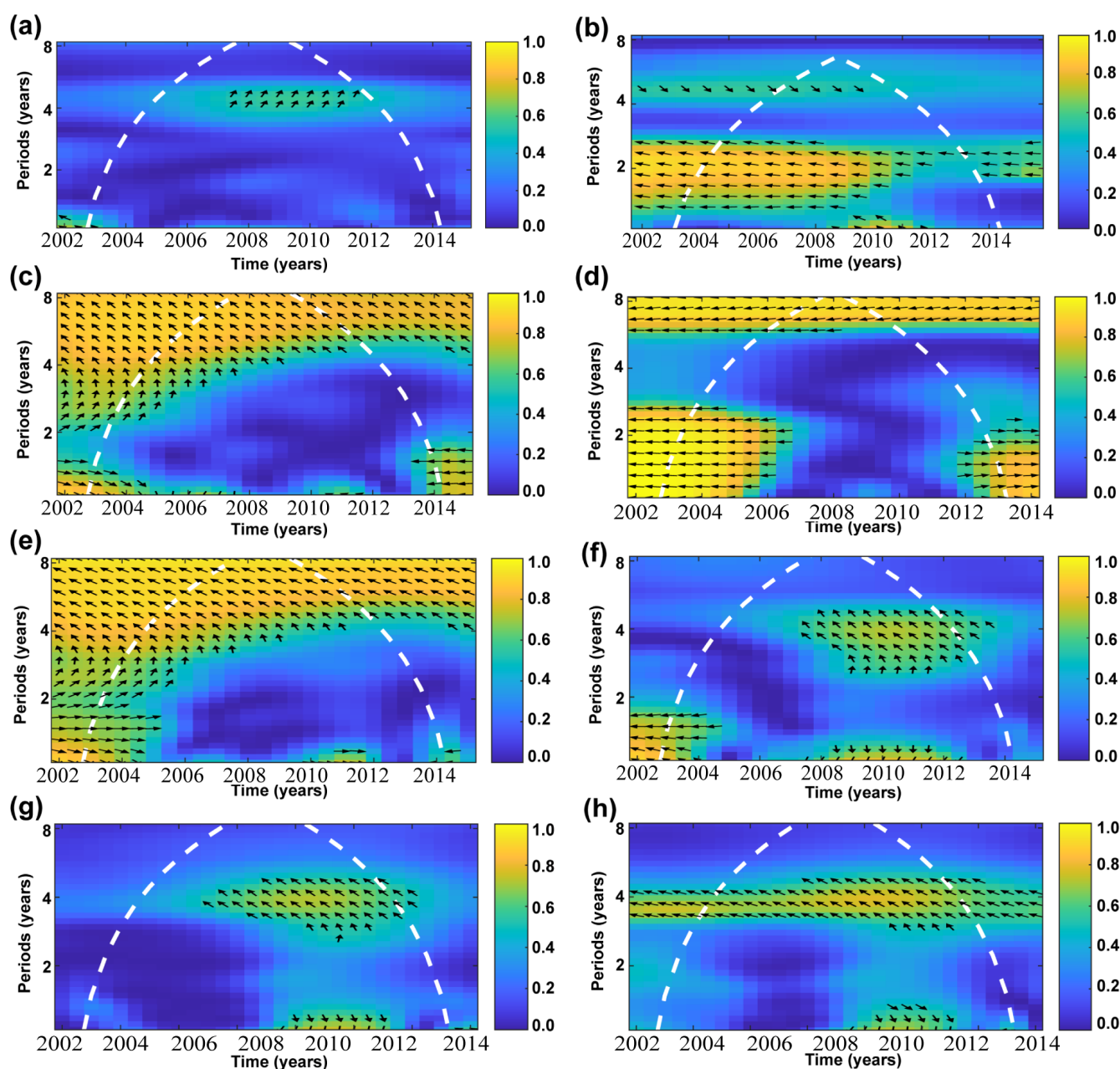


Figure 6. The results of wavelet coherence: (a) coal consumption; (b) heating fuel consumption; (c) natural gas consumption; (d) renewable energy consumption; (e) power consumption; (f) heating energy; (g) number of petrol cars; (h) number of diesel cars.

The contribution of coke ovens increased from 17.8% in 2002–2006 to 23.0% in 2007–2011 and then dropped to 7.6% in 2012–2016 (Figure 4). Coal is a critical raw material and energy source in the coke oven industry. The annual changes in energy consumption of various types of energy in Saxony from 2003 to 2016 were shown in Figure 5b. The value of coal consumption increased substantially in 2009–2010 and then declined over time after 2010. The reduction in coal consumption in recent years was related to the government’s vigorous promotion of clean energy [1,78]. The variation pattern of the coke oven was similar to that of the coal consumption sequence, indicating that the decreased contribution of the coke oven to the PAHs pollution in the study area might be attributed to the reduction in the use of coal products. In addition, the positive effects of coal consumption on PAHs emissions in the study area with a time scale of 5–6 years were observed from 2007 to

2011 (Figure 6a). The reduction in industrial coal consumption in recent years might be attributed to the European directives to limit emissions of PAHs in 2008 (2008/50/EC) and 2010 (2010/75/EU) [79,80]. However, the strength of WC was insignificant, which might indicate that variations in PAH concentrations in the river might be mainly influenced by emission sources outside the study area (e.g., upstream input). Besides, combining the analysis in Section 3.2 and other studies, the MMW-PAHs from industrial emissions (especially coal-fired power plants and the thriving mining industry) in the river might originate from the emissions in upstream [39,81].

In addition, the contribution of coal and wood combustion sources increased from 34.5% in 2002–2006 to 41.1% in 2007–2011, and then to 47.3% in 2012–2016 (Figure 4). Similar to the coke oven source, the increase in the contribution from this source between 2007 and 2011 might be related to the increase in coal consumption (Figure 5b). In addition, despite how clean energy was gradually used to replace fossil fuels in recent years, the combustion of traditional fuels (such as wood, coal, and fuel oil) was still the main source of heating for residents in the cold season [78]. The wavelet coherence results showed that the strong negative effect of fuel oil used for heating on PAHs emission appeared during 2002–2007 (Figure 6b). Fuel oil consumption for heating had dropped significantly during the period. The decrease in the use of heating oil has led to an increase in the use of traditional fuels. Research showed that the change in heating energy structure is related to the economic ability of residents [78]. The sudden change in economic conditions in 2008 led to the increase in the use of cheaper fuels in household life, which might be the important reason for the increased contribution from wood and coal combustion intensity during 2012–2016.

However, the contribution of vehicle emission sources showed a different pattern of change. The contribution of the source decreased from 47.7% in 2002–2006 to 35.9% in 2007–2011 and then increased to 45.1% in 2012–2016 (Figure 4). Figure 5a illustrates the change in the number of gasoline and diesel vehicles from 2002 to 2016. During 2002–2010, the number of diesel vehicles showed an obvious downward trend, while the number of both gasoline and diesel vehicles increased significantly after 2010. The wavelet coherence results also showed that the increase in the number of gasoline and diesel vehicles (Figure 6g,h) around 2010 had a significant positive effect on the emissions of PAHs (the relatively high WC values located in 2010–2012 with the time scale of 0–1 years). Besides, the wavelet coherence results showed that the negative effects of gasoline vehicles (Figure 6g) and diesel vehicles (Figure 6h) were concentrated in 2009–2013 and 2002–2013, respectively [71]. Using negative correlations to describe the changes in the number of cars and PAHs concentrations is inaccurate. The factors that affect the intensity of traffic emissions are often complex, especially over longer periods. Previous studies suggested that there was an obvious positive correlation between the concentration of PAHs in the urban water environment and the number of vehicles [82,83]. The implementation of vehicle emission reduction strategies and technologies in recent years could reduce the traffic emissions of PAHs (Euro 4 in 2005 and Euro 5 in 2009) [84,85]. In addition, reducing the proportion of PAHs in fuel was also an effective emission reduction measure [86]. In future work, it might be more appropriate to use estimated vehicle PAHs emissions to investigate their effects on the changes in PAHs concentrations, especially in studies on long-term changes in PAHs.

During the study period, the consumption of natural gas (Figure 6c), renewable energy (Figure 6d), and electricity (Figure 6e) had a very significant negative effect on the concentration of PAHs in suspended sediments on a time scale of 6–8 years. Compared with renewable energy, the increase in the use of natural gas and electric energy has a more significant effect on the decrease in PAHs concentration in river suspended sediments, which might be due to the fact that their consumption is significantly higher than that of renewable energy. On the other hand, natural gas is mainly used for household combustion activities, which greatly reduces the use of other high PAHs emitting fuels. These results indicate that the further promotion of renewable energy will play a crucial role in reducing PAHs emissions in the future.

3.5. Risk Assessment of PAHs

3.5.1. Potential Biological Effects

This study used the *ERL* and *ERM* values to comprehensively assess the potential ecological risk of Σ_{16} PAHs in the suspended particulate matter of the Freiburger Mulde river (Table S6). The average concentrations of NAP, ACY, and ANT were $101.0 \mu\text{g kg}^{-1}$, $39.0 \mu\text{g kg}^{-1}$, and $170.0 \mu\text{g kg}^{-1}$, lower than the respective *ERL* values of 38.6%, 11.3%, and 80.0%, respectively. However, the average value of the other PAHs ranged within the values of *ERL* and *ERM*. The maximum values of PHE and DBA were $2000.0 \mu\text{g kg}^{-1}$ and $1100.0 \mu\text{g kg}^{-1}$, respectively, which were higher than their respective *ERM* values of 25.0% and 76.3%. Excluding PHE and DBA, the maximum PAHs concentrations were lower than their respective *ERM* values. This indicated that the ecosystem of the Freiburger Mulde river might have been threatened by PAHs during the study period, which was mainly attributed to the high concentrations of PHE and DBA caused by unexpected events (such as flooding).

Table S7 summarizes the *MHQs* and *HQs* for the components of Σ_{16} PAHs at different stations. The total PAHs in the suspended particulate matter of the Freiburger Mulde river were commonly dominated by MMW-PAHs and HMW-PAHs. The *MHQ* in six stations ranged from 0.1 to 0.3; the maximum *MHQ* (0.3) was observed at the Hilbersdorf station. The *HQs* of PHE and DBA at the Hilbersdorf station were higher than other PAHs. This evidence indicated that the contamination of PHE and DBA needed considerable attention, which was mainly generated by coke ovens and vehicle emissions. In addition, in this study, all *MHQ* values (0.1–0.3) were in the range of 0.11 to 0.5, which corresponds to a 21.0% probability of toxicity to benthic fauna [87]. The results represented that the Σ_{16} PAHs in the river might have adversely affected the benthic fauna of the Freiburger Mulde river and the aquatic environment.

3.5.2. Potential Human Toxicity

The total *TEQ* calculated for Σ_{16} PAH in the study area varied from 407.0 to 1180.0 $\mu\text{g kg}^{-1}$ dry weight (Table 2). The highest *TEQ* value was observed at Halsbrücke station, which is located in the middle of the Freiburger Mulde river and was close to urban and commercial areas. The *TEQ* values declined from upstream to downstream of the river. On the one hand, it might be related to the fact that the emission sources of PAHs were concentrated in upstream. On the other hand, it might be also related to the degradation and dilution of PAHs (especially LMW-PAHs) during the migration of rivers [88].

Table 2. The total toxic benzo[a]pyrene equivalent ($\mu\text{gTEQ/kg}$) and the percentage contribution of each PAHs (%).

PAHs	Ring Number	Katzenstein		Hilbersdorf		Halsbrücke		Siebenlehn		Dobeln		Erlin	
		C × TEF	Contribution	C × TEF	Contribution	C × TEF	Contribution	C × TEF	Contribution	C × TEF	Contribution	C × TEF	Contribution
NAP	2	0.04	0.9	0.2	2.1	0.1	1.4	0.08	1.3	0.04	1.2	0.09	2.4
ACY	3	0.02	0.5	0.07	0.8	0.05	0.6	0.04	0.6	0.02	0.6	0.03	0.7
ACE	3	0.02	0.4	0.06	0.7	0.04	0.5	0.04	0.6	0.02	0.6	0.03	0.7
FLU	3	0.02	0.5	0.1	1.7	0.06	0.8	0.06	1.0	0.02	0.7	0.05	1.3
PHE	3	0.3	7.3	1.0	11.9	0.7	9.3	0.6	9.5	0.3	8.3	0.4	10.1
ANT	3	0.9	1.9	2.8	3.5	1.8	2.5	1.8	3.0	0.7	2.3	1.0	2.6
FLUH	4	0.7	16.0	1.5	18.4	1.3	18.0	1.1	18.4	0.5	17.0	0.6	16.3
PYR	4	0.6	13.3	1.2	15.2	1.0	13.9	0.9	14.2	0.4	14.1	0.5	13.1
CHR	4	4.1	9.0	6.2	7.6	6.2	8.6	5.1	8.4	2.6	8.4	3.3	8.6
BaA	4	39.5	8.7	60.7	7.4	58.0	7.9	49.0	8.1	26.0	8.3	33.6	8.8
BbF	5	39.2	8.6	55.0	6.7	63.1	8.7	48.0	8.0	27.6	8.8	29.9	7.8
BkF	5	28.5	6.3	43.9	5.4	47.8	6.6	38.6	6.3	21.7	6.9	23.5	6.1
BaP	5	409.0	9.0	549.1	6.7	547.0	7.5	455.5	7.5	258.5	8.212	308.2	5.4
DBA	5	131.3	2.9	113.5	1.4	396.5	1.4	86.2	1.4	43.6	1.4	55.2	1.4
IDP	6	37.5	8.2	47.9	5.9	52.1	7.1	39.5	6.5	23.0	7.3	25.4	6.6
BgP	6	0.3	6.6	3.8	4.7	4.0	5.4	3.2	5.3	1.9	5.9	2.1	8.0
TEQ	-	692.2	100.0	887.1	100.0	1179.8	100.0	729.7	100.0	407.0	100.0	483.8	100.0

The values and percentage contributions of *TEQ* of each PAHs are summarized in Table 2. The contributions of *TEQ* of individual PAHs were ranked in the following order: FLUH (16.0–18.4%) > PYR (13.1–15.2%) > PHE (7.3–11.9%) > CHR (7.6–9.0%) > BaP (5.4–9.0%) > BaA (7.4–8.8%) > BbF (6.7–8.6%) > IDP (5.9–8.2%) > BgP (5.3–8.0%) > BkF (5.4–6.9%) > other PAHs (less than 3.0%). MMW-PAHs and HMW-PAHs, which mainly originated from fossil fuel combustion, accounted for most of the total *TEQ* values in the river. Thus, the relatively high *TEQ* values in river suspensions during the study period were highly correlated with vehicle emissions and coal and wood combustion.

The values for *TEQ* in the suspended particulate matter from other studies are presented in Table S8. The *TEQ* values (407.0–1180.0 µgTEQ/kg dry wt.) in this study were lower than those in Naples Harbor (69.0–18,681.0 µgTEQ/kg dry wt.) (Italy) and Kaohsiung Harbor (55.0–1964.0 µgTEQ/kg dry wt.), but were higher than those in Meiliang Bay (94.0–845.0 µgTEQ/kg dry wt.) (China), Eastern Mediterranean Sea (17.0–872.0 µgTEQ/kg dry wt.) (Lebanon), and Barents Sea (638.0–1777.0 µgTEQ/kg dry wt.) (Russia). Overall, the average potential toxicity risk in the suspended particulate matter of the Freiburger Mulde river was moderate for humans from 2002 to 2016 compared to other rivers.

4. Conclusions

This study investigated the concentration levels, spatiotemporal variances, potential sources, and their driving forces, as well as ecological and human health risk assessments of Σ_{16} PAHs in suspended particulates of typical surface water. The concentrations of Σ_{16} PAHs in the study area were at moderate-high pollution levels. Higher concentrations of Σ_{16} PAHs were concentrated in the midstream and upstream of the river. The concentration of Σ_{16} PAHs in the suspension showed an obvious decreasing pattern with time. The PMF receptor model identified three potential PAH sources of coke ovens, vehicle emissions, and coal and wood combustion. Results indicated that the changes in the energy structure had a greater impact on the changes in contributions of these sources during 2002–2016. The ecological risk and human health assessment showed that a higher risk of PAHs in the given river during 2002–2016 was mainly influenced by special pollution events. The PAHs emitted from vehicle emission and coal and wood combustion posed a threat to the aquatic organisms and human health. The data provided herein suggest that policymakers might need to increase the proportion of clean energy use to reduce the pollution levels of PAHs in the water environment.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/su14116646/s1>, Section S2: Sample analysis [32]; Season S2: Mann-Kendall trend test [89–91]; Season S3: Wavelet analysis [1,45,92–94]; Table S1: The sampling information for stations; Table S2: Proposed toxicity equivalency factors (TEFs) for individual PAHs; Table S3: Global review of PAHs concentrations [23,53–61,83,95–107]; Table S4: The correlation coefficient (R^2) values between observed and predicted concentrations of the PMF model; Table S5: The results of Displacement (DISP) and Bootstrap (BS) by PMF model; Table S6: Effects range-low (ERL), effects range-median (ERM), mean and maximum concentration of 16 PAHs in µg kg^{−1}; Table S7: Hazard quotients (HQ) and mean hazard quotient (MHQ) of 16 PAHs from 2002–2016; Table S8: *TEQ* values of PAHs at different locations [54,83,108–110].

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