Supplementary Material

Occurrence of contaminants of emerging concern in different water samples from the lower part of the Danube River Middle Basin – A review

Nataša Đurišić-Mladenović[a], Jelena Živančev[a1], Igor Antić[a], Dušan Rakić[a], Maja Buljovčić[a], Biljana Pajin[a], Marta Llorca[b], Marinella Farre[b]

[a]University of Novi Sad, Faculty of Technology Novi Sad, Bulevar cara Lazara 1, 21000 Novi Sad, Serbia

[b]Institute of Environmental Assessment and Water Research (IDAEA), CSIC, C. Jordi Girona, 18-26, Barcelona, 08034, Spain

[1]Corresponding author. Tel.: +381 21 485 3860; Fax: +381 21 450 413.
E-mail addresses: jelena.zivancev@tf.uns.ac.rs, jelena.zivancev@uns.ac.rs (J. Živančev).

Text SI1

To summarize the relevant data reported so far on CECs occurrence in water samples from the countries in the lower part of the Middle Basin of the Danube River, the second-largest European river, a systematic search of Science Direct, American Chemical Society, Web of Science, Google Scholar and Springer Kluwer were performed, using the terms 'contaminants emerging concern', 'emerging pollutant', 'emerging contaminant', 'emerging compound', 'emerging substance', 'micropollutant', 'wastewater', 'Danube', and relevant names of the countries. Reference lists within the chosen studies were also looked through as an additional source of information on studies conducted in the region regarding different classes of CECs. Finally, the studies on CECs presence in various water types from Serbia, Bosnia and Herzegovina, and North Macedonia, representing a part of Western Balkan countries, and Croatia and Slovenia, as the 'upstream' EU States in the lower part of the Middle Danube Basin, have been found and included in this review.

All these countries had a similar development rate in the second half of the 20th century, when they were a part of ex-Yugoslavia, while from the 90-ties they became independent states with specific development paths. Currently, only Croatia and Slovenia are EU Member Countries, while other countries considered here are EU membership candidates, which are often termed in the political sense as 'Western Balkans'. Croatia was considered as the WB country before 2013, when it joined the EU. Similarly, in a study of Terzić et al from 2008 (Terzić et al., 2008), samples from Croatia were considered as from the WB region together with the samples from Serbia and Bosnia and Herzegovina; this is why these results were marked as 'WBC', when the origin of samples is considered in this review, while other results were marked in accordance with the exact country of origin (regardless of year of sampling).

Water resources in this region have always played an important role in the economy of the countries and have been exploited for irrigation, drinking water supply, industrial needs, livestock production and tourism (GRIDArendal, 2015). However, low investments in wastewater treatment and often direct wastewater discharge without treatment are known problems for the WB countries (European Environmental Agency, 2017). Some relevant quantitative information on countries belonging to the region of interest for this review might be found in Table SI1. It is interesting to note that shares of the population connected to at least secondary wastewater treatment plant are generally low for the countries in the region (up to

~30%), except for Slovenia with 68%, which is still less than the average for EUROSTAT estimate for EU-27 in 2021 of 81% (https://ec.europa.eu/eurostat/databrowser/view/sdg_06_20/default/table?lang=en).

As the reviewed papers contained the results of a study-specific set of CECs, the formed database is very complex. The average levels of target compounds either reported by the authors or calculated in this study for samples of surface water and wastewater were taken into consideration here. However, this approach was not applicable to groundwater samples, as some studies reported only ranges (minimum and maximum) of detected levels for some compounds; so, for groundwater, maximum values were considered for discussion here. Literature data related to the presence of CECs in drinking water come from only two studies, and only the maximum detected levels are discussed here. When data were averaged, 'non-detects', i.e. non-quantified results (reported to be not detected, nd, below limits of detection, LOD, limits of quantification, LOQ, method detection limits, MDL, or method quantification limits, MQL) were considered as 0.

In an attempt to describe the collected data in a more general way, CECs were classified arbitrarily based on the distinct final use (including also the metabolites of the parent compounds) as follows: pharmaceutically active compounds, pesticides, personal care products, industrial chemicals, and PFAS; compounds not belonging to any of these classes were gathered in a group named 'others'.

Besides the levels of detected compounds, information on the sampling procedure, preparation, and analysis was gathered, which is important for assessing the regional analytical capacities in relation to the state-of-the-art methodologies for the CECs surveillance.

Table SI2. CECs analyzed in WB water samples

Country	Location & period sampling	Matrix (number of analyzed samples)	CECs	Class (number of analyzed compounds)	Concentrations	Sampling	Sample preparation	Methods[a]	Reference
Western Balkans region (WBs; Bosnia and Herzegovina, Croatia and Serbia)	Sarajevo, Velika Gorica, Bjelovar, Čakovec, Varaždin, Vinkovci, Pula, Rijeka, Split, Zagreb, Novi Zagreb, Karlovac, Sisak, Osijek, Belišće, Zadar, Slavonski Brod, Belgrade & autumn 2004 - spring 2005	Raw municipal wastewaters (19)	Analgesics/anti-inflammatories; Antimicrobials; Sulfonamides; Fluoroquinolones; Psychiatric drugs; Antiulcer agent; Histamine H1 and H2 receptor agonists; β-blockers; Lipid regulator and cholesterol lowering drugs; Barbiturates	Pharmaceuticals (42)	nd[b] - 9450 µg/L	Composite grab samples and a few 24-hour flow-proportional composite sampling	Solid phase extraction (SPE, Oasis HLB cartridges)	LC-MS/MS	Terzić et al., (2008)
			Linear alkylbenzene sulfonates, nonylphenol, nonylphenol ethoxylates, nonylphenoxy acetic acid, nonylphenoxyethoxy acetic acid, octylphenol, octylphenoxy acetic acid	Surfactants & Metabolites (7)					
			Bisphenol A	Plasticizers (1)					
			Atrazin, atrazin–desethyl, atrazin–desisopropyl, dimethoate, epoxiconazole, metamitron, picolinafen, propiconazole, simazine, tebuconazole, terbutryn, terbutylazin, terbutylazin-desethyl	Pesticides & Metabolites (13)				GC/MS	
			Bayrepel, N-diethyl-m-toluamide	Insect repellents (2)					
			Tris-2-chloroethyl phosphate, tris-2-chloropropyl phosphate	Flame retardants (2)					
			Polycyclic musks; Nitro	Personal care					

Table SI1. Basic information (ICPDR, 2021) on the countries belonging to the lower part of the Middle Basin of the Danube River

Country/EU Member or WB region	Coverage in Danube River Basine (km²)	Percentage of Danube River Basine (%)	Population in Danube River Basine (Million)	Percentage of Danube River Basine in the country (%)	Share of the population connected to at least secondary wastewater treatment in 2021[a]
Bosnia and Herzegovina/WB region	36,636	4.6	2.9	74.9	na
Croatia/EU Member	34,965	4.4	3.1	62.5	31[b]
Macedonia/WB region	109	< 0.1	< 0.01	0.2	na
Serbia/WB region	81,560	10.2	7.5	92.3	22[c]
Slovenia/EU Member	16,422	2.0	1.7	81.0	68
Total	169,692	21.2	15.2		

[a]EUROSTAT, https://ec.europa.eu/eurostat/databrowser/view/sdg_06_20/default/table?lang=en

[b]partly neigbouring reference years if not available

[c]some sources reflect even lower shares in 2023 https://naled.rs/en/news-only-147-of-wastewater-in-serbia-gets-treated-malta-is-the-only-one-behind-us-in-europe-8615#:~:text=In%20the%20budget%20of%20the,treatment)%20list%20of%20European%20countries.

na-not available

Table SI2. CECs analyzed in WB water samples

Country	Location & period sampling	Matrix (number of analyzed samples)	CECs	Class (number of analyzed compounds)	Concentrations	Sampling	Sample preparation	Methods[a]	Reference
Western Balkans region (WBs; Bosnia and Herzegovina, Croatia and Serbia)	Sarajevo, Velika Gorica, Bjelovar, Čakovec, Varaždin, Vinkovci, Pula, Rijeka, Split, Zagreb, Novi Zagreb, Karlovac, Sisak, Osijek, Belišće, Zadar, Slavonski Brod, Belgrade & autumn 2004 - spring 2005	Raw municipal wastewaters (19)	Analgesics/anti-inflammatories; Antimicrobials; Sulfonamides; Fluoroquinolones; Psychiatric drugs; Antiulcer agent; Histamine H1 and H2 receptor agonists; β-blockers; Lipid regulator and cholesterol lowering drugs; Barbiturates	Pharmaceuticals (42)	nd[b] - 9450 µg/L	Composite grab samples and a few 24-hour flow-proportional composite sampling	Solid phase extraction (SPE, Oasis HLB cartridges)	LC-MS/MS	Terzić et al., (2008)
			Linear alkylbenzene sulfonates, nonylphenol, nonylphenol ethoxylates, nonylphenoxy acetic acid, nonylphenoxyethoxy acetic acid, octylphenol, octylphenoxy acetic acid	Surfactants & Metabolites (7)					
			Bisphenol A	Plasticizers (1)					
			Atrazin, atrazin–desethyl, atrazin–desisopropyl, dimethoate, epoxiconazole, metamitron, picolinafen, propiconazole, simazine, tebuconazole, terbutryn, terbutylazin, terbutylazin-desethyl	Pesticides & Metabolites (13)				GC/MS	
			Bayrepel, N-diethyl-m-toluamide	Insect repellents (2)					
			Tris-2-chloroethyl phosphate, tris-2-chloropropyl phosphate	Flame retardants (2)					
			Polycyclic musks; Nitro	Personal care					

			musks; Other fragrances	products (Fragrances) (7)					
Serbia	Danube River, Sava River, Tamiš River, Lake Očaga, Belgarde & 2008[c]	Surface water (14), ground water (10), waste water (2)	Antibiotic; Psychiatric drugs; Analgesics/anti-inflammatories	Pharmaceuticals (19)	Surface water: nd - 610 ng/L Ground water: nd - 100 ng/L Waste water: nd - 150 ng/L		Solid phase extraction (SPE, Oasis HLB cartridges)	LC-MS/MS	Grujić et al., (2009)
Serbia	Danube River, Sava River, Tisa River, Morava River & summer and autumn of 2009 and spring and autumn of 2010	Surface (35) and groundwater (35)	Antibiotic; Psychiatric drugs; Analgoantipyretic; Metamizole metabolites	Pharmaceutical (15)	Surface water: nd - 354 ng/L Groundwater: nd - 150 ng/L	Grab samples	Solid phase extraction (Oasis HLB)	LC–MS/MS	Radović et al. (2012)
Serbia	Sava River, Danube River & January and February 2013	Surface water (5)	Psychiatric drug; Hormones	Pharmaceutical (9)	Surface water[d]	Grab samples	liquid/liquid extraction, Solid phase extraction (Supelclaen ENVI-18, Strata C18-E)	GC–MS, LC–MS, LC-DAD	Antonijević et al. (2014)
			Methylbenzophenone 4-Methoxy-2-ethylhexylcinnamate *N,N,N',N'*-Tetraacetylethylene diamine, TAED Galaxolide Tonalide Methyl dihydrojasmonate α-Cadinol Lilial	Personal care products (8)					
			Technical additives	Plasticizers (19)					
			Acetochlor, Uvazol 236, Desethylterbutylazine, Lindane, neonicotinoid insecticides	Pesticides (8)					
			Caffeine	Stimulant/drug					

				(1)					
Serbia	Novi Sad, Zrenjanin, Bečej, Vrbas & spring 2012	Municipal waste water (1), Surface water (11), Underground water (6), Drinking water (5)	Analgesics/anti-inflammatories; Lipid regulators and cholesterol lowering statin drugs; Psychiatric drugs; Histamine H1 and H2 receptor antagonists; β-Blocking agents; Diuretic; Antidiabetic; Antihypertensives; Antiplatelet agent; Prostatic hyperplasia; To treat asthma; Anticoagulant; X-ray contrast agents; Antihelmintics; Synthetic glucocorticoid; Sedation and muscle relaxation; Tranquilizer; Antibiotics; Calcium channel blockers	Pharmaceuticals (81)	Municipal waste water: < nd - 20130 ng/L Surface water: nd - 932 ng/L Underground water: nd - 24.8 ng/L Drinking water: nd - 128 ng/L	Composite grab samples	Solid phase extraction (SPE, Oasis HLB cartridges)	LC-QqLIT-MS/MS	Petrović et al., (2014)
Serbia	Danube River (Novi Sad, Kovin), Sava River, Tisa River, Morava River, observation wells & 3-year period before 2015[c]	Surface water (30), groundwater (44)	Antibiotics; Psychiatric drugs; Analgesics/anti-inflammatories & metabolites	Pharmaceuticals (13)	Surface water: nd - 512 ng/L Ground water: nd - 150 ng/L	Grab samples	Solid phase extraction (SPE, Oasis HLB cartridges)	LC-MS/MS	Radović et al., (2015)
			Insecticides; fungicides; herbicides	Pesticides (12)					
Serbia	Danube River, Tisa River, Morava River, and Pek River & June and October,	Surface water (48)	Herbicides, Insecticides, Fungicides	Pesticides (38)	Surface water: nd - 200 ng/L	Grab samples	Solid phase extraction (SPE, Oasis HLB cartridges, ENVI_18 DSK)	LC–MS/MS, GC–MS	Antić et al., (2015)

	2009; February, April, May, June, September, October, 2010; and June and September, 2011								
Serbia	Danube River (Novi Sad) & November 2012, March 2013, May 2013, September 2013	Surface water (32)	Bisphenol A	Plasticizers (1)	nd - 693 ng/L	Grab samples	Solid phase extraction (SPE, Agilent Bond Elut Plexa cartridges)	GC-MS	Milanović et al., (2016)
Serbia	Danube River, Sava River, Velika Morava River, Tisa River & 2009-2015 autumn each year	Surface water (61); Ground water (123)	Antibiotics; Psychiatric drugs; Analgesics/anti-inflammatories & metabolites; cardiovascular pharmaceuticals	Pharmaceuticals (19)	Surface water: nd - 520 ng/L Ground water: nd to 150 ng/L	Grab samples	Solid phase extraction (SPE, Oasis HLB cartridges)	LC-MS/MS	Kovačević et al., (2017)
Serbia	Danube River, Sava River, and Sava Lake & 2016[e]	surface water (5) and groundwater (2)	Antibiotics; Psychiatric drugs; Antihypertensive; Antihyperlipidemic; Anticoagulant; Analgoantipyretic; Metamizole metabolites	Pharmaceuticals (15)	Surface water: nd - 39.1 ng/L n.d. in groundwater	grab sampling	Solid-phase extraction (Multi-walled carbon nanotubes)	LC-MS/MS	Lalović et al. (2017)
Macedonia	Kriva River, Zletovska River, Bregalnica River and	Surface water (6)	Insecticides, molluscicides, nematicides, and insect growth regulators, herbicides and their metabolites, defoliants,	Pesticides (298)	River water: nd - 549 ng/L	Grab samples	Direct injection	LC-QTOF-MS	Stipaničev et al., (2017)

	Vardar River & May 2015		fungicides and their breakdown products						
			Antiepileptics; Drugs – opioids and their metabolites; Drugs – hypnotics, anticonvulsants, and anesthetics; Drugs – analgesics, cardiovasculars, and neuroleptics; Drugs – antidepressants and their metabolites; Drugs – hallucinogens and stimulants; Drugs – antibiotics/chemotherapeutics and cannabinoids; drugs – hormones/xenoestrogens, diuretics, anticholesteremics, and antiseptics;	Pharmaceuticals (162)					
			1H-benzotriazole	Industrial chemicals (1)					
			PFOA and PFOS	Perfluorinated compounds (2)					
Serbia	Danube River & 2016	Surface water (28)	Antibiotics; Psychiatric drugs; Analgesics/anti-inflammatories	Pharmaceuticals (5);	nd - 621 ng/L	Grab samples	Solid phase extraction (SPE, Supel-Select HLB cartridges)	LC-MS/MS	Milić et al., (2018)
			Benzotriazole	Industrial chemicals (1)					
			Caffeine	Stimulant/drug (1)					

Serbia	Danube River, Tisza River, Begej River, Krivaja River, DTD irrigation canal, Lakes Tikvara and Bođani & 2014	Surface water (18)		Pesticides (398); Pharmaceutical and personal care products (29); Industrial chemicals (262); Plasticizers (14); Fire retardant (13)	nd - 5.54 µg/L	Grab samples	Liquid-liquid extraction without any sample clean-up	GC-MS	Škrbić et al., (2018)
Bosnia and Herzegovina	Bosna River & mid-October to mid-November 2012	Surface water (30)	currently used pesticides	Pesticides (52)	Surface water: nd - 488 ng/L	passive sampling (device used: polar organic chemical integrative sampler (POCIS))	Solid phase extraction (Oasis HLB)	LC-HRMS, HPLC-MS/MS	Toušová et al. (2019)
			Hormones ; Antibiotics; Antihistamins; Cancer treatment; Cardiovascular drugs; Analgesics/anti-inflammatories; Psychoactive drugs; Statins	Pharmaceutical (58)					
			2-oxo-3-hydroxy-LSD, cocaine, MDEA, MDMA, metamphetamine, methadone, oxycodone, benzoylecgonine, clindamycin_sulfoxide, N-desmethylcitalopram, norketamine, norsertraline	Illicit drugs & metabolites (12)					
			Alfuzosin, codeine, dicycloverine, ketamine, loperamide, oseltamivir_carboxylate, pizotifen, terbinafine, terbutaline, caffeine	Others (10)					
Serbia	Šabac & August – September	tertiary effluent (1)	Antibiotics; Antipsychotic; Drugs of abuse, steroids and tobacco ingredients; Hypoglycemic agents and artificial sweeteners;	Pharmaceuticals (203)	tertiary effluent: nd - 1500 ng/L	Averaged 7-day composite effluent wastewater	solid-phase extraction based on automated extraction system -	LC-QTOF, LC-MS/MS	Alygizakis et al. (2019)

	2017		Analgesics; Anesthetics; Antiepileptics; Antihypertensive drugs; Antilipidemic agents; Antiulcer drugs; Contrast agent; Diuretics; Nonsteroidal anti-inflammatory drug; Other; Selective serotonin reuptake inhibitor			sample	HORIZON SPE-DEX 4790		
			Benzotriazoles and Others; Perfluorinated substances; Phenols; Phosphates; Phthalate esters; Surfactants	Industrial chemicals (34)					
			Insecticides	Pesticides (42)					
			Benzophenone 3	UV filter (1)					
Serbia	Belgrade, Novi Sad, Zemun, Pančevo, Zrenjanin, Subotica, Šabac, Danube River, Sava River, Tisa River, DTD irrigation canal, Veliki Bačka canal, lakes Palić and Zobnatica &	Untreated municipal wastewater (30); Surface water (30); Drinking water (30)	Hormones & metabolites	Pharmaceuticals (10);	Wastewater: nd - 40.9 ng/L Surface water: nd - 31.2 ng/L Drinking water: nd - 35.6 ng/L	Grab samples	On-line solid-phase extraction (SPE)	LC-MS/MS	Čelić et al., (2020)
			Alkylphenols; Bisphenol A	Industrial chemicals (3)					

	2019*								
Serbia	"Vodokanal" Sombor municipal WWTP & September and October 2017	WW influent (3); Primary treatment WW (3); WW effluent (3)	Hormones; Lipid regulators and cholesterol lowering statin drugs; Analgesics/anti-inflammatories & meatbolites; Calcium channel blockers; Psychiatric drugs	Pharmaceuticals (13)	WW influent: nd - 33400 ng/L; Primary treatment WW: nd - 18300 ng/L; WW effluent: nd - 12300 ng/L	flow proportional composite sampling over a 10 h working day	Solid-phase extraction (SPE, Oasis HLB cartridge)	GC-MS	Bogunović et al., (2021a)
			Caffeine	Stimulant (1)					
			UV filters; Sunscreen; Parabens; Antimicrobial agent	Personal care product (13)					
			Bisphenol A, 2,2′-methylenediphenol, 4,4′-biphenol, bisphenol AF, bisphenol AP, bisphenol C, bisphenol E, bisphenol F, bisphenol FL, bisphenol M, bisphenol BP, bisphenol P, bisphenol S, bisphenol Z, 4-cumylphenol, bisphenol B, bisphenol Cl, bisphenol PH	Plastisizers (18)					
			4,4′- dihydroxydiphenyl ether	Brominated flame retardants (1)					
			mecoprop	Pesticide (1)					

			Nonylphenol	Surfactant (1)					
Serbia	Danube River, Novi Sad & 2017	Surface water (9)	UV-filters	Personal care products (2)	Surface water: Benzofenone: nd - 0.95 µg/L Benzofenone-3: nd - 0.62 µg/L Caffeine: nd - 0.7 µg/L	grab samples	Liquid-liquid extraction	GC-MS	Bogunović et al., (2021b)
			Caffeine	Stimulant (1)					
Serbia	Danube River, Belgrade, Novi Sad, Inđija, Stara Pazova, Nova Pazova, Batajnica & 2014	Municipal wastewater (8); Surface water (12)	Perfluorobutane sulfonic acid, perfluorooctane sulfonic acid, perfluorobutanoic acid, perfluorohexanoic acid, perfluoroheptanoic acid, perfluorooctanoic acid, perfluorononanoic acid, perfluorodecanoic acid, perfluoroundecanoic acid, perfluorododecanoic acid, perfluorooctane sulfon-amide	Perfluorinated compounds (11)	Municipal wastewater nd - 7.38 ng/L Surface water: nd - 14.9 ng/L	12 composite grab samples, 8 composite 24-h WW samples	Solid phase extraction (SPE, Oasis HLB cartridges)	LC-MS/MS	Buljovčić et al., (2022)

[a] Different symbols used for liquid chromatography in cited studies, either HPLC or UHPLC, have been marked uniformly as LC here, while tandem mass spectrometry (sometimes labelled as QqQ in literature) has been identified uniformly as MS-MS.
[b] nd (not detected) was used instead of different terms for minimum detection levels used in the cited studies (e.g. LOD, LOQ) to represent the reported ranges of concentrations in a uniform way
[c] assumed to be a year before the publication year
[d] For most compounds analyzed, the authors only reported whether the compound was identified or not, except for hormones (which ranges were from nd to 0.57 ng/L)
WW-wastewater, WWTP-wastewater treatment plant

Table SI3. CECs analyzed in water samples of Slovenia and Croatia

Country	Location & period	Matrix (number	CECs	Class (number	Concentration	Sampling	Sample	Method	Referenc

	sampling	of analyzed samples)		of analyzed compounds)	s		preparation	s[a]	e
Slovenia	Slovenian towns A, B, C & winter and spring 2011	River before (1) and after (1) municipality and pharmaceutical industry, hospital effluent (4), WWTP influent (2) and effluent (4), stream before (2) and after (2) effluent, river before (1) and after (2) WWTP discharge	Psychiatric drugs	Pharmaceuticals (3)	River before and after municipality - A and pharmaceutical industry: nd[b] hospital effluent: nd - 72 ng/L WWTP influent: nd - 58 ng/L WWTP effluent: nd - 133 ng/L Stream before effluent: 6-28 ng/L Stream after effluent: 19-31 ng/L river before WWTP discharge: nd - 9 ng/L river after WWTP discharge: nd - 69 ng/L	24 h time-proportional samples and grab sampling	Solid phase extraction (Oasis)	GC-MS	Kosjek et al. (2012)
Slovenia, Croatia	Lake Bled, Lake Bohinj, Lake Šobec, Lake Rakitna, and Lake Bloke, Krka River, Nadiža River, Kolpa River, Ljubljanica River (close to a WWTP	surface waters (river (6), lake (5), and sea (3))	ketoprofen, 3-ethylbenzophenone and 3-acetylbenzophenone	Pharmaceuticals (3)	River: nd -120 ng/L Lake: nd - 820 ng/L Sea: nd - 380 ng/L River (WWTP): nd - 2900 ng/L	grab sampling	Solid phase extraction (Oasis HLB)	GC-MS	Kotnik et al. (2014)
			benzophenone, 4-hydroxybenzophenone, 2-hydroxy-4-methoxybenzophenone, 2,4-dihydroxybenzophenone and 2,2′-dihydroxy-4-	UV filters (5)					

	outflow), Krka River (close to a WWTP outflow) and Kamniška Bistrica River (close to a WWTP outflow), three sites on the North Adriatic Sea (Portorož, Ankaran and Novigrad) & July–September 2013		methoxybenzophenone						
Slovenia	Maribor (aquifer Vrbanski plato), Drava River & May 2010 to October 2011	groundwater (56) and surface water (4)	Psychiatric drugs; Analgesic caffeine atrazine, desethylatrazine, deisopropylatrazine, terbuthylazine, desethylterbuthylazine, metolachlor, simazine and propazine, diuron	Pharmaceuticals (2) Stimulant (1) Pesticides (9)	*	Groundwater samples were collected from piezometers. Grab sampling for surface water	solid phase extraction		GC-MS	Koroša & Mali (2015)
Slovenia	Maribor (Vrbanski plateau aquifer), Drava River & May 2010 to October 2011	groundwater (56) and surface water (4)	Psychiatric drugs; Analgesic caffeine 2-methyl-2H-benzotriazole and 2.4-dimethyl-2H-benzotriazole atrazine, desethylatrazine, deisopropylatrazine, terbuthylazine, desethylterbuthylazine, metolachlor, simazine, propazine	Pharmaceuticals (2) Stimulant (1) Industrial chemicals (2) Pesticides (8)	Groundwater: 1.4 – 273.3 ng/L Surface water: nd – 108.4 ng/L	Groundwater samples were taken from boreholes in filter areas. Grab samples for surface water	solid phase extraction		GC-MS	Koroša et al. (2016)
Slovenia	Ljubljana & January 2014, June 2014	hospital effluents (2) and WWTP influents (2) and effluents (1)	anticancer drugs, metabolites or transformation products	Pharmaceuticals (22)	hospital effluents: nd - 60600 ng/L WWTP influents:	24 h time-proportional samples, and grab samples	Solid phase extraction (Oasis HLB Waters),		GC-MS, LC-QqLIT – MS/MS	Isidori et al. (2016)

					nd - 366 ng/L WWTP effluents: nd - 17 ng/L		on-line SPE		
Slovenia	Bled, Bohinjska Bistrica, Šoštanj, Ljubljana, Domžale-Kamnik, Rakitna, Novo mesto, Piran, Koper, Sava Bohinjka, Paka, Ljubljanica, Kamniška Bistrica, Krka & Summer, Autumn and Winter, August 2016 to February 2017	Municipal and industrial WW, influent (27) and effluent (27), surface water (43)	Bisphenol A, 2,2′-methylenediphenol, 4,4′-biphenol, bisphenol AF, bisphenol AP, bisphenol C, bisphenol E, bisphenol F, bisphenol FL, bisphenol M, bisphenol BP, bisphenol P, bisphenol S, bisphenol Z, 4-cumylphenol, bisphenol B, bisphenol Cl, bisphenol PH	Plasticizers (18)	Influent: nd -62600 ng/L Effluent: nd -7630 ng/L Surface water before discharge: nd -671 ng/L Surface water after discharge: 0.105-2630 ng/L	24-h time proportional samples for wastewater grab samples for surface water	Solid phase extraction (Oasis HLB Prime)	GC-MS	Česen et al. (2018)
			UV filters; Sunscreen; Parabens; Antimicrobial agent	Personal care products (13)					
			4,4′- dihydroxydiphenyl ether	Brominated flame retardants (1)					
			Hormones; Lipid regulators and cholesterol lowering statin drugs; Analgesics/anti-inflammatories & meatbolites; Calcium channel blockers; Psychiatric drugs	Pharmaceuticals (13)					
			Caffeine	Stimulant (1)				—	
			Herbicide	Pesticide (1)					
			Nonylphenol	Surfactant 1)					
Slovenia	Ljubljana, Domžale-Kamnik, Novo mesto, Velenje, Golnik & during August and October 2015	WW influents and effluents (18), WW influent mixed with the WW from cistern trucks from surrounding industries (4), samples of various potential source	bisphenol AF, bisphenol AP, bisphenol B, bisphenol C, bisphenol E, bisphenol F, bisphenol S and bisphenol Z	Plasticizers (8)	WW influents: nd - 403 ng/L WW effluents: nd - 85.7 ng/L WW influents from cistern trucks: nd - 85.7 ng/L WW inflows: nd - 238 ng/L	24 h time-proportional samples, and grab sampling	Solid phase extraction (Oasis HLB Waters)	GC–MS	Česen et al. (2018a)

		wastewater (WW inflows) (25)							
Slovenia	Kokra River, Ledava River, Paka River, and Savinja River, Zbilje lake & spring 2016	Surface water (6)	Psychiatric drugs; Analgesics/anti-inflammatories; β-Blocking agents; Antibiotics; Caffeine (stimulant); Selective serotonin re-uptake inhibitors; Diuretic; Lipid regulators and cholesterol lowering statin drugs; Antihypertensive; Antihiperglycemic agent; Calcium-channel blocker; Proton-pump inhibitors; Histamine H1 and H2 receptor antagonists; To treat asthma	Pharmaceuticals (n=44)	Surface water: nd – 47.23 ng/L	grab sampling	Solid-phase extraction (Strata X columns)	LC-MS/MS	Klančar et al. (2018)
Slovenia & Croatia	WWTP at Ljubljana and Zagreb, and Sava River & May – July 2017	Municipal WW effluents (12); Sava River (14)	Bisphenol A, 2,2′-methylenediphenol, 4,4′-biphenol, bisphenol AF, bisphenol AP, bisphenol C, bisphenol E, bisphenol F, bisphenol FL, bisphenol M, bisphenol BP, bisphenol P, bisphenol S, bisphenol Z, 4-cumylphenol, bisphenol B, bisphenol Cl, bisphenol PH	Plasticizers (18)	Wastewater effluents: 0.0367 - 49,600 ng/L Sava River: 0.0649 - 1390 ng/L	24-h time proportional samples except 3 grab WW samples	Solid phase extraction (SPE, Oasis HLB cartridges)	GC-MS	Česen et al., (2019)
			UV filters; Sunscreen; Parabens; Antimicrobial agent	Personal care products (13)					
			4,4′- dihydroxydiphenyl ether	Brominated flame retardants (1)					
			Hormones; Lipid regulators and cholesterol lowering statin drugs; Analgesics/anti-inflammatories & meatbolites; Calcium channel blockers; Psychiatric drugs	Pharmaceuticals (13)					
			Caffeine	Stimulant (1)					
			Herbicide	Pesticide (1)					

			Nonylphenol	Surfactant (1)					
Croatia	WWTPs of the cities of Čakovec and Velika Gorica, Sava, Drava and Danube & June 2006 and February 2007, April and May 2005	Raw municipal WW (2), WW secondary effluent (2) and river water (4)	Sulfonamides; trimethoprim; fluoroquinolones; macrolides	Pharmaceutical (13)	Raw municipal wastewater: nd - 4664 ng/L Wastewater secondary effluent: nd -3176 ng/L River water: nd -162 ng/L	grab samples	Solid phase extraction (Oasis HLB)	LC-MS/MS	Senta et al. (2008)
Croatia	Zagreb, the central wastewater treatment plant (WWTP-mechanical and biological treatment) of the city of Zagreb & February to September 2009	Raw WW (39); secondary effluent (39)	Morphine, 6-Acetyl morphine, amphetamine, 3,4-methylenedioxymethamphetamine, benzoylecgonine, cocaine, 11-nor-9-carboxy- Δ^9 -tetrahydrocannabinol, methadone, 2-ethylidene-1,5- dimethyl-3,3-diphenylpyrrolidine, codeine	Illicit drugs (10)	Raw wastewater: nd - 476 ng/L Secondary effluent: nd - 149 ng/L	24-h volume-proportional composite samples of both untreated wastewater (raw wastewater; RW) and biologically treated wastewater (secondary effluents, SE)	Solid phase extraction (SPE, Oasis HLB cartridges)	LC-MS/MS	Terzic et al., (2010)
Croatia	Belisce, Bjelovar, Cakovec, Karlovac, Novi Zagreb, Osijek, Rijeka, Slavonski Brod, Sisak, Split, Varazdin, Vinkovci, Velika Gorica, Zadar, Zagreb &	Raw municipal WW (15), WW secondary effluent (2), mechanically treated effluent (2), and mixed liquor (4) collected from the aeration tank of WWTP	Sulfonamides; trimethoprim; fluoroquinolones; macrolides	Pharmaceutical (14)	Raw municipal WW - dissolved fraction: nd -11555 ng/L Raw municipal WW - particulate	grab samples, twenty-four-hour composite samples	Solid phase extraction (Oasis HLB)	LC-MS/MS	Senta et al. (2013)

	April and May 2005 March to September 2009				fraction: nd -4380 ng/L WW secondary effluent: nd -1352 ng/L Mixed liquor, collected from the aeration tank of WWTP (aqueous phase, December 2011-January 2012): 40-9700 ng/L				
Croatia	Zagreb, WWTP, Sava River & 2016[c]	Municipal WW (3); Secondary effluent (2); River water (3)	Macrolide antibiotics, their synthesis intermediates and transformation products	Pharmaceuticals (19)	Municipal wastewater: nd - 9.70 µg/L Secondary effluent: nd - 1.06 µg/L River water: nd - 19 µg/L	Grab samples	Solid phase extraction (SPE, Oasis HLB cartridges)	LC-MS/MS	Senta et al. (2017)
Croatia	Zagreb, Sava River & 2017[c]	raw wastewater (4), secondary effluent (2) and river water (2)	opioid analgesics & metabolites (including 2 morphine glucuronide conjugates)	Pharmaceuticals (27)	Raw WW: nd - 752 ng/L Secondary effluent: nd - 890 ng/L River water: nd - 72 ng/L	24 h-composite samples, and grab sampling	Solid phase extraction (Oasis HLB)	LC–MS/MS	Krizman-Matasic et al. (2018)
Croatia	WWTP of the city of Zagreb &	WW influent (29) and secondary effluent (29)	Macrolide antibiotics, their synthesis intermediates and transformation products	Pharmaceutical (15)	Raw municipal WW: nd - 22730	Twenty-four-hour composite wastewater	Solid phase extraction (Oasis	LC-MS/MS	Senta et al. (2019)

	February to December 2017				ng/L WW secondary effluent: nd - 20178 ng/L	samples	HLB, Strata SAX cartridges)		
Croatia, Slovenia	Zagreb, Varaždin, Ljubljana, Vipap (Krško) & August, September 2017	secondary effluent (4), tertiary effluent (2)	Antibiotics; Antipsychotic; Drugs of abuse, steroids and tobacco ingredients; Hypoglycemic agents and artificial sweeteners; Analgesics; Anesthetics; Antiepileptics; Antihypertensive drugs; Antilipidemic agents; Antiulcer drugs; Contrast agent; Diuretics; Nonsteroidal anti-inflammatory drug; Other; Selective serotonin reuptake inhibitor	Pharmaceuticals (203)	secondary effluent (Zagreb): nd - 475.6 ng/L secondary effluent (Varaždin): nd - 4251.2 ng/L secondary effluent (Ljubljana): nd - 3050.7 ng/L secondary effluent (Vipap): nd - 893.5 ng/L	Averaged 7-day composite effluent wastewater sample	solid-phase extraction based on automated extraction system - HORIZON SPE-DEX 4790	LC-ESI-QTOF, LC-MS/MS	Alygizakis et al. (2019)
			Benzotriazoles and Others; Perfluorinated substances; Phenols; Phosphates; Phthalate esters; Surfactants	Industrial chemicals (34);					
			Insecticides	Pesticides (42)					
			Benzophenone 3	UV filter (1)					
Croatia	Aquifer below the Gorjak Streambed	Groundwater (4)	Macrolide antibiotics, their synthesis intermediates and transformation products	Pharmaceutical (15)	Groundwater: nd -1143 ng/L	Groundwater sampling was performed by piezometers.	Solid phase extraction (Oasis HLB)	LC-MS/MS	Senta et al. (2021)
Croatia	Jadro spring, Žrnovnica spring, Cetina River, and Gizdavac borehole &	Surface water (21), groundwater (7)	DEET, atrazine-desethyl, clothianidin, bentazone	Pesticide (4)	Surface water: 0.3 - 372 ng/L Groundwater: nd - 40.8 ng/L	grab samples		LC- Q-TOF-MS, LC-MS-MS	Selak et al. (2022)
			1H-Benzotriazole	Industrial chemicals (1)					
			Antidiabetic and antihyperglycemic;	Pharmaceutical (11)					

	March and October of 2019 March, July, September, and November 2020		Analgesic/anti-inflammatory; Psychiatric drugs; Antihypertensive; Antibiotic						
			Sucralose, caffeine, cotinine, acesulfame	Lifestyle product (4)					
			Climbazole	Personal care products (1)					
Croatia	Sava & May 2018	Surface water (5)	PFHxDA, PFODA, 1-H benzotriazole, 4-nitrophenol, 4-Nonylphenol, 4-tert-Octylphenol, Bisphenol A, HBCDD, NP1EO, NP2EO, OP1EO, OP2EO, PFBA, PFBS, PFDA, PFDoA, PFDoS, PFDS, PFHpA, PFHpS, PFHxA, PFHxS, PFNA, PFNS, PFOA, PFOS, PFPA, PFPeS, PFTA, PFTrA, PFUdA,	Industrial chemicals (31)	Surface water: nd – 1.26 µg/L	Grab samples	Direct injection	LC-QTOF-MS	Malev et al. (2022)
			Antiepileptics/Neuroleptics, Hormones, Antibiotics, Steroidal anti-inflammatory drugs, Hypnotics/Anticonvulsants/ Anesthetics, Opioids, Antiparasitics and antifungal agents Cardiovascular medicals, Antidepressants, Analgesics, Hallucinogens/Stimulants, Antibiotics, Diuretics, Cannabinoids/Illicit drugs and metabolites, Contrast agents	Pharmaceutical (247)					
			Herbicides, Insecticides, Fungicides	Pesticides (286)					
Slovenia	Domžale-Kamnik WWTP & March 2021	WWTP influent (1), primary settler influent (1), primary settler effluent	4,4'-sulfonyldiphenol, 2-[(2-hydroxyphenyl)methyl]phenol, 2-[(4-hydroxyphenyl)methyl]phenol,	Bisphenols (16)	WWTP influent: nd - 434 ng/L Primary settler influent:	Twenty-four-hour composite samples	solid-phase extraction (Oasis HLB)	GC-MS	Vehar et al. (2022)

	(1), WWTP effluent (1)	4-[(4-hydroxyphenyl)methyl]phenol, 4-[1-(4-hydroxyphenyl)ethyl]phenol, 4-[2-(4-hydroxyphenyl)propan-2-yl]phenol, 4-[2,2-dichloro-1-(4-hydroxyphenyl)ethenyl]phenol, 4-[2-(4-hydroxyphenyl)butan-2-yl]phenol, 4-[1,1,1,3,3,3-hexafluoro-2-(4-hydroxyphenyl)propan-2-yl]phenol, 4-[2-(4-hydroxy-3-methylphenyl)propan-2-yl]-2-methylphenol, 4-[1-(4-hydroxyphenyl)-1-phenylethyl]phenol, 4-[1-(4-hydroxyphenyl)cyclohexyl]phenol, 4-[(4-hydroxyphenyl)-diphenylmethyl]phenol, 4-[9-(4-hydroxyphenyl)fluoren-9-yl]phenol, 4-[2-[4-[2-(4-hydroxyphenyl)propan-2-yl]phenyl]propan-2-yl]phenol, 4-[2-(4-hydroxy-3-phenylphenyl)propan-2-yl]-2-phenylphenol	nd - 577 ng/L Primary settler effluent: nd - 591 ng/L WWTP effluent: nd - 20 ng/L

[a] Different symbols used for liquid chromatography in cited studies, either HPLC or UHPLC, have been marked uniformly as LC here, while tandem mass spectrometry (sometimes labelled as QqQ) has been identified uniformly as MS-MS.
[b] nd (not detected) was used instead of different terms for minimum detection levels used in the cited studies (e.g. LOD, LOQ) to represent the reported ranges of concentrations in a uniform way
[c] assumed to be a year before the publication year
*the reported range of concentrations is not clearly linked to the water type, so these results were not considered here
WW-wastewater, WWTP-wastewater treatment plant

Table SI4. Frequency of detection (in %) of the 8 most frequently analyzed compounds in surface water as reported in cited studies collected for this review

	Azithromycin	Bisphenol A	Caffeine	Carbamazepine	Diclofenac	Ibuprofen	Trimethoprim	Venlafaxine
				WBCs				
Stipaničev et al., 2017	67	100	100	83		100	50	33
Toušová et al. 2019	11		100	100	89		89	89
Čelić et al., 2020		33						
Milanović et al., 2016		69						
Škrbić et al., 2018		39	80	50		5		
Milić et al., 2018			100	100		54		
Lalović et al. 2017				40				
Petrović et al.,				73	27	18	9	27

2014								
	Croatia and Slovenia							
Senta et al. 2008	25						75	
Senta et al. 2017	67							
Klančar et al. 2018			100	50				67
Česen et al 2018		30	83	47	87	60		
Česen et al 2019		36	93	100	43	50		
Malev et al. 2022	40	100	100	100	80	80	100	100

Table SI5. Main strengths and gaps in knowledge and capacities for the wide-range surveillance of CECs in waters within the lower part of the Middle Danube Basin

	Strengths	Gaps/weaknesses
Issue of CECs occurrence in water	• CECs in waters are common problem regardless of the political and economic status of the countries, and different wastewater management and control systems • It is a global challange and importance of the issue goes well beyond the region, as CECs, particularly those persistent and mobile, have no barrier to their flow within river waters, affecting the quality of groundwater, drinking water, biodiversity, living, etc., with the ultimate impact on the seawater into which it flows	• There have not been any systematic monitoring within the region, only those conducted within the large international surveys (such as Join Danube Surveys) and sporadic research studies, giving snapshots on the occurrence of study-specific compounds, without possibility to follow the spatial and temporal trends, nor to perform prioritization of river basin-specific pollutants
Instrumentation	• Available GC-MS and LC-MS instrumentations, including those with HRMS	• Available HRMS instruments have not been used for screening and non-target analysis • Rare availability of specially dedicated software tools for HRMS data processing
Analytical experience and knowledge	• Well proven research experience in target analysis of environmental contaminants, especially in Croatia and Serbia	• Scarce, if any, involvement in screening and non-target analysis • Unevenly distrubuted studies on CECs throughout the region; some WB countries have not any research study on CECs in the environment (Montenegro, Albania), while in others (e.g. N. Macedonia) the available studies came out of the international cooperation without which further investigations stopped
Funding of research monitoring	• Proven examples of international projects that have covered the expensive target analysis of CECs	• Low national investments of the research activities, particularly in the WB countries, limit the scope of target

	• EU member states have more sources to fund the purchase of expensive instrumentations and consumables for the CECs analysis	analysis and the range of CECs covered, investment into the latest, highly expensive instrumentation such as modern HRMS and necessary accessories/consumables
Cooperation	• Proven international network and projects involving researchers from the region	• Rare cases of regional and/or bilateral cooperation with interest in the CECs monitoring, which hinder establishment of strong platform for regional exchange of knowledge, achievements, best practice, and support to the policy-makers, monitoring efforts, and community
Supportive legislation framework	• EU member states are obliged to follow the latest, more strigent directives on the environmental contaminants, asking for the innovative solutions and approaches also to the problems induced by CECs • Water Framework Directive and „sister" directives on drinking and groundwater requires monitoring of some CECs, while mechanism of watch lists stimulates the quest for new data and the prioritization	• The WB countries follows the EU strategy on environmental protection but there are no strict measures of control and monitoring, neither the transparent reports and communication of the data on environmental contamination

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