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UV FILTERS AS DISINFECTION BY-PRODUCTS PRECURSORS IN SWIMMING POOL WATER

FILTRY UV JAKO PREKURSORY TWORZENIA UBOCZNYCH PRODUKTÓW CHLOROWANIA WODY BASENOWEJ

Abstract: The disinfection by-products formation precursors, which are brought into swimming water with swimmers, are not only the human body fluids released during the workout, but also hair, epidermis and residues of personal care products. The cosmetics contain the whole range of substances, however UV filters have the special importance in a case of swimming water. The article presents the results of experiments on the disinfection by-products formation potential of model solutions of 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). These substances belong to different groups of cosmetics commonly used to protect skin against solar radiation. In the presented research the following by-products were analyzed: 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. For the experiments, the test of by-products formation potential in swimming water was applied, with 24 h time of swimming water samples incubation. The results were used to define which UV filters have the highest potential to form halogenated organic by-products of water chlorination.

Keywords: disinfection by-products, personal care products, UV filters, swimming pool water

Introduction

Swimming pool water is a specific environment, which new microbiological and chemical contaminants are still being introduced into by swimmers. The pool water disinfection causes DBPs formation, as a results of reaction between a chemical disinfectant and contaminants introduced by swimmers and with the filling water (tap water) [1-4]. Increased (in relation to the tap water) concentration of chemical disinfectants protect the swimmers against microbiological contaminants on one hand, and on the other they react with organic compounds introduced by them, such as the personal care products (PCPs), body fluids, introduced with urine, sweat or saliva, as well as solid contaminants, such as skin, hair of fragments of fabrics [5-9]. The filling water also contains natural organic matter, present in tap water, as well as bromides [10, 11], additional organic and non-organic precursors. Trace amounts of pharmaceuticals may be introduced with human body fluids of swimmers, as well as with filling water [12].

UV filters are ingredients of numerous PCPs, such as lipsticks, face creams, body lotions, shampoos [13, 14]. Their task is to protect against a broad range of UV radiation emitted with sun beams (280-315 nm UVB and/or 315-400 nm UVC) [13, 15, 16]. Majority

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of the organic UV filters is lipophilic (they dissolve in fats, oils and non-polar solvents); they have aromatic rings in their structure, associated with double bonds carbon-carbon [13, 17]. Although the UV filters are hydrophobic in their nature, they are washed away from the body surface, and therefore they reach water and the environment [15, 18, 19]. Only two non-organic UV filters are known (titanium dioxide and zinc oxide), while the great majority of compounds used in the PCPs for protection against sun beams are organic, from the groups of benzophenones, cinnamates, dibenzylmethanes, derivatives of camphor, para-aminobenzoates or benzimidazoles [16]. Interest in the UV filters within the water environment has recently increased also because of their potential danger related to endocrinological disruptions. Among the UV filters, potential endocrine disrupting chemicals are benzphenone-3 (BP3), 4-methylbeznylidene camphor (4-MBC), octyl-metoksycinamate, ethylhexyl methoxycinnamate (EHMC), octyl dimethyl-para-amino-benzoic acid (OD-PABA) [17, 20]. Amount of an individual UV filter, applied once on a whole body is 0.5-2.0 (if we assume that 10 g of sun cream is used, and the UV content remains on the level of 5-10%) [17]. The pool water contamination with the UV filters may be caused by the direct introduction while swimming, and rinsing off the body of a swimmer [15, 17].

The purpose of the studies presented in the article was to examine whether the UV filters are precursors of DPBs formation in the pool water. The experiments were conducted for following UV-filters: EHMC; butyl methoxydibenzoylmethane (BM-DBM); 4-MBC; octocrylene (OC); BP3; ethylhexyl salicylate (EHS), OD-PABA. Their potential to form disinfection by-products was analyzed. In the research the following compounds were analyzed: trihalomethanes (THM) - trichloromethane (TCM), bromodichloromethane (BDCM), dibromochloromethane (DBCM), tribromomethane (TBM); haloacetic acids (HAA) - monochloroacetic acid (MCAA), dichloroacetic acid (DCAA), bromochloroacetic acid (BCAA), dibromoacetic acid (DBAA), trichloroacetic acid (TCAA); haloacetonitriles (HAN) - bromochloroacetonitrile (BCAN), dibromoacetonitrile (DBAN), dichloroacetonitrile (DCAN), trichloroacetonitrile (TCAN); haloketones (HK) - 1,1-dichloro-2-propanone (1,1-DCP), 1,1,1-trichloro-2-propanone (1,1,1-TCP), chloropicrin (CP) and chloral hydrate (CH).

Materials and methods

Preparation of UV-filters solutions

In order to examine the influence of cosmetics ingredients introduced to water with swimmers (UV filters) on the DBPs formation potential in chlorinated water, the following compounds were dissolved in tap water (supplied to the water circulation system in the AGH swimming pool): EHMC, BM-DBM, 4-MBC, OC, BP3, EHS, OD-PABA. The control sample was also used, 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 ultra-pure one, to model the system of a real swimming pool. The concentration of all solutions was $1.8\cdot10^{-5}$ mol/dm^3, as proposed by Weng and Blatchley III [21]. All UV filters used in the study were manufactured by Sigma-Aldrich Company, with the exception of BP3 by ACRŌS Organics.

Chlorination experiment

The procedure of DBPs formation potential for individual samples, disinfected with chlorine was adopted after Cimetiere and De Laat [22]. Concentration of free chloride was adjusted to the level of 3.0 ±0.2 mg/dm^3, with sodium hypochlorite. After each research stage, water samples were placed in dark glass bottles (250 cm^3) with a PTFE septa. The bottles were incubated in the temperature of 25 ±2ºC. Prior the chlorination procedure, pH level in all analyzed solutions was adjusted to 7.00 ±0.02, and chlorination took place in a presence of phosphate buffer (pH = 7). After 24 h, the samples were dechlorinated, and the concentrations of DBPs were measured. The whole procedure of the DBPs formation potential test was carried out twice (the presented results are an average from two experiments). Concentration of free and total chlorine was measured with the colorimetric method with DPD (N, N-diethylphenylenediamine), according to PN-ISO 7393-2. Concentration of chlorine was measured with the spectrophotometer Aurius 2021 UV-VIS by Cecil Instruments. The detection limit for this method was 0.03 mg/dm^3.

Disinfection by-products analysis

The compounds from groups: THM, HAN, HK, CH and CP were analyzed with a gas chromatograph Trace Ultra DSQII GC-MS by Thermo Scientific. Carrier gas was helium. The compounds separation was performed on a capillary column Rxi^{TM}-5ms by Restek (film thickness 0.5 µm; column length 30 m; internal column diameter 0.25 mm). The analyzed DBPs were extracted from the water samples with MTBE (methyl tert-butyl ether) in the liquid-liquid method according to methodology recommended by US EPA [23].The detection limit for each individual compounds was 0.01 µg/dm^3.

Five compounds from HAA group were analyzed: MCAA, DCAA, TCAA, BCAA and DBAA. The HAA concentrations were measured with the method of acid esterification and GC-MS (Trace Ultra DSQII, Thermo Scientific) [24]. The detection limit was 0.50 µg/dm^3 for MCAA and 0.01 µg/dm^3 for the remaining HAA.

Statistical analysis

The goal of statistical analysis was to evaluate whether the UV filters additions influence the by-products formation potential. To realize it the concentration of individual DBPs in case of filling water chlorination with no additives was compared with the case with an addition of cosmetics components. The statistical significance of these differences was evaluated with ANOVA variation analysis with the Tukey test ($p < 0.05$). This analysis was performed with Statistica software (ver. 10.0) by StatSoft.

Results and discussion

Table 1 presents the average concentrations for individual analyzed disinfection by-products in filling water with addition of cosmetics components.

Table 1

The formation potential of disinfection by-products in filling water samples with addition of cosmetics components (UV filters)

Compounds	Disinfection by-products formation potential [μg/ dm^3]							
	Filling water	**EHMC**	**BM-DBM**	**4-MBC**	**OC**	**BP3**	**EHS**	**OD-PABA**
TCM	7.50	21.27	24.60	16.66	20.67	48.35	19.28	14.22
BDCM	1.04	3.68	1.99	1.90	1.37	5.83	2.53	2.07
DBCM	0.47	1.92	0.61	1.05	1.28	1.69	0.57	1.11
TBM	0.05	0.13	0.26	0.57	0.19	0.05	0.14	0.11
MCAA	0.91	1.61	2.87	4.10	3.19	2.46	3.15	3.53
DCAA	2.60	3.66	7.69	6.49	11.24	9.15	7.39	8.43
TCAA	0.68	3.71	6.53	5.18	9.81	12.18	6.94	7.43
BCAA	0.32	1.28	1.83	0.75	0.80	1.45	0.91	1.31
DBAA	0.06	0.39	0.20	0.27	0.18	0.49	0.37	0.52
TCAN	0.03	0.09	0.09	0.12	0.09	0.15	0.09	0.04
DCAN	0.58	1.72	1.78	1.33	1.03	1.24	1.17	1.53
BCAN	0.37	1.13	0.95	0.52	0.75	0.57	0.83	1.14
DBAN	0.44	5.50	2.26	1.48	1.49	1.01	1.06	1.43
DCP	1.79	4.60	3.71	4.54	2.51	2.76	3.20	2.84
TCP	2.43	4.82	4.43	3.81	3.92	6.65	4.52	3.12
CH	1.53	4.08	9.05	5.76	9.91	7.72	4.94	4.65
CP	0.31	1.27	0.58	1.03	0.77	0.88	0.82	1.34

Disinfection by-products formation potential

Figure 1 presents the formation potential for the sum of all the analyzed disinfection by-products (ΣDBPs-FP) in the samples of filling water and those with addition of the individual analyzed UV filters. The obtained formation potentials for the sum of analyzed DBPs (ΣDBPs) in all samples with the UV filters addition were statistically significantly higher than the result obtained for the control sample (filling water with no additions) for all of the analyzed UV filters. The highest DBPs formation potential was observed in the samples with the BP3 addition - ΣDBPs concentration in this sample was 76.89 μg/dm^3, which is by 365% higher than the concentration observed in the filling water sample (16.54 μg/dm^3). Relatively high DBPs formation potential was also observed in samples with addition of EHMC, BM-DBM and OC. Concentrations of ΣDBPs in those samples were 50.21; 50.31 and 43.99 μg/dm^3 for a samples with addition of EHMC, BM-DBM and OC, respectively. The obtained concentrations were higher than those observed in the control sample by 204% in a case of EHMC and BM-DBM, and by 166% for OC. In case of the samples with addition of 4-MBC and EHS, the DBPs formation potential amounted to 38.76 and 39.14 μg/dm^3, respectively. These concentrations were higher from the ΣDBPs in the control sample by 134% in case of the 4-MBC, and by 137% for EHS. The lowest formation potential was observed in case of the OD-PABA sample. ΣDBPs concentration in this sample was 33.61 μg/dm^3, which is still higher (by 103%) than in the control sample.

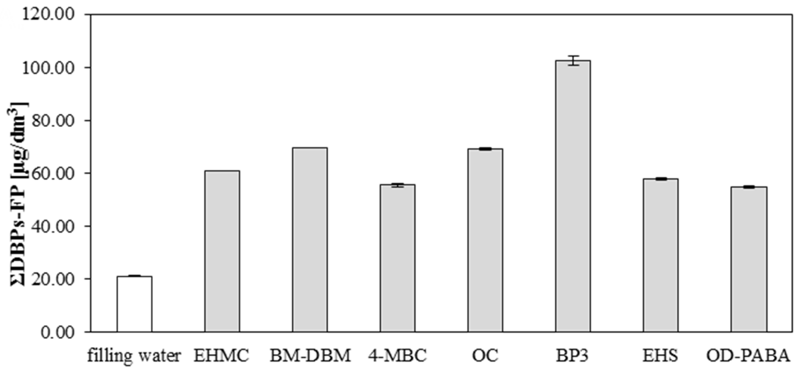

Fig. 1. The by-products formation potential (ΣDBPs-FP) in the samples of filling water with addition of cosmetics components (UV filters)

UV filters reactivity

All deliberations over an influence of the UV filters on the disinfection by-products formation potential, presented above, were related to the amount of DBPs created in a given sample (in [µg/dm^3]), without any reference to the amount of organic carbon. While, the specific DBPs formation potential represents the reactivity of 1 µg of organic carbon in relation to DBPs formation from organic matter included in a sample, thus it is possible to understand the absolute reactivity of individual precursors. Specific DBPs formation potential of the UV filters was calculated by subtracting the by-products formation potential in the control sample (filling water without additions) from the DBPs formation potential obtained in the water samples with addition of particular UV filters, and dividing the obtained result by the UV filter concentration. Figure 2 presents specific DBPs formation potential for the analyzed UV filters, calculated in such matter (shown for the carbon mass unit for particular by-products groups).

The highest specific by-products formation potential was observed for BP3 compound (19.91 ng/µg PCPs). Reactivity of that compound in relation to DBPs formation was higher by 134-197% than one of the remaining analyzed UV filters. Specific DBPs formation potential for the remaining compounds was the lowest for OD-PABA (6.78 ng/µg PCPs), and the highest for BM-DBM (8.52 ng/µg PCPs). Such a high ΣDBPs formation potential by the BP3 compound is mainly a result of a high reactiveness of this compound in relation to the THM and HAA formation. Share of THM compounds in the general amount of DBPs was as much as 57%, while in case of HAA it was 26%. A relatively high share of THM compounds (3.43 ng/µg PCPs) in the general DPBs amount (7.54 ng/µg PCPs) is also observed in case of EHMC. Percentage share of THM among all of the analyzed DBPs amounted to 46%. This compound was also characterized with a high level of reactivity in relation to HAN formation (1.34 ng/µg PCPs) and HK (0.92 ng/µg PCPs). Percentage share of those groups within the general DBPs amount was respectively 18 and 12%. All compounds, apart from the BP3, EHMC and BM-DBM, are characterized with the highest HAA share among all DBPs. In case of 4-MBC, specific ΣDBPs formation potential was 7.41 ng/µg PCPs, while the HAA share - 36%. The HAA share among all of the disinfection by-products formed by EHS was 39% and amounted to 3.15 ng/µg PCPs. In case of OC, share of HAA was 44% (3.17 ng/µg PCPs), while OD-PABA - as much as 49% (3.34 ng/µg PCPs). High percentage share of CH among all of the analyzed DBPs was observed in case of OC (17%), 4-MBC (12%) and BM-DBM (15%). 4-MBC was also characterized with a relatively high level of reactivity for HK formation (0.84 ng/µg PCPs, 11%). A characteristic quality of swimming pool water, in comparison to water distribution systems, is higher HAA concentration in relation to THM, and relatively high CH concentrations. As the presented results showed, the UV filters may influence the formation of such by-products in swimming pool water.

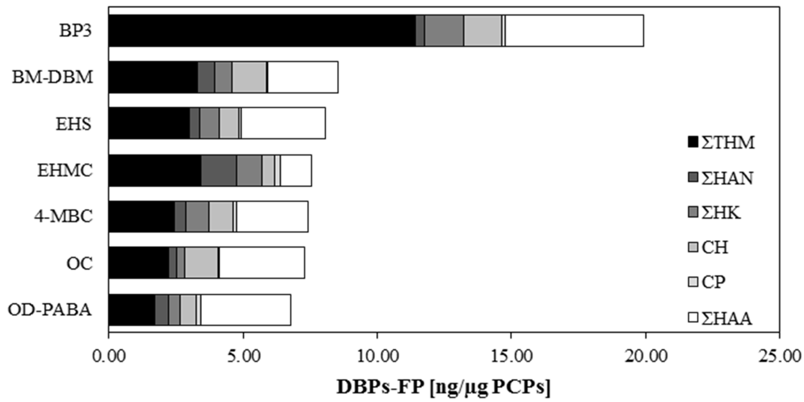

Fig. 2. The specific disinfection by-products formation potential of UV filters

Conclusions

Based on the results of experiments on influence of the UV filters on the disinfection by-products potential formation in swimming pool water, the following conclusions were made:

- All of the examined UV filters may cause an increase in formation of the disinfection by-products, but especially high influence was observed in case of haloacetic acids and chloral hydrate formation.
- BP3 was characterized with the highest potential to form trihalomethanes, haloacetic acids and haloketones, OC filter proved its significant influence on chloral hydrate and haloacetic acids formation.
- The highest formation potential of chloropicrin and haloacetonitriles was observed in case of a sample with EHMC addition. Also for this compound a high level of formation potential for the haloketones was noted.
- As suggested by the research, the highest specific disinfection by-products formation potential is showed by BP3.

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FILTRY UV JAKO PREKURSORY TWORZENIA UBOCZNYCH PRODUKTÓW CHLOROWANIA WODY BASENOWEJ

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Abstrakt: Wprowadzane z osobami kąpiącymi się do wody basenowej prekursory tworzenia produktów ubocznych dezynfekcji to nie tylko wydzieliny ciała uwalniane podczas wysiłku, ale także włosy, naskórek oraz zanieczyszczenia, znajdujące się na powierzchni skóry, pochodzące ze środków do pielęgnacji ciała. Składniki kosmetyków to cała gama związków, jednak filtry UV w przypadku wody basenowej mają szczególne znaczenie. W artykule przedstawiono wyniki badań nad potencjałem tworzenia się szeregu organicznych produktów ubocznych chlorowania modelowych roztworów zawierających filtry UV: 4-metoksycynamonian 2-etyloheksylu (EHMC); 1-(4-tert-butylofenylo)-3-(4-metoksyfenylo)propano-1,3-don (BM-DBM); 3-(4-metylobenzylideno)-d-1 kamfora (4-MBC); ester 2-etyloheksylowy kwasu 2-cyjano-3,3-difenyloakrylowego (OC); 2-hydroksy-4-metoksybenzofenon (BP3); ester 2-etyloheksylowy kwasu salicylowego (EHS), ester 2-etyloheksylowy kwasu 4-dimetyloaminobenzoesowego (OD-PABA). Związki te należą do różnych grup związków powszechnie używanych do ochrony skóry przed promieniowaniem słonecznym. W badaniach przeanalizowano potencjał tworzenia się następujących produktów ubocznych: trihalometanów (trichlorometan, bromodichlorometan, dibromochlorometan, tribromometan), kwasów halogenooctowych (kwas monochlorooctowy, kwas dichlorooctowy, kwas trichlorooctowy, kwas bromochlorooctowy i kwas dibromooctowy), haloacetonitryli (trichloroacetonitryl, dichloroacetonitryl, bromochloroacetonitryl, dibromoacetonitryl), haloketonów (1,1-dichloroproponon, 1,1,1-trichloropropanon) oraz wodzianu chloralu i chloropikryny. Badania prowadzono z wykorzystaniem testu na potencjał tworzenia się produktów ubocznych w wodzie basenowej, stosując 24 h czas inkubacji próbek wody basenowej. Analiza otrzymanych wyników pozwoliła ocenić, które związki z grupy filtrów UV mają najwyższe powinowactwo do tworzenia się halogenowych organicznych produktów chlorowania wody.

Słowa kluczowe: uboczne produkty dezynfekcji, kosmetyki, filtry UV, woda basenowa