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**Assessing UV filter inputs into beach waters during recreational activity: A field study of three French Mediterranean beaches from consumer survey to water analysis.**

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## Abstract

In order to assess the release of UV filters from the sunscreen used by beachgoers into seawater within the bathing zone, a field campaign was carried out during the summer of 2017 at three beaches in Marseille, along the French Mediterranean coast. A social survey analyzed beachgoer attendance, the quantities and types of suncare products used and the bathing frequencies, while the bathing water was analyzed spatially and temporally so as to quantify both mineral and organic UV filters directly released and recovered.

During the peak recreational time at the three beaches, both mineral and organic UV filters were detected in higher concentrations in the bathing area than offshore. In general, higher concentrations were recovered in the water top surface layer than in the water column, giving respectively 100-900 and 20-50  $\mu\text{g/L}$  for  $\text{TiO}_2$ , 10-15 and 1-3  $\mu\text{g/L}$  for ZnO, 40-420 and 30-150 ng/L for octocrylene, and 10-15 and 10-350 ng/L for avobenzone.

More than 75% of the 471 interviewees reported bathing every time they go to the beach, with 68% using a suncare product 2.6 times on average. From these data we estimated that an average mass of 52 kg/day or 1.4 t/month of suncare products are possibly released into bathing water for a beach attended by 3,000 people daily. The mass ratio of UV filters in such products typically ranges from 0.03 to 0.1, allowing us to propose theoretical maximum concentrations in the beach water. Our recovery of measured UV filter concentrations in seawater compared to the theoretical concentrations revealed two distinct scenarios for the mineral and organic filters. While up to 49% of the mineral filters used by beachgoers may be released into the seawater, the organic filters were minimally recovered in the environment, most likely due to internalization through the skin barrier or partial photodegradation.

## Keywords

sunscreen; marine contamination; nanomaterial fate; UV filter; titanium dioxide; environmental social issue

## 1. Introduction

Sunscreens are of emerging concern regarding both human and environmental health. Their regulation is constantly evolving, largely due to the potential risks related to the ingredients they contain. They typically consist of an oil-in-water emulsion in which the major active ingredients are UV filters, incorporated in high concentration. The UV filters can be organic or mineral in nature, depending on country specific regulations, and provide the desired sun protection factor (SPF) that is labeled on the product packaging (Steinberg 2007).

A debate has taken place as to the risks associated with UV filters regarding both consumer health and environmental impact. After application to the consumer's skin, such ingredients are potentially internalized and transported by the blood throughout the body (Schlumpf, Schmid et al. 2004, Gulson, McCall et al. 2010, Gulson, Wong et al. 2012, Matta, Zusterzeel et al. 2019). Several UV filters are blamed for having an endocrine disrupting capacity, including benzophenone-3, ethylhexyl methoxycinnamate, octocrylene, and 4-methylbenzylidene camphor (Schlumpf, Schmid et al. 2004, Kunz and Fent 2006, Calafat, Wong et al. 2008, Bluthgen, Zucchi et al. 2012). Benzophenone and its derivatives are also known to cause adverse effects on fecundity and reproduction in fish and rodents (Calafat, Wong et al. 2008, Kunz and Fent 2009). Furthermore, once leaving the skin either through bathing or cleaning, the UV filters contained in the sunscreen can be released into rivers, lakes, coastal waters, and/or sewage treatment plants (Giokas, Salvador et al. 2007, Hopkins and Blaney 2016). Their fate and impact in these different systems are largely determined by their chemical properties, persistence, and transformation (Auffan, Pedoutour et al. 2010, Labille, Feng et al. 2010, Botta, Labille et al. 2011). The scenario of direct release into the marine environment during recreational activity is of particular interest since UV filters have been repeatedly blamed for harmful effects toward coral reef areas (Danovaro, Bongiorni et al. 2008, Downs, Kramarsky-Winter et al. 2016, Fel, Lacherez et al. 2019) and other marine systems (Sanchez-Quiles and Tovar-Sanchez 2014, Sendra, Sanchez-Quiles et al. 2017, Rodriguez-Romero, Ruiz-Gutierrez et al. 2019, Tovar-Sanchez, Sanchez-Quiles et al. 2019)(Calafat, Wong et al. 2008, Kunz and Fent 2009).

Organic UV filters are synthetic molecules, generally dissolved in the sunscreen formulation. To date, only a few field-sampling campaigns have been completed in different coastal waters around the world to assess their environmental concentration and associated risk (Tashiro and Kameda 2013, Bargar, Alvarez et al. 2015, Rodriguez, Sanz et al. 2015, Downs, Kramarsky-Winter et al. 2016, Tsui, Lam et al. 2017, Kung, Lee et al. 2018, Mitchelmore, He et al. 2019). While the presence of organic UV filters is commonly observed in the ng/L range in areas with recreational activities, it is still unclear how these molecules partition and degrade within the different environmental compartments, and how this will impact their resulting lifetime. Degradation is mainly induced by photoisomerization and photodegradation processes in the presence of sunlight, processes which are known to be influenced by the presence of certain water constituents, such as natural organic matter, chlorides, nitrates, and bicarbonates (Giokas, Salvador et al. 2007, Santos, Miranda et al. 2012).

The mineral UV filters present in sunscreens consist of ultrafine titanium dioxide ( $\text{TiO}_2$ ) or zinc oxide ( $\text{ZnO}$ ) particles, often used in the nanoparticulate size range for improved UV blocking efficiency and transparency. These two types of minerals are both efficient UV blockers, favoring UV reflection and absorption over a wide range of wavelengths. While the environmental impact of nanotechnology has led to both tremendous lab and field research during the last 15 years, only a few studies have evaluated real mineral UV filters as found in sunscreens (Auffan, Pedeutour et al. 2010, Labille, Feng et al. 2010, Virkutyte, Al-Abed et al. 2012). Although mineral UV filters are generally considered as inert,  $\text{ZnO}$  and  $\text{TiO}_2$  minerals possess a photocatalytic character (Imanishi, Okamura et al. 2007) and are thus always surface functionalized in order to suppress the formation of undesired reactive oxygen species (King, Liang et al. 2008) as well as favor dispersion in the formulation (Faure, Salazar-Alvarez et al. 2013). This industrial surface coating controls the environmental fate, exposure, and hazard of these nanomaterials, making the bare  $\text{TiO}_2$  or  $\text{ZnO}$  species widely studied elsewhere an inappropriate reference (Sani-Kast, Labille et al. 2016).

Moreover, the detection of anthropogenic  $\text{TiO}_2$  and  $\text{ZnO}$  minerals (nano or non-nano) in aquatic environments where both Ti and Zn elements naturally occur in varying background concentrations remains an analytical challenge. Different proxies have been tested to distinguish the natural, terrigenous materials from those that are man-made. Al and Si are the most often used elements in mineral UV filter coatings, but are difficult

to use as a proxy for anthropogenic emission due to their abundance in natural systems (Gondikas, von der Kammer et al. 2014, Gondikas, von der Kammer et al. 2018). Elemental ratios using Ti, V or rare earth elements (Ga, Y, Nb, Eu, Ho, Er, Tm, Yb, Ta) have also been proposed as proxies for terrigenous material behavior in aquatic systems (Gondikas, von der Kammer et al. 2014, Reed, Martin et al. 2017).

An alternative to traditional proxies can be considered based on the simultaneous release of mineral and organic UV filters in bathing water. Both filter types may be found together in the environment, as a result of being associated in a common sunscreen or originating from different products. The organic UV filters, which are not present in the natural background, can be detected more easily and may be used as a proxy for the mineral filters. Indeed, the co-evolution of organic and mineral UV-filters has been measured in near shore fresh waters with time-dependent concentrations (Tovar-Sanchez, Sanchez-Quiles et al. 2013, Reed, Martin et al. 2017). However, a lack of knowledge remains regarding the respective environmental fate and persistence of these two types of UV-filters, which may be contrasting. The fate of mineral UV filters depends on both their solubility and their tendency to disperse or aggregate and sediment (Labille and Brant 2010). In addition, the hydrophilic or hydrophobic character of the particle surface will also affect its propensity to remain individually dispersed in the aqueous environment or to adsorb to the surface of natural suspended matter (Giokas, Salvador et al. 2007).

Overall, despite the rising interest in the environmental concern of UV filters, very few data are available on the quantification of the source of UV filter inputs in the field. Social surveys on consumer sunscreen use in recreational areas are needed to better understand the relation between the quantities of sunscreen used and the environmental concentrations of the UV filters actually detected in the water. Keller et al. (Keller, Vosti et al. 2014) estimated the amount of engineered nanomaterials released from personal care products based on a survey of consumer habits in the USA and China. For the USA, they calculated a total sunscreen consumption of 90,000 metric tons per year, involving an estimated potential release of 2,300-2,700 mt/yr of nanomaterials. Ficheux et al. performed a large survey of the French population's cosmetics consumption, including suncare products. They revealed that 40-46% of adults use sunscreen, with 80% of the consumption being concentrated during summer time (Ficheux, Wesolek et al. 2015). Using volunteers, they also determined that an average

range of 15-18 g of the sunscreen product is consumed during one application of sunscreen on the entire adult body (Ficheux, Chevillotte et al. 2016). Extrapolating this to the entire adult French population (49 millions adults), this gives 21 million users consuming 350 metric tons of sunscreen per application.

In this context, the aims of this work were (i) to estimate the daily flux of sunscreen and UV filters transferred from beachgoers into the bathing water on a standard summer day; (ii) to study the possible co-occurrence of organic and mineral UV filters in seawater, both in the water top surface layer and water column; and (iii) to determine, for the first time, the patterns of UV filter occurrence in the bathing water at three French Mediterranean beaches. This study is the first field campaign coupling chemical water analysis to quantify both organic and mineral UV filters in seawater, with a simultaneous social survey of consumer's habits on the beach. Thus, both the release and exposure to UV filters in such littoral systems could be evaluated.

## **2. Methodological approach**

### **2.1. Beach description**

The three urban beaches selected as study sites were chosen based on geographical and socio-demographic criteria. They are located in various stretches of Marseille's seafont (900.000 inh.) (Figure 1). To the north, the Lave Beach belongs to the seaside park of Corbière, the only recreational bathing area in the northern half of the city. Located beyond the northern tip of the commercial port, at the foot of limestone cliffs, it mainly welcomes people from working class neighborhoods. To the south, Pointe Rouge Beach is part of the recreational waterfront of the city's shoreline. It is characterized by high attendance and an urban environment. In the center, the Prophète Beach is characteristic of the rocky Provençal coast. Nestled at the foot of the coastal road and below a wealthy neighborhood, most of its users come from downtown Marseille. The present work focuses more on Prophète Beach than the two other beaches because its restricted access allowed us to count the exact daily attendance, and its relatively closed bathing zone enabled us to estimate the volume of bathing water. Despite their small size (less than one hectare), each of the three beaches is intensely frequented during the summer season, which requires specific management by local authorities from June to September (beach patrol, user services, additional cleaning and maintenance, etc.).



*Figure 1 around here*

Figure 1 Localization of the three urban beaches studied in this field campaign in the Marseille city area, along the French Mediterranean coast

## 2.2. Seawater analysis

### 2.2.1. Water Sampling

On Saturday, July 15th, 2017, water samples were collected at 4:00 pm, during peak recreational activity. The spatial distribution of any UV filters in the waters was assessed by sampling at three distances from the beach shoreline in Bathing Zone 1 (BZ1), Bathing Zone 2 (BZ2) and beyond the Bathing zone (BBZ). BZ1 and BZ2 were selected to sample the bathing water at the closest and farthest distance from the shoreline, respectively (Figure 2). The water depth at these sampling points ranged from 0.4 to 1.9 meters over the three beaches, depending on the coast slope (Table S).

*Figure 2 around here*

Figure 2 Localization of the sampling points (green) in Bathing Zone 1 (BZ1), Bathing Zone 2 (BZ2) and Beyond Bathing Zone (BBZ) on the three beaches studied. The yellow dotted line shows the limit of the bathing zone.

BBZ was selected to sample water beyond the bathing zone that gives a footprint of the local geochemical background. The seawater local background composition may result from several contributions, of natural or anthropogenic origin, that cannot be distinguished. Since the release of UV filters from sources other than beachgoer recreational activity could cause interference with this study, it should be noted that Marseille's treated urban wastewater is emitted into the sea more than 4 km away from the studied sites, in another bay (Figure 1). Here we assume that the BBZ water composition represents a local background, respective to each studied beach, and that any change in the bathing zone water composition can be related back to swimming activity on that beach.

Water samples were collected at two depths. The top surface layer (~ 1 cm), where hydrophobic compounds would likely be concentrated, was collected separately (250 mL) using a homemade sampler consisting of a plastic plate connected to a funnel. Samples were stored in high-density polyethylene bottles. To represent the average water column and obtain comparable data, a constant sampling depth was selected at 40

cm, which was always above the bottom of the water column. ~ 20 L of water was pumped directly from the 40 cm depth into pre-cleaned plastic drums carried on a kayak. During pumping, transects across the entire beach width were realized in order to pool the water along the BZ1, BZ2, or BBZ lines (Figure 2). Agitation due to recreational activity, waves, and wind may likely cause homogenization of the water composition over the entire water column height in the bathing zone (Table S1). Nevertheless, we assume that any contrasts in the composition between the two layers sampled (1 cm and 40 cm), should reflect the relative hydrophobic character of the components analyzed. In order to avoid any manipulation artifacts, the collaborators involved in water sampling and preparation did not use any suncare products during the campaign. Immediately following sampling, all collected waters were conserved at 4 °C in the dark until further treatment for chemical analysis.

To verify that recreational activity has an immediate impact on the bathing water composition in terms of UV filter concentration, the temporal variability of organic UV filter concentrations was assessed for the seawater at Prophète Beach. Additional samplings were conducted at 8:00 am on Saturday, July 15 and Sunday, July 16, 2017, i.e. just before and after the studied peak attendance on July 15<sup>th</sup> at 4:00 pm.

### **2.2.2. Seawater sample pre-treatment and analysis for UV filter quantification**

*Mineral UV filters* - To provide insight as to whether any mineral UV filters (i.e., TiO<sub>2</sub> and ZnO) present were aggregated or well-dispersed, the waters collected at a depth of 40 cm were fractionated into particulate (>0.5 μm) and colloidal (0.02 < x < 0.5 μm) fractions. The raw waters from each sampling point (~ 20 L) were first processed with a KrosFlow Research Ili Tangential Flow Filtration System (Spectrum Laboratories, Rancho Dominguez, CA, USA) utilizing a 0.5-μm membrane cut-off (mPES MiniKros Module: N04-P50U-10-N, 5-50L, Spectrum Laboratories), until the >0.5 μm particulate fraction had been concentrated ~100×. The resulting filtrate (<0.5 μm, ~ 20 L) was then fractionated a second time using a membrane with a 500-kD (~ 0.02 μm) cut-off (Spectrum Laboratories, mPES MiniKros Module: N04-E500-05-N, 5-50L) until the 0.02 < x < 0.5 μm colloidal fraction had been concentrated ~100×.

All water fractions (i.e., >0.5 μm and 0.02 < x < 0.5 μm), as well as surface layer samples, were subjected to total decomposition using microwave-assisted acid digestion. The samples (2 g) were digested in an UltraWAVE microwave system (Milestone Inc.) with 1 mL hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>, 30%), 1 mL nitric acid (HNO<sub>3</sub>), and 0.5 mL hydrofluoric

acid (HF). After digestion, excess HF was immediately neutralized with boric acid (0.4 g) and the resulting digest was diluted to 25 mL with 5% HNO<sub>3</sub>. Using quadrupole ICP-MS (Perkin Elmer Nexion 300X), samples were then analyzed for Ti and Zn concentrations as indicators of mineral UV filter occurrence. Al, Fe, and V elements were also quantified as potential proxies for terrestrial materials in order to better distinguish the UV filter signal apart from the local background. V, showing stable local background levels at all sampled distances, was selected for this purpose.

*Organic UV filters* - We investigated the occurrence of five commonly used organic UV filters, namely dioxybenzone (DIOXY), oxybenzone (OXY), avobenzone (AVO), 2-ethylhexyl-4-methoxycinnamate (OMC), and octocrylene (OC) (Table 1) using liquid chromatographic/mass spectrometric analyses (Table 1).

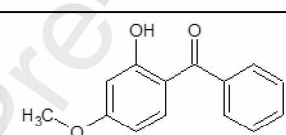
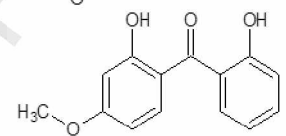
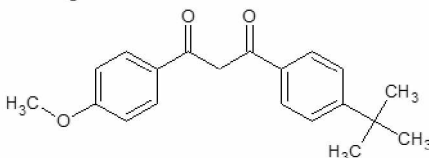
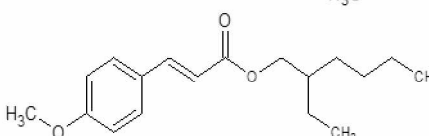
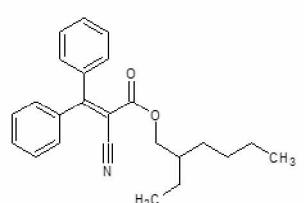
Compound/ CAS number	Formula	Molecular weight	Chemical structure	pKa	Log K <sub>OW</sub>
Benzophenone-3 (or Oxybenzone)/ 131-57-7	C <sub>14</sub> H <sub>12</sub> O <sub>3</sub>	228.247		7.56	3.79
Benzophenone-8 (or dioxybenzone)/ 131-53-3	C <sub>14</sub> H <sub>12</sub> O <sub>4</sub>	244.246		7.11	4.31
Butyl-methoxy - dibenzoylmethane (or avobenzone)/ 70356-09-1	C <sub>20</sub> H <sub>22</sub> O <sub>3</sub>	310.393		9.74	2.41
Ethylhexyl-methoxycinnamate (or octyl methoxycinnamate) /5466-77-3	C <sub>18</sub> H <sub>26</sub> O <sub>3</sub>	290.403		-	5.80
2-Ethylhexyl-2-cyano-3,3 diphenylacrylate (or octocrylene)/ 6197-30-4	C <sub>24</sub> H <sub>27</sub> NO <sub>2</sub>	361.485		-	7.35

Table 1 Chemical structure and relevant data of the target organic UV filters

Standards of OXY, DIOXY, AVO, OMC, OC and the internal standard benzophenone-d10 were purchased from Sigma-Aldrich Fluka (purity > 98 %). Standard solutions of UV filters were prepared with methanol (Ultra Chromasolv, purity > 99.9%). Sodium sulfate

(ACS reagent grade) was purchased from Sigma-Aldrich, India. Sulfuric acid (analytical grade) was purchased from Fisher Scientific, UK. Methyl tert-butyl ether (MTBE, Chromosolv, HPLC grade, Sigma-Aldrich, Germany) was used for extraction. Reconstituted seawater was prepared according to the ASTM International standard practice for the preparation of substitute ocean water (ASTM-International 2013).

Seawater samples (50 mL) were filtered using cellulose filters papers, then adjusted to pH 3.0 using concentrated sulfuric acid, and an internal standard (benzophenone-d10) was added. Liquid-liquid extraction (LLE) of the filtrates was then conducted with 5 mL MTBE by shaking vigorously for 2 min. Sodium sulfate (10 g) was added to enhance separation of the organic and aqueous phases. Extracts (5 mL) were further concentrated by exposing them to a gentle stream of nitrogen to a final volume of 0.5 mL at 50 °C. The total concentration factor thus obtained was 100 (10 by LLE and 10 by reduction of MTBE volume). Recovery ranged between 89 and 117%. Within the studied concentration range, the calibration curves were linear ( $r^2 > 0.99$ ). Limits of quantification (LOQ) were determined as a signal-to-noise ratio 3:1 and ranged between 1 and 8 ng L<sup>-1</sup>. Further details on the UPLC MS/MS analytical procedure followed for UV filter quantification is given in the Supplementary Material file (Section S1, Table S2).

### 2.3. Social survey

We assessed beach attendance through user counts and photographic images. At Prophète Beach, which is only accessible via two narrow stairways, the exact number of people entering and exiting the beach was counted at the beach entrance from 8:00 am until 8:00 pm each day. This was not possible at the other two studied beaches because of their wide and open access. Meanwhile, hourly photographic images were taken at the three beaches from 8:00 am until 8:00 pm and analyzed in order to count people on the beach and bathers into the water. Simultaneously, a questionnaire was carried out on-site between 8:00 am and 8:00 pm. We surveyed 471 people, corresponding to 112, 103 and 256 interviewees at La Lave, Pointe Rouge and Prophète beaches respectively. Our survey strategy was aimed at having equal representation of respondents by gender and age group. In addition to questions concerning their visits to the beach (frequency), activities generally practiced, individual behaviors, and the perception of the beach management by local authorities, beachgoers were asked specific questions about their sunscreen use (type, frequency, and time of application) and their opinion regarding the

possible effects of these products on the marine environment (Table S3). When applicable, the containers of the sunscreen products used by the interviewees were photographed and the information on the packaging, such as brand, SPF, and composition were recorded. A total of 124 sunscreen product compositions were reported. This enabled us to estimate the average composition and maximum UV filter (i.e., organic and mineral) quantity that could be released daily into the beach water.

### 3. Results and Discussion

#### 3.1. UV filter quantification in seawater

##### 3.1.1. Time evolution of organic UV filter concentrations in beach water

Figure 3 shows the temporal impact of bathing activity on the water chemistry by plotting the time dependent trend of the organic filter concentrations in Prophète Beach water. Three different sampling times are compared in terms of OC, AVO, and OXY concentrations. Only one sampling time corresponds to bathing activity, on Saturday, July 15<sup>th</sup> at 4 :00 pm, while the two other sampling times were at 8 :00 am the morning of the same day and 8:00 am the following day (July 16<sup>th</sup>). This clearly shows that the three UV filters are found in the water column only in the bathing zone (BZ) and during recreational activity. At 4 :00 pm on July 15<sup>th</sup>, in the water column of BZ1 and BZ2, OXY, OC and AVO levels were 15, 30, 0 and 0, 10, 300 ng/L, respectively. At the same time, they were not detected beyond the bathing zone (BBZ, Figure 2), and at the two other time points associated with low bathing activity there was also low occurrence. This trend was not as clearly observed in the top surface layer (tsl), where other factors certainly play a role, such as sun radiation, wind-induced water renewal, and tides (maximum tidal amplitude of 15 cm on July 15<sup>th</sup>). Faster photodegradation of the molecules certainly takes place, coupled with easier UV filter transport over long distances. OC has been proven to be photostable, unlike AVO and OXY, with OXY easily being photodegraded into 2,4-dimethylanisole under natural sunlight, especially in presence of natural organic matter (Santos, Miranda et al. 2012, Manasfi, Coulomb et al. 2017). Indeed, OC was always detected at 8:00 am in the top surface layer at all the distances from the shoreline (50 to 500 ng/L), probably resulting from its release in the bathing zone the day before and its subsequent transport. This was not the case for the

two other molecules that were always detected in lower concentrations in the top surface layer compared to the OC.

*Figure 3 around here*

Figure 3. Time evolution of organic UV filter concentrations at the three distances from shore line at Prophète Beach. Concentrations in water column (col) are compared to those in the top surface layer (tsl).

These results show not only the impact of bathing activity on the water chemistry, they also suggest that the residence time of the organic UV filters in the water column is quite short, since no detectable concentration was measured before, nor remained in the morning following peak attendance. Molecule degradation coupled with vertical and horizontal migration are probably the main driving factors. For this reason, in order to assess the overall UV filter release into the beach water, we focused further on the water composition during peak attendance at the three beaches, and also included the analysis of inorganic UV filters.

### **3.1.2. UV filter detection during peak bathing activity**

The concentrations of Ti and Zn recovered in the seawater at the different distances (i.e., BZ1, BZ2, and BBZ) and depths (i.e., top surface layer and 40 cm) of the bathing zone of the three beaches give insight as to the occurrence of mineral UV filters suspended in the water (Figure 4, Table 3). Since these elements occur naturally in the marine environment, it can be challenging to distinguish Ti and Zn originating from sunscreen. However, our hypothesis is that the chemical composition of the waters measured far beyond the bathing zone (BBZ), can be considered as the local background, while the waters in the bathing zone are more likely to be significantly impacted by recreational activity, and thus sun care products. Any enrichment in Zn or Ti in the bathing zone of the three studied sites as compared to BBZ concentrations could thus be attributed to mineral UV filters. Moreover, since resuspension of terrigenous materials from the sediments may likely occur in the bathing zone due to recreational activities and lead also to enrichment in those elements (Reed, Martin et al. 2017), distinction between natural and anthropogenic Zn and Ti was performed by tracking V element as a proxy of terrigenous materials (Gondikas, von der Kammer et al. 2014).

In the three beaches studied, both Zn and Ti were detected in higher concentrations in the bathing zone, i.e. at BZ1 and BZ2, compared to the BBZ site. Prophète and La Lave

displayed similar patterns of Ti concentration in the bathing zone, with a very high range of 70 – 500  $\mu\text{g/L}$  in the water top surface layer and a lower range of 10-30  $\mu\text{g/L}$  in the water column, while a much lower concentration was measured beyond the bathing zone (approx. 5  $\mu\text{g/L}$ ) (Table 3). Zn concentrations leveled off at lower values than Ti, around 10  $\mu\text{g/L}$ , and were also more concentrated, at 80-90% in the bathing zone nearest the shore (i.e., BZ1). The Ti and Zn concentrations recovered in the waters of Pointe Rouge Beach indicated very few occurrences of mineral UV filters in the top surface layer, but similar concentrations to those observed at the other beaches were found in the water column. The geographic profile of the Pointe Rouge Beach makes it more exposed to local marine currents and winds than the two other beaches, which are more protected by artificial dikes. This likely implies a more constant renewal of the water's top surface layer and thus a more limited UV filter accumulation.

*Figure 4 around here*

Figure 4 Concentrations of Ti (A) and Zn (B) found in different water fractions at 4:00 pm on July 15, 2017 in Bathing Zone 1 (BZ1), Bathing Zone 2 (BZ2), and Beyond Bathing Zone (BBZ).

The sampled water column was further fractionated into two size classes in order to distinguish the nanoparticulate UV filters free in suspension ( $0.02 < x < 0.5 \mu\text{m}$  fraction) from those existing as a part of larger aggregates ( $> 0.5 \mu\text{m}$  fraction). Indeed, this analysis revealed that the mineral UV filters are mostly concentrated in the larger size fraction, i.e. behaving in an aggregated form (Figure 4, Table 2). This size can correspond to either the original size of the sunscreen residues washed off the user's skin, or to the aggregation of the smaller mineral filters after their release in water. Salt-induced homo-aggregation of these filters or hetero-aggregation with natural particulate matter can indeed take place in such natural systems (Botta, Labille et al. 2011, Labille, Harns et al. 2015). Finally, the attribution of Ti and Zn patterns to the occurrence of mineral UV filters in seawater was well supported by Ti/V and Zn/V elemental ratios, with V being used as a proxy for geogenic materials (Figure S1 and S2). If the enrichments in Ti and Zn in the bathing area were only due to natural sediment resuspension, Ti, Zn, and V would demonstrate a co-evolution. Here the elemental ratios followed trends with the different sampled distances similar to those of Ti and Zn alone, confirming that these enrichments are not of terrigenous origin. Moreover, the fact that the same pattern of Ti and Zn is obtained for the three beaches at the same time of peak recreational activity

also supports the assumption that UV filters from the recreational area are the main contributors to this pattern.

distance		La Lave			Prophète			Pointe Rouge		
		tsl	>0.5 $\mu\text{m}$	0.02-0.5 $\mu\text{m}$	tsl	>0.5 $\mu\text{m}$	0.02-0.5 $\mu\text{m}$	tsl	>0.5 $\mu\text{m}$	0.02-0.5 $\mu\text{m}$
Ti	BZ1	91.7	29.4	0.5	70.7	10.2	3.1	5.1	8.3	0.8
	BZ2	8.3	0.2	0.3	541.4	0.3	0.4	3.6	6.4	0.1
	BBZ	5.9	0.6	0.2	4.7	0.1	0.4	4.8	0.5	0.1
Zn	BZ1	11.9	0.8	0.2	9.0	1.5	0.9	<LD	1.7	0.1
	BZ2	<LD	0.0	0.3	<LD	0.2	0.3	<LD	0.4	0.3
	BBZ	<LD	0.3	0.0	<LD	0.1	0.3	3.7	0.5	0.2
TiO <sub>2</sub>	BZ1	152.9	49.1	0.9	117.9	17.0	5.1	8.6	13.9	1.4
	BZ2	13.8	0.4	0.6	903.1	0.6	0.7	6.0	10.7	0.2
	BBZ	9.9	1.0	0.4	7.8	0.2	0.7	8.0	0.8	0.2
ZnO	BZ1	14.8	1.1	0.2	11.2	1.8	1.1	<LD	2.2	0.1
	BZ2	<LD	0.0	0.3	<LD	0.3	0.4	<LD	0.5	0.3
	BBZ	<LD	0.4	0.0	<LD	0.1	0.4	4.6	0.6	0.2

Table 2. Ti and Zn concentrations ( $\mu\text{g/L}$ ) measured in beach water for the three beaches, three distances from the shoreline and three compartments top surface layer (tsl), size fraction larger than  $0.5 \mu\text{m}$  and size fraction comprised between  $0.02$  and  $0.5 \mu\text{m}$ . Element concentrations are also converted in terms of equivalent TiO<sub>2</sub> and ZnO concentrations, based on molar masses ( $\mu\text{g/L}$ )

Among the five organic UV filters analyzed in the beach water samples, DIOXY was never detected and OMC only occurred once, but OC, AVO, and OXY were often observed in the bathing water (Figure 5, Table 3). Regarding the fractionation between the top surface layer and the water column or the spatial distribution with distance from the shoreline, there was no clear pattern for these molecules as was observed with the mineral filters. This is certainly due to the respective behavior and lifetime of the molecules at the two sampled water depths. OC and AVO were always detected in the bathing zone of the three beaches, totaling  $75\text{-}425 \text{ ng/L}$  and  $10\text{-}350 \text{ ng/L}$ , respectively. OC was generally equally distributed at both depths while AVO was mostly concentrated in the water column with a small amount in the top surface layer. OXY was only detected at Pointe Rouge and Prophète, ranging from  $50$  to  $75 \text{ ng/L}$ , with a significant contribution in the top surface layer. At La Lave and Prophète beaches, the occurrence of these molecules in the water column clearly showed again a higher concentration in the bathing zone, while they were not detected at BBZ. This was not as clear in the top surface layer, which may have been more impacted by winds that could rapidly push the UV filters offshore (e.g. OC and AVO in Prophète at BBZ).



*Figure 5 around here*

Figure 5. Concentrations of organic UV filters (ng/L) OC, AVO, OXY, OMC, and DIOXY found in water at 40 cm depth and top surface layer at 4:00 pm on July 15, 2017 in Bathing Zone 1 (BZ1), Bathing Zone 2 (BZ2), and Beyond Bathing Zone (BBZ). \*Top surface layer samples were not analyzed for Pointe Rouge.

	distance	La Lave		Prophète		Pointe Rouge	
		tsl	col	tsl	col	tsl	col
OC	BZ1	85.0	148.4	44.8	31.4	/	149.8
	BZ2	419.9	0.0	44.4	0.0	/	0.0
	BBZ	0.0	0.0	141.6	0.0	/	141.4
AVO	BZ1	10.3	12.8	15.3	0.0	/	23.5
	BZ2	0.0	10.7	31.1	309.1	/	0.0
	BBZ	0.0	0.0	48.1	0.0	/	6.1
OXY	BZ1	0.0	0.0	37.9	17.0	/	78.3
	BZ2	0.0	0.0	50.0	11.3	/	31.0
	BBZ	0.0	0.0	<LD	0.0	/	26.7
OMC	BZ1	0.0	0.0	<LD	0.0	/	8.8
	BZ2	0.0	0.0	<LD	0.0	/	2.6
	BBZ	0.0	0.0	<LD	0.0	/	0.0

Table 3. Organic UV filter concentrations (ng/L) measured in beach water for the three beaches, three distances from the shoreline and two depth compartments top surface layer (tsl) and water column (col). \*Top surface layer samples were not analyzed for Pointe Rouge.

### 3.2. Consumer habits regarding bathing and sunscreen use

For the 471 people interviewed at the three beaches, their habits regarding bathing activity are depicted in Figure 6. Bathing is a very common practice, with more than 75% of respondents having a bath every time they go to the beach and less than 3% never bathing (Figure 6). When going to the beach, attendees (40-60%) bathe 2.6 times on average and 91.5% of them practice whole body immersion. These data reflect how bathing provides an opportunity to refresh the body in a Mediterranean climate. Of note, the average air temperature measured over the week of the field campaign between 9:00 am and 7:00 pm was 25 °C (Figure S3).

*Figure 6 around here*

Figure 6. Beachgoer habits regarding bathing activity at the beach.

The use of suncare products on the beach is depicted in Figures 7. On average, 68.4% of the interviewees use a suncare product and apply it to their skin 2.6 times per visit to the beach. Details on sunscreen application and bathing frequencies are depicted in Figure S4. Of note, contrasting practices were observed between La Lave and the two other beaches, which might be related to the ethnic origins and socio-professional status of the respondents (Figure 7). Only 50% of the interviewees at La Lave Beach, which is mostly attended by people living in the nearby working class neighborhoods and having North African as well as sub-Saharan roots, use suncare products, This is compared to 70% of attendees reporting suncare product use at the two other beaches which welcome tourists and city center inhabitants. However, no clear difference was observed regarding the part of body on which the suncare product is applied, with 80% of all consumers applying it to the whole body.

*Figure 7 around here*

Figure 7. Suncare product consumption habits on the beach. The insert presents average values over the three beaches studied.

Among the list of UV filters authorized in Annex VI of the EU Cosmetic Regulation (European 2009, Sobek, Bejgarn et al. 2013), Figure 8 displays their occurrence in the suncare products consumed by the interviewees at the three beaches. Some organic UV filters are clearly more favored by the manufacturers. The six most common being avobenzone > bemotrizinol > octocrylene > octisalate > ethylexyl triazone > homosalate, which are found in 78 to 31% of the products, respectively. These percentages logically correspond to protections against both UVB and UVA ranges which are usually desired together in the formulation. Nevertheless, since UVA specific filters are available in a more limited choice for the manufacturers, the preferred candidate avobenzone is statistically more abundant on the market. The mineral UV filters, TiO<sub>2</sub> and ZnO, were found in 19% and 2% of the products, respectively.

*Figure 8 around here*

Figure 8 UV filter occurrence among the sun-care products used on the three beaches. Data recovered from photographs of 124 products. Specific UV absorption range of each filter is also given as /filter name – UV range/. For mineral UV filters, the occurrence of non-nano and nano forms are given; an overlap exists for TiO<sub>2</sub> as both forms are labeled on certain products.

### 3.3. Estimation of the UV filter flux from consumer skin to beach water

#### 3.3.1. Average UV filter flux

From the data collected on the field through the consumer survey, an assessment of the daily UV filter mass used on the consumer skin and possibly transferred to the beach water was performed. Here any retention of the product on the skin through bathing was not taken into account, so that a worst-case contamination scenario could be estimated. The flux of the sun-care product,  $m_{\text{sun-care}}$ , consumed and transferred to the bathing water can be obtained from Eq. 1.

$$m_{\text{sun-care}} = N \cdot \chi \cdot \gamma \cdot m \quad \text{Eq. 1}$$

where  $N$  is the total number of people attending the beach,  $\chi$  is the % of people that reported using sun-care products before bathing,  $\gamma$  is the average number of times the product is applied to the skin per visit to the beach, and  $m$  is the mass of product consumed during each application to the entire body. The lowest and highest values recovered from the survey over the three beaches were used for  $\chi$  and  $\gamma$  considerations to obtain a reasonable range for  $m_{\text{sun-care}}$ . People who reported applying a sun-care product before bathing represented 29, 47 and 53 % of the attendees at La Lave, Prophète, and Pointe Rouge beaches, respectively, and the average number of applications was 2.8, 2.4 and 2.9 respectively. A daily beach attendance of  $N = 3,000$  people was used for this estimation as it is typical for the Prophète Beach studied in this work (Figure S5). Finally,  $m = 15$  g was taken as the mass of sun-care product used for each application to the entire body (Ficheux, Chevillotte et al. 2016). We thus obtained:

$$m_{\text{sun-care}} = 52.5 \pm 16.5 \text{ kg/day} = 367 \pm 114 \text{ kg/week} = 1.45 \pm 0.45 \text{ t/month} \quad \text{Eq. 2}$$

From  $m_{\text{sun-care}}$ , the corresponding mass of UV filter  $i$  involved,  $m_{UVi}$ , can be obtained from the composition of the products used, following Eq. 3

$$m_{UVi} = \%_{UVi} \cdot \phi_{UVi} \cdot m_{\text{sun-care}} \quad \text{Eq. 3}$$

where  $\%_{UVi}$  corresponds to the occurrence of the UV filter  $i$  within the panel of sun-care product compositions used, given in Figure 8, and  $\phi_{UVi}$  is the average mass fraction of the

UV filter  $i$  used in the product formulation.  $\phi_{UVi}$  is not usually provided in Europe on the product packaging, only the list of components. Here, we propose a range  $0.03 < \phi_{UVi} < 0.1$  as a reasonable estimation, considering literature data (Botta, Labille et al. 2011, Matta, Zusterzeel et al. 2019), maximum UV filters concentrations authorized by EU regulation, and the fact that most of the sun care products registered in our survey display a high SPF ranging from 30 to 50+. From these values of  $\%_{UVi}$ ,  $\phi_{UVi}$  and  $m_{sun care}$ , the average and a relevant range of  $m_{UVi}$  were obtained.

UV filter $i$	$m_{UVi} av$	$m_{UVi} range$
avobenzone	2,669	845 - 5,398
bemotrizinol	2,119	671 - 4,285
octocrylene	1,899	601 - 3,840
octisalate	1,789	566 - 3,617
ethylexyl triazone	1,348	427 - 2,727
homosalate	1,073	340 - 2,170
TiO <sub>2</sub>	633	200 - 1,280
ZnO	83	26 - 167
Total mass	15,742	4,982 - 31,829

Table 4. UV filter mass consumed and potentially entering the bathing area, given in g/day, with average ( $m_{UVi} av$ ), total range. These values account for a daily attendance of 3000 beachgoers. The total mass is obtained from the sum of all the UV filters quantified in Figure 8.

These values are presented in Table 4 for the six most abundant organic UV filters and the mineral filters, ZnO and TiO<sub>2</sub>. Estimated consumption of organic UV filters is more than one kilogram per day each, and 83 and 633 g/d for the mineral filters, ZnO and TiO<sub>2</sub> respectively. Summing all the UV filters quantified in Figure 8 results in a total mass of 15.7 kg per day potentially released into the beach water. This large quantity of cosmetic product may be released or remain on the skin through bathing activity, depending on the retention factor of the product. This value accounts for a beach welcoming 3,000 users per day.

### 3.3.2. Predicted environmental concentration and measured concentration

From the  $m_{UVi}$ , based on our survey analysis, we calculated a predicted environmental concentration for the UV filter  $i$  in the bathing water,  $PEC_{UVi}$ , (Eq. 4)

$$PEC_{UVi} = m_{UVi} / V \quad \text{Eq. 4}$$

where  $V$  is the volume of water in the bathing zone.

Our aim was to investigate whether  $PEC_{UVi}$  could be compared to the actual UV filter concentrations  $C_{UVi}$ , measured in this work in the bathing zone, and used to interpret any eventual divergence in terms of UV filter retention, release, or fate. The estimation of  $PEC_{UVi}$  was only completed for the Prophète Beach, where the bathing zone is delimited between a dike and the shoreline, facilitating the estimation of  $V$ . We used 85 m length x 64 m width and bathymetric data ranging from 0 to 3 m depth, giving 4,530 cubic meters of water (Figure S8). In a coastal system such as this, constant water renewal should be considered as a source of dilution, but here water renewal is limited by the dike. To compensate for this effect, we used an average water residence time of 24 h (Basterretxea, Garces et al. 2007), giving a renewal of 29% of this volume after 7 hours of recreation time cumulated at the 4:00 pm sampling time. This gave a total water column volume of  $V_{col} = 5,850 \text{ m}^3$ . As for the top surface layer, the sampling consisted of roughly 1 cm of water layer thickness and the measured surface area of the bathing zone was 4,114  $\text{m}^2$ . This gave  $V_{tsl} = 53 \text{ m}^3$  including 29% renewal.

In order to compare our estimation of the filter concentration,  $PEC_{UVi}$ , based on the survey, to the actual UV filter concentration measured in the bath water,  $C_{UVi}$ ,  $PEC_{UVi}$  was further refined regarding the actual attendance and bathing activity at Prophète Beach during the sampling time. Indeed, the declarations from our survey reflect the users' habits, and thus do not take into account any punctual events that may disturb those habits, such as bad weather. On July 15th, 2017, the day of water sampling at the recreational peak, a cumulated beach attendance of  $N = 2,700$  people was measured at 4:00 pm, which is close to the average daily attendance (Figure S5). However, the actual number of bathers was unusually low because of the colder water temperature resulting from cold and windy weather the previous day (Figure S5-7) (water temperature on 07/15/17 = 18°C). Our daily survey during the entire week enabled us to estimate that bath practice was two times lower on July 15th than on warm days on average. A correction factor of 0.5 should thus be applied on  $m_{suncare}$ , to obtain  $PEC_{UVi}$ , on July 15th at 4:00 pm, giving Eq. 5 for water column.

$$\begin{aligned} PEC_{UVi, July\ 15, 4pm} &= \frac{\%_{UVi} \cdot \phi_{UVi} \cdot N \cdot \chi \cdot \gamma \cdot m}{V} \times 0.5 \\ &= \frac{\%_{UVi} \times 0.065 \times 2,700 \times 0.47 \times 2.4 \times 15 \times 10^6 \times 0.5}{2,600 \times 10^3} \end{aligned} \quad \text{Eq. 5}$$

$C_{UVi}$  values are obtained from Tables 3 and 4. Note that the  $C_{UVi}$  values for  $TiO_2$  and  $ZnO$  were calculated by subtracting the geochemical background measured in BBZ from the value measured in BZ1. A recovery value  $\alpha_{UVi}$  could then be calculated in both the water column and the top surface layer by normalizing  $PEC_{UVi, July 15, 4pm}$  to  $C_{UVi}$  (Table 5). If  $\alpha_{UVi} \approx 100\%$  was recovered, then the worst-case scenario previously described, where the totality of UV filters consumed are released into the bathing water, would be reality. Here, relatively high values of  $\alpha_{UVi}$  were obtained for the mineral UV filters, with 49 and 19% of  $TiO_2$  recovered in the water column and top surface layer, respectively. The same order of recovery was obtained for  $ZnO$  in the water column ( $\alpha_{ZnO, col} = 31.7\%$ ) but a lower amount was detected in the top surface layer ( $\alpha_{ZnO, tsl} = 1.3\%$ ). This may be attributed to the very low number of products containing  $ZnO$  UV filters reported in our survey ( $\varnothing_{ZnO} = 3\%$ ), which resulted in a limited range of cosmetic formulation types. Apparently these few products dispersed rather well in the water column. As for  $TiO_2$  mineral UV filters,  $\varnothing_{TiO_2} = 17\%$  resulted in a wider variety of environmental fates, leading to the filter occurrence in both the water column and the top surface layer. Different causes may be discussed to explain why the UV filter recovery is not complete. The rapid sedimentation of the UV filters at the bottom of the water column once washed off the skin is rather unlikely because the water in the bathing zone is constantly agitated during the recreation peak. The retention factor of the cosmetic product on the consumer skin certainly plays a major role in  $\alpha_{UVi}$ . Values from 25 to 50% have been proposed (Danovaro, Bongiorno et al. 2008, Slijkerman and Keur 2018), which fall in the same order as the  $\alpha_{TiO_2}$  and  $\alpha_{ZnO}$  values obtained here.

UV filter <i>i</i>	$\%_{UVi}$	$m_{UVi}/g$	$PEC_{UVi}/\mu g/L$	$C_{UVi}/\mu g/L$	$\alpha_{UVi}/\% \text{ recov.}$
TiO <sub>2</sub> col	17	251	43	21	49.0
TiO <sub>2</sub> tsl			4,724	117-895	19.0
ZnO col	3	44	7.6	2.4	31.7
ZnO tsl			834	11	1.3
AVO col	80	1,181	202	0.3	0.15
AVO tsl			22,230	0.015-0.05	0
OC col	57	842	144	0.03	0.02
OC tsl			15,840	0.04-0.14	0
OMC col	17	251	43	0	0
OMC tsl			4,724	0	0
OXY col	2	30	5	0.015	0.30
OXY tsl			556	0.04-0.05	0.01

Table 5. Estimation of UV filters fluxes on Prophète beach on Saturday 07/15/17, 4:00 pm, in terms of occurrence among the products surveyed  $\%_{UVi}$ , estimated mass involved  $m_{UVi}$ , predicted environmental concentration in bath water  $PEC_{UVi}$ , and actual concentrations measured  $C_{UVi}$ , at 40 cm depth (col) and in the top surface layer (tsl).

As for the organic UV filters, the concentrations measured,  $C_{UVi}$ , fall in the ng/L range. Despite these concentrations being in agreement with the existing literature (Tashiro and Kameda 2013, Tovar-Sanchez, Sanchez-Quiles et al. 2013, Bargar, Alvarez et al. 2015, Rodriguez, Sanz et al. 2015, Downs, Kramarsky-Winter et al. 2016, Tsui, Lam et al. 2017, Kung, Lee et al. 2018, Mitchelmore, He et al. 2019), they fall two orders of magnitude below our predicted concentrations  $PEC_{UVi}$ . Recoveries as low as 0.3, 0.15, 0.02 and 0.0 % were obtained for OXY, AVO, OC and OMC respectively in the water column and always 0.0% in the top surface layer. Such divergence with the mineral UV filters was not expected. Even if photodegradation of these molecules is a known mechanism, here, water sampling was realized precisely during the peak of recreation, which should limit this effect in the present results. Our hypothesis is that the retention factor on the skin for these molecules is much higher since they are known to penetrate through the skin barrier (Matta, Zusterzeel et al. 2019). A few field campaigns focusing on both mineral and organic UV filters have already shown, in agreement with our data, that mineral UV filters are found in bath water in concentrations 1000 times higher than organic UV filters (Tovar-Sanchez, Sanchez-Quiles et al. 2013, Reed, Martin et al. 2017). However, this is the first time that the proportion of both types of UV filters at the time

of potential release, (i.e. during consumer bathing), is known. These results clearly demonstrate the contrasted fates of mineral and organic UV filters once spread on the skin.

### 3.4. Consumer awareness and manufacturer choice

#### *Figure 9 around here*

Figure 9 Answers of the interviewees to the question « Do you think that sun care products can impact the quality of marine bath water ? »

During the survey,  $66 \pm 6$  % of the interviewees believed that sun care products do alter the quality of the bathing water, arguing first that the product is washed off and floats as an oily surface layer (28%), that the chemical components involved are of concern (37%), can pollute the environment (12%) or are bad for the aquatic fauna and flora (13%) (Figure 9). A little more than 19% of the interviewees were not aware of the issue, as they did not have any opinion. Finally, a minority but non-negligible fraction of the respondents,  $14 \pm 6$ %, think that sun care products have no effect on bathing water quality because the sea is too large and dilution effects minimize the impact. Some of them also argued that the sunscreen composition is certainly adapted by the manufacturer or regulators to take this into account. These results indicate a certain discrepancy between the claimed awareness of the consumers regarding the environmental impact of sun care products and the quasi-total absence of any product labeled as eco-friendly among those actually consumed (only 2 out of 170 products). Product composition comes as the criteria number 4 (12% of the consumers) in selecting a sun care product (Figure 10c). However, it shall be noted that only 3% of those respondents actually mention environmental concerns, while 19% of them worry about the product composition because of a possible impact on their own health. More than half, 57%, did not give any explanation and 22% mentioned their preference for the products labeled as organic (BIO). For 73% of respondents, the sun protection factor (SPF) is their paramount consideration in selecting and buying a sun care product (Figure 10c). The distribution of sunscreen SPF labels found on the packaging of the interviewees revealed a clear majority of medium and high levels of protection, namely SPF 30 and SPF 50+ (Figure 10a). This likely reflects the different expectations from the users, with highest sun protection wanted on the one hand, and tanning preferred with a



lower sun protection on the other hand. It also indicates a real awareness of the potential harmful effects of the sun. Sunscreens with SPF values between 15 and 20 were less preferred (13%), probably because the protection they provide is considered too low. The cosmetic products with SPF values from 4 to 10, found in 6% of the panel, were not considered as sunscreens. Likewise for the UV filter-free skin care products, such as monoï or other moisturizing oils, that constituted 4% of the products consumed.

The frequency of different suncare product brands used on the beach is depicted in Figure 10b. It shows that more than 50% of the local suncare product consumption is dominated by only 5 well-known brands, Garnier Ambre Solaire (L'Oréal), Nivea Sun (Beiersdorf), Avene (Lab. Pierre Fabre), La Roche Posay (L'Oréal) and Bioderma (NAOS), in accordance with the French suncare market (Euromonitor 2015).

*Figure 10 around here*

Figure 10 Distribution in % of the sun protection factors (SPF) of the suncare products used on the beach (a), of the cosmetic brands providing these products (b), and of the consumer's three primary criteria for selecting a suncare product. Data shown are averages over the three beaches studied

This suggests that consumers buy the most well-known brands, those that are more heavily advertised, or those that are the most present at points of sale. Looking for effective sun protection, and lacking means of comparison, some consumers may move towards these well-known brands because they are often associated with efficacy. Brand comes as the third most important criteria in selecting a suncare product, with 16% of the consumers actively paying attention to the brand they buy (Figure 10c). Lacking the advertising influence and financial means of many well-known brands, organic (BIO) and/or eco-designed products are often distributed by small or medium businesses which is the most likely reason for why they represent only a minority of the products consumed on the beach.

Of note, only mineral UV filters are currently allowed within products labeled as organic (BIO). This agrees with the minority of this type of filter recovered in Figure 8, namely 19% and 2% of products containing  $\text{TiO}_2$  and  $\text{ZnO}$  respectively.  $\text{ZnO}$  is used more rarely within the European sunscreen market, partly due to questions regarding its high solubility and bioavailability, which may lead to higher risk.  $\text{TiO}_2$  has been widely used as the main mineral option for organic (BIO) UV filters. Nevertheless, its under representation among the products used here suggests that manufacturers still prefer

synthetic organic UV filters. This is likely due to easier formulation processes, preferred texture, and simpler regulation. Indeed, mineral UV filters constitute a significant particle loading in the cosmetic formulation, which increases the challenge in stabilizing the product emulsion (Tyner, Wokovich et al. 2011), results in a less appreciated sensation on skin, and product whiteness in some cases. In addition, the ultrafine particles used to create more transparent formulations often fall under the definition of a nanomaterial, nanosubstance or nanoform and formulators using such materials have to deal with complex and continually changing local regulations (SCCP 2008, French-Government 2012, Sobek, Bejgarn et al. 2013, SCCS 2014, REACH 2018). Of note, 61% of the products containing TiO<sub>2</sub> UV filters were labeled with the (nano) form on their component list, while 78% were labeled as containing TiO<sub>2</sub> without the (nano) characteristic (Figure 8). The sum of both labels corresponds to 139% of the products containing TiO<sub>2</sub>. The overlap is due to manufacturers labeling both nano and non-nano forms on the same packaging, which is likely false and misleading. This illustrates some of the difficulties encountered by the manufacturers and product formulators in characterizing their material size in order to respect regulation. Nevertheless, since regulation should contribute to making our environment safer, the balance between organic and mineral UV filters should be further evaluated in terms of their respective cost/benefit ratio.

#### 4. Conclusion

A field campaign was carried out during the summer of 2017 at three beaches in Marseille along the French Mediterranean coast, with the aim of quantifying the fluxes of UV filters released from the sunscreen on the beachgoer's skin to the bathing seawater.

In the three beaches studied, both Zn and Ti were detected in higher concentrations in the bathing zone than beyond it, and behaved independently of terrigenous materials. Their occurrence could be attributed to the presence of TiO<sub>2</sub> and ZnO mineral UV filters in the seawater, with ranges of 100-900 and 20-50 µg/L for TiO<sub>2</sub> and 10-15 and 1-3 µg/L for ZnO in the top surface layer and in the water column, respectively.

Octocrylene, avobenzone, and oxybenzone were mostly observed in the bathing water during peak recreation time only at concentration ranges of 75-425, 10-350 and 50-75 ng/L respectively. They followed diverging behaviors likely due to multiple factors like

molecule lifetime, water currents, transport in the water column vs surface layer, and distance from the shoreline.

The environmental concentrations of UV filters evidenced here can be put into perspective by considering the predicted no effect concentrations (NOEC) for marine organisms. Slijkerman and Keur recently reviewed these values for different UV filters from the existing literature (Slijkerman and Keur 2018). The NOEC for nano-TiO<sub>2</sub> on crustacean mortality at 48h of exposure was 1,000 µg/L, while it was 10 µg/L for nano-ZnO at 72h of exposure. The NOEC for OC on mollusks and on algal growth inhibition at 96h of exposure was 20 and 40 µg/L, respectively. These values are higher than the UV filter concentrations measured here in seawater, suggesting that no acute effect would be expected on these living organisms. Nevertheless, since the NOEC values only represent short-term exposures, questions remain regarding UV filter chronic effects in the marine environment neighboring recreational areas.

Despite the fact that consumer awareness of this issue is rising, with 66% of the interviewees considering that sunscreen products can impact the quality of bathing water, the sunscreen composition does not appear to be a priority concern for them when buying a product. The protection efficacy of the product against sun radiation remains their paramount consideration. We were able to estimate the product quantities used on the consumer skin and those being possibly transferred to the bathing water, giving an average mass of 52 kg/day or 1.4 t/month of sunscreen products for a beach attended by 3,000 people daily. Their composition was determined from the products used by the interviewees, revealing a cumulated mass of about 15.7 kg of UV filters per day, with a net majority of organic UV filters, at the expense of mineral ones found in only 20% of the products.

Comparing our predicted environmental concentration of UV filters to the actual concentrations measured in bathing water revealed two distinct scenarios for mineral and organic filters. About 30 to 49% of the estimated mineral UV filter flux was effectively quantified in the bathing water, in the water column. This recovery is in agreement with the approaches developed here and suggests a reasonable retention factor of the product on the skin lower than 70%. On the other hand, our recovery in organic UV filters is much lower, ranging between 0 and 0.3%. This suggests a much higher retention factor for organic molecules on the skin, possibly due to transcutaneous internalization.

These results suggest that a new compromise could come into consideration when selecting sunscreen products, between mineral UV filters released to the marine environment and organic UV filters that are able to pass the skin barrier. These two aspects should be heavily weighed in future attempts toward safer-by-design sunscreen products that will have minimal impact on both human and environmental health.

## Aknowledgements

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## Graphical abstract

## Highlights

- 68% of beachgoers use sunscreen 2.6 times per visit to the beach
- 3,000 beachgoers/day = 52.5 kg sunscreen applied by bathers = 15.7 kg UV filter potentially released
- Mineral UV filters recovered in top surface layer and water column at 19 and 49% of PEC
- Low levels (ng/L) of organic UV filters recovered in seawater
- No use of suncare products labeled as eco-friendly, despite consumer awareness



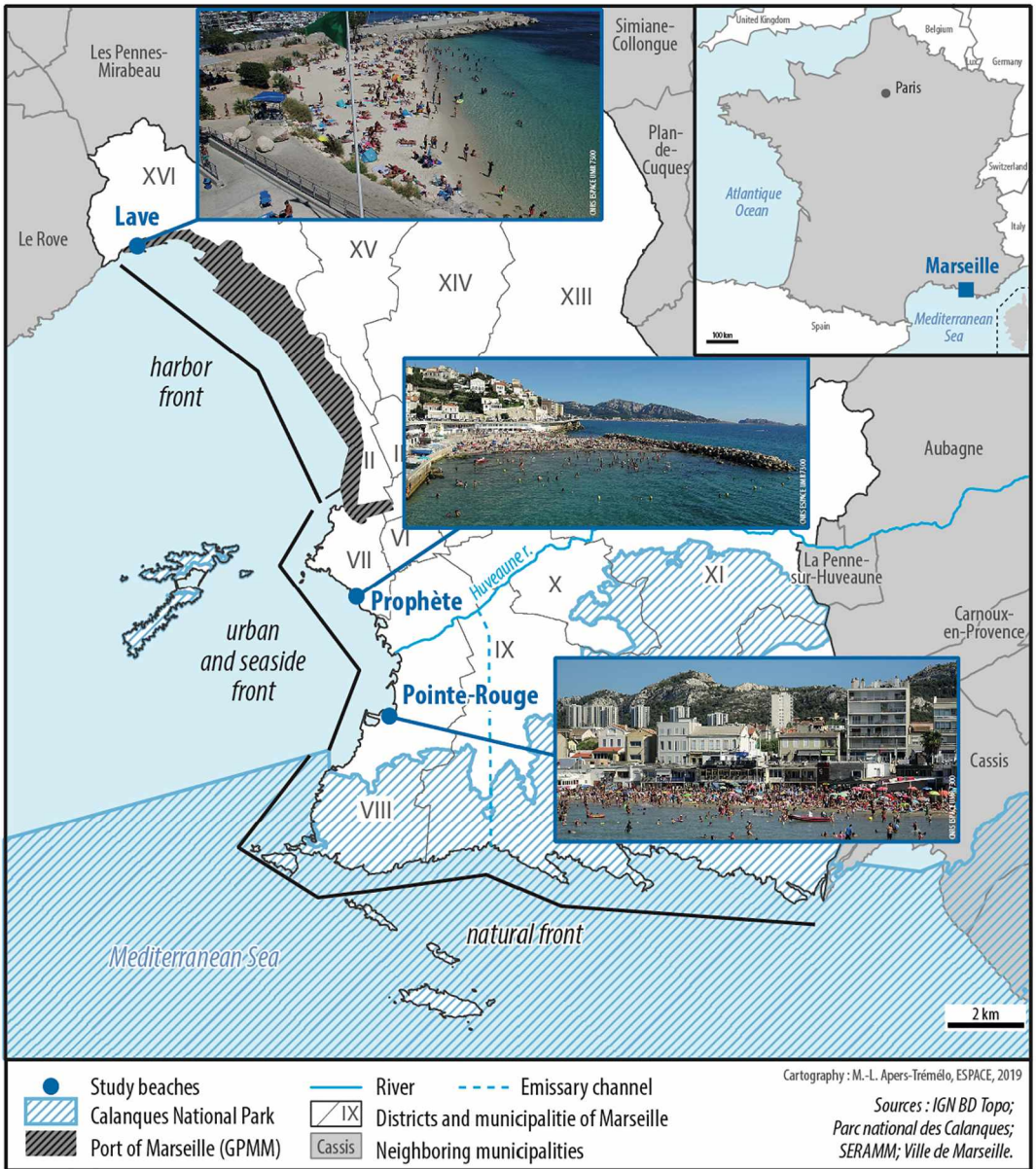
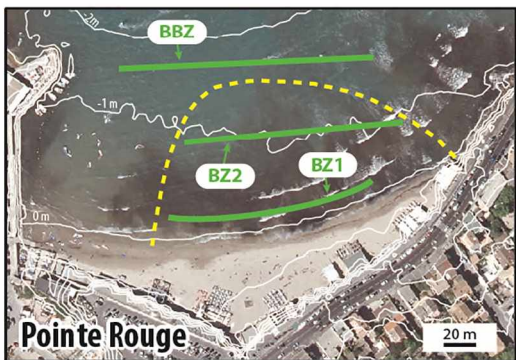
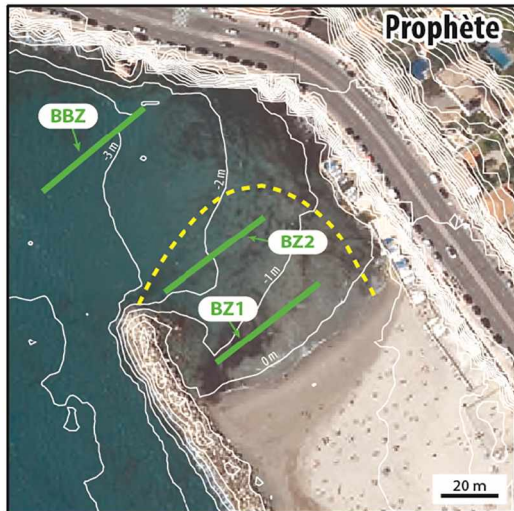
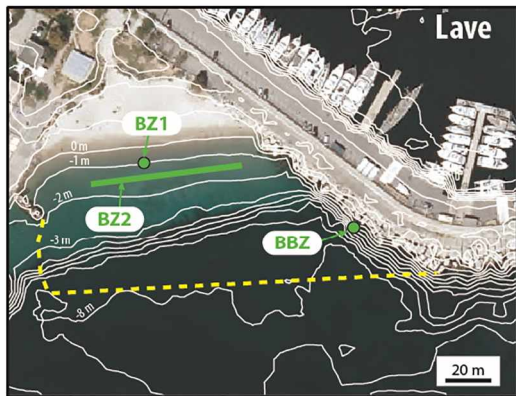





Figure 1



Cartography : M.-L. Apers-Trémelo, ESPACE, 2019

-  sampling point or transect
-  Limits of the bathing area
-  Elevation curves (1 m equidistant)

Source : Litto3D®, 2015, coproduction SHOM / IGN

Figure 2

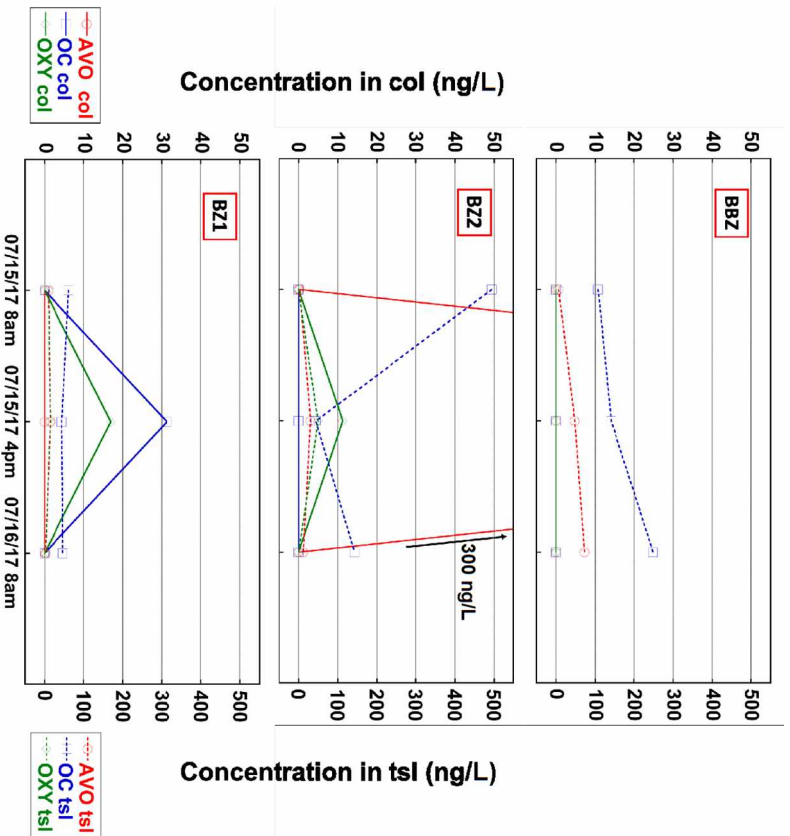
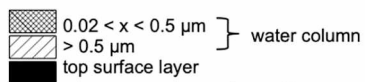


Figure 3

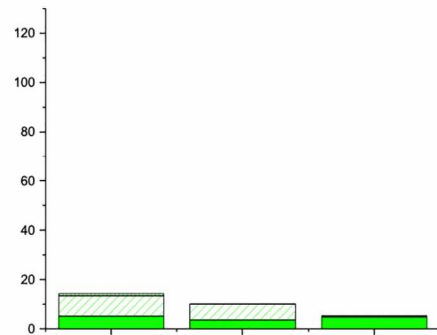
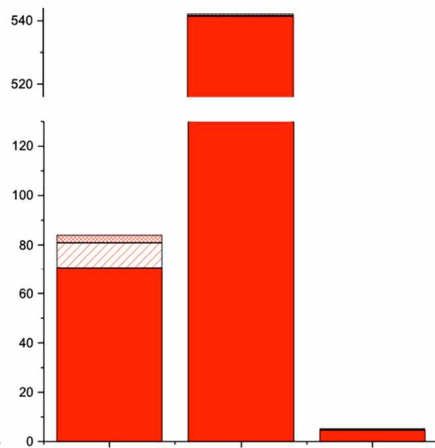
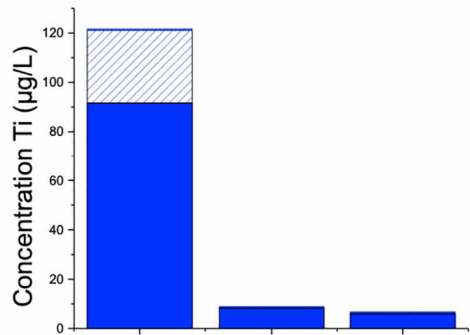


### La Lave

### Prophète

### Pointe Rouge

**A**



**B**

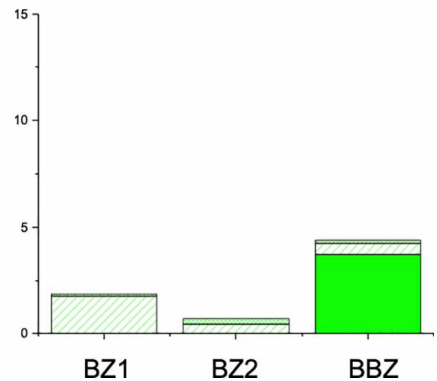
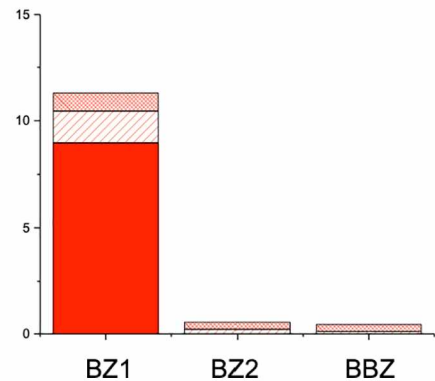
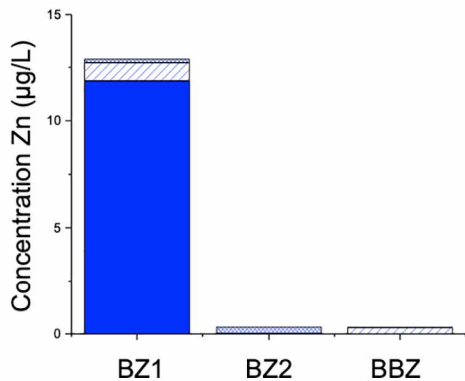
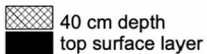
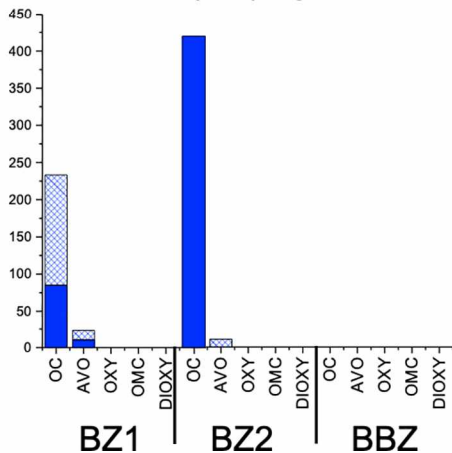


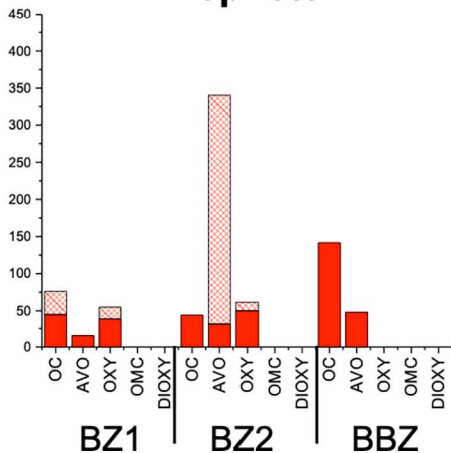
Figure 4



## La Lave



## Prophète



## Pointe Rouge

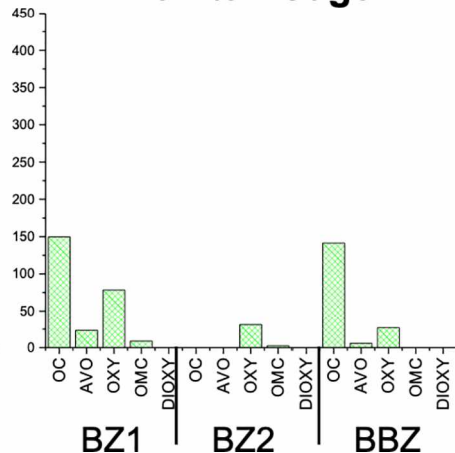
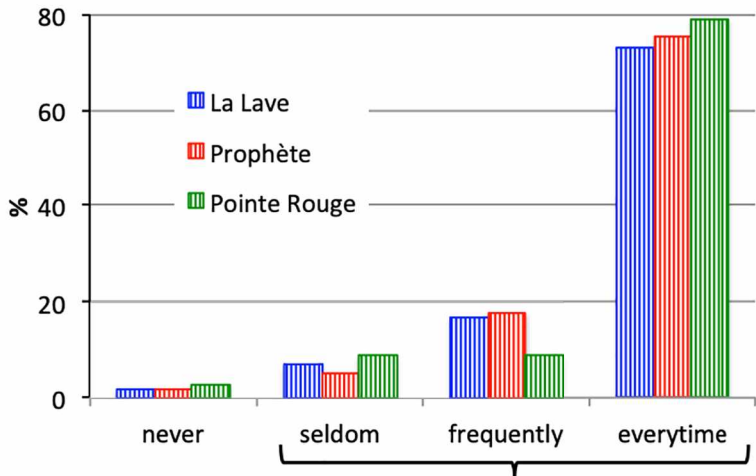
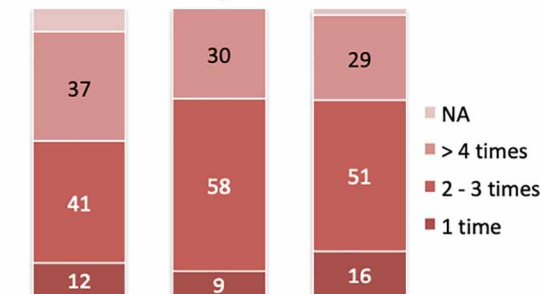


Figure 5

### How often do you practice bathing ?



### If yes, how many times do you bathe per visit?



### Do you practice whole body immersion?

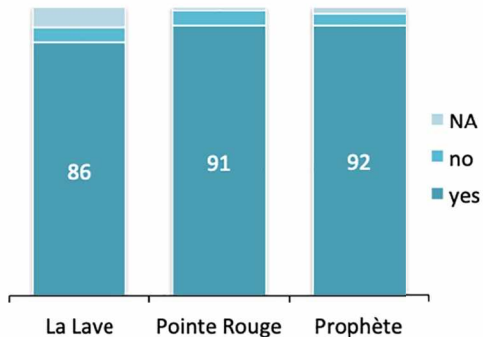


Figure 6

Do you usually use a suncare product?

If yes, do you apply it to...?

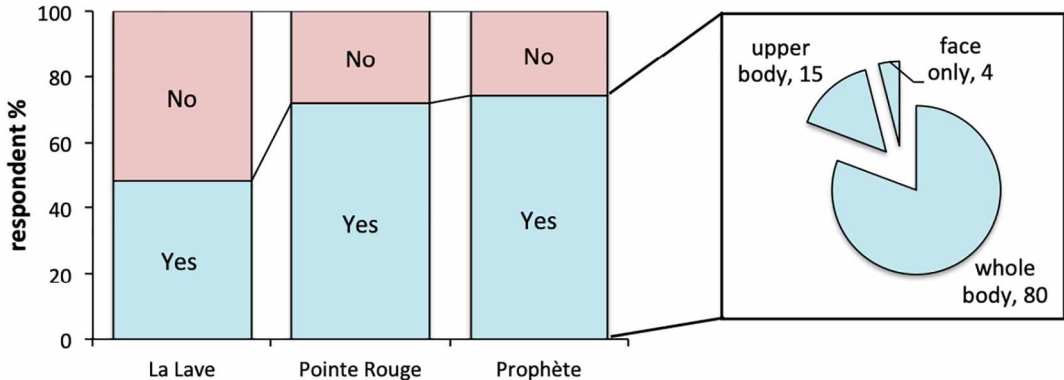


Figure 7

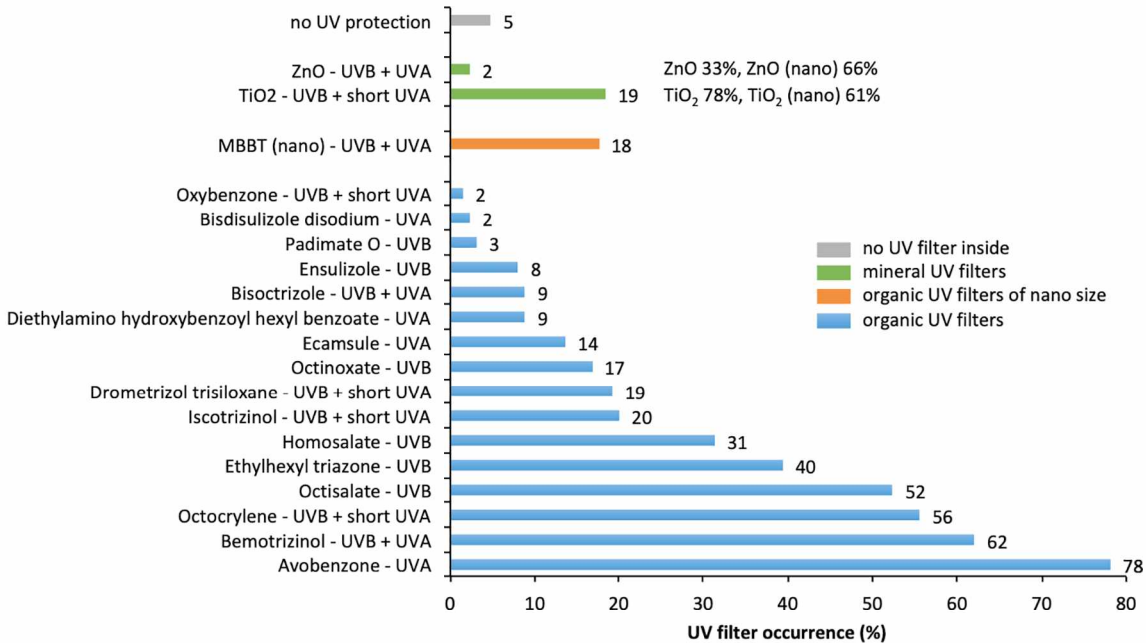


Figure 8



## Do you think that suncare products can impact the quality of marine bath water ?

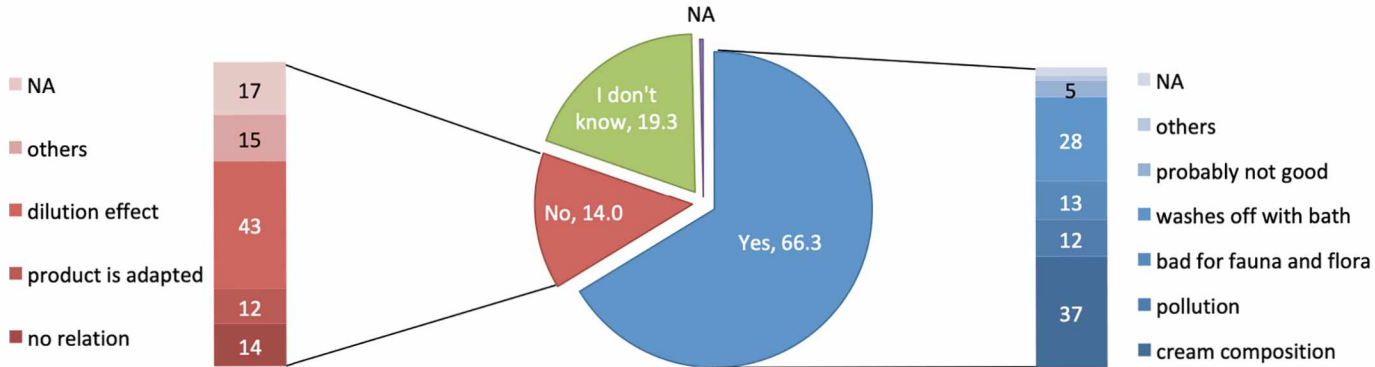
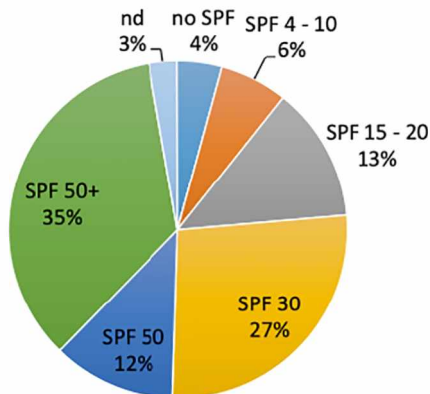
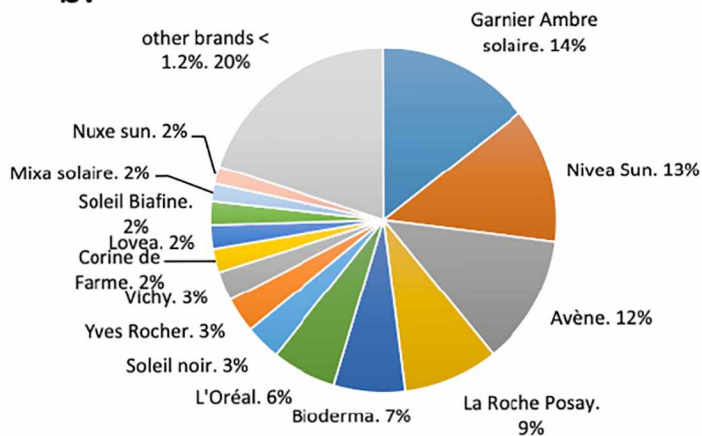


Figure 9

a.



b.



c.

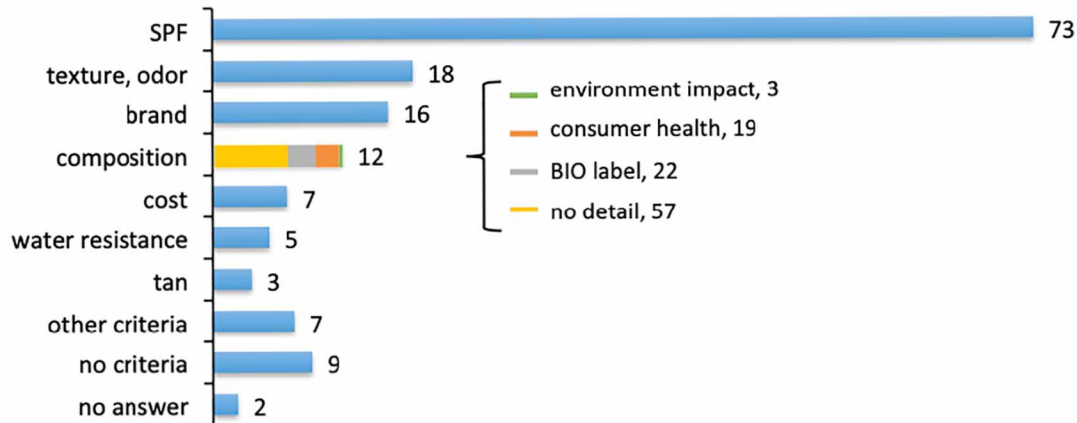


Figure 10