Intercompartment

Research Article

Distribution of Polycyclic Aromatic Hydrocarbons (PAHs) in Floodplain Soils of the Mosel and Saar River*

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Abstract

Background, Aim and Scope. Polycyclic aromatic hydrocarbons (PAHs) have gained serious attention in the scientific community due to their persistence and toxic potential in the environment. PAHs may pose a risk to ecosystem health. Along the Mosel River/Germany, a tributary of the river Rhine, PAHs were found at significantly high concentrations (> 20 mg kg^{-1}, German national guideline value Z2, LAGA 1998). These high concentrations were detected during the construction of a storm water retention basin, in which the contaminated soils had to be removed and treated as hazardous waste. This resulted in higher construction costs for implementing flood prevention measures, but did not address the origin of these PAHs and its distribution along Mosel River. Hence, for future flood prevention projects, it is necessary to estimate the extent of PAH contamination along the Mosel River. The aim of the study is to determine the extent of PAH contamination in soils collected along Mosel and Saar River, and to obtain a first insight into the origin of the PAH contamination in this region.

Materials and Methods. In total twenty six sample sites were investigated. Forty two single samples were collected along a 167 km distance of Mosel River and six samples were collected along a 20 km distance of Saar River. Soil samples were collected at a depth of 0 to 2 m with a stainless steel corer (∅ 8 cm). Each 2 m sample was further separated into two sub-samples (0–1 m and 1–2 m). The sixteen EPA PAHs and three additional PAHs (1methylnaphthalene, 2methylnaphthalene and perylene) were analysed with gas chromatography mass spectrometry (GC-MS). For soil characterisation, total organic carbon (TOC), grain size, microscope and X-ray diffraction (XRD) analysis were performed.

Results. Grain size for all soil samples was classified as a mixture of sand and silt. XRD analysis showed that all samples were dominated by quartz. Some clay minerals, such as illite and montmorillonite and feldspars, i.e. anorthoclase and orthoclase, were found in minor quantities. TOC ranged from 0.1% to 13%. Microscope analysis show black coal particles in the majority of the soils collected from the Saar River and part of the Mosel River (after Saar mouth). The black particles were not found further upstream along Mosel River. The sum of nineteen PAHs in the soil samples was up to 81 mg kg^{-1} dry weight (dw). Most soil samples showed a relationship between the presence of coal particles and PAH concentrations.

Discussion. Elevated PAH concentrations were found in all soil samples collected from Saar River and downstream Mosel River. Due to former coal mining activities in the Saarland, Germany, there is a strong evidence that the majority of the PAH contamination in the soils downstream Mosel River are linked to these mining activities. Upstream Mosel River coal particles were hardly found although PAH concentrations were high. Therefore another PAH source has to be responsible for these concentrations. PAH distribution patterns indicate a pyrogenic PAH input upstream Mosel River and a mixed input (petrogenic and pyrogenic) downstream Mosel River.

Conclusions. Due to PAH distribution patterns the contamination along the Mosel River upstream the Saar mouth is probably linked to atmospheric depositions and other sources not linked to coal mining activities. Downstream Mosel River the PAH distribution patterns reflect former coal mining activities. We could corroborate for the first time that coal mining resulted in a serious problem of an extensive PAH contamination at Saar and Mosel River floodplain soils.

Recommendations and Perspectives. Coal mining activities have a strong impact on the neighbouring regions (Johnson and Bustin 2006, Short et al. 1999, Stout et al. 2002). It is known that coals exhibit relative high PAH concentrations, especially in the low molecular weight PAHs (Chapman et al. 1996, Radke et al. 1990). However, PAHs in coals are hardly bioavailable (Chapman et al. 1996) and hence may have less adverse effects on exposed biota. They can act as sink for other hydrophobic contaminants. For the assessment of the environmental impact, a detailed study of the sorption and desorption behaviour of PAHs linked to coal particles should be carried out.

Keywords: Coal mining; coal particles; floodplain soils; Mosel river; PAH; polycyclic aromatic hydrocarbons; Saar river

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Introduction

Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous in the environment and are produced by natural and anthropogenic processes, in which the anthropogenic inputs usually outweigh the natural inputs. PAHs occur in high structural diversity and the hydrocarbon mixtures in different source materials vary in the relative contribution of their individual components. With high Kow values, they have a hydrophobic nature and hence can strongly sorb to non-aquatic phases and accumulate in soil/sediment (Youngblood and Blumer 1975, Neff et al. 2005). United States Environmental Protection Agency (US EPA) has only identified sixteen PAHs as 'priority pollutants'although there are hundreds of PAH compounds, which could be included for risk assessment (Barron and Holder 2003, Neff et al. 2005). In addition, due to the known carcinogenic and mutagenic properties of some PAHs, they are widely studied for potential environmental effects (Loibner et al. 2003, Fang et al. 2004, Neff et al. 2005, Olajire et al. 2005, Hawthorne et al. 2006). The increasing PAH concentrations in the last 100 years have been related to anthropogenic inputs (Fernández et al. 2000). Wild and Jones (1995) estimated that at least 90% of the environmental PAH burden in Great Britain is stored in soil. This estimation excluded contaminated sites like gasworks sites, petroleum refineries, or wood preservation plants.

Wieber (2005) investigated one alluvial soil collected in a rural area along the Mosel River. He found PAH concentrations of up to 25 mg kg^{-1} (Σ 16EPA PAHs) at depths of 0.3 to 0.9 m and up to 90 mg kg^{-1} at depths of 1.80 to 2.00 m. The PAH content exceeded by far the background levels of the sixteen EPA PAHs in Greenland soils in Rhineland Palatinate (2.36 mg kg^{-1} 90 percentile, LABO 2003). Rhineland Palatinate had the highest background levels of PAHs in grassland soils of Germany. PAH values reported from other federal states range from 0.43 mg kg^{-1} to 1.3 mg kg^{-1} (90 percentile).

Because of the presence of black particles (identified as coal particles in this study) in both contaminated soil layers, Wieber (2005) assumed for the first time that the PAH contaminations may be associated with these particles. However, their origin and the extent of the contamination in the region remained unclear.

The aim of this study is (a) to evaluate for the first time the distribution of PAHs in bank and alluvial soils along Mosel and Saar River and (b) to elucidate, if PAH contaminations correlate with soil black (coal) particles or to find out whether the enhanced PAH concentrations in the study area is the result of diffuse or non point source contaminations.

1 Study area and sampling sites

Mosel River is located in the German federal state of Rhineland Palatinate. It rises at the Col de Bussang (Vogues Mountains) in France and joins the Rhine River at Koblenz, Germany after 520 km. Approximately 242 km of the river is located in Germany, with several tributaries flowing into the river, including Saar River. Saar River is an important tributary of Mosel River. It flows through Lorraine in France and the Saarland in Germany, where it joins Mosel River.

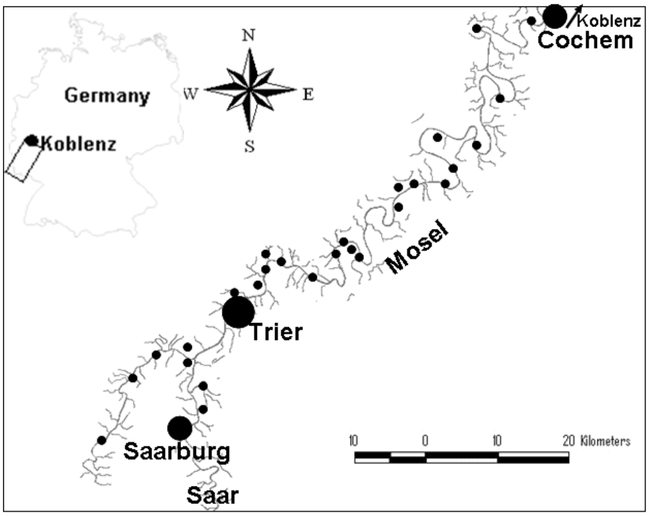

Fig. 1: Map of Mosel River in Germany with the location of the 27 sampling sites (small black dots)

Both regions, Lorraine and Saarland, are well known for coal mining. Coal mining was an important industrial sector in the Saarland. The first coal mined in Saarland can be dated back to the third century B.C. The expansion of coal mining activities since the 16th century resulted in an increase of coal particles transported along Saar River and Mosel River. Floods with water levels rising about 8 to10 m have occurred frequently and spread coal particles onto the floodplains. In addition, human activity along Mosel River has resulted in a number of possible point sources of contamination, including treated wastewater effluents being discharged into the river and surface runoff.

In this study bank soils from twenty-seven sampling sites were investigated, three sites along Saar River and twenty-four sites along Mosel River. Altogether forty-two individual samples were collected along a 167 km distance of Mosel River and six samples were collected along a 20 km distance of Saar River (**Fig. 1**).

2 Material and Methods

2.1 Sieve analysis, microscopy and Total Organic carbon (TOC)

Soil samples were gathered from a depth of 0 to 2 m with a stainless steel corer (∅ 8 cm). Each 2 m sample was further separated into two sub-samples (0 to 1m and 1 to 2 m) and then homogenised. Samples were filled in brown glass bottles and stored at 4°C in the dark. Sieve analysis was performed according to DIN ISO 11277 with the pipette method following Köhn. Different sieve sizes were used: 2 mm, 1 mm, 0.5 mm, 0.25 mm, 0.125 mm and 0.063 mm (Retsch Germany). The samples were dried at 105°C. Each dried sample was investigated with a binocular. Pulverised samples were used for total organic carbon analysis. First, inorganic carbon was removed by adding hydrochloric acid (HCl 16%, p.a. Merck) dropwise until no further reaction was visible. Afterwards the samples were washed with deionised water until a pH value of 6 was reached. After drying at 100°C, 0.1 g of each sample was analysed with a CS elemental analyser (LECO CS 225, St. Joseph, MI).

2.2 PAH analysis

Approximately 25 g to 30 g of wet soil were extracted in four cycles with an accelerated solvent extractor (ASE 300, Dionex). Two cycles were performed with acetone at 100°C and 100 mbar and the other two cycles with toluene at 150°C and 100 mbar. Perdeuterated internal standards including naphthalene d8 (Nap d8), acenaphtene d10 (Ace d10), phenanthrene d10 (Ph d10), fluoranthene d10 (Flu d10), chrysene d12 (Chr d12) and perylene d12 (Per d12) were added to the extraction solvent. Depending on their estimated PAH concentration, samples were concentrated with a rotary evaporator after sample extraction. The solvent was exchanged into cyclohexane afterwards. The perdeuterated substances were used for an internal calibration and adapted to the PAH concentrations in the soils.

PAHs were detected using gas chromatography (HP 6890) equipped with a DB-5MS column (0.25 µm film thickness, 0.25 mm x 30 m) and a mass selective detector in the single ion mode (SIM) (HP 5973). Target ions, qualifier and the retention times are given in **Table 1**.

Table 1: Analysed target ions with qualifier and retention times (Ret.-Time)

PAK	Mass	Qualifier 1	Qualifier 2	Ret.-Time [min]
Nap	128	127	102	11.17
2MNap	142	141		12.91
1MNap	142	141		13.14
Any	152	151		15.07
Ace	153	154		15.50
Fl	166	165		16.78
Ph	178	176		19.08
An	178	152		19.20
Flu	202	200		21.99
Pyr	202	101		22.58
BaA	228	114		26.62
Chr	228	226		26.75
BbF-BkF	252	253		31.20
BeP	252	125		32.08
BaP	252	250		32.28
Per	252	253		32.62
InP	276	138		36.77
DBA	278	139		36.96
BghiP	276	138		37.74

Source temperature was 300°C. The carrier gas was helium with a flow rate of 0.8 ml min^{-1}. Initial temperature of 65°C was held for 4 min, increased to 270°C and held for 10 min, and then finally increased to 310°C and held for 6.5 min. One µl of each sample was injected in splitless mode. All results were calculated on a dry weight (dw) basis. The limit of quantification for individual substances varied from 0.8 to 3.0 µg kg^{-1}. PAH concentrations were determined by using two standard solutions. One standard solution contained the sixteen EPA PAHs (naphthalene, Nap; acenaphtylene, Any; acenaphtene, Ace; fluorene, Fl; phenanthrene, Ph; anthracene, An; fluoranthene, Flu; pyrene, Pyr; benzo(a)anthracene, BaA; chrysene, Chr; benzo(b)fluoranthene – benzo(k)fluoranthene, BbF-BkF; benzo(a)pyrene, BaP; indeno (1,2,3-cd)pyrene, InP; dibenz(a,h)anthracene, DBA; benzo (g,h,i)perylene, BghiP), as well as 1methylnaphtalene, 1Mnap, and 2methylnaphtalene, 2Mnap, which were used to check the presence of the alkyl PAHs. The second solution included the 16 EPA PAHs and benzo(e)pyrene, BeP and perylene, Per. Perylene was used to check terrestrial PAH inputs. Both solutions were prepared by Dr. Ehrenstorfer GmbH (Augsburg, Germany). All solvents were in the trace analysis quality.

3 Results and Discussion

3.1 Soil characteristics

Grain size fractions, TOC, Total Inorganic Carbon (TIC) and the concentrations of the nineteen PAHs detected in the 48 soil samples from Mosel River banks and Saar River banks are listed in **Table 2** (see overleaf). Sieve analysis and fine grain analysis show the majority of the soil consisted of a sand and silt mixture. All soil samples had a similar grain size distribution with a clay content that did not exceed 19%.

The organic carbon content in most samples was around 0.2% to 3% (dw), typically found in floodplain sediments (Gocht et al. 2001). A few samples showed elevated TOC values (4–13%) (see Table 2).

The samples with high TIC values were from areas upstream of where Saar River enters Mosel River, and found in areas dominated by Lacustrine Limestone.

Coal particles were identified by microscopy analysis with a binocular in Saar River and downstream Mosel River. The picture on the right of **Fig. 2** shows a typical sifted sample

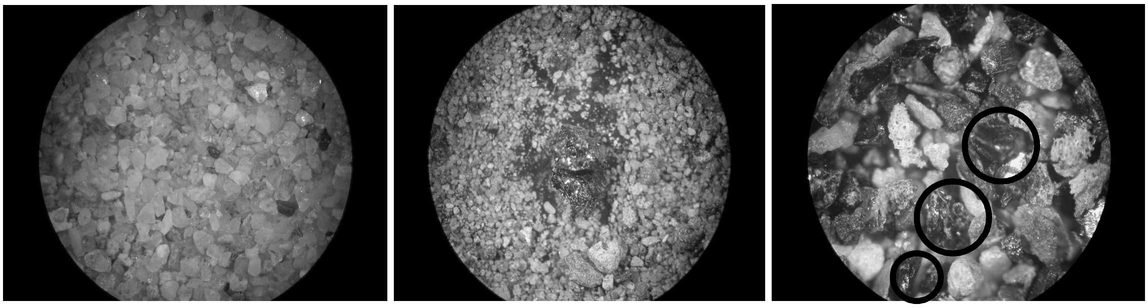

Fig. 2: Left picture: sifted sample with a dominance of quartz grains; centre picture: unsifted sample with a big coal particle in the middle, right picture: sifted sample with coal particles (circles)

Table 2: Soil properties and PAH concentrations of 27 sites (every meter is a 1 m pooled sample, n.d. = not determined)

Sampling Point (in km of the river)	Depth in m	Sand (2 mm–0.063 mm) in %	Silt (0.063 mm–0.002 mm) in %	Clay (<0.002 mm) in%	TOC in %	TIC in %	Σ 16 EPA + 3 PAH in mg kg^{-1}
229	1	63.4	23.5	13.1	0.6	0.5	0.1
229	2	78.5	9.4	12.1	0.2	0.2	0.2
211	1	61.3	30.1	8.7	1.2	3.9	18.1
211	2	56.7	25.9	17.4	0.6	2.6	1.6
204.5	1	65.6	24.1	10.3	0.8	1.8	5.1
204.5	2	71.3	25.0	3.7	0.2	7.2	0.3
0 (Saar)	1	78.8	14.6	6.6	13.3	0.3	72.6
0 (Saar)	2	n.d.	n.d.	n.d.	9.7	1.3	81.5
4 (Saar loop)	1	86.1	9.1	4.9	n.d	n.d	26.3
4 (Saar loop)	2	80.4	10.9	8.7	1.2	0.0	3.2
9 (Saar)	1	89.7	7.2	3.2	5.3	0.3	12.1
9 (Saar)	2	83.0	11.1	5.9	6.0	0.7	20.9
202	1	68.5	22.0	9.5	0.4	0.3	1.6
202	2	67.0	20.5	12.6	0.3	0.1	0.2
188	1	55.0	33.2	11.8	0.5	0.8	0.1
188	2	59.5	29.1	11.5	0.5	0.2	0.0
186	1	74.1	19.4	6.5	2.8	1.8	10.5
186	2	59.3	32.0	8.7	0.3	1.2	0.1
183	1	61.7	27.6	10.7	3.8	4.0	37.4
183	2	48.9	40.7	10.4	0.4	1.3	1.0
180	1	73.7	18.5	7.8	0.1	0.1	0.1
180	2	68.6	17.1	14.3	0.0	0.1	0.0
173	1	73.0	17.2	9.8	6.8	0.8	50.4
173	2	n.d.	n.d.	n.d.	11.9.	1.1	81.0
165.5	1	78.7	19.0	2.3	2.5	0.8	8.8
165.5	2	n.d.	n.d.	n.d.	0.2	0.9	0.1
162.5	1	62.2	29.0	8.8	6.9	0.5	17.3
162.5	2	67.3	25.0	7.7	0.8	0.9	2.7
160.5	1	n.d.	n.d.	n.d.	5.0	0.6	9.0
160.5	2	87.4	10.1	2.6	11.0	0.7	53.7
158.5	1	74.1	20.4	5.5	8.0	0.4	59.5
141	1	75.7	19.2	5.1	5.6	4.6	24.8
138	1	n.d.	n.d	n.d	2.5	2.7	14.4
135.5	1	68.7	24.1	7.2	4.8	1.3	13.1
135.5	2	55.9	35.5	8.6	0.3	0.9	0.2
132	1	67.9	26.0	6.2	0.8	2.5	4.3
132	2	70.3	23.4	6.2	0.1	0.8	0.4
126	1	65.0	24.0	11.0	0.3	0.8	0.4
126	2	69.7	22.4	7.9	0.3	0.6	0.1
118	1	70.6	21.5	7.9	4.0	2.9	42.8
118	2	51.8	36.0	12.2	3.0	2.6	24.5
109	1	53.3	32.4	14.4	1.1	1.8	3.7
109	2	43.8	38.0	18.1	0.1	0.3	0.1
88.5	1	78.4	18.1	3.5	3.2	1.0	21.3
88.5	2	49.8	34.9	15.3	2.2	2.5	6.9
74.5	1	80.1	16.2	3.7	6.6	0.8	27.6
62	1	60.4	32.7	6.9	0.2	0.8	1.3
62	2	51.7	39.4	8.9	0.3	0.3	0.8

upstream Mosel River with mainly quartz grains. The picture in the middle and on the left of Fig. 2 shows black, gleaming particles, identified as coal particles between the quartz grains and other rock fragments in the sifted samples.

3.2 PAH distribution along Mosel and Saar River

Most of the bank soils from Saar and Mosel showed elevated PAH concentrations. The concentrations of the sixteen EPA PAHs and the three individual PAHs range from non-detectable to 81 mg kg^{-1} (dw) (**Table 3**). There was no trend in the distribution of the PAH concentrations along Mosel/Saar River (see Table 3). Most of the elevated concentrations were in the first meter of the soil, except at four sample sites. The soils collected from these four sites showed the highest concentrations in the second meter. No point-sources were identified. The PAH concentrations of 11 sample sites exceeded the German guideline value of 20 mg kg^{-1} (LAGA 1998) (**Fig. 3**).

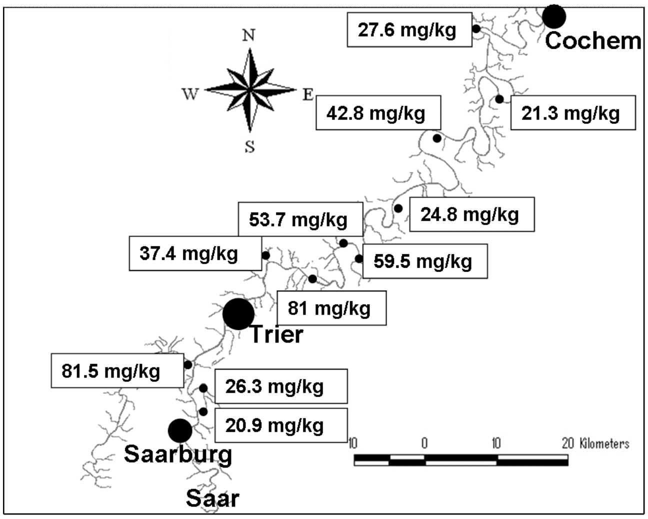

Fig. 3: 11 sampling sites with high PAH concentrations

The PAH distribution pattern of the samples is shown in **Fig. 4** (see overleaf). For the graphs medians with the standard deviations of the PAH concentrations normalised to the total concentration from samples upstream Mosel River (graph A), samples from Saar River (graph B) and samples downstream Mosel River (graph C) were calculated. Due to a higher uncertainty samples with concentrations < 0.2 mg kg^{-1} in the sum of 19 PAHs were not taken into account. The PAH distribution patterns of soil samples at the Saar and downstream Mosel River are similar. They showed high amounts of Nap and four to six ring PAHs, whereas Flu and BbF-BkF were the dominating substances. Ph, Pyr, BaP, InP and BghiP were elevated too. In contrast, upstream Mosel River lower amounts of Nap were determined. The distribution pattern of the higher molecular weight PAHs was similar for all samples.

Heavy weight PAHs are mainly formed by incomplete combustion processes of organic materials (Fernandes et al. 1997). Four to six ring PAHs are primary bound to particles like soot and engine exhaust and substituted PAHs are of minor significance (Sporstöl et al. 1983, Stout et al. 2002). Typical combustion PAHs (pyrogenic) are Flu, Py, InP and BghiP (Fernandes et al. 1997, Page et al. 1996). Lower weight PAHs and substituted PAHs are typical components of petroleum and coals (petrogenic) (Sporstöl et al. 1983, Vliex 1994, Stout et al. 2002). Vliex (1994) showed a high concentration of naphthalenes and phenanthrenes in Saar coals. This is in accordance with the soil samples from Saar River and downstream Mosel River of this study (see Fig. 4). In contrast, samples upstream Mosel River exhibited a pyrogenic distribution pattern, i.e. an atmospheric input. The soil samples from Saar River and downstream Mosel River showed a mixed pattern (pyrogenic and petrogenic). The pyrogenic part of the pattern could arise from atmospheric transported coal derived particles from coal industry, traffic emissions and other combustion processes.

Table 3: Comparison of PAHs in bank soils of Mosel and Saar River with different soils, sediments and suspended particulate matter from areas worldwide

Sites	Amount	Range in mg kg^{-1}	Reference
Semirural, agricultural, rural, and forest soils, pasture grassland (all over Great Britain)	49	0.11–54.5	Jones et al. (1989)
Semirural, agricultural soils (southeast England)	9	0.11–1.8	Jones et al. (1989)
Rural, urban and industrial soils (Estonia)	170	0.05–22.2	Trapido et al. (1999)
Rural and urban soils (Hongkong, China)	53	0.03–0.17	Zhang et al. (2006)
Pasture grassland and urban soils (Switzerland)	23	0.05–0.6	Bucheli et al. (2004)
Alluvial soil (Rhine River, Germany)	18	0.02–3.6	Gocht et al. (2001)
Rural and suburban soils (Beijing, China)	47	0.02–3.9	Ma et al. (2005)
Sediments (Gironde estuary, France)	31	0.02–4.9	Budzinski et al. (1997)
Suspended particulate matter (Seine River and estuary, France)	25	1–14	Fernandes et al. (1997)
Suspended particulate matter (Xijiang River, China)	12	0.04–0.67	Deng et al. (2006)
Sediments (Yangtze Estuary, China)	14	0.3–5.5	Liu et al. (2001)
Suspended particulate matter (Elbe River, Germany)	1 year monitoring program	0.1–1.3	ARGE Elbe (2001)
Bank soils (Mosel and Saar River, Germany)	42	0.1–81.5	This study

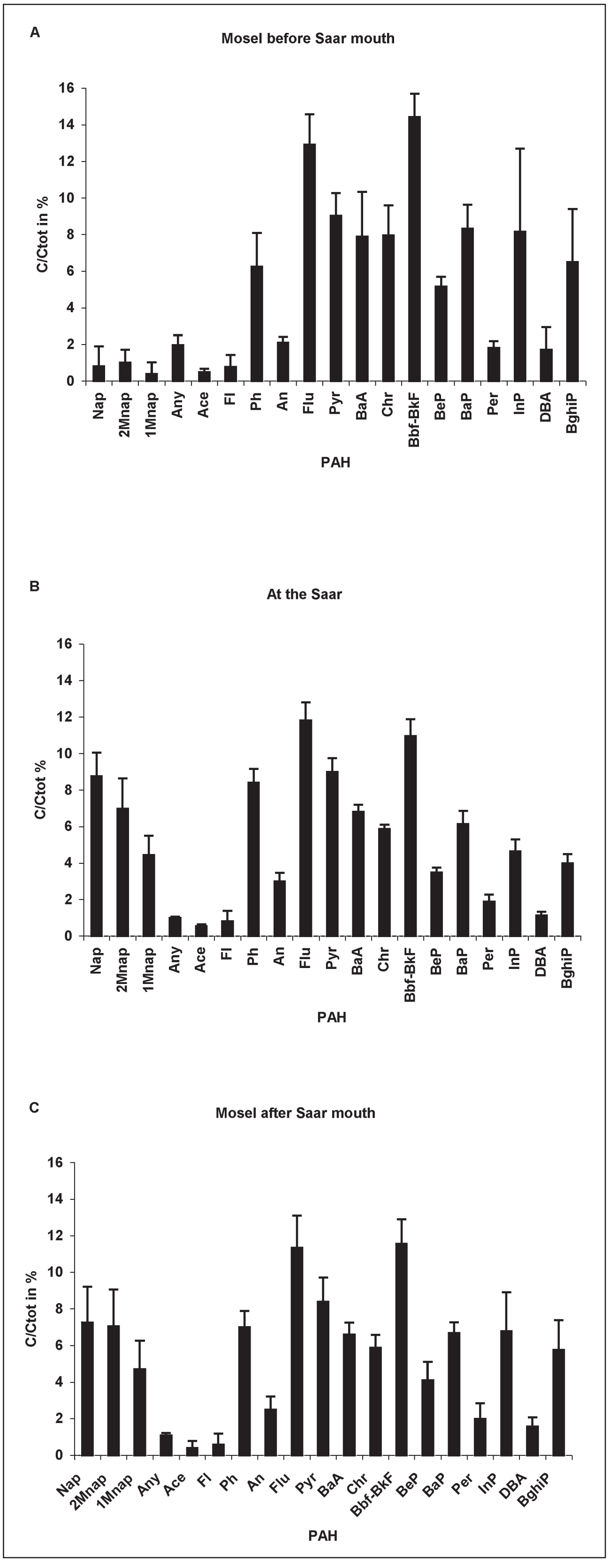

Fig. 4: PAH distribution patterns of the medians of the normalised (individual concentration to the total concentration) PAHs with A: soil samples of Mosel before Saar mouth, B: soil samples of Saar and C: soil samples of Mosel after Saar mouth

PAH concentrations investigated in this study are generally higher compared to other study areas (see Table 3), especially alluvial soils at River Rhine investigated by Gocht et al. (2001).

4 Conclusions

The investigations of this study show for the first time large scale PAH contamination in floodplain soils collected from Mosel and Saar River. Twenty six sampling sites were investigated; eleven sampling sites show elevated PAH concentrations up to 81 mg kg^{-1}. In comparison to other regions in the world these concentrations are relatively high.

In addition, for the first time coal particles were identified in most of the soil samples along Mosel and Saar River. Most of the soils with coal particles showed elevated PAH concentrations. Coal can be a source of PAHs, but also a sink due to its high sorption capacity (Barrick et al. 1984, Haenel, 1992).

Although the discharge of mine water is decreasing and less coal mining is taking place in the area (IKSMS 2005), contamination from previous mining activities is still evident by the presence of coal particles in the soils. Beside this petrogenic PAHs, the distribution patterns indicated an additional pyrogenic source downstream Mosel River and at Saar River. Upstream Mosel River only a pyrogenic source can be identified. This pyrogenic input could originate from coal industry (i.e. coal derived particles transported by atmosphere) as well as traffic emissions and combustions processes.

5 Recommendations and Perspectives

Coal mining activities have a strong impact on the neighbouring regions. (Johnson and Bustin 2006, Short et al. 1999, Stout et al. 2002). It is known that coals exhibit relative high PAH concentrations, especially of low molecular weight PAHs (Chapman et al. 1996, Radke et al. 1990). However, PAHs in coals are hardly bioavailable (Chapman et al. 1996) and hence may have less adverse effects on exposed biota. They can act as sink for other hydrophobic contaminants. Risk assessment of complex environmental samples is difficult due to the identification of the toxic components, the lack of available toxicity data and the scarcity about the knowledge of the behaviour of genotoxic substances in complex mixtures (White 2002). These difficulties get often avoided assuming that the toxicity of a mixture is the sum of the expected effects from each mixture component (EPA 1989). Some substances with high concentrations in this study (i.e. BaA, BbF-BkF as individuum, BaP, InP, DBA and BghiP) are known to have a sufficient evidence of carcinogenicity in experimental animals (IARC 1983). Hence, high contaminated soil samples in the study area are expected to have a high toxic potential. A detailed study of the sorption, desorption, and bioavailability of the PAHs linked to coal particles should be carried out.

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