

Supplementary Material

Tracing the Legacy of Historical PCB Pollution and Contemporary PAH Contamination in the Kupa River (Danube Basin, Croatia)

Snježana Herceg Romanić ¹, Ivana Jakovljević ¹, Maja Đokić ², Nina Bilandžić ², Goran Jakšić ³, Gordana Mendaš ¹, Martina Biošić ¹, Gordana Pehnc ¹, Tijana Milićević ^{4,*} and Gordana Jovanović ⁴

¹ Institute for Medical Research and Occupational Health, Ksaverska Cesta 2, P.O. Box 291, 10001 Zagreb, Croatia; sherceg@imi.hr (S.H.R.); ijakovljevic@imi.hr (I.J.); gmendas@imi.hr (G.M.); mbiosic@imi.hr (M.B.); gpehnc@imi.hr (G.P.)

² Croatian Veterinary Institute, Savska Cesta 143, 10000 Zagreb, Croatia; dokic@veinst.hr (M.D.); bilandzic@veinst.hr (N.B.)

³ Aquatika-Freshwater Aquarium Karlovac, Ulica Branka Čavlovića Čavleka 1/A, 47000 Karlovac, Croatia; gjaksic@aquariumkarlovac.com

⁴ Institute of Physics Belgrade, National Institute of the Republic of Serbia, University of Belgrade, Pregrevica 118, 11080 Belgrade, Serbia; gordana.jovanovic@ipb.ac.rs

* Correspondence: tijana.milicevic@ipb.ac.rs

Tables:

Table S1. Summary Statistics: Minimum (Min), maximum (Max), average (Mean), and median values (ng g⁻¹) of PCB congeners in 7 fish species – Northern pike (n=5), common carp (n=6), grass carp (n=7), pike-perch (n=5), Wels catfish (n=6), bream (n=4) and chub (n=4).

	Min [ng g ⁻¹]	Max [ng g ⁻¹]	Mean [ng g ⁻¹]	Median [ng g ⁻¹]	Min [ng g ⁻¹]	Max [ng g ⁻¹]	Mean [ng g ⁻¹]	Median [ng g ⁻¹]
Northern pike				Common carp				
PCB28	9.49	10.39	9.73	9.52	9.91	11.18	10.50	10.43
PCB52	14.38	16.97	15.04	14.46	16.04	23.58	17.89	16.93
PCB101	15.03	21.70	18.20	18.31	18.40	40.21	23.42	20.38
PCB118	16.88	25.95	20.66	21.17	20.15	40.61	24.38	21.02
PCB138	17.35	31.58	22.39	22.62	21.32	43.92	25.99	22.15
PCB153	17.25	32.83	22.66	22.38	21.11	41.96	25.58	22.38
PCB180	18.24	20.46	19.00	18.74	18.28	19.56	18.73	18.68
Sum	108.65	157.22	127.67	128.73	126.17	221.02	146.48	132.21
Grass carp				Pike-perch				
PCB28	9.68	11.30	10.30	10.15	9.55	9.75	9.64	9.61
PCB52	14.83	16.55	15.57	15.76	14.50	15.35	14.97	15.00
PCB101	15.24	17.06	16.21	16.39	16.14	17.89	17.03	16.98
PCB118	16.75	18.04	17.36	17.50	18.13	20.12	19.12	18.92
PCB138	17.43	18.72	17.92	17.92	19.25	21.41	20.35	20.20
PCB153	17.43	18.62	17.94	17.92	19.54	22.18	20.72	20.39
PCB180	18.19	18.26	18.21	18.20	18.39	18.73	18.52	18.50
Sum	109.65	116.28	113.50	114.53	115.48	125.21	120.34	119.70
Wels catfish				Bream				
PCB28	9.56	10.54	10.06	10.20	9.49	10.76	10.03	9.93
PCB52	14.36	18.21	16.64	17.08	15.41	16.32	15.86	15.86
PCB101	14.78	33.34	25.13	26.19	16.92	21.14	18.27	17.50

PCB118	16.55	41.98	29.66	31.06	18.36	24.41	20.32	19.25
PCB138	17.22	45.06	31.50	33.75	18.68	30.39	22.32	20.11
PCB153	17.46	45.05	32.74	35.90	20.12	31.74	23.35	20.76
PCB180	18.26	20.48	19.67	19.98	18.36	22.20	19.40	18.51
Sum	108.19	213.75	165.40	174.53	120.18	155.02	129.54	121.47
Chub								
PCB28	<LOD	<LOD	<LOD	<LOD				
PCB52	24.81	45.88	30.82	26.29				
PCB101	27.13	103.24	49.82	34.46				
PCB118	26.53	144.40	60.11	34.76				
PCB138	26.54	145.45	61.49	36.98				
PCB153	26.77	161.56	62.07	29.98				
PCB180	23.46	25.54	24.26	24.01				
Sum	172.63	624.90	299.52	200.27				

Table S2. PAH levels (ng g⁻¹ w. w.) in 7 fish species collected along Kupa (locations available in Figure 1).

Location	Sample label	Fish	Flu	Pir	BaA	Kri	BjF	BbF	BkF	BaP	DahA	BghiP	IP	sum
Karlovac	1	Northern pike	3.7	5.9	/	/	/	/	/	/	/	/	/	9.6
Rečica	2		3.6	6.0	0.1	/	/	/	/	/	/	/	/	9.7
Sela Bosiljevska	3		1.3	2.8	/	/	/	/	/	/	/	/	/	4.1
Rečica	6		0.3	1.8	/	/	/	/	/	/	/	/	/	2.0
Rečica	30		4.6	15.6	/	/	/	/	/	/	/	/	/	20.1
Brođani	8	Pike perch	1.5	5.5	/	/	/	/	/	/	/	/	/	7.0
Rečica	13		2.8	10.2	/	/	0.2	/	/	/	/	/	/	13.2
Brođani	15		2.2	7.9	/	/	0.3	/	/	/	/	/	/	10.3
Rečica	26		0.3	1.0	/	/	/	/	/	/	/	/	/	1.3
Blatnica	29		0.9	3.3	/	/	/	/	/	/	/	/	/	4.2
Husje	5	Wels catfish	0.3	0.7	/	/	/	/	/	/	/	/	/	1.0
Koritinja	16		2.1	7.4	/	/	/	/	/	/	/	/	/	9.5
Koritinja	17		2.4	8.1	/	/	/	/	/	/	/	/	/	10.5
Brođani	20		1.1	3.0	/	/	/	/	/	/	/	/	/	4.1
Šišljavić	24		0.6	2.0	/	/	/	/	/	/	/	/	/	2.6
Zamršje	28	Common carp	0.3	2.1	/	/	/	/	/	/	/	/	/	2.4
Luka Pokupska	9		5.6	16.3	/	/	/	/	/	/	/	/	/	21.8
Luka Pokupska	10		4.9	12.5	/	/	/	/	/	/	/	/	/	17.4
Rečica	18		6.9	14.9	/	/	/	/	/	/	/	/	/	21.8
Rečica	19		4.3	9.9	/	/	/	/	/	/	/	/	/	14.1
Luka Pokupska	22	Bream	7.5	18.6	/	/	/	/	/	/	/	/	/	26.1
Rečica	25		2.9	4.7	/	/	/	/	/	/	/	/	/	7.6
Rečica	12		6.0	14.1	/	/	/	/	/	/	/	/	/	20.1
Brodarci	31		3.1	5.5	/	/	/	/	/	/	/	/	/	8.6
Brodarci	32		1.5	1.8	/	/	/	/	/	/	/	/	/	3.3
Brodarci	33	Grass carp	1.4	3.1	/	/	/	/	/	/	/	/	/	4.5
Koritinja	4		0.6	0.5	0.1	0.2	/	/	/	/	/	0.1	/	1.4
Rečica	7		1.7	4.0	/	/	/	/	/	/	/	/	/	5.8
Rečica	11		1.4	1.9	/	/	/	/	/	/	/	/	/	3.3
Rečica	14		1.0	1.6	/	/	/	/	/	/	/	/	/	2.6
Zamršje	21		2.2	2.3	0.1	/	/	/	/	/	0.1	/	/	4.7

Blatnica	23		2.8	5.8	/	/	/	/	/	/	/	/	8.6
Blatnica	27		0.8	1.1	/	/	/	/	/	/	/	/	1.9
Karlovac	34	Chub	5.6	17.0	/	/	2.4	/	/	/	/	/	25.0
Karlovac	35		1.2	2.9	/	0.2	0.6	/	/	/	/	/	4.9
Karlovac	36		/	0.7	/	/	/	/	/	/	/	/	0.7
Jurovski Brod	37		1.2	9.9	0.1	0.4	/	/	/	/	/	0.1	11.7

/-below LOQ

The LOQ ranged from 0.003 ng g⁻¹ w.w. for BaA to 0.08 ng g⁻¹ w.w. for B_jF, while the LOQ for BaP, as a carcinogen PAH for humans, was 0.005 ng g⁻¹ w.w.

Table S3. Estimated daily intake (EDI; ng/kg/day) for NDL-PCBs, PCB118 and PAHs.

Location	Sam- ple No.	PCB28	PCB52	PCB10 1	PCB118	PCB118 *	PCB138	PCB153	PCB180	Flu	Pyr	Flu*	Pyr*	ΣEDI- PCB	ΣEDI- PAH
Karlovac	1	8.07	12.44	18.02	21.54	0.0006	26.21	27.25	16.98	3.08	4.89	0.0031	0.0049	130.5	7.98
Rečica	2	7.90	12.00	15.20	17.58	0.0005	18.78	19.38	16.02	2.99	4.96	0.0030	0.0050	106.9	7.95
Sela Bos- iljevska	3	8.63	14.09	17.16	18.51	0.0006	18.81	18.58	15.56	1.09	2.34	0.0011	0.0023	111.3	3.43
Koritinja	4	8.78	13.20	13.54	14.53	0.0004	14.88	15.05	15.11	0.47	0.39	0.0005	0.0004	95.1	0.86
Husje	5	7.93	11.92	12.27	13.74	0.0004	14.29	14.50	15.16	0.26	0.56	0.0003	0.0006	89.8	0.82
Rečica	6	7.88	11.96	12.70	14.09	0.0004	14.71	14.52	15.14	0.23	1.45	0.0002	0.0015	91.0	1.69
Rečica	7	8.73	13.08	13.61	14.54	0.0004	14.95	15.06	15.13	1.45	3.33	0.0014	0.0033	95.1	4.78
Brodani	8	8.02	12.64	14.85	16.70	0.0005	17.77	18.41	15.55	1.21	4.59	0.0012	0.0046	103.9	5.80
Luka Pokupska	9	8.71	14.51	17.86	17.92	0.0005	18.94	19.45	15.67	4.63	13.51	0.0046	0.0135	113.1	18.14
Luka Pokupska	10	8.59	13.93	18.22	19.27	0.0006	20.73	20.24	15.69	4.06	10.39	0.0041	0.0104	116.7	14.45
Rečica	11	8.43	13.12	13.63	14.38	0.0004	14.87	14.83	15.11	1.18	1.59	0.0012	0.0016	94.4	2.77
Rečica	12	8.08	12.79	17.55	20.26	0.0006	25.23	26.35	18.43	4.97	11.73	0.0050	0.0117	128.7	16.70
Rečica	13	8.09	12.45	14.09	15.68	0.0005	16.77	16.93	15.35	2.35	8.51	0.0024	0.0085	99.4	10.86
Rečica	14	8.08	12.43	12.65	13.91	0.0004	14.47	14.49	15.10	0.83	1.31	0.0008	0.0013	91.1	2.14
Brodani	15	7.93	12.03	13.39	15.05	0.0005	15.98	16.22	15.26	1.79	6.53	0.0018	0.0065	95.9	8.31
Koritinja	16	8.75	15.11	25.45	28.38	0.0009	29.63	31.81	16.73	1.74	6.17	0.0017	0.0062	155.9	7.91
Koritinja	17	8.44	14.69	27.68	34.85	0.0010	37.41	37.40	16.97	1.96	6.73	0.0020	0.0067	177.4	8.69
Rečica	18	8.89	14.19	15.94	16.73	0.0005	17.84	17.66	15.18	5.70	12.39	0.0057	0.0124	106.4	18.09
Rečica	19	9.28	19.57	33.38	33.71	0.0010	36.46	34.83	16.24	3.53	8.20	0.0035	0.0082	183.5	11.73
Brodani	20	7.98	12.51	16.28	19.21	0.0006	19.52	19.78	15.67	0.88	2.48	0.0009	0.0025	110.9	3.37
Zamrke	21	9.38	13.74	13.93	14.53	0.0004	14.95	14.87	15.12	1.79	1.91	0.0018	0.0019	96.5	3.70
Luka Pokupska	22	8.60	13.31	15.28	16.81	0.0005	17.70	17.70	15.34	6.24	15.47	0.0062	0.0155	104.7	21.70
Blatnica	23	8.42	12.62	14.16	14.97	0.0004	15.54	15.46	15.16	2.36	4.78	0.0024	0.0048	96.3	7.15
Išljavić	24	8.49	13.67	19.84	25.86	0.0008	28.21	31.77	16.44	0.46	1.65	0.0005	0.0017	144.3	2.12
Redo	25	8.23	13.57	15.97	16.98	0.0005	17.77	17.52	15.17	2.42	3.92	0.0024	0.0039	105.2	6.34
Rečica	26	7.98	12.74	14.54	16.22	0.0005	17.20	17.51	15.36	0.29	0.79	0.0003	0.0008	101.6	1.08
Blatnica	27	8.04	12.31	12.65	13.99	0.0004	14.47	14.47	15.10	0.66	0.94	0.0007	0.0009	91.0	1.61
Zamršje	28	8.51	14.97	23.64	25.70	0.0008	27.82	27.83	17.00	0.24	1.77	0.0002	0.0018	145.5	2.01
Blatnica	29	7.98	12.27	13.79	15.70	0.0005	16.76	16.92	15.35	0.74	2.73	0.0007	0.0027	98.8	3.47
Rečica	30	7.89	11.94	12.48	14.01	0.0004	14.41	14.32	15.15	3.80	12.92	0.0038	0.0129	90.2	16.72
Brodarci	31	8.93	13.54	14.40	15.69	0.0005	15.51	16.70	15.29	2.57	4.54	0.0026	0.0045	100.1	7.11
Brodarci	32	8.41	13.40	14.05	15.24	0.0005	16.31	16.92	15.44	1.25	1.52	0.0012	0.0015	99.8	2.77
Brodarci	33	7.88	12.93	14.65	16.27	0.0005	17.08	17.55	15.24	1.14	2.61	0.0011	0.0026	101.6	3.75
Karlovac	34	0.42	20.60	22.52	22.02	0.0007	22.03	22.44	19.47	4.67	14.08	0.0047	0.0141	129.5	18.75
Karlovac	35	0.42	21.44	24.98	24.74	0.0007	25.17	27.33	19.64	0.98	2.38	0.0010	0.0024	143.7	3.35
Karlovac	36	0.42	22.21	32.23	32.97	0.0010	36.23	22.22	21.20	0.01	0.54	0.0000	0.0005	167.5	0.55
Karlovac	37	0.42	38.09	85.70	119.87	0.0036	120.75	134.12	20.22	1.00	8.23	0.0010	0.0082	519.2	9.23

* PCB 118 (normalized to TEF); PAHs (normalized to BaP) for carcinogenic risk assessment.

Figure:

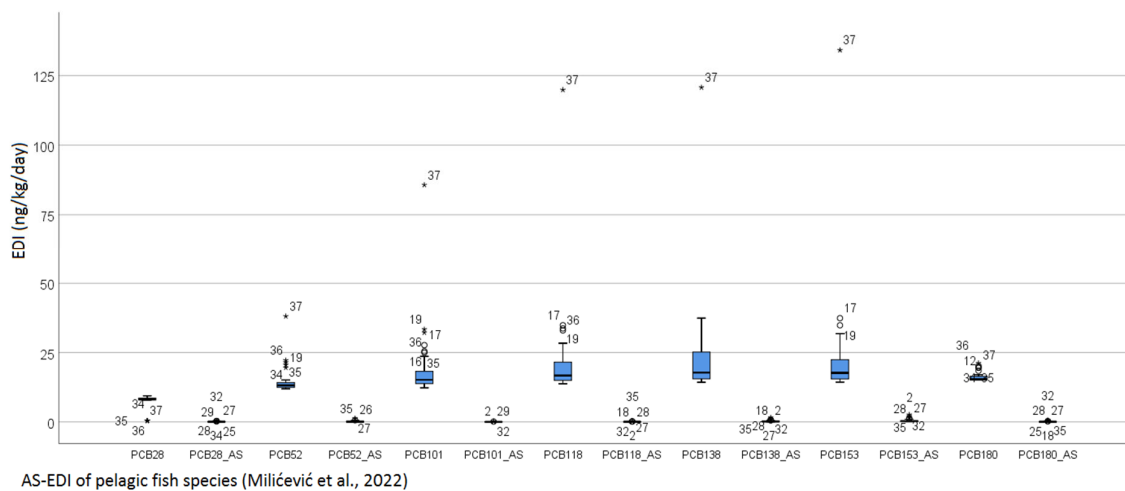


Figure S1. Estimated daily intake – EDI (ng/kg/day) of PCBs and PAHs investigated in fish species from Kupa River and EDI of PCBs investigated for Pelagic fish species (Miličević et al., 2022; *the model from the study was adapted to nowadays data fish consumption per capita in Croatia).