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Impact of UV disinfection on potential of personal care products components on chlorination by-products formation in swimming pool water

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ABSTRACT

The article presents the results of studies on an impact of UV radiation on the potential to form several organic chlorination by-products in the model solutions, containing the ingredients of body care products (i.e., UV filters and parabens). The research was conducted on seven UV filters (ethylhexyl methoxycinnamate, butyl methoxydibenzoylmethane, 4-methylbenzylidene camphor, octocrylene, benzophenone-3, ethylhexyl salicylate, octyl dimethyl-para-amino-benzoic acid) and three parabens (methyl paraben, propyl paraben, ethyl paraben). The following by-products were studied: trihalomethanes (trichloromethane, bromodichloromethane, dibromochloromethane, tribromomethane), haloacetic acids (monochloroacetic acid, dichloroacetic acid, bromochloroacetic acid, dibromoacetic acid, trichloroacetic acid), haloacetonitriles (bromochloroacetonitrile, dibromoacetonitrile, dichloroacetonitrile, trichloroacetonitrile), haloketones (1,1-dichloro-2-propanone, 1,1,1-trichloro-2-propanone), chloropicrin, and chloral hydrate. The test of by-products formation potential was applied in the studies, with 24 h time of swimming water samples incubation. The water samples were chlorinated and irradiated with UV, generated with low-pressure UV lamp by Heraeus, Germany. Three UV doses were studied: 0 kJ/m^2 (water only chlorinated), 23.5 kJ/m^2, and 47 kJ/m^2. The research results have been used to assess how UV influences the reactivity of the studied model compounds and their potential to form halogenated organic chlorination by-products.

Keywords: Disinfection by-products; UV disinfection; Swimming pool water; UV filters; Parabens

1. Introduction

Any organic compound used in the production of lotions, creams and lipsticks can flow into swimming pool water along with the swimmers and reacting with chlorine – through a series of transformations of these compounds – can lead to the formation of disinfection by-products (DBPs), such as trihalomethanes (THM), haloacetic acids (HAA), haloacetonitriles (HAN), haloketones (HK), chloropicrin (CP) and chloral hydrate (CH). Because the cosmetic ingredients are the original compounds (the substances before these chemical reactions), from which DBPs are produced of, they can be treated as the precursors of DBPs. In a case of swimming pool water the personal care products (PCPs) are mainly UV filters and parabens [1]. Most organic UV filters are lipophilic (dissolved in fats, oils and nonpolar solvents), they have in their structure the aromatic rings coupled with carbon–carbon double bonds [2,3]. Although UV filters are hydrophobic in their nature, they are washed away from the skin surface and thus flow into water and environment [4–6]. There are only two known inorganic UV filters (titanium dioxide and zinc oxide), while the majority of compounds used in PCPs for sun protection are organic compounds from the groups of

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benzophenones, cinnamates, dibenzylmethanes, derivatives of camphor, para-aminobenzoates and benzimidazoles [7]. The interest in the contamination of the aquatic environment with UV filters has also increased in recent years due to their potential risk of endocrine disruption [3,8]. There are two dominant ways of entering UV filters into swimming water: (1) directly as a result of washing from a swimmer's body and (2) indirectly from a wastewater treatment plant discharging sewage to water (an efficiency of UV filters removal by a wastewater treatment plant is 19% > 99% [3,4].

Parabens are a group of alkyl esters of the *p*-hydroxybenzoic acid with alkyl substituents of methyl, butyl or benzyl groups [9], they are widely used as preservatives in pharmaceutical and PCPs. The scientific interest in these compounds is mainly caused by their possible effect on the endocrine disorders and breast cancer [9]. A few-minute contact between the parabens and chlorine, presented in swimming water, results in the formation of bromo- and chloro-organic compounds, which are halogen derivatives of parabens [10,11].

Although PCPs do not directly affect the health of swimming users [12], they may be the precursors of the formation of DBPs. However our knowledge on the dynamics of these reactions in swimming pool water is very limited. Kim et al. [13] observed – in the DBPs formation test – the increase of concentration of CH, dichloroacetonitrile and 1,1,1-trichloro-2-propanone, as a result of an addition of cosmetic lotion (1 mg) to surface water (300 cm^3). While the concentration of trichloroacetonitrile did not change and the concentration of bromodichloromethane (BDCM) even decreased. In other studies [14], it has been shown that lotions containing UV filters can be the precursors of the halobenzoquinones formation in swimming pool water.

The application of UV radiation in pool water treatment technology also influences the dynamics of chlorination by-products formation. If chlorine-containing pool water is exposed to UV, the free radicals HO$^\bullet$ are formed, thus the UV-chlorination combination can be classified as the advanced oxidation process (AOP) [15,16]. In AOP organic compounds are effectively oxidised, often they are completely mineralised. The most important pathways of the organic compounds degradation and breakdown in AOP based on UV are the direct photolysis and the reactions with the radicals produced in AOP process [15,17]. The AOP of UV/Cl$_2$ is considered as more effective in the oxidation of some micropollutants than the UV/H$_2$O$_2$ [18]. For the typical UV doses, used in water disinfection (<500 J/m^2), chlorine photolysis caused by UV (both from low- and medium-pressure UV lamp) produces large amounts of free hydroxyl radicals, therefore the combination of chlorination and UV irradiation is an effective disinfection method, but also AOP [19,20]. However there are no publications on the effect of UV radiation on the DBPs' formation potential (FP) of UV filters and parabens.

2. Materials and methods

2.1. Preparation of precursors solutions

In order to examine the influence of cosmetics ingredients, introduced to water with swimmers, on the DBPs FP in chlorinated water, UV filters (ethylhexyl methoxycinnamate [EHMC], butyl methoxydibenzoylmethane [BM-DBM], 4-methylbenzylidene camphor [4-MBC], octocrylene [OC], benzophenone-3 [BP3], ethylhexyl salicylate [EHS], octyl dimethyl-para-amino-benzoic acid [OD-PABA]) and parabens (methyl paraben [MePB], propyl paraben [PrPB], ethyl paraben [EtPB]) were added to the tap water (supplied to the water circulation system of the swimming pool of AGH University of Science and Technology in Krakow). The control sample was also analysed, which was tap water without any additions. The type of matrix, which the organic compounds are dissolved in, is important for DBPs formation. Thus filling water (in this case – tap water) was used, and not ultrapure one, to model the system of a real swimming pool. The concentration of all solutions of PCP components was 1.8×10^{-5} mol/L. Table 1 presents the parameters of the studied UV filters and parabens. Table 2 presents physicochemical characteristics of water filling the system of the AGH swimming pool, in which the individual components of human body fluids were dissolved.

2.2. UV/chlorination experiment

The procedure of identifying the DBPs FPs of individual ingredients of PCPs in swimming pool water disinfected with UV-chlorine sequence was adopted from the work of Cimetiere and Laat [21]. In the method, proposed by these authors, water was irradiated with cumulative UV doses (23.5 and 47.0 kJ/m^2). Both before and after each irradiation free chlorine concentration was adjusted to 3 mg/L. In the experiment monochromatic UV lamp TNN 15/32 by Heraeus was used. The procedure has been adapted to the photoreactor, in which experiments were conducted

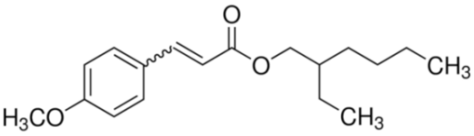
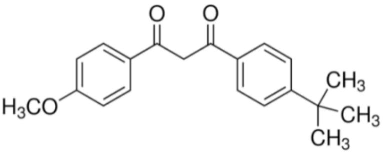
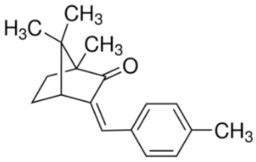
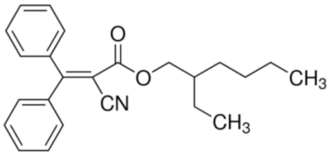
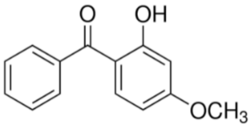
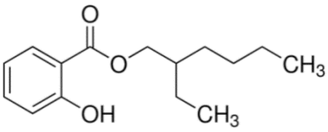
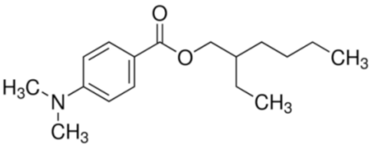
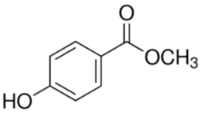
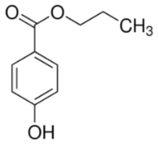
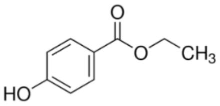
For each individual precursor three samples were taken: (1) Cl$_2$ – water only chlorinated, (2) Cl$_2$ + UV1 – water chlorinated and treated with UV dose 23.5 kJ/m^2 and chlorinated again, (3) Cl$_2$ + UV2 – water chlorinated, treated with UV dose 23.5 kJ/m^2, chlorinated, treated with UV dose 23.5 kJ/m^2 (cumulative UV dose 47.0 kJ/m^2) and chlorinated. On all stages of the experiment the free chlorine concentration was adjusted to 3.0 ± 0.2 mg/L, using sodium hypochlorite. The free chlorine concentration was determined using the N,N-diethylphenylendiamine method. The free chlorine concentration was measured using the Aurius 2021 UV-Vis spectrophotometer (Cecil Instruments, United Kingdom). The detection limit of this method was 0.03 mg/L.

After each stage of the experiment the water samples were taken into dark glass bottles of volume 250 mL, with the silicon sealing covered with polytetrafluoroethylene (PTFE). The bottles were completely filled to avoid air bubbles. The chlorinated water samples were incubated at 25 ± 2°C. After 24 h these samples were dechlorinated and DBPs were analysed. The chlorination experiments, within the studies on DBPs FP, were conducted twice for all studied precursors. The results presented in the article are an average of the results from two parallel experiments.

2.3. DBPs analysis

After 24 h incubation time the concentration of THM – trichloromethane (TCM), BDCM, dibromochloromethane (DBCM), tribromomethane (TBM); HAA – monochloroacetic

Table 1
Physicochemical properties of cosmetic ingredients (manufacturer data)

Compound	Structural formula	Form	Concentration	Manufacturer
EHMC		Light yellow liquid	>98%	Sigma-Aldrich, Fluka
BM-DBM		Powder	<100%	Sigma-Aldrich, Sigma
4-MBC		Powder	>98,5%	Sigma-Aldrich, Fluka
OC		Viscous liquid	100%	Sigma-Aldrich, Fluka
BP3		Powder	98%	ACRŌS Organics
EHS		Colourless liquid	99%	Sigma-Aldrich, Aldrich
OD-PABA		Viscous liquid	98%	Sigma-Aldrich, Fluka
MePB		Powder	99%	ACRŌS Organics
PrPB		Powder	99+%	ACRŌS Organics
EtPB		Powder	99%	ACRŌS Organics

EHMC, ethylhexyl methoxycinnamate; BM-DBM, butyl methoxydibenzoylmethane; 4-MBC, 4-methylbenzylidene camphor; OC, octocrylene; BP3, benzophenone-3; EHS, ethylhexyl salicylate; OD-PABA, octyl dimethyl-para-amino-benzoic acid; MePB, methyl paraben; PrPB, propyl paraben; EtPB, ethyl paraben.

Table 2
Physicochemical parameters of filling water

Parameter	Filling water
N_{NH4}, mg/L	0.02
N_{NO2}, mg/L	<0.002
N_{NO3}, mg/L	1.9
TN, mg/L	2.6
DOC, mg/L	1.1
SUVA, m^{-1}×L/mg	2.873
Br^-, mg/L	0.45
pH	7.8
Conductivity, mS/cm	0.438

SUVA, specific ultraviolet absorbance; DOC, dissolved organic carbon.

acid (MCAA), dichloroacetic acid (DCAA), bromochloroacetic acid (BCAA), dibromoacetic acid (DBAA), trichloroacetic acid (TCAA); HAN – bromochloroacetonitrile (BCAN), dibromoacetonitrile (DBAN), dichloroacetonitrile (DCAN), trichloroacetonitrile (TCAN); HK – 1,1-dichloro-2-propanone, 1,1,1-trichloro-2-propanone, CP and CH was analysed in all samples. Prior to the analysis of HAAs the samples were dechlorinated with sodium sulphite, while prior to the analysis of the rest of volatile DBPs the samples were dechlorinated with ascorbic acids.

The compounds from groups THM, HAN, HK, CH and CP were analysed with a gas chromatograph Trace Ultra DSQII gas chromatograph-mass spectrometer (GC-MS) by Thermo Scientific, United States. Carrier gas was helium. The compounds separation was performed on a capillary column Rxi™-5ms by Restek, United States (film thickness 0.5 µm, column length 30 m, and internal column diameter 0.25 mm). The analysed DBPs were extracted from the water samples in 40 mL dark glass bottles with a PTFE septa, with methyl tert-butyl ether (MTBE) in the liquid–liquid method, with addition of sodium sulphate (35 mL of a water sample, 6 g of Na_2SO_4, and 3 mL of MTBE) according to the methodology recommended by U.S. EPA [22]. The following temperature program was applied for volatile DBPs analysis on GC-MS – 35°C (9.5 min) to 200°C (0 min), temperature increase rate 40°C/min. The detection limit for each individual compounds was 0.01 µg/L.

The HAA concentrations were measured with the method of acid esterification and GC-MS (Trace Ultra DSQII, Thermo Scientific) [23]. Carrier gas was helium. The compounds were separated with a capillary column Rxi™-5ms by Restek (film thickness 0.5 µm, column length 30 m, and internal columns diameter 0.25 mm). HAA was extracted with acid extraction liquid–liquid with MTBE (30 mL of water samples, 12 g of Na_2SO_4, 6 g of $CuSO_4 \cdot H_2O$, 2 mL of concentrated sulphuric acid, and 3 mL of MTBE). A total of 0.9 mL of the extract was moved into 15 mL dark glass bottles, afterwards 2 mL of sulphuric acid solution in methanol (10%) was added, and the samples were placed in water bathing in the temperature of 50°C for 1 h. After this time, they were cooled down in the temperature of 4°C for 10 min, and 5 mL of copper sulphate pentahydrate and anhydrous sodium sulphate solution was added (respectively, 50 g/L and 100 g/L). The samples were shaken for 2 min and then left for 5 min. The gas chromatograph injection was carried out with an extract taken from the top layer of the solution. The chromatograph column was heated up from 40°C (0 min) to 100°C (5 min) with a temperature rate of 40°C/min, and afterwards to 200°C (0 min) with a rate of 8°C/min. The detection limit was 0.50 µg/L for MCAA and 0.01 µg/L for the remaining HAA.

2.4. Statistical analysis

To assess the UV influence on the by-products FP the statistical analyses of results were conducted. Therefore the one-way analysis of variance with post-hoc Tukey's test was used to evaluate if the concentration of each DBP was statistically significantly different (significance level < 0.05) in the analysed water samples for studied UV doses (0, 23.5 and 47 kJ/m^2). Prior the main analysis, the Lavene's test was used as the assumptions test. Those analyses were conducted for two parallel experiments (as described in Section 2.2). All statistical analyses were performed using Statistica 10.0 by StatSoft.

3. Results and discussion

Tables S1–S3 show the average concentrations of the individual DBPs analysed in model UV filters and parabens solutions in chlorinated samples (Cl_2) (Table S1); chlorinated and irradiated with UV dose 23.5 kJ/m^2 (Cl_2 + UV1) (Table S2); chlorinated and irradiated with UV dose 47 kJ/m^2 (Cl_2 + UV2) (Table S3). Figs. 1 and 2 show how UV radiation influences the FP of carbon-containing DBPs (Fig. 1) and nitrogen-containing DBPs (Fig. 2).

The statistically significant impact of UV radiation on ΣTHM FP was observed in the samples with EHMC, 4-MBC, EHS, MePB, PrPB, EtPB for both studied UV doses; and only for UV2 dose in OC, BP3 and OD-PABA. The strongest impact of UV radiation on the formation of THMs was observed in a case of the water samples with parabens. The highest ΣTHM-FP was observed in the sample with the addition of PrPB disinfected in Cl_2 + UV2 variant. The concentration of ΣTHM in this sample (121.19 µg/L) was 585% higher than in the sample only chlorinated (17.69 µg/L). In Cl_2 + UV1 variant, ΣTHM concentration was 36.85 µg/L. In the sample with an addition of EtPB, a significant increase in ΣTHM-FP was also noticed, as compared with the sample only chlorinated. In the later one ΣTHM concentration was 12.56 µg/L, while for Cl_2 + UV1 variant it was 49.21 µg/L (292% higher), and for Cl_2 + UV2 – 82.30 µg/L (555%). In the sample with an addition of MePB only chlorinated ΣTHM concentration was 11.59 µg/L, while in a case of additional UV radiation it was 38.00 µg/L (228%) and 54.74 µg/L (372%), respectively, for Cl_2 + UV1 and Cl_2 + UV2 variants.

The statistically significant impact of UV radiation on ΣHAA FP was observed for all cosmetic ingredients, however in a case of the samples with 4-MBC, OC, OD-PABA and all parabens, statistically significant differences were observed for both analysed UV doses, for the sample with EHS only for the dose UV1, and in the sample with EHMC, BM-DBM and BP3 for UV2. The strongest influence of UV radiation on the formation of HAA compounds was noticed in the sample with an addition of EtPB – its UV radiation in the cumulative dose of 47 kJ/m^2 increased ΣHAA concentration in this

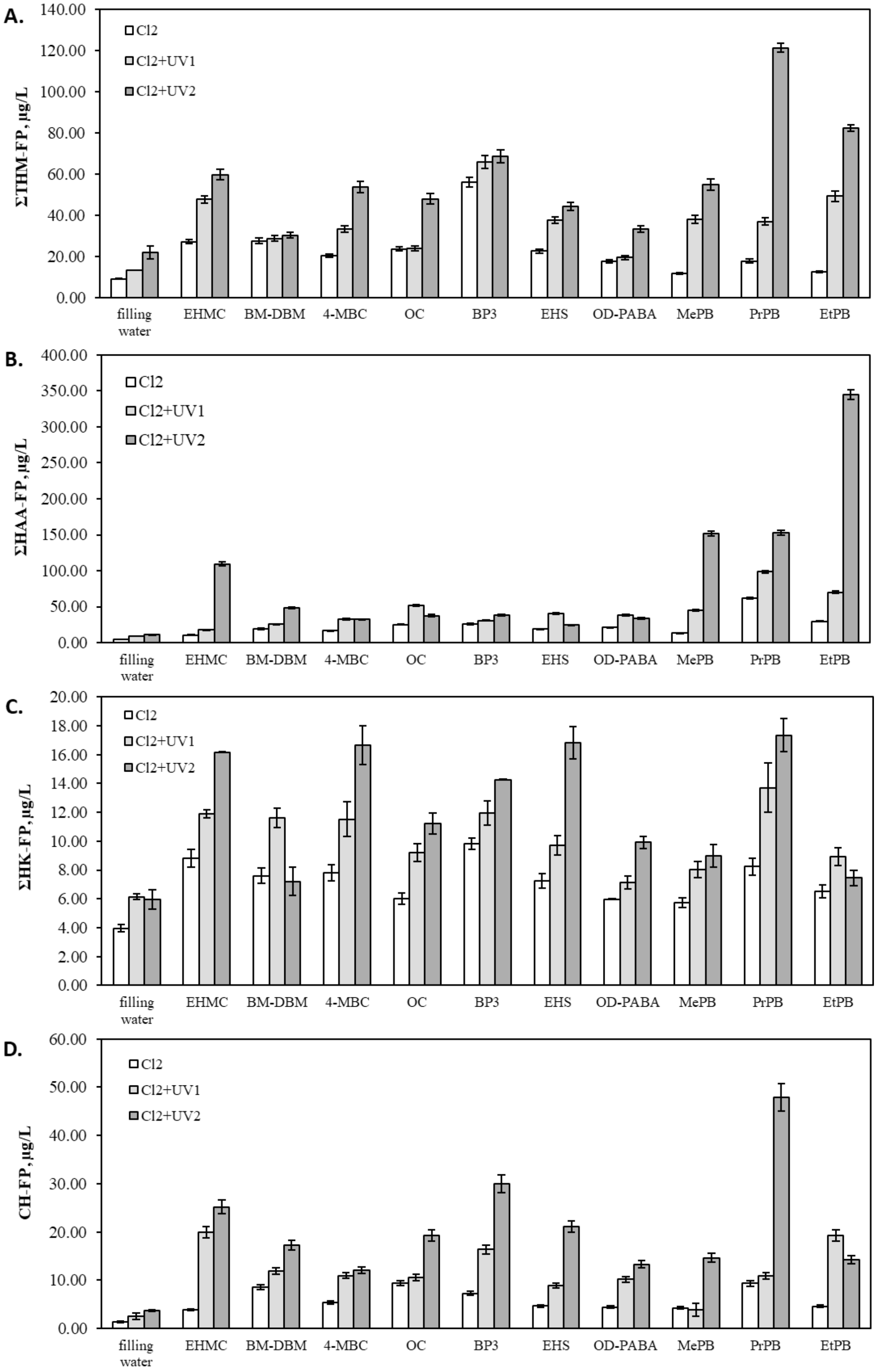


Fig. 1. UV influence on formation potential of carbon-containing DBPs: (A) trihalomethanes, (B) halogenacetic acids, (C) halo ketones, and (D) chloral hydrate.

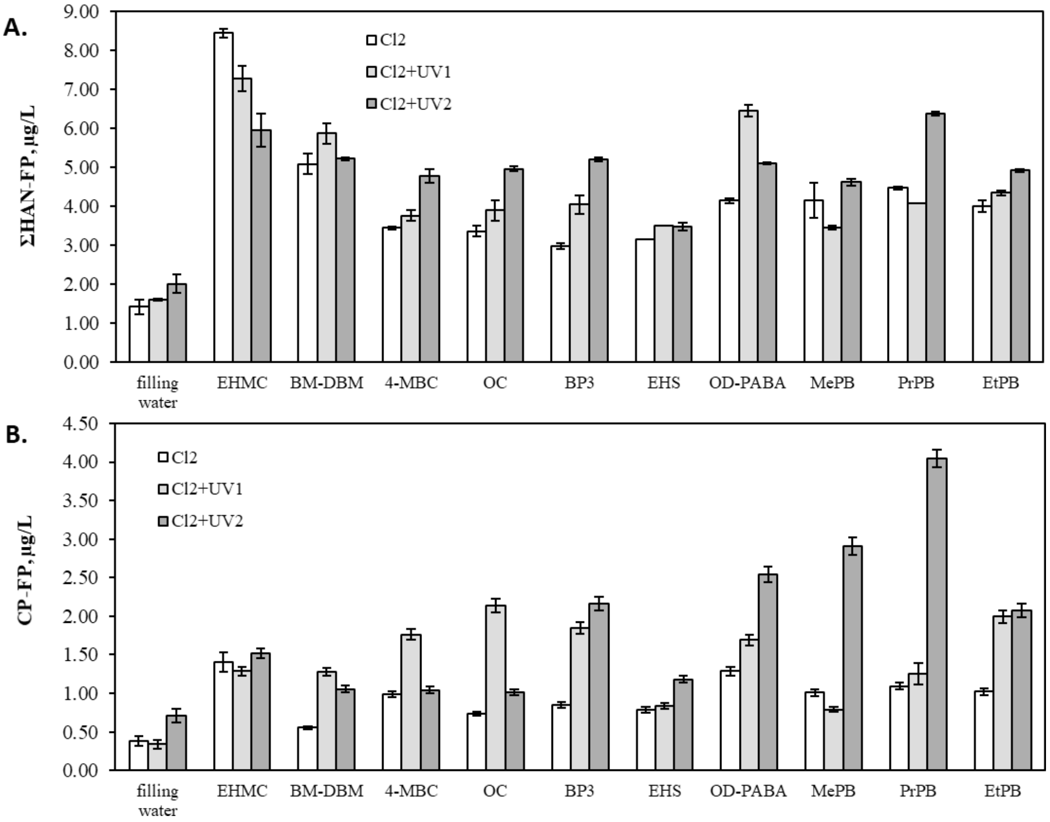


Fig. 2. UV influence on formation potential of nitrogen-containing DBPs: (A) halogenoacetonitriles, and (B) chloropicrin.

sample by 1,061% (from 29.71 µg/L in the sample only chlorinated to 344.89 µg/L).

The statistically significant influence of UV radiation on CH-FP was observed in the samples with EHMC, BP3 and EtPB for both UV doses, and in the samples with BM-DBM, 4-MBC, OC, EHS, OD-PABA, MePB and PrPB only for the dose UV2. The strongest effect of UV radiation on CH formation was noticed in the sample with an addition of PrPB for UV2 dose – CH-FP in this sample was 47.92 µg/L, which is 413% higher than in the sample only chlorinated (9.34 µg/L).

The statistically significant influence of UV radiation on ΣHK concentration was observed in the sample with an addition of 4-MBC for both UV doses, in M-DBM sample only for UV1 dose, and in the samples with EHMC, OC, BP3, EHS, OD-PABA and PrPB for UV2 dose. In the sample with MePB also for UV2 dose the statistically important effect on ΣHK-FP was observed, however ΣHK concentration did not differ statistically importantly between this sample and the control one (the sample of filling water) disinfected in the variant Cl_2 + UV2. The UV influence on HK formation in the samples with cosmetic ingredients was not as strong as in the case of THM, HAA and CH. The strongest effect was observed in the sample with EHS. The concentration of ΣHK was 7.22 µg/L in the sample only chlorinated, whereas it was 16.83 µg/L (133% higher) in the sample disinfected in Cl_2 + UV2 variant. In any of these samples the statistically significant influence of UV radiation on ΣHAN concentrations was not observed, however ΣHAN concentrations are generally higher than those observed in the control sample.

UV radiation significantly influenced the CP formation in the samples with BP3 and EtPB for both UV1 and UV2 doses; in the samples with OD-PABA, MePB and PrPB only for UV2 dose; and in the sample with OC only for UV1 dose. The strongest increase of CP-FP was noticed in the sample with PrPB disinfected in Cl_2 + UV2 variant. CP concentration was 1.09 µg/L in the sample only chlorinated, whereas it was 4.05 µg/L in the sample exposed to UV2 dose (271% higher).

As presented studies showed, UV irradiation of chlorinated water with the addition of cosmetic ingredients causes an increase in the concentration of DBPs. The FP of ΣDBPs in all samples of filling water with the addition of cosmetic ingredients differed statistically significantly from ΣDBP-FP in the sample with filling water disinfected in the same variants. The statistically significant difference between DBPs FP in chlorinated and UV-irradiated samples was also observed in all samples, for both analysed radiation doses. The influence of UV radiation on chlorination by-products formation was especially strong in the samples with EHMC, BP3 and parabens. The formation of halogenated DBPs from organic compounds in chlorinated water occurs particularly rapidly in a case of compounds with phenolic and/or amino groups [2]. Oxidation, addition and electrophilic substitution are possible chlorine reactions with organic compounds. However, taking into account the reaction kinetics, only this last reaction plays a significant role [24]. Zhang et al. [25] identified monochlorinated BP3, dichlorinated BP and 2,4,6-trichloro-3-methoxyphenol as chlorination products of BP3; while monochlorinated BP3 was formed very quickly and was very unstable. The reduction rate of dichlorinated BP3 was much slower and 2,4,6-trichloro-3-methoxyphenol was very stable and remained in chlorinated water for long time. Thus PCPs in contact with chlorine form the chlorinated derivatives, which in turn form DBPs. The identification of intermediates would allow us to understand the degradation pathways of individual PCPs, but the diagnosis of this topic would require additional detailed research, going far beyond the scope of this article.

As the studies of Negreira et al. [2] on the stability of three UV filters in the presence of chlorine (concentration up to 3 mg/L) showed – BP3 was the most stable, than OD-PABA and EHS was the least stable. It is confirmed by the results presented in this article – ΣDBPs in a sample only chlorinated was highest for BP3 solution, then OD-PABA and EHS.

While the solution with chlorine is exposed to UV radiation – AOP occurs and free radicals $HO^{\bullet}$ are formed [15], and organic compounds are effectively oxidised, often completely mineralised. The most important pathways of organic matter degradation and decay in AOP combined with UV are direct photolysis and reactions with free radicals [11,15]. During UV/Cl_2 process, the hydroxylation and bond cleavage of the compounds occur more intensively than during chlorination alone [26]. In the case of chlorophenols it can lead to the formation of resorcinol type of structures, which are the strong precursors of THM formation. The radicals $HO^{\bullet}$ can also react with organic compounds [27]. Table 3 includes the rates of second-order reaction with hydroxyl radicals for selected substances – the ingredients of analysed PCPs. Considering the high reaction rates of cinnamonian ion, benzophenone and 4-hydroxybenzoate, and the high increase of DBPs concentration resulted in irradiation of EHMC, BP3 and parabens, the assumption can be made that hydroxyl radicals may play an important role in the formation of DBPs during UV/Cl_2 disinfection of PCPs.

Outdoor swimming pools are a specific case of UV irradiation of chlorinated water containing the high amount of PCPs. As shown by studies by Simard et al. [29] carried out on 15 indoor and 39 outdoor swimming pools in Quebec, Canada, HAA concentration was significantly higher (1.5–2.5 times) in outdoor swimming pools than in indoor ones, with lower THM concentration. The authors explained this effect by the high volatility of THM and their easier transpiration to the atmosphere in outdoor facilities. However the use of outdoor swimming pools also involves the more intensive sunbathing and the higher consumption of cosmetics, containing UV filters. Anyway the presented studies evidently shows that cosmetic ingredients are primarily precursors of HAA compounds, which may be another cause of elevated HAA concentration in pool water generally, and in outdoor swimming pools particularly.

Table 3
The rates of second order reaction with hydroxyl radicals [28]

Substance	$K_{HO^{\bullet}}$, $mol^{-1}\times s^{-1}$
Cinnamonian ion	8.1×10^9
Camphor	4.1×10^9
Acrylic acid	1.5×10^9
Benzophenone	9.0×10^9
Salicylate ion	1.2×10^{10}
Benzoic acid	1.8×10^9
4-Hydroxybenzoic acid	6×10^9–9×10^9

4. Conclusions

Based on the results of experiments on influence of UV disinfection on an influence of PCPs components on the DBPs potential formation in swimming pool water, the following conclusions were made:

- UV irradiation of chlorinated water with an addition of cosmetic ingredients increases the concentration of DBPs for all analysed PCPs components;
- among analysed cosmetic components, parabens were the most sensitive to an influence of UV radiation on the DBPs formation;
- UV irradiation of parabens increased the FP of THM, HAA, CH and CP;
- among UV filters, EHMC was characterised with the highest potential to form DBPs after UV irradiation, especially HAA and CH; and
- an increase of the concentration of CH was also observed as the result of UV irradiation of OC, BP3 and EHS solutions.

The article focuses on qualitative and quantitative analyses on how the cosmetic ingredients form the analysed DBPs, and how UV radiation influences these formation processes. However the chemical reactions behind these processes are much more complex and multistage, and PCPs go through transformations, in which the variety of chlorinated subproducts is formed. The comprehensive examination of these phenomena and their kinetics is an interesting and current research challenge, undoubtedly worth the separate studies.

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Supplemental material

Table S1
Disinfection by-products formation potential in the samples of water with cosmetic ingredients disinfected only with chlorine

Compound	Disinfection by-products formation potential, µg/L										
	Filling water	EHMC	BM-DBM	4-MBC	OC	BP3	EHS	OD-PABA	MePB	PrPB	EtPB
TCM	7.50 ± 0.22	21.27 ± 1.22	24.60 ± 1.41	16.66 ± 0.96	20.67 ± 1.19	48.35 ± 2.78	19.28 ± 1.11	14.22 ± 0.82	9.55 ± 0.55	16.05 ± 0.92	9.67 ± 0.56
BDCM	1.04 ± 0.09	3.68 ± 0.17	1.99 ± 0.09	1.90 ± 0.09	1.37 ± 0.08	5.83 ± 0.27	2.53 ± 0.12	2.07 ± 0.24	1.48 ± 0.07	1.02 ± 0.05	2.21 ± 0.10
DBCM	0.47 ± 0.14	1.92 ± 0.02	0.61 ± 0.01	1.05 ± 0.01	1.28 ± 0.02	1.69 ± 0.02	0.57 ± 0.01	1.11 ± 0.05	0.47 ± 0.01	0.53 ± 0.01	0.57 ± 0.01
TBM	0.05 ± 0.01	0.13 ± 0.01	0.26 ± 0.01	0.57 ± 0.03	0.19 ± 0.01	0.05 ± 0.01	0.14 ± 0.01	0.11 ± 0.01	0.10 ± 0.01	0.09 ± 0.01	0.10 ± 0.01
ΣTHM	9.06 ± 0.27	26.99 ± 1.02	27.45 ± 1.30	20.17 ± 0.83	23.52 ± 1.08	55.81 ± 2.49	22.51 ± 0.98	17.51 ± 0.62	11.59 ± 0.48	17.69 ± 0.87	12.56 ± 0.44
MCAA	0.91 ± 0.66	1.61 ± 0.11	2.87 ± 0.19	4.10 ± 0.27	3.19 ± 0.21	2.46 ± 0.16	3.15 ± 0.21	3.53 ± 0.23	4.70 ± 0.31	4.98 ± 0.33	3.83 ± 0.25
DCAA	2.60 ± 0.50	3.66 ± 0.06	7.69 ± 0.13	6.49 ± 0.11	11.24 ± 0.19	9.15 ± 0.16	7.39 ± 0.13	8.43 ± 0.14	5.36 ± 0.09	49.63 ± 0.85	21.46 ± 0.37
TCAA	0.68 ± 0.32	3.71 ± 0.10	6.53 ± 0.17	5.18 ± 0.14	9.81 ± 0.26	12.18 ± 0.32	6.94 ± 0.18	7.43 ± 0.20	2.59 ± 0.07	6.31 ± 0.17	3.65 ± 0.10
BCAA	0.32 ± 0.03	1.28 ± 0.09	1.83 ± 0.13	0.75 ± 0.05	0.80 ± 0.06	1.45 ± 0.10	0.91 ± 0.06	1.31 ± 0.09	0.45 ± 0.03	0.55 ± 0.04	0.43 ± 0.03
DBAA	0.06 ± 0.01	0.39 ± 0.03	0.20 ± 0.02	0.27 ± 0.02	0.18 ± 0.01	0.49 ± 0.04	0.37 ± 0.03	0.52 ± 0.04	0.29 ± 0.02	0.22 ± 0.02	0.32 ± 0.03
ΣHAA	4.57 ± 0.15	10.64 ± 0.39	19.12 ± 0.63	16.79 ± 0.59	25.22 ± 0.73	25.72 ± 0.78	18.76 ± 0.61	21.23 ± 0.70	13.39 ± 0.52	61.69 ± 1.40	29.71 ± 0.77
TCAN	0.03 ± 0.01	0.09 ± 0.01	0.09 ± 0.01	0.12 ± 0.01	0.09 ± 0.01	0.15 ± 0.01	0.09 ± 0.01	0.04 ± 0.00	0.04 ± 0.00	0.24 ± 0.03	0.06 ± 0.01
DCAN	0.58 ± 0.04	1.72 ± 0.10	1.78 ± 0.09	1.33 ± 0.06	1.03 ± 0.08	1.24 ± 0.01	1.17 ± 0.06	1.53 ± 0.18	1.04 ± 0.05	1.99 ± 0.10	1.99 ± 0.10
BCAN	0.37 ± 0.19	1.13 ± 0.04	0.95 ± 0.04	0.52 ± 0.02	0.75 ± 0.03	0.57 ± 0.02	0.83 ± 0.03	1.14 ± 0.04	1.07 ± 0.32	0.95 ± 0.04	1.01 ± 0.16
DBAN	0.44 ± 0.04	5.50 ± 0.06	2.26 ± 0.21	1.48 ± 0.14	1.49 ± 0.09	1.01 ± 0.09	1.06 ± 0.10	1.43 ± 0.07	2.02 ± 0.19	1.28 ± 0.12	0.94 ± 0.09
ΣHAN	1.42 ± 0.18	8.44 ± 0.10	5.09 ± 0.25	3.45 ± 0.04	3.36 ± 0.13	2.97 ± 0.08	3.16 ± 0.00	4.14 ± 0.07	4.16 ± 0.45	4.47 ± 0.04	4.00 ± 0.14
DCP	1.79 ± 0.16	4.60 ± 0.33	3.71 ± 0.27	4.54 ± 0.33	2.51 ± 0.18	2.76 ± 0.80	3.20 ± 0.23	2.84 ± 0.20	2.30 ± 0.17	4.76 ± 0.34	3.47 ± 0.25
TCP	2.43 ± 0.09	4.82 ± 0.29	4.43 ± 0.26	3.81 ± 0.23	3.92 ± 0.23	6.65 ± 0.40	4.52 ± 0.27	3.12 ± 0.19	3.75 ± 0.17	4.08 ± 0.24	3.50 ± 0.21
ΣHK	4.22 ± 0.25	9.42 ± 0.62	8.14 ± 0.53	8.35 ± 0.56	6.43 ± 0.42	9.41 ± 0.40	7.72 ± 0.50	5.96 ± 0.01	6.05 ± 0.34	8.84 ± 0.59	6.97 ± 0.46
CH	1.53 ± 0.21	4.08 ± 0.23	9.05 ± 0.50	5.76 ± 0.32	9.91 ± 0.55	7.72 ± 0.43	4.94 ± 0.27	4.65 ± 0.26	4.52 ± 0.25	9.89 ± 0.55	4.83 ± 0.27
CP	0.31 ± 0.07	1.27 ± 0.13	0.58 ± 0.02	1.03 ± 0.04	0.77 ± 0.03	0.88 ± 0.03	0.82 ± 0.03	1.34 ± 0.05	1.05 ± 0.04	1.14 ± 0.04	1.06 ± 0.04
ΣDBPs	21.11 ± 0.09	60.85 ± 0.02	69.42 ± 0.13	55.55 ± 0.64	69.21 ± 0.51	102.62 ± 1.73	57.90 ± 0.44	54.84 ± 0.31	40.75 ± 0.23	103.72 ± 1.76	59.12 ± 0.95

EHMC, ethylhexyl methoxycinnamate; BM-DBM, butyl methoxydibenzoylmethane; 4-MBC, 4-methylbenzylidene camphor; OC, octocrylene; BP3, benzophenone-3; EHS, ethylhexyl salicylate; OD-PABA, octyl dimethyl-para-amino-benzoic acid; MePB, methyl paraben; PrPB, propyl paraben; EtPB, ethyl paraben.

Table S2
Disinfection by-products formation potential in the samples of water with cosmetic ingredients disinfected in Cl_2+UV1 variant

Compound	Disinfection by-products formation potential, μg/L										
	Filling water	EHMC	BM-DBM	4-MBC	OC	BP3	EHS	OD-PABA	MePB	PrPB	EtPB
TCM	11.51 ± 0.20	39.47 ± 2.27	25.85 ± 1.49	29.59 ± 1.70	20.99 ± 1.21	58.73 ± 3.38	32.43 ± 1.87	16.80 ± 0.97	36.12 ± 2.08	32.56 ± 1.87	46.45 ± 2.67
BDCM	1.43 ± 0.31	6.55 ± 0.30	2.13 ± 0.10	2.85 ± 0.13	2.12 ± 0.01	6.03 ± 0.28	4.63 ± 0.21	2.07 ± 0.06	1.51 ± 0.13	3.85 ± 0.18	2.26 ± 0.10
DBCM	0.27 ± 0.02	1.33 ± 0.02	0.35 ± 0.01	0.42 ± 0.01	0.59 ± 0.01	0.97 ± 0.01	0.34 ± 0.01	0.39 ± 0.00	0.28 ± 0.00	0.33 ± 0.00	0.41 ± 0.01
TBM	0.04 ± 0.02	0.19 ± 0.01	0.23 ± 0.05	0.36 ± 0.02	0.10 ± 0.02	0.06 ± 0.00	0.10 ± 0.00	0.08 ± 0.00	0.09 ± 0.00	0.11 ± 0.00	0.09 ± 0.01
ΣTHM	13.25 ± 0.15	47.54 ± 1.94	28.55 ± 1.33	33.23 ± 1.55	23.80 ± 1.15	65.78 ± 3.09	37.50 ± 1.64	19.33 ± 1.02	38.00 ± 1.95	36.85 ± 1.69	49.21 ± 2.55
MCAA	2.00 ± 0.15	3.67 ± 0.24	5.18 ± 0.34	3.01 ± 0.20	5.96 ± 0.39	3.63 ± 0.24	4.10 ± 0.27	6.25 ± 0.41	2.04 ± 0.13	4.10 ± 0.27	4.49 ± 0.29
DCAA	3.94 ± 0.60	5.34 ± 0.09	9.65 ± 0.17	18.46 ± 0.32	22.22 ± 0.38	14.62 ± 0.25	20.19 ± 0.35	15.91 ± 0.27	31.33 ± 0.54	72.04 ± 1.24	52.22 ± 0.90
TCAA	2.48 ± 0.17	7.30 ± 0.19	9.52 ± 0.25	9.38 ± 0.25	21.27 ± 0.56	10.78 ± 0.28	14.84 ± 0.39	15.02 ± 0.39	10.70 ± 0.28	21.06 ± 0.55	12.38 ± 0.33
BCAA	0.47 ± 0.05	0.88 ± 0.06	0.86 ± 0.06	1.53 ± 0.11	1.98 ± 0.14	1.44 ± 0.10	0.82 ± 0.06	1.13 ± 0.08	0.62 ± 0.04	0.79 ± 0.06	0.67 ± 0.05
DBAA	0.13 ± 0.03	0.36 ± 0.03	0.19 ± 0.02	0.29 ± 0.02	0.35 ± 0.03	0.42 ± 0.03	0.20 ± 0.05	0.31 ± 0.02	0.47 ± 0.04	0.39 ± 0.03	0.34 ± 0.03
ΣHAA	9.02 ± 0.50	17.56 ± 0.61	25.40 ± 0.83	32.67 ± 0.89	51.79 ± 1.50	30.89 ± 0.91	40.16 ± 1.11	38.62 ± 1.18	45.16 ± 1.03	98.38 ± 2.15	70.10 ± 1.59
TCAN	0.03 ± 0.01	0.12 ± 0.02	0.11 ± 0.01	0.11 ± 0.01	0.12 ± 0.01	0.08 ± 0.01	0.03 ± 0.00	0.03 ± 0.00	0.12 ± 0.01	0.03 ± 0.00	0.13 ± 0.01
DCAN	0.73 ± 0.13	1.90 ± 0.09	2.08 ± 0.02	1.46 ± 0.03	1.61 ± 0.17	1.97 ± 0.09	1.24 ± 0.06	2.73 ± 0.03	1.24 ± 0.06	2.77 ± 0.02	2.09 ± 0.10
BCAN	0.37 ± 0.13	2.13 ± 0.08	0.65 ± 0.02	0.64 ± 0.04	0.77 ± 0.03	0.97 ± 0.04	0.64 ± 0.08	1.65 ± 0.06	0.69 ± 0.11	0.75 ± 0.03	0.85 ± 0.06
DBAN	0.48 ± 0.23	3.13 ± 0.29	3.04 ± 0.28	1.55 ± 0.14	1.38 ± 0.13	1.03 ± 0.10	1.57 ± 0.15	2.04 ± 0.19	1.39 ± 0.13	0.53 ± 0.05	1.27 ± 0.17
ΣHAN	1.60 ± 0.02	7.28 ± 0.32	5.88 ± 0.26	3.76 ± 0.14	3.89 ± 0.26	4.04 ± 0.24	3.49 ± 0.00	6.45 ± 0.15	3.44 ± 0.05	4.08 ± 0.01	4.34 ± 0.07
DCP	1.69 ± 0.63	3.25 ± 0.32	4.51 ± 0.21	4.44 ± 0.71	2.57 ± 0.19	3.62 ± 1.30	3.36 ± 0.24	2.80 ± 0.20	2.98 ± 0.22	3.38 ± 0.24	3.97 ± 0.29
TCP	4.65 ± 0.43	8.35 ± 0.03	7.79 ± 0.46	8.29 ± 0.49	7.23 ± 0.43	7.48 ± 0.45	7.03 ± 0.42	4.79 ± 0.25	5.60 ± 0.33	8.62 ± 1.96	5.56 ± 0.33
ΣHK	6.34 ± 0.20	11.59 ± 0.29	12.30 ± 0.68	12.73 ± 1.21	9.80 ± 0.62	11.10 ± 0.86	10.39 ± 0.66	7.58 ± 0.46	8.58 ± 0.55	12.00 ± 1.71	9.53 ± 0.62
CH	3.12 ± 0.67	21.14 ± 1.17	12.65 ± 0.70	11.60 ± 0.64	11.20 ± 0.62	17.33 ± 0.96	9.47 ± 0.53	10.78 ± 0.60	5.15 ± 1.28	11.50 ± 0.64	20.44 ± 1.13
CP	0.28 ± 0.06	1.34 ± 0.05	1.33 ± 0.05	1.83 ± 0.07	2.23 ± 0.09	1.92 ± 0.08	0.87 ± 0.03	1.76 ± 0.07	0.82 ± 0.03	1.12 ± 0.13	2.07 ± 0.08
ΣDBPs	33.61 ± 1.44	106.56 ± 0.72	86.12 ± 0.66	95.82 ± 1.12	102.71 ± 1.41	131.07 ± 1.76	101.87 ± 0.69	84.53 ± 1.13	101.16 ± 1.00	163.93 ± 0.75	155.69 ± 0.80

EHMC, ethylhexyl methoxycinnamate; BM-DBM, butyl methoxydibenzoylmethane; 4-MBC, 4-methylbenzylidene camphor; OC, octocrylene; BP3, benzophenone-3; EHS, ethylhexyl salicylate; OD-PABA, octyl dimethyl-para-amino-benzoic acid; MePB, methyl paraben; PrPB, propyl paraben; EtPB, ethyl paraben.

Table S3
Disinfection by-products formation potential in the samples of water with cosmetic ingredients disinfected in Cl_2+UV2 variant

Compound	Disinfection by-products formation potential, μg/L										
	Filling water	EHMC	BM-DBM	4-MBC	OC	BP3	EHS	OD-PABA	MePB	PrPB	EtPB
TCM	20.13 ± 3.34	50.10 ± 2.88	26.71 ± 1.54	49.87 ± 2.87	44.56 ± 2.56	60.59 ± 3.48	37.17 ± 2.14	28.94 ± 1.66	49.94 ± 2.87	115.49 ± 2.54	77.66 ± 0.37
BDCM	1.69 ± 0.17	8.02 ± 0.37	2.98 ± 0.14	2.96 ± 0.14	2.57 ± 0.12	7.31 ± 0.34	6.14 ± 0.28	3.76 ± 0.17	4.48 ± 0.21	5.34 ± 0.25	4.40 ± 1.17
DBCM	0.17 ± 0.02	1.05 ± 0.01	0.29 ± 0.00	0.79 ± 0.01	0.59 ± 0.06	0.42 ± 0.01	0.80 ± 0.01	0.37 ± 0.00	0.28 ± 0.02	0.28 ± 0.00	0.16 ± 0.03
TBM	0.03 ± 0.00	0.39 ± 0.02	0.27 ± 0.01	0.13 ± 0.01	0.12 ± 0.01	0.11 ± 0.01	0.07 ± 0.00	0.06 ± 0.00	0.05 ± 0.00	0.08 ± 0.03	0.09 ± 0.00
ΣTHM	22.02 ± 3.15	59.57 ± 2.48	30.25 ± 1.38	53.75 ± 2.72	47.84 ± 2.50	68.43 ± 3.14	44.19 ± 1.84	33.13 ± 1.48	54.74 ± 2.69	121.19 ± 2.26	82.30 ± 1.50
MCAA	1.15 ± 0.38	8.16 ± 0.53	4.20 ± 0.28	2.18 ± 0.13	8.01 ± 0.52	6.04 ± 0.39	4.69 ± 0.31	2.78 ± 0.18	1.52 ± 0.10	2.53 ± 0.17	4.94 ± 0.32
DCAA	5.36 ± 0.04	63.38 ± 1.09	26.00 ± 0.45	17.50 ± 0.30	16.17 ± 0.28	17.05 ± 0.29	11.16 ± 0.19	17.38 ± 0.30	111.38 ± 1.91	114.49 ± 1.97	272.54 ± 4.68
TCAA	3.42 ± 0.15	32.42 ± 0.85	15.95 ± 0.42	11.31 ± 0.30	10.79 ± 0.28	13.42 ± 0.35	7.42 ± 0.20	10.83 ± 0.28	35.63 ± 0.94	33.75 ± 0.89	63.35 ± 1.67
BCAA	0.77 ± 0.23	5.15 ± 0.36	1.83 ± 0.13	1.30 ± 0.09	2.42 ± 0.17	0.79 ± 0.05	0.84 ± 0.06	2.07 ± 0.14	2.43 ± 0.17	1.68 ± 0.12	3.67 ± 0.26
DBAA	0.16 ± 0.01	0.19 ± 0.02	0.19 ± 0.02	0.15 ± 0.01	0.18 ± 0.01	0.55 ± 0.04	0.20 ± 0.02	0.34 ± 0.03	0.45 ± 0.04	0.25 ± 0.02	0.39 ± 0.03
ΣHAA	10.86 ± 0.80	109.31 ± 2.85	48.18 ± 1.28	32.43 ± 0.57	37.56 ± 1.27	37.84 ± 1.14	24.32 ± 0.74	33.40 ± 0.94	151.42 ± 3.15	152.69 ± 3.16	344.89 ± 6.96
TCAN	0.02 ± 0.01	0.11 ± 0.01	0.08 ± 0.01	0.09 ± 0.01	0.04 ± 0.01	0.16 ± 0.02	0.06 ± 0.01	0.03 ± 0.00	0.19 ± 0.02	0.11 ± 0.01	0.07 ± 0.01
DCAN	1.05 ± 0.45	2.02 ± 0.01	2.81 ± 0.13	1.98 ± 0.10	2.11 ± 0.10	3.16 ± 0.30	1.59 ± 0.08	2.64 ± 0.30	1.96 ± 0.09	3.57 ± 0.17	2.32 ± 0.11
BCAN	0.37 ± 0.09	0.84 ± 0.03	0.63 ± 0.01	0.64 ± 0.02	0.84 ± 0.03	0.51 ± 0.02	0.78 ± 0.08	1.41 ± 0.42	1.50 ± 0.06	0.93 ± 0.03	0.64 ± 0.02
DBAN	0.57 ± 0.30	2.98 ± 0.46	1.70 ± 0.16	2.07 ± 0.09	1.97 ± 0.08	1.37 ± 0.21	1.06 ± 0.10	1.02 ± 0.09	0.97 ± 0.09	1.76 ± 0.16	1.88 ± 0.17
ΣHAN	2.01 ± 0.23	5.95 ± 0.42	5.22 ± 0.03	4.78 ± 0.18	4.96 ± 0.06	5.20 ± 0.05	3.48 ± 0.09	5.09 ± 0.02	4.62 ± 0.08	6.37 ± 0.05	4.91 ± 0.03
DCP	1.87 ± 0.51	2.89 ± 0.35	4.20 ± 0.76	4.80 ± 0.56	3.00 ± 0.22	4.70 ± 0.59	4.96 ± 0.36	3.96 ± 0.29	2.98 ± 0.39	2.90 ± 0.21	4.18 ± 0.30
TCP	4.75 ± 0.17	13.25 ± 0.32	4.02 ± 0.24	13.18 ± 0.78	8.97 ± 0.53	9.54 ± 0.57	12.99 ± 0.77	6.38 ± 0.13	6.80 ± 0.41	13.28 ± 1.37	3.81 ± 0.23
ΣHK	6.62 ± 0.67	16.14 ± 0.03	8.22 ± 1.00	17.98 ± 1.35	11.97 ± 0.75	14.24 ± 0.02	17.98 ± 1.13	10.34 ± 0.42	9.79 ± 0.80	16.19 ± 1.16	7.98 ± 0.53
CH	3.89 ± 0.17	26.67 ± 1.48	18.22 ± 1.01	12.79 ± 0.71	20.43 ± 1.13	31.74 ± 1.76	22.35 ± 1.24	14.10 ± 0.78	15.52 ± 0.86	50.73 ± 2.81	15.09 ± 0.84
CP	0.63 ± 0.08	1.58 ± 0.06	1.10 ± 0.04	1.08 ± 0.04	1.06 ± 0.04	2.25 ± 0.09	1.23 ± 0.05	2.65 ± 0.10	3.03 ± 0.12	3.93 ± 0.11	2.16 ± 0.08
ΣDBPs	46.03 ± 3.42	219.22 ± 1.46	111.18 ± 1.93	122.82 ± 0.23	123.82 ± 0.75	159.72 ± 0.22	113.52 ± 1.22	98.71 ± 0.78	239.10 ± 2.33	351.11 ± 2.49	457.34 ± 6.87

EHMC, ethylhexyl methoxycinnamate; BM-DBM, butyl methoxydibenzoylmethane; 4-MBC, 4-methylbenzylidene camphor; OC, octocrylene; BP3, benzophenone-3; EHS, ethylhexyl salicylate; OD-PABA, octyl dimethyl-para-amino-benzoic acid; MePB, methyl paraben; PrPB, propyl paraben; EtPB, ethyl paraben.