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ORIGINAL PAPER

Sediments in the mangrove areas contribute to the removal of endocrine disrupting chemicals in coastal sediments of Macau SAR, China, and harbour microbial communities capable of degrading E2, EE2, BPA and BPS

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Abstract The occurrence of endocrine disrupting chemicals (EDCs) is a major issue for marine and coastal environments in the proximity of urban areas. The occurrence of EDCs in the Pearl River Delta region is well documented but specific data related to Macao is unavailable. The levels of bisphenol-A (BPA), estrone (E1), 17α-estradiol (αE2), 17β-estradiol (E2), estriol (E3), and 17α-ethynylestradiol (EE2) were measured in sediment samples collected along the coastline of Macao. BPA was found in all 45 collected samples with lower BPA concentrations associated to the presence of mangrove trees. Biodegradation assays were performed to evaluate the capacity of the microbial communities of the surveyed ecosystems to degrade BPA and its analogue BPS. Using sediments collected at a WWTP discharge point as inoculum, at a concentration of 2 mg l^{-1} complete removal of BPA was observed within 6 days, whereas for the same concentration BPS removal was of 95% after 10 days, which is particularly interesting since this compound is considered recalcitrant to biodegradation and likely to accumulate in the environment. Supplementation with BPA improved the degradation of bisphenol-S (BPS). Aiming at the isolation of EDCs-degrading bacteria, enrichments were established with sediments supplied with BPA, BPS, E2 and EE2, which led to the isolation of a bacterial strain, identified as *Rhodoccoccus* sp. ED55, able to degrade the four compounds at different extents. The isolated strain represents a valuable candidate for bioremediation of contaminated soils and waters.

Keywords Endocrine disrupting chemicals (EDCs) · Mangrove · Biodegradation · EDCs-degrading bacteria · Bisphenols · Estrogens

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Introduction

Endocrine disrupting chemicals (EDCs) are anthropogenic and natural chemicals which mimic or block hormonal action, thus triggering or preventing a cellular response and interfering with essential

endocrine functions such as reproduction, growth, and development (Flint et al. 2012). Several EDCs are not acute toxicants at environmental concentrations, and the consequences on wildlife and humans become apparent only during early adulthood affecting the reproductive system (Colborn et al. 1994; Flint et al. 2012). The specific interaction between the hormone and the receptor implies that a slight variation of concentration may cause a significant physiological response and may result in a permanent change, even in mature organisms and subsequent generations (Colborn et al. 1994; Diamanti-Kandarakis et al. 2009).

The occurrence of EDCs in the environment is associated with various products such as pesticides, fuels, surfactants, heavy metals, industrial chemicals, plastics and plasticizers, food additives, food and drink packaging, personal care products, and pharmaceutical agents which ultimately end up in the environment during their manufacture, use, or disposal. The presence of EDCs in municipal and industrial effluents is a great concern for marine and coastal environments near urban centers (Yamazaki et al. 2015). Wastewater treatment plants (WWTPs) do not completely eliminate EDCs (Kim et al. 2007) and the physical properties of different EDCs, such as solubility and sorption, can affect their biodegradation during wastewater treatment (Ying and Kookana 2003). Also, the aquatic ecosystems in the proximity of contamination sources, such as hospitals and pharmaceutical production facilities, are affected by specific EDCs with high levels detected in the water (Lin and Tsai 2009). In China, EDCs were detected in wastewater effluents and surface waters and in riverine and marine sediments (Wang et al. 2011; Zhang et al. 2014; Sun et al. 2014), including the Pearl River Delta (Peng et al. 2008; Zhao et al. 2009; Gong et al. 2011), Taiwan (Lin and Tsai 2009), and Hong Kong (Xu et al. 2014).

Recently, significant attention has been given to bisphenol-A (BPA) as it is one of the most common EDCs in the environment whose potential negative effects on human health and wildlife has been reviewed (Vandenberg et al. 2009, 2013) such as its carcinogenic potential (Seachrist et al. 2016) or effects on female fertility (Ziv-Gal and Flaws 2016). Concerns about the risks associated with BPA exposure urged several governments to regulate the usage of this chemical and to force manufacturers and retailers to shift to BPA-free products (Brede et al. 2003). The substitution of BPA with analogue compounds such as bisphenol-S (BPS) is not regulated, and endocrine-disruptive effects of BPA-substitutes have been reported (Molina-Molina et al. 2013; Mersha et al. 2015; Eladak et al. 2015; Rochester and Bolden 2015). Estrogens have been classified carcinogens and long-term exposure can disrupt sexual development in animals, in particular in humans (Chen et al. 2017). There are reports in which the concentrations of theses hormones detected in the effluents of WWTPs was higher than PNECs for ecological toxicity to aquatic organisms, representing risks to aquatic ecosystems (Tran et al. 2018).

Different studies are present in literature about the degradation of BPA in river water, sediments and sludge from WWTP (Ike et al. 2000; Kang and Kondo 2002; Ying and Kookana 2003; Suzuki et al. 2004; Zhao et al. 2008; Mohapatra et al. 2010). Although several different metabolic pathways have been described, the structure, effects and fate of the intermediate metabolites still have to be clarified (Im and Löffler 2016). BPS has been reported to degrade slower than BPA, and the degradation of BPA and BPS in saline coastal environment appears to be limited (Danzl et al. 2009; Kang and Kondo 2005). Several estrogen-degrading bacteria have been isolated from wastewater, however less is known about estrogen degradation on other environments and the synthetic hormone (EE2) was found to be more resistant to biodegradation than natural estrogens (Cajthaml et al. 2009). Moreover, knowledge about biodegradation kinetics and estrogenic potential of estrogen metabolites is crucial to understand their fate in the environment (Yu et al. 2013).

Due to EDCs high bioactivity, ubiquitous nature, toxicity and persistence, it is important to investigate the occurrence and degradation these compounds. The main aim of this study was to analyze the occurrence of selected EDCs, estrone (E1), 17β-estradiol (E2), 17α-estradiol (αE2), estriol (E3), ethinylestradiol (EE2), and BPA, in sediment samples collected along the coastal areas of Macao, a city located in the estuary of the Pearl River, opposite to Hong Kong, and one of the most densely populated regions in the world. Moreover, the influence of the presence of mangrove trees on BPA levels found on the surveyed sites was assessed. Another objective was to access the biodegradation potential of bacterial consortia

retrieved from those sediments, aiming at the isolation of EDCs-degrading bacterial strains, which may be used for bioremediation applications.

Methodology

Chemicals and reagents

BPA, BPS, E1, αE2, E2, EE2, and E3 standards with 97% purity or higher were purchased from Sigma-Aldrich (Oakville, ON, Canada and Steinheim, Germany). Internal standards, BPA-d_{16} and E2-d_3, were also acquired from Sigma-Aldrich. HPLC-grade methanol (CH_3OH), acetonitrile (C_2H_3N), ethyl acetate ($C_4H_8O_2$), n-hexane (C_6H_{14}) and dichloromethane (CH_2C_{l2}) were purchased from Merck (Darmstadt, Germany) and Sigma-Aldrich. Analytical grade acetone was purchased from Bodi Chemical (Tianjin, China). Dansyl chloride was obtained from Alfa Aesar (Johnson Matthey, MA, USA). Trifluoroacetic acid (TFA, analytical grade) was obtained from Merck (Darmstadt, Germany). Ultra-pure water was supplied by a Milli-Q water system (Merck Millipore, Billerica, MA, USA). Silica gel (100 mesh size) was purchased from Hai Lixin Company (Qingdao, China). Minimal salts medium (MM) (Moreira et al. 2013) was prepared with analytical grade chemicals purchased from Sigma-Aldrich. The nutrient broth and agar were purchased from Sigma-Aldrich.

For the monitoring of EDCs along the coastal area, stock solutions of BPA, E1, αE2, E2, EE2, and E3 were prepared in methanol/acetonitrile (50:50) and diluted to obtain working solutions. A standard mixed solution containing 200 ppm of each compound was prepared in methanol for determining recovery rates. An internal standard solution of BPA-d_{16} was prepared in methanol and of E2-d_3 was prepared in acetone. An internal standard mixed solution was prepared in methanol at final concentrations of 50 ppm of BPA-d_{16} and E2-d_3. Derivatization agent dansyl chloride and $NaHCO_3$ solutions were prepared in acetone and stored at 4 °C. Stock solution, working solution, and internal standard solutions were stored at -20 °C. For the biodegradation assays, stock solutions of BPA, BPS, E2 and EE2 were prepared in methanol and diluted to obtain working solutions.

Sample collection

Sediment samples (45) were collected along the Macau coastline, in Taipa and Coloane Islands, and Cotai area. Considering land use and zoning, 6 different sampling areas were defined (Fig. 1) as follows: the discharge point of the Coloane WWTP; the Cotai Ecological Zone, a protected coastal area mostly covered with mangrove trees; the waterway along the Cotai cycling track; the waterway along the Taipa cycling track; industrial areas—an abandoned shipyard and the Coloane-A Power Station; and beach areas. All samples were collected at a depth of 10 cm. In the mangrove areas, the samples were collected from the rhizosphere and 1 m away from the roots. Each sample consisted in a composite of 3 individual samples which were pooled together in situ. The samples were collected with a small metallic shovel, wrapped in clean aluminum foil, sealed in polyethylene bags and kept cold at 4 °C during the transportation and then stored at -80 °C until being processed for analytical determinations. From the samples collected for the environmental survey, some samples were also used to perform biodegradation experiments: from Cotai Ecological Zone, Coloane WWTP, Taipa cycling track and industrial areas (near the Coloane-A Power Station and near an abandoned shipyard). The samples used for biodegradation experiments were those collected from mangrove areas. Mangrove species identified in the sampled areas were *Acanthus ilicifolius*, *Aegiceras corniculatum*, *Kandelia obovata*, *Avicennia marina*, and *Sonneratia apetala*. The samples were placed into an autoclaved Schott bottle and kept at 4 °C until the start of biodegradation experiments.

Biodegradation assays

Biodegradation of BPA and BPS by microbial community from sediment samples

Some samples collected from mangrove areas for the environmental survey were also used to perform biodegradation experiments: Cotai Ecological Zone, Coloane WWTP, Taipa cycling track and industrial areas (near the Coloane-A Power Station and near an abandoned shipyard). For biodegradation assays in liquid media sediment samples (5 g) were added to 50 mL of minimal salts medium (MM) (Moreira et al.

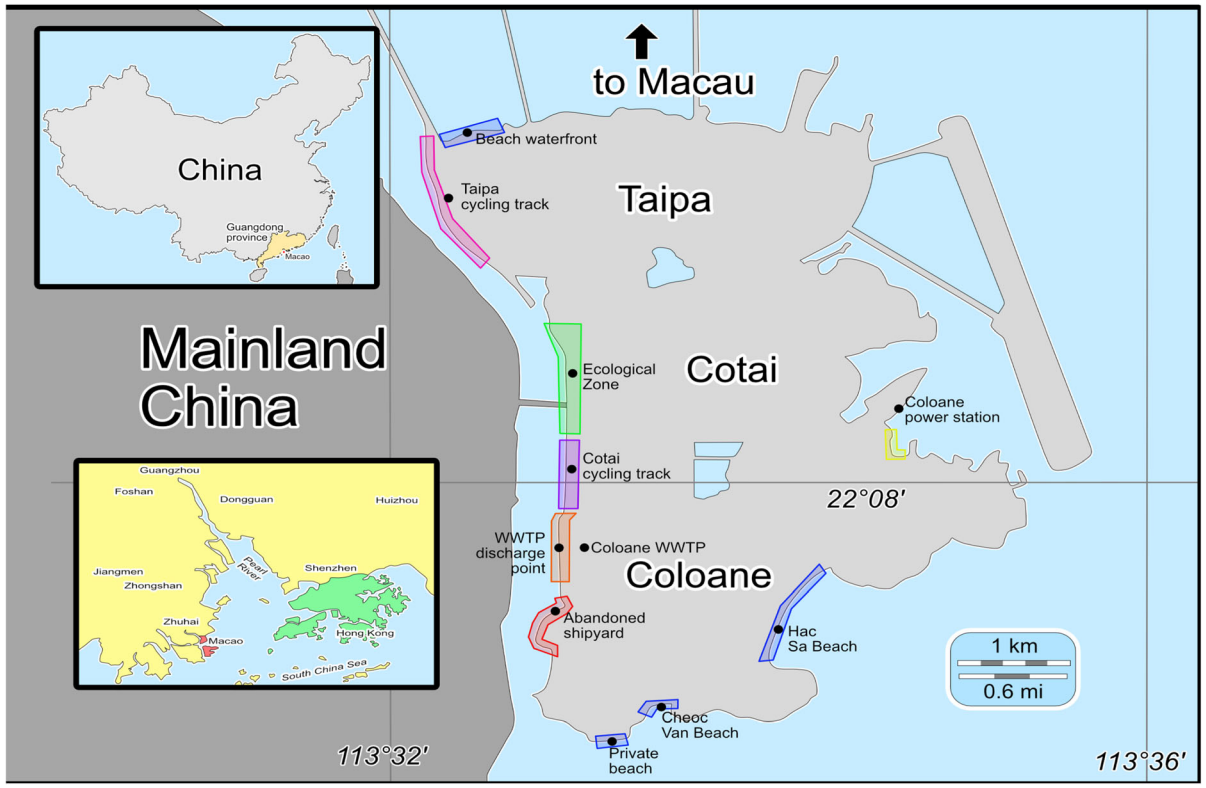

Fig. 1 Map of Taipa and Coloane Islands, Macau, S.A.R., China showing the location where the sediment samples we collected according to different land use and zonage: Cotai cycling track, intracoastal area near the Coloane WWTP, Cotai Ecological Zone, Taipa Cycling track, Coloane-A power station, Coloane abandoned shipyard, and beach waterfronts

2013); salinity was adjusted to 10 ppt with NaCl. Each flask was supplement with 2 mg l^{-1} of BPA or BPS. Additionally, assays containing mixtures of both substrates (2 mg l^{-1} BPA and 2 mg l^{-1} BPS) were also prepared for each sediment sample. Experiments were performed in triplicate under sterile conditions. Control assays inoculated with autoclaved sediments were established in order to evaluate adsorption and abiotic degradation. The flasks were protected from light with aluminum foil and incubated in an orbital shaker, at 30 °C, and 150 rpm. The experiment lasted 10 days. Samples were collected on days 0, 1, 2, 3, 6 and 10, to assess substrate removal.

Selective enrichments on BPA, BPS, E2 and EE2

For the establishment of the selective enrichments the samples from Coloane WWTP discharge area were used. Enrichments were performed in MM (75 ml), inoculated with sediment samples (5 g) and supplemented with one of the following compounds: BPA or BPS, at a final concentration of 15 mg l^{-1}; E2 or EE2 at a final concentration of 4.0 mg l^{-1}. Cultures were incubated in an orbital shaker at 25 °C and 130 rpm, protected from light. Half of the suspension was removed and replaced with fresh medium after 28 days. The whole enrichment experiment lasted 56 days. After the enrichment, degradation by the obtained bacterial consortia was tested also in MM (75 ml) supplied with 10 mg l^{-1} of each target compound. Cultures were incubated in an orbital shaker at 25 °C and 130 rpm, protected from light. Degradation by the consortia was assessed during two months. All experiments were done in triplicate and controls without inoculum were also monitored. Samples were collected at regular intervals to evaluate the degradation of target compounds.

Identification of bacterial isolates from BPA, BPS, E2 and EE2 degrading consortia

The culturable fraction of the obtained consortia was characterized. After the enrichment period, samples of the cultures were spread onto Nutrient Agar to obtain bacterial isolates. Bacterial isolates were grouped according to species similarity, based on RAPD profiles, as previously described by Amorim et al. (2014). Bacterial identification of isolates was performed by 16S rRNA sequencing analysis (Amorim et al. 2014). Genomic DNA extraction and further amplification by polymerase chain reaction (PCR) was performed as described elsewhere using universal bacterial 16S rRNA primers 27F and 1492R (Amorim et al. 2014). Sequencing was performed at Macrogen Inc. (Seoul, Republic of Korea) using universal bacterial 16S rRNA primer (f27). Identification and phylogenetic classification was performed using the BLAST software at the National Centre of Biotechnology Information website (http://www.ncbi.nlm.nih.gov/). The partial 16S rRNA gene sequences were submitted to the GenBank database.

Degradation of EDCs by the bacterial isolates

Bacterial isolates were streaked onto MM agar plates containing 25 mg l^{-1} of each compound, and incubated at 25 °C for 5 days in order to carry out the selection of the isolates with the ability to grow in the presence of the respective compound as sole carbon source. Isolates with the ability to grow on selective plates were re-inoculated into liquid MM containing 10 mg l^{-1} of each compound as a sole carbon and energy source to evaluate biodegradation of the target compound, using as initial biomass cultures with an OD600 of ca. 0.05. Compounds concentrations were determined by HPLC based on a method published elsewhere (Ribeiro et al. 2010). The biodegradation was monitored after 30 days. Purity of the cultures was evaluated through plating on NA plates.

Analytical methods

Analysis of EDCs for the monitoring along the coastal area

Simultaneous analysis of the six EDC's in sediments was performed by using a combination of ultrasonicated and solid phase extraction, followed by silica gel fractionation, derivatization with dansyl chloride, and determination by UHPLC/MS analysis by the method developed and validated by Yu et al. (2011). Recovery tests were performed by spiking matrix samples with 10 and 50 ppb of each compound in the extracted sediments; 3 samples were spiked for each concentration. A procedural blank and an instrumental blank were included in every batch of 12 field samples. Recovery rates from spiked samples were between 96.1 and 119.6% with a standard deviation between 9.97 and 37.42 ng g^{-1} dw. The recovery rates and limits of detection for each compound are presented in Table S1. The results in the present work were not corrected by surrogate recoveries.

EDCs analysis for biodegradation experiments by HPLC

The biomass was previously removed from culture samples by centrifugation at 14,000 rpm for 10 min at 4 °C. BPA and BPS concentration was determined by HPLC based on a method published elsewhere (Ribeiro et al. 2010). The HPLC analyses were performed on a System Gold 126 (Beckman Coulter, Fullerton, USA) using a reversed phase 250–4 HPLC-Cartridge LiChrospher 100 RP-18 column (Merck), operated in isocratic mode at room temperature, with a flow rate of 1.0 ml/min and an injection volume of 20 µL. Acetonitrile/water (60:40, V/V) acidified to pH 2 with TFA was used as the mobile phase. The eluted peaks were monitored at 230 nm.

Degradation rate constants were calculated assuming first-order kinetics. With this model, the residual concentration changes with time (t) were determined according to the following relationship: $C = C_0 e^{-kt}$, where C_0 is the initial concentration and k is the degradation rate constant. The half-life of biodegradation ($t_{1/2}$) is estimated from k using: $t_{1/2} = \ln 2/k$.

Statistical analysis

Statistical analysis was performed with IBM SPSS Statistics for Windows, Version 22.0. Armonk, NY: IBM Corp. Released 2013.

Results

Monitoring of EDCs along the coastal area

The six target EDCs were detected in the sediment samples from Macau coastline (Table 1 and S2). BPA was found with the highest detection frequency (100%), having been detected in all 45 surface sediments samples in concentrations ranging from 1.38 to 34.44 ng g^{-1} dw. In relation to the steroid hormones, E1, E2 and αE2 were present in more than 70% of the samples. The maximum detected concentrations were 9.05 ng g^{-1} dw for E1 and 1.01 ng g^{-1} dw for E2 and αE2. On the other hand, E3 and EE2 were detected in less than 20% of the analysed samples, in concentrations ranging from ND to 0.29 and 0.96 ng g^{-1} dw. The concentration of BPA did not differ between the 6 sampled areas (one-way ANOVA $F_{6,42} = 1.488$, $p = 0.206$). The only significant difference between sampling sites was found in the beach areas where the levels of E1, E2 and αE2 were below detection limit. These hormones were also not detected in some samples collected along the Taipa cycling track, located further away from the Coloane WWTP discharge point and industrial areas, in which the detection frequency was below 70%. EE2 was also not detected in beach areas and cotai cycling track and in the other areas the detection frequency was below 20%. In all sampling sites, except for the beach areas where no mangroves were present and the cotai cycling track, samples were collected from sections with mangrove and sections without mangrove. Lower concentrations of BPA were measured in samples associated to the presence of mangrove trees. The BPA concentration in the sediments collected from the mangrove areas was 7.45 ± 4.31 ng g^{-1} dw and 14.91 ± 9.62 ng g^{-1} dw in the absence of mangrove (Table 2) and these differences were statistically significant (one-way ANOVA, $F_{2,37} = 9.367$, $p = 0.001$). Post hoc comparisons showed that BPA levels in samples collects in the rhizosphere were significant lower as compared to samples collected in the absence of mangrove (Tukey HSD, $p = 0.000$ and $p = 0.004$, respectively). Additionally, the samples collected from the mangrove rhizosphere and in areas without mangrove were compared within each site. A repeated measure ANOVA detected a difference between the samples ($F_{2,14} = 14.025$, $p = 0.000$) and the Mauchly's test ($\chi 2(2) = 5.253$, $p = 0.072$) did not indicate any violation of sphericity. For the same sampling point, pairwise comparisons showed that the mean of BPA concentration from the samples collected inside the mangrove rhizosphere is lower than the samples collected in the areas without mangrove ($p = 0.018$ and $p = 0.16$).

Biodegradation of BPA and BPS by microbial community from sediment samples

To evaluate the ability of the autochthones microbial communities to degrade the target compounds and to select the most promising location for the isolation of EDCs-degrading bacterial strains, degradation experiments were set up using sediment samples from the surveyed sites as inocula. All the sediments allowed for the complete removal of BPA was observed within a few days when supplemented as single or mixed substrate. BPA was completely removed till day 6 in all the conditions tested (Fig. 2). The BPA decrease in the sterilized controls represented ca. 27% removal in the first day, stabilizing thereafter, which is consistent with adsorption of the compound to the sediments. The BPA and BPS degradation rate constants were well fitted to the first-order kinetics (the value of $R^2 = 0.911$–0.997) (Table 3). BPA degradation was slower in the assays inoculated with sediments from the Ecological Zone when compared to the other inocula. BPA degradation was slower when supplemented in mixture with BPS.

Complete removal of BPS was only observed when supplemented in mixture with BPA, in the experiments inoculated with sediments collected from discharge point of Coloane WWTP and industrial areas (Fig. 2) whereas almost complete removal (95%) was achieved as single substrate in the assays inoculated with samples from the WWTP discharge point. In the other experiments the removal of BPS was not significant when compared to the controls with inactivated bacteria, indicating no biodegradation of this compound. These results revealed a clear trend of BPS degradation being achieved in the experiments performed using inocula from industrial/WWTP areas in opposition with non-industrial zones. Considering the cases in which total removal was achieved, degradation of BPS was faster when supplemented in mixture with BPA (Table 3).

Table 1 Quantification of selected EDCs (ng/g dw) from coastal sediments from Macao SAR, China

Sampling site	EDCs	Frequency (n)	Frequency (%)	Range	Mean	Median
Cotai cycling track	BPA	6	100	2.77–9.63	5.72	5.15
	E1	6	100	1.15–4.70	2.23	1.40
	E2	6	100	0.38–0.49	0.41	0.39
	αE2	6	100	0.38–0.43	0.40	0.40
	E3	0	0	ND	ND	ND
	EE2	0	0	ND	ND	ND
WWTP discharge area	BPA	10	100	1.38–17.01	8.11	6.28
	E1	10	100	0.83–9.05	2.36	1.62
	E2	10	100	0.41–1.01	0.51	0.44
	αE2	9	90	ND–1.01	0.44	0.41
	E3	0	0	ND	ND	ND
	EE2	1	10	ND–0.96	0.10	0
Ecological Zone	BPA	8	100	1.83–34.44	11.02	8.52
	E1	7	88	ND–4.01	1.78	1.51
	E2	7	88	ND–0.52	0.37	0.42
	αE2	7	88	ND–0.47	0.36	0.40
	E3	1	13	ND–0.14	0.02	0
	EE2	1	13	ND–0.35	0.04	0
Taipa	BPA	10	100	4.28–31.31	12.13	10.29
	E1	7	70	ND–2.98	1.16	1.32
	E2	6	60	ND–0.58	0.28	0.36
	αE2	6	60	ND–0.58	0.28	0.36
	E3	2	20	ND–0.17	0.03	0
	EE2	2	20	ND–0.48	0.08	0
Industrial Areas	BPA	6	100	3.33–23.53	10.33	6.85
	E1	4	67	ND–1.49	0.79	0.95
	E2	4	67	ND–0.45	0.26	0.37
	αE2	4	67	ND–0.46	0.27	0.37
	E3	0	0	ND	ND	ND
	EE2	1	17	ND–0.38	0.06	0
Beach areas	BPA	5	100	4.77–18.60	10.58	7.78
	E1	0	0	ND	ND	ND
	E2	0	0	ND	ND	ND
	αE2	0	0	ND	ND	ND
	E3	5	100	0.11–0.29	0.17	0.15
	EE2	0	0	ND	ND	ND
Total sampling area	BPA	45	100	1.38–34.44	9.77	7.47
	E1	34	76	ND–9.05	1.50	1.27
	E2	33	73	ND–1.01	0.33	0.41
	αE2	32	71	ND–1.01	0.31	0.39
	E3	8	18	ND–0.29	0.03	0
	EE2	5	11	ND–0.96	0.05	0

Table 2 Quantification of selected EDCs (ng/g dw) in mangrove and non-mangrove sampling sites

Sampling site	EDCs	Frequency (n)	Frequency (%)	Range	Mean	Median
Mangrove	BPA	31	100	1.38–19.0	7.45	6.62
	E1	30	97	ND–9.05	1.84	1.4
	E2	29	94	ND–1.49	0.42	0.42
	αE2	28	90	ND–1.01	0.39	0.39
	E3	1	3	ND–0.17	0.01	0
	EE2	4	13	ND–0.96	0.07	0
No Mangrove	BPA	14	100	ND–34.44	14.91	14.75
	E1	4	29	ND–3.51	0.76	0
	E2	4	29	ND–0.52	0.14	0
	αE2	4	29	ND–0.58	0.14	0
	E3	7	50	ND–0.17	0.08	0.06
	EE2	1	7	ND–0.38	0.03	0

EDCs removal during the enrichment to isolate degrading bacterial strains

Following the observed trend for higher removal achieved with microbial communities from industrial areas and especially from the WWTP discharge point in the first experiment, enrichments aiming at isolating bacterial strains with degrading abilities for the target compounds proceeded with sediments from those promising provenances. The objective was the isolation of culturable strains which may be applied for bioaugmentation to assist decontamination of polluted areas or wastewater treatment. Samples from sediments collected near the Coloane WWTP discharge point were used for the establishment of selective enrichments with BPA, BPS, E2 and EE2. In the beginning of the enrichment experiment, the estrogens (4.0 mg l^{-1}) were completely removed from the liquid media. The E2 was not detected after 7 days and the EE2 was not detected after 21 days (Fig. 3a). In relation to bisphenols, BPA was totally removed in 3 days while for BPS a 91% removal was observed in the beginning of the enrichment experiments, fed with 15 mg l^{-1}, during 28 days (Fig. 3b). The observed removal includes degradation and adsorption to particles from sediments.

The biodegradation by the consortia retrieved after two months of enrichment, was evaluated in assays supplemented with 10 mg/l of the target compound during 15 days. For the hormones, the degradation of EE2 was 77% of the supplied amount. E2 was completely degraded in one day by the obtained consortium (Fig. 3c). In relation to bisphenols, BPA was totally degraded in 3 days, while BPS was not degraded (Fig. 3d). BPS degradation was observed in the beginning of the enrichment but was not maintained in the final obtained consortium. No decrease on the concentration of EDCs on abiotic controls was observed, indicating that no abiotic losses occurred during the experiment.

Degradation of EDCs by bacterial isolates

A total of 27 strains were isolated from the degrading consortia according to morphological analysis of colonies, genotyping by RAPD-PCR and sequencing of the 16sRNA gene of the isolates displaying distinct RAPD profiles. The partial 16S rRNA gene sequences of the isolated strains were submitted to the GenBank database under accession numbers presented at Table 4. Among them, 13 of the isolates were affiliated with representatives of the β-Proteobacteria, 6 of the isolates were affiliated with microorganisms belonging to γ-Proteobacteria and 5 with Firmicutes. The remaining 5 isolates clustered with α-Proteobacteria, Actinobacteria and Bacteroidetes.

The ability of the bacterial isolates to degrade the target EDCs was evaluated first assessing their growth on minimal salts medium agar plates containing the compounds and, and then in liquid minimal salts medium with the EDCs as sole carbon source. From the 27 bacterial isolates, 18 have shown ability to degrade the tested hormones at some extent, as pure cultures, while only two isolates (*Rhodococcus* sp. ED55 and *Staphylococcus* sp. ED63) were able to degrade between 19 and 34% of both bisphenols and

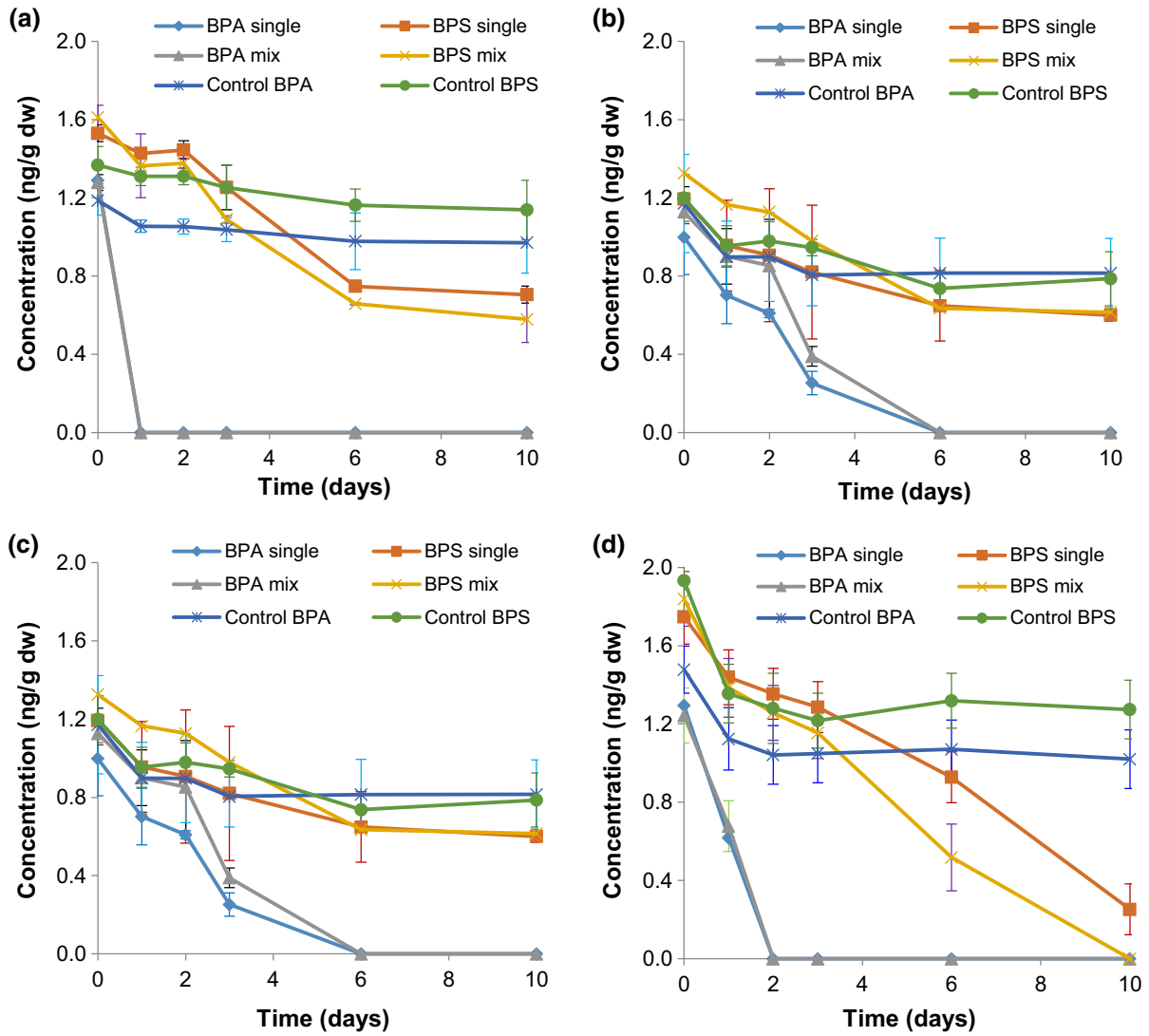

Fig. 2 Degradation of BPA and BPS by consortia from coastal sediment collected at **a** Cotai cycling track, **b** ecological zone, **c** Coloane WWTP discharge point and **d** industrial areas. BPA and BPS were added as a single or mixed (mix) substrate. Control experiments were performed with inactivated biomass

two other isolates were able to degrade around 20% of BPS (*Achromobacter* sp. ED64 and *Paenibacillus* sp. ED38). The higher extent of degradation by a bacterial isolate observed was 67% for EE2 and 100% for E2, supplied at 4.5 mg l^{-1}, and 23% for BPA and 34% for BPS, supplied at 9.5 mg l^{-1}, during 28 days. The achieved degradation for BPS is particularly interesting, since no degradation was observed by the consortium. Promising results were obtained by the bacterial isolate *Rhodococcus* sp. ED55, able to degrade the four compounds at different extents (Table 4).

Discussion

Monitoring of EDCs along the coastal area

The monitoring of EDCs revealed a ubiquitous contamination of sediment samples of the coastal area of Macau with BPA. Detection frequencies of 100%

Table 3 First-order rate constant (k) and half-life ($t_{1/2}$) for degradation of BPA and BPS by microbial community from sediment samples

	Supplementation	BPA				BPS			
		Degradation (%)	Abiotic (%)	K	t1/2	Degradation (%)	Abiotic (%)	K	t1/2
Cotai cycling track	Single	100	11	ND	< 1	54	17	0.083	8.35
	Mix	100		ND	< 1	64		0.105	6.6
Ecological zone	Single	100	30	0.239	2.9	50	34	0.14	4.97
	Mix	100		0.176	3.94	54		0.121	5.73
Coloane WWTP discharge point	Single	100	34	0.392	1.77	95	24	0.553	1.25
	Mix	100		0.348	1.99	100		0.301	2.30
Industrial areas	Single	100	31	ND	< 1	86	34	0.301	7.15
	Mix	100		ND	< 1	100		0.148	4.70

for BPA is similar with frequencies found in other locations in China (Liu et al. 2017; Wang et al. 2016; Zhang et al. 2014) and India (Tiwari et al. 2016). Moreover, the concentration range of BPA in the studied sediments was in the same order of magnitude of that reported for sediments of the Three Gorges Reservoir Region (4.7–41.1 ng g^{-1} dw) (Wang et al. 2016), Songhua River (1.60–17.3 ng g^{-1} dw) (Zhang et al. 2014) and Fen river (Liu et al. 2017) and also estuarine sediments from Mumbai, India (16.3–35.79 ng g^{-1} dw) (Tiwari et al. 2016). For the sediments of the Pearl River system previous studies reported values varying from ND–296 ng g^{-1} dw, with detection frequencies of 83 to 100% in South China (Liuxi River, Zhujiang River and Shijing River) (Zhao et al. 2011).

The synthetic hormone (EE2) was the compound detected at lower frequency. E3 and EE2 were also the less abundant EDCs. Similar pattern for these hormones was also observed in sediments of the Songhua River, in China (Zhang et al. 2014), while higher detection frequencies and concentrations were observed on sediments of the Three Gorges Reservoir Region (Wang et al. 2016). In the present study it was observed that the average concentrations were ranked in the order of E1 > E2 > E3, which is in accordance with those reported for effluents from WWTP in China (Huang et al. 2014). Higher concentration of E1 in relation to the other steroid hormones can be related with the fact that E2 has been reported to be transformed into E1 in WWTPs, and as E1 is more recalcitrant that explains the higher concentrations of E1 in the effluent and its accumulation on receiving environments (Liu et al. 2009). Similar results were reported for sediments in Fen River in China (Liu et al. 2017), river Ouse in UK (Labadie and Hill 2007) and rivers in the north of France (Kinani et al. 2010), in which EE2 was not detected. Zhao et al. (2011) also detected E1 with the highest frequency and the maximum concentration in comparison with E2 and EE2 in sediments of the Pearl River system. However, the steroid hormones concentrations reported were higher than that detected in the present study. On the other hand, E1 and E2 were not detected in sediments from the Yellow river, which was explained by its high content of sand and low organic content (Wang et al. 2012b). On the other hand, average occurrence of steroid hormones in sediment samples from Thane Creek area (India) were EE2 > E1 > E2, with high concentrations observed and associated with large inputs from urban and industrial wastewater (Tiwari et al. 2016). The same was observed in surface sediments from mangrove areas in Brazil (Froehner et al. 2012).

Compared with steroid estrogens, BPA presented not only higher frequency but also significantly higher levels, which corresponded to their wide application in industrial products and household wares, such as plastics, food packaging, and thermal paper (Noszczyńska and Piotrowska-Seget 2018). While the concentration of BPA was not significantly different between the sampled areas, significant differences

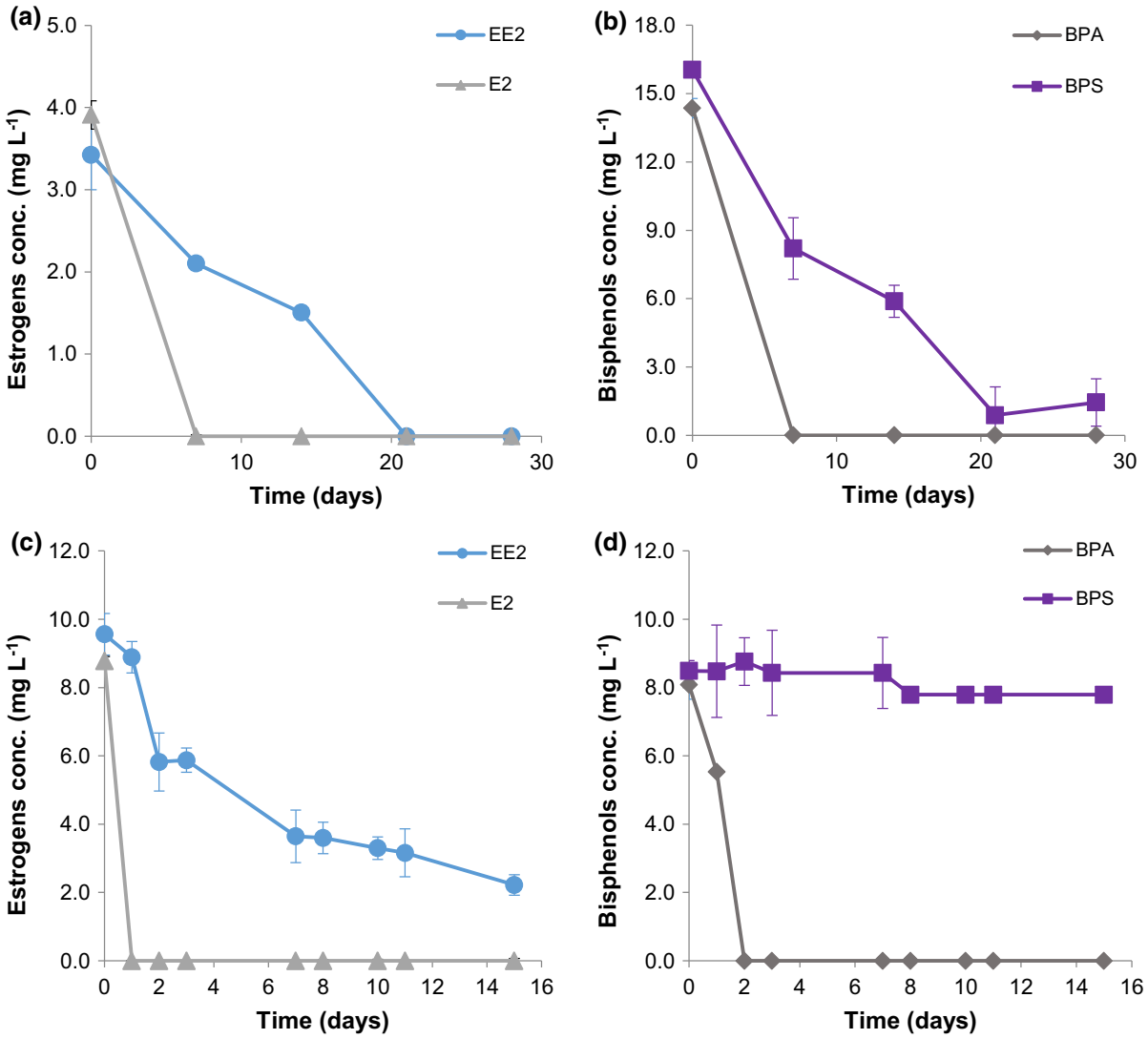

Fig. 3 EDCs removal in the first 28 days of enrichments, using as inoculum sediment samples collected at Coloane WWTP discharge point: **a** EE2 (circle) and E2 (triangle) and **b** BPA (diamond) and BPS (square). Degradation of EDCs obtained microbial consortia (after two months enrichment): **c** EE2 (circle) and E2 (triangle) and **d** BPA (diamond) and BPS (square)

were observed for the hormones in the beach areas. It is well known that due to their hydrophobic properties, steroid hormones tend to sorb to sediments (Wang et al. 2016; Zhang et al. 2016). There are some studies that indicate that this sorption tend to correlate with organic matter in sediments (Lai et al. 2000; Zhang et al. 2014). This can explain the lower concentration of steroid hormones observed in beach areas when compared with the other areas.

In relation to EE2, the higher concentration was detected in the sample from WWTP discharge area. Since EE2 is a synthetic hormone used in oral contraceptives, WWTP effluents are a major source to the environment. Also for the natural hormones, with exception for E3, a gradient with higher concentration detected near WWTP discharge area was observed, indicating that effluents discharge from WWTPs is a major way for these compounds to enter into the environment, as previously recognized (Zhang et al. 2016).

Another interesting observation was the significant differences on BPA concentration between samples

Table 4 Degradation of EDs compounds by bacterial isolates from enriched consortia

Isolate	Acession no	Phylogenetic affiliation	Closest relative (accession no.)	Similarity (%)	Growth on agar plates				Degradation on liquid media (%)			
					EE2	E2	BPA	BPS	EE2	E2	BPA	BPS
ED15	MT496779	*β-proteobacteria*	*Achromobacter xylosoxidans* strain GD003A	99	+ +	+ +	+ +	+ +	34.0	54.9	1	5.5
ED17	MT496780	*γ-proteobacteria*	*Stenotrophomonas maltophilia* strain ODW 2.4.3	97	−	−	−	−	−	−	−	−
ED19	MT496781	*γ-proteobacteria*	*Pseudomonas putida* strain LPK411	98	+ +	+ +	+ +	+ + +	28.0	53.7	1.78	10.2
ED24	MT496782	*Actinobacteria*	*Gordonia iterans* strain IFM 10,348	98	+ +	+ + +	+ + +	+ + +	35.1	52.8	0.8	5.9
ED38	MT496783	*Firmicutes*	*Paenibacillus* sp. H4-2–1 H	98	−	+ +	−	+ + +	−	56.3	−	21.9
ED55	MT496785	*β-proteobacteria*	*Rhodococcus ruber strain TH-22*	100	+ +	+ + +	+ + +	+ +	67.4	79.4	23.2	33.5
ED63	MT496786	*Firmicutes*	*Staphylococcus* sp. strain Atelim2I	99	+ +	+ + +	+ + +	+ + +	0	27.7	20.4	19.2
ED64	MT496787	*β-proteobacteria*	*Achromobacter denitrificans* strain SMV191#5	99	−	−	−	+ +	−	−	−	19.0
ED70	MT496788	*β-proteobacteria*	*Alicycliphilus* sp. UF5H	99	+ +	+ +	+	+ +	52.2	61.9	3.1	17.6
ED73	MT496789	*Firmicutes*	*Paenibacillus* sp. strain KB5	99	−	−	−	−	−	−	−	−
ED86	MT496792	*β-proteobacteria*	*Castellaniella* sp. strain WP-1	98	+ +	+	+ +	+ + +	67.4	55.2	8.5	11.1
ED89	MT496793	*β-proteobacteria*	*Castellaniella* sp. TCOB-5	99	+ +	+	+ +	+	25.2	55.7	7.4	10.2
ED95	MT496794	*γ-proteobacteria*	*Pseudomonas putida* strain VTs-23	96	+ +	+	+ +	+	30.9	72.6	0	9.5
ED96	MT496795	*γ-proteobacteria*	*Pseudomonas plecoglossicida* strain SLr02	99	−	−	−	−	−	−	−	−
ED106	MT496796	*α-proteobacteria*	*Ochrobactrum cytisi* strain HiB1	97	+ + +	+ + +	+ + +	+	6.3	49.3	2.8	7.2
ED113	MT496797	*β-proteobacteria*	*Cupriavidus malaysiensis* strain USMAA1020	99	+ + +	+ +	+ +	+	60.9	100	0	0
ED115	MT496798	*β-proteobacteria*	*Alicycliphilus* sp. R-24604	99	−	−	−	−	−	−	−	−
ED117	MT496799	*α-proteobacteria*	*Xanthobacter* sp. strain DMO-3	97	+ +	+ +	+ +	+ +	26.8	76.8	2.8	14.3
ED136	MT496801	*Bacteroidetes*	*Chitinophaga arvensicola strain B10*	98	−	−	−	−	−	−	−	−
ED138	MT496802	*Actinobacteria*	*Microbacterium* sp. strain Actino-49	97	−	−	−	−	−	−	−	−
ED158	MT496803	*γ-proteobacteria*	*Citrobacter freundii* strain I-N-1–1-1	97	−	−	−	−	−	−	−	−
ED174	MT496804	*γ-proteobacteria*	*Pseudomonas plecoglossicida* strain ZZYC13-1	97	+	+	+ +	+	33.1	65.7	9.6	11.4
ED185	MT496805	*β-proteobacteria*	*Alcaligenes faecalis* strain O1R4	97	+ +	+ +	+ +	+	64.7	62.0	4.4	11.4
ED190	MT496806	*β-proteobacteria*	*Achromobacter* sp. QUEBA08	99	+ +	+ +	+	+	61.0	63.2	0	9.2
ED206	MT496807	*β-proteobacteria*	*Alcaligenes faecalis* strain UWI9	94	+ + +	+ + +	+	+	58.9	65.3	1.8	3.3

Table 4 continued

Isolate	Acession no	Phylogenetic affiliation	Closest relative (accession no.)	Similarity (%)	Growth on agar plates				Degradation on liquid media (%)			
					EE2	E2	BPA	BPS	EE2	E2	BPA	BPS
ED209	MT496962	*Firmicutes*	*Paenibacillus lautus* strain JCM 9073	96	+	+ +	−	+	39.0	43.8	-	11.4
ED210	MT496808	*Actinobacteria*	*Microbacterium sp. strain 4*	98	−	−	−	−	−	−	−	−

from mangrove rhizosphere and samples collected outside mangrove areas. These results suggest a positive influence of mangrove trees for remediation of BPA. Removal of BPA from contaminated wastewater by the mangrove tree *Bruguiera gymnorhiza* was previously reported (Saiyood et al. 2013). In another study, removal of BPA was accelerated in the rhizosphere of *Phragmites australis*, while persisting in the absence of the plant, suggesting a positive interaction between the plant and the bacteria present in the rhizosphere for the removal of BPA from sediments (Toyama et al. 2009).

Biodegradation assays

Degradation of BPA was effective in cultures obtained from sediments collected along the coastal line of Macau whereas degradation of BPS was achieved when using as inocula sediments collected from WWTP and from industrial areas. Bacterial strains with the ability to degrade BPA, BPS, E2 and EE2 were isolated by enrichment from samples from the WWTP discharge point, the most promising location according to the first experiment. Focus on the enrichment of cultural degrading strains aimed at the attainment of bacteria that may be used in bioaugmentation for bioremediation of contaminated sites or treatment of wastewater. Degradation of BPA has been extensively studied while less attention has been given to BPA analogues, which are being used as BPA-replacers. Additionally, coastal environments are not well-studied as freshwater counterparts and it has been reported that BPA is much more recalcitrant to degradation in seawater than in river water (Kang and Kondo 2005). In the present study, salt concentration was adjusted in order to reproduce the natural conditions of the estuary ecosystems of Macau, revealing that degradation of BPA and BPS may occur within a few days with coastal sediments in brackish conditions. The coastal environment of Macau is contaminated with BPA, as revealed in the present study. It is accepted that autochthonous microorganisms present in polluted environments may be key players in the degradation of contaminants and bioremediation of polluted sites, since they are well adapted to the particular environmental conditions (Azubuike et al. 2016). In this study, microbial cultures from those polluted environments were able to degrade 2 mg l^{-1} of BPA in < 1 to 6 days (maximum half-life of 3.94 days), while similar studies reported half-life of 14.5 days in marine sediments spiked with 1 µg/g of BPA (Ying and Kookana 2003). In relation to BPS, 95% of 2 mg l^{-1} was degraded as single substrate with inocula from the WWTP discharge point, while other studies reported degradation of BPA (4–9 mg l^{-1}) in 3–12 days in microcosms but no degradation of BPS (Danzl et al. 2009). BPS is more difficult to degrade, thus suggesting a higher risk of accumulation in the environment.

Several bacterial strains have been isolated from different environments for its ability to degrade BPA (Zhang et al. 2013). Some of these bacteria are able to grow on BPA as sole carbon source (Masuda et al. 2007; Oshiman et al. 2007; Zhang et al. 2007; Fujiwara et al. 2016; Vijayalakshmi et al. 2017) while in other cases bacteria were able to transform BPA after grow on other substrates (Zühlke et al. 2017) or in co-metabolism with other compounds (Heidari et al. 2017). Degradation of BPA by *Cupriavidus basilensis* JF1, which was able to degrade BPA as sole carbon source, was significantly enhanced with the addition of phenol as co-substrate (Fischer et al. 2010). However, knowledge about BPA degradation by bacteria from coastal environments is scarce. Kamaraj et al. (2014) described the isolation of bacterial species with ability to degrade BPA from seawater and sediments from coastal regions of India. In the present study two bacterial strains (*Rhodococcus* sp. ED55 and *Staphylococcus* sp. ED63) with ability to degrade 20% of the supplied BPA as only carbon source were isolated. The BPA degradation by the isolates was lower than the degradation achieved with the bacterial consortium, in which total degradation was achieved in 3 days. Similar results were obtained by Peng et al. (2015), who described more efficient degradation by a microbial consortium (10 mg l^{-1} BPA within 28 h) then by isolated single strains. These results point out the importance of microbial diversity and cooperation for efficient degradation of recalcitrant pollutants. Interesting is the fact that the same isolates were also able to degrade BPS at similar extents. The availability of degrading strains is very important since these bacteria can be used for developing of bioremediation strategies. Previous reports have documented the benefits of bioaugmentation using isolated BPA-degrading microorganisms for degradation of BPA in polluted soil (Matsumura et al. 2015) and water–sediment microcosms (Xiong et al. 2017). Moreover,

the ability to degrade BPA and BPS can be of particular importance due to the increasing use of BPA analogues, resulting in environmental co-contamination (Yamazaki et al. 2015).

Estrogen degrading bacteria have also been isolated from various environments. The degradation can occur using the estrogens as only carbon source or in co-metabolism with growth substrates, as reviewed by Yu et al. (2013). The capacity to degrade E2 or to biotransform E2 into E1 have been identified in phylogenetically diverse bacterial strains (Yu et al. 2007; Jiang et al. 2010) namely *Rhodococcus* sp. strains (Kurisu et al. 2010; Liu et al. 2016; Yu et al. 2016). E2 degrading bacteria were isolated from sediments from deep sea (Fernández et al. 2017). The capacity to degrade EE2 is less observed and this synthetic hormone is considered more difficult to degrade in comparison with the natural estrogens. Larcher and Yargeau (2013) reported the degradation of EE2 by heterotrophic bacteria. Co-metabolic degradation of EE2 by nitrifying bacteria has shown to have a significant effect on EE2 removal (Song et al. 2017). An *Acinetobacter* sp. strain isolated from activated sludge demonstrated to be capable of cometabolic biodegradation of E2 and EE2. In the present study, several bacterial isolates revealed ability to degrade estrogens as only carbon source. Namely, *Cupriavidus* sp. 113 was able to completely degrade the supplied amount of E2 and 60.9% of EE2. These removal efficiencies by the isolates were very similar with those obtained by the consortia for the estrogens, unlike what happened for BPA.

Very interesting was the result obtained for the isolate ED55, which was identified as belonging to the genus *Rhodococcus*, able to degrade the bisphenols and the tested estrogens at significant extent, which is an impressive trait when compared with previous studies. To the best of our knowledge, this capacity has only been observed for white rot fungus (Křesinová et al. 2017). The enzyme Laccase from *Trametes versicolor* was shown to be able to oxidize E2, EE2 and BPA but not BPS, in spite of the structural similarity of the latter to BPA (Beck et al. 2018). Lignin peroxidase from the white-rot fungus *Phanerochaete sordida* YK-624 has also been shown to be able to degrade E2, EE2 and BPA, but it was not tested for BPS degradation (Wang et al. 2012a). Microbial strains have also shown capacity to degrade some of these the same contaminants, but not all of them. For example, the *Sphingomonas* strain KC8 was reported as having the capacity to degrade E2, E1 and testosterone, but could not degrade BPA (Roh and Chu 2010). *Pseudomonas putida* SJTE1 isolated from an enrichment culture of sludge was able to degrade natural estrogens, such as E2, and some estrogenic chemicals but it could not use EE2 or BPA (Wang et al. 2019). *Pseudomonas putida* strain YC-AE1 isolated as a BPA degrader from polluted soil, was able to also degrade related compounds, namely BPS, but was not tested for estrogens (Eltoukhy et al. 2020). In a previous enrichment experiment for the isolation of bacteria from activated sludge for the degradation of pharmaceutical and personal care products, two bacterial strains with the ability to degrade E2 and BPA were isolated but the isolation of bacteria capable of degrading EE2 failed (Zhou et al. 2013). Villemur et al. (2013) also isolated estrogen degrading bacteria from enrichments cultures but none of the isolates degraded EE2. The strains isolated in the present study that have shown biodegradation abilities represent valuable candidates for bioremediation of contaminated soils and waters. EDCs-degrading bacteria isolated from specific environments can well adapt to the unfavorable conditions and be use for bioremediation purposes (Zhang et al. 2016).

Conclusion

The simultaneous assessment of multiple EDCs in sediments from the coastal areas of Macau revealed that BPA was found to be ubiquitous in that environment. On the other hand, for the steroid hormones a gradient with higher concentration detected near WWTP was observed, pointing out the contribution of WWTP effluents to the environmental contamination with these compounds and the need to develop wastewater treatments able to remove micropollutants. Lower concentration of BPA was detected in the samples collected from the rhizosphere of mangrove trees in comparison with the samples outside mangrove areas, highlighting the benefits and potential of phytoremediation. Biodegradation experiments revealed the capacity of the bacterial communities from sediment samples for biodegradation of EDCs. This study is the first reporting complete degradation of BPS by bacterial communities from coastal sediments. Enrichments experiments allowed the isolation

of EDCs-degrading bacterial strains, which may be used for bioremediation proposes. Of particular importance was the isolation of the *Rhodoccocus* sp. ED55, which has shown the ability to degrade the four tested compounds at significant levels.

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Authors' contributions All authors contributed to the study conception and design. Sample collection and statistical analysis were performed by AL. Biodegradation experiments and isolation of EDCs degrading bacteria were performed by AL and ISM. Analysis of EDCs for the monitoring was developed by XP. ISM and AL wrote the paper. PMLC and DG finalized the paper. All authors have read, commented and approved the final manuscript.

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Declarations

Conflict of interest The authors declare that they have no conflict of interest.

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