Long-Term Trends and Patterns of Legacy and Emerging Cosmetic UV Filters in German Waters

Additional file 1

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Table S1: General information on the UV filters investigated.

Analyte	Acronym	CAS No.	Purity (%)	Supplier
Camphor benzalkonium methosulfate	CBM	52793-97-2	99.8	trc-canada (Canada)
4-aminobenzoic acid	PABA	150-13-0	99.1	LGC Labor GmbH (Germany)
Methoxypropylamino cyclohexenylidene ethoxyethylcyanoacetate	S87	1419401-88-9	99.7	WITEGA Laboratorien Berlin-Adlershof GmbH (Germany)
Ensulizole	PBSA	27503-81-7	93.7	LGC Labor GmbH (Germany)
Sulisobenzone	BP-4	4065-45-6	95	HPC Standards (Germany)
2,2',4,4'-Tetrahydroxybenzophenone	BP-2	131-55-5	97	Merck (Germany)
2,3,4-Trihydroxybenzophenone	THB	1143-72-2	97	Merck (Germany)
Benzylidene Camphor Sulfonic Acid	BCSA	56039-58-8	85	LGC Labor GmbH (Germany)
Ecamsule	TDSA	92761-26-7	99.4	Merck (Germany)
Oxybenzone	BP-3	131-57-7	99.9	HPC Standards (Germany)
5-Chloro-2-hydroxybenzophenone	BP-7	85-19-8	99	Merck (Germany)
Amiloxate	IMPC	71617-10-2	98	Merck (Germany)
3-Benzylidenebornan-2-one	3-BC	15087-24-8	97	Merck (Germany)
Drometrizole	UV-P	2440-22-4	99	abcr (Germany)
4-Methylbenzylidene camphor	4-MBC	36861-47-9	99.9	Merck (Germany)
Avobenzone	BMDBM	70356-09-1	99	HPC Standards (Germany)
Homosalate	HS	118-56-9	98.7	LGC Labor GmbH (Germany)
Padimate O	OD-PABA	21245-02-3	98	Merck (Germany)
Octinoxate	EHMC	5466-77-3	98	Merck (Germany)
Octisalate	EHS	118-60-5	99.1	LGC Labor GmbH (Germany)
1,2,4-Triazine, 3,3'-(1,4-phenylene)bis[5,6-diphenyl-; 3,3'-(1,4-Phenylene)bis[5,6-diphenyl-1,2,4-triazine]	S86	55514-22-2	97.5	LGC Labor GmbH (Germany)
Diethylamino hydroxybenzoyl hexyl benzoate	DHHB	302776-68-7	98	Merck (Germany)
Octocrylene	OC	6197-30-4	99.9	HPC Standards (Germany)
Tris-biphenyl triazine	TBT	31274-51-8	98	abcr (Germany)
Bemotrizinol	BEMT	187393-00-6	99.0	Merck (Germany)
Iscotrizinol	DBT	154702-15-5	96	Merck (Germany)
Bisoctrizole	MBBT	103597-45-1	99.5	LGC Labor GmbH (Germany)
Ethylhexyl triazone	EHT	88122-99-0	99.4	HPC Standards (Germany)
Oxybenzone-d$_5$	BP-3-d$_5$	1219798-54-5	99	HPC Standards (Germany)
Octocrylene-[^{13}C]$_3$	OC-[^{13}C]$_3$		99.7 / 99.7	trc-canada (Canada)
4-Methylbenzylidene camphor-d$_4$	4-MBC-d$_4$	1219806-41-3	98 / 98.3	trc-canada (Canada)
Drometrizole-d$_3$	UV-P-d$_3$		97 / 98.3	trc-canada (Canada)
Iscotrizinol-d$_4$	DBT-d$_4$		99.7 / 98.1	trc-canada (Canada)
Avobenzone-[^{13}C]-d$_3$	BMDBM-[^{13}C]-d$_3$		98 / 99.5	trc-canada (Canada)

Table S2: Information on LC-MS/MS system and parameters.

LC/Q-TOF equipment and conditions			
Instrument	Agilent 6545 LC/Q-TOF. Quadrupole Time of Flight LC/MS, LC-System: Agilent 1290 Infinity II, Ion Source: Jet Stream Technology Ion Source (AJS), Agilent Technologies Deutschland GmbH, Waldbronn.		
Pre-Column	SecurityGuard Cartridges, Phenyl 4 × 2.0 mm ID. Phenomenex Ltd. Deutschland, Aschaffenburg.		
Column	ZORBAX RR Eclipse XDB-C18. Dimensionen: 80Å, 2.1 × 150 mm, 3.5 µm, Agilent Technologies Deutschland GmbH, Waldbronn.		
Mobile Phase A	0.1 % formic acid in water		
Mobile Phase B	0.1 % formic acid in acetonitrile		
Flow rate	0.5 mL/min		
Gradient	Time	Profile	
	min	%A	%B
	0.0	100	0
	0.0	98	2
	1.0	98	2
	2.0	80	20
	16.5	2	98
	23.0	2	98
	23.1	98	2
	25.1	98	2
Run Time	26.1 min		
Column Temperature	40 °C		
Injection Volume	20 µL		
Q-TOF-Program	**Time**	**LC-Stream**	**Ionpolarity**
	0.00 min	Waste	positive
	2.00 min	MS	positive
	19.00 min	MS	negative
	22.00 min	Waste	positive

ESI-Parameter	**positive**	**negative**
Gas Temp (°C)	278	278
Drying Gas (L/min)	9.2	8.7
Nebulizer (psi)	19	39
Sheath Gas Temp (°C)	264	264
Sheath Gas Flow (L/min)	11.6	10.1
VCap (V)	3899	2444
Nozzle Voltage (V)	1148	92
Fragmentor (V)	158	375
Skimmer (V)	65	65
Oct 1 RF Vpp (V)	750	750

Table S3: Information on substance-specific parameters for analytes detected by LC-MS/MS.

Analyte	Prec (m/z)	Quantifier (CE) Qualifier (CE)	Retentiontime (min)	Acquisition Time (ms/spec)
S87	323.1962	233.1284 (15) 201.1017 (30)	8.35	300
BP-3	229.0858	77.0389 (40) 105.0336 (20)	11.00	300
BP-3-d_5	234.1170	151.0384 (20) 82.0700 (40)	11.00	75
UV-P-d_3	229.1159	121.0617 (20) 82.0735 (50)	13.00	75
UV-P	226.0972	77.0387 (40) 107.0491 (15)	13.05	300
3-BC	241.1579	91.0545 (40) 143.0847 (20)	13.20	250
4-MBC-d_4	259.1993	216.1433 (20) 160.1195 (15)	14.00	75
4-MBC	255.1739	105.0699 (30) 111.0800 (20)	14.05	300
IMPC	249.1475	161.0594 (15) 179.0695 (5)	14.30	300
DHHB	398.2314	149.0233 (30) 240.1016 (30)	15.35	150
BMDBM	311.1638	161.0955 (30) 135.0432 (30)	15.70	200
BMDBM-[^{13}C]-d_3	315.1850	161.0952 (30) 139.0644 (30)	15.70	75
OC	379.2376	250.0854 (10) 232.0756 (20)	16.07	75
OC-[^{13}C]$_3$	382.2476	253.0960 (10) 235.0858 (20)	16.08	75
OD-PABA	278.2113	151.0625 (40) 166.0860 (20)	16.00	300
S86	541.2139	129.0448 (60) 335.1294 (40)	17.01	150
DBT	764.4530	273.1604 (50) 216.1137 (60)	20.30	300
DBT-d_4	768.4774	273.1603 (60) 220.1391 (60)	20.30	300
Δ Retentiontime (min): 2				
Iso. Width: ~1.3 m/z				
Z: 1				

Table S4: LOD, LOQ, and recoveries (%) for the UV filters investigated (assumed sample weight 1 g).

Analyte	Internal Standard	LOD (ng/g dw)	LOQ (ng/g dw)	Recoveries (%) (Level of spiked amounts)		Quantifiable
				20 ng/g dw (n = 8)	60 ng/g dw (n = 8)	
S87	DBT-d_4	0.02	0.05	99 ± 13	99 ± 8	yes
BP-3	BP-3-d_5	7.8	23	102 ± 25	101 ± 21	yes
UV-P	UV-P-d_3	1.6	4.6	34 ± 9	64 ± 18	semi
3-BC	4-MBC-d_4	9.6	29	36 ± 46	130 ± 48	semi
4-MBC	4-MBC-d_4	3.0	9.1	81 ± 53	107 ± 49	semi
IMPC	DBT-d_4	1.1	3.2	62 ± 6	86 ± 29	semi
DHHB	DBT-d_4	0.04	0.12	79 ± 14	87 ± 18	yes
BMDBM	BMDBM-[^{13}C]-d_3	4.4	13	73 ± 41	98 ± 41	semi
OC	OC-[^{13}C]$_3$	16	48	138 ± 61	92 ± 69	semi
OD-PABA	DBT-d_4	0.04	0.11	75 ± 11	81 ± 12	yes
S86	DBT-d_4	0.02	0.05	22 ± 6	25 ± 5	semi
DBT	DBT-d_4	1.1	3.4	93 ± 3	97 ± 5	yes

Table S5: Detections of UV filters in suspended particulate matter (SPM) and detection frequencies (%) in water at Koblenz, 2022, using targeted- and non-targeted screening.

Acronym	CAS No.	xlogP (PubChem†)	Target study: Detection, SPM	NTSPortal: Detection, SPM	NTSPortal: FoA** of Substances, Water
DPDT	180898-37-7	-2.5*	na	na	na
CBM	52793-97-2	-0.7*	na	yes	0%
PEG-25 PABA	116242-27-4	-0.5	na	na	na
PABA	150-13-0	0.8	na	yes	0%
S87	1419401-88-9	1.7	no	no	0%
PBSA	27503-81-7	2.0	na	no	78%
BP-4	4065-45-6	2.2	na	no	8%
BP-2	131-55-5	2.4	na	no	0%
THB	1143-72-2	2.8	na	no	0%
BCSA	56039-58-8	2.9	na	no	0%
TDSA	92761-26-7	3.1	na	no	0%
HAA299	919803-06-8	3.4*	na	na	na
BP-3	131-57-7	3.6	no	no	0%
BP-7	85-19-8	3.9	na	no	0%
IMPC	71617-10-2	3.9	no	no	0%
3-BC	15087-24-8	4.1	no	no	0%
UV-P	2440-22-4	4.3	yes	no	0%
4-MBC	36861-47-9	4.5	yes	yes	0%
BMDBM	70356-09-1	4.8	yes	yes	0%
HS	118-56-9	5.0	na	na	na
OD-PABA	21245-02-3	5.0	yes	no	0%
EHMC	5466-77-3	5.3	na	no	0%
EHS	118-60-5	5.7	na	na	na
S86	55514-22-2	6.4	no	no	0%
DHHB	302776-68-7	6.5	yes	yes	0%
Drometrizole trisiloxane	155633-54-8	6.9*	na	na	na
OC	6197-30-4	7.1	yes	yes	0%
TBT	31274-51-8	9.8	na	na	na
BEMT	187393-00-6	10.4	na	na	na
DBT	154702-15-5	11.8	yes	na	na
MBBT	103597-45-1	12.8	na	na	na
EHT	88122-99-0	14.5	na	na	na
†	Kim, 2016; Kim et al., 2025, 2021, 2019, 2016				
*****	log K_{OW} calculated by OPERA 2.9 (Mansouri et al., 2024, 2021, 2020, 2019, 2018, 2016a, 2016b)				
******	Frequency of Appearance (Water: Percentage of detections in all samples in 2022 at Koblenz (Rhine))				
na	not analyzed				

Table S6: SPM sampling stations with discharge information.

SPM station	River	Code	Next station with discharge information	Source of discharge information	Years with data	Mean discharge	Standard deviation of discharge	Maximum discharge
						[m³/s]		
Ulm	Danube	D1	Neu-Ulm Bad Held	Bayerisches Landesamt für Umwelt, 2026a	1953-2025	160	78	966
Jochenstein	Danube	D3	Passau-Ilzstadt	Bayerisches Landesamt für Umwelt, 2026b	1998-2025	1391	599	9300
Prossen	Elbe	E1	Schöna	Wasserstraßen- und Schifffahrtsverwaltung des Bundes, 2026a	2001-2026	280	248	8710
Zehren	Elbe	E2	Dresden	Wasserstraßen- und Schifffahrtsverwaltung des Bundes, 2026b	2000-2026	296	258	4590
Cumlosen	Elbe	E4	Wittenberge	Wasserstraßen- und Schifffahrtsverwaltung des Bundes, 2026c	2000-2026	600	425	4330
Blankenese	Elbe	E5	Neu Darchau	Bundesanstalt für Gewässerkunde (BfG), 2026	1874-2024	690	-	4070
Dessau	Mulde	Mu	Dessau-Leopoldshafen	Landesportal Sachsen-Anhalt, 2026a	2025-2026	181	77	689
Wettin	Saale	Sa	Halle-Trotha UP	Landesportal Sachsen-Anhalt, 2026b	2025-2026	50	18	155
Weil am Rhein	Rhine	R1	Basel	Bundesamt für Umwelt BAFU, 2026	2020-2026	1009	464	4620
Koblenz	Rhine	R3	Andernach	Wasserstraßen- und Schifffahrtsverwaltung des Bundes, 2026d	2000-2026	2011	1008	8710
Bimmen	Rhine	R4	Emmerich	Wasserstraßen- und Schifffahrtsverwaltung des Bundes, 2026e	2000-2026	2222	1084	9520
Güdingen	Saar	S1	Sankt Arnual	Wasserstraßen- und Schifffahrtsverwaltung des Bundes, 2026f	2000-2026	40	47	670
Rehlingen	Saar	S2	Fremersdorf	Wasserstraßen- und Schifffahrtsverwaltung des Bundes, 2026g	2000-2026	69	82	1350

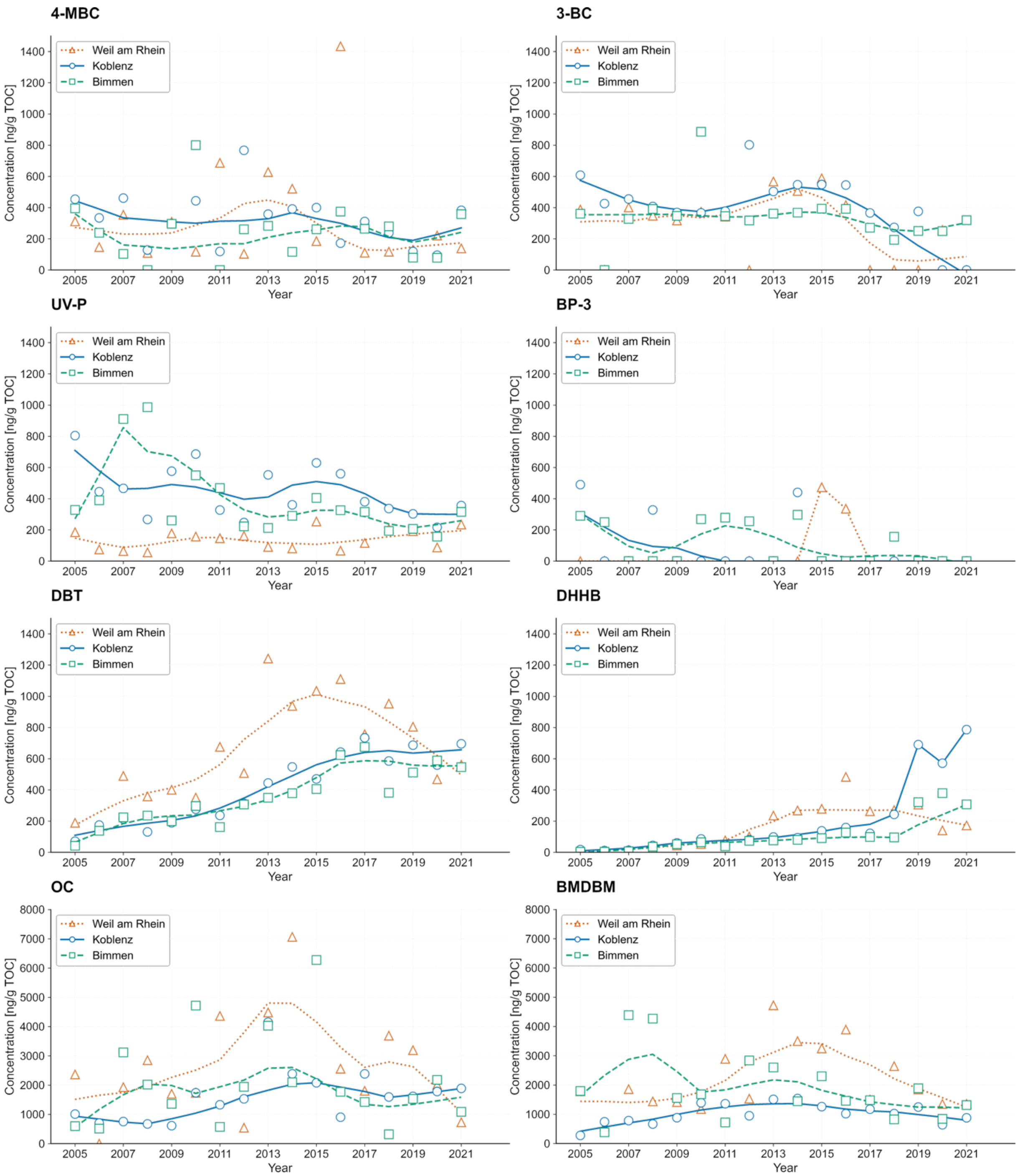

Figure S1: Time trends of the concentrations of organic UV filters in SPM (dry weight, dw) in ng/g TOC. Displayed are LOWESS-smoothed trends (smoothing span: 38.9% of data points) and individual measurements from the Koblenz, Bimmen, and Weil am Rhein stations (2005-2022). Values below the limit of detection (LOD) were set to 0 ng/g TOC, and those between the LOD and the limit of quantification (LOQ) were set to 0.5 × LOQ before normalizing to TOC content. The scale on the y-axis for OC and BMDBM is different, due to the high concentrations found.

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