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Spatial Differentiation of Ecosystem Risks of Soil Pollution in Floodplain Areas of the Czech Republic

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Abstract

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Extensive soil sampling and screening assessment of ecosystem risks combined with a multidimensional statistical analysis were used to estimate and spatially characterize the ecosystem risks stemming from the contamination of floodplain soils in the Czech Republic. We proved structural differences in regional pollution patterns where different regional/local pollution sources led to various nature and extent of the environmental load of floodplain soils. The established spatial patterns helped reveal the areas where soils do not meet soil quality standards and where the ecosystem risks were elevated. Furthermore, the results allowed to establish priority contaminants of flood affected areas in various catchments in the Czech Republic. Combining both the magnitudes of estimated ecosystem risks and structural characteristics of pollution profiles, the highest estimated risks showed the localities with high contents of organochlorine pesticides, several samples connected to geochemical anomalies of metallogenic zones (deposits), and localities with a long history of industrial load. Since extreme weather events have recently become more frequent, our results highlight the importance of a continual monitoring of pollutant turnover in floodplain soils with a high flood frequency.

Keywords: floods; persistent organic pollutants; soil contamination; spatial patterns; trace elements

In recent years, extreme weather events have become still more frequent and intense (Beniston & Stephenson 2004). As for the Czech Republic, intensive floods caused significant damages in last two decades. A high and long term anthropogenic alteration of floodplains in the consequence of a high population density (Tockner & Standford 2002) resulted in enhanced anthropogenic pressures on fluvial ecosystem. Although the quality of surface waters in Czech rivers underwent significant changes during the 1990s due to a decline in industrial and agricultural activity associated with political changes (Langhammer *et al.* 2010), increased concentrations of various pollutants can still be found in alluvial soils in Central Europe (Podlešáková *et al.* 1994; Stachel *et al.* 2003). As for the floodplains in the Czech Republic, soil contamination in the alluvial zones of the sites strongly affected by mining and metallurgy has been studied (Borůvka *et al.* 1996; Ettler *et al.* 2006; Žák *et al.* 2009). Regional studies attempted to describe the contamination situation in the Elbe catchment (Podlešáková *et al.* 1994; Heinisch *et al.* 2007) or in the Morava River basin (Grygar *et al.* 2012; Nováková *et al.* 2013) and especially the flood impacts on soil contamination in case studies (Vácha *et al.* 2003; Elhottová *et al.* 2006; Hilscherová *et al.* 2007; Pulkrabová *el al.* 2008). Since our knowledge regarding magnitudes and pollution profiles of floodplain contamination in the Czech Republic is still fragmentary, a rigorous analysis of soil contamination was conducted by an extensive soil sampling and screening assessment of ecosystem risks combined with a multidimensional statistical analysis. The study has provided

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a comprehensive spatial analysis of ecosystem risks of soil contamination in cultivated floodplains in the Czech Republic with the objectives of revealing both the spatial patterns of pollution magnitude as well as the composition of pollutants.

MATERIAL AND METHODS

Soil sampling. Since we endeavoured to characterize pollution profiles of fertile soils and potential ecological threats in cultivated floodplains, the sampling sites (n = 100) were preferably located in the areas where a high flood frequency has met a high agriculture intensity. An extensive spatial analysis of land use and soil diversity in Czech floodplains (Skála *et al.* 2013) had forgone field work so that the target sampling areas could be established using a spatial overlap of the 5-year flood inundation areas from the DIBAVOD geo-database (Digital database of water management data) and agricultural areas from the digital Land Parcel Identification System (LPIS). For each sampling site, a set of 10 individual probes was collected from an area of 1 ha using handheld auger (samples were homogenized by quartation). The sample depth was 0–10 cm for pastures and 0–30 cm for arable land. The soil samples were analyzed for a wide range of pollutants (Table 1) and basic soil properties (e.g. total organic carbon, soil texture characteristics).

Laboratory analysis. Analyses for trace elements (TEs) and soil properties were performed in the Research Institute for Soil and Water Conservation, Prague. Trace elements were determined using the aqua regia digestion (ISO 11466 1995) followed by atomic absorption spectrometry in flame (VARIAN FAAS 240; Agilent Technologies, Inc., Mulgrave, Australia – Cu, Ni, Pb, Zn), atomic absorption spectrometry with electrothermal atomization (VARIAN ETA 240Z; Varian, Mulgrave, Australia – Cd) and hydride generation mode (As). The total Hg content was assessed using the AMA method (AMA254). The analytical results underwent rigorous quality assurance procedures – analytical replicates with a relative standard deviation tolerance lower than 5%, the sample spiking and certified reference materials analyses (RM 7001, 7003). Chemical measurements for persistent organic pollutants (POPs) were performed in the Laboratory of trace analyses in the Research Centre for Toxic Compounds in the Environment, Brno. Contents of POPs were determined by gas chromatography–mass spectrometry (GC-MS) (an Agilent 7890 GC coupled to an Agilent 7000B Series Triple Quadrupole GC/MS System; Agilent Technologies Inc., Wilmington, USA) after dichloromethane extraction of samples spiked with recovery standards and clean up. Certified reference materials (Cambridge Isotope Labs soil standard RM-0002) and laboratory blanks were analyzed with each set of POPs samples.

Estimation of ecosystem risks. A screening risk assessment of soil contamination was accomplished using the Dutch method from the National Institute for Public Health and the Environment (Lijzen *et al.*

Table 1. List of pollutants determined in soil samples and their serious risk concentration (SRC) values

Pollutant	SRC (mg/kg)
Trace elements	
As	85
Cd	13
Cu	96
Hg	36
Ni	100
Pb	580
Zn	350
Organochlorines	
ΣPCBs (7)	3.4
PeCB	16
HCB	2.0
α-HCH	17
β-HCH	13
Lindane	1.2
o,p'-DDE, p,p'-DDE	1.3
o,p'-DDD, p,p'-DDD	34
o,p'-DDT, p,p'DDT	1.0
Polycyclic aromatic hydrocarbons	
Naphthalene	17
Phenanthrene	31
Anthracene	1.6
Fluoranthene	260
Benzo(a)anthracene	2.5
Chrysene	35
Benzo(k)fluoranthene	38
Benzo(a)pyrene	7.0
Indeno(123-cd)pyrene	1.9
Benzo(ghi)perylene	33

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2001; Van Vlaardingen *et al.* 2007) . The method is based on the estimation of potential ecological risks by a confrontation of environmental concentrations (PEC) with referenced values for potential ecosystem risks of soil contamination (serious risk concentration – SRC) (Eq.1). The SRC values (Table 1) are simultaneously derived using all reliable toxicity data reflecting both acute and chronic toxicity for species representing different trophic levels as well as using data for microbial processes and enzymatic reactions, where the final choice is most frequently the lowest value representing the most sensitive ecological response (RIVM 2007). The SRC values provide the effect base level where some ecotoxicological effects may be expected.

$$\mathrm{RISK_{ECO}} = \frac{\mathrm{PEC}_i}{\mathrm{SRC}_i} \tag{1}$$

A resultant ecological hazard index for n pollutants at each locality ($\mathrm{HI_{ECO}}$ in Eq. (2)) is then calculated as a sum of quotients calculated for each i pollutant in Eq. (1) and poses an estimation of potential ecosystem risks (ERs).

$$\mathrm{HI_{ECO}} = \Sigma_{i=1}^{n}\mathrm{RISK_{ECO}} \tag{2}$$

In the last step (Eq. (3)), we calculated relative contributions of each element/substance to the overall hazard index ($\mathrm{HI_{ECO}}$).

$$p_{i\mathrm{RISK_{ECO}}} = \frac{\mathrm{RISK}_{\mathrm{ECO}i}}{\mathrm{HI_{ECO}}} \tag{3}$$

Regionalization of ecosystem risks. Regional patterns of ERs were assessed by a hierarchical cluster analysis (HCA) using an average linkage clustering. A matrix transformation of relative contributions of each analyte to total $\mathrm{HI_{ECO}}$ in each sample was performed prior to the HCA. The dataset was transformed by a range transformation after the matrix had been constant-row-sum normalized to generate homogeneity of variance among all variables according to Miesch (1976). The constant-row-sum transformation can reduce the influence of samples with high proportions, while the range transformation reduces the influence of determinants with a high variability. An optimal number of clusters was determined by the Mantel test as an algebraic equivalent of the Pearson's correlation between the values in the original distance matrix and binary matrices computed from the dendrogram cut at various levels (Legendre & Legendre 1998). The Kruskal-Wallis test was used to verify significant differences in pollutant concentrations by the cluster grouping. All the processes were performed using R programme (R Core Team 2012). The results were spatially visualized using ArcGIS 10.2 (ESRI, Redlands, USA).

RESULTS AND DISCUSSION

Regional patterns of soil pollution profiles. Cluster analysis was processed to verify whether soil pollution composition tends to form regional patterns in the dataset. The HCA results proved a satisfactory cophenetic correlation coefficient (r = 0.8) with the optimal number of 9 clusters (dendrogram in Figure 1). Basic statistics for main pollutants (Table 2) and average relative contributions of pollutants to total estimations of ERs (Figure 2) were calculated after the dataset partitioning in the HCA.

The most substantial cluster was formed of localities characterized by high contributions of TEs (Ni ~ Zn > Hg ~ Cd ~As) together with elevated contributions of industrial POPs (PCBs, HCB). Lower contributions of pesticides (especially lindane) and individual PAHs were also characteristic for this cluster. The cluster constituents reached lower or average magnitudes of estimated ERs (Figure 4). Searching for a spatial scatter of the cluster, it can be concluded that member localities can be found in all major Czech river systems (Figure 3). Cluster 1 can be considered as a characteristic profile of an overall fluvial pollution in the dataset.

Cluster 2 can be characterized by high contributions of PAHs compounds to estimated ERs. The increased PAHs contents were evident in fluvial systems of the broader North Moravian region (in the Odra and Bečva River basins). In North Moravia, manifold anthropogenic pollution sources (coal processing, metallurgy) have generated higher Cd and PAHs contamination of agricultural soils (Vácha *et al.* 2015). The elevated PAHs contents were also recorded along the Jizera River past the industrial centre of Mladá Boleslav. Impacts of local industrial enterprises in the Jizera floodplain were proved in a complex geochemical study of TEs enrichment in the floodplain sedimentary fill by Grygar *et al.* (2013). High PAHs contributions together with above-average values of $\mathrm{HI_{ECO}}$ were surprisingly found in the upper courses of the Elbe and the Morava Rivers. This could only be explained by a high propensity of PAHs to atmospheric transport that may result in high concentrations of

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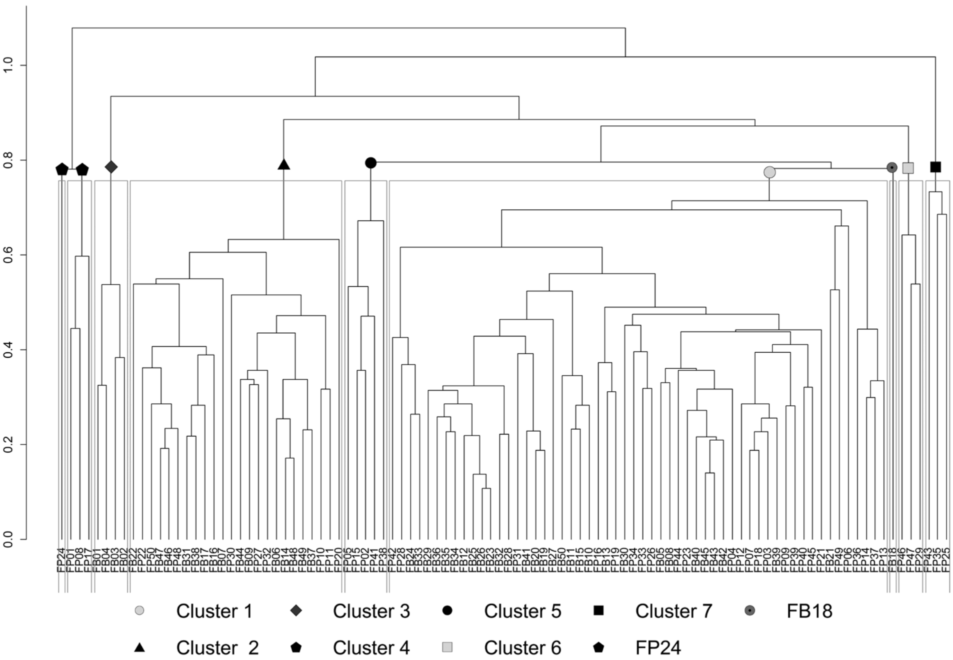

Figure 1. Dendrogram characterizing the partitioning of sampling localities according to relative contributions of pollutants to estimation of ecosystem risks (ERs)

airborne POPs in remote and unpolluted freshwater ecosystems (Grimalt *et al.* 2004).

Cluster 3 involved a toposequence of 4 soil samples near the confluence of the Morava and the Dřevnice Rivers. There was a series of higher concentrations of organochlorine pesticides (especially HCHs and PeCB) in this region (Figure 2a). Studying POPs dynamics in the dissolved phase in the water column of the Morava and the Dřevnice Rivers, Prokeš *et al.* (2012) found the highest concentrations of organochlorine hydrocarbons (especially PCBs, HCHs) at the Dřevnice sampling site.

There were higher contributions of organochlorine pesticides (DDT and its metabolites) to estimated

Table 2. Medians for main pollutants after the data partitioning in hierarchical cluster analysis (HCA) and the results of Kruskal-Wallis (K-W) test for significant differences on pollutant concentrations by the cluster grouping

Sample (n)	ΣPCB	PeCB	HCB	ΣHCH	ΣDDX	ΣPAHs	As	Cd	Cu	Hg	Ni	Pb	Zn
	(μg/kg)						(mg/kg)						
Dataset (100)	1.32	0.12	1.21	1.58	9.7	362	8.8	0.38	24.3	0.11	25.6	23.7	96.6
Cluster 1 (56)	1.21	0.12	1.21	1.58	9.2	249	8.5	0.31	23.2	0.11	25.7	22.1	94.0
Cluster 2 (24)	2.47	0.13	1.27	1.64	3.8	1 748	8.2	0.40	24.8	0.11	25.1	23.4	93.4
Cluster 3 (4)	1.59	1.52	2.26	29.50	10.1	755	8.5	0.34	18.8	0.07	28.2	17.9	67.9
Cluster 4 (3)	1.72	0.14	0.60	2.94	715	404	12.7	0.95	37.5	0.13	27.2	40.1	128
Cluster 5 (5)	1.12	0.10	1.48	1.48	72.0	262	16.3	1.50	106	0.12	25.8	47.6	188
Cluster 6 (3)	1.16	0.07	2.47	1.71	8.3	400	9.8	0.55	35.5	0.25	23.7	516	180
Cluster 7 (3)	1.13	0.04	0.96	1.58	14.5	151	35.4	0.23	19.5	0.08	20.1	16.2	76.1
FB18	0.70	0.10	1.12	0.82	157	147	9.0	0.29	20.2	0.07	69.9	16.5	79.6
FP24	3.16	0.16	0.92	1.26	3 419	434	3.0	0.74	28.8	0.15	24.9	45.9	136
K-W test	–	*	–	*	**	***	**	**	**	–	–	**	*

– not significant; *, **, ***α = 0.05, 0.01 and 0.001, respectively

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ERs characteristic for Cluster 4. The cluster was spatially confined to the catchment of the Berounka River and to the lower reach of the Ohře River near the confluence with the Elbe River. Enhanced Cd and Cu contributions to ERs estimation followed the organochlorine pesticides dominance in this cluster. Pollution patterns observed in the cluster may refer to a prevalence of agricultural activity in those areas.

Cluster 5 can be characterized as a complex pollution profile where various pollutants contributed to estimated ERs and involved most of the sampling sites along the Ohře River. High TEs contributions (especially of Cd, As, Cu) may be attributed to a spatial interaction of various pollution sources in the North Bohemian region (long-term airborne pollution due to combustion of fossil fuel and chemical industry, geochemical anomalies of metallogenic zones) (Vácha *et al.* 2015). Higher TEs contents are accompanied by higher contents of various POPs of industrial as well as agricultural origin (HCB, HCHs, DDT, and metabolites). Similar ecosystem contamination caused by anthropogenic sources was reported by Kohušová *et al.* (2011) in the adjacent watershed of the Bílina River.

In Cluster 6, dominant Pb and Zn contributions to estimated ERs were reported and there prevailed localities with soils contaminated by mining- and smelting-related pollutants in historical Pb-Zn-Ag mining districts of the Příbram and Stříbro deposits. The previous studies showed a strong effect of the metal works on the sediment contamination from

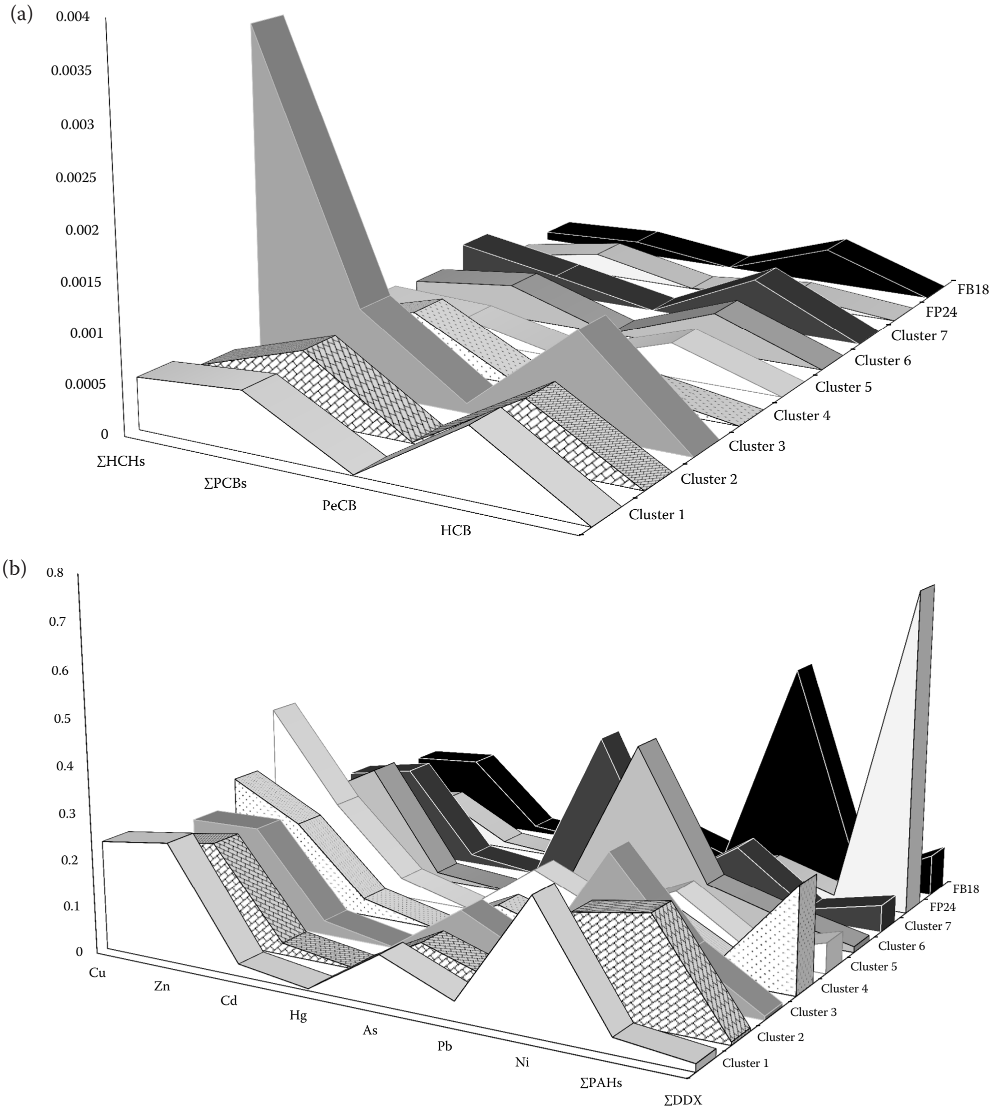

Figure 2. Average relative contribution of individual pollutants (groups) to the total estimation of ecosystem risks (ERs) for individual clusters: (a), (b) – major and minor pollutant groups, respectively

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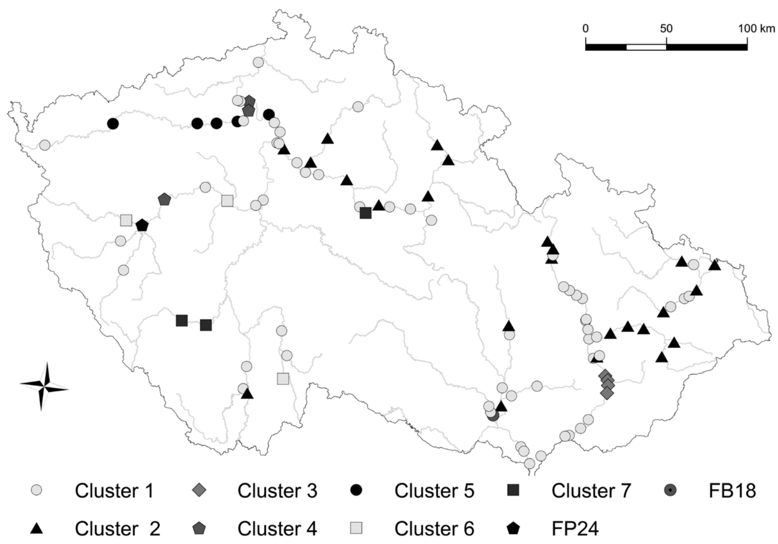

Figure 3. Spatial regionalization of the cluster analysis results

the Litavka stream (Borůvka *et al.* 1996; Ettler *et al.* 2006; Žák *et al.* 2009) as well as on the overall contamination of flood sediments of the Berounka River past the Litavka stream inflow (Navrátil *et al.* 2008). The enhanced Pb concentration was also recorded in the sample from a wet meadow in a foreland of the Rožmberk fishpond. The high Pb content trapped by the fluvic Gleysol may correspond to organic matter binding (C_{org} = 8.5) and might be connected with the regional lead glassworks (Veselý 1994). Further work incorporating broader analysis with a more detailed sampling programme would be needed to verify the lead origin.

Cluster 7 involved 3 samples that are featured by high contributions of As to estimated ERs. One sample was located in a floodplain of the Klejnárka stream draining the Kutná Hora ore region renowned for its high contamination by TEs (especially As, Zn) (Horák & Hejcman 2013). The predominant As contribution to estimated ERs was accompanied by elevated contributions of Cu and Zn in the sample. Other two samples were derived from the floodplains of the Otava River where the higher As contributions reflected the influence of the geological situation as well as an abundance of gold deposits with a high accessory As content (Fillipi *et al.* 2004; Skála *et al.* 2011).

There were reported two singular localities in the dendrogram cut at the optimum level. The first locality (No. FB18) may be considered as an outstanding branch of Cluster 1 (the dendrogram in Figure 1). A

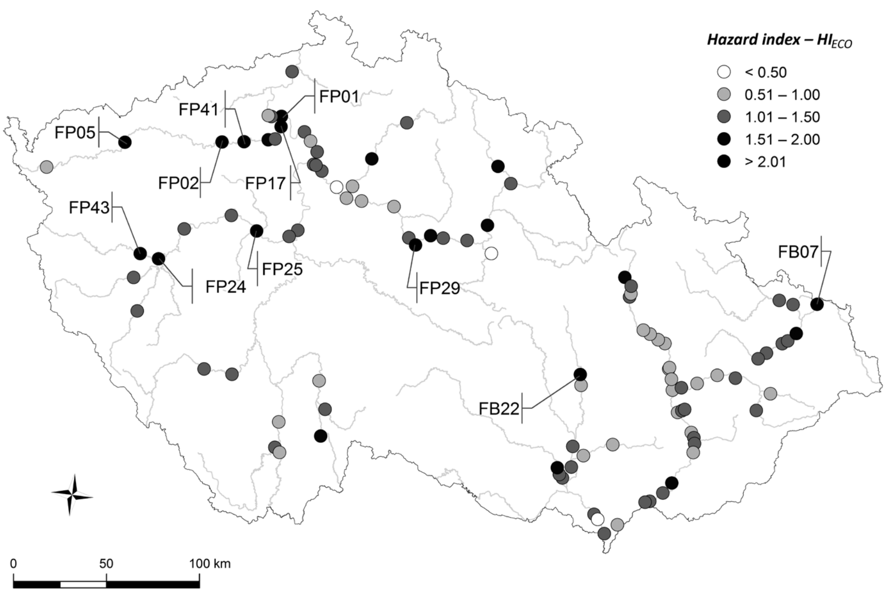

Figure 4. Spatial differentiation of ecological hazard index (HI_{ECO}) magnitude (ecosystem risks estimation) and regional hot spots (HI_{ECO} > 2.0)

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Table 3. Priority pollutants for samples with topmost estimation of ecosystem risks (HI_{ECO} > 2.0) – their concentrations and relative contribution to HI_{ECO}

Sample	Cluster	HI_{ECO}	Priority pollutants (relative contribution) [measured concentration; mg/kg]		
			1	2	3
FB22	2	3.3	ΣPAHs (0.66) [14.6]	Cu (0.12) [36.9]	Ni(0.10) [33.2]
FB07	2	2.5	ΣPAHs (0.40) [7.3]	Zn (0.23) [209]	Cu(0.13) [31.6]
FP17	4	2.6	Cu (0.42) [104]	ΣDDX (0.24) [0.72]	Zn (0.14) [128]
FP01	4	2.5	ΣDDX (0.28) [0.82]	Zn (0.20) [177]	Cu (0.15) [37.5]
FP05	5	3.0	Cu (0.55) [157]	Zn (0.18) [188]	Ni (0.08) [23.7]
FP02	5	2.7	Cu (0.40) [106]	Zn (0.22) [208]	As (0.14) [33.6]
FP41	5	3.5	Cu (0.34) [116]	Zn (0.16) [201]	ΣDDX (0.15) [0.59]
FP43	6	3.5	Zn (0.36) [438]	Pb (0.33) [680]	Cu (0.11) [35.5]
FP25	6	2.5	Pb (0.35) [516]	Zn (0.20) [180]	Cu (0.19) [44.9]
FP29	7	2.6	As (0.37) [82.9]	Zn (0.27) [246]	Cu (0.18) [45.8]
FP24	–	4.1	ΣDDX (0.72) [3.4]	Zn (0.10) [136]	Cu (0.07) [28.8]

strong influence of high Ni content and DDTs content and low contributions of industrial pollutants (PCB, HCB, PAHs) distinguished it from Cluster 1 (Figure 2a). The higher Ni content may be associated with its geochemical position in regional ultrabasic rocks (Quantin *et al.* 2008). Similarly, the singular locality FP24 is spatially as well as structurally connected to Cluster 5 because an exceptional predominance of DDT and its metabolites was proved in this sample from the Berounka River.

Regional differentiation of the estimated ecological hazard index. Since magnitudes of the ecological hazard index were calculated (Eq. (2)) and cartographically represented (Figure 4) for all sampling sites, regional hot spots of potential ecological impacts of complex soil pollution can be determined. Higher values of estimated HI_{ECO} were proved for the samples collected along the Ohře River (especially at its lower course). An increase of HI_{ECO} was recorded for the Elbe River past the Pardubice centre and floodplains adjacent to the Neratovice site confirming spatial pollution patterns in various environmental compartments of the Elbe River basin reported by Podlešáková *et al.* (1994), Heinisch *et al.* (2007), Randák *et al.* (2009), and Kolaříková *et al.* (2012). Higher magnitudes of HI_{ECO} were recorded for the whole length of the Berounka River with its increase in a consequence of some hot spots (the Litavka stream inflow or Ag-Pb-Zn deposit in Stříbro). Similarly, the elevated HI_{ECO} followed the Odra River with a regional increase near the Ostrava agglomeration. Several local contamination abnormalities were detected in a consequence of spatially confined pollution sources (Kutná Hora deposit, industrial centre of Mladá Boleslav or the Svitava River near Boskovice).

Relative pollution profiles for the localities with elevated values of hazard index (HI_{ECO} > 2.0) are reported in Table 3. When combining both the magnitudes of estimated ERs and constituent compositions of pollution profiles, 11 samples (from 6 clusters) are to be explained. Three samples (FP01, FP17, FP24) can be characterized by high contributions of organochlorine pesticides (especially DDT and its metabolites) to total ERs. A high dominance of some TEs (As, Cd, Pb, Zn) featured several samples which are spatially associated with geochemical anomalies of metallogenic zones and deposits (FP24, FP29, FP43). A pair of samples (FB07, FB22) with priority pollutants from PAHs group also exhibited elevated $HI_{ECO.}$ Last but not least, the samples from the Ohře catchment with a more complex anthropogenic load (As, Cu, Cd, Zn, HCB, DDT) reached an upper limit of HI_{ECO} variability.

CONCLUSION

The general pollution profile of flood affected areas in the Czech Republic and several regional pollution abnormalities with different pollution characteristics were defined. Regional hot spots of the potential ecosystem impacts of complex soil pollution were estimated using the screening risk assessment of soil contamination. When combining

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both the magnitudes of estimated ERs and structural characteristics of pollution profiles, the highest estimated ERs were associated with several localities characterized by enhanced contents of organochlorine pesticides, samples connected to geochemical anomalies of metallogenic zones (deposits), and samples with a higher anthropogenic load (airborne PAHs and TEs contamination). This suggests that the soil contamination (magnitude, pollutant associations) of Czech floodplain soils followed the character of local pollution sources (geochemical anomalies) as well as regional diffuse contamination sources (organochlorine pesticides, PAHs). The screening risk assessment of sample contamination estimated only potential ecotoxicological effects for the most vulnerable trophic levels for particular pollutants. A high probability of ecological impacts of soil pollution may be expected at several sampling sites however full consequences and details of ecotoxicological effects are beyond the scope of this paper. Obviously there is a need for the present results coupling with our consecutive results of ecotoxicity testing using various bioassays similarly to Vašíčková *et al.* (2013).

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