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Special Series

Investigating the exposure and impact of chemical UV filters on coral reef ecosystems: Review and research gap prioritization

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EDITOR'S NOTE:

This article is part of the special series "Consequences of Sunscreen Product Use on Aquatic Environments." This series documents the current state of knowledge concerning potential impacts of chemicals derived from sunscreen products on freshwater and marine ecosystems, including coral reefs. Specific topics discussed include use patterns, environmental loadings, potential exposures, toxicological effects, and future research needs.

Abstract

Coral reefs are among the world's most productive and biologically diverse ecosystems. In recent decades, they have experienced an unparalleled decline resulting from various anthropogenically induced stressors. Ultraviolet (UV) filters found in personal care products, such as sunscreen, are chemical pollutants that are emerging as a growing toxic threat to reef organisms. In this study, a systematic literature review was conducted to (1) determine the current understanding of spatial distribution and the occurrence of UV filters exposed to the marine environment, (2) synthesize current ecotoxicological thresholds of relevant reef organisms under various UV-filter exposures, (3) identify research gaps related to both exposure and toxicity of UV filters in coral reef ecosystems. With gaps identified, a survey was developed and distributed to experts in the field representing academic, governmental, not-for-profit, and industry researchers in order to prioritize research gaps and inform future research efforts. The survey identified the need for better understanding of the impacts of co-stressors, long-term exposure, mixture, and degradation product exposure and realistic environmental conditions. Ultimately, this review will help guide priority research efforts to understand the risks of UV-filter exposure to coral reef ecosystems. *Integr Environ Assess Manag* 2021;00:1–15. © 2021 The Authors. *Integrated Environmental Assessment and Management* published by Wiley Periodicals LLC on behalf of Society of Environmental Toxicology & Chemistry (SETAC).

KEYWORDS: Coral reefs, Ecotoxicity, Exposure, UV filters

INTRODUCTION

Coral reefs are among the world's most productive and biologically diverse ecosystems (Odum & Odum, 1955; Reaka-Kudla, 1997), performing many important ecological roles and supporting a variety of fish, invertebrate, and algal species (Briones-Fourzán et al., 2012). Shallow warm-water coral reefs cover less than 0.1% of the seabed but support approximately 1–9 million inhabitant species (Fisher et al., 2015; Reaka-Kudla, 1997). Cold-water coral reefs have now been discovered in every ocean, extending to depths of 3000 m and forming vital habitats for thousands of other

species including many that are commercially important (Hoegh-Guldberg et al., 2017). Coral reefs are essential to the livelihoods of many coastal communities, because many benefits can be derived from their ecosystem goods and services, including food supplies, shoreline protection, recreational activity, and financial stability through fisheries and tourism (Cinner et al., 2009; Costanza et al., 2014). Despite their significance to humans, biological diversity, and productivity, warm- and cold-water coral reefs are vulnerable to natural and anthropogenically induced disturbances, both locally and globally (Burke et al., 2011; Hall-Spencer et al., 2002).

Coral reef ecosystems around the world have experienced an unparalleled decline, with more frequent and extended bleaching events (Chen et al., 2015; Hughes et al., 2018). Climate change and elevated ocean temperatures are regarded as the main drivers of global coral decline (Hughes et al., 2018); however, an array of physical, biological, and chemical stressors have been shown to have adverse effects on their distribution, abundance, and survival (e.g.,

Hoegh-Guldberg, 2014; van Dam et al., 2011). Chronic and acute anthropogenic stressors include coastal development, overfishing, destructive activities, and various forms of pollution (Burke et al., 2011; Harvell et al., 2002; Pandolfi et al., 2003). There is growing concern over chemical contaminant threats such as sunscreens and personal care products (PCPs), which may present challenges for reefs in areas of high population density, growing tourism, and recreational activity hotspots (Mitchelmore et al., 2019).

Ultraviolet (UV) filters are a common ingredient of sunscreens and cosmetic products that are added to protect the skin against deleterious UVA, UVB, and UVC light radiation (Mitchelmore et al., 2019; Salvador & Chisvert, 2005). Growing concern over the negative health effects of UV radiation in humans has caused an increase in sunscreen usage and subsequent release into marine ecosystems (Brausch & Rand, 2011; Mitchelmore et al., 2019). Sunscreens have received greater attention as a result of their extensive application, and their ubiquity and pseudo-persistence in the environment raises concerns over potential toxicological impacts on coral reefs (Giokas et al., 2007; Mitchelmore et al., 2019). Traditionally, they have been largely unregulated and, to date, very few studies concerning the distribution, toxicity, and effects of UV filters in the marine environment have been conducted.

Given the current health and state of coral reef ecosystems caused by present climate trends, it is important to consider the additional risks posed by exposure to these chemicals in order to mitigate further degradation. This study will review the current understanding of the exposure, distribution, and effects of UV filters on coral reef ecosystems by (1) assessing the spatial distribution and occurrence of UV filters measured in surface waters in relation to coral reefs; (2) assessing current knowledge of the ecotoxicological risks within the marine environment; (3) identifying major research gaps from the synthesized literature; (4) draw on the expertise of researchers from academia, government, industry, and nonprofit organizations through a survey to prioritize these research gaps. The results of this analysis will help guide future research and regulatory efforts on the impacts of UV filters on marine ecosystems.

METHODS

Systematic literature search

To identify the occurrence and toxicity of UV filters as an emerging marine pollutant, a systematic literature review was conducted using the search engines Google Scholar and Web of Science. Keywords included sunscreen, UV filters, UV chemicals, organic, marine ecosystem, coral reefs, ecotoxicity, occurrence, and distribution; they were used either alone or together in queries. In all, 458 papers were identified, of which 35 occurrence and distribution articles and 14 toxicology articles met the criteria for evaluation. To determine the distribution and occurrence of UV filters in the marine environment, the following information was obtained from each study where possible:

compound name, concentration detected in surface water, and location of sample sites. To determine the toxicity and risk of UV filters to marine coral reef species, the following information was obtained from each study where possible: compound, test organism, species, toxicity endpoint, measurement endpoint, exposure time, and concentration. Measured occurrence data were then compared with toxicity endpoints to determine instances where concentrations were potentially inducing biological stress. This study focuses primarily on organic UV filters and does not include in-depth analysis of the effects of inorganic mineral sunscreen ingredients such as zinc or titanium dioxide (nanoparticles or otherwise).

Prioritization survey development

Relevant literature identified in the systematic search was reviewed and research gaps were identified. The following eight general research areas were identified and are presented in Table 3 (along with 24 specific knowledge gaps): (1) effects of UV-filter exposure on different biotic parameters, (2) effects of UV-filter exposure on different study condition parameters, (3) effects of different spatial and temporal parameters, (4) effects of UV filters when coinciding with additional climate-induced environmental stressors, (5) effects of pulse exposure to marine organisms, (6) effects of whole product/co-exposure testing, (7) effects of UV-filter exposure on ossification of organisms, and (8) effects of UV-filter exposure on organism biological processes.

To prioritize research gaps, expert opinion was sought to rank the importance of specific knowledge gaps. A mixed-mode anonymous online survey approach was developed to collect quantitative and qualitative data to incorporate expert opinion into the ranking of these research gaps using Qualtrics (Provo, Utah, USA). Survey participants and their contact information was extracted from the author lists in subject-relevant papers identified by the systematic literature review. Sixty participants were contacted, and 18 responded. In addition, industry researcher contributions were sought through dissemination of the survey to a relevant trade organization. This European Union General Data Protection Regulation (GDPR) compliant survey was composed of seven closed-ended questions with multiple-choice and rank formatted response options. Closed-ended questions were developed to make quantitative evaluation easier to standardize (Fink, 2002). The identified research gaps were grouped into four categories and presented to survey respondents. Participants were asked to rank the gaps in each group from highest to lowest in order of importance. These categories included "general research gaps" and an additional three specific categories to determine further prioritization, including "spatial and temporal parameters," "study condition parameters," and "biotic parameters." Each position was given a numerical value for which respondents were asked to rank 1–4, 1–5, or 1–8, depending on the number of research gaps in each category, with 1 indicating the highest priority. Average

rankings were calculated for each research gap within each category. Thus, the lowest value indicates the highest priority for research. For example, if all respondents rank the same gap most important, it would have an average score of one. An additional, open-ended response question was included to allow respondents to suggest additional research gaps not identified by the review. Survey questions are provided in the Supporting Information.

UV FILTERS

Economics, regulation, and legislation

Demand for sun care products is expected to increase as marine and coastal tourism continues to rise globally, attracting an estimated 1.56 billion tourists worldwide (Honey & Krantz, 2007). The US Food and Drug Administration (FDA) average recommended amount of sunscreen application for adequate sun protection is 20 g at any one time. However, Giokas et al. (2007) suggest that consumers are likely to apply considerably larger amounts. An estimated 10 000 tons of UV filters are produced annually for the global market, with sunscreen product sales higher than half a billion US dollars in 2005 (Shaath, 2005). Despite the lack of current knowledge of the effects of sunscreen on the marine environment, recent studies have raised the issue for increased regulation (Osterwalder et al., 2014). A study by Sobek et al. (2013) investigated the UV-filter environmental hazard classifications according to the regulation on classification, labeling, and packaging (CLP) of substances and mixtures. In Europe, environmental risk assessments are not required for PCPs (including all UV filters), and the CLP does not include cosmetics in their regulatory process (Sobek et al., 2013). The study found that if all PCPs, including cosmetics were to be included in the regulatory processes, 46% of all currently approved UV filters in the EU would meet the CLP classification as "hazardous to the aquatic environment." In 2015, Hawaiian legislators approved a ban on the commonly used oxybenzone largely because of a study presented by Downs et al. (2016); however, ongoing debate has postponed the date of enforcement (Johnsen, 2018).

Sunscreen chemical composition and their physiochemical properties

Sunscreen or cosmetic products are typically composed of three to eight separate UV filters, and these can be either organic (UV-absorbing chemicals) or inorganic (UV-reflective minerals; Brausch & Rand, 2011; Gago-Ferrero et al., 2012). Commercial sunscreen formulas often comprise a mixture of both organic and inorganic UV filters of up to 20 or more compounds (Danovaro et al., 2008). This is to cover a wider spectrum of protection, because no singular compound is sufficient for human protection at current permitted concentrations (Sánchez-Quiles & Tovar-Sánchez, 2015). Organic filters commonly utilized in sunscreen are derivatives of para-aminobenzoates (PABA), salicylates cinnamates and camphor (absorb UV-B), and benzophenone

and dibenzoylmethane (absorb UV-A; Jallad, 2017). They are classified according to their structure and key physiochemical properties (Table 1), with most exhibiting characteristics typical of priority organic pollutants (Eljarrat & Barcelo, 2003). The presence of an aromatic moiety with a side chain is a collective feature between filters demonstrating different degrees of unsaturation, high lipophilicity, and stability against biotic degradation (Díaz-Cruz et al., 2008; Vila et al., 2017). Organic filters are generally less photostable than inorganic and undergo photolysis with potential transformation into hazardous by-products and harmful free-oxygen radicals that may affect coral reef ecosystem health (Abid et al., 2017; Danovaro et al., 2008; Downs et al., 2016). However, evidence of toxicological effects on marine life is scarce, and further research is required (Nash & Tanner, 2014). All compound abbreviations discussed in this paper are defined in Table 1, along with their physiochemical properties and corresponding structure.
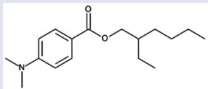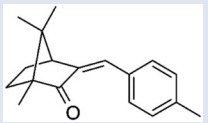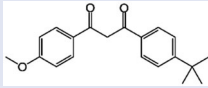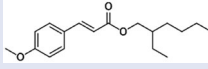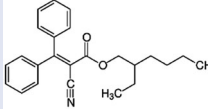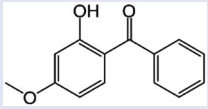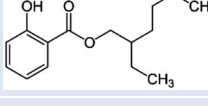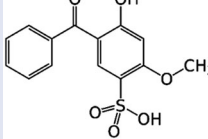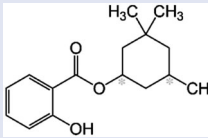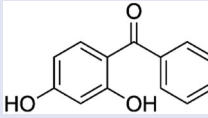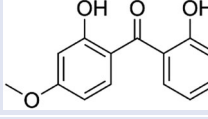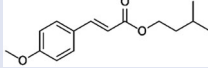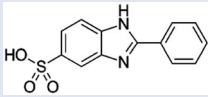
SOURCES, OCCURRENCE, AND DISTRIBUTION OF UV FILTERS IN THE MARINE ENVIRONMENT

Sources

At least 10% of coral reefs worldwide are at risk from UV-filter exposure (Danovaro et al., 2008). Of those, 40% are located along coastal areas and may be at greater risk of exposure, because an estimated 6000 and 14 000 tons of sunscreen lotion containing 1–10% of the UV chemical oxybenzone is released annually into coral reef ecosystems (Danovaro et al., 2008; Downs et al., 2016; Wilkinson & Souter, 2008). Despite a wide reporting of UV filters in the aquatic environment, only a few studies have reported environmental concentrations in seawater surrounding coral reefs located around the world (e.g., Bargar et al., 2015; Downs et al., 2016; Tsui et al., 2017).
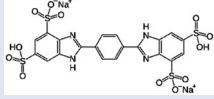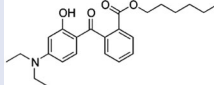
Organic UV filters enter the marine environment via two principle pathways, directly from human skin washed off during recreational actives such as swimming and diving, and indirectly through industrial discharges, wastewater effluents, runoff, and domestic use (Cadena-Aizaga et al., 2020; Cuderman & Heath, 2007). Approximately 25% of sunscreen applied to the skin is not absorbed, and residues are released into surrounding waters through bathing activity or into wastewater systems via shower 20 min post-application (Danovaro et al., 2008). Many of the chemicals that are absorbed are then excreted in urine, where they enter wastewater systems (Calafat et al., 2008; DiNardo & Downs, 2018; Gonzalez et al., 2006). Many UV filters are hydrophobic and are expected to partition into organic sediment and marine organism tissues; however, studies investigating the presence of UV filters in sediments from coral reef locations are scarce and only a few have demonstrated bioaccumulation of chemicals in coral tissues (e.g., Mitchelmore et al., 2019; Tsui et al., 2014b, 2017).

TABLE 1 Structure and physicochemical properties of common organic UV filters found in sunscreen[b]

Abbreviation	Compound	CAS no.	Molecular formula	Molecular weight (g/mol)	pKa	Structure
OD-PABA	2-Ethylhexyl 4-(dimethylamino) benzoate	21245-02-3	$C_{17}H_{27}NO_2$	277.40	2.9	
4-MBC	4-Methylbenzylidene camphor	36861-47-9	$C_{18}H_{22}O$	254.37	–	
BMDBM	Avobenzone	70356-09-1	$C_{20}H_{22}O_3$	310.39	9.74[a]	
OMC	Octinoxate	5466-77-3	$C_{18}H_{26}O_3$	290.41	–	
OC	Octocrylene	6197-30-4	$C_{24}H_{27}NO_2$	361.48	–	
BP-3	Oxybenzone	131-57-7	$C_{14}H_{12}O_3$	228.24	7.1	
EHS	2-Ethylhexyl salicylate	118-60-5	$C_{15}H_{22}O_3$	250.34	8.13[a]	
BP-4	Sulisobenzone	4065-45-6	$C_{14}H_{12}O_6S$	308.31	$pK_a1 = -2.4$ $pK_a2 = 7.6$	
HMS	Homosalate	118-56-9	$C_{16}H_{22}O_3$	262.35	8.09[a]	
BP-1	2,4-Dihydroxybenzo-phenone	92092-63-2	$C_{13}H_{10}O_3$	214.22	$pK_a1 = 7.1$ $pK_a2 = 8.0$	
BP-8	Dioxybenzone	131-53-3	$C_{14}H_{12}O_4$	244.24	6.78	
IMC	Isoamyl 4-methoxycinnamate	71617-10-2	$C_{15}H_{20}O_3$	248.32	–	
PMDSA	Ensulizole	27503-81-7	$C_{13}H_{10}N_2O_3S$	274.30	−0.87[a]	

(Continued)

TABLE 1 (Continued)

Abbreviation	Compound	CAS no.	Molecular formula	Molecular weight (g/mol)	pK_a	Structure
DPDT	Bisdisulizole disodium	180898-37-7	$C_{20}H_{14}N_4Na_2O_{12}S_4$	676.58	-0.27^a	
DHHB	Diethylamino hydroxybenzoyl hexyl benzoate	302776-68-7	$C_{24}H_{31}NO_4$	397.51	7.29^a	

[a]From Cadena-Aizaga et al. (2020).
[b]Data obtained from https://pubchem.ncbi.nlm.nih.gov.

Wastewater treatment plants (WWTPs) are a main source of UV-filter exposure to marine ecosystems (Mitchelmore et al., 2019). UV filters enter the sewage system from showering, clothes laundering, and urination. These compounds are inefficiently removed because of their physiochemical properties (Díaz-Cruz et al., 2008; Li et al., 2007; Schneider & Lim, 2019). Removal rate efficiency can be as low as 28%–43% (Li et al., 2007); however, efficiency has improved with advancements in treatment technologies (Margot et al., 2015). BP-4, BP-3, and many other benzophenones and benzotriazoles have been reported in WWTP effluent across the world (Coronado et al., 2008; Rodil et al., 2008; Zhang et al., 2011), ranging from 19 ng/L of BP-3 in samples from New York City (Coronado et al., 2008) to 1481 ng/L of BP-4 in Spain (Rodil et al., 2008). Ramos et al. (2016) provide an exhaustive list of chemicals detected in wastewater, including UV filters. Identifying the source from a comprehensive mixture of UV-filter-containing products is particularly challenging; however, seasonal fluctuations reported in WWTP and concentrations in coastal waters during summer seasons may indicate the significance of sunscreen use in total UV-filter exposure (Danovaro et al., 2008; Plagellat et al., 2006).

Occurrence and distribution

Various studies have detected different UV-filter concentrations in ocean surface water samples around the world; however, published reports have focused primarily on European countries, China, and the USA. Global detections of UV filters were collated from synthesized literature and plotted along with documented distributions of cold- and warm-water coral reef locations (Figure 1; Burke et al., 2011;
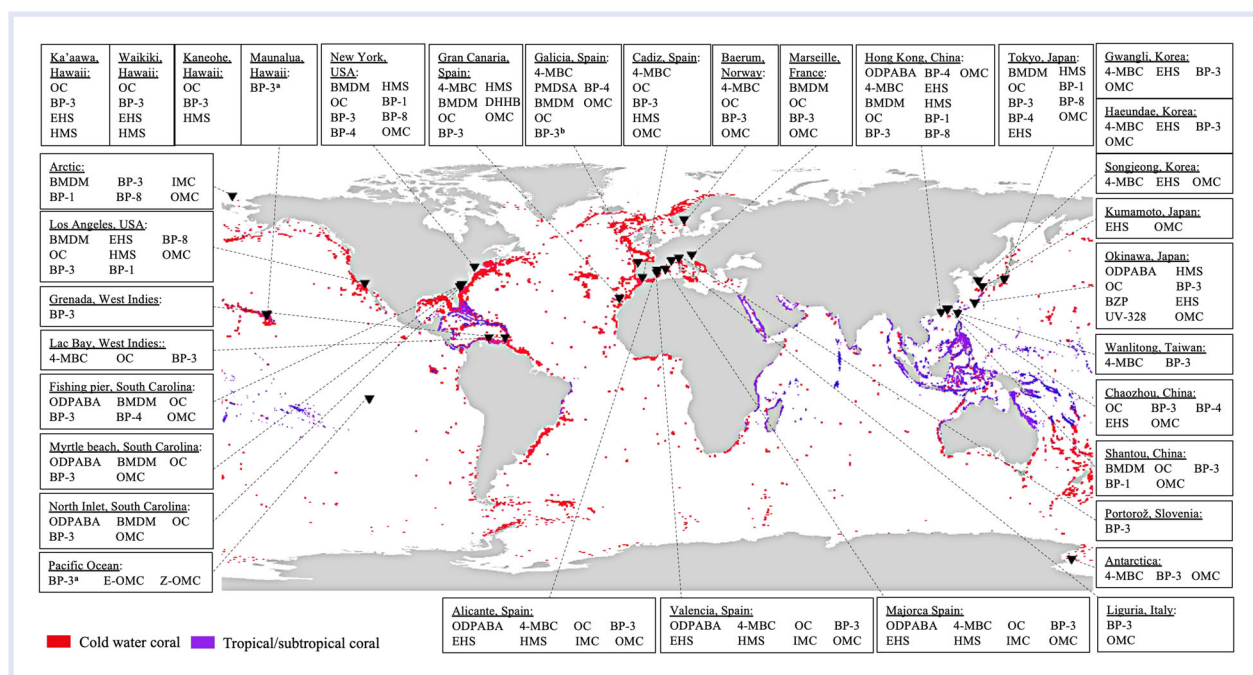
FIGURE 1 Global occurrence of UV filters (max observed concentrations) detected in marine surface water in relation to cold- and warm-water coral reefs. Coral reef locations obtained from available databases (Burke et al., 2011; Freiwald et al., 2017). Triangles indicate approximate sample site and list of UV filter present (see Table S1 for full concentration list and corresponding citations). [a]Exceedance of EC_{50}; [b]Exceedance of LC_{50}

Freiwald et al., 2017). Only the highest concentrations observed at a location were included. For example, Figure 1 and Table S1 show maximum concentrations obtained for Hong Kong by Tsui et al. (2014a). More recently, Tsui et al. (2017, 2019) have reported data with lower observed concentrations for this location. Maximum observed concentrations of the detected compounds are presented by region in the Supporting Information (Figures S1–S6). Compounds covering almost all UV-filter families (Benzophenones, *p*-aminobenzoic acid and derivatives, salicylates, cinnamates, camphor derivates, benzimidazole derivatives, dybenzoyl methane derivatives, and crylenes) were detected across the various locations (Figure 1). BP-3 is the most frequently detected UV filter reported from all sample areas (excluding Kumanmoto, Japan) in concentrations ranging from 6.2 to 692 000 ng/L, followed by OMC (8.8–4043 ng/L) and OC (21.7–30 000 ng/L). The reoccurrence of BP-3 is likely the result of its slow degradation rate compared with others (Santos et al., 2012) and is one of the most frequently utilized UV filters in over-the-counter products, because its use is permitted in all countries (Kim & Choi, 2014; Tarazona et al., 2010).

The highest BP-3 concentration (692 000 ng/L) was found in Galicia, Spain, from a water sample collected at a bathing spot during the summer season (Vila et al., 2016a, 2016b). High concentrations of OC (30 000 ng/L) and BMDM (72 000 ng/L) were also reported from the same water sample (Figures 1 and S2). The second highest concentrations of chemicals observed in the literature were reported in samples at Maunalua Bay (Oahu, Hawaii, USA) and in the Pacific Ocean at 19 200 and 34 310 ng/L BP-3, respectively. Samples from Maunalua Bay were collected at a popular visitor beach receiving more than 500 swimmers a day during peak seasons in June (Downs et al., 2016). According to Burke et al. (2011), Hawaii has approximately 3834 km^2 of coral coverage (Figure 1) and, owing to the isolated nature of the islands, they support many endemic species with uniquely distinct marine assemblages compared with those elsewhere. Downs et al. (2016) also reported a concentration of 1 395 000 ng/L BP-3 detected at Trunk Bay of St. John Island, US Virgin Islands. However, this value is abnormally large and may be an anomaly; a general paucity of data with which to compare complicates the interpretation and significance of a single detection. Therefore, continuous and extensive monitoring within the bay and globally is required to confirm environmental exposure.

High concentrations of UV chemicals such as BP-3 have been detected mostly during summer seasons, emphasizing high direct influxes and increased concentrations of UV chemicals due to the higher capacity of tourists (Schaap & Slijkerman, 2018). Kim et al. (2017) report seasonal concentration increases up to 4.4 times greater than the preholiday period leading up to June and August. The highest recorded concentrations for each compound at each location are reported in Table S1. High concentrations have also been detected in areas such as Hong Kong during cooler off-peak tourism seasons and may be representative of their location's proximity to wastewater effluent (Tsui et al., 2014a). High concentrations of BP-3 (5429 ng/L) have been detected in the Victoria Harbour channel, located near wastewater effluent emissions in Hong Kong during the summer season from June to August (Tsui et al., 2014a). The proximal WWTP handles 70% of the population's discharges (Tsui et al., 2014a). In other areas such as Japan and Korea, concentrations reported are generally lower during winter seasons and indicate seasonal variations in these locations (Kim et al., 2017; Sankoda et al., 2015).

A surprisingly high range of UV-filter concentrations have also been detected in areas of little to no tourist activity, such as Pacific Ocean (three UV filters: 8484–19 200 ng/L) and Arctic (five UV filters: 36–75 ng/L) samples, where recreational wash-off and direct local effluent input are not assumed to be the primary source. There are two possible pathways for the occurrence and distribution of these UV filters: (1) ocean transport via ocean currents, or (2) atmospheric transport, either long or short range (Tsui et al., 2014a). Ineffective wastewater treatment facilities may lead to direct discharge and dissipation of untreated or undertreated wastewater into marine ecosystems via ocean currents (Gunnarsdottir et al., 2013). A study by Rodil et al. (2009) identifies high photostability characteristics of BP-3 and OC against UV irradiation, with half-lives of nearly 72 h. Therefore, it is possible for UV filters such as these to undergo short- and long-range transportation (Rodil et al., 2009).

The relatively small number of monitoring studies shown in Figure 1 highlights the urgent need to monitor environmental occurrence and distribution of UV filters, particularly in tropical and subtropical coral reef regions, because these often attract large numbers of tourists. Ecotoxicological data regarding both UV filters and their degradation products is lacking. However, sunscreen chemicals may remain in the marine environment for decades, even at trace concentrations, and the long-term risks associated with pseudo-persistence are still largely unknown (Maipas & Nicolopoulou-Stamati, 2015). Therefore, their bioaccumulation capability in both organisms and substrata should be determined in addition to surface water occurrence (Johnsen, 2018).

EVIDENCE OF ECOTOXICOLOGICAL RISK

Ecotoxicological assays

There is a paucity of UV-filter toxicity data reported in the literature, with only a few studies including coral species and even fewer assessments on other coral reef inhabitants (Tables 2 and S2). As of 2008, various legislation has permitted international use of 50 organic and inorganic UV filters within commercial sunscreen products, but only approximately 16 have been analyzed in marine toxicity assays (Sánchez-Quiles & Tovar-Sánchez, 2015). General toxicity assays of freshwater organisms are more comprehensive and widely accepted by the scientific community than of marine organisms (Baker et al., 2014). The complex

TABLE 2 Current reported ecotoxicological values (LC$_{50}$, EC$_{10}$, EC$_{20}$, LOEC; [μg/L]) of varying organic UV-filter exposure and endpoints measured on different marine species

Compounds	Test organism	Species	Toxicity endpoints	Measurement endpoint	Exposure time	Conc. (μg/L)	Reference
BP-2	Coral planula	*Stylophora pistillata*	Mortality	LC$_{50}$	24 h	165 (light) 508 (dark)	Downs et al. (2014)
	Coral planula	*S. pistillata*	Deformity	EC$_{50}$	24 h	315 (light) 1.05 (dark)	Downs et al. (2014)
BP-3	Hard coral	*Pocillopora damicornis*	Metabolomic change	Effect reported	7 days	2000	Stien et al. (2020)
	Coral planula	*S. pistillata*	Mortality	LC$_{50}$	24 h	139 (light) 779 (dark)	Downs et al. (2016)
	Coral planula	*S. pistillata*	Deformity	EC$_{50}$	24 h	49 (light) 10.4 (dark)	Downs et al. (2016)
	Sea urchin larvae	*Paracentrotus lividus*	Larval size	EC$_{50}$	48 h	3280	Paredes et al. (2014)
	Mussel	*Mytilus galloprovincialis*	Normal D-larvae	EC$_{50}$	48 h	3472.59	Paredes et al. (2014)
	Hard coral	*Acropora sp.*	Bleaching	Bleaching rate	48 h	2376	Danovaro et al. (2008)
	Hard coral	*Acropora pulchra*	Bleaching	Bleaching rate	96 h	3600	Danovaro et al. (2008)
OMC	Hard coral	*Seriatopora caliendrum*	Mortality	LOEC	7 days	1000	He et al. (2019a)
	Hard coral	*S. caliendrum*	Total polyp reaction	LOEC	7 days	10	He et al. (2019a)
	Hard coral	*P. damicornis*	Total polyp reaction	LOEC	7 days	1000	He et al. (2019a)
	Sea urchin larvae	*P. lividus*	Larval size	EC$_{50}$	48 h	283.69	Paredes et al. (2014)
	Mussel	*Mytilus galloprovincialis*	Normal D-larvae	EC$_{50}$	48 h	3118.18	Paredes et al. (2014)
	Hard coral	*Acropora sp.*	Bleaching	Bleaching rate	24 h	2000	Danovaro et al. (2008)
	Hard coral	*Acropora pulchra*	Bleaching	Bleaching rate	96 h	3030	Danovaro et al. (2008)
OC	Hard coral	*P. damicornis*	Metabolomic change	Effect reported	7 days	50	Stien et al. (2020)
	Hard coral	*P. damicornis*	Mitochondrial function change	Effect reported	7 days	50	Stien et al. (2020)
	Hard coral	*Seriatopora caliendrum*	Total polyp reaction	LOEC	7 days	10 000	He et al. (2019a)
	Hard coral	*P. damicornis*	Total polyp reaction	LOEC	7 days	10 000	He et al. (2019a)
	Hard coral	*P. damicornis*	Polyp retraction	Effect reported	7 days	300	Stien et al. (2019)
	Hard coral	*P. damicornis*	Metabolomic change	Effect reported	7 days	50	Stien et al. (2019)
	Mussel	*M. galloprovinciali*	Embryogenesis	EC$_{10}$	48 h	162	Giraldo et al. (2017)
	Sea urchin larvae	*P. lividus*	Growth rate	EC$_{10}$	48 h	511	Giraldo et al. (2017)

(*Continued*)

TABLE 2 (Continued)

Compounds	Test organism	Species	Toxicity endpoints	Measurement endpoint	Exposure time	Conc. (µg/L)	Reference
BP-8	Coral nubbin	*Seriatopora caliendrum*	Mortality	LOEC	14 days	100	He et al. (2019b)
	Coral planula	*S. caliendrum*	Mortality	LOEC	14 days	500	He et al. (2019b)
	Coral planula	*S. caliendrum*	Bleaching	LOEC	14 days	250	He et al. (2019b)
BP-4	Sea urchin larvae	*P. lividus*	Larva size	EC$_{50}$	48 h	<10 000	Paredes et al. (2014)
	Mussel	*M. galloprovincialis*	Normal D-larvae	EC$_{50}$	48 h	>10 000	Paredes et al. (2014)
4-MBC	Sea urchin larvae	*P. lividus*	Larval size	EC$_{50}$	48 h	853.74	Paredes et al. (2014)
	Mussel	*M. galloprovincialis*	Normal D-larvae	EC$_{50}$	48 h	587.17	Paredes et al. (2014)
	Flatfish larvae	*Solea senegalensis*	Mortality	EC$_{50}$	96 h	439	Araújo et al. (2018)
	Hard coral	*Acropora sp.*	Bleaching	Bleaching rate	48 h	1053	Danovaro et al. (2008)
	Hard coral	*Acropora pulchra*	Bleaching	Bleaching rate	62 h	1596	Danovaro et al. (2008)

Abbreviations: LC50, lethal concentration required to kill 50% of the population; EC$_{10}$/EC$_{50}$, effective concentrations, which adversely affect 10% and 50% of the population; LOEC, lowest observed effect concentration.

nature of salt water makes marine toxicity studies more complex, and determining the thresholds for particular UV filters in a diverse range of marine organisms presents many challenges (Baker et al., 2014).

Physiochemical and molecular stress responses in corals under UV chemical exposure can be expressed through various mechanisms, depending on the test species and their life stage (Morgan et al., 2001). All reef organisms vary substantially in their sensitivity and response to individual chemicals (Morgan et al., 2001). Sensitivity distribution is a crucial concept for ecotoxicological risk assessments to predict a species community composition and response to chemical stress, and to determine the probable community restoration success (Posthuma et al., 2001; van Woesik et al., 2012). Three main factors determine the toxic threshold of a chemical: the test chemical's structure, the amount absorbed by the target organism, and the organism's ability to depurate or detoxify the chemical (Johnsen, 2018). Coral structures' sensitivity to UV-filter chemicals is likely the result of their thin, lipid-dense, outer layer of tissue (approximately 11 µm) that lines the calcium carbonate skeleton, promoting uptake of lipophilic UV filters (Peters et al., 1997).

Toxicity assays developed for UV filters have investigated various toxicological endpoints, including expulsion of symbiotic zooxanthellae and mucous production, embryogenesis, functional and structural cell failure and necrosis, larval settlement inhibition, and endocrine disruption (Tables 2 and S2; e.g., Danovaro et al., 2008; Downs et al., 2014). UV-filter toxicity values median lethal concentration (LC$_{50}$) and effect concentrations (EC$_{50}$, EC$_{10}$) identified from the literature review are displayed in Table 2, and no observed effect concentration values (NOECs) for various compounds are presented in Table S2. Tables 2 and S2 highlight available ecotoxicological data specific to marine and coral reef ecosystems in peer-reviewed literature. The high concentrations of BP-3 (692 000 ng/L) observed in Galicia, Spain (Figures 1 and S2), exceed lethal toxicity concentrations (LC$_{50}$) established for coral planula larvae (Table 2) as reported by Downs et al. (2016), suggesting the potential for coral mortality. From the same water sample, significant OC (30 000 ng/L) and BMDM (72 000 ng/L) concentrations do not exceed any lethal toxicity thresholds assessed for corals (Table 2). However, these OC values do exceed NOEC values reported for the sea urchin (*Paracentrotus lividus*) larval-growth rate and mussel (*Mytilus galloprovincial*) embryogenesis, which are likely in this area (Table S2).

Few studies have investigated the ecotoxicological effects of UV-filter exposure, with only a handful testing a small range of coral species. In acute-toxicity, laboratory-based studies, Scleractinian coral have demonstrated substantial coral bleaching (loss of symbiotic algae from host coral; Danovaro et al., 2008) and expulsion of zooxanthellae in both coral planulae and mature fragments (Downs et al., 2014, 2016; He et al., 2019a) when exposed to varying concentrations of BP-2, BP-3, OC, OMC, and BP-8. Typical

toxicity thresholds of effect used in risk assessments have been determined only for BP-3 and BP-2 (Mitchelmore et al., 2019). For example, Downs et al. (2016) reported thresholds for mortality (LC_{50}; 139 μg/L) and deformity (EC_{50}; 10.4 μg/L) of *Stylophora pistillata* coral planulae after 24 h of exposure to BP-3 and in vitro coral cell mortality (LC_{50}; 8–340 μg/L) after 4 h. However, it should be noted that the exposure concentrations were not analytically verified (Mitchelmore et al., 2019).

Concentrations of BP-3 measured within Maunalua Bay, Hawaii (19.2 μg/L), exceed EC_{50} toxicity values recorded when exposed under dark conditions (10.4 μg/L) but are below the EC_{50} determined for daylight conditions (49 μg/L). BP-3's phototoxicant properties cause the induction of different toxicities to coral planulae when exposed to the UV filter under light and dark conditions (Downs et al., 2016). Many significant biological processes, such as reproduction of certain coral species, occur during the night. For example, many reef-building corals typically brood and release planulae or spawn gametes nocturnally (Boch et al., 2011; Gleason & Hofmann, 2011). Conversely, significantly lower concentrations of BP-3 under light conditions were observed to induce 50% mortality (LC_{50}) compared with dark conditions for *Stylophora pistillata* coral planula (Table 2). Planula larvae of broadcast spawners are positively buoyant and reside on the water column's surface for 2–4 days before settlement (e.g., Fadlallah, 1983; Shlesinger & Loya, 1985). Natural environmental light conditions in tropical regions can reach levels five times higher than those replicated in some laboratory tests and may magnify phototoxicities of certain UV filters (Downs et al., 2016). Therefore, it is likely that planula larvae may be more at risk from both exposure toxicities from UV filters and diurnal differences, which should be considered when evaluating toxicity in future assays.

Danovaro and Corinaldesi (2003) investigated the effects of sunscreen compounds on marine ecosystems and observed increased viral production in marine bacterioplankton via prophage induction when exposed to UV filters. Further development in this research included field experiments (Celebes Sea, the Caribbean Sea, the Andaman Sea, and the Red Sea), using plastic bags containing coral (*Acropora* spp.), sunscreen and/or active ingredients (Danovaro et al., 2008). This work has demonstrated rapid and complete coral bleaching caused by viral infections at low concentrations of BP-3, OMC, and 4-MBC (Table 2). The loss of coral intracellular endosymbionts caused by expulsion under UV-filter exposure has a detrimental impact on the biodiversity and functioning of coral reef ecosystems (Gago-Ferrero et al., 2012).

Ecotoxicological assessments of species that are representative of the marine ecosystem structure and functionality, other than coral species, are sparse. This includes ecologically important or indigenous species, with only a few reporting the effects of different UV filters. It is important to include these species within toxicological assays, because they play important roles in maintaining ecosystem functionality (Tsui et al., 2017). A study by Paredes et al. (2014) presents effective concentrations (EC_{50}) of BP-4, 4-MBC, and OMC to inhibit the growth of sea urchin (*P. lividus*) larvae and deform mussel (*M. galloprovincialis*) larvae (Table 2). Their methodologies, however, fail to measure cellular stress responses. The effects of acute BP-3 exposure to fish are thought to be similar to those of mammals, causing endocrine disruption by modulating estrogen receptor signaling pathways, inducing reproductive pathologies, and thus reducing reproductive fitness (Downs et al., 2016). Chronic exposure reduces egg production and embryo hatching, and promotes production of vitellogenin proteins in males, suggesting the potential for shifts in gender (Coronado et al., 2008). With just a handful of studies of coral and other marine species, further work to understand the impact of UV-filter exposure is imperative.

The data presented in Table 2 demonstrates the general paucity of marine toxicological data available for chemical UV filters. To extrapolate these data for use in a species sensitivity distribution, or other statistical extrapolation protocols, to derive predicted no-effect concentrations for regulation, additional data would be required. For example, the European Registration, Evaluation, Authorization and Restriction of Chemicals (REACH) regulations require a minimum of 10 NOECs for different species covering at least eight taxonomic groups, with species ecologically relevant to the marine environment (European Chemicals Agency [ECHA], 2010). Based on available data (Table 2), this could not be done for any of the UV-chemical compounds.

Limitations

Existing toxicity assessment methods performed in laboratories may be insufficient owing to the variability of external ecosystem parameters, such as light levels and salinity (Johnsen, 2018). It has been argued that, although controlled ex situ conditions are convenient, they are unable to accurately replicate current environmental conditions or predict toxicity thresholds for coral reef populations (Chapman, 2007). Realistic field investigations need to be applied to better understand the interactions between marine organisms and surrounding UV-filter contaminants. Additional compounds concomitantly added with UV filters in sunscreens have not been measured in ecotoxicological studies or in conjunction with active ingredients (He et al., 2019b). Toxicity assays evaluated within this review were typically performed on a single UV compound and do not realistically represent the ecological risks of complex mixtures used to achieve high sun protection factors (SPF) for sunscreen (Gago-Ferrero et al., 2012). Few studies have reported significant synergistic effects of mixed UV-filter combinations at NOECs of the individual chemicals (e.g., Brian et al., 2005; Fent et al., 2008; Kunz & Fent, 2006, 2009). Therefore, future toxicity studies of sunscreen UV filters on marine species should not focus solely on the UV-active ingredients, but rather on whole-product assessment and mixture toxicity.

TABLE 3 Summary of knowledge gaps on the effects of UV-filter exposure in the marine environment and additional issues not presented in the survey identified by respondents

Description	Knowledge gap
1. Effects of UV-filter exposure on different biotic parameters	1. Coral species type (soft, hard)
	2. Life-cycle stage of test species (larvae, adult structure)
	3. Planulae larval production (brooding, broadcast spawning, settled, swimming)
	4. Test species origin (farmed or wild)
	5. Structural sample regions of coral polyp (stalk, tip)
	6. Ecosystem structure reflective test species (ecologically important species, indigenous species)
2. Effects of UV-filter exposure on different study condition parameters	7. Natural vs. artificial sea water
	8. Natural vs. artificial sunlight
	9. Diurnal differences (light or dark)
	10. Functional complexity of ecosystem mesocosms 11. In situ studies
3. Effects of different spatial and temporal parameters	12. Seasonal variation (wet and dry)
	13. Depth in the water column at which test species is harvested
	14. Occurrence and distribution in surface water of UV filters with regards to geographical location
	15. Water conditions (wastewater, coastal recreational activities, and low water renewal)
	16. Chronic exposure (long-term monitoring)
	17. Accumulation rate in ocean food chains/trophic transfer
4. Effects of UV filters when coinciding with additional climate-induced environmental stressors	18. Temperature, pH, salinity, ocean acidification
5. Effects of pulse exposure to marine organisms	19. Ability to recover and build resilience to previous exposures (physiological acclimation)
6. Effects of whole product/co-exposure testing	20. Ecotoxicity of UV-filter degradation products and metabolites
	21. Mixture toxicity of UV filters and other concomitant chemical exposure (inorganic, organic, additional sunscreen products)
	22. Bioavailability boosting of UV filters from other sunscreen ingredients
7. Effects of UV-filter exposure on ossification of organisms	23. Coral species (and other calcium structured organisms) skeletal formation under acute and chronic exposure of UV-filter chemicals
8. Effects of UV-filter exposure on organism biological processes	24. Metabolic capabilities, viral infection rates oxidative stress, endocrine disruption processes
Gaps identified by survey respondents	25. Potential significant contributors of UV filters to marine environment other than sunscreen (packaging, plastic, textiles, fishing equiptment, paints, coatings, etc.)
	26. Appropriate population endpoints for coral species in laboratory environments
	27. Optimal exposure conditions for corals in laboratory environments
	28. Cost–benefit analysis or socioeconomic analysis of UV-filter removal from the environment and using alternatives for UV protection (hats, protective clothing, etc.)
	29. Increasing risks of coral disease
	30. Photosynthesis-important light levels for wild corals

TABLE 4 Weighted average ranking scores of general research gaps and, spatial and temporal parameters, study condition parameters and biotic parameter research gaps

Research gap	Average rank	
General	Effects of spatial and temporal parameters of UV-filter chemical exposure on coral reef ecosystems	4.57
	Realistic test conditions (representative species, environment, abiotic factors) of coral reef ecosystems	3.79
	Biotic parameters (e.g., coral type, life-cycle stage) appropriate to evaluating risk in actual reef ecosystems	5.14
	Impacts of co-stressors for example, temperature, pH, salinity, ocean acidification on UV-filter toxicity	3.21
	Long-term consequences of UV-filter exposure to coral reef ecosystem species and impacts on recovery	3.43
	Ecotoxicological effects of UV-filter degradation products and metabolites on coral reef ecosystems	3.93
	Mixture toxicity of UV filters and other concomitant chemical exposure	5.50
	Coral species (and other calcium structured organisms) skeletal formation under acute and chronic exposure of UV-filter chemicals	6.43
Spatial and temporal	Occurrence and distribution in surface water of UV filters with regards to geographical location	1.93
	Exposure time (acute, chronic)	2.14
	Water conditions (wastewater, area of high recreational activities)	2.64
	Seasonal variation (wet and dry)	4.00
	Depth in the water column at which we harvest test species	4.28
Study condition	Functional complexity of ecosystem mesocosms	1.92
	Diurnal differences in UV-filter exposure	2.54
	Natural vs. artificial sunlight	2.69
	Natural vs. artificial seawater	2.85
Biotic	Life-cycle stage (larvae, adult structure)	1.77
	Planulae larvae reproduction (brooding, broadcast spawning, settled, swimming)	2.69
	Coral type (soft, hard)	2.85
	Structural sample regions of coral polyp (stalk, tip)	3.61
	Farmed or wild individuals	4.07

Note: A low score indicates the highest priority for research.

PRIORITIZATION OF RESEARCH GAPS

The current understanding and data available are insufficient to fully determine the distribution and ecotoxicological risks of UV-filter exposure in coral reef ecosystems. The lack of occurrence and toxicity data for most UV filters and metabolites hinders reliable and integral environmental risk assessments to aid comprehensive reef ecosystem protection (Gago-Ferrero et al., 2012). The 24 specific knowledge gaps revealed in this review can be grouped in eight general research areas and are presented in Table 3. The number of knowledge gaps highlights the currently limited knowledge of this topic.

To help focus future research efforts, we developed an anonymous survey and distributed it to experts in the field of marine UV-filter exposure, as identified by author contributions to papers that are reviewed herein and industry

representatives via relevant trade organizations. Of the 18 respondents, approximately 42% were from academia, 15.7% from nonprofit, 26.3% from industry and 15.7% were from governmental affiliations. Of these, 55% are currently involved in active research in this area. Most (72.2%) felt that both the ecotoxicological risks and exposure of UV filters to coral reef ecosystems are not well understood.

The identified research gaps were grouped into four categories and presented to survey respondents. Participants were asked to rank the gaps within each group based on their perceived order of importance. These categories included general research gaps, spatial and temporal parameters, study conditions, and biotic parameters. In keeping with the EU GDPR, respondents could leave any questions unanswered or stop the questionnaire altogether at any point.

When asked to rank the eight general research gaps in order of importance, four areas emerged as high priority, with a weighted average ranking less than 4 (Table 4): (1) effects of UV-filter toxicity when coinciding with additional climate-induced environmental stressors (Temperature, pH, salinity, ocean acidification), (2) long-term consequences of UV-filter exposure for corals and the impacts on recovery, (3) realistic test conditions (e.g., species, environment, abiotic factors) of coral reef ecosystems, and (4) the ecotoxicological effects of UV-filter degradation products and metabolites. That these four research areas received an average ranking ranging from 3.21 to 3.93 indicates that there was little consensus between the priority of these general research gaps, with each of these thought to be high priority by different respondents. Of the respondents, 66% from governmental and regulatory affiliations believed it was most important to prioritize research into the long-term consequences and recovery impacts of coral, whereas 50% of those from industry affiliations ranked realistic test conditions of reef ecosystems to be of greatest importance. Respondents from nonprofit backgrounds equally ranked the effects of UV-filter toxicity with environmental co-stressors and the ecotoxicological effects of UV-filter degradation and metabolites to be most important. Finally, those from academic affiliations expressed mixed priorities across most general areas.

Participants were then asked to rank specific research gaps within three research areas (Biotic Parameters, Study Conditions, Spatio-Temporal Parameters); the results are summarized in Table 4. Respondents felt that the occurrence and distribution in surface water of UV filters with regard to geographical location were of highest priority (1.93) from a spatio-temporal standpoint (Table 4). This was followed by UV-filter exposure time and water conditions according to location (i.e., proximity to wastewater emissions and areas of extensive recreational activity). A large percentage of respondents across all affiliations ranked occurrence and distribution highly; however, 60% of those in academia ranked exposure time to be of highest priority. Weighted average rankings of study conditions were all very narrow, indicating that respondents believe

that all parameters are of equal importance (Table 4). All (100%) of the respondents from government and regulatory affiliations and 50% from nonprofit and industry ranked the functional complexity of mesocosms most important when carrying out future studies. Respondents of academic affiliations expressed mixed priorities over all study conditions, supporting the need for priority research in all areas.

Respondents strongly agreed that the life-cycle stage of individuals (e.g., larval, adult structure) was of highest priority (1.77) with regards to biotic parameters when assessing the toxicological effects of UV filters. This was followed by reproductive strategy (2.69) and coral type (2.85; Table 4). All (100%) of the respondents from nonprofit research affiliations ranked the life-cycle stage as the highest priority. Half (50%) of those from governmental and regulatory affiliations and 40% of those from academia agreed; however, 50% from industry and the other 50% from governmental and regulatory affiliations ranked coral type as the highest priority.

Respondents were then given the opportunity to provide additional research gaps they felt were missing from those discussed in this review. These additional research gaps, as well as the full list of current major knowledge gaps in the effects of UV-filter exposure in the marine environment identified through this systematic literature search, are provided in Table 3, gaps 25–30.

CONCLUSIONS

Growing concern over the effects of UV solar radiation on human health has led to extensive use of PCPs containing UV-filter compounds, including sunscreen for skin protection. Because these compounds can enter the environment at the points of manufacture and use, understanding their potential impact on sensitive marine ecosystems, such as coral reefs, is increasingly important. Because the products that use these compounds have significant implications for human health, clear scientific evidence must back any action to restrict the use of specific compounds or products. Given the declining status of coral reef ecosystems and the many stressors they already face, it is important to identify the potential occurrence and toxicological risks associated with UV-filter exposure to reef ecosystems. The systematic literature review demonstrates that our current understanding and extent of knowledge of these issues are limited, and extensive research is required. Singular UV-filter compounds have demonstrated toxic effects on marine organisms; however, exposure assessments are often acute and simplistic, lacking important biotic, spatial, temporal, and environmental condition parameters. Additionally, environmental conditions may either increase or decrease the response of an organism to toxicants, making it difficult to establish the true toxicological risks. Concerning levels of UV filters in surface waters have been detected, but robust monitoring studies are rare. There is an urgent need for substantial long-term environmental occurrence and

distribution monitoring, particularly in tropical and sub-tropical coral reef areas.

Although toxicity data for UV-filter compounds exist, most research has focused on freshwater organisms and ecosystems. The unique ecology and biochemistry of the marine environment, and coral reefs in particular, make translation of this into regulation dubious. Therefore, ecotoxicological testing on coral reef biota representing multiple taxa is needed. Although vertebrate testing for PCPs is prohibited in most cases, it may become necessary to complete a relevant environmental risk assessment.

Experts and industry representatives in the field of marine UV-filter exposure support the call for priority research in these particular areas. Results from the expert survey highlight four general areas of highest priority: (1) the need for a better understanding of UV-filter toxicity to coral reefs when coinciding with additional climate-induced environmental stressors; (2) that future research should focus on the long-term consequences of UV-filter exposure to coral reefs and their inhabitants, including characterizing their potential to recover; (3) toxicity assessments should represent realistic exposure conditions including the functional complexity of ecosystems; (4) there is a need to understand the toxicological effects of UV-filter degradation products and metabolites on coral reefs. The lack of occurrence and toxicity data for most UV filters and their metabolites hinders reliable and integral environmental risk assessments to support comprehensive reef ecosystem protection. Based on the literature, there are not enough marine species endpoints to determine a statistically extrapolated, safe limit for any of the organic UV-filter compounds in the marine environment.

Research addressing the priority research areas identified in this study will allow regulators and policy makers to improve conservation and management policies that ensure the safe use of UV filters to promote human health, while reducing their impact on coral reef ecosystems.

DATA AVAILABILITY STATEMENT

Data are available upon request from corresponding author J Brett Sallach (brett.sallach@york.ac.uk).

SUPPORTING INFORMATION

FIGURE S1. Occurrence of UV filters (max observed concentrations) in marine seawater sample sites of the Pacific Ocean. *Y*-axis are in ng/L. Yellow bars indicate concentrations exceeding lowest reported BP-3 toxicity value (EC_{50} [Red]).

FIGURE S2. Occurrence of UV filters (max observed concentrations) in marine seawater sample sites of Spain. *Y*-axis are in ng/L. Yellow bars indicate concentration exceeding lowest reported toxicity values (LC_{50} [Black] and EC_{50} [Red]; BP-3).

FIGURE S3. Occurrence of UV filters (max observed concentrations) in marine seawater sample sites of Europe (excluding Spain). *Y*-axis are in ng/L and line of lowest reported toxicity values (EC_{50} [BP-3]).

FIGURE S4. Occurrence of UV filters (max observed concentrations) in marine seawater sample sites of the USA. *Y*-axis are in ng/L and line of lowest reported toxicity values (EC_{50} [BP-3]).

FIGURE S5. Occurrence of UV filters (max observed concentrations) in marine seawater sample sites of East Asia. *Y*-axis are in ng/L and line of lowest reported toxicity values (EC_{50} [BP-3]).

FIGURE S6. Occurrence of UV filters (max observed concentrations) in marine seawater sample sites of (a) the West Indies and (b) Polar regions. *Y*-axis are in ng/L and line of lowest reported toxicity values (EC_{50} [BP-3]).

TABLE S1. Max observed concentrations (ng/L) of UV filters in marine seawater samples worldwide.

TABLE S2. Current reported No Observed Effect Concentration (NOEC) values (µg/L) of organic UV filters for marine organisms.

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