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# Bioaccumulation and toxicological effects of UV-filters on marine species.

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UV-filters are of emerging concern and their toxicity have been demonstrated in many papers. Organic and mineral UV-filters are active ingredients found in sunscreens. Due to the presence of UV-filters in marine waters, studies on these compounds bioaccumulating in organisms have been carried out and this has been complemented by toxicity studies, with reports of detrimental effects to a variety of organisms. This chapter gives an overview of the bioaccumulation and the toxicity of sunscreen UV-filters on marine species. The toxicity of both inorganic and organic UV-filters is summarized as well as their bioaccumulation in marine biota. Ecotoxicological effects of UV-filters suffer from a lack of standardization across studies. We highlighted the difficulties to make comparisons between studies and emphasize a need for harmonization.

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**Abbreviation list of UV-filters**

<b>Abbreviation</b>	<b>INCI</b>	<b>INN/USAN name</b>	<b>CAS #</b>
4-MBC	4-Methylbenzylidene camphor	Enzacamene	36861-47-9
BEMT	bis-Ethylhexyloxyphenol methoxyphenyl triazine	Bemotrizinol	187393-00-6
BM	Butyl methoxydibenzoylmethane	Avobenzone	70356-09-1
BP1	Benzophenone-1	2,4-Dihydroxybenzophenone	131-56-6
BP2	Benzophenone-2	2,2',4,4'- Tetrahydroxybenzophenone	131-57-5
BP3	Benzophenone-3	Oxybenzone	131-57-7
BP4	Benzophenone-4	Sulisobenzone	4065-45-6
BP6	Benzophenone-6	2,2'-Dihydroxy-4,4'- dimethoxybenzophenone	131-54-4
BP8	Benzophenone-8	Dioxybenzone	131-53-3
DHHB	Diethylamino hydroxybenzoyl hexyl benzoate	-	302776-68-7
DT	Drometrizole trisiloxane	-	155633-54-8
EMC	Ethylhexyl methoxycinnamate	Octinoxate	5466-77-3
ES	Ethylhexyl salicylate	Octisalate	118-60-5
ET	Ethylhexyl triazone	Ethylhexyl triazone	88122-99-0
HS	Homosolate	-	118-56-9
IMC	Isoamyl <i>p</i> -methoxycinnamate	Amiloxate	71617-10-2
MBBT	Methylene bis-benzotriazolyl tetramethylbutylphenol	Bisotrizole	103597-45-1
OC	Octocrylene	Octocrilene	6197-30-4
OD-PABA	Ethylhexyl dimethyl PABA	Octyldimethyl PABA, Padimate O	21245-02-3
PABA	PABA	Aminobenzoic acid	150-13-0
TDSA	Terephthalidene dicamphor sulfonic acid	Ecamsule	90457-82-2

# 1. Introduction

UV-filters are recent anthropogenic pollutants encountered in the marine environment. Several studies have reported their occurrence in diverse ecosystems (Rainieri *et al.* 2016). Marine biota has not dealt with these compounds in their evolutionary life history. Therefore, questions have been raised as to how aquatic organisms are coping with emerging contaminants such as sunscreens. In the last decade, research has been carried out and received some attention. Benzophenone-3 (BP3) and ethylhexyl methoxycinnamate (EMC), also known as oxybenzone and octinoxate, respectively, have been banned in Hawaii and Key West, Florida because of their detrimental effects to coral (Downs *et al.* 2016) among other marine organisms (Paredes *et al.* 2014). The Republic of Palau and U.S Virgin Islands have also banned these two UV-filters, but they also included octocrylene (OC).

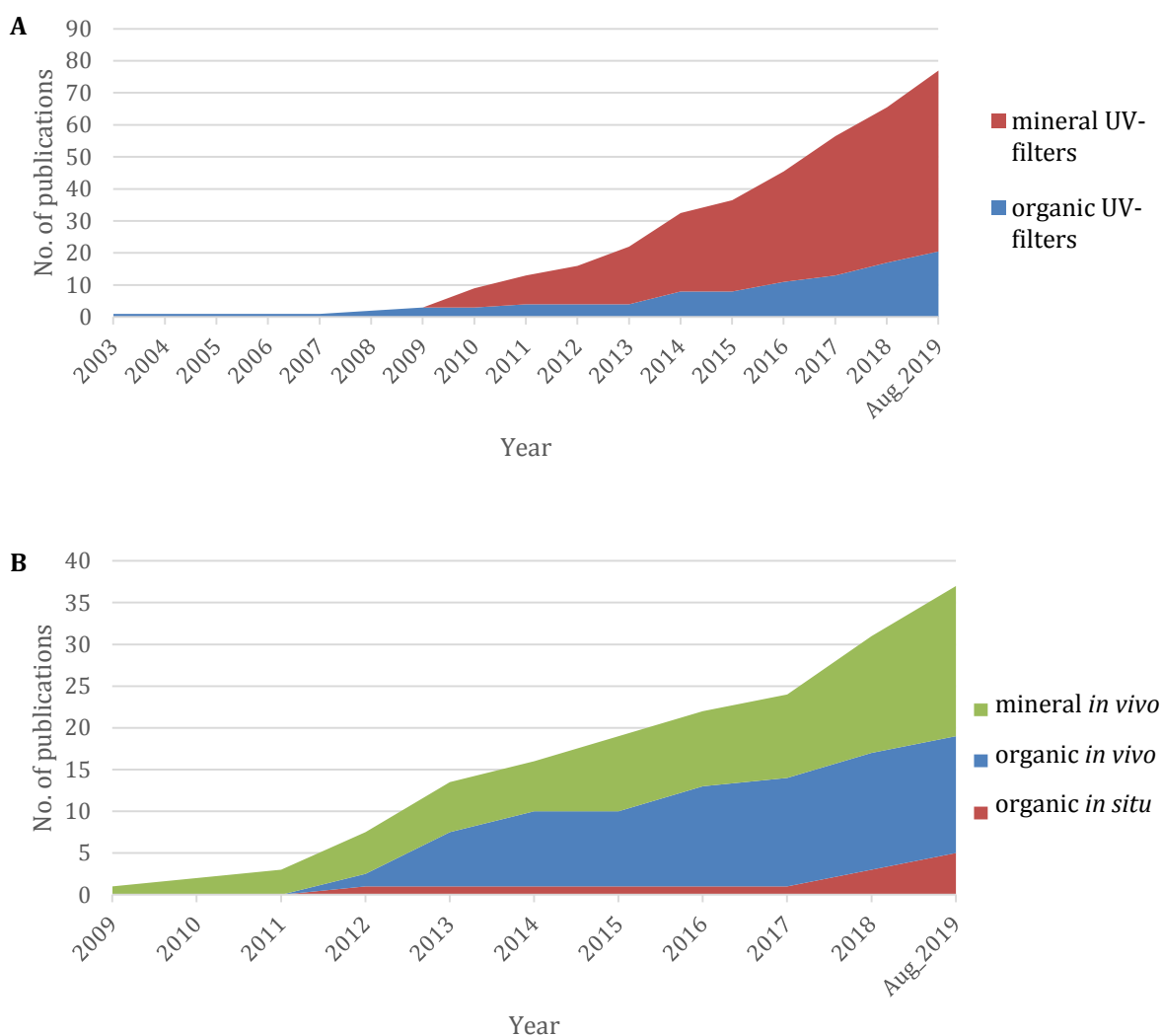
Species richness is higher in the shallow seas compared to below 100m (Costello and Chaudhary, 2017) and species in the coastal waters are subjected to anthropogenic impacts to a greater degree. Coastal waters allow for different ecosystems that support a wide variety of biodiversity. The shallow waters allow for thriving flora which provide shelter for different species and nursery grounds for juveniles, allowing fish populations to increase in yield. This natural wealth helps the economy that being tourism and fisheries, that are prolific sectors important for the socio-economic health. For these reasons, it is essential to explore the impact of emerging contaminants on marine biota.

This chapter will review the toxicity of both organic and inorganic UV-filters and their bioaccumulation on marine species. Critical analysis and scientific trends are presented as well as the main challenges and needs related to ecotoxicological studies.

## 2. Current trends of UV-filters

The first paper to report on the toxicity of sunscreen agents with reference to marine life was Danovaro and Corinaldesi in 2003. They observed an increase in viral abundance with the addition of sunscreen that altered the ratio between the viral phage and the marine bacterial host. No papers were published following this in terms of sunscreens until Danovaro and colleagues published another paper in 2008 showing that changes in the viral:bacterial ratio due to the presence of sunscreen, induced coral bleaching. The authors included three brands of sunscreen and tested single molecules to target the cause.

Studies on mineral UV-filters, especially nanoparticles, are heavily weighted in terms of effort compared to the organic UV-filters. As mineral UV-filters can be found in a variety of manufactured products, some researchers did not focus on sunscreen components *per se*. Although the toxicity of ZnO nanoparticles dedicated for industrial or cosmetic use has been shown to be similar (Spisni *et al.* 2016). Therefore, any studies found with ZnO or TiO<sub>2</sub> mineral UV-filters were included in this review. Nanoparticle research for mineral UV-filters began to gain traction in 2011, whilst organic UV-filters were just being explored (Figure 1).



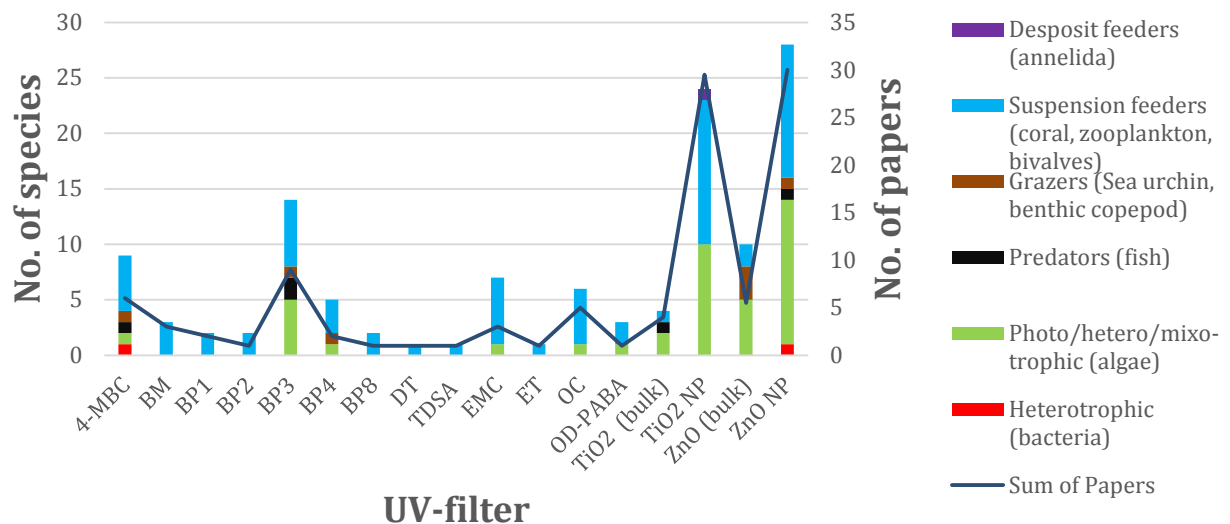
**Figure 1.** Cumulative number of publications for UV-filter, sunscreen and marine biota related searches from Google Scholar, Web of Science and NCBI databases. Toxicity of UV-filters (A) and bioaccumulation (B) in marine biota.

There is an unbalance regarding the choice of UV-filter that has been examined in the literature. There are 60 UV-filters that are permitted in sunscreens worldwide (Pirota, 2016) yet only 15 appear in literature regarding the toxicity on marine biota. Furthermore, only nine of them appear in more than one paper with BP3 and nanoparticles dominating (Figure 2). Homosalate (HS) is not examined as single molecule, yet it is commonly found in sunscreens.

The same pattern is also observed for *in vivo* bioaccumulation studies with more studies performed on mineral nanoparticles and an underrepresented range of organic UV-filters. However, mineral UV-filters have not been targeted for *in situ* bioaccumulation studies, which calls attention to a gap in the research. Perhaps this is because minerals such as zinc oxide (ZnO) and titanium dioxide (TiO<sub>2</sub>) are naturally occurring which presents a difficulty to distinguish between natural and manufactured occurring levels (Montaño *et al.* 2014). The analytical methods for these minerals require more effort to identify and quantify.

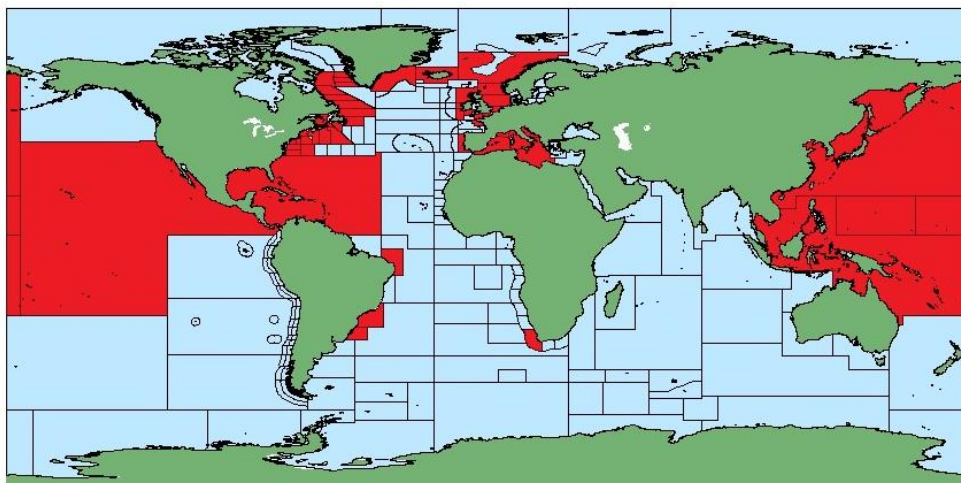
This review shows that there is a preference on the model, that being algae and coral. Marine fish have not been well represented in the toxicity studies except in recent papers that saw the use of clownfish (Barone *et al.* 2019), gilt-head bream (Ziarrusta *et al.* 2018), sole flatfish (Araújo *et al.* 2018) and medaka (Cong *et al.* 2017) as models (Figure 2). The impacts on marine bacteria are often overlooked yet they are a major contributor to the oceans total biomass (Bar-On *et al.* 2018). They are

also key players in nutrient cycling and important symbionts to larger species. Zooplankton were examined by a few papers but mostly using the brine shrimp as a model (Ates *et al.* 2013; Bhuvaneshwari *et al.* 2018; Clemente *et al.* 2014; Li *et al.* 2017; Lu *et al.* 2018; Miglietta *et al.* 2011). These pelagic suspension feeders are also exposed to contaminants as they sieve through the water column collecting food. As the seafloor is the “sink” for contaminants, it is surprising that only two papers to date examined the effects of contaminated sediments for UV-filters; ZnO on the ragworm, the peppery furrow shell (Buffet *et al.* 2012) and TiO<sub>2</sub> on the lugworm (Galloway *et al.* 2010).



**Figure 2.** Number of marine species and their feeding mode for each UV-filter examined to date. Abbreviations: 4-MBC, 4-Methylbenzylidene camphor; BM, Butyl methoxydibenzoylmethane; BP1, Benzophenone-1; BP2, Benzophenone-2; BP3, Benzophenone-3; BP4, Benzophenone-4; BP8, Benzophenone-8; DT, Drometrizole trisiloxane; TDSA, Terephtalidene dicamphor sulfonic acid ; EMC, Ethylhexyl methoxycinnamate; ET, Ethylhexyl triazone; OC, Octocrylene; OD-PABA, Ethylhexyl dimethyl PABA.

There have been 350 published papers on the ingestion of plastics to marine fauna in 2015 and it continues to escalate (Provencher *et al.* 2017). Comparatively, there have only been 20 published papers for *in situ* bioaccumulation of organic UV-filters to date. This may highlight that there is a greater concern regarding marine plastics but also that we are at the beginning stages of understanding sunscreens as an emerging pollutant. UV-filter’s presence in marine biota has been detected across the globe but there are still uncertainties for many waters (Figure 3).



**Figure 3.** An overview of FAO fishing areas where UV-filter bioaccumulation has been reported to date. Red sections indicate the fishing areas with marine biota containing UV-filter pollutants.

The next step is to address if UV-filters are true persistent organic pollutants. It was only after the long-term use of DDT (dichlorodiphenyltrichloroethane) that the true environmental impacts were detected, and a worldwide ban was actioned by the Stockholm Convention on persistent organic pollutants. To understand the future impacts of UV-filters, the following should be explored more thoroughly; biomonitoring focused on marine megafauna, *in vivo* testing, examining the contaminations passed on through generations and chronic toxicity assessments.

### 3. Toxicity of sunscreens

Common endpoints that were used to measure the toxicity effects of UV-filters were observed in this literature review. Oxidative stress causes free radicals resulting in damage to the cell by reactive oxygen species (ROS) and other reactive intermediates. This was mostly measured by assessing antioxidative enzymes, which is a common endpoint for mussels (Barmo *et al.* 2013; Ciacci *et al.* 2012; D'Agata *et al.* 2014; Giraldo *et al.* 2017; Hanna *et al.* 2013; Huang *et al.* 2016; Libralato *et al.* 2013; Wang *et al.* 2014) and algae (M. Bhuvaneshwari *et al.* 2018; Giraldo *et al.* 2017; Miller *et al.* 2012; Paredes *et al.* 2014; M. Sendra *et al.* 2017; Seoane *et al.* 2017; Thiagarajan *et al.* 2019; Wong and Leung, 2014; Xia *et al.* 2015). Growth inhibition was also a repetitive parameter for mineral UV-filters that was applied to different organisms such as bacteria, amphipods, bivalves but mostly for algae (Deng *et al.* 2017; Fabrega *et al.* 2012; Li *et al.* 2017; Manzo *et al.* 2013(a); Sendra *et al.* 2017; Wang *et al.* 2016; Xia *et al.* 2015). Crustaceans saw mortality as a common endpoint (Paredes *et al.* 2014; Wong *et al.* 2010; Wong and Leung, 2013) and corals saw endpoints such as bleaching, polyp retraction, mortality (Downs *et al.* 2016, 2014; He *et al.* 2019) and metabolic alterations (Stien *et al.* 2019). DNA damage was studied on bivalves, lugworm, coral and algae (D'Agata *et al.* 2014; Downs *et al.* 2016; Galloway *et al.* 2010; Jovanović and Guzmán, 2014; Wang *et al.* 2017). Mortality, behavioral and morphological abnormalities were the most studied parameters for Fish (Araújo *et al.* 2018; Barone *et al.* 2019; Cong *et al.* 2017). Table 1 summarizes the extent of toxicity for UV-filters across different studies.

#### 3.1 4-Methylbenzilydene camphor

4-Methylbenzilydene camphor (4-MBC) is one of the most studied UV-filters in marine environments. Coral bleaching effects have been demonstrated with exposure concentrations of 33 µg/L (Danovaro *et al.* 2008), as well as developmental disruptive effects in sea urchin larvae at 0.8 µg/L (Torres *et al.* 2016), an environmentally relevant concentration (Sánchez-Rodríguez *et al.* 2015; Román *et al.* 2011).



Its toxicity was also shown for fish (Araújo *et al.* 2018) and copepod (Chen *et al.* 2018) at 25 and 10 µg/L, respectively.

### 3.2 Benzophenones

Benzophenone derivatives are by far the most studied compounds, with the greatest diversity of parameters studied. Among the compounds investigated: BP1, BP2, BP3, BP4 and BP8; BP4 is the only one that showed no toxicity with different organisms (algae, mussels, sea urchins, crustaceans, fish and coral) (Paredes *et al.* 2015; He *et al.* 2019; Barone *et al.* 2019). BP3, which is commonly used in cosmetics, induced bleaching in coral species such as *Seriatopora caliendrum*, *Stylophora pistillata* and *Acropora* sp. (Danovaro *et al.* 2008; He *et al.* 2019; Downs *et al.* 2016) from 33 µg/L. Downs *et al.* (2016) reported increased bleaching in the presence of light, as well as DNA damage from 2.8 µg/L and mortality from 22.8 µg/L; concentrations are the same range as those reported in literature (Kim and Choi, 2014) but concentrations have been cited to be as high as 1.4 mg/L in the U.S. Virgin Islands (Downs *et al.* 2016).

### 3.3 Ethylhexyl methoxy cinnamate

EMC has been studied for algae, mussels, sea urchins, and crustacea by Paredes *et al.* (2014) with the lowest EC<sub>50</sub> concentration at 74.73 µg/L for algae. EMC showed a species-dependent effect on corals species. While it did not increase mortality in *P. damicornis* from 0.1 to 1000 µg/L, EMC increased the mortality of *S. caliendrum* and induced bleaching at 1 mg/L (He *et al.* 2019(a)). Polyp retraction were observed from 10 µg/L for the same species (He *et al.* 2019). 91% of bleaching was observed at concentrations of 33 µg/L and 50 µg/L for *Acropora* sp. Comparatively, occurrence study highlighted its presence up to 4 µg/L (Tsui *et al.* 2014).

### 3.4 Octocrylene

OC was detected in sea water at concentrations up to 7 µg/L (Tsui *et al.* 2014; Langford *et al.* 2015). Several studies have highlighted detrimental effects against several marine species from 40 µg/L. Its effect on coral species appeared to be nuanced (Danovaro *et al.* 2008; Fel *et al.* 2018). However, He *et al.* (2019) demonstrated that it induced polyp's retraction at 1 mg/L and increased mortality in *P. damicornis* if combined with EMC. A recent metabolomic study showed that *P. damicornis* accumulated OC in fatty acid-conjugated forms from 5 µg/L and is associated with mitochondrial dysfunctions at 50 µg/L, whereas polyps were only retracted at 300 µg/L (Stien *et al.* 2019). This study showed that usual ecotoxicological parameters such as mortality or polyps state may result in an underestimation of a compound toxicity.

### 3.5 Other organic UV-filters

Some chemicals, such as TDSA, BM, DT, ES and ET have undergone very little investigation on marine organisms, with only one or two studies on coral species (Danovaro *et al.* 2008; Fel *et al.* 2018) with the papers concluding no major threat to cause harm for the single molecule toxicity. The toxicity of approximately 45 globally authorized sunscreen UV-filters have not been assessed on marine biota. Therefore, it is difficult to draw conclusions about their effects and the risk associated with their use.

### 3.6 Inorganic UV-filters

Mineral UV-filters have been broadly advertised as eco-friendly in the public market. Nevertheless, several recent studies on ZnO and TiO<sub>2</sub> reported their toxicity on marine biota (see Table 2 for full reference list). Corinaldesi and colleagues (2018) presented severe bleaching to stoney coral due to exposure of zinc nanoparticles at 6.3 mg/L over 48 hours. Alterations in the surrounding microbial abundance for the stoney coral were also presented after exposure to mineral UV-filters at the same concentration. Digestive and immune systems of bivalves were affected by mineral UV-filter (Buffet *et*

*al.* 2012; D'agata *et al.* 2014; Canesi *et al.* 2010(a); Xia *et al.* 2017). Because of their feeding strategy, these organisms are subjected to nanoparticle ingestion.

Mineral UV-filters can display different sizes and shapes. These characteristics are potentially influenced by abiotic variables that can affect their toxicity. Wong and Leung (2013) proposed that temperature sequentially affects the toxicity by influencing the size of ZnO aggregates. Miao *et al.* (2010) demonstrated that the dissolution rate, influenced by the pH and the nanoparticles shape, altered the toxicity of ZnO on the algae *Thalassiosira pseudonana*. D'Agata and colleagues (2014) observed that bulk TiO<sub>2</sub> was more toxic causing DNA damage compared to TiO<sub>2</sub> nanoparticles in mussels, even if the nanoparticles are more likely to be accumulated. Hazeem *et al.* (2016) looked at chronic exposures of ZnO and only showed growth inhibition at the early growth stages of *Picochlorum* sp. due to aggregation and sedimentation occurring at the later stages. Conversely, some authors highlighted that smaller particles are more toxic for example Xia *et al.* (2015) demonstrated a growth inhibition at lower concentrations for *Nitzschia closterium* algae with EC<sub>50</sub> values of 88.8 mg/L for 21 nm TiO<sub>2</sub> nanoparticles but 118.8 mg/L for 60 nm particle size. The shape is another characteristic that should be considered as Peng *et al.* (2011) demonstrated that rod shaped ZnO particles were more toxic than sphere shaped particles to marine algae when also measuring the growth inhibition.

Two crystalline forms are commonly used in mineral sunscreens, rutile and anatase. Doyle *et al.* (2018) demonstrated minimal differences between the bulk anatase and bulk rutile crystalline forms for TiO<sub>2</sub>, when looking at the hemocyte responses from the Atlantic oyster, *Crassostrea virginica*. This feature is yet to be explored for nanoparticles. As described above, the characteristics of mineral filters can strongly influence ecotoxicology studies. The literature revealed a lack of detail on the characteristics of the filters studied. Some authors provided information on the anatase/rutile percentage, diameter and shape of the particles (Miller *et al.* 2010), while others did not specify on the nanoparticle nature (Barmo *et al.* 2013; Guan *et al.* 2018). Ciacci and colleagues (2012) confirmed the size distribution using transmission electron microscope and noted that they were different to the declared 42nm size at purchase. These results highlight the difficulty of comparing toxicity studies for mineral filters.

### 3.7 Combination study

Emollients and emulsifiers make up typically 20% of the sunscreen ingredients as with other components of sunscreen such as sensory enhancers, boosting ingredients and rheology modifiers (Osterwalder *et al.* 2014). The toxicity of these compounds has not been thoroughly explored. Danovaro and Corinaldesi (2003) indicated that sunscreens can alter carbon, nitrogen and phosphate biogeochemical cycling in the Ocean. Further studies demonstrated that commercial sunscreen increased the nutrient content in seawater (Rodríguez-Romero *et al.* 2019; Tovar-Sánchez *et al.* 2013). Danovaro *et al.* (2008) showed that excipients like butylparaben preservatives which are commonly found in sunscreens and other cosmetic products caused coral bleaching by encouraging viruses and resulted in a 70-90% zooxanthellae release. Furthermore, sunscreens have also been shown to be a source of hydrogen peroxide production (Sánchez-Quiles and Tovar-Sánchez, 2014; Sendra *et al.* 2017). Toxicities can vary depending on the sunscreen formulation. For example, spray sunscreens have a greater bioavailability according to Tovar-Sánchez and colleagues (2013) as they demonstrated a higher toxicity in growth inhibition to phytoplankton compared to a cream. Only a few papers investigated the toxicity of a complete formulation instead of a single UV-filter (Barone *et al.* 2019; Danovaro *et al.* 2008; Danovaro and Corinaldesi, 2003; Mccoshum *et al.* 2016; Fastelli and Renzi, 2019). Toxicities were seen to be greater in the formulation compared to a single molecule. For example, Sendra *et al.* (2017) showed that commercial sunscreens with TiO<sub>2</sub> nanoparticles were more toxic to micro algae compared TiO<sub>2</sub> nanoparticles alone due to greater H<sub>2</sub>O<sub>2</sub> production. ZnO and TiO<sub>2</sub> nanoparticles have been shown to negatively impact marine algae growth to a greater extent when they are combined (Hazeem *et al.* 2016).

Laboratory assays are performed in controlled environments and not all environmental variables can be considered. Variation in abiotic parameters such as temperature, salinity, nutrient content, organic matter and UV radiation strongly shape the fate of chemicals in marine biota. For example, oxygen content (Wang *et al.* 2014) and UV radiations (Miller *et al.* 2012; Sendra *et al.* 2017(a)) can influence the toxicity of TiO<sub>2</sub>. As UV-filter's transformation products could pose a greater risk than the native compound, a better understanding of their release and formation is needed. Literature data indicate that the following compounds remain stable under direct light exposure in experimental conditions: BP3 (Liu *et al.* 2011; Rodil *et al.* 2009), OC (Herzog *et al.* 2009; Rodil *et al.* 2009), bis-ethylhexyloxyphenol methoxyphenyl triazine (BEMT) (Herzog *et al.* 2009) and 4-MBC (Rodil *et al.* 2009). On the other hand, EMC (Jentzsch *et al.* 2016; MacManus-Spencer *et al.* 2011), Isoamyl *p*-methoxycinnamate (IMC) (MacManus-Spencer *et al.* 2011), ethylhexyl dimethyl PABA (OD-PABA) (Rodil *et al.* 2009), Ethylhexyl triazone (ET) (Damiani *et al.* 2010; Herzog *et al.* 2009), butyl methoxydibenzoylmethane (BM) (Damiani *et al.* 2010; Herzog *et al.* 2009) demonstrate a lower photostability. In addition, the impact of organic matter on the fate of UV-filters was demonstrated, acting as photosensitizer through its triplet-excited states. Hence, we can distinguish between a direct and indirect photodegradation process (Li *et al.* 2016). The involvement of biotic factors on the fate of UV-filters in marine environments is poorly documented. Beel *et al.* (2013) highlighted the biodegradation of BP4 in contact with activated sludge of municipal wastewater treatment plant. Badia-Fabregat *et al.* (2012) have demonstrated fungus mediated degradation of 4-MBC. Microcosms experiments showed the biodegradation of BP3 (Liu *et al.* 2012; Volpe *et al.* 2017). Volpe *et al.* (2017) demonstrated that 4-MBC was stable whereas OD-PABA was biodegraded in marine sediments.

**Table 1.** Summary of organic UV-filter toxicity studies.

UV-filter	Species Model [Common name]	Endpoint	Tested conc. range	Toxic Effects [Conc with toxic effect]	Reference
4-Methylbenzilidene camphor (4-MBC)	<i>Acropora pulchra</i> [Staghorn coral]	bleaching	50 µ/L	95% bleaching rate over 62 hours [50 µg/L]	Danovaro <i>et al.</i> 2008
	<i>Acropora sp.</i> [Stoney coral]	bleaching	33 µ/L	63% bleaching rate over 48 hours [33 µg/L]	Danovaro <i>et al.</i> 2008
	<i>Isochrysis galbana</i> [Unicellular algae]	growth rate	N/A	EC <sub>50</sub> = 171.45 µg/L over 72h	Paredes <i>et al.</i> 2014
	<i>Mytilus galloprovincialis</i> [Mediterranean Mussel]	morphologies		changes in D-larvae, EC <sub>50</sub> = 587.17 µg/L over 48h	
	<i>Paracentrotus lividus</i> [Purple sea urchin]	larval length	0.32–5000 µg/L	5% decrease in larval length [2 µg/L]	Torres <i>et al.</i> 2016
		morphological abnormalities	0.32–5000 µg/L	20% of deformed larvae [0.8 µg/L]	
		larval growth	N/A	EC <sub>50</sub> = 854 µg/L over 48h	Paredes <i>et al.</i> 2014
	<i>Senegalese sole</i> [Sole flatfish]	enzymatic activity	25–216 µg/L	increase of AChE activity [85 µg/L]	Araújo <i>et al.</i> 2018
		enzymatic activity		decrease in LDH activity [25 µg/L]	
		swimming behavior	68–360 µg/L	decrease in swimming distance [360 µg/L]	
		mortality	235–935 µg/L	40 +/- 18% mortality at 48hpf; 85+/-5% mortality at 72hpf [935 µg/L]	
		growth	68–360 µg/L	decreased length [360 µg/L]	
		hatching success	235–935 µg/L	decrease of 40% in hatching rates [447 µg/L]	
	<i>Siriella armata</i> [Crustacea]	mortality	N/A	EC <sub>50</sub> = 192.63 µg/L over 96h	Paredes <i>et al.</i> 2014
<i>Tigriopus japonicus</i> [Copepod]	mortality	10–5000 µg/L	100% mortality over 72h [1000 µg/L]	Chen <i>et al.</i> 2018	
	survival	0.5–10 µg/L	52% survival rate [10 µg/L]		
Benzophenone–1 (BP1)	<i>Acartia tonsa</i> [Copepod]	DT <sub>1/2</sub> values	0.051–2 mg/L	89-296 (h) (time elapsed until 50% of the organisms had developed into copepodites) [concentrations were pooled]	Kusk <i>et al.</i> 2011
		hatching success		70-95% hatching success (depending on environment)	
		survival success		64-92% survival success (depending on environment)	
		mortality	0.5–10 mg/L	LC <sub>50</sub> = 2.6 mg/L at 24 and 48h	
		larval development		EC <sub>50</sub> = 1.5 mg/L with 15% salinity EC <sub>50</sub> = 0.59 mg/L with 25% salinity	
	<i>Pocillopora damicornis</i> [Cauliflower coral]	mortality	0.1–1000 µg/L	mortality on the nubbin = 100% on day 7 [1000 µg/L]	He <i>et al.</i> 2019
		bleaching rate		33% bleaching on the nubbin [1000 µg/L]	
	<i>Seriatopora caliendrum</i> [Stoney coral]	mortality		no mortality on the larvae	
		mortality		100% mortality on the nubbin [1000 µg/L]	
		bleaching rate		57% white, 25% pale brown for the larvae [1000 µg/L]	
bleaching rate			33% bleaching for the nubbin [1000 µg/L]		
Benzophenone–2 (BP2)	<i>Stylophora pistillata</i> [Hood coral]	chlorophyll fluorescence	2.46 µg–246 mg/L	photoinduced toxicity – coral bleaching [2.46 µg/L]	Downs <i>et al.</i> 2014
		DNA abasic lesions		increase of DNA lesions (greater toxicity if exposed to light) [24.6 µg/L]	

UV-filter	Species Model [Common name]	Endpoint	Tested conc. range	Toxic Effects [Conc with toxic effect]	Reference
		cellular morphology		morphological perturbation and an increase in calicoblast cells from 2.46 µg/L [2.46 µg/L]	
<b>Benzophenone-3 (BP3)</b>	<b><i>Acropora pulchra</i></b> [Staghorn coral]	bleaching	50 µl/L	93% bleaching rate over 96 hours [50 µl/L]	Danovaro <i>et al.</i> 2008
	<b><i>Acropora sp.</i></b> [Stoney coral]	bleaching	33 µl/L	86% bleaching rate over 48 hours [33 µl/L]	Danovaro <i>et al.</i> 2008
	<b><i>Amphiprion ocellaris</i></b> [Clown fish]	mortality	1–100 mg/L	25% of mortality (entire sunscreen was tested) [100 mg/L]	Barone <i>et al.</i> 2019
		feeding behavior		20% of fishes with abnormal behavior at 30 mg/L, 100% at 100 mg/L (entire sunsreen was tested) [30 mg/L]	
		swimming behavior		100% of fishes with abnormal movements (entire sunsreen was tested) [100 mg/L]	
	<b><i>Isochrysis galbana</i></b> [Unicellular algae]	growth rate	N/A	EC <sub>50</sub> = 13.87 µg/L over 72h	Paredes <i>et al.</i> 2014
	<b><i>Mytilus galloprovincialis</i></b> [Mediterranean Mussel]	percentage of normal D- larvae		EC <sub>50</sub> = 3472.59 µg/L over 48h	
	<b><i>Paracentrotus lividus</i></b> [Purple sea urchin]	larval growth		EC <sub>50</sub> = 3280 µg/L over 48h	
	<b><i>Seriatopora caliendrum</i></b> [Stoney coral]	mortality (larvae)	0.1–1000 µg/L	mortality 5% on day 7 [1000 µg/L]	He <i>et al.</i> 2019
		mortality (nubbin)		mortality 100% on day 6/7 [1000 µg/L]	
		bleaching rate (larvae)		95% white, 5% pale brown [1000 µg/L]	
		bleaching rate (nubbin)		50% bleaching [1000 µg/L]	
	<b><i>Siriella armata</i></b> [Crustacea]	mortality	N/A	EC <sub>50</sub> = 710.76 µg/L over 96h	Paredes <i>et al.</i> 2014
	<b><i>Skeletonema pseudocostatum</i></b> [Algae]	growth inhibition		EC <sub>50</sub> = 1.1 µM over 72h	Petersen <i>et al.</i> 2014
	<b><i>Sparus aurata</i></b> [Gilt-head bream]	metabolic alterations	50 mg/L	alterations in lipid metabolism, altered metabolites in liver (phenylalanine metabolism and the tyrosine-related metabolites) exposure over 14 days	Ziarrusta <i>et al.</i> 2018(a)
		physical changes		no changes observed in weight or length over 14 days	
mortality			no mortality over 14 days of exposure		
<b><i>Stylophora pistillata</i></b> [Hood coral]	chl fluorescence	2.28 µg–228 mg/L	photoinduced toxicity – coral bleaching [2.28 µg/L]	Downs <i>et al.</i> 2016	
	DNA abasic lesions		increase of DNA lesions (greater toxicity if exposed to light) [22.8 µg/L]		
	cellular morphology mortality		altered morphology and motility of planulae [22.8 µg/L] increased mortality (greater toxicity if exposed to light) [22.8 µg/L]		
<b><i>Tetraselmis suecica</i></b> [Unicellular algae]	cell size	5–40pg/cells (250–2000 µg/L)	increase of cell size [40 pg/cells (2000 µg/L)]	Seoane <i>et al.</i> 2017	
	cell autofluorescence growth		increase of autofluorescence [10 pg/cells (500 µg/L)] increased growth rate [10 pg/cells (500 µg/L)]		
	membrane integrity		membrane depolarization [10 pg/cells (500 µg/L)]		

UV-filter	Species Model [Common name]	Endpoint	Tested conc. range	Toxic Effects [Conc with toxic effect]	Reference
		metabolic activity		increase of metabolic activity [20 pg/cells (1000 µg/L)]	
<b>Benzophenone-4 (BP4)</b>	<i>Isochrysis galbana</i> [Unicellular algae]	growth rate	N/A	no effect over 72h	Paredes <i>et al.</i> 2014
	<i>Mytilus galloprovincialis</i> [Mediterranean Mussel]	percentage of normal D-larvae		no effect over 48h	
	<i>Paracentrotus lividus</i> [Purple sea urchin]	larval growth		no effect over 48h	
	<i>Seriatopora caliendrum</i> [Stoney coral]	mortality bleaching rate	0.1–1000 µg/L	no effect on the larvae no effect on the larvae	He <i>et al.</i> 2019
	<i>Siriella armata</i> [Crustacea]	mortality	N/A	no effect over 96h	Paredes <i>et al.</i> 2014
<b>Benzophenone-8 (BP8)</b>	<i>Pocillopora damicornis</i> [Cauliflower coral]	mortality	0.1–1000 µg/L	mortality 100% on day 3 (nubbin) [1000 µg/L]	He <i>et al.</i> 2019
		bleaching rate	0.1–1000 µg/L	100% bleaching (nubbin) [1000 µg/L]	
	<i>Seriatopora caliendrum</i> [Stoney coral]	mortality	0.1–1000 µg/L	mortality 30% on day 9 (larvae) [1000 µg/L]	
		mortality	0.1–1000 µg/L	mortality 100% on day 2 (nubbin) [1000 µg/L]	
		bleaching rate	0.1–1000 µg/L	75% white, 25% pale brown (larvae) [1000 µg/L]	
		bleaching rate	0.1–1000 µg/L	100% bleaching (nubbin) [1000 µg/L]	
<b>Butyl Methoxydibenzoylmethane (BM)</b>	<i>Acropora sp.</i> [Stoney coral]	bleaching	33 µl/L	no bleaching. Sunscreen formulations were tested and demonstrated different effects (see article)	Danovaro <i>et al.</i> 2008
	<i>Stylophora pistillata</i> [Hood coral]	photosynthetic efficiency	10–5000 µg/L	photosynthetic efficiency reduced to 0 on day 14 [5000 µg/L]	Fel <i>et al.</i> 2018
<b>Drometrizole Trisiloxane (DT)</b>	<i>Stylophora pistillata</i> [Hood coral]	photosynthetic efficiency	10–5000 µg/L	no change photosynthetic efficiency	Fel <i>et al.</i> 2018
<b>Ethylhexyl Dimethyl PABA (OD-PABA)</b>	<i>Isochrysis galbana</i> [Algae]	growth rate	1– 200 µg/L	10% decrease growth rate [100 µg/L] (EC <sub>50</sub> = 59 µg/L) over 72h	Giraldo <i>et al.</i> 2017
	<i>Mytilus galloprovincialis</i> [Mediterranean mussel]	% of normal larvae	1– 200 µg/L	10% reduction of normal larvae [100 µg/L] (EC <sub>50</sub> = 130 µg/L)	
	<i>Paracetrotus lividus</i> [Purple sea urchin]	early larval growth	1– 200 µg/L	40% inhibition [200 µg/L] (EC <sub>50</sub> = 279 µg/L)	
<b>Ethylhexyl methoxy cinnamate (EMC)</b>	<i>Acropora pulchra</i> [Staghorn coral]	bleaching	50 µl/L	91% bleaching rate over 96 hours [50 µl/L]	Danovaro <i>et al.</i> 2008
	<i>Acropora sp.</i> [Stoney coral]	bleaching	33 µl/L	91% bleaching rate over 24 hours [33 µl/L]	Danovaro <i>et al.</i> 2008
	<i>Isochrysis galbana</i> [Unicellular algae]	growth rate	N/A	EC <sub>50</sub> = 74.73 µg/L over 72h	Paredes <i>et al.</i> 2014
	<i>Mytilus galloprovincialis</i> [Mediterranean mussel]	percentage of normal D-larvae	N/A	EC <sub>50</sub> = 3118.19 µg/L over 48h	
	<i>Paracentrotus lividus</i> [Purple sea urchin]	larval growth	N/A	EC <sub>50</sub> = 284 µg/L over 48h	
	<i>Pocillopora damicornis</i> [Cauliflower coral]	mortality	0.1–1000 µg/L	no mortality with single molecule. 100% mortality with sunscreen products.	He <i>et al.</i> 2019(a)
	<i>Paracentrotus lividus</i> [Purple sea urchin]	polyp retraction		retracted polyp [1000 µg/L]	

UV-filter	Species Model [Common name]	Endpoint	Tested conc. range	Toxic Effects [Conc with toxic effect]	Reference
		visual bleaching		no bleaching. Bleaching occurred with combined UVFs and entire sunscreens	
		zooxanthellae density		no effect	
	<b>Seriatopora caliendrum</b> [Stoney coral]	polyp retraction	0.1–1000 µg/L	retracted polyp [10 µg/L]	
		visual bleaching		83.3% of bleached coral [1000 µg/L]	
		zooxanthellae density		No significant decrease [1000 µg/L]	
		mortality		33.3% mortality [1000 µg/L]	
	<b>Siriella armata</b> [Crustacea]	mortality		EC <sub>50</sub> = 199.43 µg/L over 96h	Paredes <i>et al.</i> 2014
<b>Ethylhexyl salicylate (ES)</b>	<b>Acropora sp.</b> [Stoney coral]	bleaching	33 µg/L	no bleaching. Sunscreen formulations were tested and demonstrated different effects (see article)	Danovaro <i>et al.</i> 2008
<b>Ethylhexyl Triazone (ET)</b>	<b>Stylophora pistillata</b> [Hood coral]	photosynthetic efficiency	10–5000 µg/L	increased photosynthetic efficiency	Fel <i>et al.</i> 2018
<b>Octocrylene (OC)</b>	<b>Acropora sp.</b> [Stoney coral]	bleaching	33 µg/L	no bleaching. Sunscreen formulations were tested and demonstrated different effects (see article)	Danovaro <i>et al.</i> 2008
	<b>Isochrysis galbana</b> [Algae]	growth rate	1–300 µg/L	10% decrease growth rate [80 µg/L] (EC <sub>50</sub> ≥ 150 µg/L) over 72h	Giraldo <i>et al.</i> 2017
	<b>Mytilus galloprovincialis</b> [Mediterranean Mussel]	% of normal larvae	1–750 µg/L	10% reduction of normal larvae [40 µg/L] (EC <sub>50</sub> ≥ 650 µg/L)	
	<b>Paracetrotus lividus</b> [Purple sea urchin]	early larval growth	1–750 µg/L	40% inhibition [40 µg/L] (EC <sub>50</sub> = 737 µg/L)	
	<b>Pocillopora damicornis</b> [Cauliflower coral]	polyps state	5–1000 µg/L	most polyps closed [300 µg/L]	Stien <i>et al.</i> 2019
		metabolism	5–1000 µg/L	abnormal fatty acid metabolism increased acylcarnitines [50 µg/L]	
		mortality	0.1–1000 µg/L	no mortality was observed in single molecule exposure. 50% mortality in EMC + OC combined. 100% mortality in sunscreen products.	He <i>et al.</i> 2019(a)
		polyp state		total polyp retraction [1000 µg/L]	
	<b>Seriatopora caliendrum</b> [Stoney coral]	mortality	0.1–1000 µg/L	no mortality was observed in single molecule exposure. 100% mortality in EMC + OC combined. 100% mortality in sunscreen products.	
		polyp state		total polyp retraction [1000 µg/L]	
	<b>Stylophora pistillata</b> [Hood coral]	photosynthetic efficiency	10–5000 µg/L	slight decrease in photosynthetic efficiency [5000 µg/L]	Fel <i>et al.</i> 2018
<b>Terephthalidene dicamphor sulfonic acid (TDSA)</b>	<b>Stylophora pistillata</b> [Hood coral]	photosynthetic efficiency	10–5000 µg/L	no significant results for photosynthetic efficiency	Fel <i>et al.</i> 2018

**Table 2.** Summary of inorganic UV-filter toxicity studies.

UV-filter	Species Model [Common name]	Endpoint	Tested conc. range	Toxic Effects [Conc with toxic effect]	Reference
TiO <sub>2</sub> (nanoparticles)	<i>Acropora</i> sp. [Stoney coral]	abundance of zooxanthellae	6.3 mg/L	increase of zooxanthellae released into seawater from coral. (24 + 48h)	Corinaldesi <i>et al.</i> 2018
		damaged zooxanthellae		increase of damaged zooxanthellae (24 + 48h)	
		bleaching		slight color loss in 6–7% (no visual bleaching)	
		microbial enrichment		reduced number of prokaryotes and viruses in the surrounding seawater of the exposed coral.	
	<i>Alexandrium tamarens</i> [Dinoflagellate]	growth	2–320 mg/L	EC <sub>50</sub> = 85.08 mg/L (4 days) EC <sub>50</sub> = 140.09 mg/L (13 days) recovery observed depending on the exposure conc. (increase in antioxidative enzymes and toxins)	Li <i>et al.</i> 2019
	<i>Amphidinium carterae</i> [Unicellular algae]	enzymatic activity	1–10 mg/L	high H <sub>2</sub> O <sub>2</sub> produced at first 12h, combined with UV. Sunscreen products produced a higher concentration of H <sub>2</sub> O <sub>2</sub> [10 mg/L]	Sendra <i>et al.</i> 2017
		cell density		reduced cell density (combined UV) [1 mg/L]	
	<i>Arenicola marina</i> [Lugworm]	DNA damage	1–3 g/kg	13% DNA damage [2 g/kg]	Galloway <i>et al.</i> 2010
		cytotoxicity effects		reduced lysosomal stability [2 g/kg]	
	<i>Artemia salina</i> [Brine shrimp]	synergistic effect	5, 400 mg/L	nTiO <sub>2</sub> increased the toxicity Cd <sup>2+</sup> by 57.5% [5 mg/L]	Lu <i>et al.</i> 2018
		% mortality	0.1–10 mg/L	increase in mortality (48h) LC <sub>50</sub> = 4.21 mg/L	Bhuvaneshwari <i>et al.</i> 2018
		mobility	1–80 mg/L	EC <sub>50</sub> = 4 mg/L (48h) exposed to UV exposure. 5–80 mg/L (anatase) 1–20 mg/L (anatase/rutile)	Clemente <i>et al.</i> 2014
		growth rate	0.06, 0.6 mg/L	no effect (96h) (anatase and anatase/rutile mix) exposed to UV + visible light.	
<i>Chaetoceros gracilis</i> [Unicellular algae]	enzymatic activity	1–10 mg/L	high H <sub>2</sub> O <sub>2</sub> produced at first 12h, combined with UV. Sunscreen products produced a higher concentration of H <sub>2</sub> O <sub>2</sub> [10 mg/L]	Sendra <i>et al.</i> 2017	
	cell density		reduced cell density (combined UV) [1 mg/L]		
<i>Chlamys farreri</i> [Marine scallop]	oxidative stress	1 mg/L	SOD & CAT responses were triggered in the gill and digestive gland over 14 days [1 mg/L]	Xia <i>et al.</i> 2017	
	enzymatic activity		increased AChE [>14 days]		
	enzymatic activity		histopathological alterations of the gill and digestive gland [>14 days]	Wang <i>et al.</i> 2017	
	LMS	1 mg/L	increased lysosomal membrane destabilization but		



UV-filter	Species Model [Common name]	Endpoint	Tested conc. range	Toxic Effects [Conc with toxic effect]	Reference
				recovered on day 15 [day 14]	
		genotoxicity		tail DNA% increased to 55.56% [day 14]	
	<b>Chlorella sp.</b> [Unicellular algae]	oxidative stress	0.1–10 mg/L	ROS generation and LPO production, EC <sub>50</sub> = 1.6 mg/L (UVA), 5 mg/L (dark conditions)	Thiagarajan <i>et al.</i> 2019
	<b>Crassostrea virginica</b> [Atlantic oyster]	phagocytosis	0.1–1 mg/L	phagocytic activity of hemocytes (dark, anatase) [1 mg/L]	Doyle <i>et al.</i> 2018
phagocytosis			phagocytic activity of hemocytes (light, rutile) [0.1, 0.5 mg/L]		
ROS production			ROS production increased when photoactivated in UV conditions [0.5 mg/L]		
	<b>Dunaliella salina</b> [Unicellular algae]	viability	0.25–16 mg/L	loss in cell viability at 48h LC <sub>50</sub> = 11.35mg/L	Bhuvaneshwari <i>et al.</i> 2018
	<b>Dunaliella tertiolecta</b> [Unicellular algae]	oxidative stress	1–7 mg/L	increase OH <sup>•</sup> production [3 mg/L]	Miller <i>et al.</i> 2012
		genotoxicity	N/A	DNA damage [20 mg/L]	Schiavo <i>et al.</i> 2016
	<b>Haliotis diversicolor supertexta</b> [Marine abalone]	enzymatic activity	1–10 mg/L	SOD significantly increased [1 mg/L]	Zhu <i>et al.</i> 2011
		non–enzymatic activity		GSH decreased [1 mg/L]	
		lipid peroxidation		LPO dose–dependent increase but no significant difference was reported for this range	
	<b>Isochrysis galbana</b> [Unicellular algae]	oxidative stress	1–7 mg/L	increase OH <sup>•</sup> production [1 mg/L]	Miller <i>et al.</i> 2012
	<b>Karenia brevis</b> [Dinoflagellate]	growth inhibition	5–30 mg/L	EC <sub>50</sub> = 10.69 mg/L over 72h	Li <i>et al.</i> 2015
		ROS production	10 mg/L	hydroxyl radical production in chloroplast	
	<b>Mytilus coruscus</b> [Hard shelled mussel]	immune function	2.5–10 mg/L	impairment of immunocompetence, reduced phagocytosis & lysosomal content over 14days [10 mg/L]	Huang <i>et al.</i> 2016
		ROS production		increase in ROS production (14 days) [10 mg/L]	
		enzyme activities	2.5, 10 mg/L	reduced digestive enzyme activities over 14 days (weak recoveries after a 7-day repair period)	Kong <i>et al.</i> 2019
		immune function	0.1, 1, 10 mg/L	-reduced total hemocyte count -increased hemocyte mortality -reduced lysosomal content -reduced phagocytosis activity -increased ROS activity smaller the NP the higher the toxicity. over 21 days [10 mg/L]	Wang <i>et al.</i> 2019
	<b>Mytilus galloprovincialis</b> [Mediterranean mussel]	immune function	1–100 µg/L	alterations in the gene expression, decreased mRNA levels for the antimicrobial peptides [1 µg/L]	Barmo <i>et al.</i> 2013

UV-filter	Species Model [Common name]	Endpoint	Tested conc. range	Toxic Effects [Conc with toxic effect]	Reference
		immune function	1–100 µg/L	particle internalization in the digestion gland (ROS formed) over 96h [100 µg/L]	
		immune function	10 mg/L	gills: Influx of haemocytes, histomorphological + histochemical alterations over 4days [10 mg/L]  digestive glands: vacuolation, histomorphological + histochemical alterations [10 mg/L]	D'Agata <i>et al.</i> 2014
		DNA damage		DNA strand breaks, over expression of <i>mt20</i> gene [10 mg/L]	
		embryotoxicity	0–64 mg/L	alterations in the developmental stages (light/dark conditions) [4 mg/L]	Libralato <i>et al.</i> 2013
		LMS	1–10 µg/L	decrease in lysosomal membrane stability [5 µg/L]	Ciacci <i>et al.</i> 2012
		phagocytosis		80% phagocytic activity, activity reduced at higher concentrations [1 µg/L]	
		hemocytes activity		total oxyradical production (extracellular). Superoxide anion (O <sub>2</sub> <sup>-</sup> ) production [5 µg/L]	
		LMS	0.05–5 mg/L	74% lysosomal membrane destabilization in both the hemocytes and the digestive gland (24h). 7-fold increase in lysosomal lipofuscin accumulation [1 mg/L]	Canesi <i>et al.</i> 2010
		enzymatic activity		157% increased activity of the antioxidant enzyme catalase in digestive gland [1 mg/L]	
		enzymatic activity		155% increase activity of glutathione transferase in digestive gland [0.2 mg/L]	
		LMS	1, 5, 10 µg/ml	no lysosomal membrane destabilization.	Canesi <i>et al.</i> 2010(a)
		ROS production		increased extracellular ROS (dose dependent increase) [5 µg/ml]	
		nitrite production		increase nitrite at 2h followed by a reduction [1 µg/ml]	
		enzymatic activity		increased phosphorylation (activation) on p38 MAP Kinase after 5mins [10 µg/ml]	
	<b><i>Nitzschia Closterium</i></b> [Unicellular algae]	growth inhibition	5–100 mg/L	reduced cell count, EC <sub>50</sub> =88.78 mg/L for 21 nm TiO <sub>2</sub> NPs (96h)	Xia <i>et al.</i> 2015
		growth inhibition	10–360 mg/L	reduced cell count, EC <sub>50</sub> =118.80 mg/L for 60 nm TiO <sub>2</sub> NPs (96h)	
		oxidative stress	5 mg/L	increased LPO production, decreased membrane integrity, measured over 2 – 48h. 2h gave a hormesis effect	

UV-filter	Species Model [Common name]	Endpoint	Tested conc. range	Toxic Effects [Conc with toxic effect]	Reference
		particle internalization		particle internalization occurred at 48h	
	<b><i>Nannochloropsis gaditana</i></b> [Unicellular algae]	enzymatic activity	1–10 mg/L	high H <sub>2</sub> O <sub>2</sub> produced at first 12h, combined with UV. Sunscreen products produced a higher concentration of H <sub>2</sub> O <sub>2</sub> [10 mg/L]	Sendra <i>et al.</i> 2017
		cell density		reduced cell density (combined UV) [1 mg/L]	
	<b><i>Orbicella faveolata</i></b> [Star coral]	zooxanthellae count	0.1 + 10 mg/L	reduced abundance of zooxanthellae (17 days) (slight bleaching) [10 mg/L]	Jovanović and Guzmán, 2014
		gene expression		gene for heat shock protein 70 (HSP.70) was upregulated	
	<b><i>Perna viridis</i></b> [Green-lipped mussel]	mortality	2.5–10 mg/L	increase hemocyte mortality over [10 mg/L]	Wang <i>et al.</i> 2014
		phagocytosis		phagocytosis decreases at 24h [10 mg/L]	
		lysosomal content		decreased lysosomal content [10 mg/L]	
		total hemocyte count		increase hemocyte response [10 mg/L]	
	<b><i>Phaeodactylum tricorutum</i></b> [Unicellular algae]	growth inhibition	0.1–400 mg/L	growth reduced, EC <sub>50</sub> = 1.98 mg/L (+UVA exposure) 144 mg/L (visible light) (3–72h)	Sendra <i>et al.</i> 2017a
		cell wall damage		34% cell membrane damage [2 mg/L]	
		ROS production	2, 5 mg/L	intracellular production of ROS under additional UVA exposure, (3–72h) [2 mg/L]	
		EPS production		increasing production of exo-polymeric substances with increasing concentrations (dose dependant)	
		growth inhibition	5–200 mg/L	EC <sub>50</sub> = 12.65 mg/L over 1 day	Wang <i>et al.</i> 2016
		cell wall damage		25% cell wall damage due to aggregates formed over 5 days, dose-response effect [10 mg/L]	
		oxidative stress		ROS production, only short-term stress (15min exposure) recovered after longer exposure	
		growth inhibition		shading affect, chlorophyll a reduction was observed over 5 days [10 mg/L]	
		growth inhibition	2.5–40 mg/L	growth inhibition of 49.59% at 48h [40 mg/L]	Deng <i>et al.</i> 2017
		chlorophyll a content		increase in chlorophyll a at 24h exposure [10 mg/L]	
		enzymatic activity		increase in SOD and POD activity [2.5 mg/L]	

UV-filter	Species Model [Common name]	Endpoint	Tested conc. range	Toxic Effects [Conc with toxic effect]	Reference
	<i>Pleurochrysis roscoffensis</i> [Unicellular algae]	enzymatic activity	1–10 mg/L	high H <sub>2</sub> O <sub>2</sub> produced at first 12h, combined with UV. Sunscreen products produced a higher concentration of H <sub>2</sub> O <sub>2</sub> [10 mg/L]	Sendra <i>et al.</i> 2017a
		cell density		reduced cell density (combined UV) [1 mg/L]	
	<i>Skeletonema costatum</i> [Diatom]	growth inhibition	5–30 mg/L	EC <sub>50</sub> = 7.37 mg/L over 72h	Li <i>et al.</i> 2015
	<i>Tegillarca granosa</i> [Blood cockle]	gene regulation	0.1–10 mg/L	neurotransmitter genes were down regulated (DA, GABA, and AChE) over 96h [0.1 mg/L]	Guan <i>et al.</i> 2018
enzymatic activity			lower AChE activity over 96h [0.1 mg/L]		
	<i>Thalassiosira pseudonana</i> [Diatom]	growth rates	1–10 mg/L	reduced growth [3 mg/L]	Miller <i>et al.</i> 2012
TiO <sub>2</sub> (bulk)	<i>Amphiprion ocellaris</i> [Clown fish]	mortality	1–100 mg/L	6.7% of mortality [100 mg/L]	Barone <i>et al.</i> 2019
		feeding behavior		10% of fishes with abnormal behavior [1 mg/L]	
		swimming behavior		10% of fishes with abnormal movements [1 mg/L]	
	<i>Mytilus galloprovincialis</i> [Mediterranean mussel]	immune function	10 mg/L	gills–Hypoplasia, loss in cilia (histomorphological + histochemical alterations) over 4 days [10 mg/L]	D'Agata <i>et al.</i> 2014
		immune function		digestive glands– vacuolation & haemocyte influx (histomorphological + histochemical alterations) over 4 days [10 mg/L]	
	<i>Nitzschia closterium</i> [Unicellular algae]	growth inhibition	20–500 mg/L	reduced cell count, EC <sub>50</sub> = 179 mg/L (96h)	Xia <i>et al.</i> 2015
	<i>Phaeodactylum tricornutum</i> [Unicellular algae]	growth inhibition	0.1–400 mg/L	growth inhibition, EC <sub>50</sub> = 6.58 mg/L (+UVA exposure) 185 mg/L (visible light), 3–72h	Sendra <i>et al.</i> 2017a
cell wall damage			no damage (unlike with TiO <sub>2</sub> nanoparticles)		
ROS production		2, 5 mg/L	ROS production in visible light, (3–72h) [5 mg/L]		
EPS production			increasing production of exo- polymeric substances with increasing concentrations (dose dependent)		
ZnO (nanoparticles)	<i>Acropora sp.</i> [Stoney coral]	abundance of zooxanthellae	6.3 mg/L	high abundance of zooxanthellae released into seawater from coral. (24 + 48h) [6.3 mg/L]	Corinaldesi <i>et al.</i> 2018
		damaged zooxanthellae		high increase of damaged zooxanthellae (24 + 48h) [6.3 mg/L]	
		bleaching		severe bleaching occurred [6.3 mg/L]	
		microbial		high increase in prokaryotes and viral abundance in the	

UV-filter	Species Model [Common name]	Endpoint	Tested conc. range	Toxic Effects [Conc with toxic effect]	Reference
		enrichment		surrounding seawater of the exposed coral. [6.3 mg/L]	
	<b>Alexandrium minutum</b> [Dinoflagellate]	ROS production	1–100 mg/L	ROS production increased [10 mg/L]	Castro-bugallo and Barreiro, 2014
	<b>Artemia salina</b> [Brine shrimp]	mortality	133 mg/L	mortality (24h ~6%) ( 48h ~30%) [133 mg/L]	Miglietta <i>et al.</i> 2011
		mortality	10–100 mg Zn/L	EC <sub>50</sub> = 58 mg/L at 96h,	Schiavo <i>et al.</i> 2017
		mortality	0.03 –0.5 mg Zn/L	EC <sub>50</sub> = 0.02 mg/L at 14days	
		mortality	10–100 mg/L	<20% mortality was observed at 96h [10 mg/L]	Ates <i>et al.</i> 2013
		lipid peroxidation		total malondialdehyde (MDA) increased with higher concentration and time	
	<b>Chaetoceros gracilis</b> [Unicellular algae]	growth inhibition	10–80 mg/L	complete cessation of growth [2.59 mg/L]	Wang and Wang, 2014
	<b>Corophium volutator</b> [Amphipod]	growth inhibition	200–1000 µg/l	delayed growth and affected reproduction [1000 µg/L]	Fabrega <i>et al.</i> 2012
		growth inhibition		growth rate reduced by 11% [1000 µg/L]	
	<b>Crassostrea Gigas</b> [Pacific oyster]	mortality	50 µg–50 mg/L	LC <sub>50</sub> = 37.2 mg/L at 96h	Trevisan <i>et al.</i> 2014
		enzymatic activity	4 mg/L	decrease in glutathione reductase activity at 24h for gills, 48h for digestive gland [4 mg/L]	
	<b>Dunaliella tertiolecta</b> [Unicellular algae]	growth inhibition	0.1–10 mg/L (Zn)	EC <sub>50</sub> = 1.94 mg/L (Zn)	Manzo <i>et al.</i> 2013a
		growth inhibition	0.1–10 mg/L (Zn)	EC <sub>50</sub> = 2.2 mg/ (Zn) over 24h	Schiavo <i>et al.</i> 2017
		genotoxicity	5–100 mg/L (Zn)	55% of nuclei seriously damaged [25 mg/L]	Schiavo <i>et al.</i> 2016
		viability	10–100 mg/L (Zn)	32% and 26% viable cells at 50 and 100 mg/L, respectively over 72h	Miller <i>et al.</i> 2010
		growth rates	10–1000 µg/L	significant reduced growth [1000 µg/L]	
		growth inhibition	133 mg/L	100% growth inhibition at 96h [133 mg/L]	Miglietta <i>et al.</i> 2011
		growth inhibition	0.081–810 mg/L	IC <sub>50</sub> = 0.88 mg/L over 24h, (using f/2 media) [0.88 mg/L]	Aravantinou <i>et al.</i> 2015
	<b>Dunaliella tertiolecta</b> [Unicellular algae]	growth inhibition	1–5 mg/L (Zn)	cell division inhibition over 72h [2 mg/L (Zn)]	Schiavo <i>et al.</i> 2016
	<b>Isochrysis galbana</b> [Prymnesiophytes]	growth rates	10–1000 µg/L	reduced growth [1000 µg/L]	Miller <i>et al.</i> 2010
		growth inhibition	133 mg/L	100% growth inhibition at 96h [133 mg/L]	Miglietta <i>et al.</i> 2011
	<b>Lytechinus pictus</b> [Sea urchin]	larval morphology	1-50 µM of Zn	EC <sub>50</sub> = 99.5 µg/L	Manzo <i>et al.</i> 2013
	<b>Melita longidactyla</b> [Amphipod]	mortality	0.1–10 mg/L	LC <sub>50</sub> = 0.04 mg/L at 96h with a temperature of 25°C	Wong and Leung, 2014

UV-filter	Species Model [Common name]	Endpoint	Tested conc. range	Toxic Effects [Conc with toxic effect]	Reference
	<b><i>Mytilus galloprovincialis</i></b> [Mediterranean Mussel]	LMS	1–10 µg/L	decrease in lysosomal membrane stability [1 µg/L]	Ciacci <i>et al.</i> 2012
		phagocytosis		80% phagocytic activity observed, activity reduced at higher concentrations [1 µg/L]	
		hemocytes activity		total oxyradical production (extracellular) [1 µg/L]	
		respiration rate	0.1–2 mg/L	respiration rate increased with increasing concentrations, 12week exposure [<2 mg/L]	Hanna <i>et al.</i> 2013
		growth		decrease in shell total length, 41–47% less growth in 12weeks [2 mg/L]	
	<b><i>Nereis diversicolor</i></b> [Rag worm]	survival		62% large mussels and 23% small mussels survived after 12 weeks [2 mg/L]	
		behavior	3 mg/kg	reduced feeding rate (16-day exposure)	Buffet <i>et al.</i> 2012
		oxidative stress		increased immune enzymes; CAT, GST, SOD [3 mg/kg]	
		enzymatic activities		reduced thiobarbituric acid reactive substances (TBARS) and AChE [3 mg/kg]	
	<b><i>Oryzias melastigma</i></b> [Marine medaka]	mortality	0.1–10 mg/L	40% of embryo mortality [10 mg/L]	Cong <i>et al.</i> 2017
		heartbeat		increased embryo heart beats [10 mg/L]	
		hatching success		16.7% hatched [10 mg/L]	
		hatching time		delayed hatching of embryos [0.1 mg/L]	
		abnormalities		increased malformation % of newly hatched larvae, (increased with increasing concentrations)	
	<b><i>Paracentrotus lividus</i></b> [Purple sea urchin]	growth	133 mg/L	growth of embryos reduced, 100% toxic effect observed [133 mg/L]	Miglietta <i>et al.</i> 2011
		spermiotoxicity	1–50 µM Zn	LOEC = 0.30 mg/L (Zn)	Manzo <i>et al.</i> 2013
		spermiotoxicity	0.11–30 µM Zn	EC <sub>50</sub> = 0.92 µM (Zn 100nm) for DNA damage EC <sub>50</sub> = 1.23 µM (Zn 100nm) for offspring quality NOEC = < 0.1 DNA damage DNA damage in sperm cells higher at lower conc.	Oliviero <i>et al.</i> 2019
	<b><i>Phaeodactylum tricorutum</i></b> [Diatom]	growth inhibition	10–80 mg/L	no effect	Peng <i>et al.</i> 2011
		growth inhibition	0.1–10 mg/L (Zn)	EC <sub>50</sub> = 1.09 mg/L (Zn)	Li <i>et al.</i> 2017
		growth inhibition	1–100 mg/L	growth reduction [10 mg/L]	Castro-bugallo <i>et al.</i> 2014
	<b><i>Picochlorum sp.</i></b> [Unicellular algae]	growth inhibition	10 mg/L	only inhibited in first 5 days, after viability of cells increased as aggregates of metals formed (5 weeks)	Hazeem <i>et al.</i> 2016
		chlorophyll a		no effect	

UV-filter	Species Model [Common name]	Endpoint	Tested conc. range	Toxic Effects [Conc with toxic effect]	Reference
	<b>Ruditapes philippinarum</b> [Manila clams]	enzyme activities	1 and 10 µg/L	SOD activity increased [10 µg/L]	Marisa <i>et al.</i> 2016
enzyme activities			CAT activity increased [10 µg/L]		
enzyme activities			AchE activity increase [1 µg/L]		
	<b>Scrobicularia plana</b> [Peppery furrow shell]	behavior	3 mg/kg	reduced feeding rate (16 days exposure) [3 mg/kg]	Buffet <i>et al.</i> 2012
oxidative stress			increased immune enzymes; CAT, GST, SOD [3 mg/kg]		
enzymatic activities			increased thiobarbituric acid reactive substances (TBARS) and AchE [3 mg/kg]		
	<b>Skeletonema costatum</b> [Unicellular algae]	growth inhibition	0–10 mg/L (Zn)	complete growth inhibition, EC <sub>50</sub> = 3.6 mg/L (Zn)	Zhang <i>et al.</i> 2016
growth inhibition		0–10 mg/L (Zn)	complete growth inhibition, EC <sub>50</sub> = 2.36 mg/L (Zn)	Wong and Leung, 2014	
chlorophyll fluorescence		0.5–16 mg/L	IC <sub>50</sub> = 12 mg/L or 1.18mg Zn/L over 96h at 25°C		
growth inhibition		4–40 mg/L	IC <sub>50</sub> = 2.36 mg/L	Wong <i>et al.</i> 2010	
	<b>Skeletonema marinoi</b> [Unicellular algae]	growth rates	10 –1000 µg/L	growth reduced by a factor of 2 [1000 µg/L]	Miller <i>et al.</i> 2010
	<b>Stylophora pistillata</b> [Hood coral]	photosynthetic efficiency	10–5000 µg/L	photosynthetic efficiency decreased to 0 on day 14 [1200 µg/L]	Fel <i>et al.</i> 2018
	<b>Synechococcus elongatus</b> [Cyanobacteria]	growth inhibition	1, 5, 10 mg/L	reduced optical density for coated and uncoated ZnO [5, 10 mg/L]	Vicente <i>et al.</i> 2019
	<b>Tetraselmis suecica</b> [Unicellular algae]	growth inhibition	133 mg/L	75% growth inhibition over 96h [133 mg/L]	Miglietta <i>et al.</i> 2011
growth inhibition		0.1–10 mg/L (Zn)	EC <sub>50</sub> = 3.91 mg/L (Zn)	Li <i>et al.</i> 2017	
growth inhibition		1–100 mg/L	no growth inhibition	Castro-bugallo and Barreiro, 2014	
inhibitory concentrations (using f/2 media)		0.081–810 mg/L	IC <sub>50</sub> = 0.13 mg/L over 24h	Aravantinou <i>et al.</i> 2015	
	<b>Thalassiosira pseudonana</b> [Unicellular algae]	growth inhibition	4–40 mg/L	IC <sub>50</sub> = 4.56 mg/L	Wong <i>et al.</i> 2010
growth inhibition		10–80 mg/L	complete cessation of growth at 2.59 mg/L (EC <sub>50</sub> = 0.82 mg/L) (nanospheres and nanoneedles examined)	Peng <i>et al.</i> 2011	
growth rates		10–1000 µg/L	growth reduced by a factor of 3 [500 µg/L]	Miller <i>et al.</i> 2010	
growth inhibition		see effects	growth ceased, see paper for concentrations as different treatments were used	Miao <i>et al.</i> 2010	
chlorophyll a		see effects	chlorophyll a content decreased, see paper for concentrations as different treatments were used		
growth inhibition		0.5–50 mg/L	IC <sub>50</sub> = 5 mg/L over 96h (for the	Yung <i>et al.</i> 2015	

UV-filter	Species Model [Common name]	Endpoint	Tested conc. range	Toxic Effects [Conc with toxic effect] average ocean salinities)	Reference
		growth inhibition	0.1–100 mg/L	IC <sub>50</sub> ≤ 10 mg/L over 96h	Yung <i>et al.</i> 2017
		cell damage	10 mg/L	cell wall damage over 4 days [10 mg/L]	
	<b>Thalassiosira weissflogii</b> [Centric diatom]	growth inhibition	0.1-1 mg/L	reduced algal cell density, control had 5/6-fold increase [0.1-1 mg/L]	Bielmyer-Fraser <i>et al.</i> 2014
	<b>Tigriopus japonicus</b> [Crustacean]	mortality	4–40 mg/L	LC <sub>50</sub> = 0.85 mg/L	Wong <i>et al.</i> 2010
	<b>Vibrio fischeri</b> [Bacteria]	bioluminescence test	0.3–40 mg/L (Zn)	EC <sub>50</sub> = 17 mg/L (Zn) over 30mins	Schiavo <i>et al.</i> 2017
<b>ZnO (bulk)</b>	<b>Artemia salina</b> [Brine shrimp]	mortality	10–100 mg Zn/L	EC <sub>50</sub> = 38 mg/L over 96h (Zn)	Schiavo <i>et al.</i> 2017
		mortality and body growth	0.03–0.5 mg Zn/L	EC <sub>50</sub> = 0.02 mg/L over 14days (Zn)	
	<b>Elasmopus rapax</b> [Crustacean]	mortality	4–40 mg/L	LC <sub>50</sub> = 0.37 mg/L	Wong <i>et al.</i> 2010
	<b>Dunaliella tertiolecta</b> [Unicellular algae]	growth inhibition	0.1–10 mg/L (Zn)	EC <sub>50</sub> = 3.57 mg/L (Zn)	Schiavo <i>et al.</i> 2017
	<b>Paracentrotus lividus</b> [Mediterranean Sea urchin]	embryotoxicity	1 –50 µM of Zn	EC <sub>50</sub> = 0.059 mg/L (Zn)	Manzo <i>et al.</i> 2013
		spermiotoxicity		LOEC = 0.18 mg/L (Zn)	
		spermiotoxicity	0.11–30 µM Zn	EC <sub>50</sub> = 1.59 µM (Zn) for DNA damage EC <sub>50</sub> = 0.92 µM (Zn) for offspring quality	Oliviero <i>et al.</i> 2019
	<b>Phaeodactylum tricorutum</b> [Unicellular algae]	growth inhibition	0.1–10 mg/L (Zn)	EC <sub>50</sub> = 3.91 mg/L	Li <i>et al.</i> 2017
	<b>Skeletonema costatum</b> [Unicellular algae]	growth inhibition	0–10 mg/L (Zn)	complete growth inhibition observed, EC <sub>50</sub> = 5.5 mg/L	Zhang <i>et al.</i> 2016
		mortality	4–40 mg/L	LC <sub>50</sub> = 6.65 mg/L	Wong <i>et al.</i> 2010
	<b>Tetraselmis suecica</b> [Unicellular algae]	growth inhibition	0.1–10 mg/L (Zn)	EC <sub>50</sub> = 7.12 mg/L (Zn)	Li <i>et al.</i> 2017
	<b>Thalassiosira pseudonana</b> [Unicellular algae]	growth inhibition	4–40 mg/L	LC <sub>50</sub> = 2.97 mg/L	
	<b>Tigriopus japonicus</b> [Crustacean]	mortality	4–40 mg/L	LC <sub>50</sub> = 0.43 mg/L	

Abbreviations: EC<sub>50</sub>, half maximal effective concentration; IC<sub>50</sub>, half maximal inhibitory concentration; LC<sub>50</sub>, lethal concentration; LOEC, Lowest Observed Effect Concentration; DA, dopamine; EPS, exopolymer substances; GABA, gamma-aminobutyric acid; GHS, glutathione; LMS, lysosomal membrane stability; LPO, lipid peroxidation; MDA, malondialdehyde; ROS, reactive oxygen species; SOD, superoxide dismutase; CAT, catalase; LDH, lactate dehydrogenase; AchE, acetylcholinesterase; POD, peroxidase

## 4. Standardized toxicity studies

Today, there are no guidelines or standards specifically set for UV-filters for marine organisms. The Organization for Economic Co-operation and Development (OECD) has developed guidelines for toxicity testing on fish (OECD test guideline N°203, Fish, Acute toxicity Testing), although they recommend the use of the following marine species: *Pagrus major*, *Dicentrarchus labrax* and *Cyprinodon variegatus*. From an overall perspective, these guidelines are not specifically designed for marine organisms, let alone for the testing of highly hydrophobic chemicals such as UV-filters.



Standards that are globally harmonized would facilitate comparisons between studies by normalizing toxicological endpoints, exposure media, materials, tested concentrations and exposition times. In the literature, exposure concentrations vary greatly among the different studies ranging from a wide range (0.32 – 5000 µg/L, Torres *et al.* 2016) to conservative ones (0.5-10 µg/L, Chen *et al.* 2018). It is noteworthy to underline that some authors measured the exposure concentration with methods that are hardly comparable between studies, due to the UV-filter's physicochemical properties. In addition, with similar organisms, exposure time is a parameter displaying variation from several hours (Downs *et al.* 2014; Seoane *et al.* 2017) to several days (He *et al.* 2018; Stien *et al.* 2019). Hydrophobic UV-filters tend to adhere to plastic and glass. Materials used for toxicity studies differ, some authors used PTFE (Teflon) (Downs *et al.* 2016; Sendra *et al.* 2017) or glass (Kusk *et al.* 2011; He *et al.* 2019(a)), while it is not mentioned in most studies. Experimental studies performed in our group have highlighted that some UV-filters such as BP3 can evaporate during extraction process resulting in underestimated concentrations. This point emphasizes a need for standardization of analytical methods.

## 5. Bioaccumulation of UV-filters

Bioaccumulation may be a better indicative measure of the spread of UV-filters in the Ocean. These studies are pivotal to better understand to which extent marine organisms are exposed to UV-filters. Two types of studies can be distinguished: those that measure the concentration of different compounds from organisms collected *in situ* and those that expose the organism to a given concentration *in vivo*, in order to measure the bioaccumulation fraction.

Organic UV-filters are poorly soluble compounds, characterized by a high octanol-water partition coefficient (Log  $K_{ow}$ ) (Apel *et al.* 2018; Fagervold *et al.* 2019). These hydrophobic properties result in bioaccumulation of UV-filters in marine species. Interestingly, this literature analysis reveals the lack of studies for most UV-filters. Indeed, only a few were targeted in bioaccumulation analyses with 14 sunscreen UV-filters, the others being benzotriazole derivatives. A positive point that can be drawn from the literature is that studies are approaching a worldwide coverage, including the North & South Atlantic Ocean, the Pacific Ocean and the Mediterranean Sea.

### 5.1 *In situ*

Accumulation of UV-filters in mussels has been shown in different environments: Atlantic (Castro *et al.* 2018), Mediterranean Sea (Bachelot *et al.* 2012) and Pacific (Nakata *et al.* 2012) with concentrations up to 3,992 ng/g dw for OC and 1,765 ng/g dw for EMC (Picot Groz *et al.* 2014). UV-filter prevalence in marine organisms is becoming a health concern as they are detected in seafood. Mussels, clam, mackerel, tuna, salmon and cod were shown to accumulate multiple UV-filters (Cunha *et al.* 2015; Langford *et al.* 2015). Cunha and coworkers (2018) highlighted the presence of nine UV-filters in shrimp, pangasius and salmon collected in an aquaculture farm. In 2019, Rodil *et al.* assessed the occurrence at 5 timepoints throughout a year, of multiple emerging pollutants, including BP3, EMC, OD-PABA, OC and 4-MBC, in bivalves. If they do not highlight a clear temporality in UV-filters abundance, they identified UV-filters as the most abundant persistent pollutants, behind polycyclic aromatic hydrocarbons.

The fate of sunscreen agents in species of higher trophic level has been poorly documented. To date, three studies were carried out on dolphins, which are the first assessments of UVF in marine mammals. Dolphins have been identified to accumulate organic pollutants and are good candidates as sentinel species for Ocean health, due to biomagnification (Bossart, 2011; Tanabe *et al.* 1994). Benzotriazole UV stabilizers were identified in blubbers of finless porpoises in Japan with a bioaccumulation factor as high as 33,300, which is ten times higher than those reported for small fish (Nakata *et al.* 2010). Alonso *et al.* (2015) demonstrated maternal transfer of UV-filters in Franciscana

Dolphin, demonstrating that 4-MBC, EMC, OD-PABA and OC can be considered as transplacental contaminants. These results suggest that UV-filters biomagnify through the marine food web. Further analyses are needed to better apprehend the biomagnification of these emerging pollutants.

**Table 3.** *In situ* summary of UV-filter bioaccumulation studies.

Reference	Organism model	Sunscreen components (abbreviations)	Concentration range	Location
Alonso <i>et al.</i> 2015	Dolphin ( <i>Pontoporia blainvillei</i> & <i>Sotalia guianensis</i> )	EMC 4-MBC OC OD-PABA	ND–250 ng/g lw ND–855 ng/g lw ND–11,130 ng/g lw ND–1385 ng/g lw	South Atlantic Ocean
Bachelot <i>et al.</i> 2012	Mussels ( <i>Mytilus edulis</i> & <i>Mytilus galloprovincialis</i> )	EMC OC OD-PABA	3–256 ng/g dw 1–200 ng/g dw ND	French Atlantic, Mediterranean Sea
Barbosa <i>et al.</i> 2018	Gilt-head bream, mackerel, salmon, cod, monkfish and mussels. (Seafood species, wild and farmed origins.)	ES HS BP6 OC BP1 4MBC DHMB	5–12 µg/kg ww >LOD–10 µg/kg ww 20–110 µg/kg ww 4–28 µg/kg ww 9–27 µg/kg ww ND (detected after steaming) ND (detected after steaming)	North Sea, Atlantic Ocean, Mediterranean Sea
Castro <i>et al.</i> 2018	Mussels ( <i>Mytilus galloprovincialis</i> & <i>Mytilus edulis</i> )	ES BP3 4MBC IMC EMC	ND–59.2 ng/g dw ND–622.1 ng/g dw ND–88.3 ng/g dw ND–43.1 ng/g dw ND–181.8 ng/g dw	North Atlantic Ocean
Cunha <i>et al.</i> 2015	Mussels, mullet, clam & flounder	BP1 BP3 OD-PABA HS DHMB IMC OC EMC ES DHMB 4-MBC	ND ND – <MQL (ng/g dw) ND ND ND – <MQL (ng/g dw) ND ND – <LOQ (ng/g dw) ND – <LOQ (ng/g dw) ND ND ND	North Atlantic Ocean, North Sea, Mediterranean Sea
Cunha <i>et al.</i> 2018	Mackerel, tuna, salmon, seabream, cod, monkfish, crab, shrimp, octopus, plaice, hake, mussel & sardine (Seafood species, wild, farmed and industrial origins)	BP1 BP3 ES 4-MBC EMC IMC DHMB OD-PABA OC DHMB	ND–98.9 µg/kg dw ND–98.7 µg/kg dw ND–72.1 µg/kg dw ND–56.2 µg/kg dw ND–74.4 µg/kg dw ND–66.7 µg/kg dw ND–90.7 µg/kg dw ND ND–103.3 µg/kg dw ND	Mediterranean Sea, North Sea, North Atlantic Ocean, Pacific Ocean
Gago-Ferrero <i>et al.</i> 2013	Dolphin ( <i>Pontoporia blainvillei</i> )	OC	89–782 ng/g lw	South Atlantic Ocean
Horricks <i>et al.</i> 2019	Lionfish ( <i>Pterois volitans</i> )	4MBC EMC BP3	2.11 µg/kg ww ND or below LOD 0.12–2.90 µg/kg ww	Caribbean Sea

		OD-PABA	ND or below LOD	
Kim <i>et al.</i> 2011	Several fish species ( <i>see article</i> )	UV-P *	ND–222 µg/g lw	South China Sea
		UV–326 *	ND–71.3 µg/g lw	
		UV–328 *	ND – 563 µg/g lw	
		UV–329 *	ND – 96.7 µg/g lw	
		UV–9 *	ND – 16.2 µg/g lw	
		UV–234 *	ND – 126 µg/g lw	
		UV–320 *	ND – 28.7 µg/g lw	
		UV–327 *	ND – 61.5 µg/g lw	
Langford <i>et al.</i> 2015	Crab, Prawn & Cod ( <i>Carcinus meanas</i> , <i>Pandalus borealis</i> & <i>Gadus morhua</i> )	BP3	<20–1037 ng/g dw	Norwegian Sea
		OD-PABA	<20–21.3 ng/g dw	
		EMC	<30–36.9 ng/g dw	
		OC	<20–11875 ng/g dw (cod's liver)	
		OC	<10–23.1 ng/g dw	
		UV–234 *	<10 ng/g dw	
		UV–327 *	<51.8 ng/g dw	
		UV–328 *	<19.5 ng/g dw	
		UV–329 *	<25 ng/g dw	
Mitchelmore <i>et al.</i> 2019	Coral (Coral tissue from sites around Oahu, Hawaii)	HS	100–600 ng/g dw	North Pacific Ocean
		ES	100–800 ng/g dw	
		BP3	0–600 ng/g dw	
		OC	0–800 ng/g dw	
Molins-Delgado <i>et al.</i> 2018	Mullet ( <i>Mugil liza</i> )	BP1	>LOQ–17.1 ng/g dw	South Atlantic Ocean
		BP3	10.8–81.4 ng/g dw	
		4-MBC	12.6–31.5 ng/g dw	
		EMC	>LOQ–98.8 ng/g dw	
		OD-PABA	>LOQ	
		OC	5.03–68 ng/g dw	
		4-HB **	40.1–152 ng/g dw	
		4-DHB **	43.7–462 ng/g dw	
Nakata <i>et al.</i> 2009	55 samples including different organisms ( <i>see article</i> )	UV–320 *	>0.05–0.60 ng/g ww	East China Sea (Ariake Sea)
		UV–326 *	>0.1–0.91 ng/g ww	
		UV–327 *	>0.12–0.98 ng/g ww	
		UV–328 *	>0.15–0.80 ng/g ww	
Nakata <i>et al.</i> 2010	Finless porpoises ( <i>Neophocaena phocaenides</i> )	UV–328 *	11–64 ng/g ww	East China Sea (Ariake Sea)
		UV–327 *	4.5–31 ng/g ww	
		UV–320 *	<0.05	
Nakata <i>et al.</i> 2012	Green mussels & Blue mussels ( <i>Perna viridis</i> & <i>Mytilus edulis</i> )	UV–320 *	ND–33 ng/g lw	Pacific Ocean
		UV–326 *	ND–450 ng/g lw	
		UV–327 *	ND–150 ng/g lw	
		UV–328 *	ND–220 ng/g lw	
Pacheco-Juárez <i>et al.</i> 2019	Macroalgae (12 species of macroalgae)	UV-P *	0–80 ng/g dw	North Atlantic Ocean
		UV–329 *		
		UV–326 *		
		UV–328 *		
		UV–327 *		
		MBBT	0–115 ng/g dw	
Peng <i>et al.</i> 2017	17 species of fishes, 2	BP3	>43.40 ng/g lw	South China Sea

	species of cephalopods (squids), and 5 species of crustaceans (see article)	OC	>31.8 ng/g lw	
		4-MBC	2–38.9 ng/g lw	
		BM	ND–21 ng/g lw	
		EMC	ND–13 ng/g lw	
		UV531 *	ND–294 ng/g lw	
		UVP *	1–10.64 ng/g lw	
		UV329 *	ND–25 ng/g lw	
		UV326 *	ND–8.97 ng/g lw	
		UV234 *	0.7–14.54 ng/g lw	
		UV328 *	ND–23 ng/g lw	
		UV327 *	0.8–9 ng/g lw	
Picot Groz et al. 2014	Mussel ( <i>Mytilus galloprovincialis</i> )	OD-PABA	0–833 ng/g dw	North Atlantic Ocean
		OC	0–3992 ng/g dw	
		EMC	0–1765 ng/g dw	
		UV-P *	ND	
		UV-326 *	recovery too low 33%	
Rodil et al. 2019	Wild and cultivated mussels, cockle and clam ( <i>Mytilus galloprovincialis</i> , <i>Cerastoderma edule</i> , <i>Ruditapes decussatus</i> )	4MBC	0–49 ng/g dw	North Atlantic Ocean
		BP4	0–87 ng/g dw	
		BP3	0–63 ng/g dw	
		EMC	0–94 ng/g dw	
		OD-PABA	0–12 ng/g dw	
		OC	0–141 ng/g dw	
Tsui et al. 2017	Coral ( <i>Platygyra acuta</i> , <i>P. Porites</i> sp., <i>Pavona decussata</i> , <i>Acropora valida</i> & <i>Favites abdita</i> )	BP1	below LOD	South China Sea
		BP8	3.9–24.7 ng/g ww	
		BP3	2–31.8 ng/g ww	
		OC	1.7–7.9 ng/g ww	
		OD-PABA	2.8–12.4 ng/g ww	

Abbreviations: dw, dry weight; ww, wet weight; lw, lipid weight; ND, not detected; LOD, limit of detection; MQL, method quantification limit.

Abbreviations for organic UV-filters: 4-MBC, 4-Methylbenzylidene camphor; BM, Butyl methoxydibenzoylmethane; BP1, Benzophenone-1; BP3, Benzophenone-3; BP4, Benzophenone-4; BP6, Benzophenone-6; BP8, Benzophenone-8; EMC, Ethylhexyl methoxycinnamate; OC, octocrylene; OD-PABA, Ethylhexyl dimethyl PABA; IMC, Isoamyl *p*-methoxycinnamate; DHHB, Diethylamino hydroxybenzoyl hexyl benzoate; PABA, *para*-Aminobenzoic acid; ES, Ethylhexyl salicylate; MBBT, Methylene bis-benzotriazolyl tetramethylbutylphenol

\* Benzotriazole derivatives used in sunscreens

\*\* Benzophenone metabolites

## 5.2 *In vivo*

*In vivo* bioaccumulation studies are scarce, with only 5 and 12 studies on organic and mineral UV-filters, respectively. All studies carried out with organic UV-filters demonstrated bioaccumulation, regardless of the molecule and the organism studied (Table 4, 5). Exposed concentrations ranged from 1 to 1000 µg/L, for organic UV-filters, with three modes of contamination: waterborne (WB), sediment exposure (SE) or dietary (D), the latter imitating trophic transfer mechanisms. Only one study has investigated bioaccumulation using contaminated sediment (Buffet *et al.* 2012), although it is the medium containing the higher concentrations of UV-filter (Fagervold *et al.* 2019; Mitchelmore *et al.* 2019). Vidal-Liñán *et al.* (2018) exposed mussels to different UV-filters at 1 µg/L, which is the lowest concentration noticed in bioaccumulation studies, via the WB route for 14 days. OD-PABA and BP3 underwent the least bioaccumulation with 15.5 ng/g and 67 ng/g dw while OC and 4-MBC were found at 559.5 ng/g and 801 ng/g dw, respectively. As previously mentioned, Stien *et al.* (2019) demonstrated that *Pocillopora damicornis* accumulated and metabolized OC under fatty acid conjugated forms. This study raised the importance of conducting metabolomic studies to ensure that the effects and concentration of anthropogenic contaminants are not underestimated. Regarding the accumulation of mineral filters, there are no studies with bulk filters. In addition, the characteristics of the nanoparticles used are rarely mentioned. Both ZnO and TiO<sub>2</sub> have been demonstrated to encounter assimilation by several organisms such as turbot, oysters or algae (Trevisan, *et al.* 2014; Wang *et al.*, 2016; Wang *et al.*, 2017). Bhuvaneshwari and co-workers (2018) differentiated two different mechanisms: accumulation and uptake. The first is the stable assimilation of the compound while the second is used if the organism can excrete the contaminate over time.

**Table 4.** Summary of *In vivo* studies reporting the bioaccumulation of organic UV–filters.

Organic UV filter	Species Model [Common name]	Exposed conc	Exposure time [exposition route]	Measured conc	Reference
EMC	<i>Tetraselmis suecica</i> [Algae]	100 µg/L	6h [WB]	12.7 µg/L	Gomez <i>et al.</i> 2012
		11.9 µg/L	48h [D – <i>T. suecica</i> ]	38 ng/g dw	
OC	<i>Mytilus galloprovincialis</i> [Mussel]	11.6 µg/L	48h	>33 ng/g dw	He <i>et al.</i> 2019
BP3	<i>Pocillopora damicornis</i> & <i>Seriatopora caliendrum</i> [Coral]	10–1000 µg/L	7 days [WB]	1.42–67.51 ng/g dw	
BP1		10–1000 µg/L		1.02–53.08 ng/g dw	Stien <i>et al.</i> 2019
BP8		10 and 100 µg/L		0.94–37.76 ng/g dw	
OC	<i>Pocillopora damicornis</i> [Coral]	5–1000 µg/L	7 days [WB]	OC–fatty acid conjugates detected	Stien <i>et al.</i> 2019
ES				bioaccumulation under fatty–acid conjugated form	
4-MBC	<i>Mytilus galloprovincialis</i> [Mussel]	1 µg/L	14 days [WB]	801 ng/g dw	Vidal-Liñán <i>et al.</i> 2018
BP3				67 ng/g dw	
BP4				520 ng/g dw	
OC				559.5 ng/g dw	
OD–PABA				13.5 ng/g dw	
BP3	<i>Sparus aurata</i> [Gilt-head bream]	50 ng/ml	14 days [WB]	9-160 ng/g (note; up to 20 by products were detected.)	Ziarrusta <i>et al.</i> 2018

**Table 5.** Summary of In vivo studies reporting the bioaccumulation of inorganic UV–filters.

Mineral UV filter	Species Model [Common name]	Exposed conc	Exposure time [exposition route]	Measured conc	Reference	
nZnO	<i>Artemia salina</i> (larvae) [Brine shrimp]	100 mg/L	24h [WB]	135 µg/g (Zn) or 735 µg/g ZnO	Ates <i>et al.</i> 2013	
		100 mg/L	96h	220 µg/g (Zn) or 1301 µg/g ZnO		
nTiO <sub>2</sub>	<i>Artemia salina</i> [Brine shrimp]	1–7 ng/g dw	48h [D – <i>D. salina</i> ]	0.001 mg/g dw (accumulated)	Bhuvaneshwari <i>et al.</i> 2018	
		10 mg/L	[WB]	0.5 mg/g dw (accumulated)		
		1–7 ng/g dw	[D – <i>D. salina</i> ]	0.12 mg/g dw (uptake)		
		10 mg/L	[WB]	15 mg/g dw (uptake)		
nZnO	<i>Thalassiosira weissflogii</i> [Centric diatoms]	0.1mg/L	7 days [WB]	1x10 <sup>-9</sup> µgZn/cell	Bielmyer-Fraser <i>et al.</i> 2014	
nZnO	<i>Scrobicularia plana</i> [Clam]	3 mg/kg	16 days [SE]	5.4 µg/g ww	Buffet <i>et al.</i> 2012	
	<i>Nereis diversicolor</i> [Rag worm]	3 mg/kg	[SE]	3.7 µg/g ww		
nZnO	<i>Mytilus galloprovincialis</i> [Mussel]	2 mg/L	2 weeks [WB]	350–400 µg/L dw	Hanna <i>et al.</i> 2013	
	<i>Thalassiosira weissflogii</i> [Algae]	168 µg/L	7 days [WB]	17 µg/g dry wt	Jarvis <i>et al.</i> 2013	
	<i>Acartia tonsa</i> [Copepod]	200,000 cells/mL/d	7 days [D – <i>T. weissflogii</i> ]	toxic effects were observed		
	<i>Orbicella faveolata</i> [Star coral]	0.1, 10 mg/L	17 days [WB]	6.19, 16.58 mg/L	Jovanović and Guzmán, 2014	
	<i>Ruditapes philippinarum</i> [Clam]	1, 10 µg/L	7 days [WB]	130 µg/g dw (Zn)	Marisa <i>et al.</i> 2016	
	<i>Mytilus galloprovincialis</i> [Mussel]	10 mg/L	4 days [WB]	880 µg/g dw (Zn)	Montes <i>et al.</i> 2012	
	<i>Crassostrea Gigas</i> [Oyster]	4 mg/L	24h [WB]	0.55 mg/kg (gills)	Trevisan <i>et al.</i> 2014	
			24h	0.28 mg/kg (digestive gland)		
	nTiO <sub>2</sub>	<i>Perinereis aibuhitensis</i> [Clamworm]	10, 50, 100 mg/L	24h [WB]	32.7–59.3 µg/g dw	Wang <i>et al.</i> 2016(b)
		<i>Scophthalmus maximus</i> (juvenile) [Turbot]	32.7 –59.3 µg/g dw	24h [D – clamworms]	toxic effects were observed	
<i>Nitzschia closterium</i> [Algae]		1 mg/L	24h [WB]	ingestion occurred	Wang <i>et al.</i> , 2017	
<i>Chlamys farreri</i> [Scallop]			14 days [D]	4.44–50.18 mg/kg dw		
			14 days [WB]	2.5–65 mg/kg dw		

Abbreviations: dw, dry weight; ww, wet weight; WB, waterborne; D, dietary; SE, sediment exposure. Species mentioned alongside the dietary “D” were previously exposed to UVF before being used as feed.

## 6. Conclusion

This review provides an analysis of the UV-filter toxicity and bioaccumulation studies conducted up to August 2019. This analysis has allowed us to identify the progress made so far, and what remains to be explored. We have highlighted a disparity in the studies. Although, the number of published papers is increasing from year to year, the majority of compounds authorized for cosmetic use have not been studied, while 4-MBC, BPs or OC are at the core of several investigations. In addition, a trend on the models studied has been identified, particularly for organic filters, for which no sedimentary organisms have been studied, though sediments are the most concerned biotopes. It should also be noted that there are no standardized methods specifically designed to comparatively assess the toxicity of these chemicals and marine species. Bioaccumulation studies have shown that these compounds biomagnify through trophic levels. In addition, it is important to note that no *in situ* bioaccumulation studies have been conducted for inorganic filters. Intensive research is therefore needed to better understand the effect of UV-filters, in order to adapt the regulations with a science-based approach.

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