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PII: S0375-6742(17)30409-0
DOI: doi: 10.1016/j.gexplo.2017.06.008
Reference: GEXPLO 5933

To appear in: *Journal of Geochemical Exploration*

Received date: 25 January 2016
Revised date: 1 September 2016
Accepted date: 12 June 2017

Please cite this article as: Sanja Sakan, Bojana Ostojić, Dragana Đorđević , Persistent organic pollutants (POPs) in sediments from river and artificial lakes in Serbia, *Journal of Geochemical Exploration* (2017), doi: 10.1016/j.gexplo.2017.06.008

Persistent organic pollutants (POPs) in sediments from river and artificial lakes in Serbia

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Abstract

In an attempt to evaluate the toxicological condition of the most important river and artificial lakes in Serbia, 52 grab-samples were collected from different locations. Sediment samples were analyzed for PCBs (PCB 28, PCB 52, PCB 101, PCB 138, PCB 153, PCB 180, and PCB 194), organochlorine pesticides (*α*-HCH, *β*-HCH, *γ*-HCH, benzene hexachloride, heptachlor, heptachlor epoxide, aldrin, endrin, dieldrin, DDE, *p,p'*-DDD, *p,p'*-DDT, *o,p'*-DDT, and methoxychlor), triazine herbicides (atrazine, simazine, and propazine) and PAHs (fluoranthene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, benzo [g,h,i]perylene, indeno [1,2,3,-c,d]pyrene). The principal objective was to establish the of the considered basins. The content of triazine herbicides were below the detection limits (<MDL) in all sediment samples. Total contents of PCBs ranged from <MDL to 57.0 μg kg^{-1} and PAHs from <MDL to 728 μg kg^{-1}. The concentrations of organochlorine pesticides ranged from <MDL to 113 μg kg^{-1}. The most abundant organochlorine pesticide was the DDT and its degradation products. Obtained result shows ongoing recent inputs of DDTs to the rivers. The evaluations suggest that environmental persistent toxic substances levels are generally relatively low, although organochlorine pesticides may be an issue in some areas of the basin (Tisa, Sava, and Topčiderska river).

Keywords: Persistent organic compounds, organochlorine pesticides, polyaromatic hydrocarbons, polychlorinated biphenyls, triazine herbicides, river sediment

Introduction

Persistent organic pollutants (POPs) are a group of organic chemicals that pose a threat to the environment despite no longer being in common use (Heath et al. 2010). Generally, POPs are hydrophobic and therefore, readily bind to the particle fraction in lake and river waters.

Subsequently, via sedimentation processes, these contaminants are deposited to the basin bed. They remain very long in sediment processes, due to their long half-life times (Minh et al. 2007). The sources of organic micropollutants are very different, such as industrial processes and combustion sources, including traffic and agricultural uses (Kelderman et al. 2012). Soil and sediments play an important role in the global distribution and fate of POPs. They not only have a large retention capacity but also, as a secondary source, they re-emit POPs into the environment and from particles into water under favorable conditions (Aly Salem et al. 2013).

Polychlorinated biphenyls (PCBs), organochlorine pesticides (OCPs), polyaromatic hydrocarbons (PAHs) and triazine herbicides represent an important group of POPs that have caused worldwide concern as toxic environmental contaminants (Baumard et al., 1998; Papadopoulou and Samara, 2002; Kanzari et al., 2012; Bahroumi et al., 2014; Montouri et al. 2014). The input sources of PCBs and OCPs into river environments include discharge of domestic sewage and industrial wastewater, runoff from nonpoint sources and direct dumping of wastes into the river (Montuori et al. 2014). OCPs were used on a large scale in agriculture, forestry and public health. In agriculture, they act as insecticides, acaricides and fumigants to control pests in orchards, vegetable, grain, cotton and tobacco fields and vineyards (Montuori et al. 2014). Despite their ban or restricted use, PCBs and pesticides studied are among the most prevalent environmental pollutants and can be found in various environmental compartments, abiotic (air, water, sediment, soil) as well as biotic (from plankton to humans). The surficial sediments constitute one of the primary locations of PCB accumulation, as confirmed by numerous studies (Cardellicchio et al., 2007; Vane et al., 2007; Albanese et al., 2010; Zhao et al., 2010; Barhoumi et al., 2014; Kanzari et al., 2014).

PAHs represent a complex mixture of compounds (family of more than 100 organic molecules comprising at least two aromatic cycles) originating from the incomplete combustion of organic matter. They are classified as persistent organic pollutants (POPs) (Kanzari et al. 2012). Alkyl PAHs bioaccumulate more, and their toxicity, phototoxicity, and carcinogenicity can be equal or more pronounced than that of the parent molecules. They tend to persist for a longer time than the parent PAHs and are often more abundant than parent compounds (at least those alkyl PAHs originating from petrogenic sources) (Irwin et al., 1997). Recently, methods of the computational chemistry (Ab initio and DFT) were used to calculate the electronic properties of alcylated PAHs, threemethyl naphthalene (TMN) and dimethylanthracenes (DMA) for the prediction of

their biodegradation rates (Ostojić and Đorđević 2012; Ostojić et al., 2015) and mutagenicity of 1-nitro-6-azabenzo[a]pyrene N-oxide (1-N-6-ABPO) and 3-nitro-6-azabenzo[a]pyrene N-oxide (3-N-6-ABPO) (Ostojić and Đorđević, 2015a) and Nitro derivatives of azaphenanthrenes (Ostojić and Đorđević, 2015b).

Examination of POPs levels in sediment may give basic information on the contamination status, sources and ecological risk of POPs in the aquatic environments. The research, which provides information on the degree of contamination of river and lake sediments in the territory of Serbia are insufficient and poor, especially in the field of organic, persistent and emergent substances. In Teodorović (2009) was stated that several hot spots of heavy freshwater pollution and sediment contamination have been identified in Serbia resulting from outdated environmental legislation, negligible amounts of properly treated waste waters and accidental spills. Heavy metals appear to be the most prominent problem, although the elevated contents of organic pollutants (polychlorinated biphenyls, organochlorine pesticides, polyaromatic hydrocarbons, organic micro pollutants, even emerging substances) have also been registered in some areas.

Comprehensive studies on the environmental pollution of persistent organic pollutants in Serbian river system have not been carried out in recent years. In this study, surface sediments from river and artificial lakes collected from different locations in Serbia were examined by chemical analysis for POPs compounds (organochlorine pesticides, triazine herbicides, polychlorinated biphenyls, and polycyclic aromatic hydrocarbons), providing baseline data the environmental state of river and artificial lakes, spatial distribution and the possible origins of the studied POPs.

Materials and methods

Study area

The rivers Danube, Sava and Tisa belongs to the Black Sea river basin and they flow through many European countries, while in Serbia they flow through the Pannonian flatland. Transboundary river courses and river' catchment areas are not restricted to the geographical territory of a particular country.

The waters in Danube River Basin serve people for many purposes: drinking water preparation, use for industrial and agricultural activities, recreation, hydropower generation and navigation (Nagy et al. 2013). Because of that, the Danube River Basin is under great pressure from diverse human activities, which can be allocated to the municipal, industrial and agricultural sectors.

Agriculture is a traditionally prominent economic sector, especially in the middle and lower Danube Basin countries (Paleari et al. 2005). The organochlorine compounds (lindane and DDT) show the same spatial profile, with an increasing pattern from the upper and middle to the lower Danube. The concentrations of polyaromatic hydrocarbons (PAHs) in sediments were usually lower than the quality target, while their concentration in mussels showed an increasing trend as one moved downstream to the Danube delta (Paleari et al. 2005). In Nagy et al. (2013) is shown that PAHs concentrations in water and sediment samples at different sampling stations along the whole Danube showed wide variations. Also, the concentration profiles downstream in the Danube suggest that PAHs arise from diffuse sources. For Danube, in Nagy et al. (2013) is shown that transboundary pollution arrived approximately ten times during the 13 years.

Sediment sampling and analysis

Sediment samples (Fig. 1, Table 1) were collected from 15 rivers and 6 artificial lakes in Serbia (52 samples in total). River samples were taken from the Danube, Tisa, Begej, Tamiš, Vrbas DTD, Sava, Kolubara, Topčiderska river, Great Morava, West Morava, Ibar, South Morava, Toplica, Nišava, Pek, and Porečka river. Lake sediment samples were taken from Bovan, Ćelije, Vrutci, Barje, Bojnik, and Garaši.

The granulometric fraction (< 63 μm) of the bottom sediment samples ("grab"-the sample) were used to determinate POPs content. The sampling of sediments in this research was conducted using a Van Veen grab sampler, designed to collect an accurate representative sample of the sediment. Sediments in surface water are frequently heterogeneous due to small-scale changes in hydrological regime and geomorphologic changes in the catchment area. Equal volumes of sediment from nearby locations (five subsamples) were mixed for each site in order to make a composite sample to expand the area represented by the sample.

The sediment were analyzed for PCBs (PCB 28, PCB 52, PCB 101, PCB 138, PCB 153, PCB 180, and PCB 194), organochlorine pesticides (α-HCH, β-HCH, γ-HCH, benzene hexachloride, heptachlor, heptachlor epoxide, aldrin, endrin, dieldrin, DDE, *p,p′*-DDD, *p,p′*-DDT, *o,p′*-DDT, and methoxychlor), triazine herbicides (atrazine, simazine, and propazine) and PAHs (fluoranthene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo [a]pyrene, benzo [g,h,i]perylene, indeno [1,2,3,-c,d]pyrene). The POPs concentration in the sediment were determined according to the following methods: EPA 8270D:2007/GC-MS (PAHs and PCBs),

EPA 8270D:2007/GC-MS and SRPS EN ISO 6468:2008/GC-ECD (OCls) and UP 1.124/PC12 /GC-MS and SRPS EN ISO11369:2008/LC-UV (triazine herbicides). The measurements uncertainty ranged between 20-25% for all studied parameters. Method detection limits for studied compound are shown in Table 2.

Mean, minimum and standrad deviation were determined for all data. Q mode of Cluster analysis (CA) was performed to group of samples in terms of similarity/dissimilarity among sediments. The hierarchical clustering joining of the most similar observations, and successively of the next most similar observations was employed in the presented research. All statistical analyses were carried out using SPSS 21 for Windows.

Results and Discussion

Organochlorine pesticides

In river sediments (Fig. 1a), content of organochlorine pesticides (α-HCH, β-HCH, γ-HCH, benzene hexachloride, heptachlor, heptachlor epoxide, aldrin, endrin, and *o,p'*-DDT) were below detection limit. Dieldrin was only detected in one sample (Tisa, M7) with 4.2 μg kg^{-1} (Table 3,4). Dieldrin is a synthetic organochlorine pesticide and also the main persistent degradation product of the synthetic organochlorine pesticide aldrin. Dieldrin has penetrated in aquatic systems primarily as surface runoff from lands treated lumber, and from deposition following volatization and aerial transport. Because of its hydrophobicity and affinity for organic materials, dieldrin in aquatic systems tends to become bonded to particulate matter and accumulate in sediments. DDE, *p,p'*-DDD, *p,p'*-DDT, and methoxychlor were extracted from river sediments samples (Fig. 2). Sum of DDTs and theirs degradation products were found to be between <MDL–295 μg kg^{-1}. The pesticide DDT and its degradation products, DDD and DDE, mainly derive from agricultural sources; the use of DDT has been banned in the Serbia since 1989 and the residual levels found in the river sediments can probably be ascribed to the strong persistence of this pesticide in the environment. *p,p'*-DDT is extracted mostly from majority of samples, and the highest content (295 μg kg^{-1}) was observed in Topčiderska river (at the site 26). The content of other organochlorine pesticides that were determined was mainly less than 16 μg kg^{-1}, but the highest DDE content was detected in Topčiderska river (at the site 26) also.

In studied sediments, the ratio of DDT/(DDE+DDE) generally over 0.1 points to recent rather than historical DDT inputs (Kelderman et al. 2012). The results suggest that although the use of

organochlorine pesticides is generally prohibited or restricted, they can be found in households because of their stocks. This assumption is confirmed by comparing the results obtained for the Danube sediment in this study and research in 2007 (Škrbić et al. 2007). Compared with the results obtained by Škrbić et al. (2007) for the Danube sediment (6.31 μg kg^{-1} *p,p′*-DDT), the results obtained in this study were higher (up to 94 μg kg^{-1} *p,p′*-DDT in the Danube sediments). In general, DDT and other persistent organochlorine pesticides continue to penetrate surface waters from sources such as atmospheric deposition and erosion of soils that have been contaminated by past use. Field studies indicate that DDT half-lives in soil are 15 years or longer (USGS, 2015).

In artificial lakes sediments in Serbia (Fig. 3a), content of organochlorine pesticides (*α*-HCH, *β*-HCH, *γ*-HCH, benzene hexachloride, heptachlor, heptachlor epoxide, aldrin, endrin, dieldrin, *p,p′*-DDD and *o,p′*-DDT) were below detection limit. *p,p′*-DDT is the most abundant organochlorine pesticide extracted from all samples.

Triazine herbicides

The content of triazine herbicides (atrazine, simazine, and propazine) were below detection limits in all sediment samples (both, rivers and lakes), which may indicate that these pesticides were not used in the research areas.

PCB

All the studied PCBs were extracted. PCB 28, PCB 52, PCB 101 and PCB 194 were determined at some localities, while PCB 138, PCB 153, and PCB 180 were extracted from almost all the investigated sediments (Fig. 2b and Fig. 3b), and with similar content (Table 4). ∑7 determined PCB_s was in range MDL–57 μg kg^{-1} (mean 7.4 μg kg^{-1}). Comparison of the obtained results for PCBs with the content of PCBs in the sediment of the Danube river (Novi Sad) (Škrbić et al. 2007) indicate that the content in the studied sediments at this site (0.41 μg kg^{-1}) was lower than ∑7 PCBs in the Danube sediments examined in this study (to 10.2 μg kg^{-1}).

The highest extracted PCBs content was observed in Tisa River (sample 15, locality Novi Bečej) and in Topčiderska River (sample 26). In the report on the state of the environment in Serbia (Ministry of Agriculture and Environment, 2013), also shown an increased content of the sum of PCBs (0.402 mg kg^{-1}). These results indicate that the wider areas of the measuring points are

contaminated and sources of PCBs contamination. Topčiderska River was synonymous with highly contaminated aquifer for many years, because the hot water from a number of illegal housing and rural households and industrial waste water from Rakovica basin in this watercourse untreated vented. In the lower part of Topčiderska river an open concrete collector for receiving wastewater from Rakovica basin is. It should be emphasized that in Serbia are no permanent storage sites or decontamination or disposal of equipment contaminated with PCBs, as well as plants for the degradation of the fluid-based PCB (Veljković et al. 2014). Preliminary inventory established that in many places in the Republic of Serbia are transformers and capacitors that are unused and represent waste. Most of the equipment is temporarily taken care of, but in some cases leaked capacitors, or contaminated fluid oils based on PCBs are placed in barrels and tanks, while a number is not stored temporarily (Veljković et al. 2014).

In some of samples were extracted PCB 138, PCB 153 and PCB 180, while others PCBs have not been found in the artificial lakes. Content of determined PCBs in the lakes is lower compared to their content in the river sediment.

PAH

From studied PAHs, all of these compounds were detected in river sediments (Fig. 2c), but not in all of the samples. From some of sediments were extracted benzo [g,h,i]perylene and indeno [1,2,3,-c,d]pyrene. Fluoranthene, benzo [b]fluoranthene, benzo[k]fluoranthene, and benzo [a]pyrene are detected in most of samples, whereby the most abundant PAH was fluoranthene. The range of PAH concentration (∑6 PAH) in river sediment from Sava (240 – 395 μg kg^{-1}) was lower than in the Danube sediments (<MDL – 689 μg kg^{-1}) and lower than in Tisa (86.1 – 728 μg kg^{-1}). The obtained values for Sava and the Danube were similar to the values published by Crnković et al. (2008). According to the classification proposed by Baumard et al. (1998), pollution levels with PAHs can be characterized as low, moderate, high, and very high when total PAH concentrations are 0–100 ng g^{-1}, 100–1000 ng g^{-1}, 1000–5000 ng g^{-1}, and >5000 ng g^{-1} , respectively (Papadopoulou and Samara 2002; Ünlü et al. 2010). Therefore, it can be stated that levels found in river sediments reflect a low to moderate level of pollution due to PAHs. In 46% of all studied river sediments pollution was low, but moderate in 54%. All sediments of artificial lakes (Fig. 3c) had low pollution level with PAHs, i.e. sum of all values for studied PAHs were below 100 μg g^{-1}.

Combustion of fossil fuels, waste incineration, and oil spills are the potential sources of PAHs in the environment (Neammtu et al. 2009). PAH diagnostic ratios may provide an important tool for the identification of pollution emission sources (Tobieszewski and Namieśnik, 2012). In this paper, for PAH diagnostic Ipy/(Ipy+BPe) ratios have been used. Values of this ratio < 0.2 indicate petrogenic source, from 0.2 to 0.5 petroleum combustion and values > 0.5 indicate grass, wood and coal combustion as PAHs dominate sources (Katsoyiannis et al 2007, Brandli et al. 2007). In this research, all values for Ipy/(Ipy+BPe) was > 0.2, indicating dominate PAHs origin from pyrogenic sources. Results show that in the Danube River (sample 3), Tisa River (samples 7 and 10), Sava River (sample 21) and Topciderska River (sample 26) values Ipy/(Ipy+BPe) was > 0.5 (Danube – sample 0.57; Tisa – samples 0.56 and 0.58; Sava – sample 0.86 and Topčiderska river – sample 0.58), indicating origin from grass/coal/wood combustion and only in Tisa River Ipy/(Ipy+BPe) was < 0.5 (0.44) indicating origin from fuel combustion (one sample – sample 17 from Tisa-Titel). From studied PAHs, in all lake sediment samples have been determined fluoranthene, benzo [b]fluoranthene, and benzo [k] fluoranthene. Indeno [1,2,3,-c,d] pyrene are not detected in sediment samples, while benzo [a] pyrene, and benzo [g,h,i] perylene are determined at some localities.

It should be emphasized that the quality of some watercourses flowing into Sava and the Danube are worse than Sava and Danube quality (Tisa river, Topčiderska river for example). The primary reason for this is that most of them gravitate to urban and semi-urban sewage systems and industrial processes from which no purified waste water is discharged.

Cluster analysis

The results of Q mode of CA (Pearson method) are presented in the dendrogram (Figure 4). The 39 river sediments samples were hierarchically clustered. Two big (cluster I and II) and three small clusters (III, IV, and V) could be distinguished. Cluster I and cluster II contain most of sediments samples. The clusters III, IV and V are represented by only one station (III – Danube (Bezdan), IV - Nišava (Niš) and V - South Morava (Vladičin Han)). These stations are similar since that on studied localities are not extracted POPs from sediments. The clusters I and II contain samples of river sediments with different content of studied POPs. Cluster I contains mainly sediment samples from Vojvodina (North Serbia) as well as one sample from Great Morava (Central Serbia). In these regions, agriculture is still one of the most important activities.

The average content of PAHs in river sediment form Cluster I (219 μg kg^{-1}) is much higher than average content of PAHs in cluster II (72.9 μg kg^{-1}). The rivers in sediments from cluster I (Danube, Tisa and Sava) are international rivers under strong influence of contamination along the whole drainage basins. The high content of PAH compounds in rivers from Cluster I may be consequence of accumulation and long-distance transport of the PAHs along the entire rivers courses. The cluster II is composed of local rivers from the territory of Serbia. In some of samples from Cluster II is noted high content of DDTs. The dominance of DDTs in these sediments indicates slow degradation of DDTs or recent inputs of fresh DDTs at these locations.

Comparison with Sediment quality guidelines and POPs content in surface sediments from various locations in the world

The ecotoxicological significance of sedimentary POPs levels was evaluated by comparison with Sediment quality guidelines (SQGs). Results of comparison of obtained results with SQGs (MacDonald et al. 2000; Canadian sediment quality guidelines-CCME, 2001) indicate that at some localities DDE, DDD and DDT content (sediments from rivers and lakes) were higher than ISQG (interim sediment quality guidelines – this is generally based on the threshold effect levels) and TEC (*i.e.*, below which harmful effects are unlikely to be observed), respectively higher than values at which the expected minimal effect, while at some localities for DDT (one sample of the Danube and Tisa, 2 samples from Sava, Kolubara, Topčiderska river, one sample from Great Morava, West Morava, Ibar, South Morava, Toplica, Nišava, Porečka river, artificial lakes as Bovan, Barje, Bojnik and Garaši) content was higher than PEC (*i.e.*, above which harmful effects are likely to be observed). Values for DDT content (total) were below PEC proposed by MacDonald et al. 2000 at studied localities, indicating that sum DDT content are below content at which harmful effects are likely be observed. It can be concluded that content of DDT (total) is in the possible effects range within which adverse effects occasionally occur.

The content of dieldrin in Tisa are higher than ISQ and TEC, but lower than PEC and PEL (MacDonald et al. 2000; CCME, 2001). In some of the river sediments were detected certain POPs and these values over PEL and TEL (Σ PCBs in the sample Tisa and Topčiderska river, fluoranten (the Danube), and benzo [a]pyrene (Dunav, Sava, Tisa, Vrbas), while in sediment samples of the lake's these POPs compounds were not detected. For Tisa river, obtained values

for pesticide residues and metabolites in sediment are expected due to the proximity of agricultural land with a long history of use of plant protection products in this area.

Results of comparison of obtained results with literature POPs data are shown in Table 5. A quantitative comparison across reported POPs data is difficult due to variances in the number and type of individual species determined in each study, the analyzed sediment fraction, and the analytical method used. In general., the ∑PAHs, ∑PCBs and ∑DDTs levels found in the present study are consistent with those reported in Table 5. The large difference between the minimum and maximum content of sum values extracted PAHs, PCBs and pesticides (mainly DDT forms) are due to the fact that in the research were included river and lake systems from the whole territory of Serbia which is very heterogeneous system with great inter-difference.

By comparing the obtained results with literature data and sediment quality guidelines, it can be concluded that from detected organochlorine pesticides or their metabolites, DDT may pose a risk for the ecosystem. Besides the fact that their use is restricted or prohibited, some organochlorine pesticides are still present in the environment. The comparison of sedimentary PAH and PCB data with presented criteria, show these compounds are not expected to be a primarily cause for ecochemical and ecological degradation in studied sediments in Serbia.

Conclusion

This study demonstrates DDTs as major organochlorine contaminants of sediments in river systems in Serbia. Although use of DDT was officially restricted in Serbia, the result showed evidence of inputs of DDT into the rivers but sum DDT content are below content at which harmful effects are likely be observed. Toxicological assessment suggests that, in general, PAH and PCB contents is not expected to be primarily cause for ecochemical and ecological degradation in studied sediments.

The data gathered in this study can be used as baseline reference for further watershed management. The presented results of POPs chemicals in rivers and lakes sediments of Serbia point the need to control these pollutants in various environmental media. More comprehensive studies would be needed in order to clarify the pathway of inputs DDTs to the aquatic environment as well as to investigate further contamination by organic pollutants groups.

Acknowledgments

This study was supported by the Ministry of Education, Science and Technological Development of Serbia, Grant Nos. 172001 and 43007. In addition, we would like to thank the Republic Hydrometeorological Service of Serbia for the sediment samples and others data.

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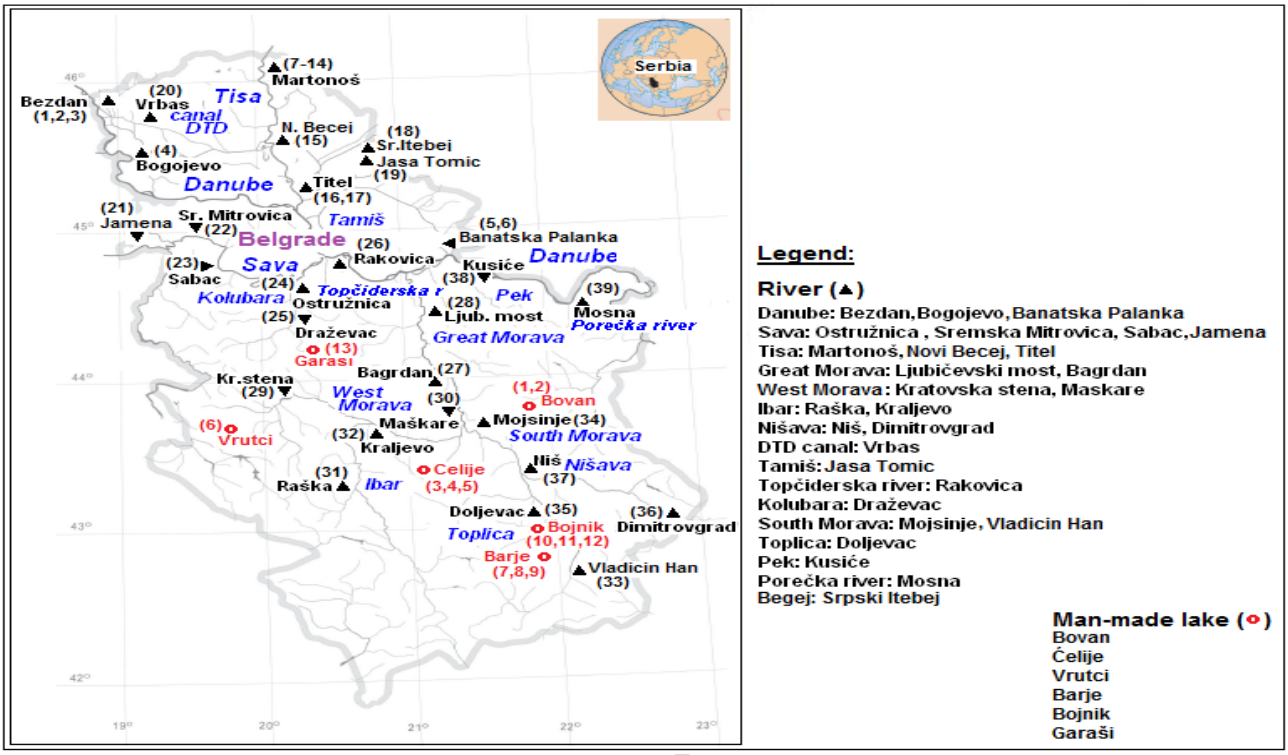
Figures captions

Fig. 1 Investigated rivers and man-made lakes

Fig. 2. The distribution content of POPs compounds in Serbian river sediment: a) content of all studied pesticides; a1) content of studied pesticides without p,p-DDT (p,p-DDT not shown because of its high content); b) content of studied PCBs; c) content of studied PAHs

Fig. 3 The distribution content of POPs compounds in sediment from Serbian artificial lakes: a) content of studied pesticides; b) content of studied PCBs; c) content of studied PAHs

Fig.4 Hierarchical dendrogram for studied sediments



Legend:

River (▲)

Danube: Bezdan, Bogojevo, Banatska Palanka
Sava: Ostružnica , Sremska Mitrovica, Sabac, Jamena
Tisa: Martonoš, Novi Becej, Titel
Great Morava: Ljubičevski most, Bagrdan
West Morava: Kratovska stena, Maskare
Ibar: Raška, Kraljevo
Nišava: Niš, Dimitrovgrad
DTD canal: Vrbas
Tamiš: Jasa Tomic
Topćiderska river: Rakovica
Kolubara: Draževac
South Morava: Mojsinje, Vladicin Han
Toplica: Doljevac
Pek: Kusiće
Porečka river: Mosna
Begej: Srpski Itebej

Man-made lake (○)

Bovan
Ćelije
Vrutci
Barje
Bojnik
Garaši

Fig. 1

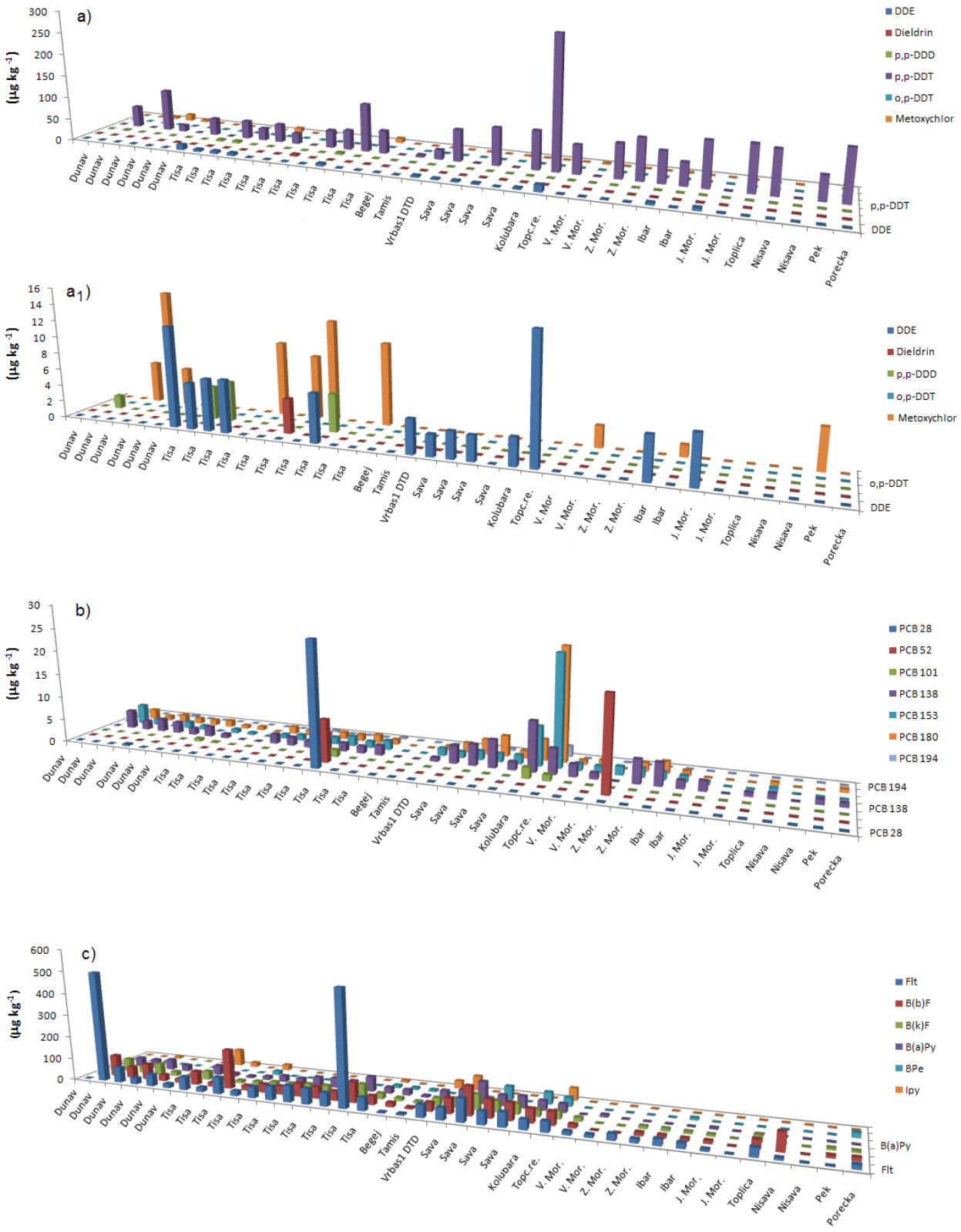


Fig. 2

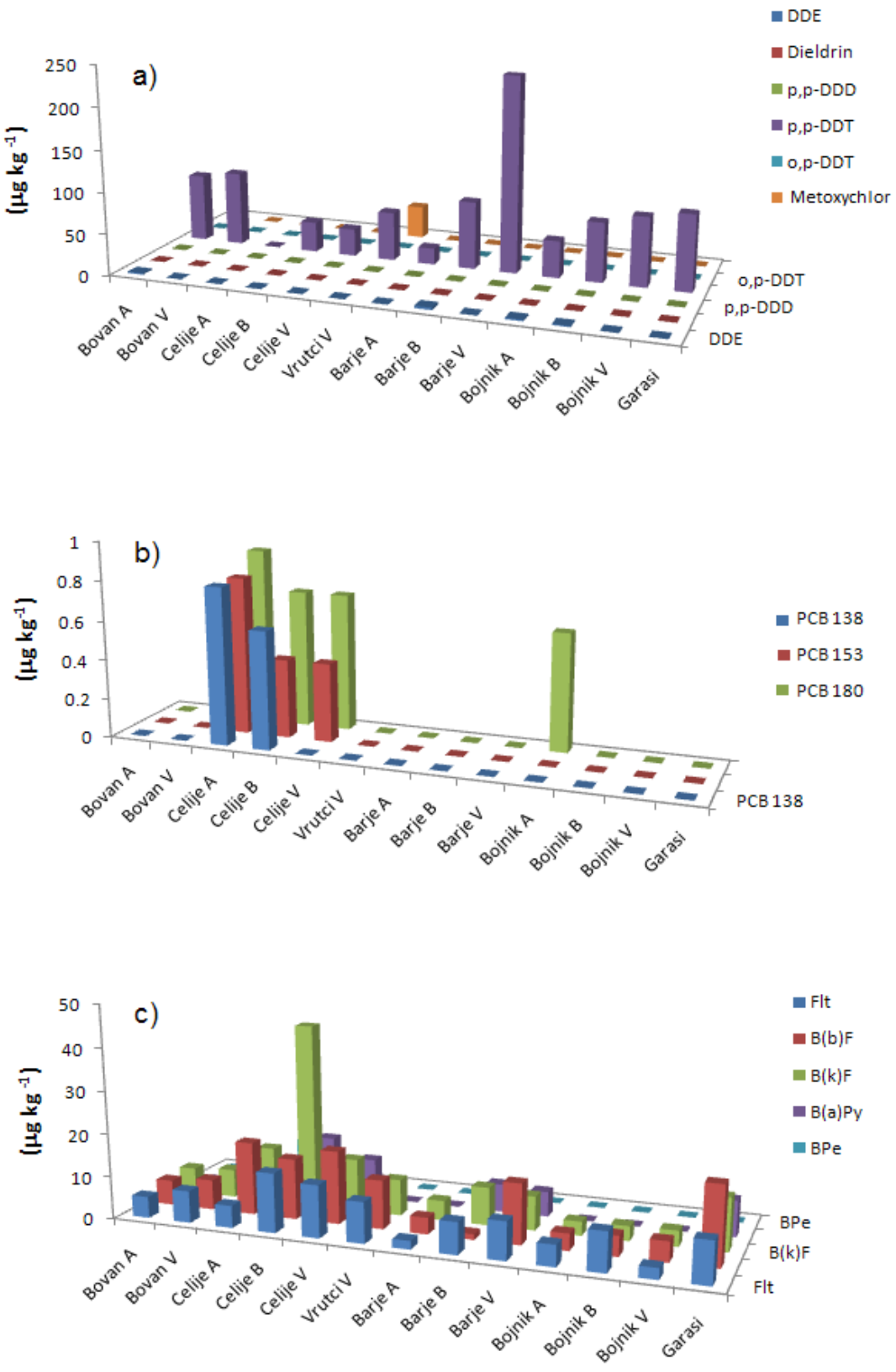


Fig. 3

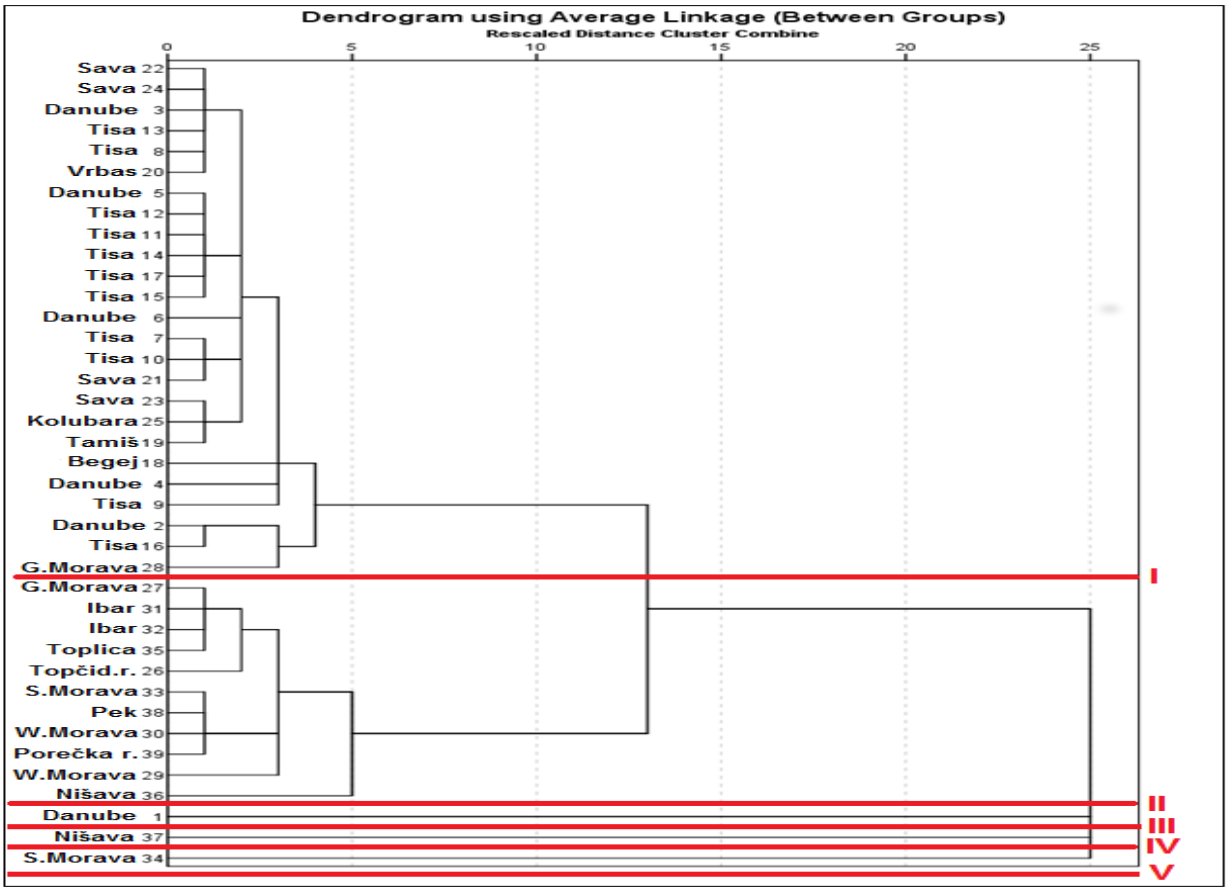


Fig. 4

Table 1. Sampling locations.

	Sampling location	Number of sample
River		
Danube (Dunav)	Bezdan	3
Danube (Dunav)	Bogojevo	1
Danube (Dunav)	Banatska Palanka	2
Tisa	Martonoš	8
Tisa	Novi Bečej	1
Tisa	Titel	2
Begej	Srpski Itebej	1
Tamiš	Jaša Tomić	1
Vrbas 1DTD	Vrbas 1	1
Sava	Jamena	1
Sava	Sremska Mitrovica	1
Sava	Šabac	1
Sava	Ostružnica	1
Kolubara	Draževac	1
Topčiderska river	Rakovica	1
Great Morava (Velika Morava)	Bagrdan	1
Great Morava (Velika Morava)	Ljubičevski most	1
West Morava (Zapadna Morava)	Kratovska stena	1
West Morava (Zapadna Morava)	Maskare	1
Ibar	Raška	1
Ibar	Kraljevo	1
South Morava (Južna Morava)	Vladičin Han	1
South Morava (Južna Morava)	Mojsinje	1
Toplica	Doljevac	1
Nišava	Dimitrovgrad	1
Nišava	Niš	1
Pek	Kusiće	1
Porečka river	Mosna	1
Man-made lake		
Bovan		2
Ćelije		3
Vrutci		1
Barje		3
Bojnik		3
Garaši		1

Table 2. List of compounds, family, abbreviation and method detection limit (MDL, µg kg^{-1}).

Full name	Abbreviation	MDL
PAHs		
Fluoranthene	Flt	0.1
Benzo [b]Fluoranthene	B[b]F	0.2
Benzo[k]Fluoranthene	B[k]F	0.2
Benzo [a]pyrene	B[a]Py	0.2
Benzo [g,h,i]perylene	BPe	0.2
Indeno [1,2,3,-c,d]pyrene	Ipy	0.2
OCls		
α-Hexachlorocyclohexane	α-HCH	0.5
β-Hexachlorocyclohexane	β-HCH	0.5
γ- Hexachlorocyclohexane (Lindane)	γ-HCH	0.5
Benzene hexachloride	BHC	0.1
Heptachlor	HC	0.4
Heptachlor epoxide	HCE	0.5
*	Aldrin	0.5
**	Endrin	1.0
***	Dieldrin	0.6
Dichloro diphenil dichloroetylene	DDE	0.6
p,p'-Dichloro diphenil dichloroetane	*p,p'*-DDD	0.1
p,p'-Dichloro diphenil trichloroetane	*p,p'*-DDT	0.5
o,p'-Dichloro diphenil trichloroetane	*o,p'*-DDT	0.5
Methoxychlor	MTC	0.2
Triazine herbicides		
Atrazine	ATR	1.3
Simazine	SIM	2.0
Propazine	PRO	0.9
PCBs		
2,4,4'-Trichlorobiphenyl	PCB 28	0.2
2,2',5,5'-Tetrachlorobiphenyl	PCB 52	0.2
2,2',4,5,5'-Pentachlorobiphenyl	PCB 101	0.3
2,2',3,4,4',5'-Hexachlorobiphenyl	PCB 138	0.3
2,2',4,4',5,5'-Hexachlorobiphenyl	PCB 153	0.2
2,2',3,4,4',5,5'-Heptachlorobiphenyl	PCB 180	0.2
2,2',3,3',4,4',5,5'-Octachlorobiphenyl	PCB 194	0.4

* 1,2,3,4,10,10-Hexachloro-1,4,4a,5,8,8a-hexahydro-1,4:5,8-dimethanonaphthalene

** 1R,2S,3R,6S,7R,8S,9S,11R)-3,4,5,6,13,13-Hexachloro-10-oxapentacyclo[6.3.1.13,6.02,7.09,11]tridec-4-ene

*** 1a*R*,2*R*,2a*S*,3*S*,6*R*,6a*R*,7*S*,7a*S*)-3,4,5,6,9,9-hexachloro-1a,2,2a,3,6,6a,7,7a-octahydro-2,7:3,6-dimethanonaphtho[2,3-*b*]oxirene

Table 3. POPs statistical analysis, river and lake sediment (content in µg kg^{-1})

Rivers

	DDE	Dieldrin	p,p-DDD	p,p-DDT	MTC	PCB28	PCB52	PCB101	PCB138
Min	<MDL[1]	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL
Max	15	4.2	4.8	295	14.3	26.4	19.6	2.3	10.6
	PCB153	PCB180	PCB194	Flt	B[b]F	B[k]F	B[a]Py	BPe	Ipy
Min	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL
Max	23.7	24.3	2.3	515	176	67.8	97.3	60.7	73.7

Lakes

	DDE	Dieldrin	p,p-DDD	p,p-DDT	MTC	PCB28	PCB52	PCB101	PCB138
Min	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL
Max	2.8	<MDL	<MDL	236	39.5	<MDL	<MDL	<MDL	0.8
	PCB153	PCB180	PCB194	Flt	B[b]F	B[k]F	B[a]Py	BPe	Ipy
Min	<MDL	<MDL	<MDL	2	1.3	3.3	<MDL	<MDL	<MDL
Max	0.8	0.9	<MDL	13.9	18.4	43.2	13.8	9.5	<MDL

[1] Below detection limit

Table 4. POPs content in sediments (μg kg^{-1}) from rivers (R, 1-39) and man-made lakes (L, 1-13)

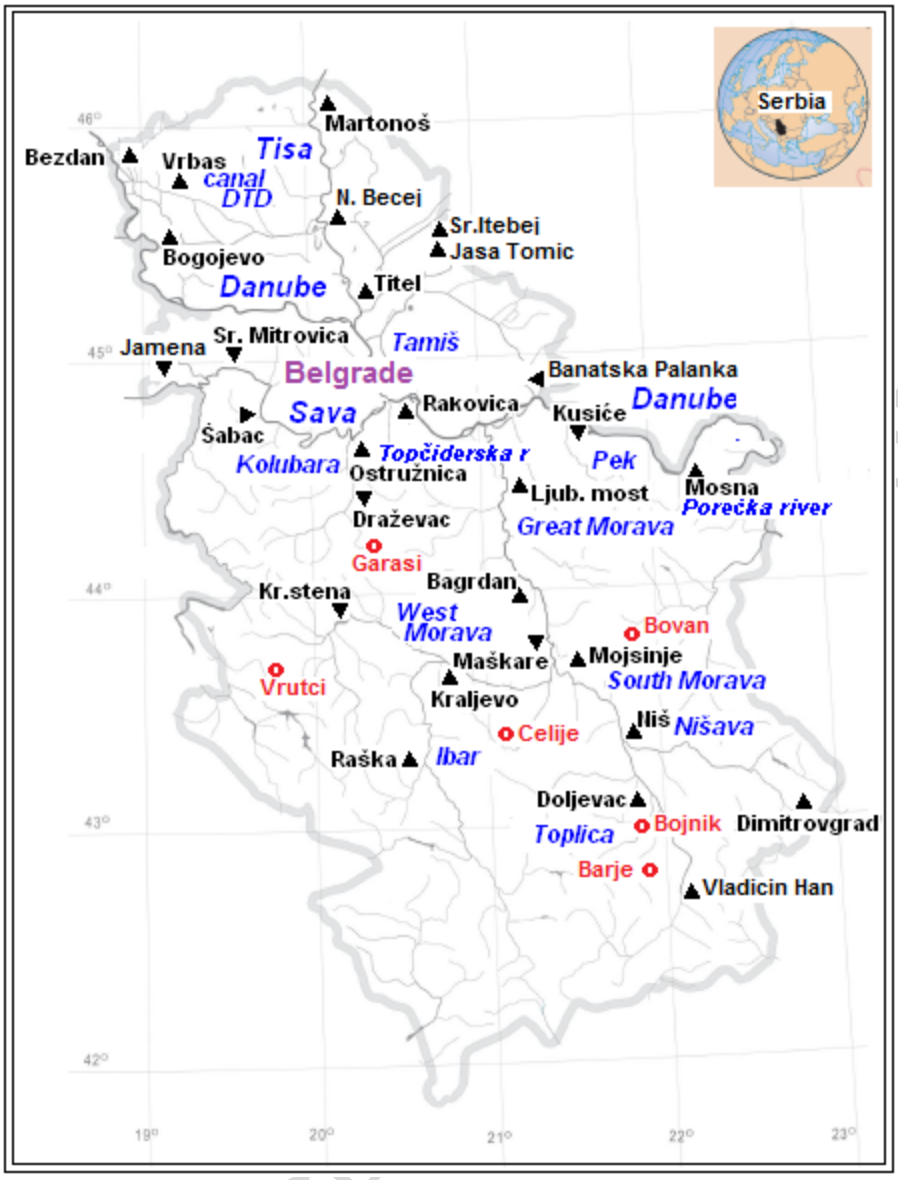
	DDE[5]	DDD[5]	[6]pp DDT	DDT total	PCB 138	PCB 153	PCB 180	PCB sum	Flt	B[b]F	B[k]F	B[a]Py	BPe	D[7]	Ipy	PAH sum
R																
1D	<gd	<gd	<gd	<gd	<gd	<gd	<gd	<gd	<gd	<gd	<gd	<gd	<gd	<gd	<gd	<gd
2D	<gd	1.6	48.2	49.8	3.90	4.20	2.10	10.2	499	94.9	56.0	39.2	<gd	<gd	<gd	689
3D	<gd	<gd	<gd	<gd	1.90	1.30	1.00	4.20	69.3	48.9	29.8	38.0	7.00	<gd	9.30	202
4D	<gd	<gd	94.6	94.6	2.70	<gd	1.60	5.10	28.5	69.9	48.9	46.1	<gd	<gd	<gd	193
5D	<gd	<gd	14.4	14.4	2.40	1.40	1.20	5.00	53.5	28.9	20.3	27.6	<gd	<gd	<gd	130
6D	<gd	<gd	<gd	<gd	1.40	.90	1.20	3.50	19.4	18.5	13.0	<gd	<gd	<gd	<gd	50
7T	12.2	<gd	38.5	50.7	2.10	<gd	1.40	4.10	59.2	61.1	38.4	42.0	58.8	<gd	73.7	333
8T	5.60	4	<gd	9.6	0.90	0.90	0.70	2.70	20.4	18.7	13.6	13.7	<gd	<gd	19.7	86.1
9T	6.30	4.8	40.4	51.5	<gd	0.50	0.80	1.30	75.1	176	12.5	9.20	<gd	<gd	<gd	273
10T	6.40	<gd	27.3	33.7	<gd	<gd	<gd	<gd	22.0	22.0	16.6	16.0	17.9	<gd	24.5	119
11T	<gd	<gd	40.9	40.9	2.10	0.90	1.50	4.50	49.5	37.2	23.6	27.0	<gd	<gd	<gd	137
12T	<gd	<gd	23.9	23.9	1.80	1.20	1.20	4.20	60.7	36.3	25.7	24.3	<gd	<gd	<gd	147
13T	<gd	<gd	<gd	<gd	1.40	1.30	1.00	4.10	72.8	58.5	31.5	38.7	<gd	4.2	<gd	202
14T	<gd	<gd	39.5	39.5	2.20	1.50	1.50	5.20	69.2	51.5	33.1	42.9	<gd	<gd	<gd	197
15T	5.90	4.6	43.6	54.1	1.80	2.10	1.40	42.3	59.5	41.7	32.2	39.1	<gd	<gd	<gd	17
16T	<gd	<gd	108	108	1.50	1.40	1.70	4.60	515	89.7	57.1	66.1	<gd	<gd	<gd	728
17T	<gd	<gd	51.2	51.2	2.10	2.00	1.10	5.60	57.8	40.0	25.3	27.0	11.8	<gd	9.2	171
18B	<gd	<gd	<gd	<gd	<gd	<gd	<gd	<gd	4.6	19.2	14.3	25.0	10.0	<gd	<gd	73.1
19T	<gd	<gd	5.20	5.2	<gd	<gd	<gd	<gd	7.6	8.60	7.9	7.6	9.30	<gd	<gd	41.0
20V	4.10	<gd	21.2	25.3	0.80	1.60	0.90	3.30	56.8	39.4	33.0	39.1	<gd	<gd	33.8	202
21S	2.60	<gd	71.0	73.6	3.80	1.30	2.00	7.10	54.2	67.3	38.7	51.2	10.8	<gd	64.3	286
22S	3.20	<gd	<gd	3.2	4.50	1.90	3.20	9.60	102	128	67.8	97.3	<gd	<gd	<gd	395
23S	3.00	<gd	84.1	87.1	5.90	1.80	4.40	12.6	58.8	81.4	49.5	47.4	60.7	<gd	<gd	298
24S	<gd	<gd	<gd	<gd	1.70	1.90	1.60	5.20	64.3	77.4	46.1	52.2	<gd	<gd	<gd	240
25K	3.30	<gd	85.9	89.2	10.6	8.40	4.50	25.8	43.9	61.4	31.9	41.4	52.8	<gd	<gd	231
26Tr	15.0	<gd	295	310	5.40	23.7	24.3	57.0	46.5	59.3	34.3	37.3	39.0	<gd	54.8	271
27V	<gd	<gd	64.1	64.1	2.60	1.50	.90	5.00	15.6	16.1	15.8	<gd	<gd	<gd	<gd	47.5
28V	<gd	<gd	<gd	<gd	1.30	1.40	1.40	4.10	13.4	<gd	<gd	10.0	<gd	<gd	<gd	23.4
29Z	<gd	<gd	76.6	76.6	<gd	1.80	<gd	21.4	27.0	<gd	<gd	<gd	<gd	<gd	<gd	27.0
30Z	<gd	<gd	92.7	92.7	5.00	1.90	1.80	8.70	17.0	14.2	11.0	<gd	<gd	<gd	<gd	42.2
31I	5.10	<gd	69.8	74.9	4.80	1.60	2.50	8.90	27.8	13.8	11.5	<gd	<gd	<gd	<gd	53.1
32I	<gd	<gd	50.5	50.5	1.70	1.30	1.00	4.00	23.0	11.5	8.90	<gd	9.30	<gd	<gd	52.7
33J	5.80	<gd	101	106	2.10	0.60	<gd	2.70	9.40	12.5	8.20	5.90	<gd	<gd	<gd	36.0
34J	<gd	<gd	<gd	<gd	<gd	<gd	<gd	<gd	<gd	<gd	<gd	<gd	<gd	<gd	<gd	<gd
35To	<gd	<gd	103	103	0.80	<gd	<gd	0.80	37.6	39.6	27.3	<gd	<gd	<gd	<gd	104
36N	<gd	<gd	96.7	96.7	1.10	0.80	0.60	2.50	9.00	80.0	6.00	<gd	<gd	<gd	<gd	95.0
37N	<gd	<gd	<gd	<gd	<gd	<gd	<gd	<gd	<gd	<gd	<gd	<gd	<gd	<gd	<gd	<gd
38Pe	<gd	<gd	53.4	53.4	1.00	<gd	<gd	1.00	<gd	10.2	7.80	<gd	<gd	<gd	<gd	18.0
39Po	<gd	<gd	113	113	0.80	<gd	0.70	1.50	17.9	13.3	7.70	<gd	20.8	<gd	<gd	59.7
L																
1B	1.6		82.6	84.2	<gd	<gd	<gd	<gd	5	5.9	6	<gd	<gd	<gd	<gd	16.9
2B	<gd		90	90	<gd	<gd	<gd	<gd	7.5	7.1	6.6	<gd	<gd	<gd	<gd	21.2
3C	<gd		<gd	<gd	0.8	0.8	0.9	2.5	5.2	17.1	13	<gd	9.5	<gd	<gd	44.8
4C	<gd		36.8	36.8	0.6	0.4	0.7	1.7	13.9	14.2	43.2	13.8	<gd	<gd	<gd	85.1
5C	<gd		33.2	33.2	<gd	0.4	0.7	1.1	12.3	17.1	12.3	9.4	<gd	<gd	<gd	51.1
6V	<gd		59	59	<gd	<gd	<gd	<gd	9.6	11.4	8.5	<gd	<gd	<gd	<gd	29.5
7Ba	<gd		19.8	19.8	<gd	<gd	<gd	<gd	2	3.9	4.6	<gd	<gd	<gd	<gd	10.5
8Ba	2.8		82	84.8	<gd	<gd	<gd	<gd	7.5	1.3	8.9	6.6	<gd	<gd	<gd	24.3
9Ba	<gd		236	236	<gd	<gd	<gd	<gd	8.9	14.1	8	5.9	<gd	<gd	<gd	36.9
10Bo	1.8		44.9	46.7	<gd	<gd	0.6	0.6	5	4	3.3	<gd	<gd	<gd	<gd	12.3
11Bo	1.2		72.5	73.7	<gd	<gd	<gd	<gd	9.2	4.8	3.5	<gd	<gd	<gd	<gd	17.5
12Bo	<gd		84.3	84.3	<gd	<gd	<gd	<gd	2.6	4.8	3.7	<gd	<gd	<gd	<gd	11.1
13G	<gd		92.4	92.4	<gd	<gd	<gd	<gd	9.8	18.4	12.4	8.5	<gd	<gd	<gd	49.1
SQGs																
TEC[1]	3.16	4.88	4.16	5.28				59.8	423			150		1.9		1,620
PEC[2]	31.3	28.0	62.9	572				676	2,230			1,450		61.8		22,800
ISQ[3]	1.42	3.54	1.19					34.1	111			31.9		2.85		
ISQ[4]	6.75	8.51	4.77					277	2,355			782		6.67		

[1] Consensus based TEC (*i.e.*, below which harmful effects are unlikely to be observed), MacDonald et al. (2000)
[2] Consensus based PEC (*i.e.*, above which harmful effects are likely to be observed), MacDonald et al. (2000)
[3] ISQG (interim sediment quality guideline - this is generally based on the threshold effects level), CCME (2001)
[4] PEL (probable effects level), CCME (2001)
[5] Sum of *p,p'* and *o,p'* isomers
[6] Represent also sum DDT, since content of opDDT for all sediment samples was below detection limit (river sediment)
[7] Dieldrin

Table 5. Comparison of PAHs, PCBs and DDT content (μg kg^{-1}) in surface sediments from various locations in the world

Location	Year	∑PAHs	∑PCBs	∑DDTs	References
Tonghui River, China	2002		0.78–8.74 (∑12PCBs)	0.11–3.78[b]	Zhang et al. (2004b)
Firth of Clyde, Scotland	1989		0.5–500 (∑7 PCBs)	0.5–262[e]	Kelly and Campbell (1995)
Gulf of Lions, France	1987/91		4.1–8.9 (∑12PCBs)	3.5–11.5[d]	Tolosa et al. (1995)
Aegean Sea, Greece	2001	185-654			Hatzianestis and Sklivagou (2001)
Taihu, China	2005	13-963			Peng et al (2005)
Iznek, Turkey	2010	17-835			Ünlü et al (2010)
Serbian river, this study	2008	MDL-728	MDL-57 (∑7 PCBs)	MDL-310	

[b] Sum of p,p′-DDE, p,p′-DDD and p,p′-DDT
[e] Sum of 4,4-DDE, 4,4-DDD and 4,4-DDT
[d] Sum of 2,4′-DDE, 4,4′-DDE, 2,4′-DDD, 4,4′-DDD, 2,4′-DDT and 4,4′-DDT



Graphical abstract

Highlights

- The persistent organic pollutants (POPs) in rivers and man-made lakes sediments of Serbia were studied.
- The investigated sediments are mainly influenced by pesticides and PAHs
- All investigated sediments accumulated p,p-DDT