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Synthetic ultraviolet light filtering chemical contamination of coastal waters of Virgin Islands national park, St. John, U.S. Virgin Islands



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ABSTRACT

Contamination of surface waters by synthetic ultraviolet light (UV) filtering chemicals is a concern for the Virgin Islands National Park (VINP). Discrete water samples were collected from VINP bays to determine UV filter chemical presence in the coastal waters. Spatial distribution and the potential for partitioning between subsurface waters and the sea surface microlayer (SML) were also examined. The UV filter chemicals 4-methylbenzylidene camphor, benzophenone-3, octinoxate, homosalate, and octocrylene were detected at concentrations up to 6073 ng/L (benzophenone-3). Concentrations for benzophenone-3 and homosalate declined exponentially ($r^2 = 0.86$ to 0.98) with distance from the beach. Limited data indicate that some UV filter chemicals may partition to the SML relative to the subsurface waters. Contamination of VINP coastal waters by UV filter chemicals may be a significant issue, but an improved understanding of the temporal and spatial variability of their concentrations would be necessary to better understand the risk they present.

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1. Introduction

Contaminants of emerging concern are those chemicals that have no regulatory standard and whose presence in natural waters at potentially deleterious concentrations is becoming more apparent (USEPA, 2008). Synthetic ultraviolet light filtering (UV filter) chemicals are among the many chemicals of emerging concern. They are incorporated within many cosmetic and plastic products to minimize the damaging effects of ultraviolet light (Giokas et al., 2007; Diaz-Cruz et al., 2008). Detection of several UV filter chemicals in human placental tissue indicates these chemicals are absorbed into the human body (liménez-Díaz et al., 2013) and may be excreted into septic systems and be a component of municipal waste waters. Other studies have indicated that UV filters washing off the skin during recreational activities (i.e. swimming) can be a significant source for surface waters (Giokas et al., 2005; Lanford and Thomas, 2008; Sankado et al., 2015). As a result, UV filter presence in the environment is being widely reported (Balmer et al., 2005; Buser et al., 2006; Fent et al., 2010; Kameda et al., 2011; Rodil et al., 2012; Magi et al., 2013).

Concentrations for several of the UV filters are being reported in multiple aquatic ecosystem compartments (i.e., sediment, water, and biota). Perhaps the compartment for which UV filter contamination is best documented is surface water where the reported concentrations range from 0.5–6,812 ng/L (Balmer et al., 2005; Rodil et al., 2008; Fent

* Corresponding author. E-mail address: tbargar@usgs.gov (T.A. Bargar). Nguyen et al., 2011; Tovar-Sanchez et al., 2013; Paredes et al., 2014; Tsui et al., 2014). The highest reported concentrations were from marine waters near high use public beaches (Tsui et al., 2014). Contamination of aquatic biota is reported less often, but demonstrates contamination throughout the food chain. For example, the UV filter chemicals 4-MBC (4-methylbenzylidene camphor), BP-3 (benzophenone-3), EHMC (ethylhexyl methoxy cinnamate), and OC (octocrylene) were detected in fish from freshwater lakes (Balmer et al., 2005) and small rivers (Buser et al., 2006) in Switzerland. In addition, based on data from passive semipermeable membrane devices (SPMD). OC and 4-MBC showed relatively greater capacity for bioconcentration (Balmer et al., 2005). In fact, several UV filter chemicals have $\log K_{OW}$ values greater than 5 indicating the potential for bioconcentration (Diaz-Cruz et al., 2008; Gago-Ferrero et al., 2012). Further, Fent et al. (2010) reported EHMC in tissues from organisms over multiple trophic levels from benthic invertebrates (crustaceans - Gammarus sp.) to birds (cormorants). Lastly, Kameda et al. (2011) reported numerous UV filter chemicals in sediments from Japanese rivers and lakes. A number of toxicological responses have been reported for aquatic

et al., 2010; Bratkovics and Sapozhnikova, 2011; Kameda et al., 2011;

A number of toxicological responses have been reported for aquatic organisms exposed to UV filter chemicals. The most often reported effects are associated with endocrine disruption including vitellogenin induction (EHMC, 4-MBC, ethyl-4-para-aminobenzoic acid [Et-PABA], and BP-3 as reported in Inui et al., 2003; Kunz and Fent, 2006; Coronado et al., 2008), altered fecundity (BP-3 reported in Coronado et al., 2008), and steroid hormone receptor agonism or antagonism (4-MBC, EHMC, OD-PABA, and homosalate [HMS] reported in Schreurs







et al., 2002; Inui et al., 2003; Ozaez et al., 2013). Other reported effects include altered development in exposed invertebrates (BP-3 and 3-benzyldene camphor reported in Scheil et al., 2008; Paredes et al., 2014) and oxidative stress in corals (benzophenone-2 reported in Downs et al., 2014). The effect concentration of 246 ng/L reported in Downs et al. (2014) is lower than other reported effect concentrations and is within the range of concentrations typically reported for surface waters.

The combination of the occurrence of UV filter chemicals and reported effects in exposed aquatic organisms has led to concerns by resource managers for potential adverse effects in the coastal ecosystems of the VINP. Contamination of Virgin Islands National Park (VINP) habitats by those chemicals is likely given the high level of tourism on St. John, USVI and the VINP and that most of those tourists likely use products containing UV filter chemicals. The VINP encompasses sensitive marine habitats that are in proximity to high public use areas. In addition, several threatened scleractinian coral species inhabit coral reefs within those habitats and may be exposed to and affected by those UV filter chemicals. Therefore, the present study reports on UV filter chemical contamination of coastal waters of the VINP.

2. Materials and methods

This project occurred within the VINP, which is located on the island of St. John of the U.S. Virgin Islands (Fig. 1) in the eastern Caribbean Sea. Three different questions were addressed by three different sampling approaches. The first question was whether or not UV filter chemicals were present in VINP coastal waters at detectable concentrations. All of the VINP bays freely exchange with the ocean enabling rapid dilution of introduced contaminants including UV filter chemicals. Water samples were collected at midday from six bays (Great Lameshur, Brown, Leinster, Maho, Cinnamon, and Trunk) for analysis of UV filter chemical concentrations. The selected bays represented a presumed range of visitation. Trunk and Cinnamon Bays experience relatively heavy tourism pressure being closer to the largest community on St. John (Cruz Bay). In addition Trunk Bay is the destination for many daytime excursions from cruise ships that visit neighboring St. Thomas. Maho and Leinster Bays likely experience

a relatively moderate level of tourist pressure being further from Cruz Bay, while Brown and Great Lameshur Bays likely experience the least pressure both being the furthest from Cruz Bay and requiring either an arduous hike (Brown Bay) or a drive on a rough, unimproved road (Great Lameshur Bay) to access.

In April of 2013, 3 discrete water samples were collected from each of 3 locations (A, B, C – 9 L total) in each bay (Fig. 2). No lotions or sunscreen lotions were worn by the personnel during sample collection to minimize inadvertent contamination during sampling. All samples were collected in pre-cleaned 1 L amber bottles from approximately 1 m below water surface. Each bottle was rinsed once with site water before collecting the sample. In late May or early June of 2013, the bays were visited to repeat the discrete water sampling. The collected water samples were placed into a cooler with ice packs and transported to the field laboratory where they were held in a refrigerator for no more than 24 h before processing.

Partitioning of the UV filters between the sea surface microlayer (SML) and subsurface waters was also investigated. Water samples were collected from three high-use public beaches (Trunk, Cinnamon, and Hawksnest Bays) in December, 2013. One liter of SML water and 3×1 L of subsurface water were collected at about mid-day from each bay on three different days over a one-week period. Each 1 L SML sample was collected as described in Harvey and Burzell (1972). Briefly, a sheet of glass (28 cm \times 43 cm) was held vertically, repeatedly lowered into and raised out of the water, and allowed to drain for a short period of time (~5 s) before collecting into a pre-cleaned 1-L amber jar the remaining water that drained from the glass. Unlike in Harvey and Burzell (1972), no squeegee was used to force water into the jar so that cross contamination would be minimized. In order to minimize cross contamination among samples, the glass sheet $(28 \text{ cm} \times 43 \text{ cm})$ used to collect the SML sample was rinsed on shore in succession with acetone and deionized (DI) water, and was then dipped in site water prior to collection of the samples. The subsurface water was collected within a few meters of the SML sample collection location as described above for the April and June 2013 collections. The collected samples were placed into a cooler with ice packs, transported to the field laboratory, and held in a refrigerator until processing (within 24 h).



Fig. 1. Location of bays (black stars) on St. John, U.S. Virgin Islands (USVI) from which water was sampled during the present study.

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