Beach showers as sources of contamination for sunscreen pollution in marine protected areas and areas of intensive beach tourism in Hawaii, U.S.A.

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Highlights

- Beach showers are a point-source of contamination to coastal surface waters
- Sunscreen contamination was highest at beaches with the highest visitation rates
- Concentrations of UV sunscreens pose a threat to terrestrial and aquatic receptors
- Apply mitigation options to reduce sunscreen pollution from beach showers

Keywords: oxybenzone, benzophenone-3, octocrylene, risk assessment, coral reef, octisalate, octinoxate,

ABSTRACT

In 2019, sands in nearby runoff streams from public beach showers were sampled on three islands in the State of Hawaii and tested for over 18 different petrochemical UV filters. Beach sands that are directly in the plume discharge of beach showers on three of the islands of Hawaii (Maui, Oahu, Hawai'i) were found to be contaminated with a wide array of petrochemical-based UV-filters that are found in sunscreens. Sands from beach showers across all three islands had a mean concentration of 5,619 ng/g of oxybenzone with the highest concentration of 34,518 ng/g of oxybenzone at a beach shower in the Waikiki area of Honolulu. Octocrylene was detected at a majority of the beach shower locations, with a mean concentration of 296.3 ng/g across 13 sampling sites with the highest concentration of 1,075 ng/g at the beach shower in Waikiki. Avobenzone, octinoxate, 4-methylbenzylidene camphor and benzophenone-2 were detected, as well as breakdown products of oxybenzone, including benzophenone-1, 2,2'-dihydroxy-4-methoxybenzophenone, and 4-hydroxybenzophenone. Dioxybenzone (DHMB) presented the highest concentration in water (75.4 ng/mL), whereas octocrylene was detected in all water samples. Some of these same target analytes were detected in water samples on coral reefs that are adjacent to the beach showers. Risk assessments for both sand and water samples at a majority of the sampling sites had a Risk Quotient >1, indicating that these chemicals could pose a serious threat to beach zones and coral reef habitats. There are almost a dozen mitigation options that could be employed to quickly reduce contaminant loads associated with discharges from these beach showers, like those currently being employed (post-study sampling and analysis) in the State of Hawaii, including banning the use of sunscreens using petrochemical-based UV filters or educating tourists before they arrive on the beach.

1. Introduction

Mass tourism has created a demographic phenomenon that congregates a high density of people into a relatively small and confined space (Chong, 2020; Dodd and Butler, 2019; Dodds and Butler, 2022). For example, locations such as Hanauma Bay (Hawaii, USA), Maya Bay (Krabi, Thailand), La Concha Bay (Biscay, Spain), Playa Delfines (Cancun, Mexico), and Trunk Bay (U.S. Virgin Islands) can see between 2,000 to 15,000 people per day (Downs et al., 2011; Castillo-Pavón and Mendez-Ramirez, 2017; Downs et al., 2021; Kainthola et al., 2022). This demographic density, especially in a natural resource setting, can often be destructive to nearby wildlife habitats (Downs et al., 2022; Guabiroba et al., 2022). One contributing factor to the declining ecological integrity of aquatic ecosystems is sunscreen pollution (Casas-Beltrán et al., 2021; Downs et al., 2022). Some locations can easily receive hundreds of thousands to millions of visitors a year resulting in sunscreen discharges directly from swimmers into the water (Downs et al., 2016; Downs et al., 2021a). It is estimated that a thousand visitors on a beach using sunscreen following the recommended product instructions with a 50% sunscreen-shedding rate from swimmers could deposit more than 36 kilograms/day of sunscreen into the aquatic environment (**Supplemental Table 1**; U.S. CDC; Diffey, 2001; Heerfordt et al., 2018; Cancer Council 2022). Sunscreen pollution, as a consequence of unmanaged tourism, can pose potential threats to the integrity of near-shore habitats and ecosystems that are in proximity to these popular tourist sites (NOAA, 2002).

Some of the most common petrochemical sunscreen UV-filters include avobenzone, oxybenzone, octinoxate, octocrylene, octisalate and homosalate, and can induce toxicities to wildlife at environmentally relevant concentrations (Carve et al., 2021; Downs et al., 2021). Oxybenzone, being the most studied, can induce a range of reproductive and early-stage developmental toxicities in fish and invertebrates based on mechanisms of endocrine disruption, genotoxicity, inhibition of cell migration, and cell death (Schlumpf et al., 2004; Schlumpf et al., 2008; Downs et al., 2016; Wnuk et al., 2018; Xu et al., 2021). Oxybenzone is also toxic to algae and plants, acting as a multi-functional herbicide, inhibiting aspects of both photosynthesis and mitochondrial oxidative phosphorylation (Mao et al., 2017; Zhong et al., 2019a, 2019b; Zhong et al., 2020). Octocrylene is a developmental and metabolic disruptor (*i.e.*, obesogen) in species ranging from corals and fish to mammals, and can inhibit the Calvin Cycle of photosynthesis, as well as oxidative phosphorylation in plant mitochondria (Stien et al., 2019; Zhong et al., 2020; Ko et al., 2022). Furthermore, octocrylene is known to degrade into benzophenone, a highly regulated chemical that is an endocrine disruptor, a carcinogen and a patented herbicide (Downs et al., 2021b; Foubert et al., 2021). Homosalate and octisalate are derivatives of salicylic acid, and imparts a level of genotoxicity, teratogenicity, carcinogenic proliferation and inhibits intracellular signaling pathways (Alamer and Darbre, 2018; Yang et al., 2018; Yazar and Ertekin, 2019; Thorel et al., 2020; DiNardo and Downs, 2021). Octinoxate also exhibits toxicities to algae and plants, as well as thyroid disruption, disruption to sexual maturation, and

toxicities across multi-generational exposures in fish and invertebrates (Inui et al., 2003; Lee et al., 2019; Zhong et al., 2020; Chu et al., 2021; Tian et al., 2021). These UV filters pose not just a threat by themselves, but may also act additively or synergistically with global-level pollutants, such as climate change factors and marine plastic debris (Wijgerde et al., 2020; Song et al., 2021; Liu et al., 2022; Downs et al., 2022). The environmental contamination of these UV filters is past the threshold of emerging concern, as a preponderance of scientific evidence has allowed these chemicals to be jurisdictionally regulated as a means of mitigating their pollution (Downs et al., 2022).

These petrochemical UV-filters are known to contaminate receiving waters from alpine lakes, river systems (including the respective shallow aquifers), pan-tropical coral reefs, coastal waters in both polar regions, as well as municipal potable water sources (Balmer et al., 2005; Tsui et al., 2014; Díaz-Cruz and Barceló, 2015; Emnet et al., 2015; Claudia and Magrini, 2017; Mandaric et al., 2017; Diaz-Cruz et al., 2019; Dominguez-Morueco et al., 2020). The most obvious direct source of sunscreen pollution comes from recreational water activities such as swimming, where sunscreen products applied to the skin are shed as a result of sweating and submersion (Stokes and Diffey, 1999; Poh Agin, 2005; Sivamani et al., 2010; Puccetti and Fares, 2014; O'Malley et al., 2021). Non-point sources of sunscreen pollution can come from sewage discharges, municipal wastewater treated effluents, as well as from cesspits and septic discharges (Balmer et al., 2005; Li et al., 2007; Gago-Ferrero et al., 2011b; Cabeza et al., 2012; Bratkovics et al., 2015; Molins-Delgado et al., 2017; Wang and Kannan, 2017; He et al., 2019).

A recently recognized source of sunscreen petrochemical pollution is found associated with agriculture; reclaimed water from wastewater treatment effluent and sludge biosolids are used in irrigated agriculture and as soil/fertilizer amendments to commercial agricultural settings (Plagellat et al., 2006; Zhang et al., 2011; Eljarrat et al., 2012; Jurado et al., 2014; Sunyer-Caldú and Diaz-Cruz, 2021; Cadena-Aizaga et al., 2022). This point-source of pollution can have adverse impacts to crop yields, contaminate crops with these petrochemical UV-filters, and be associated with agricultural runoff within a watershed, ultimately discharging into groundwater, and freshwater and marine receiving-waters (Loraine and Pettigrove, 2006; Molins-Delgado et al., 2016; Serra-Roig et al., 2016; Cabrera-Peralta and Pena-Alvarez, 2018; Glover et al., 2021; Ramos et al., 2021; Bigott et al., 2022). Plastic aquatic debris are both a point-source for some UV-filter pollution (e.g., oxybenzone, benzotriazole, benzophenone-8) as well as a concentrator of petrochemical UV-filters; UV-filters adhere to the plastic particle surface, increasing the concentration of UV-filter-exposure if consumed by aquatic wildlife (Rani et al., 2017; Hahladakis et al., 2018; O'Donovan et al., 2020; Na et al., 2021; Santa-Viera et al., 2021; Cui et al., 2022). Aerosol/atmospheric distribution and deposition is a newly recognized source, coming directly from the use of aerosol sunscreens, volatilization from waste-water facilities, and through aerosolization of wave action along the shoreline (Wan et al., 2015; Shoeib et al., 2016; Afshar-Mohajer et al., 2018; Pegoraro et al., 2020; Du et al., 2022).

In many tourist-beach locations, both public and private, a freshwater shower will be available on the beach to allow tourists to rinse off sand and seawater from their bodies after swimming or sunbathing. A recent study found that the sands around the beach showers of Hanauma Bay, Hawaii, U.S.A. were highly contaminated with petrochemical sunscreen residues (Downs et al., 2021a). The showers at Hanauma Bay were not plumbed to a municipal wastewater treatment system, but were discharged directly into the shallow ground or the effluent would pool and create runoff rivulets that ran directly through the shoreline to the water's edge. This is a case of a shower being a point-source of pollution for sunscreen contaminants; sunscreen residues on the skin can be washed off in this shower, with or without soap or shampoo, which can also contain petrochemical sunscreen filters, such as oxybenzone and octocrylene (**Supplemental Figure 2**). Beach showers are global amenities; they are found abundantly on the public beaches along the Mediterranean, the Caribbean and Central American coastlines, all through the South Pacific and Oceana region, as well as along the multiple coastlines of the United States, including on most of the islands of Hawaii. Boat showers can also be sources of sunscreen contamination; many recreational vessels have freshwater shower amenities that discharge directly into receiving waters.

The first objective of this study is to determine if other beach showers within the Hawaiian Islands are also point-sources of sunscreen contamination. The second objective is to examine in a single case study whether sunscreen contamination occurs near boat moorings that use on-board showers. The third objective of this study is to determine whether the concentrations of these petrochemical sunscreens pose a threat to terrestrial and aquatic wildlife at these sampling locations. Finally, resource managers, conservation specialists, community leaders, and legislators need to know the variety of mitigation options that are available to reduce the pollution loads of sunscreens. In terms of how various Hawaiian jurisdictions respond to this contamination discovery, we provide a summary of the different mitigation options employed in Hawaii to combat this threat of sunscreen pollution, especially resulting from the data contained in this study.

2. Materials and Methods

2.1 Sample Collection

Seawater samples from Hawaii, U.S.A. were collected using precleaned one-liter amber glass bottles with Teflon-lined lids (I-Chem, 300 series, VWR), at a depth of 30 cm below the surface of the water. Sediment samples were collected using certified pre-cleaned 250 mL amber glass bottles with Teflon-lined lids. On the sand, vertices on a 15 cm x 15 quadrat were marked, and within the quadrat, the top 0.5-1.0 cm of surface sand was collected using a precleaned Teflon spatula. Sample locations are indicated in **Figures 1-4**.

On the island of Oahu (**Figure 1**), sand samples were collected on November 19, 2019 at the Kuhio Beach shower in the Waikiki district in the City of Honolulu (**Supplemental Figure 2**).Sand was collected from the shower run-off at Wailupe Beach Park (**Supplemental Figure 3**).

On November 13, 2019, sand samples were collected on the Island of Maui (**Figure 2**) from the showers at Kalama Park (**Supplemental Figure 4**), Kamaole Beach Part 2 (**Supplemental Figure 5**), and Polo Beach Park (**Supplemental Figure 6**). A reference sand sample was collected at the beach area of the Kihei, Maui boat ramp recreational area (**Figure 2**, **Supplemental Figure 7**). All of these sampling sites sit along the boundary of the U.S. Hawaiian Islands Humpback Whale National Marine Sanctuary.

From October 26 to November 13, 2019, sand and water samples were collected on the Island of Hawai'i (**Figures 3 and 4**). Sand samples were collected at two locations from the beach shower at the beach park that is the land access to Waialea Bay Marine Conservation and Land District (**Figure 3B**). Water samples were collected at a depth of approximately 30 cm below the surface of the water within Waialea Bay at the locations designated in **Figure 3C**. Water samples were collected near the Captain Cooke Monument within Kealakekua Bay Marine Conservation District. Two water samples were collected approximately 30 cm below the surface of the water on October 26, 2019 indicated by arrows in Figure 3D. Approximately 100 mL of shower discharge from the commercial recreational vessel was collected as it was flowing from the boat into the Kealakekua Bay receiving waters. Sand samples were collected at two locations (proximal and distal to the shower) at Mauna Lani Beach (Waikoloa Village, Hawaii; **Fig. 4A**; **Supplemental Figure 8**), at Black Sand Beach (Puako, Hawaii; **Figure 4B**; **Supplemental Figure 9**), and at Kahalu'u Beach Park (Kahaluu-Keauhou, Hawaii; **Figure 4C; Supplemental Figure 10**). As a reference for the Island of Hawai'i, sand was collected at the shore of Kapakuukapu (Manini Beach; **Supplemental Figure 11**). Five water samples were collected within Kahalu'u Bay (**Figure 4B**).

2.2 Chemicals and standards

Chemicals - Methanol (MeOH), acetonitrile (ACN), dichloromethane (DCM), ethyl acetate (EtAc), formic acid and HPLC-grade water were purchased from J.T. Baker (Deventer, The Netherlands) and were all ≥99.9% purity and included Certificates of Analysis. Aluminum oxide (≥99.9% purity), used as a clean-up agent in the pressurized liquid extraction (PLE) cells, was obtained from Merck (Darmstadt, Germany). Nitrogen and argon gasses (purchased from Air Liquid – Barcelona, Spain) were of 99.999% purity. Glass fiber filters (1 µm) and nylon membranes (0.45 µm) from Whatman International Ltd. (Maidstone, UK) and syringe filters supplied by Dionex Corporation (Sunnyvale, CA, USA) were used.

The following analytical standards (> 97% purity) were used: 2-Hydroxy-4-metoxybenzophenone (oxybenzone (BP3), dihydroxy benzophenone (benzophenone-1, BP1), 5-benzoyl-4-hydroxy-2-methoxybenzene-1-sulfonic acid (benzophenone-4, BP4), 4-hydroxybenzophenone (4HB), 4,4'-dihydroxy benzophenone (4DHB), 3-(4-tert-butylphenyl)-1-(4-methoxyphenyl)propane-1,3-dione (avobenzone,AVO), drometrizole (UVP), dimethyl benzotriazole (DMBZT), octocrylene (OC), 3,3,5-trimethylcyclohexyl 2-hydroxybenzoate (homosalate, HMS), 2-ethylhexyl salicylate (octisalate,OS), and BP-13C were purchased from Sigma-Aldrich (Darmstadt, Germany). Benzophenone-2 (BP-2), 2,2'-dihydroxy-4-methoxybenzophenone (DHMB, benzophenone-8), ethyl p-aminobenzoic acid (benzocaine, EtPABA), ethylhexyl methoxycinnamate (octinoxate,EHMC), and 1H-benzotriazole (BZT) were purchased from Merck (Darmstadt, Germany). 4-Methylbenzylidene camphor was obtained from Dr. Ehrenstorfer (Augsburg, Germany). MeBZT was acquired from TCI (Zwijndrecht, Belgium). BP3-^{13}C, BP3-d5, 4MBC-d4 and BZT-d4 were obtained from CDN isotopes (Quebec, Canada).

Stock solutions of the UV-filters and isotopically labeled standards were prepared at 100 mg/L in MeOH, and stored in the dark at -20 °C. Separate working solutions with UV-filters \ and isotopically-labeled internal standards were prepared at 10 mg/L in MeOH, stored in the dark at -20 °C, and refreshed weekly.

2.3 Sample pre-treatment and analytes extraction

The optimized and validated analytical methods used for the multiresidue determination of the target compounds and isotopically labelled standards in the standard solutions and in the sample extracts, were based in previous procedures developed in our laboratory (Gago-Ferrero et al., 2011; Downs et al., 2021a) and expanded for new compounds (**Supplemental Table 2**).

2.3.1 Sediment

The extraction of the selected analytes from the sediment samples was carried out according to the expanded method based on Gago-Ferrero *et al.* (2011). Samples were isolated and in-cell purified by pressurized liquid extraction (PLE) using an automatic system ASE 350 from Dionex. One gram of the lyophilized sediments was mixed with 1 g of activated aluminum oxide (previously activated at 130 °C for 24 h.) in the PLE cells. Before extraction, the surrogate standard, i.e. BP·-

^{13}C, was spiked into the samples and allowed to equilibrate overnight. MeOH and the mixture MeOH/water (1:1, v:v) were used as extracting solvents. The 20 mL extraction was brought to 25 mL with MeOH in a volumetric flask. An aliquot of 6 mLof this solution was passed through a 0.45 µm syringe PFTE filter to a UHPLC-vial and further evaporated until dryness under a nitrogen stream using a Turbo-Vap (Zymark, Hopkin, MA). Finally, the dried extract was reconstituted with 500 µL of the IS mixture solution containing the rest of the isotopically labelled standards, i.e. BP3-d5, 4MBC-d4 and BZT-d4.

2.3.2 Water

The extraction of the 20 petrochemical, ultraviolet filters (parent and transformation products) and hormones from water samples followed the expanded method based on Downs et al. (2021). Briefly, 100-200 mL of water sample was loaded onto Strata™-X 33 µm polymeric reversed phase C18 solid phase extraction (SPE) cartridges (500 mg/12 mL; Phenomenex) at a flow rate of 4 mL/min, for isolation and purification of the target analytes. Then, the cartridges were washed with 3 mL of HPLC-grade water and dried under a current of N_2. The cartridges were eluted at a flow rate of 3 mL/min, first with 8 mL of a mixture solution of EtAC and DCM (EtAc/DCM (1:1, v:v), and then with 2 mL of DCM. The two extracts were joined and evaporated with nitrogen until near dryness and then transferred into a UHPLC-vial for full evaporation. Reconstitution of the dried extract was performed with 0.5 mL of HPLC-grade water containing the isotopically labelled internal standards.

2.4 UHPLC-(ESI)-Orbitrap MS analysis

The chromatographic separation of all analytes was performed in an Acquity UHPLC C18 column (100 Å, 1.8 µm, 2.1 ×100 mm) with a guard column containing the same material, using an Acquity ultra-high-performance liquid chromatograph (UHPLC) (Waters Corporation, Milford, Ma, USA) coupled to a Q-Exactive Orbitrap mass spectrometer (Thermo Scientific, Waltham, Ma, USA). Electrospray ionization in positive (ESI+) and negative (ESI-) modes was selected as the ionization technique. In ESI+, the mobile phase consisted of a mixture of MeOH and HPLC-grade water, both 0.1% in formic acid. The elution gradient started with 5% of MeOH, increasing to 75% at min. 7 and to 100% at min. 10. Afterwards, it was decreased from initial conditions for at least 17 min and were maintained 20 min until equilibration.

Five of the 18 compounds were analysed under negative ionization mode (i.e., BP4, E1, E3, HMS, and OS). In ESI- mode, the mobile phase consisted of the same solvents as in positive polarity, but in this case, both were 5 mM in ammonium acetate. The elution gradient started with 5% of MeOH, increasing to 50% at 3 min., to 90% at 6 min., and to 100% at 13 min. Then, it was decreased to 5% in one minute and maintained until 20 min. for equilibration.

In both modes of analysis, the flow rate was set at 0.3 mL/min, the oven temperature at 40 ºC and the injection volume at 10 µL. Mass spectrometry detection was performed under parallel reaction monitoring (PRM), specifying the compounds of interest in the inclusion list. Precursor selection combined with high-resolution product ion scanning was tuned to provide enhanced selectivity.

All samples were measured in duplicate and the reported value corresponds to the mean of the two determinations. All the compounds were quantified and confirmed with the two most intense transitions by the isotope dilution approach.

Supplemental Tables 3 and **4** report the validation parameters for the target substances in water and sand samples, respectively. The methods showed generally good average recoveries calculated from two spiking levels in sand (77 – 125%, spiking levels 5 and 50 ng/g) and water (56-110%, spiking levels 5 and 50 ng/L): n=5, RSD: 5-22% and low LODs ranging between 0.01– 0.06 ng/g dw in sand and 0.001– 0.007 ng/mL in water, and LOQs ranging between 0.03– 0.11 ng/g dw in sand and 0.005- 0.0022 ng/mL in water) in a wide linear range (1-700 ng/mL for water and 1-700 ng/g for sand) for all compounds (r^2 >0.991), allowing the reliable analysis of the target UV filters in water and sediment samples. LODs and LOQs, both instrumental and methodological, were calculated as the concentration of each compound giving a signal-to noise ratio of 3 and 10, respectively. The precision of the method was evaluated by analyzing five consecutive times the corresponding matrices (water or sand extracts) spiked with a standard mixture of the analytes at 100 ng/L; intraday values were < 12% and inter-day precision was < 23%) indicating a good method precision.

2.5 Analytical chemistry quality assurance and quality control

Background contamination in the laboratory is known to be potentially an issue in petrochemical UV filter analysis at environmentally relevant concentration levels. To avoid this, all non-volumetric glassware used was previously washed sequentially with HPLC grade water, ACN, MeOH and acetone, and heated overnight at 380 °C. In addition, gloves were used throughout the handling and preparation of the samples. Labware was always wrapped with aluminium foil, the PLE extraction vials were used from amber glass, and they were stored in the dark to avoid photodegradation. In every batch of samples, a procedural blank, a control standards mixture solution (Quality Control solutions, to check for instrumental drift in response factors), and two pure MeOH blanks (indicators of instrument contamination) were inserted randomly among samples to be measured.

2.6 Risk Assessment Method

The European Commission guidance was used to calculate risk quotients (RQs) for the oxybenzone and octocrylene in water samples, and oxybenzone, octocrylene, and 4-methylbenzylidene camphor in sand samples. The European Commission methodology has been adopted in the development of several ecological risk assessment guidelines (ECHA 2008; European Commission 1996, 2003; Environment Canada 2013; European Medicines Agency 2006; Dussault et al., 2008; Hernando et al., 2006). Using this method, the actual or predicted environmental concentration (MEC) is compared to a derived known or Predicted No-Effect Concentration (PNEC). In cases where the NOEC was not known, but the Lowest Observable Effect Concentration (LOEC) was known, the LOEC was divided by two to calculate a predicted NOEC (ECHA, 2008). For RQ determinations, an assessment factor of 1000 was selected to address the differences between laboratory data and natural conditions to account for interspecies differences and intraspecies differences. Thus, the RQ = (MEC)/(PNEC, NOEC,LC_{50} or EC_{50}) x 1000 (Chapman et al., 2009; Dussault et al., 2008; Means et al., 1993; Environment Canada, 2013; Belanger et al., 2021). Toxicity reference values were obtained from the published literature (**Tables 1 and 2, Supplemental Table 5**).

A number of endpoints not commonly used as regulatory toxicological endpoints are included in **Tables 1-2 and Supplemental Table 5**. However, all of these toxicity endpoints can be argued to reflect aspects necessary for population-level survival and reproductive fitness in real world situations (Goulson, 2013; Moore et al., 2004; Ruel and Ayres, 1999; Schafer et al., 1994).

The criteria for Levels of Concern for organisms in ecosystems for interpreting the RQ is based on a four-tier ranking system (European Commission 1996; Sanchez-Bayo et al. 2002; Hernando et al. 2006). Based on the American National Standards Institute recommendations for Hazard Communications, a color scheme is used for ease of visualization of the Levels of Concern for this methodology (**Tables 1 and 2, Supplemental Table 5**). Red boxes represent RQ values greater than 1, indicating an unacceptable risk requiring immediate action, and is the standard criteria for the Level of Concern within the European Commission framework. Orange boxes represent values between 0.5 and 1.0; a moderate concern of an acute impact. Yellow boxes represent values between 0.1 and 0.49, indicating a lower risk of impact. White boxes indicate no concern of danger with values below 0.1.

3. Results

3.1 Measured Environmental Levels

Petrochemical UV-filters were measured at 12 locations across three islands of Hawaii (**Figs. 1-4**). For oxybenzone, the highest concentrations were found at popular tourist beach showers; the highest being the Kuhio Beach shower in Waikiki at 34 mg/g dw (dry weight) (**Fig. 1**). Two beach showers did not have quantifiable oxybenzone concentrations: Wailupe Beach park shower in Oahu and Polo Beach Park shower in Maui (**Figs. 1 and 2**). Both parks are visited predominantly by locals and not by tourists. At shower locations that sampled both a proximal and distal length from the shower, only the proximal locations had measurable concentration of oxybenzone, while the distal locations had non-quantifiable levels or much lower levels as compared to the proximal location (**Fig 3B, Fig 4A-C**). The two non-shower sites, Kihei Boat Beach (**Fig. 2B, Supplemental Figure 6**) and Kapakuukapu (Manini Beach; **Supplemental Figure 11**) had no quantifiable levels of oxybenzone or any other target analytes.

Breakdown products associated with oxybenzone, including benzophenone-1 and DHMB were concomitant with oxybenzone contamination; the exception was Black Sand Beach. At most oxybenzone-contaminated sites, 4-hydroxybenzophenone (4HB) was detected with the exception of Polo Beach (**Fig. 2B**).

Avobenzone levels were highest at 1, 085 ng/g dw at Kuhio Beach and ranged from 100 ng/g dw to 500 ng/g at the high-density tourist beaches in Maui and Hawai'i Islands. Avobenzone levels at the less-frequented beaches such as Wailupe, Polo, and Wailea beaches were around 1-5 ng/g dw (**Figs. 1, 2B, 3A**). Mauna Lani (**Fig. 4A**) and Black Sand (**Fig. 4B**) beaches were groomed (all biological and plastic debris remove, sands were raked) by local grounds keepersthe evening or morning before the samples were collected and had no quantifiable levels of avobenzone.

Kuhio Beach had the highest levels of octocrylene (1,075 ng/g dw; **Fig. 1**), while the other three highly visited tourist-beach sites (Kalama, Kamaole, and Kahalu'u beach parks) had octocrylene ranging from 204 ng/g dw to 686 ng/g dw of octocrylene. The more reclusive sites had relatively low concentrations such as 7 ng/g dw (Wailupe Beach Park, **Fig. 1**) to no quantifiable levels at Mauna Lani and Black Sand beaches, with the exception of the distal site of Mauna Lani, which had 61 ng/g octocrylene (**Fig. 4A**).

Octisalate and homosalate were not detected in any of the sand samples. 4-methylbenzylidene camphor was only detected at the Wailupe Beach Park (**Fig. 1A, Supplemental Fig. 3**).

A lateral transect was designed and sampled at Waialea Bay (Fig. 3C). Site 3 water-sample from Waialea Bay was closest to the rivulet that ran from the Waialea Bay beach shower to the tidal shoreline. Site 3 had the highest concentrations of all measurable target analytes, except for octisalate. It should be noted that benzophenone-2 was also detected in all three water samples.

A four-point transect, and a two-point transect was designed to collect water samples in Kahalu'u Bay where Site 1 is the origin of both transects and the entry point for many swimmers as well as the receiving waters for the rivulet that flows from the beach shower (**Fig. 4D, Supplemental Figure 10**). Site 1 had the highest concentrations of avobenzone, oxybenzone and its break-down products. Site 2 had no quantifiable levels of oxybenzone. Concentrations for octocrylene were equal between Site 1 and Site 5 but were highest at Site 2. There was no consistent pattern regarding concentrations among the five sites for octisalate.

The water samples were collected on the northern end of Kealakekua Bay, near the Captain Cooke monument. There is no beach shower at this location, but visitors can wash off with fresh water if they board a large commercial tourism vessel, similar to the one in **Fig. 3D**. which can hold as many as 16 to 90 swimming customers. The shower is near the water-entry/exit point on the vessel, and shower-discharge was sampled as it was cascading into the receiving waters of Kealakekua Bay. No target analytes were detected. Measurable concentrations of target analytes were found at the two collection sites near the moored vessel shown in **Fig. 3D**.

3.2 Risk Analysis

Oxybenzone concentrations in beach sands at Kuhio, Kalama, Kamaola, Waialea (Site 1), Black Sand (Site 2) and Kahalu'u (Site 1) exhibited RQs all above 1 (**Table 1**). Mauna Lane (Site 2) and Black Sand (Site 1) exhibited lower risk levels for both mortality and growth rate in *Eisenia fetida*. Kahalu'u (Site 2) exhibited moderate to no risk for all of the multi-species parameters examined.

Octocrylene concentrations in beach sands at Kuhio, Kalama, Kamaola, Wailea (Site 1), and both sites within Mauna Lani and Kahalu'u beach sites had RQ values above 1 (**Table 1**). Octocrylene was measurable at the Wailupe Beach Park site, but exhibited a moderate level of risk (RQ = 0.71).

4-Methylbenzylidene camphor is not a U.S. Food & Drug Administration approved sunscreen active ingredient, but it is used in fragrances as a UV-stabilizer. It was detected in sands only at the Wailupe Beach Park site. Despite it not being allowed as an active SPF ingredient in U.S.-based sunscreen products, we cannot rule out that tourists from countries

where this UV filter is approved, such as the European countries and Australia, which is allows up to 4% as a sunscreen ingredient. The RQ values in the three species included in this risk assessment range from moderate to serious concern to ecological integrity (**Supplemental Table 5**).

Risk quotients were generated for all water samples from sites that had measurable amounts of oxybenzone, which means Kealakekua (Site 1) and Kahalu'u (Site 2) had an RQ of 0 (**Table 2**). All oxybenzone-contaminated samples exhibited RQ values for all coral cell species and all species parameters above 1, with the exception for Kealakekua Bay (Site 2) which had RQ values of serious concern. Risk quotients generated for octocrylene at all 8 water-sampled sites predominantly exhibited RQ values above 1, indicating a severe condition to coral and other invertebrates, as well as fish (**Table 2**).

4. Discussion

The U.S. Environmental Protection Agency (U.S. EPA) defines a point-source of pollution as "any single identifiable source of pollution from which pollutants are discharged" and that a pollutant is a substance at a specific concentration that alters the chemical, biological, or physical characteristics of a navigable receiving water (U.S. NOAA; U.S. Federal Water Pollution Control Act Amendments of 1972). The risk assessment conducted in this study allows for the argument that the sunscreen chemical concentrations measured at these locations can be classified as pollutants, which makes beach showers a point-source of pollution. Under the U.S. Clean Water Act, beach showers should cease discharging or apply for a permit under the National Pollutant Discharge Elimination System (U.S EPA, 2022).

There was an unexpected incongruence in the concentration of sunscreen contaminants in sands at locations where proximal and distal samplings were conducted (*e.g.*, Black Sands Beach, Kahalu'u Beach). Our first conjecture for this incongruence was based on hydrophobicity (*i.e.* n-Octanol/water partition coefficient, K_{ow}) of the compounds in relation to basaltic or aragonitic sands or that microplastics in the sand were a confounding factor (**Supplemental Table 2**) (Schaffer et al., 2012; Santa-Viera et al., 2021). Instead, after speaking with State of Hawaii resource managers and resort grounds keepers, we learned that beaches are groomed almost every day between 4 am to 6 am. This is done to remove gullies created by shower runoff and surface depressions made by beach-visitors for both aesthetic and liability issues (*i.e*. these can be obstacles that physically harm beach-visitors). This grooming includes both raking and plowing the beach, so that a final smooth beach surface is obtained, and both biological and marine plastic debris are removed. This daily disturbance of plowing under surface sands and bringing-up sands 4-6 cm below the surface could easily skew expected concentrations.

Tidal flux can liberate these sunscreen contaminants from sands during high tides and result in diurnal pulses of contamination into the adjacent water column. Some workers make claims that water solubility is a critical factor for seawater contamination. There is some merit to this line of inquiry, but also a susceptibility to abuse; it should never be assumed that seawater, especially coastal seawater in highly developed coastlines is akin to "pure laboratory seawater" and that natural waters are without a high level of biological or dissolved organic/carbon matrix. The reality is just the opposite, many of these waters are loaded with high levels of sea dissolved-organic-matter (SeaDOM) and total dissolved carbon, partially resulting from terrestrial runoff (agricultural runoff, road runoff, residential landscape runoff, etc), and point and non-point source sewage discharges. Even shedding of sunscreens from swimmers contributes to the SeaDOM composition of a receiving water (*i.e.* manifestation of the sunscreen sheen on the surface of the water). This increase in dissolved organic matter can increase the solubility and suspension of petrochemical UV-filters and any other polyaromatic hydrocarbons within the water column versus its hydrophobic partitioning to the surface of the water

(Whitehouse, 1985; Bejarano et al., 2005; Lou et al., 2006; Mopper et al., 2007; Shang et al., 2015).

This study provides an anecdotal observation that there is a relationship between beach visitation rate and UV filter-contaminant concentration, the higher the visitation, the higher the concentration. This seems reasonable, but a more formal study should be conducted that examines the relationship and the strength of this correlation. If this relationship is true, then resource managers may want to focus on mitigating contamination loads at the most visited sites. Measuring UV filter contamination analysis can be expensive and readily accessible for most locations. The estimation for sunscreen loads at a site based on the formula in Supplemental Table 1 can be used to estimate the threat of a visitors if a "no mitigation policy" is enacted, as well as hypothesize/estimate the impact of a proposed mitigation policy. The Carrying Capacity estimation for a natural resource is notoriously inaccurate and fraught with conflicted or contrived political and economic influences (Lindberg & McCool, 1998; Hawkins et al., 1999; McCool and Lime, 2001; Ponting and O'Brien, 2015; Singh, 2015; Tacconi and Williams, 2020; Wall, 2020). Sunscreen load calculation, with accurate visitation rates at a location, could provide a basis for more exact carrying capacities, especially if integrated with simple hazard or risk assessments (Thomas et al.,2005; Butler & Dodd, 2002). This study should hopefully inspire risk assessors and social and municipal managers to develop new methods for generating accurate tourism carrying capacity models of aquatic and coastal natural resources based on sound ecological risk assessment theory (Raimondo & Forbes, 2022; Sun et al., 2022).

Beach showers may not be the only point-source of sunscreen pollution; boat showers are another possible point-source of pollution. In Kealakekua, the commercial vessel we collected discharge from did not contain detectable levels of sunscreen contaminants. This vessel is an irregular example, because it visits the Kealakekua marine protected area and must adhere to a state-issued mooring permit not to sell or have on board petrochemical-UV sunscreen products (**Supplemental Figure 12**). Furthermore, this company (Fair Winds Cruises) has an active customer-education program that engages all their customers in using non-petrochemical-UV sunscreens, even providing free mineral sunscreens to all customers (**Supplemental Figure 13**). The measurable levels of sunscreen contaminants in Kealakekua indicates that the likely source is coming from individuals on other personal and commercial craft that can become point-sources of sunscreen contamination because they are not required to adhere to the permit regulations prohibiting petrochemical sunscreens (**Supplemental Figure 12**). To reduce the impact of sunscreen pollution within the Kealakekua marine protected area, mitigation options should be considered and implemented that include all visitors to Kealakekua.

The threat to beach and near-shore habitats based on the risk assessments is considerable (**Tables 1 and 2, Supplemental Table 4**). Beach showers are all located in the Upper Zone of beach habitats and can contaminate middle and lower beach zones via rivulets created from the

shower discharge. For terrestrial risk quotients, the only species available were soil species not often associated with beach habitats, or freshwater larval forms that associate with submerged sediments (Gautam et al. 2022). Sunscreen pollution intrudes upon all three zones of beach habitats, and model species that are more appropriately representative of beach habitats should be included in ecotoxicological studies to create more relevant risk assessments. These include Ghost crabs (subfamily Ocypodinae), fishery-related crabs such as Kona (*Ranina ranina*), Kuahonu (*Portunus sanguinolentus*), and Samoan (*Scylla serrata*) crabs where part of their lifecycle is spent within beach habitats, as well as plants and macroalgae (e.g., grass, Hawaiian *limu*) and even sea turtle egg development (Titcomb et al., 1978; Thomas et al., 2013).

4.1 Sunscreen pollution mitigation options

Within social and political frameworks, it is often hoped that science provides a context and justification for action in mitigating pollution. Mitigation of pollution usually proceeds along three main strategies: reduction of contamination (Strategy 1), prohibition of contamination (Strategy 2), and the removal of the contamination (Strategy 3). In the case of Hawaii, based on the data presented within this study and in other concordant scientific efforts, all three strategies are being implemented to mitigate the impacts of specific aspects of sunscreen pollution (e.g., a specific sunscreen product ingredient) (Downs et al., 2016, 2021a).

The dominant strategy used to reduce contamination (Strategy 1) of a pollutant is public outreach and education. In the case of Hawaii, and specifically in several Hawaiian beach parks and Marine and Land Conservation Districts (i.e., marine protected area), tourists and locals are encouraged to wear sunscreen products that do not contain ingredients that may pose a threat to biological/ecological integrity. In the case of Keleakakekua Marine and Land Conservation District, the major commercial tourist vessel company exhibits literature and educational videos about the problem of sunscreen pollution, and how consumers can contribute individually to addressing the problem (**Supplemental Figure 13**). In the case of Kahalu'u Bay Beach Park, the Kahalu'u Bay Education Center directly engages with tourists visiting the beach regarding the issue of sunscreen pollution, and advocates which ingredients they should avoid. Besides advocating for using only U.S. Food and Drug Administration's category-1 GRASE (Generally Recognized As Safe and Effective) sunscreen products, they recommend that visitors wear UPF (Ultraviolet Protection Factor) sun-protective clothing which reduces the use of sunscreen product application to a body by more than 50%, while ensuring continuous high-level UPF protection that does not degrade and shed from the skin while swimming (**Supplemental Figure 14**).

Employing rational and compelling arguments based on sound scientific information to tourists regarding what their sunscreen products could do if levels reach an action threshold of pollution is thought to increase the likelihood of adopting conservation-consumerism behavior,

thereby reducing sunscreen chemical contamination in a given area. Kahalu'u Bay Education Center was one of the first organizations to both conduct environmental contamination surveys of their marine managed area, as well as soliciting U.S. NOAA to generate risk assessment quotients of their contaminant data (**Supplemental Figure 15**). This information was translated into easy-to-access infographics and educational material and presented to tourists as banners and handout literature.

A second tactic in the strategy to reduce contamination loads (Strategy 1) is to regulate the number of visitors to a geographic location. One example of this was the complete shutdown of Maya Bay, Thailand for almost three years to allow for ecological recovery. After this approach, Maya Bay regulators took a more nuanced approach by limiting the number of visitors per day to a location, or to close the location to visitors for a set number of days per week. Hanauma Bay, Hawaii had once adopted both approaches, limiting the number of visitors to 1,500 persons/day and shutting the Bay down to visitors Monday and Tuesday of every week (https://www.honolulu.gov/parks-hbay/information-fees.html). In the 2nd quarter of 2021, despite ecological impacts of overtourism, the city of Honolulu abandoned the former policy, and now visitor numbers are no longer regulated. Hanauma Bay sees more than 2,000 visitors/day (City of Honolulu, 2022), contributing to more than an estimated metric ton of sunscreen per month (**Supplemental Table 1**). This tactic's effectiveness could be optimized with a commitment to conservation over commercialization so a legitimate carrying capacity model, merged with contaminant surveys and risk assessments, can measurably mitigate visitor impacts.

A second strategy of mitigation is the prohibition of the sale or use of targeted products that contain specific UV-filters, such as oxybenzone, octinoxate, and octocrylene. Within this strategy are a number of tactics. One tactic of Strategy 2 is the prohibition of specific UV-filter containing sunscreen products in a geographic area. The prohibition of petrochemical UV-filter products as part of the mooring permit in the Keleakakekua Marine and Land Conservation District is an example of prohibition in a geographic jurisdiction, especially a marine protected area (**Supplemental Figure 12**). A second tactic of Strategy 2 is the "selective prohibition" which bans the sale of specific ingredients in a jurisdiction, the most famous example of this is the 2018 Hawaii Act 104 that regulates the sale and distribution of oxybenzone and octinoxate sunscreen products in the State of Hawaii (**Supplemental Figure 16**). This study was conducted immediately before the COVID lockdowns of 2020 and before the full implementation of Hawaii Act 104 (implemented on January 1, 2021), so future studies will be able to document the effectiveness of this mitigation tactic.

A third tactic of Strategy 2 is the Precautionary Principle approach, which is the prohibition of environmental contaminants that are scientifically documented to induce ecotoxicities at environmentally relevant concentrations (Downs et al., 2022). A measure based on this precautionary approach was adopted by the County of Maui which prohibited all

petrochemical sunscreens that have not been recognized as GRASE (Generally Recognized As Safe and Effective) by the U.S. FDA and would remain prohibited until these chemicals can be proven *not* to pose a threat to ecological receptors. In November 2021, the Maui County Council passed Ordinance 5306 (https://www.mauicounty.gov/DocumentCenter/View/130826/Ord-5306; **Supplemental Figure 17**), banning the sale, distribution and use (without a prescription) of all U.S. FDA non-GRASE sunscreens, effective October 1, 2022. This legislative action was supported by the Hawaii State Department of Land and Natural Resources (DLNR) as a crucial policy to protect its terrestrial, freshwater and marine ecosystems (DLNR, 2022 press release).

Implementation of this Precautionary approach regarding petrochemical sunscreen prohibition requires public education efforts (Strategy 1, Tactic 1) to ensure both broad public compliance and persistent behavioral fidelity. Especially in the early period of the ordinance's implementation, public education could be coupled with providing free-access to FDA-GRASE sunscreen products to beach visitors. It could aid compliance and public endorsement of the policy, especially in the early period of Ordinance 5306, by coupling public and consumer education with accessibility to beach visitors with a U.S. FDA-GRASE sunscreen. If sunscreen is one of a number of measures to protect public health from the damaging effects of over-exposure to the sun, then ecologically safer sunscreen products can be made accessible to ensure compliance to mitigation tactic #3 (Precautionary Approach). Fortunately, pursuant to the passage of Ordinance 5306, DLNR installed four mineral sunscreen dispensers at two South Maui marine protected areas. The 2022 Hawaii State Legislature is debating whether to expand this tactic and to install similar dispensers throughout the State of Hawaii. Along with greatly diminishing the harmful chemical impacts, the new law may also benefit local businesses through stimulating increased demand for FDA-GRASE mineral sunscreens, several of which are produced and sold by local Maui companies.

A fifth tactic of Strategy 2 is the temporal prohibition of the use of a sunscreen product in a geographic/jurisdictional locality. This is often employed during the most sensitive aspects of the lifecycle of a keystone species (*e.g.*, lunar-influenced spawning of coral, fish, sea urchins, among others). For the case of Kahalu'u Bay Beach Park, the entire Park and the Bay are closed to visitors for a duration of time (*i.e.* 2-7 days) during the lunar spawning of the coral genus, *Pocillopora meandrina* (**Supplemental Figure 18**). Many marine species, especially corals and fish, will have a very defined duration in which to spawn (full moon) of a single month for the entire year. Several studies have shown that certain petrochemical UV-filter ingredients can be toxic to gametes and may even prevent successful fertilization events (Blüthgen et al., 2012; Coronado et al., 2008; Ghazipura et al., 2017; Rehfeld et al., 2018; Xu et al., 2021). Fertilization/spawning events may not be the only critical facet of an organism's life cycle. In corals, fish and mammals, environmentally relevant oxybenzone-exposure *in utero* or during embryonic development may result in disease, ranging from morbid deformities and bleaching in coral planulae to Hirschsprung Disease from exposure during the first trimester in mammals and

abnormal development of the nervous system in fish (Downs et al., 2016; Huo et al., 2016; DiNardo and Downs, 2019; Wang et al., 2021; Han et al., 2022).

The third strategy is removal of the contaminating effluent. There are theoretical technologies that are not commercially available at the time of publication that propose to scavenge UV-filter aromatics by adsorption using floating macrobeads released into receiving waters, which are then recollected for disposal (Stoye, 2017). The most effective tactic is to prevent the discharge of beach/boat showers into receiving waters altogether, and instead to either (a) install a collection drain that pumps the shower waters directly into a municipal wastewater treatment system (WWTS) or (b) collects the shower waters into a secure cesspit container, which then can be pumped out and delivered to a WWTS for processing.

Unfortunately, rudimentary WWTS (those that only remove solids) can be a significant source of environmental contamination. A majority of petrochemical UV-filters, pharmaceuticals, micro- and nano-plastic ingredients in personal care products, and other cosmetic chemicals are not degraded or made innocuous by rudimentary sewage treatment. Instead, the WWTS effluent can be one of the major sources of sunscreen pollution of a coastal, lake and river environments. With water scarcity and fertilizer resources becoming a global issue, reclaimed waters from a WWTS or its biosolids are often used in agricultural settings, and their run-off becomes non-point sources of sunscreen pollution. For an effective WWTS to manage sunscreen pollution, it requires an advanced design that has a secondary and tertiary phase that removes these sunscreens contaminants through a singular process or a mixture of biological degradation, absorption to activated carbon, or extensive chemical/radiation degradation (Bavumiragira et al., 2022; Morin-Crini et al., 2022).

Effective wastewater treatment systems (WWTS) are expensive to obtain and maintain. New construction of rudimentary WWTS and sewage collection lines can cost over U.S. $1million for 1,000-5,000 people (Bode and Grünebaum, 2000). Addition of secondary treatment (i.e.., aeration and clarifiers) and disinfection systems are additional costs to the wastewater utility. For instance, the application of advanced oxidation processes (AOPs) has proven to be fairly effective in removing emerging contaminants, especially petrochemical sunscreens (Rizzo et al., 2019; Imamovi ć et al., 2022); however, its cost, estimated to be in excess of €2 million a year hinders its wide implementation (Mainardis et al., 2020). Advanced oxidative process (AOP) is a chemical process which has been found to be the most effective technologies in eliminating most biological and organic micropollutants from the water. The AOP technologies have been applied to degrade and remove insecticides, herbicides and a wide variety of organic pollutants (Martinez-Huitle & Ferro, 2006; Xiao et al., 2016; Guelfi et al., 2017; Martınez-Huitle & Penizza, 2018). These advanced technologies have been shown to successfully remove more than

90% of the residual pharmaceuticals, endocrine disrupting chemicals (EDCs), and personal care products from the wastewater effluent (Esplugas et al., 2007; Trapido et al, 2007; Ikehata et al, 2008; Lester et al., 2011; Angeles et al, 2020; Bermudez et al, 2021; Mousel et al., 2021).

Granulated activated carbon (GAC) is an excellent adsorption media and commonly used in drinking and wastewater industries to remove organic compounds. It is also used to improve the odor, color, and taste of the water in the drinking water industry, as well as reclaimed water for irrigated agriculture. It is a passive system to remove contaminants from the water at very low cost. GAC works by attracting the organic pollutants to attach to its surface. In the wastewater domain GAC has been used to remove micropollutants and phosphorus (Altmann et al., 2016; Benstoem et al., 2017), antibiotics (Choi et al., 2008), perfluoroalkyl acid (PFAA) (Inyang & Dickenson, 2017) and perfluorinated surfactants (Ochoa-Herrera and Sierra-Alvarez, 2008) among other pollutants. GAC adsorption is an effective treatment for wastewater when the dissolved oxygen carbon (DOC) contents are between 10 to 20 mg/L (Bui et al., 2016). However, GAC-based systems, by themselves, are not efficient in removing many pollutants of high concern including UV sunscreens (Glover et al., 2021).

Ideally, Hawaii's and any coastal beach-shower wastewater should be collected into a holding tank and processed through an AOP treatment system to degrade organic pollutants including the UV sunscreen chemicals followed by a nanomembrane (NM) filtration system. This combination of AOP-electrical chemical reactor and nanomembrane filter could degrade and remove a majority of residuals from the shower water and produce a clean effluent that can be safely released into receiving waters. If the objective is to eliminate only biological pollution such as bacteria and viruses, an AOP system based on ozone, H_2O_2, or peracetic acid can be a viable solution. However, if the goal is to eliminate organic chemicals and micropollutants from the wastewater effluent then an electric-chemical reactor-based AOP treatment system followed by nanofiltration should be a strong consideration.

5. Conclusion

Beach shower and boat discharges can be point-sources of sunscreen pollution. There is a trend where the most visited the beach by tourists, the higher the concentration of sunscreen pollutants. In 2018, The State of Hawaii passed a law that would ban oxybenzone and octinoxate beginning in 2021. The public education campaign associated with being "Hawaii Compliant", even in 2019, may have contributed to the reduced contamination of oxybenzone at these Hawaii beach locations. Beach shower discharges, especially with the use of beach grooming, can potentially result in distributing the contaminants over the entirety of the beach, impacting crab, annelid, monk seal, sea turtles, migratory birds, and plant/algal species within these beach habitats.

Industry and governments need to invest in the ecotoxicological sciences to determine the effects of sunscreen ingredients on beach habitat species, so that risk assessments can be incorporated into the drafting of more valid and verifiable carrying capacity models. Many of these locations sampled in this study are multi-jurisdictional marine protected areas, arguing that management plans for all marine protected areas need to consider the impact of tourists, especially that of sunscreen pollution. There are a number of mitigation options that can possibly reduce pollution discharges and effects. There is a need for studies to determine the effectiveness of these policies so that other jurisdictional and management organizations can implement and optimize solutions for the conservation of their own natural resources.

DECLARATIONS

Ethical Approval: Not applicable.

Consent to Participate: Not applicable.

Consent for publication: Not applicable.

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Availability of Data and Materials

LC-MS chromatograms for the sediment/water analysis can be obtained from Dr. Silvia Diaz Cruz, Spanish Research Council (email: sdcqam@cid.csic.es). Calculations for the E.U. risk quotients can be obtained from Dr. Cheryl Woodley, U.S. National Oceanic and Atmospheric Administration (email: cheryl.woodley@noaa.gov).

References

Afshar-Mohajer, N., Li, C., Rule, A.M., Katz, J., Koehler, K. (2018) A laboratory study of particulate and gaseous emissions from crude oil and crude oil-dispersant contaminated sweater due to breaking waves. Atmospheric Environment 179:177-186. https://doi.org/10.1016/j.atmosenv.2018.02.017

Akkanen, J., Penttinen, S., Haitzer, M., & Kukkonen, J. V. (2001). Bioavailability of atrazine, pyrene and benzo[a]pyrene in European river waters. Chemosphere, 45(4-5), 453–462. https://doi.org/10.1016/s0045-6535(01)00038-8

Alamer, M., & Darbre, P. D. (2018). Effects of exposure to six chemical ultraviolet filters commonly used in personal care products on motility of MCF-7 and MDA-MB-231 human breast cancer cells in vitro. Journal of Applied Toxicology38(2), 148–159. https://doi.org/10.1002/jat.3525

Almeida, S.D.S., Rocha, T.L., Qualhato, G., Oliveira, L.A.R., Amaral, C.L.D., Conceicao, E.C.D., Saboia-Morais, S.M.T., Bailao, E., 2019. Acute exposure to environmentally relevant concentrations of benzophenone-3 induced genotoxicity in *Poecilia reticulata*. Aquatic Toxicology 216, 105293. https://doi.org/10.1016/j.aquatox.2019.105293.

Altmann, J., Rehfeld, D., Trader, K., Sperlich, A. and Jekel., M. Combination of granular activated carbon adsorption and deep-bed filtration as a single advanced wastewater treatment step for organic micropollutant and phosphorus removal. Water Research 92, 131-139, 2016

Balmer, M. E., Buser, H. R., Müller, M. D., & Poiger, T. (2005). Occurrence of some organic UV filters in wastewater, in surface waters, and in fish from Swiss Lakes. Environmental Science & Technology, 39(4), 953–962. https://doi.org/10.1021/es040055r

Bavumiragira, J. P., Ge, J., & Yin, H. (2022). Fate and transport of pharmaceuticals in water systems: A processes review. The Science of the Total Environment, 823, 153635. https://doi.org/10.1016/j.scitotenv.2022.153635

Belanger, S. E., Beasley, A., Brill, J. L., Krailler, J., Connors, K. A., Carr, G. J., Embry, M., Barron, M. G., Otter, R., & Kienzler, A. (2021). Comparisons of PNEC derivation logic flows under example regulatory schemes and implications for ecoTTC. Regulatory Toxicology and Pharmacology: RTP, 123, 104933. https://doi.org/10.1016/j.yrtph.2021.104933

Bejarano, A.C., Chandler, G.T., Decho, A. W. (2005) Influence of natural dissolved organic matter (DOM) on acute and chronic toxicity of the pesticides chlorothalonil, chlorpyrifos and fipronil on the meiobenthic estuarine copepod *Amphiascus tenuremis*. Journal of Experimental Marine Biology and Ecology 321(1):43-57. https://doi.org/10.1016/j.jembe.2005.01.003

Benstoem, F., Nahrstedt, A., Boehler, M., Knopp, G., Montag, D., Siegrist, H. and Pinnekamp, J. (2017) Performance of granular activated carbon to remove micropollutants from municipal wastewater-A meta-analysis of pilot- and large-scale studies. Chemosphere, 185, 105-118.

Bigott, Y., Gallego, S., Montemurro, N., Breuil, M. C., Pérez, S., Michas, A., Martin-Laurent, F., & Schröder, P. (2022). Fate and impact of wastewater-borne micropollutants in lettuce and the root-associated bacteria. The Science of the Total Environment, 831, 154674. https://doi.org/10.1016/j.scitotenv.2022.154674

Blüthgen, N., Zucchi, S., Fent, K. (2012). Effects of the UV filter benzophenone-3 (oxybenzone) at low concentrations in zebrafish (Danio rerio). Toxicology and Applied Pharmacology, 263(2), 184–194. https://doi.org/10.1016/j.taap.2012.06.008

Bode, H., Grünebaum, T. (2000) The cost of municipal sewage treatment – structure, cost, minimization – methods of fair cost comparison and allocation. Water Science & Technology 41(9):289-298. https://doi.org/10.2166/wst.2000.0226

Boyd, A., Stewart, C.B., Philibert, D.A., How, Z.T., El-Din, M.G., Tierney, K.B., Blewett, T.A., 2021. A burning issue: The effect of organic ultraviolet filter exposure on the behaviour and physiology of *Daphnia magna*. Science of The Total Environment 750, 141707. https://doi.org/10.1016/j.scitotenv.2020.141707.

Bratkovics, S., Wirth, E., Sapozhnikova, Y., Pennington, P., & Sanger, D. (2015). Baseline monitoring of organic sunscreen compounds along South Carolina's coastal marine environment. Marine Pollution Bulletin, 101(1), 370–377. https://doi.org/10.1016/j.marpolbul.2015.10.015

Bui, XT, Vo, TPT, Ngo, HH, Guo, WS, and Nguyen, TT. Multicriteria assessment of advanced treatment technologies for micropollutants removal at large-scale applications. Sci. Total Environ., 563-564, 1050-1067, 2016

Butler, R.W. and Dodds, R. (2022), "Overcoming overtourism: a review of failure", Tourism Review, Vol. 77 No. 1, pp. 35-53. https://doi.org/10.1108/TR-04-2021-0215

Cabeza, Y., Candela, L., Ronen, D., & Teijon, G. (2012). Monitoring the occurrence of emerging contaminants in treated wastewater and groundwater between 2008 and 2010. The Baix Llobregat (Barcelona, Spain). Journal of hazardous materials, 239-240, 32–39. https://doi.org/10.1016/j.jhazmat.2012.07.032

Cabrera-Peralta, J. & Pena-Alvarez, A. (2018). Simple method for the determination of personal care product ingredients in lettuce by ultrasound-assisted extraction combined with solid-phase microextraction followed by GC-MS. Journal of Separation Science, 41(10), 2253–2260. https://doi.org/10.1002/jssc.201701244

Cadena-Aizaga, M.I., Montesdeoca-Esponda, S., Sosa-Ferrera, Z., Santana-Rodriguez, J.J. (2022) Occurrence and environmental hazard of organic UV filters in seawater and

836 wastewater from Gran Canaria Island (Canary Islands, Spain). Environmental Pollution
837 300: 118843. https://doi.org/10.1016/j.envpol.2022.118843

838 Campos, D., Gravato, C., Fedorova, G., Burkina, V., Soares, A., Pestana, J.L.T., 2017a.
839 Ecotoxicity of two organic UV-filters to the freshwater caddisfly *Sericostoma vittatum*.
840 Environmental Pollution, 228, 370-377. https://doi.org/10.1016/j.envpol.2017.05.021.
841
842 Campos, D., Gravato, C., Quintaneiro, C., Golovko, O., Zlabek, V., Soares, A., Pestana, J.L.T.,
843 2017b. Toxicity of organic UV-filters to the aquatic midge *Chironomus riparius*. Ecotoxicology
844 and Environmental Safety 143, 210-216. https://doi.org/10.1016/j.ecoenv.2017.05.005.
845

846 Cancer Council (2022) About sunscreen: when to use, how to apply.
847 https://www.cancer.org.au/cancer-information/causes-and-prevention/sun-safety/about-
848 sunscreen Accessed 17 March, 2022

849 Carve, M., Nugegoda, D., Allinson, G., Shimeta, J. (2021). A systematic review and ecological
850 risk assessment for organic ultraviolet filters in aquatic environments. Environmental
851 Pollution 268(Pt B), 115894. https://doi.org/10.1016/j.envpol.2020.115894
852
853 Casas-Beltrán DA, Febles-Moreno K, Hernandez-Yac E, Gallaher CM, Alvarado-Flores J, Leal-
854 Bautista RM, Lenczewski M. (2021). Impact of tourist behavior on the discharge of
855 sunscreen contamination in aquatic parks, sinkholes, and beaches of the Mexican
856 Caribbean. Applied Sciences, 11(15):6882. https://doi.org/10.3390/app11156882
857
858 CASTILLO-PAVÓN, Octavio; MÉNDEZ-RAMÍREZ, José Juan. (2017) Los desarrollos
859 turísticos y sus efectos medioambientales en la Riviera Maya, 1980-2015. Quivera
860 Revista de Estudios Territoriales, [S.l.], v. 19, n. 2, p. 101-118. ISSN 1405-8626

861 CDC (U.S. Centers for Disease Control) https://wwwnc.cdc.gov/travel/page/sun-exposure
862 Accessed March 17,2022

863 Chen, T.H., Wu, Y.T., Ding, W.H., 2016. UV-filter benzophenone-3 inhibits agonistic behavior
864 in male Siamese fighting fish (Betta splendens). Ecotoxicology 25(2), 302-309.
865 https://doi.org/10.1007/s10646-015-1588-4.
866
867 Choi, K.J., Kim, S.G., and Kim, S.H. (2008) Removal of antibiotics by coagulation and granular
868 activated carbon filtration. Journal of Hazardous Materials, 151, 38-43.
869 https://doi.org/10.1016/j.jhazmat.2007.05.059
870
871 Chong, K.L. (2020) The side effects of mass tourism: the voices of Bali islanders, Asia Pacific
872 Journal of Tourism Research, 25:2, 157-169, DOI: 10.1080/10941665.2019.1683591
873
874 Chu, S., Kwon, B. R., Lee, Y. M., Zoh, K. D., & Choi, K. (2021). Effects of 2-ethylhexyl-4-
875 methoxycinnamate (EHMC) on thyroid hormones and genes associated with thyroid,
876 neurotoxic, and nephrotoxic responses in adult and larval zebrafish (Danio rerio).
877 Chemosphere, 263, 128176. https://doi.org/10.1016/j.chemosphere.2020.128176
878

City of Honolulu (2022) https://www.honolulu.gov/rep/site/dpr/hanaumabay_docs/Hanauma_Bay_Attendance_Metrics_Dec_2020_to_Feb_2022_Honolulu_Parks_and_Rec.pdf Accessed 2 March, 2022.

Claudia, J. & Magrini, G. (2017). Cosmetic ingredients as emerging pollutants of environmental and health concern. A mini-review. Cosmetics, 4(2), 11–29. https://doi.org/10.3390/cosmetics4020011

Coronado, M., De Haro, H., Deng, X., Rempel, M. A., Lavado, R., & Schlenk, D. (2008). Estrogenic activity and reproductive effects of the UV-filter oxybenzone (2-hydroxy-4-methoxyphenyl-methanone) in fish. Aquatic toxicology 90(3), 182–187. https://doi.org/10.1016/j.aquatox.2008.08.018

Cui, R., Jong, M. C., You, L., Mao, F., Yao, D., Gin, K. Y., & He, Y. (2022). Size-dependent adsorption of waterborne Benzophenone-3 on microplastics and its desorption under simulated gastrointestinal conditions. Chemosphere, 286(Pt 3), 131735. https://doi.org/10.1016/j.chemosphere.2021.131735

Díaz-Cruz, M.S. & Barceló, D (2015) Personal Care Products in the Aquatic Environment, The Handbook of Environmental Chemistry, Vol 36.Springer, Switzerland.

Díaz-Cruz, M. S., Molins-Delgado, D., Serra-Roig, M. P., Kalogianni, E., Skoulikidis, N. T., & Barceló, D. (2019). Personal care products reconnaissance in EVROTAS river (Greece): Water-sediment partition and bioaccumulation in fish. The Science of the Total Environment, 651(Pt 2), 3079–3089. https://doi.org/10.1016/j.scitotenv.2018.10.008

Diffey B. (2001). Sunscreen isn't enough. Journal of Photochemistry and Photobiology. B, Biology, 64(2-3), 105–108. https://doi.org/10.1016/s1011-1344(01)00195-6

DiNardo, J. C. and Downs, C.A. (2019). Can oxybenzone cause Hirschsprung's disease? Reproductive Toxicology, 86, 98-100.

DiNardo, J.C., and Downs, C.A. (2021) Fall from GRASE: The sunsetting of the Sunscreen Innovation act. American Journal of Fermatological Research and Reviews 4:39.

Dodds, R. and Butler, R. (2019), "The phenomena of overtourism: a review", International Journal of Tourism Cities, Vol. 5 No. 4, pp. 519-528. https://doi.org/10.1108/IJTC-06-2019-0090

Downs CA, Woodley CM, Fauth JE, Knutson S, Burtscher MM, May LA, Avadanei AR, Higgins JL, Ostrander GK. (2011) A survey of environmental pollutants and cellular-stress markers of Porites astreoides at six sites in St. John, U.S. Virgin Islands. Ecotoxicology 20(8):1914-31. doi: 10.1007/s10646-011-0729-7.

Downs, C.A., Kramarsky-Winter, E., Segal, R., Fauth, J., Knnutson, S., Bronstein, O. et al. (2016). Toxicopathological effects of the sunscreen UV filter, oxybenzone (Benzophenone-3), on coral planulae and cultured primary cells and its environmental contamination in Hawaii and the U.S. Virgin Islands. *Archives of Environmental Contamination & Toxicology*, 70, 265-288. https://doi.org/10.1007/s00244-015-0227-7

Downs, C.A., Bishop, L., Diaz Cruz, M.S., Haghshenas, S.A., Stien, D., Rodrigues, A.M.S. et al. (2021a). Oxybenzone contamination from sunscreen pollution and its ecological threat to Hanauma Bay, Oahu, Hawaii, U.S.A. *Chemosphere*, 132880. https://doi.org/10.1016/j.chemosphere.2021.132880

Downs, C. A., DiNardo, J. C., Stien, D., Rodrigues, A., & Lebaron, P. (2021b). Benzophenone accumulates over time from the degradation of octocrylene in commercial sunscreen products. Chemical Research in Toxicology, 34(4), 1046–1054. https://doi.org/10.1021/acs.chemrestox.0c00461

Downs, C.A., Cruz, O.T., Remengesau, T.E. Jr (2022) Sunscreen pollution and tourism governance: science and innovation are necessary for biodiversity conservation and sustainable tourism. Aquatic Conservation: Marine and Freshwater Ecosystems. 1-11. https://doi.org/10.1002/aqc.3791

Du, B., He, Y., Liang, B., Li, J., Luo, D., Chen, H., Liu, L. Y., Guo, Y., & Zeng, L. (2022). Identification of triazine UV filters as an emerging class of abundant, ubiquitous pollutants in indoor dust and air from South China: Call for more concerns on their occurrence and human exposure. Environmental Science & Technology, 56(7), 4210-4220. https://doi.org/10.1021/acs.est.1c08909

Du, Y., Wang, W.Q., Pei, Z.T., Ahmad, F., Xu, R.R., Zhang, Y.M., Sun, L.W., 2017. Acute toxicity and ecological risk assessment of benzophenone-3 (BP-3) and benzophenone-4 (BP-4) in ultraviolet (UV)-filters. International Journal of Environmental Research and Public Health 14(11), 1414. https://doi.org/10.3390/ijerph14111414.

Eljarrat, E., Díaz-Cruz, M.S., Farré, M., de Alda, M.J.L., Petrović, M., Barceló, D. (2012). Analysis of Emerging Contaminants in Sewage Sludge. In: Vicent, T., Caminal, G., Eljarrat, E., Barceló, D. (eds) Emerging Organic Contaminants in Sludges. The Handbook of Environmental Chemistry, vol 24. Springer, Berlin, Heidelberg. https://doi.org/10.1007/698_2012_155

Emnet, P., Gaw, S., Northcott, G., Storey, B. & Craham, L. (2015). Personal care products and steroid hormones in the Antarctic coastal environment associated with two Antarctic research stations, McMurdo Station and Scott Base. Environmental Research, 136:331–342. https://doi.org/10.1016/j.envres.2014.10.019

Fel, J.-P., Lacherez, C., Bensetra, A., Mezzache, S., Béraud, E., Léonard, M., Allemand, D., Ferrier-Pagès, C., 2019. Photochemical response of the scleractinian coral *Stylophora pistillata* to some sunscreen ingredients. Coral Reefs 38(1), 109-122. https://doi.org/10.1007/s00338-018-01759-4.

Fitt, W.K., Hofmann, D.K., 2020. The effects of the UV-blocker oxybenzone (benzophenone-3) on planulae swimming and metamorphosis of the Scyphozoans *Cassiopea xamachana* and *Cassiopea frondosa*. Oceans 1(4), 174-180. https://doi.org/10.3390/oceans1040013.

Foubert, K., Dendooven, E., Theunis, M., Naessens, T., Ivanova, B., Pieters, L., Gilissen, L., Huygens, S., De Borggraeve, W., Lambert, J., Goossens, A., & Aerts, O. (2021). The presence of benzophenone in sunscreens and cosmetics containing the organic UV filter octocrylene: A laboratory study. Contact Dermatitis, 85(1), 69–77. https://doi.org/10.1111/cod.13845

Gago-Ferrero P, Díaz-Cruz MS, Barceló D (2011a) Fast pressurized liquid extraction with in-cell purification and analysis by liquid chromatography tandem mass spectrometry for the determination of UV filters and their degradation products in sediments. Anal Bioanal Chem 400:2195–2204.

Gago-Ferrero, P., Díaz-Cruz, M. S., & Barceló, D. (2011b). Occurrence of multiclass UV filters in treated sewage sludge from wastewater treatment plants. Chemosphere, 84(8), 1158–1165. https://doi.org/10.1016/j.chemosphere.2011.04.003

Gautam, K., Seth, M., Dwivedi, S., Jain, V., Vamadevan, B., Singh, D., Roy, S.K., Downs, C.A., Anbumani, S. (2022). Soil degradation kinetics of oxybenzone (Benzophenone-3) and toxicopathological assessment in the earthworm, *Eisenia fetida*, Environmental Research, https://doi.org/10.1016/j.envres.2022.113689.

Giraldo, A., Montes, R., Rodil, R., Quintana, J.B., Vidal-Linan, L., Beiras, R., 2017. Ecotoxicological evaluation of the UV filters ethylhexyl dimethyl p-aminobenzoic acid and octocrylene using marine organisms *Isochrysis galbana*, *Mytilus galloprovincialis* and *Paracentrotus lividus*. Archives of Environmental Contaminants and Toxicology 72(4), 606-611. https://doi.org/10.1007/s00244-017-0399-4.

Ghazipura, M., McGowan, R., Arslan, A., & Hossain, T. (2017). Exposure to benzophenone-3 and reproductive toxicity: A systematic review of human and animal studies. Reproductive Toxicology 73, 175–183. https://doi.org/10.1016/j.reprotox.2017.08.015

Glover, C. M., Liu, Y., & Liu, J. (2021). Assessing the risk from trace organic contaminants released via greywater irrigation to the aquatic environment. Water Research, 205, 117664. https://doi.org/10.1016/j.watres.2021.117664

Gorito, A. M., Ribeiro, A. R., Almeida, C., & Silva, A. (2017). A review on the application of constructed wetlands for the removal of priority substances and contaminants of emerging concern listed in recently launched EU legislation. Environmental Pollution 227, 428–443. https://doi.org/10.1016/j.envpol.2017.04.060

Guabiroba, H.C., Vilar, C.C., Pinheiro, H.T., Joyeux, J-C. (2022) Limited human access is linked to higher effectiveness in a marine sanctuary. Journal of Environmental Management 311:114838. https://doi.org/10.1016/j.jenvman.2022.114838

Guelfi, DR., Gozzi, F., Sires, I., Brillas, E., Machulek, A., de Oliviera, SC. Degradation of the insecticide propoxur by electrochemical advanced oxidation process using a boron-doped diamond/air-diffusion cell. Environ. Sci. Pollut. Res., 24, 6083-6095, 2017.

Hahladakis, J. N., Velis, C. A., Weber, R., Iacovidou, E., & Purnell, P. (2018). An overview of chemical additives present in plastics: Migration, release, fate and environmental impact during their use, disposal and recycling. Journal of hazardous materials, 344, 179–199. https://doi.org/10.1016/j.jhazmat.2017.10.014

Han, X., Lu, T., Hu, Y., Duan, J., Guan, Y., Huang, X., Zhou, J., Huang, R., Tang, M., Sun, R., Wang, Y., Song, L., Xia, Y., Wang, X., Chen, M., & Chen, T. (2022). A metabolomic study on the effect of prenatal exposure to Benzophenone-3 on spontaneous fetal loss in mice. Ecotoxicology and environmental safety, 233, 113347. https://doi.org/10.1016/j.ecoenv.2022.113347

He, K., Hain, E., Timm, A., Tarnowski, M., & Blaney, L. (2019). Occurrence of antibiotics, estrogenic hormones, and UV-filters in water, sediment, and oyster tissue from the Chesapeake Bay. The Science of the total environment, 650(Pt 2), 3101–3109. https://doi.org/10.1016/j.scitotenv.2018.10.021

Heerfordt, I. M., Philipsen, P. A., & Wulf, H. C. (2020). A Handful of Sunscreen for Whole-Body Application. Advances in experimental medicine and biology, 1268, 381–385. https://doi.org/10.1007/978-3-030-46227-7_19

He, T., Tsui, M.M.P., Tan, C.J., Ma, C.Y., Yiu, S.K.F., Wang, L.H., Chen, T.H., Fan, T.Y., Lam, P.K.S., Murphy, M.B., 2019a. Toxicological effects of two organic ultraviolet filters and a related commercial sunscreen product in adult corals. Environmental Pollution, 245, 462-471. https://doi.org/10.1016/j.envpol.2018.11.029.

Heerfordt, I. M., Philipsen, P. A., & Wulf, H. C. (2020). A handful of sunscreen for whole-body application. Advances in Experimental Medicine and Biology, 1268, 381–385. https://doi.org/10.1007/978-3-030-46227-7_19

Huo, W., Cai, P., Chen, M., Li, H., Tang, J., Xu, C., Zhu, D., Tang, W., & Xia, Y. (2016). The relationship between prenatal exposure to BP-3 and Hirschsprung's disease. Chemosphere, 144, 1091–1097. https://doi.org/10.1016/j.chemosphere.2015.09.019

Imamović , B.; Trebše, P.; Omeragić, E.; Bečić, E.; Pečet, A.; Dedić, M. (2022). Stability and removal of benzophenone-type UV filters from water matrices by Advanced Oxidation Processes. Molecules, 27, 1874. https://doi.org/10.3390/molecules27061874

Inui, M., Adachi, T., Takenaka, S., Inui, H., Nakazawa, M., Ueda, M., Watanabe, H., Mori, C., Iguchi, T., & Miyatake, K. (2003). Effect of UV screens and preservatives on vitellogenin and choriogenin production in male medaka (Oryzias latipes). Toxicology, 194(1-2), 43–50. https://doi.org/10.1016/s0300-483x(03)00340-8

Inyang, M. and Dickenson, ER., The use of carbon adsorbents for the removal of perfluroalkyl acids from potable reuse systems. Chemosphere, 184, 168-175, 2017

Jurado, A., Gago-Ferrero, P., Vàzquez-Suñé, E., Carrera, J., Pujades, E., Díaz-Cruz, M. S., & Barceló, D. (2014). Urban groundwater contamination by residues of UV filters. Journal of hazardous materials, 271, 141–149. https://doi.org/10.1016/j.jhazmat.2014.01.036

Jung, J.W., Kang, J.S., Choi, J., Park, J.W., 2021. A novel approach to derive the predicted no-effect concentration (PNEC) of benzophenone-3 (BP-3) using the species sensitivity distribution (SSD) method: suggestion of a new PNEC value for BP-3. International Journal of Environmental Research and Public Health 18(7). https://doi.org/10.3390/ijerph18073650.

Kainthila, S., Tiwari, P., Chowdhary, N. (2022) Overtourism: striking a balance between delimiting and managing tourism growth in Asia. In: Sharma, A., Hassan, A. (eds) Future of Tourism in Asia. Springer, Singapore. https://doi.org/10.1007/978-981-16-1669-3_14

Kaiser, D., Sieratowicz, A., Zielke, H., Oetken, M., Hollert, H., & Oehlmann, J. (2012). Ecotoxicological effect characterisation of widely used organic UV filters. Environmental Pollution 163, 84–90. https://doi.org/10.1016/j.envpol.2011.12.014

Kinnberg, K.L., Petersen, G.I., Albrektsen, M., Minghlani, M., Awad, S.M., Holbech, B.F., Green, J.W., Bjerregaard, P., Holbech, H., 2015. Endocrine-disrupting effect of the ultraviolet filter benzophenone-3 in zebrafish, *Danio rerio*. Environmental Toxicology and Chemistry 34(12), 2833-2840.

Ko, H., An, S., Ahn, S., Park, I. G., Gong, J., Hwang, S. Y., Oh, S., Ki, M. W., Jin, S. H., Choi, W. J., & Noh, M. (2022). Sunscreen filter octocrylene is a potential obesogen by acting as a PPARγ partial agonist. Toxicology letters, 355, 141–149. https://doi.org/10.1016/j.toxlet.2021.12.001

Lee, S.-H., Xiong, J.-Q., Ru, S., Patil, S.M., Kurade, M.B., Govindwar, S.P., Oh, S.-E., Jeon, B.-H., 2020. Toxicity of benzophenone-3 and its biodegradation in a freshwater microalga *Scenedesmus obliquus*. Journal of Hazardous Materials, 389, 122149. https://doi.org/10.1016/j.jhazmat.2020.122149.

Lee, I., Lee, J., Jung, D., Kim, S., & Choi, K. (2019). Two-generation exposure to 2-ethylhexyl 4-methoxycinnamate (EHMC) in Japanese medaka (Oryzias latipes) and its reproduction and endocrine related effects. Chemosphere, 228, 478–484. https://doi.org/10.1016/j.chemosphere.2019.04.123

Lingberg, K., & McCool, S.F. (1998) A critique of environmental carrying capacity as a means of managing the effects of tourism development. Environmental Conservation 25:291-292. https://doi.org/10.1017/S0376892998000368

Li, W., Ma, Y., Guo, C., Hu, W., Liu, K., Wang, Y., & Zhu, T. (2007). Occurrence and behavior of four of the most used sunscreen UV filters in a wastewater reclamation plant. Water Research, 41(15), 3506–3512. https://doi.org/10.1016/j.watres.2007.05.039

Liu, Y., Wang, Y., Li, N., & Jiang, S. (2022). Avobenzone and nanoplastics affect the development of zebrafish nervous system and retinal system and inhibit their locomotor behavior. The Science of the Total Environment, 806(Pt 2), 150681. https://doi.org/10.1016/j.scitotenv.2021.150681

Loraine, G. A., & Pettigrove, M. E. (2006). Seasonal variations in concentrations of pharmaceuticals and personal care products in drinking water and reclaimed wastewater in southern California. Environmental science & technology, 40(3), 687–695. https://doi.org/10.1021/es051380x

Lou, T., Xie, H., Chen, G., & Gagné, J. P. (2006). Effects of photodegradation of dissolved organic matter on the binding of benzo(a)pyrene. Chemosphere, 64(7), 1204–1211. https://doi.org/10.1016/j.chemosphere.2005.11.043

Mainardis, M., Buttazzoni, M., De Bortoli, N., Mion, M. y Goi, D. (2020) 'Evaluation of ozonation applicability to pulp and paper streams for a sustainable wastewater treatment', Journal of Cleaner Production, 258, 120781. https://doi.org/10.1016/j.jclepro.2020.120781

Mandaric, L., Diamantini, E., Stella, E., Cano-Paoli, K., Valle-Sistac, J., Molins-Delgado, D., Bellin, A., Chiogna, G., Majone, B., Diaz-Cruz, M. S., Sabater, S., Barcelo, D., & Petrovic, M. (2017). Contamination sources and distribution patterns of pharmaceuticals and personal care products in Alpine rivers strongly affected by tourism. The Science of the total environment, 590-591, 484–494. https://doi.org/10.1016/j.scitotenv.2017.02.185

Mao, F., He, Y., Kushmaro, A., & Gin, K. Y. (2017). Effects of benzophenone-3 on the green alga *Chlamydomonas reinhardtii* and the cyanobacterium *Microcystis aeruginosa*. Aquatic Toxicology, 193, 1–8. https://doi.org/10.1016/j.aquatox.2017.09.029

Martinez-Huitle C.A. and Ferro S. (2006): Electrochemical oxidation of organic pollutants for the wastewater treatment: direct and indirect processes. Chemical Society Review, 35,1324–1340. https://doi.org/10.1039/B517632H

Martinez-Huitle C.A. and Panizza, M. (2018). Electrochemical oxidation of organic pollutants for wastewater treatment. Current Opinion in Electrochemistry, 11, 62-71 https://doi.org/10.1016/j.coelec.2018.07.010

McCool S.F., and Lime, D.W. (2001) Tourism Carrying Capacity: Tempting Fantasy or Useful Reality?, Journal of Sustainable Tourism, 9:5, 372-388, DOI: 10.1080/09669580108667409

Meng, Q., Yeung, K., Kwok, M.L., Chung, C.T., Hu, X.L., Chan, K.M., 2020. Toxic effects and transcriptome analyses of zebrafish (*Danio rerio*) larvae exposed to benzophenones. Environmental Pollution, 265(Pt A), 114857. https://doi.org/10.1016/j.envpol.2020.114857.

Miller, I.B., Moeller, M., Kellermann, M.Y., Nietzer, S., Di Mauro, V., Kamyab, E., Pawlowski, S., Petersen-Thiery, M., Schupp, P.J., 2022. Towards the development of standardized bioassays for corals: Acute toxicity of the UV filter benzophenone-3 to scleractinian coral larvae. Toxics 10(5), 244. https://doi.org/10.3390/toxics10050244.

Molins-Delgado, D., Gago-Ferrero, P., Díaz-Cruz, M. S., & Barceló, D. (2016). Single and joint ecotoxicity data estimation of organic UV filters and nanomaterials toward selected aquatic organisms. Urban groundwater risk assessment. Environmental Research, 145, 126–134. https://doi.org/10.1016/j.envres.2015.11.026

Molins-Delgado, D., Távora, J., Silvia Díaz-Cruz, M., & Barceló, D. (2017). UV filters and benzotriazoles in urban aquatic ecosystems: The footprint of daily use products. The Science of the total environment, 601-602, 975–986. https://doi.org/10.1016/j.scitotenv.2017.05.176

Mopper, K., Stubbins, A., Ritchie, J. D., Bialk, H. M., & Hatcher, P. G. (2007). Advanced instrumental approaches for characterization of marine dissolved organic matter: extraction techniques, mass spectrometry, and nuclear magnetic resonance spectroscopy. Chemical Reviews, 107(2), 419–442. https://doi.org/10.1021/cr050359b

Morin-Crini, N., Lichtfouse, E., Fourmentin, M. et al. (2022). Removal of emerging contaminants from wastewater using advanced treatments. A review. Environmental Chemistry Letters 20, 1333–1375. https://doi.org/10.1007/s10311-021-01379-5

Na, J., Song, J., Achar, J. C., & Jung, J. (2021). Synergistic effect of microplastic fragments and benzophenone-3 additives on lethal and sublethal toxicity. Journal of Hazardous Materials 402: 123845. https://doi.org/10.1016/j.jhazmat.2020.123845

NOAA - U.S. National Oceanic & Atmospheric Administration. (2002). A national coral reef action strategy. Report to Congress on implementation of the coral reef conservation act of 2000 and the national action plan to conserve coral reefs in 2002–2003. Available at: https://www.coris.noaa.gov/activities/actionstrategy/action_reef_final.pdf

Ochoa-Herrera, V., and Sierra-Alvarez, R., Removal of perfluorinated surfactants by sorption onto granular activated carbon, zeolite and sludge. Chemosphere, 72, 1588-1593, 2008

O'Donovan, S., Mestre, N. C., Abel, S., Fonseca, T. G., Carteny, C. C., Willems, T., Prinsen, E., Cormier, B., Keiter, S. S., & Bebianno, M. J. (2020). Effects of the UV filter, oxybenzone, adsorbed to microplastics in the clam Scrobicularia plana. The Science of the total environment, 733, 139102. https://doi.org/10.1016/j.scitotenv.2020.139102

O'Malley, E., McLachlan, M. S., O'Brien, J. W., Verhagen, R., & Mueller, J. F. (2021). The presence of selected UV filters in a freshwater recreational reservoir and fate in

controlled experiments. The Science of the total environment, 754, 142373. https://doi.org/10.1016/j.scitotenv.2020.142373

Pablos, M.V., Garcia-Hortiguela, P., Fernandez, C., 2015. Acute and chronic toxicity of emerging contaminants, alone or in combination, in *Chlorella vulgaris* and *Daphnia magna*. Environmental Science and Pollution Research International, 22(7), 5417-5424. https://doi.org/10.1007/s11356-015-4119-1.

Paredes, E., Perez, S., Rodil, R., Quintana, J.B., Beiras, R., 2014. Ecotoxicological evaluation of four UV filters using marine organisms from different trophic levels Isochrysis galbana, *Mytilus galloprovincialis, Paracentrotus lividus*, and *Siriella armata*. Chemosphere 104, 44-50. https://doi.org/10.1016/j.chemosphere.2013.10.053.

Pegoraro, C. N., Harner, T., Su, K., & Ahrens, L. (2020). Occurrence and Gas-Particle Partitioning of Organic UV-Filters in Urban Air. Environmental science & technology, 54(20), 12881–12889. https://doi.org/10.1021/acs.est.0c02665

Plagellat, C., Kupper, T., Furrer, R., de Alencastro, L. F., Grandjean, D., & Tarradellas, J. (2006). Concentrations and specific loads of UV filters in sewage sludge originating from a monitoring network in Switzerland. Chemosphere, 62(6), 915–925. https://doi.org/10.1016/j.chemosphere.2005.05.024

Poh Agin P. (2006). Water resistance and extended wear sunscreens. Dermatologic clinics, 24(1), 75–79. https://doi.org/10.1016/j.det.2005.08.002

Ponting, J. & O'Brien, D. (2015) Regulating "Nirvana": sustainable surf tourism in a climate of increasing regulation. Sport Management Review 18:99-110. https://doi.org/10.1016/j.smr.2014.07.004

Puccetti, G., & Fares, H. (2014). A new approach for evaluating the water resistance of sunscreens on consumers: tap water vs. salt water vs. chlorine water. International journal of cosmetic science, 36(3), 284–290. https://doi.org/10.1111/ics.12126

Raimondi, S., & Forbes, V.E. (2022). Moving beyond risk quotients: advancing ecological risk assessment to reflect better, more robust and relevant methods. Ecologies 3:145-160. https://doi.org/10.3390/ecologies3020012

Rani, M., Shim, W. J., Han, G. M., Jang, M., Song, Y. K., & Hong, S. H. (2017). Benzotriazole-type ultraviolet stabilizers and antioxidants in plastic marine debris and their new products. The Science of the total environment, 579, 745–754. https://doi.org/10.1016/j.scitotenv.2016.11.033

Ramos, S., Homem, V., & Santos, L. (2021). Uptake and translocation of UV-filters and synthetic musk compounds into edible parts of tomato grown in amended soils. The Science of the total environment, 792, 148482. https://doi.org/10.1016/j.scitotenv.2021.148482

Rehfeld, A., Egeberg, D. L., Almstrup, K., Petersen, J. H., Dissing, S., & Skakkebæk, N. E. (2018). EDC IMPACT: Chemical UV filters can affect human sperm function in a progesterone-like manner. Endocrine connections, 7(1), 16–25. https://doi.org/10.1530/EC-17-0156

Rizzo, L., Malato, S., Antakyali, D., Beretsou, V. G., Đolić, M. B., Gernjak, W., Heath, E., Ivancev-Tumbas, I., Karaolia, P., Lado Ribeiro, A. R., Mascolo, G., McArdell, C. S., Schaar, H., Silva, A., & Fatta-Kassinos, D. (2019). Consolidated vs new advanced treatment methods for the removal of contaminants of emerging concern from urban wastewater. The Science of the total environment, 655, 986–1008. https://doi.org/10.1016/j.scitotenv.2018.11.265

Santa-Viera, S., Montesdeoca-Esponda, S., Sosa-Ferrera, Z., Santana-Rodrigues, J.J. (2021) UV filters and UV stabilisers adsorbed in microplastic debris from beach sand. Marine Pollution Bulletin 168:112434. https://doi.org/10.1016/j.marpolbul.2021.112434

Schaffer, M., Boxberger, N., Börnick, H., Licha, T., & Worch, E. (2012). Sorption influenced transport of ionizable pharmaceuticals onto a natural sandy aquifer sediment at different pH. Chemosphere, 87(5), 513–520. https://doi.org/10.1016/j.chemosphere.2011.12.053

Schlumpf, M., Schmid, P., Durrer, S., Conscience, M., Maerkel, K., Henseler, M. et al. (2004). Endocrine activity and developmental toxicity of cosmetic UV filters – an update. *Toxicology*, 205(1-2), 113-122. https://doi.org/10.1016/j.tox.2004.06.043

Schlumpf, M., Durrer, S., Faas, O., Ehnes, C., Fuetsch, M., Gaille, C. et al. (2008). Developmental toxicity of UV filters and environmental exposure: a review. *International Journal of Andrology*, 31(2), 144-151. https://doi.org/10.1111/j.1365-2605.2007.00856.x

Schmitt, C., Oetken, M., Dittberner, O., Wagner, M., Oehlmann, J., 2008. Endocrine modulation and toxic effects of two commonly used UV screens on the aquatic invertebrates *Potamopyrgus antipodarum* and *Lumbriculus variegatus*. Environmental Pollution, 152(2), 322-329. https://doi.org/10.1016/j.envpol.2007.06.031.

Seoane, M., Cid, A., Herrero, C., Esperanza, M., 2020. Comparative acute toxicity of benzophenone derivatives and bisphenol analogues in the Asian clam *Corbicula fluminea*. Ecotoxicology. https://doi.org/10.1007/s10646-020-02299-wSerra-Roig, M. P., Jurado, A., Díaz-Cruz, M. S., Vázquez-Suñé, E., Pujades, E., & Barceló, D. (2016). Occurrence, fate and risk assessment of personal care products in river-groundwater interface. The Science of the total environment, 568, 829–837. https://doi.org/10.1016/j.scitotenv.2016.06.006

Shang, J., Chen, J., Shen, Z., Xiao, X., Yang, H., Wang, Y., & Ruan, A. (2015). Photochemical degradation of PAHs in estuarine surface water: effects of DOM, salinity, and suspended particulate matter. Environmental science and pollution research international, 22(16), 12374–12383. https://doi.org/10.1007/s11356-015-4543-2

Shoeib, M., Schuster, J., Rauert, C., Su, K., Smyth, S. A., & Harner, T. (2016). Emission of poly and perfluoroalkyl substances, UV-filters and siloxanes to air from wastewater treatment plants. Environmental Pollution 218: 595–604. https://doi.org/10.1016/j.envpol.2016.07.043

Singh, S. (2015) What's wrong with carrying capacity for Tourism? Tourism Recreation Research 31:67-72. https://doi.org/10.1080/02508281.2006.11081263

Sivamani, R. K., Ghiya, M., & Maibach, H. I. (2010). Shedding light on sunscreens and their labels: Testing policies need to match actual use. American Journal of Preventive Medicine, 38(6), 679–681. https://doi.org/10.1016/j.amepre.2010.02.009

Song, J., Na, J., An, D. & Jung, J. (2021). Role of benzophenone-3 additive in chronic toxicity of polyethylene microplastic fragments to *Daphnia magna*. *Science of the Total Environment*, 800, 149638. https://doi.org/10.1016/j.scitotenv.2021.149638

Stien, D., Clergeud, F., Rodrigues, A.M.S., Lebaron, K., Pillot, R., Romans, P. et al. (2019). Metabolics reveal that octocrylene accumulates in *Pocillopora damicornis* tissues as fatty acid conjugates and triggers coral cell mitochondrial dysfunction. *Analytical Chemistry,* 91(1), 990-995. https://doi.org/10.1021/acs.analchem.8b04187

Stokes, R. P., & Diffey, B. L. (1999). The water resistance of sunscreen and day-care products. The British journal of dermatology, 140(2), 259–263. https://doi.org/10.1046/j.1365-2133.1999.02659.x

Stoye, E. (2017). Magnetic beads mop up subblock in the sea. https://www.chemistryworld.com/news/magnetic-beads-mop-up-sunblock-in-the-sea/3007881.article Article accessed 3 March, 2022.

Sun, J., Ma, Y., Qin, H., Li, Z., Pan. L. (2022) An integrated approach using chemical ecological risk assessment and multi-integrated biomarker indexes approach to assess pollution: a case study of Ruditapes phillippinarum in four bays on the Shandong Peninsula in China. Environmental Research 203: 111793. https://doi.org/10.1016/j.envres.2021.111793

Sunyer-Caldú, A., & Diaz-Cruz, M. S. (2021). Development of a QuEChERS-based method for the analysis of pharmaceuticals and personal care products in lettuces grown in field-scale agricultural plots irrigated with reclaimed water. Talanta, 230, 122302. https://doi.org/10.1016/j.talanta.2021.122302

Tacconi, L., and Williams, D.A. (2020) Corruption and anti-corruption in environmental and resource management. Annual Review of Environment and Resources 45:305-329. https://doi.org/10.1146/annurev-environ-012320-083949

Tao, J., Bai, C., Chen, Y., Zhou, H., Liu, Y., Shi, Q., Pan, W., Dong, H., Li, L., Xu, H., Tanguay, R., Huang, C., Dong, Q., 2020. Environmental relevant concentrations of benzophenone-3 induced developmental neurotoxicity in zebrafish. Science of the Total Environment, 721, 137686. https://doi.org/10.1016/j.scitotenv.2020.137686.

Thomas, B.N., Pigozzi, B.W., Sambrook, R.A. (2005) Tourist carrying capacity measures: Crowding Syndrome in the Caribbean. The Professional Geographer 57:13-20. https://doi.org/10.1111/j.0033-0124.2005.00455.x

Thomas, L.R., DiNardo, G.T., Lee, H-H., Piner, K.R., Kahng, S.E. (2013). Factors influencing the distribution of Kona crabs Ranina ranina catch rates in the main Hawaiian Islands. Journal of Crustacean Biology 33(5):633-640. https://doi.org/ 10.1163/1937240X-00002171

Thorel, E., Clergeaud, F., Jaugeon, L., Rodrigues, A., Lucas, J., Stien, D., & Lebaron, P. (2020). Effect of 10 UV Filters on the Brine Shrimp Artemia salina and the Marine Microalga Tetraselmis sp. Toxics, 8(2), 29. https://doi.org/10.3390/toxics8020029

Tian, L., Huang, L., Cui, H., Yang, F., & Li, Y. (2021). The toxicological impact of the sunscreen active ingredient octinoxate on the photosynthesis activity of Chlorella sp. Marine Environmental Research, *171*, 105469. https://doi.org/10.1016/j.marenvres.2021.105469

Titcomb, M., Fellows, D.B., Pukui, M.K., Devaney, D.M. (1978) Native use of marine invertebrates in Old Hawaii. Pacific Sciences, 32(4):325-386.

Tsui, M.M.P., Leung, H.W., Wai, T-C., Yamashita, N., Taniyasu, S., Liu, W. et al. (2014). Occurrence, distribution, and ecological risk assessment of multiple classes of UV filters in surface waters from different countries. *Water Research*, 67, 55-65. https://doi.org/10.1016/j.watres.2014.09.013

U.S. Federal Water Pollution Control Act Amendments of 1972. https://www.govinfo.gov/content/pkg/STATUTE-86/pdf/STATUTE-86-Pg816.pdf Accessed 28 March, 2022

U.S. EPA (2022) National Pollutant Discharge Elimination System. https://www.epa.gov/npdes Accessed 28 March, 2022.

U.S NOAA (2022) Point Source. https://oceanservice.noaa.gov/education/tutorial_pollution/03pointsource.html Accessed 28 March, 2022.

Valhondo, C., Martínez-Landa, L., Carrera, J., Díaz-Cruz, S. M., Amalfitano, S., & Levantesi, C. (2020). Six artificial recharge pilot replicates to gain insight into water quality enhancement processes. Chemosphere, 240, 124826. https://doi.org/10.1016/j.chemosphere.2019.124826

Vieira Sanches, M., Oliva, M., De Marchi, L., Cuccaro, A., Puppi, D., Chiellini, F., Freitas, R., Pretti, C., 2021. Ecotoxicological screening of UV-filters using a battery of marine bioassays. Environmental Pollution. https://doi.org/10.1016/j.envpol.2021.118011.

Wall, G. (2020). From carrying capacity to overtourism: a perspective article. Tourism Review 75:212-215.

Wan, Y., Xue, J., & Kannan, K. (2015). Occurrence of benzophenone-3 in indoor air from Albany, New York, USA, and its implications for inhalation exposure. The Science of the total environment, 537, 304–308. https://doi.org/10.1016/j.scitotenv.2015.08.020

Wang, W., & Kannan, K. (2017). Mass loading and emission of benzophenone-3 (BP-3) and its derivatives in wastewater treatment plants in New York State, USA. The Science of the total environment, 579, 1316–1322. https://doi.org/10.1016/j.scitotenv.2016.11.124

Wang, J., Meng, X., Feng, C., Xiao, J., Zhao, X., Xiong, B., & Feng, J. (2021). Benzophenone-3 induced abnormal development of enteric nervous system in zebrafish through

MAPK/ERK signaling pathway. Chemosphere, 280, 130670. https://doi.org/10.1016/j.chemosphere.2021.130670

Whitehouse, B. (1985). The effects of dissolved organic matter on the aqueous partitioning og polynuclear aromatic hydrocarbons. Esturine, Coastal and Shelf Science 20(4):393-402. https://doi.org/10.1016/0272-7714(85)90084-8

Wijgerde, T., van Ballegooijen, M., Nijland, R., van der Loos, L., Kwadijk, C., Osinga, R. et al. (2020). Adding insult to injury: effects of chronic oxybenzone exposure and elevated temperature on two reef-building corals. *Science of the Total Environment*, 1, 139030. https://doi.org/10.1016/j.scitotenv.2020.139030

Wnuk, A., Rzemieniec, J., Staron, J., Litwa, E., Lason, W., Bojarski, A. et al. (2018). Prenatal exposure to benzophenone-3 impairs autophagy, disrupts RXRs/PPARγ signaling, and alters epigenetic and post-translational statuses in brain neurons. *Molecular Neurobiology*, 56, 4820-4837. https://doi.org/10.1007/s12035-018-1401-5

Xiao, H., Lv, B., Gao, J. and Zhao, G. (2016). Hydrothermal electrocatalytic oxidation for the treatment of herbicides wastewater. Environmental Science and Pollution Research, 23, 10050-10057. https://doi.org/10.1007/s11356-016-6218-z

Xu, M, Zheng, D. & Gong, S. (2021). Effects of low concentration benzophenone-3 exposure on the sex ratio and offspring development of zebrafish (*Danio rerio*). *Bulletin of Environmental Contamination & Toxicology*, 106, 740-746. https://doi.org/10.1007/s00128-021-03166-y

Yan, S., Liang, M., Chen, R., Hong, X., Zha, J., 2020. Reproductive toxicity and estrogen activity in Japanese medaka (*Oryzias latipes*) exposed to environmentally relevant concentrations of octocrylene. Environmental Pollution, 261, 114104. https://doi.org/10.1016/j.envpol.2020.114104.

Yang,C., Lim, W., Bazer, F.W., Song, G., (2018) Homosalate aggravates the invasion of human trophoblast cells as well as regulates intracellular signaling pathways including PI3K/AKT and MAPK pathways. Environmental Pollution 243:1263-1273. https://doi.org/10.1016/j.envpol.2018.09.092

Yazar, S., Ertekin, S.K. (2019) Assessment of the cytotoxicity and genotoxicity of homosalate in MCF-7. Journal of Cosmetic Dematology 19 :246-252. https://doi.org/10.1111/jocd.12973

Zhang, P., Lu, G., Liu, J., Yan, Z., Wang, Y., 2020. Toxicological responses of *Carassius auratus* induced by benzophenone-3 exposure and the association with alteration of gut microbiota. Science of the Total Environment, 747, 141255. https://doi.org/10.1016/j.scitotenv.2020.141255.

Zhang, Z., Ren, N., Li, Y. F., Kunisue, T., Gao, D., & Kannan, K. (2011). Determination of benzotriazole and benzophenone UV filters in sediment and sewage sludge. Environmental science & technology, 45(9), 3909–3916. https://doi.org/10.1021/es2004057

Zhong, X., Downs, C.A.,Che, X., Zhang, Z., Li, Y., Liu, B. et al. (2019a). The toxicological effects of oxybenzone, an active ingredient in suncream personal care products, on

prokaryotic *Arthrospira* sp. and eukaryotic alga *Chlorella* sp. *Aquatic Toxicology*, 216, 105295. https://doi.org/10.1016/j.aquatox.2019.105295

Zhong, X., Li, Y., Che, X., Zhang, Z., Li, Y., Liu, B., Li, Q., & Gao, H. (2019b). Significant inhibition of photosynthesis and respiration in leaves of Cucumis sativus L. by oxybenzone, an active ingredient in sunscreen. Chemosphere, 219, 456–462. https://doi.org/10.1016/j.chemosphere.2018.12.019

Zhong, X., Downs, C.A., Li, Y., Zhang, Z., Li, Y., Liu, B. et al. (2020). Comparison of toxicological effects of oxybenzone, avobenzone, octocrylene, and octinoxate sunscreen ingredients on cucumber plants. *Science of the Total Environment*, 714, 136879. https://doi.org/10.1016/j.scitotenv.2020.136879

Figure and Table Legends

Figure 1. Sample site location on Oahu, Hawaii, U.S.A. and concentration of UV-filter target analytes in nanogram of target analyte per gram of sand (dry wt).

Figure 2. *Panel A* – sampling area for Maui, Hawaii, U.S.A. *Panel B* – Location and concentration of UV-filter target analytes in nanogram of target analyte per gram of sand (dry wt).

Figure 3. *Panel A* – Sampling area location on the Island of Hawai'i, Hawaii, U.S.A. *Panel B* – Sampling locations for the beach shower at Waialea Bay that serves the Waialea Bay Marine and Land Conservation District and concentration of UV-filter target analytes in nanogram of target analyte per gram of sand (dry wt). *Panel C* – Sampling location of Waialea Bay water samples and concentration of UV-filter target analytes in nanogram of target analyte per liter of water. *Panel D* – Sampling location within Kealakekua Bay Marine and Land Conservation District and concentration of UV-filter target analytes in nanogram of target analyte per liter of water.

Figure 4. *Panel A* - Sampling locations for the beach shower at Mauna Lani Bay's beach that serves the Island of Hawai'i, Hawaii, U.S.A. and concentration of UV-filter target analytes in nanogram of target analyte per gram of sand (dry wt). *Panel B* - Sampling locations for the beach shower at 49 Black Sand Beach on the Island of Hawai'i, Hawaii, U.S.A. and concentration of UV-filter target analytes in nanogram of target analyte per gram of sand (dry wt). *Panel C* - Sampling locations for the beach shower at Kahalu'u Bay's beach that serves the Island of Hawai'i, Hawaii, U.S.A. and concentration of UV-filter target analytes in nanogram of target analyte per gram of sand (dry wt). *Panel D* - Sampling locations for collecting water samples at Kahalu'u Bay in the Island of Hawai'i, Hawaii, U.S.A. and concentration of UV-filter target analytes in nanogram of target analyte per liter of water.

Table 1. Risk Quotient for Acute Toxicity of oxybenzone and octocrylene in sand samples collected in the State of Hawaii associated with Figures 1-4 using the European Union method for Cnidarian species, invertebrate (non-Cnidarian) species, plant and algae species, and fish species. Color chart: RED= Severe condition for a potential toxic effect ≥1; Orange = Moderate threat condition for a potential toxic effect = 0.5 to 1.0; Yellow= Condition of concern 0.49 to 0.1.

Table 2. Risk Quotient for Acute Toxicity of oxybenzone and octocrylene in water samples collected in the State of Hawaii associated with Figures 1-4 using the European Union method for Cnidarian species, invertebrate (non-Cnidarian) species, plant and algae species, and fish species. Color chart: RED= Severe condition for a potential toxic effect ≥1; Orange = Moderate threat condition for a potential toxic effect = 0.5 to 1.0; Yellow= Condition of concern 0.49 to 0.1.