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Polycyclic aromatic hydrocarbons in various Lithuanian water bodies and a positive matrix factorization-based identification of pollution sources

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Abstract. In this study, results on PAHs distribution, origin and ecological risks from several surveys are combined with a special focus on naphthalene. The study area comprises a large part of the Lithuanian territory and includes two biggest Lithuanian rivers, Nemunas and Neris (sampling locations with different hydrodynamics and varying anthropogenic pressure), four different lakes and the central part of the Curonian Lagoon. $\sum$16PAHs concentrations in the surface sediment ranged between 74.5 and 6377 ng g^{-1} dw. In 9 locations out of 14, naphthalene concentration exceeded TEL and posed a threat to the environment. Naphthalene was a significant contributor to the total PAHs concentration in the study area with concentrations ranging from 0.78 to 106 ng g^{-1} dw. Despite the highest naphthalene concentration recorded in Lake Talkša, naphthalene had only a minor contribution to the PAH profile in this lake. Even a lower impact of naphthalene was observed in the Curonian Lagoon. Naphthalene was found to be the prevailing compound in the Nemunas River Delta region, in a relatively clean Lithuanian Lake Dusia and in some Neris River locations. The highest contribution of naphthalene was observed in the areas with low urbanization degree, but close to peatlands. The main naphthalene source in the study area might be related to peatlands, albeit additional naphthalene sources, such as food cooking, burning of agricultural residues, wood and coal could not be completely rejected either. The sources of other PAHs are attributed to the vehicular emission, emissions from industry, biomass and coal burning.

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INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) are undoubtedly among the most ubiquitous organic pollutants present in the aquatic environment. Due to their toxicity, persistency and a tendency to bioaccumulate, 16 PAHs, including naphthalene (Naph), have been listed as priority pollutants by the United States Environmental Protection Agency (Keith, Telliard 1979). PAHs might enter the environment from various

sources, most of them related to human activities: spillage of petroleum or refinery products, combustion of fossil fuel and biomass (Rimayi *et al*. 2017). In addition to that, PAHs could also originate from biogenic sources (Pichler *et al*. 2021). For instance, perylene was reported to originate from the perylenequinones which are found in the plants, insects, crinoids and fungi (Guo, Liao 2020; Hanke *et al*. 2019; Varnosfaderany *et al*. 2015). However, anthropogenic activities play the most prominent role in PAHs pollution: PAHs

loadings in the environment were observed to correlate positively with the increasing intensity of industrialization (Karaca 2016; Liu *et al.* 2016).

PAHs are highly mobile, they can easily migrate from the areas of high anthropogenic loading to the aquatic environment by atmospheric deposition and surface runoff (Hadibarata *et al.* 2019). Once entered the aqueous environment, PAHs tend to settle and accumulate onto sediments due to their hydrophobicity (Varnosfaderany *et al.* 2015). Sediments act as a medium for the accumulation of organic pollutants and is a relevant secondary pollution source (Ranjbar *et al.* 2020).

The PAHs distribution pattern in the sediments of a water body is usually dominated by high molecular weight (HMW) PAHs (Kanzari *et al.* 2014; Zeng *et al.* 2018). However, while conducting several researches in various Lithuanian water bodies, we noticed that low molecular weight (LMW) PAH naphthalene is often found to exceed sediment quality guidelines (TEL) and/or be the prevailing compound in the sediment of lakes and rivers (Stakėnienė *et al.* 2019). Similar results were reported by several researchers worldwide. In some cases, high Naph concentrations were closely related to the coal mining industry (Achten, Hofmann 2009; Pies *et al.* 2007), albeit more often its prevalence has not

been comprehensively discussed (Baran *et al.* 2002; Lacorte *et al.* 2006; Shi *et al.* 2005). Naphthalene is classified as a probable human carcinogen (Jia, Batterman 2010). It was rather often found to exceed TEL in the sediment of various Lithuanian water bodies, which makes it a matter of concern.

In this study, results on PAHs distribution in sediments, possible origin and ecological risks from several different surveys are combined with a special focus on naphthalene. We seek to detect the main naphthalene sources and estimate its potential effects on the environment.

MATERIALS AND METHODS

Study area

The study area comprises a large part of the Lithuanian territory and includes two biggest Lithuanian rivers, Nemunas and Neris, four different lakes, and the Nemunas inflow zone of the Curonian Lagoon, which is a transitory zone between the Nemunas River and the Baltic Sea. Several locations with different hydrodynamics and varying anthropogenic pressure were sampled in the rivers. Lakes of different type, anthropogenic pressure and trophic state were studied (Fig. 1).
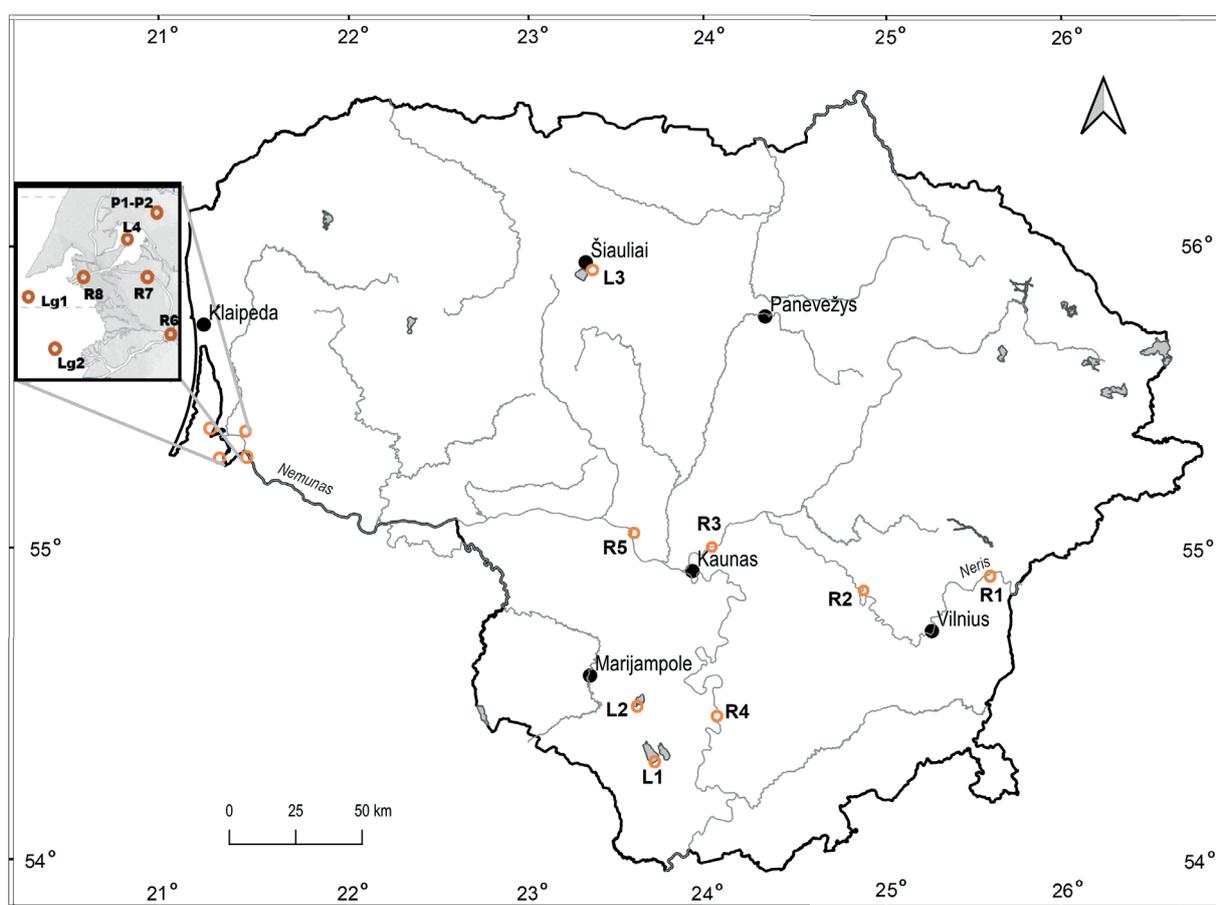
Fig. 1 Map of the study area. Red circles - sampling sites, the sedimentary environment indicated by letter: R – river, L – lake, Lg – lagoon, P – peatland.

Three stations were situated in different sections of the Neris River (R1–R3, Fig. 1). The Neris River (R1–R3) is the biggest tributary of the Nemunas and the second largest river in Lithuania, with a catchment area of 24,942 km^2. Flowing through the capital city Vilnius, the river receives municipal wastewater together with industrial waste. Five sampling stations were located in the Nemunas River arms (R4–R8), and 2 stations in the Nemunas inflow zone in the Curonian Lagoon (Lg1 and Lg2). The Nemunas River (R4–R8) is the biggest river in Lithuania, with a catchment area of 97,864 km^2 (Rimkus *et al*. 2013). It collects water together with both industrial and agricultural pollutants from 72% of the country's territory. In the Nemunas River Delta, just south of the Aukštumala Bog (Fig. 1, P1–P2), a shallow (depth 2–3 m) lake Krokų Lanka is situated (Fig. 1, L4). This adjacent eutrophic lake was formed when the deposit of the Nemunas River isolated part of the Curonian Lagoon. Two soil samples were collected in the Aukštumala Bog (peatland).

The lakes Dusia (L1) and Simnas (L2) are attributed to the basin of Lake Žuvintas. Lake Dusia is considered to be one of the cleanest Lithuanian lakes, whereas Lake Simnas is located close to the Simnas town and is connected with the fishery ponds (Taminskas *et al*. 2006). The vicinity of the city and the inflow from fishery ponds increase the amount of biogenic substances and pollutants in the lake. Lake Talkša (L3) experiences the influence of the surrounding city and landfill leachates (Taraškevičius, Gregorauskas 1993; Gregorauskienė 2006). Moreover, large amounts of untreated industrial sewage affected this lake significantly in the past. All these factors make Talkša one of the most polluted lakes of Lithuania.

The surface (0–3 cm) sediment samples were collected in the summer of 2015–2016 using a Van Veen grab sampler. Sediment samples were stored at $-20\,°C$ in aluminium boxes before further processing.

The Aukštumala Bog was sampled differently: Searching for possible sources of PAHs, floating sphagnum mats from a drainage ditch crossing a peatland area affected by a fire were sampled. The fire had taken place five years before the sampling. The surface of the drainage ditch was sampled using a telescopic stick and a brown glass vessel. Samples were stored on ice both in the field and during transportation.

Sample preparation and analysis

Sediment and floating sphagnum mat samples were prepared for GC-MS analysis following the procedure described by Martinez *et al*. (2004): 5–20 g of a dry sediment sample were extracted in an ultrasonic bath with 10 ml of n-hexane (Sigma-Aldrich, HPLC grade) and methylene chloride (1:1) (Sigma-Aldrich, HPLC grade) for 10 min. The procedure was repeated 3 times, the obtained extracts were combined, centrifuged and filtered through glass fibre filters. The volume of the as-prepared extracts was reduced to approx. 1 ml using a rotary evaporator. Florisil cartridges (Chromabond 6 ml/500 mg) were applied for sample cleaning. PAHs were eluted with 20 ml of dichlormethane, and a purified sample was evaporated to dryness under a gentle stream of nitrogen. Analytes were reconstituted in 200 μL of acetonitrile (Merck, HPLC grade). Blank samples were prepared following the same analytical procedure.

The targeted PAHs were analysed by the GC-MS-QP 2010 mass spectrometer operating in the selected ion monitoring mode. The injection mode was splitless, and the temperatures of the injector and ion source were both set to 250°C. Each compound was separated using a fused silica Rxi 5Sil Ms capillary column (30 m × 0.25 mm i.d. × 0.25 μm). The following temperature programme was applied: 75°C for 1 min, 75–250°C at 25°C/min, 250 to –310°C at 3°C/min and then 310°C for 7 min. The carrier (helium) flow rate was 1.18 mL/min. Compound identification was based on the retention time and m/z ratio of a PAH mixed standard (Restek), as well as on the literature data (Macias-Zamora *et al*. 2002). Concentrations of each individual PAH (16 PAHs: naphtalene (Naph), acenaphthylene (Acy), acenaphthene (Ace), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Fluo), pyrene (Pyr), benzo(a)anthracene (BaA), chrysene (Chr), benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo(a)pyrene (BaP), indeno(123-cd)pyrene (Incdp), dibenzo(ah)anthracene (DBA) and benzo(ghi)perylene (BghiP)) were determined using an external standard method, the calibration curve ranged from 0.01 to 1 μg/ml.

Replicate samples (n = 3) and procedural blanks were used for quality control. Relative standard deviation (RSD) was less than 10%, no traces of analytes of interest have been detected in the chromatograms of blank samples. The procedural blanks were periodically analysed for each batch of 10 samples. The recovery efficiency of the procedure was determined by analysing matrix samples spiked with a known amount of the EPA Method 8310 PAH Mixture (Restek). PAH recoveries were between 75% and 112%. Naph and Ace demonstrated the lowest recoveries: 81% and 75%, respectively, whereas the highest recoveries were obtained for higher molecular weight compounds: 112 for BghiP and 104 for InP. Recoveries for the rest of the analytes ranged from 88% to 97%.

The limit of detection (LOD) and the limit of quantification (LOQ) of the method were determined

by streaming five sets of analytical blanks. The LOD and LOQ of each PAH were determined by calculating three and ten multiples of standard deviation, respectively, of the mean of the signal-to-noise (S/N) ratio for ten measurements, in comparison to the baseline noise close to the peak of each PAHs blank. LOD was in the range of 0.015–0.2 while LOQ varied from 0.05–0.7.

Source apportionment: Positive Matrix Fractionation (PMF)

The US EPA's PMF 5.0 model (Paatero, Tapper 1994; Taghvaee *et al*. 2018) was applied to identify the main PAH sources and to quantify their contributions in the study area. Analysis was done following the PMF 5.0 user guide (US EPA 2014). The multivariate receptor model rotates the data matrix x_{ij} into a factor contribution matrix g_{ik} and a factor profile matrix fkj (Paatero, Tapper 1994):

$$x_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij}. \qquad (1)$$

Here: x_{ij}, e_{ij} are matrix i by j dimensions, g_{ik} is matrix i by k dimensions, f_{kj} is matrix k by j dimensions, i refers to the individual sample, j corresponds to the PAH compound, k refers to the decomposed source factor, p is the number of source factors. For the model optimization, minimizing of the objective function Q was performed (Paatero 1997):

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

Here: n and m indicate the number of measured samples and studied PAH species, respectively; u_{ij} is the uncertainty in the x_{ij} measurement.

To obtain the optimal number of factors, the model was first run with a different factor number (ranging from 2 to 6). The run resulting in the minimum Q(Robust)/Qexp value was chosen as the optimum PMF model. With the increasing number of factors, the values of the determination coefficient were slightly increasing. However, a higher number of factors resulted in lower robustness. Finally, 4 was selected as the optimal number of factors. The individual PAHs were categorized into strong, weak and bad as recommended by USEPA. Concentrations of the PAHs analysed were reproduced well by the model, except for BghiP. Hence, this PAH was set as weak and excluded from the model. For the remaining PAHs, the regression predictions approximated well the real data points, and the coefficient of determination (r^2) was in the range of 0.91–1.0.

RESULTS AND DISCUSSION

Polycyclic aromatic hydrocarbon loadings in bottom sediments

In the study area, concentrations of $\sum$16PAHs ranged between 74.5 and 6377 ng g^{-1} dw (Fig. 2, b). The lowest PAHs loadings were detected in the Nemunas River Delta (74.5 and 96.1 ng g^{-1} dw in the sampling stations R7 and R6, respectively) and in Lake Dusia (L1; 114 ng g^{-1} dw). The highest concentrations of 16 PAHs were observed in the sampling stations L3 (Lake Talkša). Baumard *et al*. (1998) classified sediment contamination with PAHs as low (0–100 ng g^{-1} dw), moderate (100–1000 ng g^{-1} dw), high (1000–5000 ng g^{-1} dw) and very high (>5000 ng g^{-1} dw). Following this system, sediments analysed in this study covered all the range of sediment contamination classes: from low to very highly contaminated. The Nemunas River Delta (R6–R8) could be classified as low contaminated, Lake Talkša (L3) corresponds to a very high contamination level, while the remaining sampling stations could be classified as moderately contaminated with PAHs (Fig. 1).

Figure 2a demonstrates that naphthalene was a significant contributor to the total PAHs concentration in the study area. The Curonian Lagoon was characterised by the lowest naphthalene concentrations (0.78 ng g^{-1} dw in Lg1 and 4.4 ng g^{-1} dw in Lg2). The highest naphthalene concentrations were recorded in Lake Talkša (L3, 106 ng g^{-1} dw), R6 (84.6 ng g^{-1} dw), R8 (73.3 ng g^{-1} dw), R4 (74 ng g^{-1} dw) and R7 (59.9 ng g^{-1} dw). Moreover, in 9 locations out of 14, naphthalene concentration exceeded TEL (Fig. 2a). Despite the highest naphthalene concentration recorded in the sampling station L3, naphthalene had only a minor contribution to the PAH profile in Lake Talkša (Fig. 3). Even lower impact of naphthalene was observed in the Curonian Lagoon which was characterised by the lowest values of Naph. In contrast, Naph was found to be the prevailing compound in the Nemunas River Delta region (stations R6–R8, L4), in a relatively clean Lithuanian Lake Dusia and in some Neris River locations (R1 and R3). The PAH composition pattern revealed that relatively polluted sites, situated in the cities or anthropogenic activities impact zones, are characterised by significantly lower contributions of Naph than the relatively clean sites located in the rural and relatively undisturbed areas. If compared to other studies, the prevalence of Naph has been observed in rivers and lakes of Poland, Spain, China (Baran *et al*. 2002; Lacorte *et al*. 2006; Shi *et al*. 2005). However, this phenomenon was not comprehensively studied and the sources of naphthalene were not identified.

Figure 3 also reveals that naphthalene was prevailing in the samples of the Aukštumala Bog. It should be noted that the analysis of the peatland samples

plays only a supportive role and helps us to identify PAH sources more precisely. These two samples were not collected from sediment like the rest of the samples, they were prepared in a completely different way. The main results of the study include PAHs distribution in sediments and their spatial variation. The peatland samples only play a supportive role and are not to be compared to the sediment samples.

Source apportionment: positive matrix factorization model

In this study, a positive matrix factorization (PMF) model was applied as a reliable multivariate source identification method (Fig. 4). It should be noted that BghiP was excluded from the model due to poor regression predictions and poor reproducibility. For the
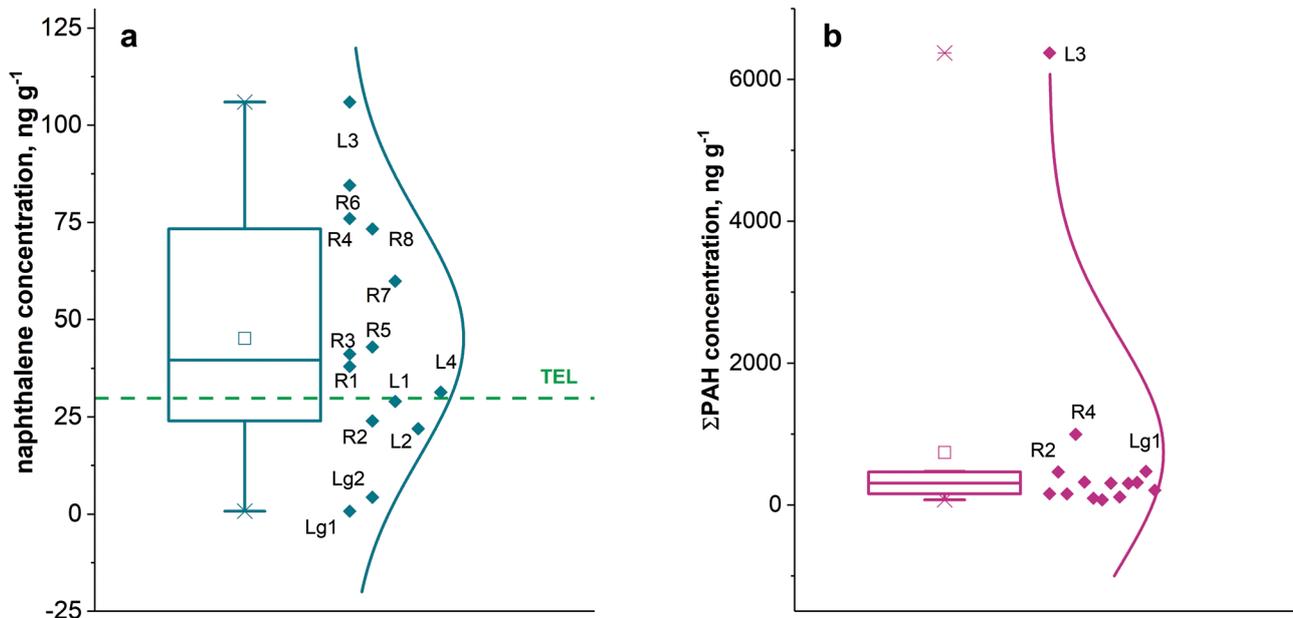
Fig. 2 Concentrations of naphthalene (a) and 16 PAHs (b) in the sediment samples of the study area
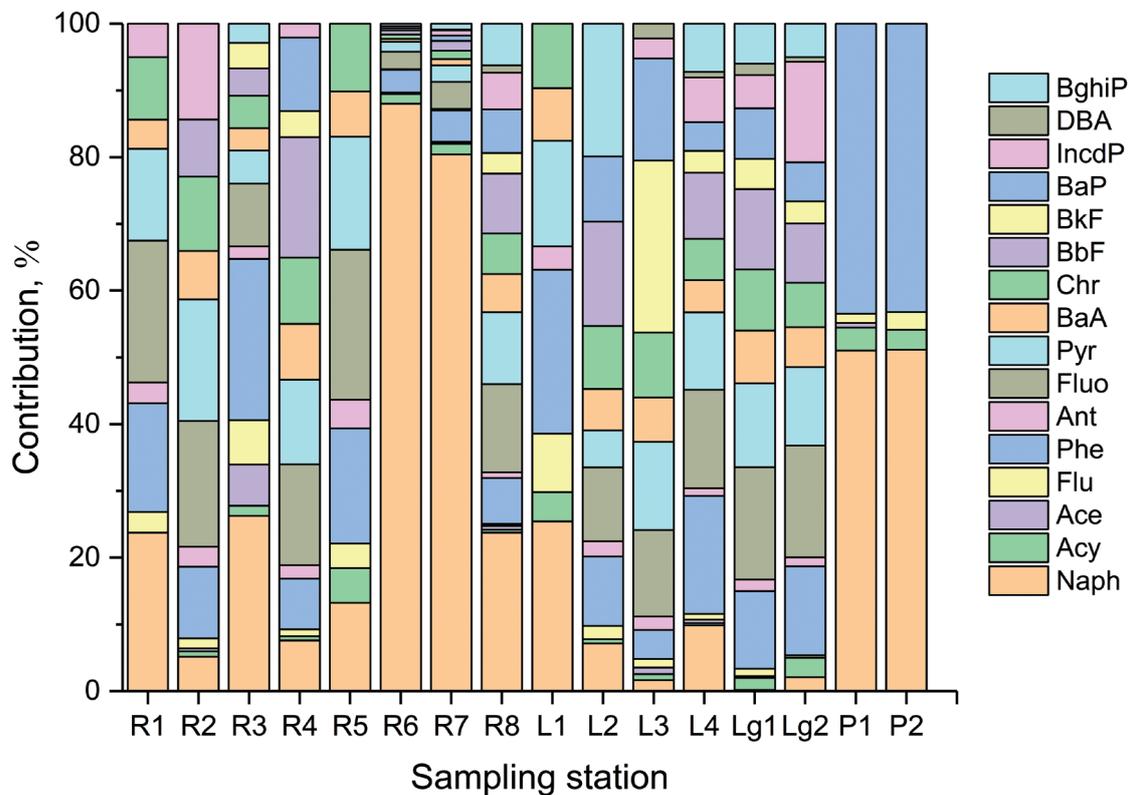
Fig. 3 Distribution pattern of individual PAHs

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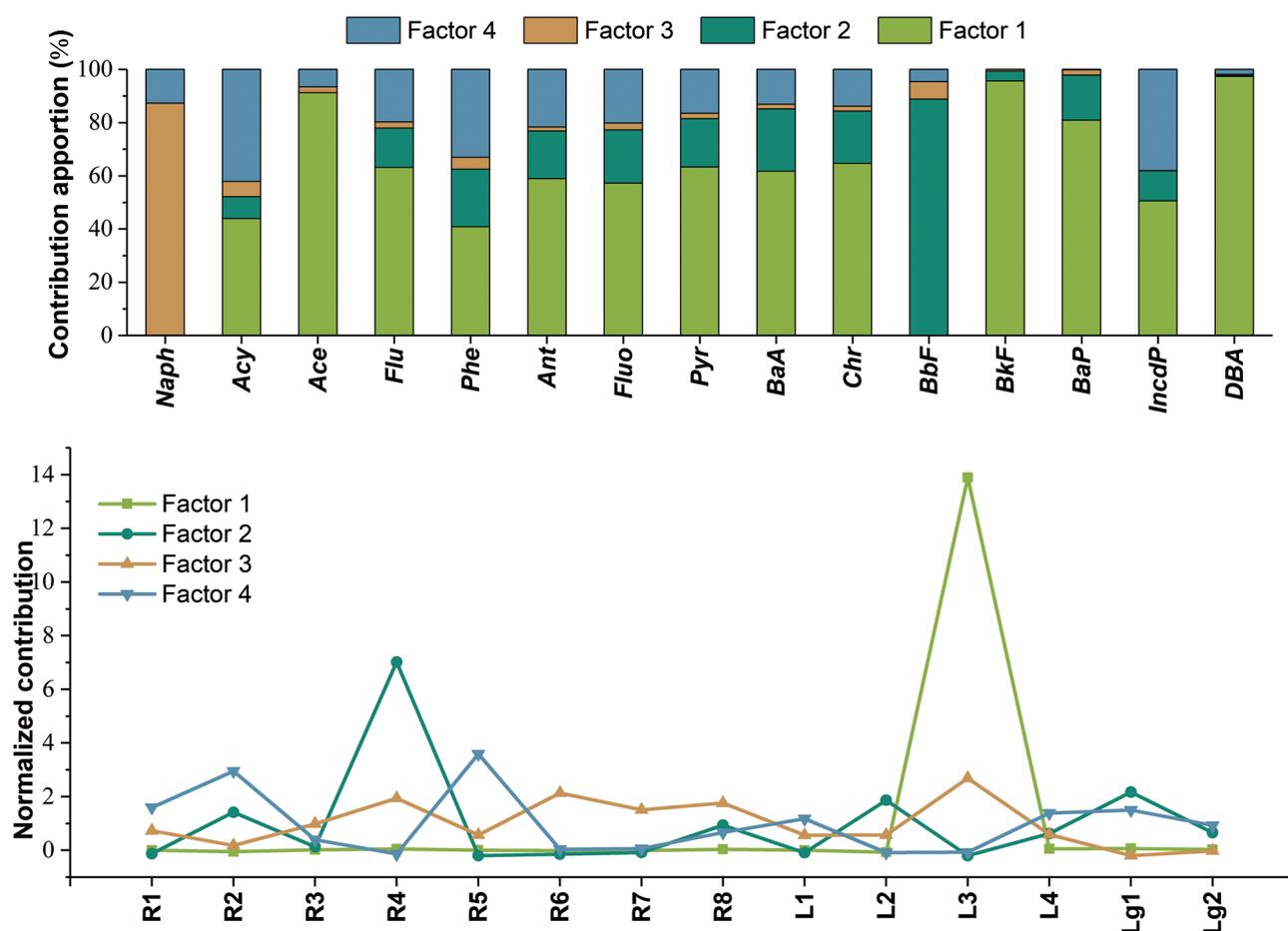
Fig. 4 Contribution of different factors for each compound determined by the PMF model and each factor contribution in the sampling stations of the study area

remaining PAHs, the regression predictions approximated well the real data points, and the coefficient of determination (r^2) was in the range of 0.91–1.0.

The first factor (F1) was defined by high loadings of Ace, BkF and DBA. Moreover, all the remaining PAHs, except for Naph and BbF, contributed significantly to this factor as well. Such profile is highly indicative of pyrogenic sources (Wang *et al*. 1999), and high levels of Flu, BkF and BaP were reported to originate from the diesel engine emission sources (Tavares *et al*. 2004; Wang 2020; Wu *et al*. 2014). Diesel vehicle emissions also contain significant concentrations of Pyr and Chr (Fang *et al*. 2019; Wang *et al*. 2020). However, it should be noted that Ace and Acy are tracers of industrial processes (Kong *et al*. 2013; Wu *et al*. 2014). DBA, Flu, Pyr, BaA, BaP, Pyr and Chr are also reported to be closely related to industrial emissions (Mougin 2002). Spatially, F1 showed a high impact on the sampling station L3 (Lake Talkša) and no significant effect on the remaining locations. Lake Talkša is located in the urbanised territory. By rain water collectors, surface runoff gets into the lake from the busiest street of the city. Thus, vehicle emissions and the influence of the surround-

ing city could not be refuted. Moreover, the lake is known to be heavily affected by post-industrial processes: for a long period of time, it has been polluted with unthreatened sewage from leather industry. Considering all of the above, F1 was described as a mixed source of industrial and vehicle emissions.

The second factor (F2) was loaded with BbF and showed some minor impact from Pyr, BaA and Chr. It should be noted that Phe, Ant and Flu loadings are significant for both F2 and F4, but are slightly higher on the latter. Hence, they were attributed to the fourth rotated component. BbF is found in vehicular emissions of gas and diesel (Davis *et al*. 2019; Fang *et al*. 2006). BaA and Chr hold higher emission factors for both gasoline and diesel vehicles (Chen *et al*. 2013). Phe, Flu, Pyr, BaA and Chr are considered to originate from coal combustion (Li *et al*. 2015; Wang *et al*. 2020). Taking these facts into account, factor 2 was considered a mixed source of coal combustion and vehicular emissions (diesel and gasoline). This factor played the most important role in the sampling station R4 and had some contribution in the stations R2, R8, L2, L4, Lg1, Lg2.

F3 was predominantly loaded with Naph. This factor was significant in all sampling stations, except

for the Curonian Lagoon and R2 (Neris River downstream of Vilnius city). The highest influence of this factor was observed in the Nemunas River Delta (R6–R8). F3 was also important in the sampling station R4 and Lake Talkša (L3), where relatively high naphthalene concentrations were recorded. LMW PAHs are known to be more labile and to have a short half-life if compared with higher molecular weight PAHs (Ravindra *et al*. 2008). Thus, relatively high concentrations of Naph suggest either the existence of a local emission source or a relatively recent introduction of these chemicals. Naphthalene might be emitted from human activities: biomass burning, gasoline and oil combustion, tobacco smoking, the use of mothballs, fumigants, food cooking (Jia, Batterman 2010). In addition to that, natural sources might also contribute to Naph contamination. As it was previously stated out by Stakėnienė *et al*. (2019), peatlands might be an important source of LMW PAHs. Peatland soil is known to contain high amounts of LMW PAHs, especially naphthalene (Tsibart *et al*. 2014; Turetsky *et al*. 2014). PAHs could be transported via surface runoff from peatlands into the surrounding water bodies and streams. The results of this study support this assumption. The highest contribution of F3 was observed in the Nemunas River Delta, which could be affected by the largest Lithuanian peatland Aukštumala Bog (P1, P2). Several peatlands (including Rėkyva) are also located around the Šiauliai city and Lake Talkša (L3) (Lietuvos durpynų... 1995). The northern part of Lake Dusia (L1) is known to be in the process of swamping and contains several peaty areas (Valatka *et al*. 2018; Lietuvos durpynų… 1995). Seeking to check whether peatlands might contribute to the increased Naph concentration in the surrounding water basins, an additional analysis was performed: several extracts from floating mats collected from a fire-affected peatland area in the Aukšumala Bog were analysed for PAHs. The Aukštumala Bog (P1–P2) is located close to the Nemunas River Delta, where the highest Naph contribution has been detected (Fig. 1). Indeed, naphthalene was found to be the prevailing compound in each of the peatland samples studied (Fig. 2), confirming that peatland fire-related naphthalene source in the Lithuanian rivers and lakes should not be rejected. Certainly, we could not claim that peatlands are the only source of Naph in the study area. Despite a potentially important role of these wetland ecosystems, the burning of agricultural residues, wood and coal, food cooking could be additional naphthalene sources (Jia, Batterman 2010).

F4 was loaded with Acy, Phe, Inp and, to a lesser extent, Fl, Ant and Flu. Acy was identified as a wood combustion marker (Jang *et al*. 2013; McDonald *et al*. 2000), while Oanh *et al*. (2015) and Wei *et al*. (2014) related higher loadings of Acy to the straw burning. Phe abundance was recorded in various pollution sources (Marr *et al*. 2004; Wang *et al*. 2020), but several studies relate Phe to the biomass combustion sources (Wang *et al*. 2020; Zhang *et al*. 2008). Even though the prevalence of LMW PAHs is characteristic of this factor, a quite significant loading of Inp should not be neglected either. Inp is detected in vehicular emissions from diesel and gas engines (Afshar-Mohajer *et al*. 2016; Harrison *et al*. 1996; Wang *et al*. 2009). Acy has also been used as a tracer for vehicles (Afshar-Mohajer *et al*. 2016). In view of the above, F4 represents mixed source biomass combustion and vehicular emissions, in particular emissions from diesel engines. The impact of F4 was the most obvious in the rivers (R1, R2 and R5), lakes (L1 and L4) and Curonian Lagoon (Lg1, Lg2) sampling stations (Fig. 1).

The fact that vehicle emission was found to contribute significantly in 3 factors out of 4 might seem confusing. However, one should keep in mind that a particularly broad territory was studied with many more or less urbanised areas. Vehicular emissions should thus contribute in most of the sampling stations, and such results should not be surprising.

CONCLUSIONS

Sediments analysed in this study covered all the range of sediment contamination classes: from low to very highly contaminated. Naphthalene was found to be a significant contributor to the total PAHs concentration in the sediment of various Lithuanian rivers and lakes. In more than 60% of the sampling stations, naphthalene concentration exceeded TEL. The highest naphthalene concentrations were recorded in Lake Talkša which was also found to be the most polluted sampling site. However, naphthalene had only a minor contribution to the total PAH profile in this lake. The PMF model revealed that Lake Talkša is mostly affected by industrial and vehicle emissions. Vehicular emission was also found to contribute significantly to PAH pollution in most of the sampling stations. We assume that the main naphthalene source in the study area might be related to the peatlands, albeit additional naphthalene sources, such as food cooking, burning of agricultural residues, wood and coal should not be completely rejected either. In order to understand such trend better, a more comprehensive studies focusing on the origin, transportation and distribution of naphthalene are required.

ACKNOWLEDGMENTS

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