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ORIGINAL RESEARCH ARTICLE

UV filters are an environmental threat in the Gulf of Mexico: a case study of Texas coastal zones

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Summary UV filters are the main ingredients in many cosmetics and personal care products. A significant amount of lipophilic UV filters annually enters the surface water due to large numbers of swimmers and sunbathers. The nature of these compounds cause bioaccumulation in commercial fish, particularly in estuarine areas. Consequently, biomagnification in the food chain will occur. This study estimated the amount of four common UV filters (ethylhexyl methoxycinnamate, EHMC; octocrylene, OC; butyl methoxydibenzoylmethane, BM-DBM; and benzophenone-3, BP3), which may enter surface water in the Gulf of Mexico. Our data analysis was based on the available research data and EPA standards (age classification/human body parts). The results indicated that among the 14 counties in Texas coastal zones, Nueces, with 43 beaches, has a high potential of water contamination through UV filters; EHMC: 477 kg $year^{-1}$; OC: 318 kg $year^{-1}$; BM-DBM: 258 kg $year^{-1}$; and BP by 159 kg $year^{-1}$. Refugio County, with a minimum number of beaches, indicated the lowest potential of UV filter contamination. The sensitive estuarine areas of Galveston receive a significant amount of UV filters. This article suggests action for protecting Texas estuarine areas and controlling the number of tourists and ecotourism that occurs in sensitive areas of the Gulf of Mexico.

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1. Introduction

Ultraviolet (UV) filters are common ingredients in many cosmetics and personal care products such as sunscreens, soap, shampoos, and hair sprays (Li et al., 2007; Sharifan et al., 2016; Silvia Díaz-Cruz et al., 2008). UV filters and their transformation products, which are washed off from the skin and clothes during swimming and bathing, enter the surface water (Giokas et al., 2007; Li et al., 2007; Nakajima et al., 2009; Plagellat et al., 2006; Poiger et al., 2004; Ramos et al., 2016) and are considered to be a source of surface water contamination (Ekpeghere et al., 2016; Poiger et al., 2004; Ramos et al., 2016). UV filters are added to consumer sunscreen products at different concentrations due to sunscreen formulations (Amine et al., 2012; Kupper et al., 2006; Li et al., 2007; Plagellat et al., 2006; Silvia Díaz-Cruz et al., 2008). The water contamination by UV filters is an increasing public concern due to the secondary effects (i.e. bioaccumulation) of pharmaceuticals and personal care products (PPCPs) in receiving waters, which may reach detectable and potentially toxic concentration levels (Gago-Ferrero et al., 2013; Sharifan et al., 2016).

Furthermore, due to the lipophilic characteristics of UV filters, they can bioaccumulate and biomagnify through the food chain, and their presence is associated with estrogenic effects (Broniowska et al., 2016; Mueller et al., 2003; Vila et al., 2016). Ultimately, these filters can bioaccumulate in humans (Broniowska et al., 2016; Valle-Sistac et al., 2016). Due to a high log octanol—water partition coefficient (log K_{ow}) of UV-filters (3.8—5.9), these compounds are associated with a high accumulation rate in fish (Broniowska et al., 2016; Ekpeghere et al., 2016; Kim and Choi, 2014).

Fish has a strong tendency to accumulate UV filters (Giokas et al., 2007; Liu et al., 2015). Reported concentrations of UV filters in fish ranged from 9 to 2400 ng g^{-1} lipid weight (Gago-Ferrero et al., 2015). For example, two fish species of perch and roach accumulated UV filters, respectively, by 2000 ng g^{-1} and 500 ng g^{-1} lipids (Li et al., 2007). Though the accumulation rate of UV-filters in fish has been studied both in the field and in laboratories (Blüthgen et al., 2014; Gago-Ferrero et al., 2013; Liu et al., 2015), the toxicokinetic mechanisms of these compounds in fish remain unclear.

In addition to accumulating in the food chain, UV filters have shown severe effects on coral reefs by bleaching corals at very low concentrations (Danovaro et al., 2008). Recently, the UV filters were detected at concentration levels greater than 3700 ng L^{-1} along the coastal areas of South Carolina in the USA (Bratkovics et al., 2015). This concentration may actively link to the life of U.S. endangered coral species such as *Acropora palmata* at the Flower Garden Banks National Marine Sanctuary in the northwestern Gulf of Mexico (Zimmer et al., 2006).

The long shoreline in South Texas (approximately 367 miles/590 km) is a center of recreational activities throughout the year. All 14 counties of this shoreline have 169 beaches for water activities (EPA, 2013). Every year, due to millions of beach visitors and swimmers, significant amounts of UV filters directly or indirectly (i.e. through mistreatment of wastewater, contamination of sand, etc.) enter the surface water in the Gulf of Mexico. However, UV filter concentration information is geographically restricted to some European and Asian countries, as well as Australia, whereas data from other regions, namely the Americas, is missing (Ramos et al., 2016). The potential release of these compounds has never been studied in the Gulf of Mexico. A major challenge for the potential risk effects of UV filters on aquatic life and the food chain is the availability of reliable analytical procedures that determine these substances in aquatic systems (Giokas et al., 2004, 2005; Rodil and Moeder, 2008). However, the empirical research (laboratory experiments and field surveys) is strictly limited due to financial and practical constraints (Arnot and Gobas, 2003; Korsman et al., 2015).

In order to fill the knowledge gap on the ecotoxicity of UV filters in the Gulf of Mexico region, this study aims to identify potentially hazardous substances in an effective and conservative manner. The objective of this study is to estimate the amount of UV filters: ethylhexyl methoxycinnamate (EHMC), octocrylene (OC), butyl methoxydibenzoylmethane (BM-DBM), and benzophenone-3 (BP3) entering the Gulf waters from Texas beaches.

2. Material and methods

2.1. Study area of Gulf of Mexico (Texas)

Based on an EPA report, the total number of beaches in the Texas shoreline, all 14 counties, contain 169 beaches (EPA, 2013), which are aquatic centers for swimmers and beachgoers. The counties of Nueces and Galveston have the highest number of beaches, 43 and 36, respectively. Since 1970, the population of this region increased more than 50% by 2003 based on available statistical data (Lynch et al., 2003). Texas coastal zones have the second largest number of beach visitors (3.8 million) and swimmers (3.07 million) in the entire USA (Lynch et al., 2003). Fig. 1 shows the geographical distribution of Texas counties along the Gulf of Mexico.

2.2. Chemicals

Currently, 14 organic UV filters are authorized in the USA (Ao et al., 2015; Rodil et al., 2009). Four commonly-used UV filters, which are authorized in the USA were studied herein (Santos et al., 2012; Scalia and Mezzena, 2009). The chemical structures of these four compounds, BP3, EHMC, OC and BM-DBM, are described in Table 1, which are presenting the typical structure of chemical UV filters with an aromatic moiety and a side-chain indicating different degrees of unsaturation (Silvia Díaz-Cruz et al., 2008).

2.3. Concentration of UV filters

The average content of each UV filter in cosmetic products (Table 1) was calculated as a weighted average from the composition of individual products via Eq. (1), which was developed by Poiger et al. (2004). The data of UV filter content in sunscreen products used in this study were extracted from a study by Poiger et al. (2004).

$$c_{j,\mathrm{av}} = \frac{\sum n_j c_{j,i}}{\sum n_j}. \qquad (1)$$

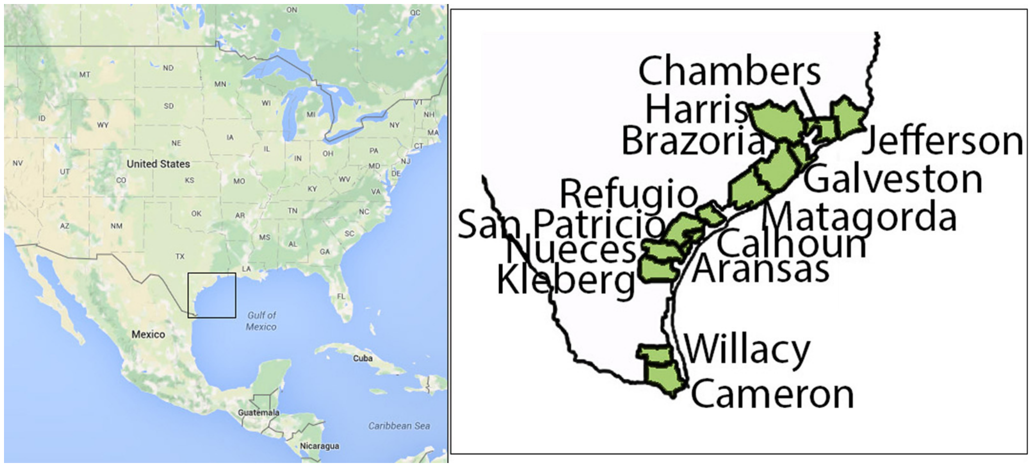


Figure 1 The map of the beaches in 14 counties along the Gulf coast of Texas.
Source: EPA (EPA, 2013), Google.

In this equation $c_{j,av}$ is the average content of UV filter j in the products used during the survey, n_i is the number of people using product i, and $c_{j,i}$ is the concentration of UV filter j in product i.

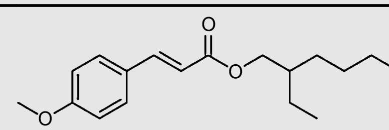
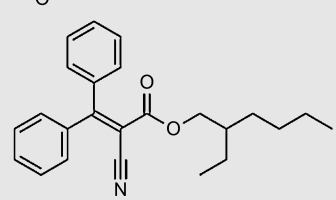
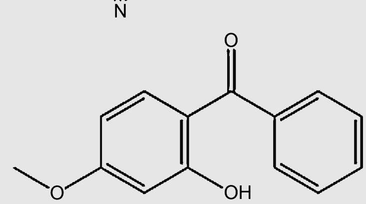
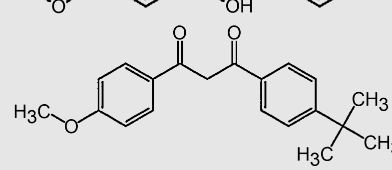
2.4. Surface area of the body

In this study, the surface area of the body parts for different standard age groupings as recommended by the EPA (Table 2) was applied to estimate how much each individual body part could contribute to the release of UV filters into the surface water. The age classes between 1 and 21 are combined by gender. Both genders of male and female are classified from age 21 to 80 (EPA, 2011).

2.5. Release estimation

The direct input of UV filters to the surface water is a function of the fraction of UV filters released from the skin (wash-off rate) during swimming and sunbathing at the beach. The

Table 1 Chemical characteristics, structures and corresponding abbreviations of the surveyed UV filters.

INCI name[a]	Abbreviation	Solubility ([mg L^{-1}] at 25°C)[b]	Average UV filter content in sunscreen products [mg g^{-1}]	Log K_{ow}	Structure
Ethylhexyl methoxycinnamate	EHMC	0.15	2.4	5.8	
Octocrylene	OC	0.02	1.6	6.88	
Benzophenone-3	BP3	68.56	0.8	3.79	
Butyl methoxydibenzoylmethane	BM-DBM	2.2	1.3	4.51	

[a] International Nomenclature of Cosmetic Ingredients.
[b] *Source*: https://pubchem.ncbi.nlm.nih.gov.

Table 2 Surface area of each individual body part [m^2] for different age classifications.

Age class	Face	Trunk	Arms	Hands	Legs	Feet
1—<2	0.029	0.188	0.069	0.03	0.122	0.033
2—<3	0.017	0.25	0.088	0.028	0.154	0.038
3—<6	0.02	0.313	0.106	0.037	0.195	0.049
6—<11	0.022	0.428	0.151	0.051	0.311	0.073
11—<16	0.024	0.63	0.227	0.072	0.483	0.105
16—<21	0.025	0.759	0.269	0.083	0.543	0.112
>21 male	0.045	0.827	0.314	0.107	0.682	0.137
>21 female	0.038	0.654	0.237	0.089	0.598	0.122

lipophilic and hydrophilic UV filters account for 50 and 100% fraction rate (wash-off rate from the skin), respectively. Such a high fraction rate was studied on swimmers on beaches in Galveston County, Texas (Wright et al., 2001). In order to estimate the amount of release in each beach — because of a lack of demographical data on age classification of visitors to beaches — the average surface body of each adult swimmer, 1.94 m^2 recommended by the EPA (>21 years), was applied in this study to account for the worst case of water contamination with all adult swimmers (EPA, 2011). The empirical formula used for this estimation is shown in Eq. (2), which was developed in the previous study by author (Sharifan et al., 2016).

$$c_{j,\mathrm{rel}} = c_{j,\mathrm{av}} \times \alpha \times \beta \times S \times A. \tag{2}$$

The index $c_{j,\mathrm{rel}}$ indicates an estimation of the UV filters released from skin surface area for an adult swimmer (average of male and female), α is the amount of sunscreen cm^{-2} of skin, β is the application rate (dimensionless), S represents the surface area of the body and A represents the percentage of body which was covered by sunscreen products. Table 3 presents the experimental values of these parameters used by researchers in recent studies. In this study, $A = 87\%$, $\alpha = 2$ and $\beta = 1.5$ have been assumed (Poiger et al., 2004; Sharifan et al., 2016; Wright et al., 2001).

The number of swimmers on each beach was estimated by evenly distributing the total number of swimmers in Texas. The potential release rate of UV filters to the surface water in the Gulf of Mexico (index $c_{j,\mathrm{rel,p}}$) was calculated based on experimental Eq. (3), which was developed in this study.

$$c_{j,\mathrm{rel,p}} = c_{j,\mathrm{rel}} \times \emptyset \times P. \tag{3}$$

In this equation, $\emptyset$ indicates the fraction of UV filter (50%) and index P shows the number of swimmers or bathers who are visiting the beach during the swimming season (Sharifan et al., 2016). The annual number of swimmers in Texas was estimated to be approximately 3.07 million (Lynch et al., 2003).

3. Results and discussion

3.1. Release of UV filters from the body parts

The body surface area data (Table 2) and Eq. (2) that was used to calculate the potential release of UV filters from the surface area of swimmers is presented in Fig. 2, highlighting the impact of the different body parts on UV filter release. Evaluation of the data achieved from Eq. (3) showed that the release of UV filters significantly increased proportionately with the age of the groups. An adult male has the greatest majority of UV filter release from their skin due to higher surface area. Adult females show a significant rate of release except for the amount of EHMC, which is greater for the age group of 16—21. This difference can be explained by a larger surface area for teenagers. EHMC has the highest potential of release among the three other UV filters, which can be explained by a higher content in sunscreen products compared to other UV filter ingredients. OC showed greater value than BM-DBM, and BP3 indicated a lower value than BM-DBM in all average surface body parts in all ages. OC has higher K_{ow} (6.8) and has the highest content amount after EHMC compound. The trunk of the body has the largest surface area and shows the maximum potential for release. The hands and feet, respectively, have the lowest potential for release in the water. Trunk surface of the body for an adult can release a considerable concentration of UV filters that for EHMC could be the maximum level of approximately 200 mg per square meter of skin. Both hands and feet for a child (between 1 and 11 years) may release the minimum amount of UV filters due to smaller surface areas of the body.

Table 3 Experimental parameters for the application of sunscreens, which may vary in different studies.

References	Experimental parameters		
	α (amount of sunscreen [mg] cm^{-2} of skin)	β (application rate)	A (percentage of body [%])
Giokas et al. (2007); Poiger et al. (2004)	2, 3, 8	—	—
Poiger et al. (2004)	1	1.5	87
Neale et al. (2002)	1.5	1.03	80
Wright et al. (2001)	0.5, 1.5, 2	—	—

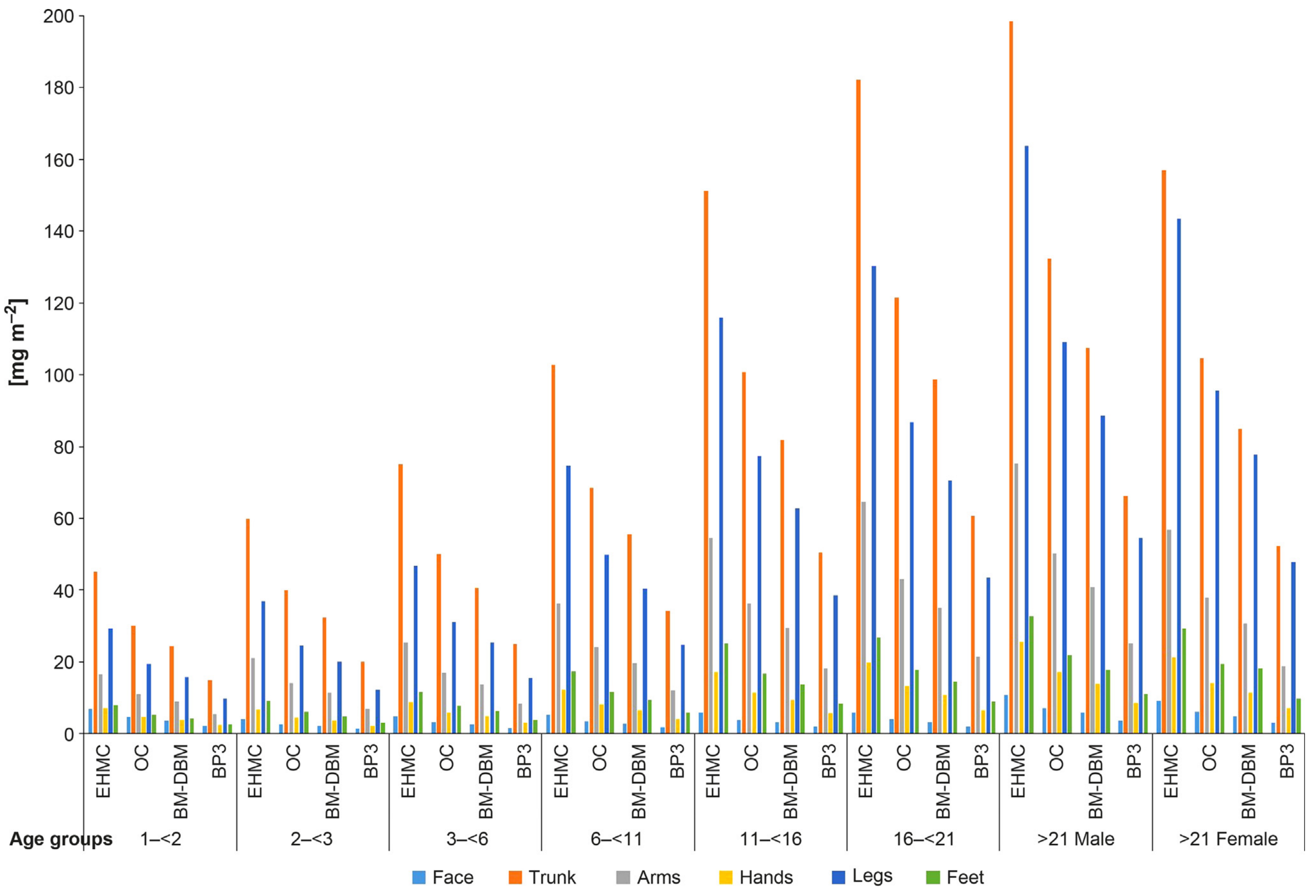


Figure 2 Results for UV filter release per body parts for different age classifications.

With respect to solubility, BP3 has the highest solubility in water (68.5 mg L^{-1}) among the other UV filters studied; therefore, it may have more potential for release in the surface water. However, BP3 may indicate a lower lipophilicity and bioaccumulation rate.

Fig. 3 shows that all UV filters have a higher potential release from the body surface of an adult male, and the age grouping of 16—21 has the second largest potential to release UV filters in surface water. This potential is due to the greater size of the body compared to other age classifications. Though there was no statistical data on the number of male and female visitors to the Gulf of Mexico in Texas, if women have a more than 50% higher tendency than men to apply sunscreens (Wright et al., 2001) on their body, the rate of release may significantly increase. However, regardless of a tendency to apply sunscreens, this analysis indicated that an average surface body of an adult female, after a male body, has a relatively high potential to release UV filters.

Due to the higher solubility of BP3 in the water, it may wash off faster than any other UV agents and may require reapplying the sunscreens. BP3 is approximately 4500 times more soluble in water than EHMC; and, at the same concentration levels, BP3 may enter the water at significantly larger amounts compared to EHMC. A study on beachgoers in Texas shows that half of beach visitors are more likely to stay in the sun longer when applying sunscreen, and approximately 70% of them believe the sunscreen will last at least 3 h without reapplying (Wright et al., 2001). Therefore, preference to stay longer periods of time on the beach may strengthen the hypothesis of higher wash-off rate from skin, as long as exposure times to the sunlight increase.

3.2. Release of UV filters to the surface water

The amount of UV filters that directly enters the Gulf further depends on the amount of UV filters released from the skin during swimming/bathing. The results of this analysis were used to estimate the average input of UV filters discharged to surface water in Texas coastal zones including 14 counties. Estuarine areas of the Texas coastline are productive aquatic systems for providing recreational and commercial marine species such as crabs, shrimp, and fish (Cai et al., 2007; Sager, 2002). For example, the Galveston Bay area has the second greatest number of beaches after Nueces, which is in the vicinity of the second largest populated region in Texas. This area has been contaminated by organic and inorganic aromatic compounds through anthropogenic sources (Glenn and James Lester, 2010; Liu et al., 2016). Therefore, an evaluation of the number of swimmers and potential release of organic UV filters from their body to the surface water is critical for the protection of the marine ecosystem of the Gulf and estuarine areas of the Texas region.

The results of this study (Table 4) estimated the Nueces with 43 beaches has the highest amount of sunscreen release to the water by 477 kg of EHMC, 318 kg of OC, 258 kg of BM-DBM and 159 kg of BP3. The county of Refugio with minimum

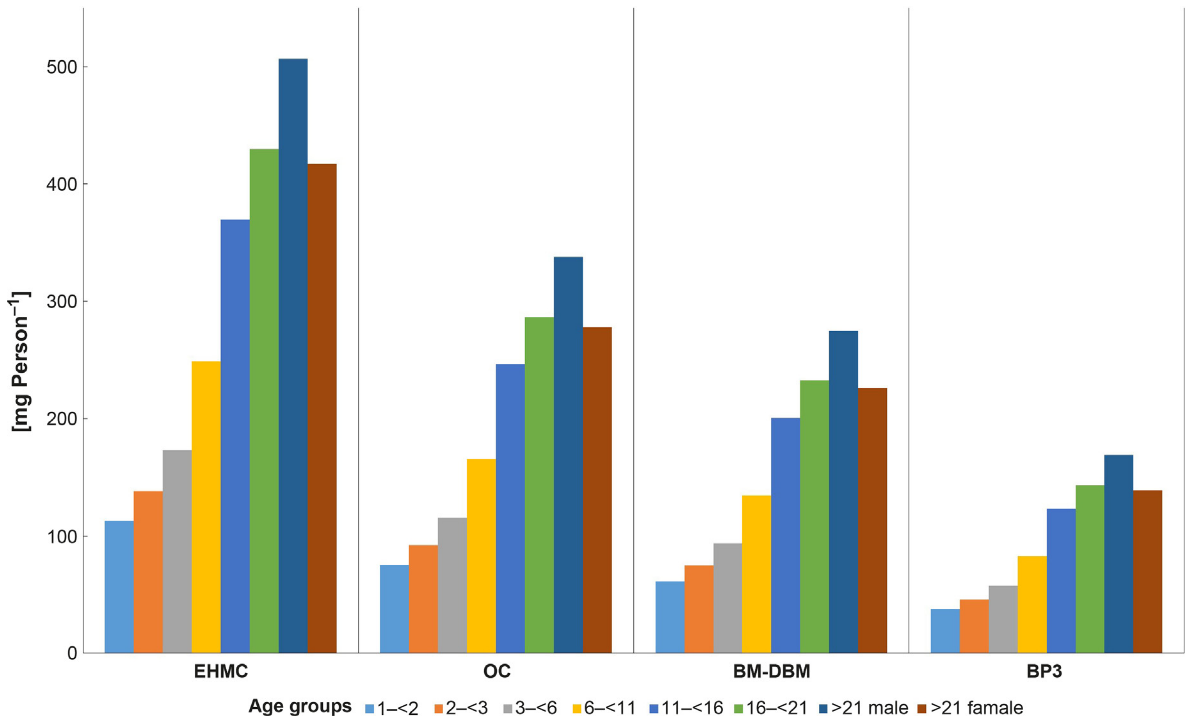


Figure 3 Results of releasing UV filters from the whole surface of a human body at different ages to the receiving water.

beach areas indicated the lowest potential of contamination with sunscreen products at the rate of approximately EHMC = 11 kg, OC = 7.5 kg, BM-DBM = 6 kg and BP3 = 4 kg. The quantity of sunscreen released during the swimming season could be far higher than what was estimated in this study because the number of beachgoers in Texas was greater than three million per year, which was not considered in this analysis. In addition, due to water activities (e.g. swimming, surfing, etc.) in the upstream of discharging rivers to the Gulf, a significant amount of UV filters may enter the water through a variety of streams, which increases the concentrations of UV filters in the Gulf.

Table 4 Estimated input of UV filters to surface water from swimmers in each county on the Texas coastline.

County	No. of beaches [a]	Beach percentage [b]	Swimmers (million) [c]	Estimated input of UV filters to surface water from swimmers [kg] [d]			
				EHMC	OC	BM-DBM	BP3
Aransas	9	0.05	0.16	99.8	66.5	54.1	33.3
Brazoria	10	0.06	0.18	110.9	73.9	60.1	37.0
Calhoun	18	0.11	0.33	199.6	133.1	108.1	66.5
Cameron	12	0.07	0.22	133.1	88.7	72.1	44.4
Chambers	2	0.01	0.04	22.2	14.8	12.0	7.4
Galveston	36	0.21	0.66	399.2	266.1	216.2	133.1
Harris	8	0.05	0.15	88.7	59.1	48.0	29.6
Jefferson	2	0.01	0.04	22.2	14.8	12.0	7.4
Kleberg	7	0.04	0.13	77.6	51.7	42.0	25.9
Matagorda	12	0.07	0.22	133.1	88.7	72.1	44.4
Nueces	43	0.25	0.78	476.8	317.8	258.3	158.9
Refugio	1	0.01	0.02	11.1	7.4	6.0	3.7
San Patricio	6	0.04	0.11	66.5	44.4	36.0	22.2
Willacy	3	0.02	0.05	33.3	22.2	18.0	11.1
Total	169	1.00	3.08	1873.8	1249.2	1015.0	624.6

[a] Data from EPA report on Texas Beaches (EPA, 2013).
[b] Percentage of the beaches based on the EPA report on Texas Beaches.
[c] Calculated based on total population of swimmers in Texas reported by Lynch et al. (2003) and assumption of even distributions between beaches.
[d] Extrapolated from calculated data of potential release of UV filters from an adult (average of male and female) from the EPA report (EPA, 2015) and estimation of swimmers in a year.

However, the concentration of a variety of pyrogenic polyaromatic compounds (PAH) in the Gulf of Mexico has been increasing during the last decades (Ruiz-Fernández et al., 2016). The high concentration of UV filters released from the human body along with significant amounts of other PAH may have accumulative effects in estuarine areas of the Gulf coast of Texas. Due to chemical/microbiological stability, low water solubility, lipophilic properties, and vapor pressure, these aromatic-based compounds may accumulate highly in both aquatic and terrestrial estuary areas (Adhikari et al., 2016; Park et al., 2001). As an interpretation, the combination of both UV filter compounds and residues of petroleum pollution to the marine ecosystem may transform the oil into compounds with less volatility and longer residence time in the sediment. The carcinogenic, toxic, mutagenic and persistent nature of PAHs (Adhikari et al., 2016; Singleton et al., 2016) and the high tendency of UV filters to bioaccumulate (Giokas et al., 2007; Liu et al., 2015) may adversly affect the large fishing industry (e.g. mutagenic and estrogenic) in the Gulf of Mexico (Klimová et al., 2015; Ozáez et al., 2016), and make them a critical group of organic pollutants that need to be monitored thoroughly.

4. Conclusion

This study was a scientific approach based on the analysis of the available research data and EPA standards (age classification/body parts of human). Through this method, the release of UV filters from the surface body was estimated and the release of UV filters in susceptible coastal areas in Texas was predicted to provide bioaccumulation data. This information can be used to determine the hazard risks to aquatic wildlife of the region, which is linked to the food web.

Texas coastal zones consist of several sensitive estuarine that may be significantly affected by cumulative effects of UV filter release and contamination by PAHs. Rather than direct release through wash off from the skin, a considerable amount of UV filters may be released through showering or rubbing off with towels or clothes. This number may increase more during laundering or showering by using other personal care products containing UV filters (i.e. shampoos, cosmetics, etc.) and indirectly be discharged to the surface bodies through wastewater. Further studies are needed to investigate the ecotoxicological effects of the UV filters in aquatic organisms, particularly the cumulative effects of PAH compounds in estuarine areas of Texas coastal zones. Therefore, several bay areas in Texas can be considered as a sensitive ecosystem which are exposed to a significant amount of UV filters. Research on this issue may affect the environmental policies for the protection of the reservoirs such as zoning of marine areas as well as make markets reconsider the sunscreen formulation for a safe combination of ingredients.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.oceano.2016.07.002.

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