



Emerging contaminants in aquatic environments and coastal waters affected by urban wastewater discharge in Thailand: An ecological risk perspective

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ABSTRACT

Emerging contaminants such as synthetic musks and UV-filters as ingredients personal care products were widely used in human daily life in Thailand. The occurrence and fate of four synthetic musks and nine UV-filters were investigated in eight full-scale sewage treatment plants (STPs) and their receiving aquatic environments in Bangkok and Pattaya, Thailand. All target compounds were detected in every single sample from STPs and surface water with magnitude from ng/L to µg/L. HHCB-lactone and HMS were found as the predominant musk and UV filter in influent and effluent of STPs, respectively. HHCB-lactone was also found with the highest concentration up to 79501 ng/g (dw) in the sludge. Low removal efficiency range from -37% (HHCB-lactone) to 58% (AHTN) were found for four musks in the STPs. The total emission of Σ₄musks and Σ₉UV-filters were estimated to be up to 16.7 mg/person/day and 0.28 mg/person/day by the STPs. Three synthetic musks and seven UV-filters were detected in fish from the receiving river. Concentration and emission of musks and UV filters found in this study from Thailand were much higher than those reported in many other countries worldwide. The preliminary ecological risk assessment showed that Musk xylene, 4-MBC and OC may pose high risk to aquatic organisms in the riverine and estuarine environment in Thailand.

1. Introduction

Large amounts of synthetic musks and UV filters are used as ingredients in personal-care products (PCPs) in Thailand, and some of these chemicals pose significant human and environmental health risks (Gago-Ferrero et al., 2011; Homem et al., 2015; Ramos et al., 2016). Some synthetic musks are restricted and banned in European countries (European Commission, 2011). Synthetic musks have been used as fragrance ingredients in almost all scented consumer products, such as lotions, perfumes, shampoos and air fresher (Rimkus, 1999). UV filters are commonly used in sunscreen and skincare products to absorb

200–400 nm UV irradiation to reduce skin cancer. (Kupper et al., 2006; Serpone et al., 2007).

The presence of synthetic musks and UV-filters in surface water, sediment, sea water, and even biota in European countries, America, Australia, and China have been reported (Clara et al., 2011; Liu et al., 2012; Mitchelmore et al., 2019; Villa et al., 2012; Yao et al., 2019). Musks and UV filters have shown adverse effects on aquatic organisms, e.g. AHTN, Musk xylene, 4-MBC, and OC have exhibited endocrine disrupting effects (estrogenic, antiestrogenic, androgenic and anti-androgenic) in both *in vitro* and *in vivo*. (Balázs et al., 2016; Ozáez et al., 2013; Paredes et al., 2014; Yamauchi et al., 2008). Effluent discharge

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and sludge application from sewage treatment plants (STPs) could be the main source of musks and UV filters to receiving environments. It is important to explore the emission of musks and UV filters in STPs as well as the removal potential of these STPs for each group, in order to estimate their final environmental pollution load and the ecological risk they may pose.

So far, limited information on synthetic musks and UV-filters is available in Southeast Asia especially in Thailand, which is located at the center of peninsular Southeast Asia and was the tenth-highest tourist country in the world in 2013 (World Tourism, 2015). The high volume of synthetic musks and UV-filters has been used in Thailand due to tropical climate, especially in the capital city Bangkok and another popular tourist destination Pattaya, both high urbanized (Nitivattanon and Srinonil, 2019). Therefore, firstly, the occurrence and removal of synthetic musks and UV-filters in STPs and the receiving rivers and coastal environment in Bangkok and Pattaya need to be studied.

This study aimed to investigate the occurrence and removal of four synthetic musks and nine UV-filters in eight full scale STPs and the receiving rivers and coastal areas in Bangkok and f Pattaya, Thailand. The pollution load and emission of these compounds were calculated for STPs. A screening level risk assessment was conducted to evaluate the ecological risks of these compounds in the aquatic environment in Thailand.

2. Materials and methods

2.1. Chemicals and materials

Commonly consumed four synthetic musks and nine UV-filters were selected as target compounds in this study. Monitored target contaminants included synthetic musks [Toxalide (AHTN), Galaxolide (HHCB), Galaxolidone (HHCB-lactone) and Musk xylene] and UV-filters [3,3,5-trimethylcyclohexyl salicylate (HMS), 2-Ethylhexyl 2-cyano-3,3-diphenylacrylate (OC), 2-Ethylhexyl salicylate (EHS), Octyl4-methoxycinnamate (OMC), 3-(4'-Methylbenzylidene) camphor (4-MBC), 2-(2H-benzotriazole-2-yl)4-methyl (UV-P), 2-(5-Chloro-2H-benzotriazol-2-yl)-4,6-bis(2-methyl-2-propanyl) phenol (UV-327), 2-(2H-Benzotriazole-2-yl)-4,6-bis(2-methyl-2-buatanyl)phenol (UV-328) and 2-(2'-hydroxy-5'-octylphenyl)-benzotriazole (UV-329)]. The physico-chemical properties of the target compounds are listed in Table S1.

2.2. Sampling

Wastewater, sludge, water, sediment and fish samples were obtained from eight STPs and receiving river and coastal areas in Bangkok and Pattaya, Thailand. The sampling campaigns were carried out in June 2016. Eight major and biggest STPs distributed in Bangkok for whole

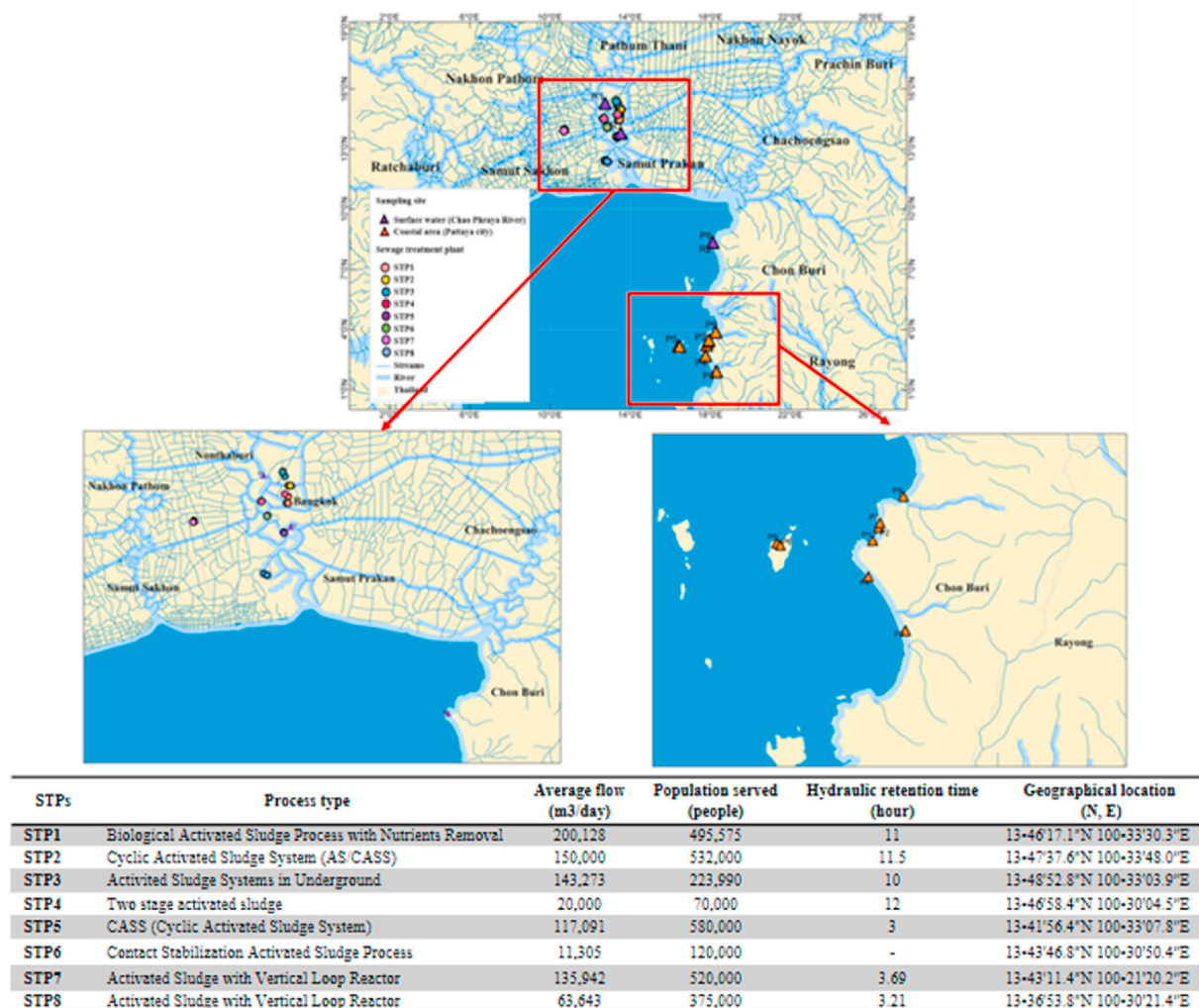


Fig. 1. The sampling site map showing the study area in Bangkok and Pattaya. (STP: sewage treatment plant, R: Chao Phraya river, and P: Pattaya city). Circle symbols represent sample locations of 8 major STPs in Bangkok, while purple triangle symbols represent sample locations of Chao Phraya River in Bangkok and red triangle symbols represent sample locations of coastal area in Pattaya. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

municipal wastewater treatment were selected as sampling sites in this study. Activated sludge treatment is the main operated treatment process in all eight STPs. The basic information of each STPs and location of sampling sites are shown in Fig. 1 and Table S2. Water samples from the Influent, effluent and sludge samples were collected from the STPs, and surface water and sediment samples were sampled from Chao Phraya River and urban streams. Besides, fish samples (*Oreochromis niloticus*) were captured by hook and line from six urban streams. In Pattaya, sea water and sediment samples were collected from nine sites in the coastal area. Each environmental sample was collected by 3 replicated sampling in each study site. Briefly, the samples from STPs were collected in 1 L brown glass bottles by 24h composite sampling, whereas the surface and sea water samples were collected about 0.5 m deep from the water surface in 1 L brown glass bottles as grab samples in each sampling location. 50 mL of methanol and 100 μ L of 4 M sulphuric acid were immediately added into water samples to inhibit microbial activity after sampling. Sludge samples were also collected from the activated sludge tank with 3 replicated sampling in each plant. As well as 3 replicated sediment samples (about 50 cm depth of surface sediment) were grabbed in each location. Sludge and sediment samples were collected in 0.5 L glass jar. All the samples were kept in coolers and directly transported to the laboratory. The water samples were store in a cold room (4 °C) and extracted within 48 h. Whereas, sludge and sediment samples were stored at -20 °C and were freeze dried within a week before extraction. The pH, temperature, DO, COD of the water samples were recorded and summarized in Tables S3–S4.

2.3. Sample preparation and extraction

The water, sludge, sediment, and fish samples were extracted and analyzed by using our previously reported methods (Liu et al., 2012; Yao et al., 2018a). Briefly, water samples were filtered using 0.7 μ m glass fiber filters (GF/F, Whatman) and spiked with the internal standard before solid phase extraction (SPE). Then the Oasis HLB cartridges (500 mg, 6 mL) were conditioned with 10 mL methanol and 10 mL Milli-Q water. The flow rate of water samples through the cartridge were load at 5–10 mL/min. The sample bottle was rinsed with 2 aliquots of 50 mL of 5% (v/v) methanol in Milli-Q water. The target compounds were eluted using 5 mL methanol, 4 mL ethyl acetate and 3 mL Dichloromethane, respectively. Then, the elutants were evaporated to nearly dryness under a gentle stream of nitrogen, and re-dissolved in 1 mL methanol before passing through 0.22 μ m organic membrane filter, and then kept at -18 °C prior to the instrumental analysis. The detailed water extraction and elution are summarized in Fig. S1.

For sludge and sediment, freeze-dried sludge (0.5 g each) and sediment (2 g each) in 30 mL centrifuge tube were extracted by using QuEChERS method. The samples were kept in the fridge (4 °C) overnight after spiking an internal standard mixture. 5 mL of Milli-Q was added into the centrifuge tube containing with sample and vortexed for 1 min, 10 mL acetonitrile containing 1% acetic acid, salt set containing 6 g anhydrous magnesium sulfate, and 1.5 g anhydrous sodium acetate were added. Then, the tubes were hand-shaken for 2–3 min immediately and centrifuged at 3500 rpm for 10 min. The 7 mL of extract supernatant was transferred into the dispersive solid-phase extraction tube (containing 900 mg magnesium sulfate, 150 mg primary-secondary amine, and 150 mg C18 bulk sorbent), and vortexed for 1 min followed by centrifuged at 3500 rpm for 10 min. Again, 5 mL of supernatant extract was transferred into the 10 mL glass tube, and were evaporated under a gentle stream of nitrogen. The final extract was re-dissolved in 1 mL methanol, filtered and kept in a freezer (-20 °C) until analysis. Flow chart of extraction and purification procedures in sludge and sediment are shown in Fig. S2.

Fish tissues including bile, plasma, muscle, and liver were extracted separately for the target compounds for each fish. 50 μ L bile sample and 50 μ L plasma samples were transferred to a 2 mL glass vial, spiked with 20 μ L (2 mg/L) internal standard mixture. The bile samples were loaded into Captiva ND^{Lipid} tubes, with 500 μ L of acetonitrile (containing 1% (v/v)

v) acetic acid) added previously and vortexed for 30 s followed by pumped under a vacuum to collect acetonitrile eluates into d-SPE tubes. After that, 200 μ L of acetonitrile acidified with 1% (v/v) acetic acid was used for rinsing the Captiva ND^{Lipid} tube walls, and 5.2 mL acetonitrile containing 1% (v/v) acetic acid were added. Then, d-SPE tubes were shaken by hand for 2–3 min and centrifuged at 2364 g for 10 min. The 3 mL supernatant acetonitrile phases were transferred to 10 mL glass vials and dried under a gentle nitrogen stream and reconstituted in 0.2 mL methanol a freezer (-20 °C) until analysis.

For plasma samples, loaded into Captiva ND^{Lipid} tubes, with 500 μ L of acetonitrile (acidified with 1% (v/v) acetic acid) added beforehand. The tubes were vortexed for 30 s and then pumped under a vacuum to collect eluates into 10 mL glass vials. Later, captiva ND^{Lipid} tube walls were rinsed once with 200 μ L of acetonitrile acidified with 1% (v/v) acetic acid, and the acetonitrile eluates were collected together and dried under a gentle nitrogen stream. The dried extract was reconstituted in 0.2 mL methanol a freezer (-20 °C) until analysis.

For muscle and liver extraction, 1 g liver or 0.5 g muscle (dry weight), with 40 μ L of 1 mg/L of corresponding internal standards, 5 mL of Milli-Q water, and two ceramic homogenizers, was added to a 50 mL polypropylene conical bottom centrifuge tube, then vortexed about 1 min to homogenize the tissue sample sufficiently. Subsequently, 10 mL of acetonitrile with 1% acetic acid and a packet of QuEChERS extraction salts (consisting of 6 g anhydrous magnesium sulfate and 1.5 g anhydrous sodium acetate) were added into the tube, and shaken for 2–3 min. After centrifugation at 2364g for 10 min, 7 mL of the supernatant was transferred into a d-SPE tube, and vortexed and centrifuged. A final 5 mL of supernatant extract was concentrated under a gentle nitrogen stream and redissolved in 0.2 mL methanol and kept in a freezer (-20 °C) until analysis.

2.4. Target compounds analysis

The targeted chemicals were analyzed by GC–MS (Agilent 6890N–5975B, USA) in EI mode. An Agilent DB-5MS column (30 m \times 0.25 mm, 0.25 μ m film thickness) was used to separate target compounds. The injection volume was 2.0 μ L in the splitless mode, and the splitless time and split flow were set at 1 min and 100 mL/min, respectively. The detailed operating conditions can be referred to our previously published method (Lai et al., 2014; Yao et al., 2018b). Strict quality assurance and quality control (QC/QA) were conducted in all the analysis procedures, blanks and control samples were run for every 10 samples to check for any carryover, background contamination, precision and accuracy of the recovery. The recovery, limit of detection (LOD) and limit of quantification (LOQ) for each target compounds are listed in Tables S5–S6.

2.5. Data analysis

2.5.1. Statistical analysis

The contamination profiles of synthetic musks and UV-filters in samples were described in the range, mean value, median value of concentrations and detection frequency. When calculating the range, median and mean values, the value less than LOQ (method quantitation limit) was substituted with a value of half of LOQ, and for the data of ND (not detected), a zero was substituted. The detection frequency was calculated including the data below LOQ, but ND results were not counted. The descriptive statistic was performed with Microsoft Excel 2010 and Sigma Plot 10.0.

2.5.2. Mass balance and emission calculation

Mass balance analysis was performed by theory of mass balance:

$$100\% = Mf_{Inf}(\%) = Mf_{Eff}(\%) + Mf_{Sludge}(\%) + Mf_{Loss}(\%) \quad (1)$$

Where Mf_{Inf} , Mf_{Eff} , Mf_{Sludge} and Mf_{Loss} are the mass fraction of influent,

effluent, sludge, and mass fraction loss, respectively; $M_{\text{Eff}}(\%)$ or $M_{\text{Sludge}}(\%)$ was calculated by the mass load of effluent (M_{Eff}) or sludge (M_{Sludge}) divided by the mass load of influent (M_{Inf}). The mass load of each compound was calculated using the following equation:

$$M_{\text{Inf}} = C_{\text{Inf}} \times Q / 10^6 \quad (2)$$

$$M_{\text{Eff}} = C_{\text{Eff}} \times Q / 10^6 \quad (3)$$

$$M_{\text{Sludge}} = C_{\text{Sludge}} \times DP \times (1 - MC) / 10^3 \quad (4)$$

Where C_{Inf} , C_{Eff} , and C_{Sludge} represent the concentrations of each target compound in influent, effluent and sludge, respectively; Q is the average of water flow (m^3/d); DP is the daily production of sludge (t/y , wet weight); MC is the moisture content of sludge (%).

Emission calculation were performed by pollution load per capita in influent, effluent and sludge with the following equations:

$$\text{Pollution loads per capita in influent} = M_{\text{inf}} \text{ divided by population served} \quad (5)$$

$$\text{Pollution loads per capita in effluent} = M_{\text{eff}} \text{ divided by population served} \quad (6)$$

$$\text{Pollution loads per capita in sludge} = M_{\text{sludge}} \text{ divided by population served} \quad (7)$$

2.5.3. Bioaccumulation factors

Bioaccumulation factors (BAFs) in fish plasma, bile, muscle, and liver bile and plasma were calculated using the following equation:

$$\text{BAF}_{\text{plasma}} = (C_{\text{plasma}} / C_{\text{water}}) \times 1000 \quad (8)$$

$$\text{BAF}_{\text{bile}} = (C_{\text{bile}} / C_{\text{water}}) \times 1000 \quad (9)$$

$$\text{BAF}_{\text{muscle}} = (C_{\text{muscle}} / C_{\text{water}}) \times 1000 \quad (10)$$

$$\text{BAF}_{\text{liver}} = (C_{\text{liver}} / C_{\text{water}}) \times 1000 \quad (11)$$

Where $\text{BAF}_{\text{plasma}}$, BAF_{bile} , $\text{BAF}_{\text{muscle}}$ and $\text{BAF}_{\text{liver}}$ are the bioaccumulation factor of fish plasma, bile, muscle and liver, respectively; C_{water} is target compound concentration in surface water with units of ng/L ; C_{plasma} and C_{bile} are target compound concentration in fish plasma and bile, muscle and liver, respectively, with units of $\mu\text{g}/\text{L}$; C_{muscle} and C_{liver} are target compound concentration in fish plasma and bile, muscle and liver, respectively, with units of $\mu\text{g}/\text{kg}$.

2.6. Preliminary risk assessment

The potential environmental risks were assessed by using the risk quotient (RQ) approach according to the European ecological risk assessment guideline (Van Leeuwen, 2003) which follow the equation:

$$\text{RQ} = \text{MEC} / \text{PNEC} \quad (12)$$

Where MEC is the measured environmental concentration and PNEC is predicted no-effect concentration of each compound.

The risk classification was based on risk ranking criteria in which $\text{RQ} < 0.01$, minimal risk; $0.01 \leq \text{RQ} < 0.1$, low risk; $0.1 \leq \text{RQ} < 1$, medium risk; and $\text{RQ} \geq 1$, high risk.

3. Results

3.1. Concentrations, removal and emission of synthetic musks and UV-filters in STPs

For synthetic musks and nine UV-filters as target compounds were detected in all influent, effluent and sludge samples (Fig. 2). Basic

information and physicochemical properties measurement of each sampling site are shown in Tables S2 and S3. Higher median value concentration were found for most of the synthetic musks than UV-filters. For synthetic musks, HHCB-lactone was found as the predominant musk in influent, effluent and sludge. All synthetic musks were detected in every single samples from STPs, with four musks concentration ranging from 271 to 39142 ng/L in influent, 127–63982 ng/L in effluent, and 971–79501 ng/g in sludge. HHCB-lactone was found as dominant synthetic musks with the highest median level in influent, effluent, and sludge samples from STPs. While 100% detected of nine UV-filters were found in both influent and effluent with median value concentration ranging from 7.23 to 2352 ng/L and 6.60–439 ng/L , respectively. OC was the predominant found in influent, followed by HMS, UV-329, EHS and OMC, whereas about 1–9 times decrease found in effluent by the median value, except UV-327.

The removal efficiency of synthetic musks and UV-filters are given in Fig. S6 and Fig. S7. Compare to synthetic musks, UV filters shown higher removal in all STPs. AHTN was removed by 58%, followed by HHCB (34%) in all eight STPs. For UV-filters, the removal of EHS was up to 81%, followed by OC (79%), HMS (65%), OMC (60%) and 4-MBC (45%). While negative removal rates were observed for HHCB-lactone, Musk xylene and UV-327, probably due to variations in the influent and effluent concentrations of these compounds.

Mass balance analysis could better the understanding of the degradation/transformation behaviors and pollution discharges of synthetic musks and UV filters in STPs of Bangkok. We calculated the mass fractions (%) of target compounds in influent, effluent and sludge of eight STPs (Fig. S8). More than 42% and 66% (mean value) of AHTN and HHCB, respectively, were found in effluent, which will be discharged into receiving river, and 4% and 10% of them were adsorbed to sludge, with 49% and 29% degraded through the treatment of STPs. The negative mass fraction loss of HHCB-lactone and Musk xylene were calculated, which is similar to the aqueous phase removal. The findings indicated that some co-occurring compounds/metabolites may have transformed to HHCB-lactone and Musk xylene during the wastewater treatment process in STPs.

Usage per capita were calculated based on the severed population of STPs and the influent concentrations of all detected synthetic musks and UV-filters in STPs in Bangkok. The highest usage of synthetic musks and UV filters were HHCB-lactone (2.77 $\text{mg}/\text{person}/\text{day}$) and HMS (0.24 $\text{mg}/\text{person}/\text{day}$), respectively (Fig. S9; Fig. S10). Based on the concentration in effluent and sludge, and the discharge volume of wastewater in STPs, the emission amount of target compounds was estimated. As shown in Fig. S9, the low levels of pollution load per capita were found in sewage sludge, ranged from 0.002 to 1.64 $\text{mg}/\text{person}/\text{day}$. UV-329 was the major UV-filter with the mean value of 0.09 $\text{mg}/\text{person}/\text{day}$. The mean input of $\Sigma 9\text{UV}$ -filters to the STPs was 0.001–0.93 $\text{mg}/\text{person}/\text{day}$, while 0.01–0.28 $\text{mg}/\text{person}/\text{day}$ of target pollutants were discharged into the rivers through the effluent. Hence, these results implied that the STPs effluent could be one of the important sources of synthetic musks and UV-filters pollution in the aquatic environment.

3.2. Concentrations of synthetic musks and UV-filters in receiving environments

High level concentrations were investigated in receiving water bodies for all four synthetic musks and nine UV-filters in both water and sediment parts, as reported in Fig. 3. Among all target synthetic musks, HHCB-lactone was also identified as the highest concentrations in water portion, with the median value of 4498 ng/L . However, the median values of each individual synthetic musk in water were lower than those in the corresponding sediment, except Musk xylene which higher than in water portion. For UV-filters, the environmental median value concentration of individual UV filter was generally higher than 100 ng/L , except EHS, 4-MBC, UV-327, and UV-328 in river water samples. Although HMS was detected as the highest median value of

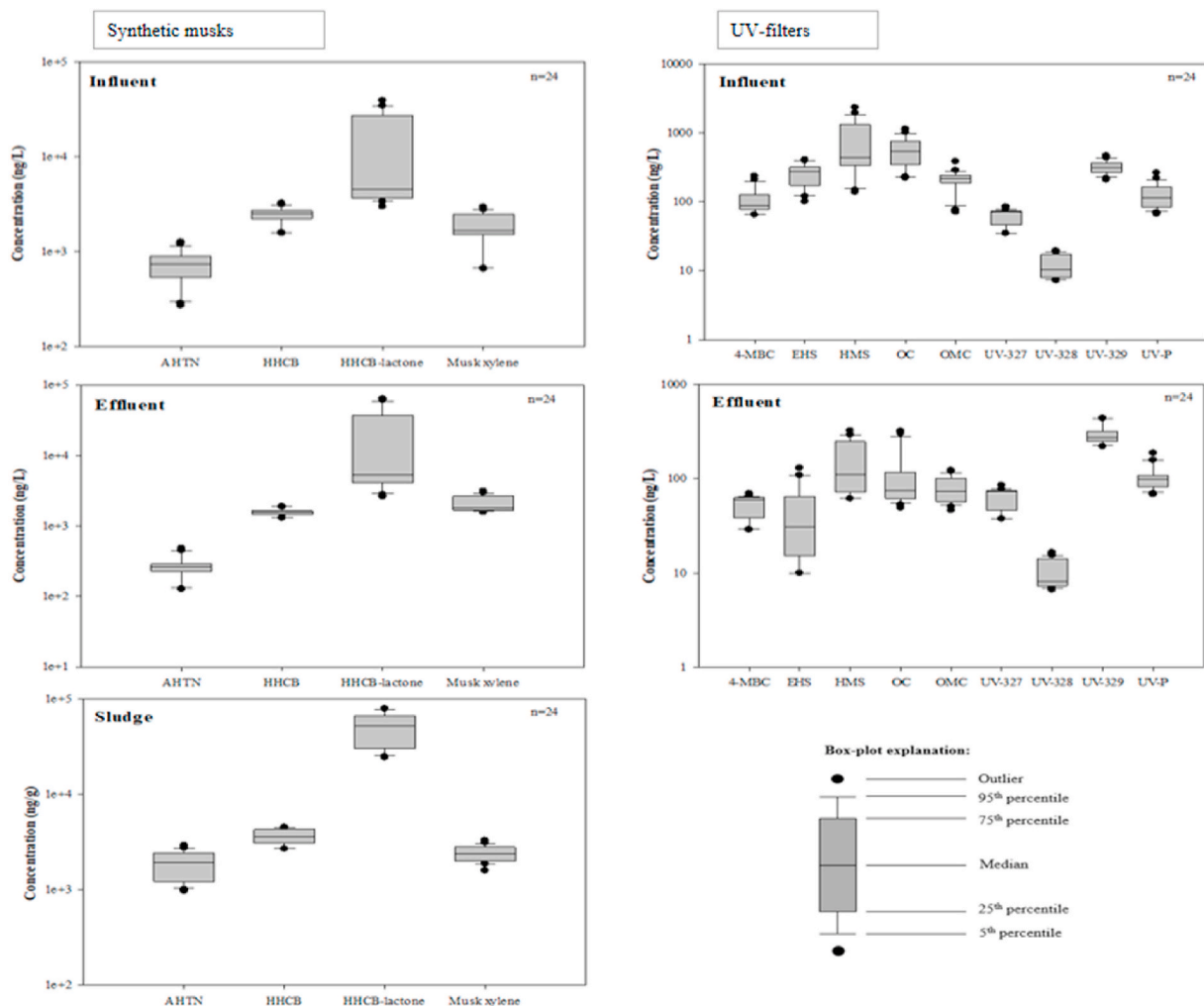


Fig. 2. The target synthetic musks and UV-filters concentration levels in eight major STPs in Bangkok.

concentration in both river water and sea water.

The widespread occurrence of synthetic musks and UV-filters at coastal area were shown. Most synthetic musks were found in all samples, except HHCB-lactone was found 89% in sea water. Furthermore, only AHTN and Musks xylene were found in all estuarine sediment samples. Musk xylene was presented the highest median values in both sea water and estuarine sediment. For UV-filters, most UV-filters were found in all samples, except UV-329 was found 89% in sea water. In general, the sea water levels of UV filters were higher than those in river water. This could be probably due to the coastal recreational activities (Tsui et al., 2017).

3.3. Accumulation of synthetic musks and UV-filters in fish

Fish samples were also attended in this study as the contamination of synthetic musks in biota. Fish (*Oreochromis niloticus*) were investigated from six receiving rivers in Bangkok. The occurrence of these compounds in fish suggested a potential for biota accumulation. AHTN and HHCB were found as the predominant musks in. The average concentrations of these synthetic musks in fish samples are presented in Fig. S8 and Table S7. The concentrations of AHTN and HHCB were found with a relatively similar trend in fish organs, HHCB ranged from 519 to 1380 ng/g (liver), ND - 1034 ng/g (muscle), however, HHCB-lactone was only found in plasma with concentration ranged from ND to 9158 ng/L.

The bioaccumulation factor (BAFs) values for synthetic musks and UV-filters in plasma, bile, muscle, and liver were calculated to evaluate

the fish uptake potencies of the target compounds by using our previously reported methods (Zhao et al., 2015). The data are shown in Table S8. For synthetic musks, AHTN and HHCB were detected in bile, muscle and liver with the range of log BAFs were -1.06 to -0.46 , -1.40 to -0.24 and -0.98 to -0.20 , respectively, with the median values of log BAFs for HHCB > AHTN. For UV-filters, seven target UV-filters were detected in fish muscle between 2.42 and 4.24, and the median value of log BAFs for all detected UV-filters were higher than 2, with the following order: EHS > HMS > UV-328 > UV-329 > UV-P > OC. For fish liver samples, the five detected UV-filters were investigated in the range of log BAFs between 2.65 and 4.98, which the mean and median values for all were more than 3. The median of log BAFs showed the following order: HMS > EHS > UV-328 > UV > 327 > UV-329. Generally, the mean and median of log BAFs in fish liver were higher than muscle for detected synthetic musks and UV-filters. Salicylate derivatives (HMS and EHS) presented the maximum values BAFs among all various fish tissues, as well as the log BAFs values of detected UV-filters showed higher than detected synthetic musks in all fish samples.

4. Discussion

4.1. Ecological risks assessment

The wide presence of target synthetic musks and UV-filters in STPs and aquatic environments with concentration levels in the range of ng/L to $\mu\text{g/L}$ in Thailand were found in this study. A screening-level risk

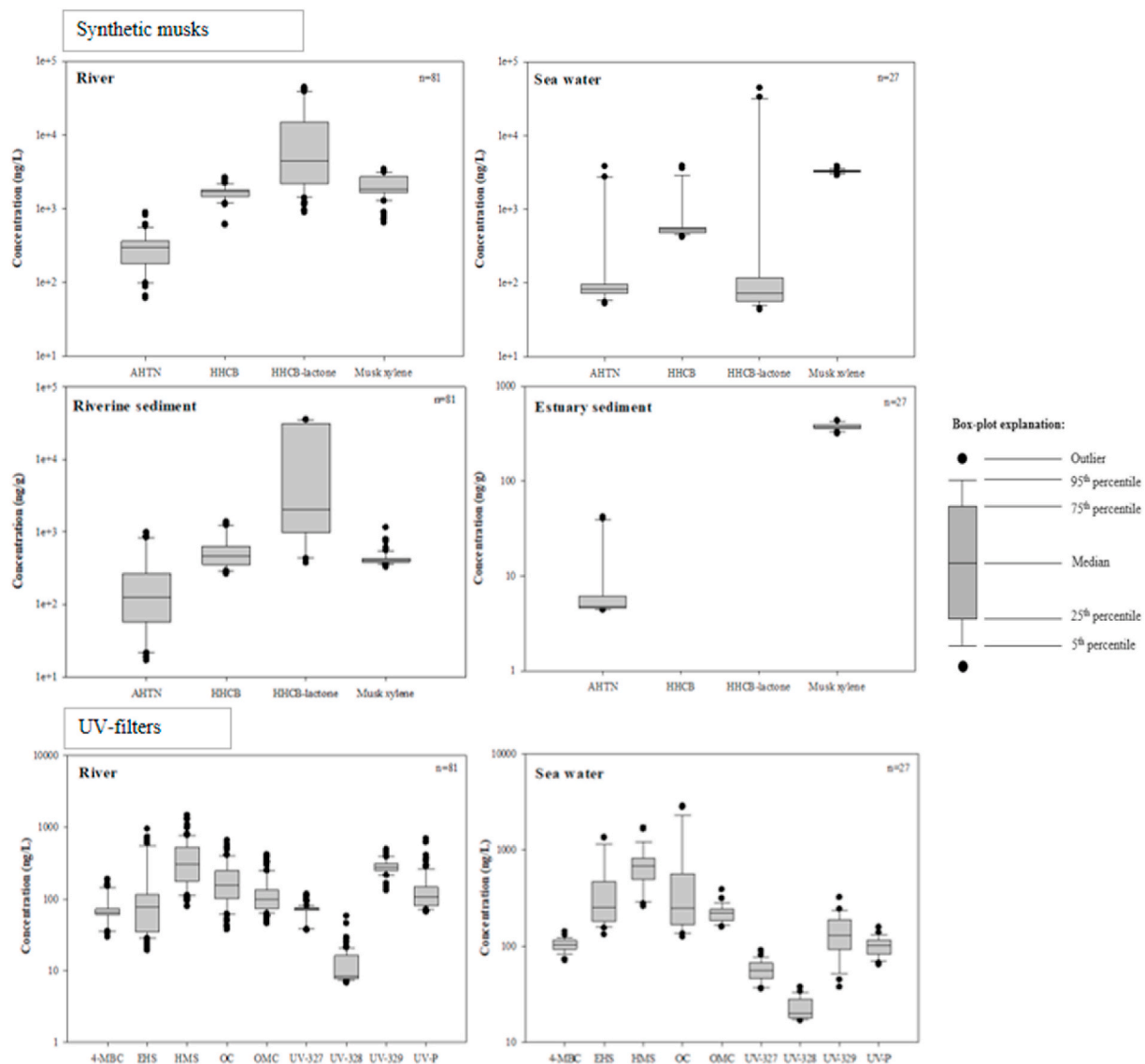


Fig. 3. The target synthetic musks and UV-filters concentration levels in riverine and coastal environments.

assessment to the aquatic organism in riverine, and coastal area were performed based on the measured concentrations of target synthetic musks and UV-filters, and PNECs derived from literature data (Sánchez Rodríguez et al., 2015; Peng et al., 2017). Risk evaluation could only be performed for three synthetic musks and five UV-filters including AHTN, HHCB, Musk xylene, 4-MBC, UV-P, OC, UV-328, and UV-329, since the PNEC values were not available for rest target compounds. The results of risk assessment and PNEC values are summarized in Fig. 4 and Tables S9–S10. The results demonstrated that receiving riverine and coastal area faced with ecological risks from minimal risk to high risk. Musk xylene were found with 93% and 100% high risk in receiving riverine and coastal area, respectively. While AHTN, 4-MBC, UV-P, and OC were showed high risk in some sampling sites. And most of the target compounds were also indicated as medium risk in both receiving riverine and coastal area.

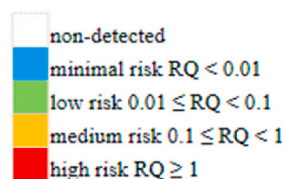
Owing to the results from the present study, Musk xylene presented a huge usage and emission, and the serious contamination in STPs and receiving environments was found. Also, high risk for Musk xylene in riverine and coastal area in Thailand may result in adverse effects to aquatic organisms and human health. Mersch-Sundermann et al. (1996) reported that Musk xylene is a strong inducer of phase 1 liver enzymes and act as co-genotoxicants (Mersch-Sundermann et al., 1996). Musk

xylene is the nitro musk and introduced into the market since the last century (Hutter et al., 2010; Rimkus, 1999). On the other hand, the enormous consumption in drinking water and fish, likewise using personal care products that contain synthetic musks and UV-filters might be the cause of serious exposure to humans. The contamination of Musk xylene in human blood, milk, and adipose tissue were also reported (Eisenhardt et al., 2001; Hutter et al., 2010; Liebl and Ehrenstorfer, 1993; Müller et al., 1996; Rimkus et al., 1994). Musk xylene was banned in the European Commission under the new chemical regulation REACH and Japan (Registration, Evaluation, Authorization and Restriction of Chemicals). (European Commission, 2011 and Wong et al., 2019). But Musk xylene is still produced and use in Thailand. So, the potential human health risk of these compounds should be paid more attention to since this river was the source of water supply for drinking water and fishery. Therefore, with the intention to better control synthetic musks and UV-filters contamination and mitigate the risk in aquatic environment in Thailand, more research in the contaminated monitoring, usage control and production policies and wastewater removal enhancements are needed as soon as possible.

Surface water	AHTN ^a	HHCB	Musk xylene	4-MBC	UV-P	OC	UV-328	UV-329
*PNEC (ng/L)	1000 ^a	6800 ^a	1000 ^a	100 ^b	130 ^c	500 ^d	78 ^d	100000 ^d
STP1-Upstream								
STP1-Discharge point								
STP1-Downstream								
STP2-Upstream								
STP2-Discharge point								
STP2-Downstream								
STP3-Upstream								
STP3-Discharge point								
STP3-Downstream								
STP4-Upstream								
STP4-Discharge point								
STP4-Downstream								
STP5-Upstream								
STP5-Discharge point								
STP5-Downstream								
STP6-Upstream								
STP6-Discharge point								
STP6-Downstream								
STP7-Upstream								
STP7-Discharge point								
STP7-Downstream								
STP8-Upstream								
STP8-Discharge point								
STP8-Downstream								
R1 (River-Upstream)								
R2 (River-Middle point)								
R3 (River-Downstream)								

Sea water	AHTN ^a	HHCB	Musk xylene	4-MBC	UV-P	OC	UV-328	UV-329
*PNEC (ng/L)	1000 ^a	6800 ^a	1000 ^a	100 ^b	130 ^c	500 ^d	78 ^d	100000 ^d
P1								
P2								
P3								
P4								
P5								
P6								
P7								
P8								
P9								

The criteria of RQ levels were classified as follows:



*The PNECs values are taken from previous reports of
^aFeng-Jiao Pent et al., 2017,
^bA.Sanchez Rodriquez et al., 2015,
^cNICNAS, 2017.
^dNai-Sheng Zhang et al., 2015,

Fig. 4. The calculated risk quotient values (RQs) of the target synthetic musks and UV-filters in receiving riverine environment and coastal environment in Thailand.

4.2. Synthetic musks and UV-filters comparison with other regions in the world

The comparison in the occurrence of synthetic musks and UV filters was summarized in Table 1 and Table 2. Musk xylene found in Thailand from present study were higher than other countries, which most of them banned this compounds, in addition, AHTN and HHCB in influent of STPs were higher than those in China, Canada, Germany, and USA (Bester, 2004; Horii et al., 2007; Lishman et al., 2006; Yang and Metcalfe, 2006; Zhang et al., 2008), but lower than those detected in Austria, Spain, and Switzerland (Carballa et al., 2004; Clara et al., 2011; Kupper et al., 2006). Interestingly, the results were compared with those in Austria and Spain (Carballa et al., 2004; Clara et al., 2011), found that influent concentrations were higher, but effluent concentrations were lower than the present study. This spot may be caused by different wastewater treatment potential in a different area. For UV filters, The higher concentrations were found for benzotriazole ultraviolet stabilizers (BUVs; including UV-P, UV-327 and UV-329) in influent by median values, excepting UV-328 than those in Canada (Lu et al., 2017). HMS and EHS were found at higher levels than those in influent from Hong Kong and Germany (Rodil et al., 2009; Tsui et al., 2014). Despite, the occurrence of 4-MBC in influent from other studies showed 1.3 to 42 times higher median value than those from this study (Balmer et al.,

2005; Bester, 2005; Li et al., 2007; Lu et al., 2017; O'Malley et al., 2019; Rodil et al., 2009; Tsui et al., 2014). Additionally, the most detected target UV-filters concentration in effluent were higher than those previously reported in Switzerland, China, Japan, Hong Kong, Slovenia and Canada (Balmer et al., 2005; Cuderman and Heath, 2007; Kameda et al., 2011; Li et al., 2007; Lu et al., 2017; Tsui et al., 2014).

The ratio of HHCB/AHTN in the influent of this study was 3.4:1 which indicated that the greater production and usage of HHCB compared with AHTN in Thailand. The occurrence of these synthetic musks in receiving river in this study were in accordance with (Homem et al., 2015; Hu et al., 2011a; Lange et al., 2015), which means discharge from STPs is a very important pathway for musks and UV filters to the environment. (Homem et al., 2017; Hu et al., 2011b; Lange et al., 2015). The increase of HHCB-lactone might be caused by the formation of a lactone through the oxidation of HHCB (Horii et al., 2007; Kuhlich et al., 2011; Lange et al., 2015). Moreover, the concentration levels of some synthetic musks and UV filters in the riverine environment were higher than levels detected in STPs, reflecting that some other wastewater could discharge into the river by direct discharge or no-point source in Bangkok (Molins-Delgado et al., 2017).

Synthetic musks concentrations in riverine environment, the detected concentration in surface water and sediment from this study were higher than previous studies from Korea, Germany, Austria, China and

Table 1

The occurrence of synthetic musk (median values) in difference environmental media from this study compare with the literature from other regions in the world.

Synthetic musks	Environmental samples ^a	Thailand	Spain ^d	Switzerland ^e	Austria ^f	Korea ^g	USA ^{h,i,p}	China ^{q,r}	Canada ⁱ	Germany ^{k,s}	Canada ^m	UK ⁿ	Italy ^t
AHTN	Influent	742	1530	1370	1004	770 ^a	712	695	687	587	79.8	–	–
	Effluent	265	160	300	<LOQ		328	87	–	206	298	129	–
	Sludge	1927	–	3440	1900 ^a	11850	2700	3600	–	1525	771	–	–
	Surface water	294	–	–	<40	120 ^a	ND	11.9	–	60	–	–	97
	Riverine sediment	123	–	–	<10	2.3 ^b	0.85	7.2	–	–	–	–	927
HHCB	Influent	2512	3180	4450	4350	3480 ^a	5524	2152	1701	1933	373	–	–
	Effluent	1563	500	720	<LOQ		2960	310	876	669	–	1735	–
	Sludge	3561	–	10140	14200 ^a	37350	11000	10400	–	3038	3107	–	–
	Surface water	1703	–	–	150	650 ^a	ND	18.8	–	260	–	–	463
	Riverine sediment	461	–	–	67	6.3 ^b	1.43	5.8	–	–	–	–	1349
HHCB-lactone	Influent	4533	–	–	–	–	247	–	–	230	–	–	–
	Effluent	5297	–	–	–	–	–	–	–	370	–	–	–
	Sludge	52464	–	–	–	–	4000	–	–	–	–	–	–
	Surface water	4498	–	–	–	–	–	–	–	100	–	–	–
	Riverine sediment	2032	–	–	–	–	–	–	–	–	–	–	–
Musk xylene	Influent	1656	–	–	–	–	ND	<LOQ	–	–	13.9	–	–
	Effluent	1796	–	–	–	–	ND	<LOQ	–	–	–	5.5	–
	Sludge	2389	–	–	–	–	–	–	–	–	83.2	–	–
	Surface water	1838	–	–	–	–	ND	–	–	–	–	–	–
	Riverine sediment	405	–	–	–	–	–	–	–	–	–	–	–

oGuo, 2010.

cmaximum value, <LOQ: below of limit of quantitation, ND: not detected. Hu et al., 2011c; Sumner et al., 2010; Sun et al., 2014; Zhou et al., 2009

^a The concentration of synthetic musks in difference environmental media: Influent (ng/L), Effluent (ng/L), Sludge (ng/g), Surface water (ng/L), and Riverine sediment (ng/g).^b mean value.^d Carballa et al. (2004).^e Kupper et al. (2006).^f Clara et al. (2011).^g Lee et al. (2010).^h Chase et al. (2012).ⁱ Zhang et al. (2008).^j Lishman et al. (2006).^k Bester (2004).^l Horii et al. (2007).^m Yang and Metcalfe (2006).ⁿ Sumner et al., 2010^p Sun et al., 2014^q Zhou et al., 2009^r Hu et al., 2011c^s Lange et al. (2015).^t Villa et al. (2012).

USA (Chase et al., 2012; Clara et al., 2011; Hu et al., 2011a; Lange et al., 2015; Lee et al., 2010), except for concentration in sediment from Italy (Villa et al., 2012). Moreover, Fish contamination by musks and UV filters were higher than previous studies in China (He et al., 2013; Yao et al., 2019).

The resulting trend was similar with detected UV-filters in surface water, organic UV-filters concentration including HMS, EHS and OC were higher than those reports from other regions (Table 2). Especially, most of detected UV-filters concentration from the present study were increased from the previous study in Bangkok, which reflects that UV-filters consumption and production are growing up in Thailand.

The occurrence of UV-filters compare to some other famous sea and beach tourism regions, the UV-filters concentrations levels in Thailand were higher than Hawaii, Spain, Norway, USA and Japan (Bargar et al., 2015; Langford and Thomas, 2008; Mitchelmore et al., 2019; Sánchez Rodríguez et al., 2015; Tashiro and Kameda, 2013). From the overall comparison, the contamination of synthetic musks and UV filters in STPs, receiving environments and coastal environments were more serious in Thailand than the most of other countries from previous reports, and the pollution and environmental risk of these compounds should be concern for safety marine ecosystems.

5. Conclusions

Four synthetic musks and nine UV-filters were found in every single sample collected in eight major STPs and receiving water in Thailand. Low removals of these compounds were observed in the STPs. HHCB-lactone and Musk xylene, as well as HMS and UV-329, are the major synthetic musks and UV-filters found in effluent and receiving water. As a result, municipal wastewater seems to be the major emitters of synthetic musks and UV-filters into the environment. The higher level of UV-filters were detected in receiving river water than the effluent from STPs, hence, direct discharge from households may be another source of these compounds. Concentration and emission of musks and UV filters, especially for HHCB-lactone and Musk xylene in Thailand were much higher than lots of other countries. High risk of Musk xylene were found in riverine and coastal areas. Future investigation should look into the mechanisms for the degradation of synthetic musks and UV filters and the behavior of these compounds in the aquatic environment.

Author contribution

Kanokthip Juksu: Investigation, Data curation, Writing-Original draft preparation; **You-Sheng Liu:** Conceptualization, Writing-Review & Editing, Founding acquisition; **Jian-Liang Zhao:** Investigation; **Li Yao:** Formal analysis; **Charoon Sarin:** Investigation, Writing - Review &

Table 2The occurrence of UV-filters (median values) in different environmental media from this study compare with the literature from other regions in the world [Cunha, 2015](#).

UV-filters	Environmental samples (ng/L)	Thailand	Bangkok ^b	Switzerland ^{c,d}	Australia ^e	China ^{f,g}	Arctic ^g	Hong Kong ^g	Portugal ^h	Germany ⁱ	Canada ^j	Japan ^{g,k,l}	Slovenia ^m	USA ^{g,n}	Norway ^o	Spain ^p
HMS	Influent	441	-	-	-	-	-	105	-	<LOQ	-	-	-	-	-	-
	Effluent	111	-	-	-	-	-	<LOD	-	-	-	ND	<LOD	-	-	-
	Surface water	306	29	-	-	<LOD	<LOD	66	-	<LOQ	-	65	<LOD	142	-	-
OC	Sea water	682	-	-	-	-	-	-	-	-	-	10	-	144	-	-
	Influent	542	-	2100	-	86	-	LOD	104 ^a	5322 ^a	-	-	-	-	-	-
	Effluent	75.3	-	20	-	55	-	<LOD	254	-	-	ND	<LOD	-	-	-
EHS	Surface water	156	153	<2	-	75	26	103	-	4381 ^a	-	87	<LOD	117	-	-
	Sea water	249	-	-	-	-	-	-	-	-	-	6.5	-	3.2	12.5	110
	Influent	279	-	-	-	-	-	93.2	-	753 ^a	-	-	-	-	-	-
UV-P	Effluent	30.7	-	-	-	-	-	<LOD	-	-	-	-	-	-	-	-
	Surface water	76.6	283	-	-	121	<LOD	61	-	748 ^a	-	71	-	53	-	-
	Sea water	252	-	-	-	-	-	-	-	-	-	-	-	-	-	-
4MBC	Influent	116	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	Effluent	98.3	-	-	-	-	-	-	-	-	-	7	-	-	-	-
	Surface water	108	-	-	-	-	-	-	-	-	-	ND	-	-	-	-
UV327	Sea water	101	-	-	-	-	-	-	-	-	-	ND	-	-	-	-
	Influent	89	-	3750	230 ^a	1048	-	117	66.6	278 ^a	-	-	-	-	-	-
	Effluent	59.7	-	650	-	647	-	<LOD	<LOQ	-	-	ND	<LOD	-	-	-
UV328	Surface water	64.5	<LOD	7	-	-	-	-	-	1140 ^a	-	-	<LOD	-	-	-
	Sea water	104	-	-	-	-	-	-	-	-	-	-	-	ND	2.2	6.3
	Influent	72.6	-	-	-	-	-	-	-	-	ND	-	-	-	-	-
UV329	Effluent	73.8	-	-	-	-	-	-	-	-	ND	2	-	-	-	-
	Surface water	73.1	-	-	-	-	-	-	-	-	-	ND	-	-	-	-
	Sea water	55.2	-	-	-	-	-	-	-	-	-	ND	-	-	-	-
UV329	Influent	10.4	-	-	-	-	-	-	-	-	45.1	-	-	-	-	-
	Effluent	8.11	-	-	-	-	-	-	-	-	3.61	62	-	-	-	-
	Surface water	8.27	-	-	-	-	-	-	-	-	-	18.6	-	-	-	-
UV329	Sea water	19.9	-	-	-	-	-	-	-	-	-	6.7	-	-	-	-
	Influent	313	-	-	-	-	-	-	-	-	8.82	-	-	-	-	-
	Effluent	273	-	-	-	-	-	-	-	-	ND	ND	-	-	-	-
UV329	Surface water	279	-	-	-	-	-	-	-	-	-	ND	-	-	-	-
	Sea water	122	-	-	-	-	-	-	-	-	-	ND	-	-	-	-

^a Mean value, <LOQ: below of limit of quantitation, <LOD: below of limit of detection ND: not detected.^b [Tsui et al. \(2014\)](#).^c [Balmer et al. \(2005\)](#).^d [Bester \(2005\)](#).^e [O'Malley et al. \(2019\)](#).^f [Li et al. \(2007\)](#).^g [Tsui et al. \(2014\)](#).^h [Cunha, 2015](#)ⁱ [Rodil et al. \(2009\)](#).^j [Lu et al. \(2017\)](#).^k [Kameda et al. \(2011\)](#).^l [Tashiro and Kameda \(2013\)](#).^m [Cuderman and Heath \(2007\)](#).ⁿ [Mitchellmore et al. \(2019\)](#).^o [Langford and Thomas \(2008\)](#).^p [Sánchez Rodríguez et al. \(2015\)](#).

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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