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Microbial degradation of hydrophobic emerging contaminants from marine sediment slurries (Capbreton Canyon) to pure bacterial strain

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A B S T R A C T

Despite emerging contaminants (ECs) are more and more monitored in environmental matrices, there is still a lack of data in marine ecosystems, especially on their fate and degradation potentials. In this work, for the first time, the degradation potential of synthetic musks (galaxolide and tonalide), UV filters (padimate O and octo-crylene) and a pharmaceutical compound (carbamazepine) was studied in marine sediment samples, under laboratory conditions using sediment slurry incubations under biotic and abiotic conditions. Minimum half life times under biotic conditions were found at 21 days, 129 days and 199 days for padimate O, galaxolide and carbamazepine, respectively. Enrichments conducted under anoxic and oxic conditions demonstrated that de-gradations after one month of incubation either under both biotic and abiotic conditions were limited under anoxic conditions compared to oxic conditions for all the contaminants. Novel aerobic bacteria, able to degrade synthetic musks and UV filters have been isolated. These novel strains were mainly related to the Genus *Bacillus*. Based on these results, the isolated strains able to degrade such ECs, can have a strong implication in the natural resilience in marine environment, and could be used in remediation processes.

1. Introduction

As a result of the last century, Man created and is still creating new synthetic substances. Although well known persistent organic

contaminants, such as organochlorine pesticides or polycyclic aromatic hydrocarbons, are regulated in industrialized countries (European Commission, US, Japan) for several decades, regulation for emerging contaminants (ECs) arised only in the early of the 21th century and

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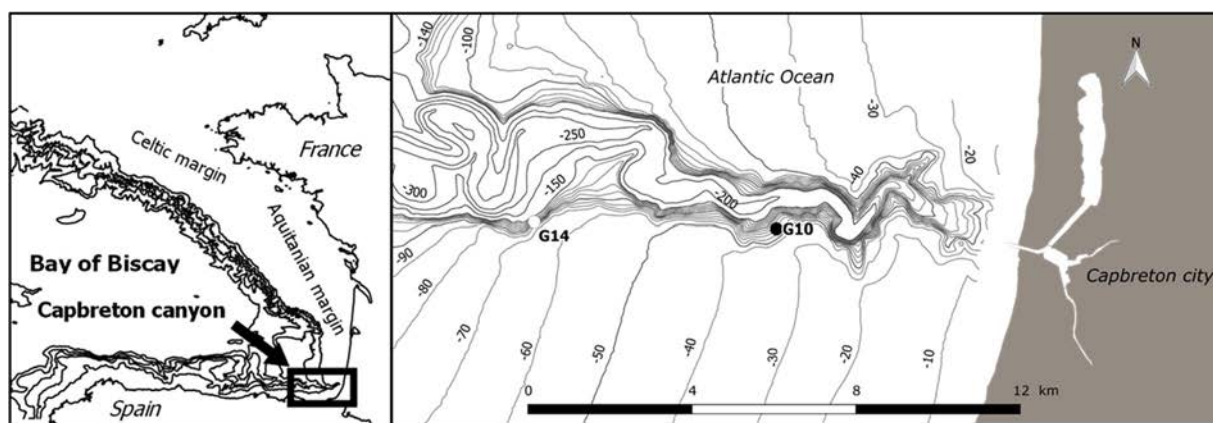


Fig. 1. Sediment sampling sites in the Capbreton Canyon (dark circle) and on the adjacent continental shelf (white circle). Isobath data from SEDIMAQ3 (Gillet 2012).

their update is still ongoing in Europe (Directive 2008/105/EC and Commission implementing decision 2018/840). Indeed, the scarce information available of their occurrence, reactivity and impact have led to a rising interest in identifying and screening these new compounds in the environment (Field et al., 2006). Among those ECs, pharmaceuticals and personal care products (PPCPs) are substances widely consumed and continuously released in the environment, mainly through wastewater, both treated and untreated (Li, 2014; Rainieri et al., 2017).

Marine ecosystem is the final receptor for these organic ECs. They were found in marine sediments, mainly close to the high density population coast nearby the main estuaries (Beretta et al., 2014; Huang et al., 2016; Pintado-Herrera et al., 2017; Azaroff et al., 2020).

Marine sediments act as integrative matrices reflecting the pollution state in a given area (Bellas et al., 2011; Roberts, 2012). The affinities of contaminants with the suspended particulate matter (SPM) lead them to be readily scavenged from the water column and to be deposited in the sediments. Submarine canyons are known to act as transfer zones of suspended particulate matter and contaminants between the continent and the open ocean where organic pollutants can be accumulated (Azaroff et al., 2020; Paull et al., 2002; Costa et al., 2011) into these productive ecosystems containing important stocks of commercially important fishes. Capbreton Canyon is located nearby the coast, with important urban and agricultural activities.

Indeed, since the Capbreton canyon is connected to the Adour estuary, its geomorphology and the local currents lead to transfer particles and micropollutants further, amplified during storm events (e.i. turbid currents) (Mulder et al., 2001; Salles et al., 2008; Brocheray et al., 2014). For instance, in the Capbreton canyon sediments collected in the first 25 km from the coast, UV filters and synthetical musks have been detected where the highest concentrations were observed in sediment collected at 25 km from the coast (Azaroff et al., 2020). This trend was also observed for mercury compounds (Azaroff et al., 2019). Additionally, wastewater treatment plants (WWTPs) are known to be an important pathway for introduction of ECs both through the treated effluents and particles released in aquatic environment (Miège et al., 2009; Verlicchi et al., 2012). In coastal areas, those WWTPs might be the main source of ECs in coastal and submarine sediments (Azaroff et al., 2020).

Microorganisms play a key role in ecological processes such as biogeochemical cycling and among them the carbon cycle and the organic compounds degradation. This bioremediation provides an important ecosystem service for the maintenance of the environment quality. The physicochemical properties of the sediment, such as organic carbon, grain size or pH, drive most of its interactions with the contaminants (Nowell et al., 2013). The bioavailability for the dwelling benthic organisms of these ECs is dependent on the adsorption, desorption and transformation processes which are themselves under

control of the biogeochemical parameters. Even at low concentrations, those ECs can be hazardous for the sediment-dwelling benthic organisms. It is widely accepted that their microbial co-metabolization could be the main degradation pathway leading to limited structural changes and incomplete mineralization (e.g. formation of degradation products). Consequently, the fates of ECs in submarine canyon sediments depend on physicochemical properties, and on the presence and activity of microorganisms

Several studies were performed to explore bioremediation by microorganisms of priority contaminants such as polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyl (PCBs) (Perelo, 2010; Duran and Cravo-Laureau, 2016). Few studies have shown the potential of bacteria and microbial consortia involvement in the ECs remediation (Ncibi et al., 2017). Biotransformation potential of ECs in natural and oceanic environments is still unknown and it is urgent to better understand their fate in marine ecosystems (Gavrilescu et al., 2015).

The aim of this present work was to study the degradation potentials of ECs such as synthetic musks (galaxolide, HHCB; tonalide, AHTN), UV filters (padimate O, OD-PABA; octocrylene, OC) and an antiepileptic (carmabazepine, CBZ) in marine sediments in order to highlight, for the first time their natural resilience in a submarine canyon sediments. Moreover, a specific focus was put on the isolation of pure strains exposed to these hazardous substances in order to estimate their capacities in biotransformation of these contaminants in natural ecosystems and more particularly in marine sediments

2. Material and methods

2.1. Study area and sampling strategy

The Capbreton Canyon (South-Eastern Bay of Biscay, NE Atlantic) begins 250 m from the coastline and reaches up to 3000 m water depth. In this study, we focused on two surface sediments (G10 and G14) sampled in the Capbreton Canyon area during the oceanographic cruise HAPOGE organized in July 2017. Surface sediments (0–10 cm) were sampled with Shipeck sampler grab. First sampling station was into the canyon (G10) and the second one was on the adjacent continental shelf (G14) at 4.4 and 9 km from the coast, and at 70 and 120 m of depth, respectively (Fig. 1). After collection, the sediment samples were placed into sterile polyethylene bags sealed and stored in the dark at 4 °C until slurry incubations in the laboratory (described further) within 48 h. In parallel, for each sample, contaminants analysis and geochemical parameters were analysed (Azaroff et al., 2020, 2019). Briefly, G10 sediments were more muddy than those in G14 and were characterized with particulate organic matter of 1.60 % and 0.92 %, respectively and fine grain size (< 63 µm) of 76 % and 46 %, respectively.

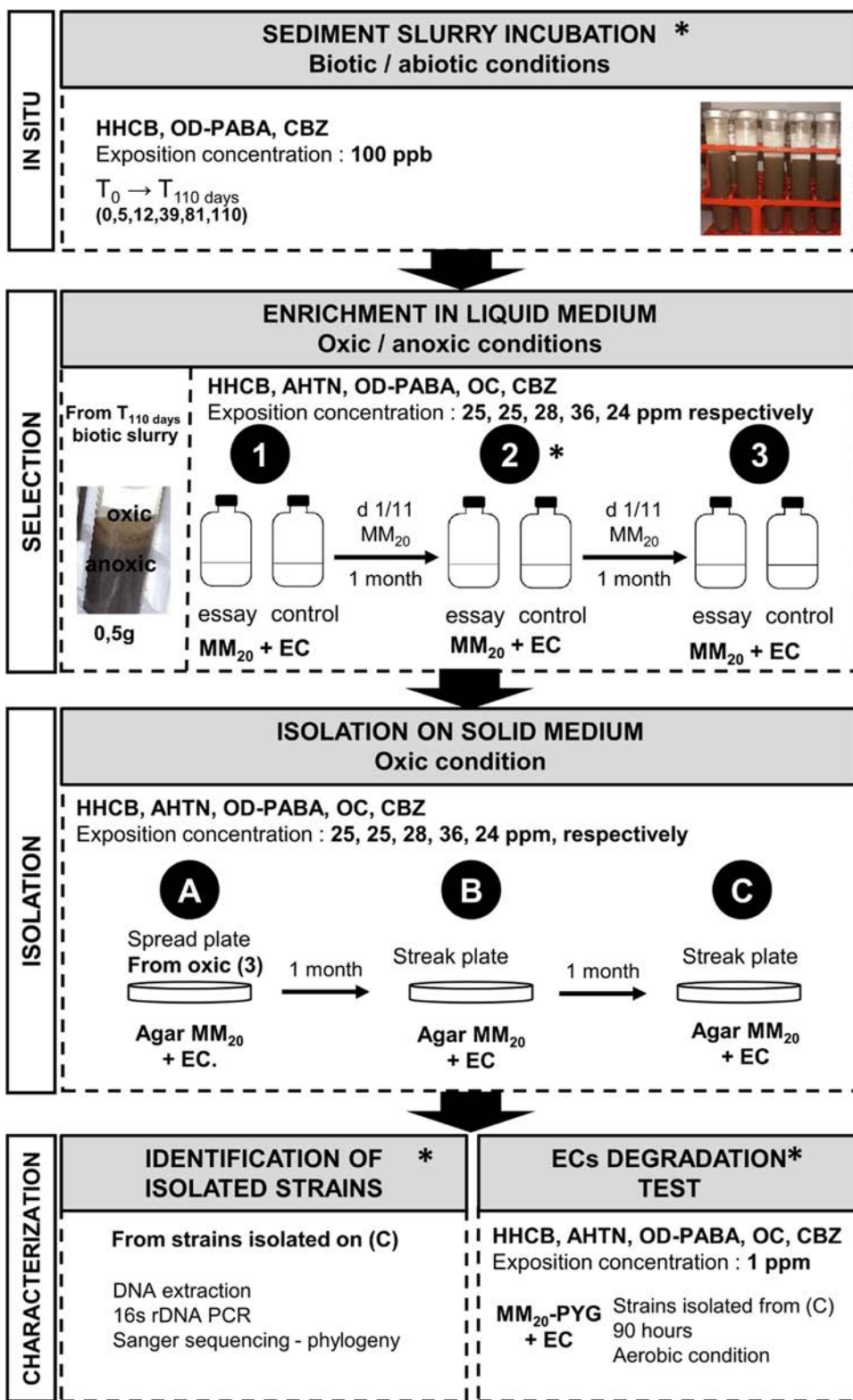


Fig. 2. Design of experiments to determine natural resilience and microbial strains involved in the degradation of emerging contaminants (ECs): Galaxolide (HHCb), Padimate O (OD-PABA), Carbamazepine (CBZ), Tonalide (AHTN) and Octocrylen (OC). First step was the determination of *in situ* degradation constant rates from slurry sediments spiked with three ECs under biotic and abiotic conditions at 100 ppb (AHTN and OC were not monitored). Second step was the selection of strains from the last slurry sediments incubation (day 110) through enrichments performed under oxic/anoxic and biotic/abiotic conditions at 25, 25, 28, 36 and 24 ppm of HHCb, AHTN, OD-PABA, OC and CBZ, respectively (corresponding to 100 μ M). These enrichments were repeated three times (1, 2, 3) with liquid multipurpose medium at 20 g L⁻¹ of NaCl, called MM₂₀. Third step was strains isolation from the last enrichment (3) where microbial consortia were separated (spread plates) on agar MM₂₀ at same ECs exposition concentrations than previously (A). After one month of growth, colonies were twice isolated by streak plating (B, C). The last step was the characterisation of isolated strains through both their identification and the determination of their ECs degradation capacities. Isolated strains were identified by 16s rDNA amplification and Sanger sequencing. ECs degradation tests were performed at 1 ppm of ECs using liquid MM₂₀ supplemented with peptone (5 g L⁻¹), yeast extract (2.5 g L⁻¹) and glucose (5 g L⁻¹), called MM₂₀-PYG. * indicates that results are presented in the figures.

2.2. Chemicals

Ethyl acetate (EtOAc) and methanol were of analytical grade and supplied by Sigma Aldrich (Saint-Louis, USA). Acetone (laboratory reagent. 99.5 %) used for cleaning the glassware was supplied by Fisher (Hampton, USA). Ultrapure water was obtained with a PURELAB Classic water purification system from Veolia (Paris, France). Reference standards of galaxolide (HHCb), octocrylene (OC), padimate O (OD-

PABA) and carbamazepine (CBZ) were purchased from Sigma-Aldrich. Tonalide (AHTN) was purchased from LGC Standards (Molsheim, France). Internal standards musk xylene-d15 (MX-d15) (100 ng μ L⁻¹ in acetone) was purchased from LGC Standards and carbamazepine-d10 (CBZ-d10) (100 ng μ L⁻¹ in methanol) was purchased from Sigma Aldrich.

2.3. Determination of half life: sediment slurry incubations

In the laboratory slurry incubations spiked with HHCB, OD-PABA and CBZ were performed for 110 days in order to estimate the degradation potential (Fig. 2). Briefly, for each station (G10 and G14), a slurry was prepared by mixing fresh sediment with underlying water (50:50, w:w). Incubation experiments were performed in 10 mL glass tubes sealed with PTFE stoppers filled with 10 g of slurry. For all assays under abiotic and biotic conditions, HHCB, OD-PABA and CBZ were added independently, to have a final concentration at $100 \mu\text{g g}^{-1}$. Initial time assays were stopped immediately by storing samples at -80°C whereas incubated assays were placed in the dark at 14°C (*in situ* temperature) and stopped at different elapsed times until 110 days. Slurries under abiotic condition (control), were twice sterilized for 20 min at 120°C . All assays were performed in triplicate.

Based on the assumption that degradation reaction of these contaminants are of pseudo-first-order (1), biodegradation rate constants (k) were estimated with the following Eq. (2) :

$$\ln(C) = f(t) \quad (1)$$

$$k = \ln(C_0/C_t) \quad (2)$$

Where C_t is the concentration of the contaminant at the kinetic time t; C_0 is the initial contaminant concentration; k is the biodegradation rate constant and t is the time. Then, half time reactions were also determined by the following Eq. (3):

$$t_{1/2} = \ln(2)/k \quad (3)$$

2.4. Enrichment experiments in liquid medium

In order to do a selective enrichment of microbial populations potentially involved in the ECs degradation, 0.5 g of oxic (surface, 0.1 cm, clear yellow color) and 0.5 g of anoxic (dark color) sediments (Fig. 2) were subsamples from the last days of sediment slurry incubations and were diluted with modified multipurpose medium (Widdel and Bak, 1992) at 20 g L^{-1} of NaCl, called MM₂₀. The medium contained : 1 L of MilliQ water, 1 mL of SL₁₂, 1 mL SeTg, HEPES 10 mM, yeast extract 0.1 g L^{-1} , 20 g NaCl, 3 g MgCl₂·6H₂O, 0.15 g CaCl₂·H₂O, 0.25 g NH₄Cl, 0.5 g KCl. pH was adjusted at pH = 7.5 before sterilization and autoclave (20 min at 120°C). After autoclaving, 1 mL of solution V7 (vitamins) and KH₂PO₄ (final concentration of 0.2 g L^{-1}) were added with a syringe and a cellulose nitrate $0.2 \mu\text{m}$ filter (Fisher).

While we determined the half life for 3 ECs, the enrichment experiments in liquid medium were performed for 5 ECs, HHCB, AHTN, OD-PABA, OC and CBZ. Since AHTN has the same molecular mass than HHCB but with a different chemical structure (Table 1), we performed the enrichment experiment for both from the HHCB sediment slurry incubation. Then, OC was added to this list due to its occurrence in coastal sediments (Combi et al., 2016; Mitchelmore et al., 2019). ECs

were added independently at $100 \mu\text{M}$ final concentration (corresponding to 25, 25, 28, 36 and 24 ppm, respectively) according previous experiments (Vallecillos et al., 2017; Balk and Ford, 1999; König et al., 2016; Sauvêtre et al., 2018). These ECs have low solubility in water (Table 1), and they were added first into the tube with organic solvent (EtOAc, or methanol for CBZ) and evaporated at ambient temperature, under microbiological safety cabinet (MSC). This step was performed in order to release organic solvent, a potential carbon source (Delgado-Moreno et al., 2019). Enrichment was performed in an agitator (120 rpm) (Vallecillos et al., 2017) under oxic and anoxic conditions, at 27°C for one month. This enrichment was repeated three times, with each time a dilution at approximately 1/11 of the previous assay. Briefly, under MSC, micropollutant was added in a falcon tube (50 mL) or penicillin tube (100 mL) (for oxic and anoxic conditions, respectively). Then 5.5 mL of the MM₂₀ were added before the addition of 0.5 g of anoxic and oxic sediments from the slurry incubations (1 st step, Fig. 2). After one month in the agitator, new tubes were prepared with ECs by using the same protocol. Then 5 mL of MM₂₀ were added and inoculated with 0.5 mL of the previous enrichment, corresponding to a 1/11 dilution (2nd and 3rd steps, Fig. 2).

2.5. Strains isolation experiments

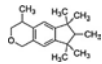
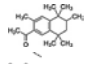
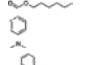
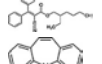
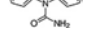
After the third enrichment step, in order to isolate the strains previously selected and enriched, solid medium of MM₂₀ (with 20 g L^{-1} of agar bacteriologic, previously rinsed 3 times with ultrapure water) were prepared in Petri plates. Under MSC, micropollutants were added (274, 274, 306, 498 and 254 μL of HHCB, AHTN, OD-PABA, OC and CBZ at 1000 ppm, respectively) at the surface of the solid medium and organic solvent was evaporated. Then, inocula of the last enrichments were spread at the surface of the solid medium with sterile inoculator and sealed with parafilm before incubation for 1 month, at 27°C to favour the growth of colonies. After one month, colonies were selected and isolated by streak plates as described (A, B, C steps, Fig. 2). Then all strains isolated were stored at -80°C in sterile LB (20 g L^{-1}) supplemented with NaCl (20 g L^{-1}), and glycerol 30 % (v:v).

2.6. Identification of isolated strains: DNA extraction, 16s rDNA gene amplification, sequencing and phylogenetic analysis

DNA was amplified from isolated strains after growth in LB Lennox medium. Amplification of the 16S rRNA gene was done with the universal primers 63 F (5'-CAG GCC TAA CAC ATG CAA GTC-3') and 1387R (5'-GGG CGG WGT GTA CAA GGC-3') (Marchesi et al., 1998). PCR amplification was performed using ampliTaq Gold® 360 master mix (Applied Biosystems, CA, USA), $0.2 \mu\text{M}$ of each primer and $1 \mu\text{L}$ of strain. PCR cycling was as following : after 10 min of initial denaturation at 95°C (lysis of cells), 35 cycles of 40 s denaturation at 95°C , 40 s annealing at 58°C and 60 s elongation at 72°C with 7 min final elongation at 72°C . Amplicons were sequenced by SANGER sequencing at

Table 1

Characteristics of the emerging contaminants used in this study. Log K_{ow} is octanol/water partition coefficient.

Compound	Family	Formula		Molar mass g mol^{-1}	Solubility in water mg L^{-1}	log K_{ow}	References
Galaxolide (HHCB)	Musk	C ₁₈ H ₂₆ O		258.4	1.75	5.70	(Field et al., 2006)
Tonalide (AHTN)	Musk	C ₁₈ H ₂₆ O		258.4	1.25	5.90	(Field et al., 2006)
Padimate O (OD-PABA)	UV filter	C ₁₇ H ₂₇ NO ₂		277.4	0.54	6.15	(Li, 2014)
Octocrylen (OC)	UV filter	C ₂₄ H ₂₇ NO ₂		361.5	0.36	6.88	(Rainieri et al., 2017)
Carbamazepine (CBZ)	Pharmaceutical	C ₁₅ H ₁₂ N ₂ O		236.3	17.7	2.45	EPA's EPI Suite™

GATC (Köln, Germany). Sequences were trimmed with ChromasPro (Technelysium software) and were aligned with MUSCLE (Edgar, 2004). A tree was generated using MEGA X software (Kumar et al., 2018), with the Maximum Likelihood method and Tamura-Nei model (Tamura and Nei, 1993) (with n replication bootstraps = 500). Phylogenetic analysis were processed with NCBI (<https://www.ncbi.nlm.nih.gov/>) and corresponding reference type strains as defined by the bacteriological code of nomenclature for Prokaryotes (<http://www.bacterio.net/~classifphyla.html>). Sequences are archived in GenBank under accession numbers MT658667 to MT658707.

In parallel, in order to compare with the isolated strains, DNA of *in situ* samples and from the final kinetic time of the slurry incubations were studied to determine the global diversity. DNA was extracted from frozen sediments with the QIAGEN DNeasy Powersoil kits (Qiagen Inc., Netherlands) according to the manufacturer's instructions. Diversity of the 16S rDNA were determined by sequencing the V4-V5 hypervariable regions of the 16S rDNA with universal primers V4–515F (5' GTGY-CAGCMGCCGCGGTA 3') and V5–928R (5' ACTYAAKGAATTGRCG-GGG 3') (Parada et al., 2016; Walters et al., 2015; Wang and Qian, 2009). PCR was performed using ampliTaQ Gold® 360 master mix (Applied Biosystems, CA, USA), 0.5 µM of each primer and 3 ng of extracted DNA. PCR cycling was as following : after 10 min of initial denaturation at 95 °C, 30 cycles of 30 s denaturation at 95 °C, 30 s annealing at 60 °C and 40 s elongation at 72 °C with 7 min final elongation at 72 °C. Amplicons were sequenced using MiSeq 250-pair-end technology (Illumina, CA, USA) with V3 kit version, in Get-plage sequencing platform (INRA, Toulouse, France). Data were preprocessed using Galaxy FROGS pipeline (Escudé et al., 2018). Chimera and PhiX reads were removed, Operational Taxonomic Units (OTUs) clustering, after a de-noising step allows building fine clusters with minimal differences, with an aggregation distance equal or above 3. Data were normalized with the minimum number of reads. Taxonomic affiliation was performed using the Silva database v.128 (Pruesse et al., 2007). Sequences data *in situ* and from the final kinetic time of the slurry incubations have been deposited in Genbank under the accession number PRJNA608532 and PRJNA640934, respectively.

2.7. Experimental ECs degradation test

Isolated strains were re-cultivated in medium MM₂₀-PYG (MM₂₀ medium supplemented with peptone, 5 g L⁻¹, yeast extracts, 2.5 g L⁻¹ and glucose at 5 g L⁻¹) à 37 °C for 90 h (Popa et al., 2014). Optical density (DO) at 600 nm was measured during the growth until the maximum growth using spectrophotometer (Spectronic 20). Degradation tests were performed in glass tubes with contaminants (HHCB, AHTN, OD-PABA, OC and CBZ) evaporated (Delgado-Moreno et al. 2019) to be at 1 ppm final concentration, with 1 mL of the cultivated strains, 9 mL of MM₂₀-PYG at 27 °C in agitator (120 rpm) (Vallecillos et al., 2017; Popa et al., 2014). 10 µL of micropollutant at 1000 ppm were added with sterile syringe and PTFE filter 0.2 µm (Fisher). Degradation was stopped when DO₆₀₀ reached the maximum value. For initial and final times, chemical analysis were performed (in triplicates) to evaluate the ECs degradation capacity of the isolated strains under controlled conditions (described below).

2.8. ECs analysis

HHCB, AHTN, OD-PABA, OC and CBZ were extracted by a liquid:liquid extraction, for both the second enrichment in liquid medium (3) and the ECs degradation test (Fig. 2) at the initial and final time of each experiment. Briefly, 10 and 2 mL of EtOAc were added directly into the essay tube before to vortex for 1 min, respectively. Then, 20 µL of the supernatant was diluted with EtOAc (960 µL) in GC-vial and spiked with 20 µL of an internal standard mixture at 10 µL ng⁻¹ (CBZ-d10 and MX-d15) before analysis. Extracts were analysed by 7890A gas chromatograph coupled with 5975C mass spectrometer (GC/MS) with an Electron

Ionization (EI) source using a Large Volume Injection (LVI) (Agilent Technologies). The GC/MS system was equipped with a single taper ultra-inert liner with glass wool and a HP-5MS UI capillary column (30 m length × 0.25 mm diameter and 0.25 µm film thickness). Carrier gas was helium (He) with a purity greater than 99.999 % (Linde). Separation was performed at a constant He flow of 1.5 mL min⁻¹, and the GC oven temperature was programmed to hold at 50 °C for 3 min, then increase at 25 °C min⁻¹ until 195 °C (hold 1.5 min), then 8 °C min⁻¹ until 265 °C (hold 1 min) and 20 °C min⁻¹ until 310 °C (hold 5 min). Injection volume was 20 µL in splitless mode. Instrument control, data acquisition and data treatment were performed using Agilent Chemstation software. Quantification was carried out in Selected Ion Monitoring (SIM) mode, selecting two characteristics fragments ions for each compound. Calibrations with the target analytes (HHCB, AHTN, OD-PABA, OC and CBZ) were used to quantify target compounds. A six-point calibration curve was performed in EtOAc spiked with increasing pollutants concentration levels ranging from 0 to 500 µg L⁻¹ as well as internal standards at 50 µg L⁻¹ transferred into GC vials (analytical performances are compiled in Table S1, supporting information). Recoveries achieved for all target ECs in MM₂₀ medium spiked at 100 µg L⁻¹ ranged from 91 % to 102 % for musk and UV-filter compound and was at 74 % for CBZ (Supplementary information, Table S1).

3. Results and discussion

3.1. Degradation of HHCB, OD-PABA and CBZ in sediment slurry incubations

Based on kinetic degradation for 110 days under both abiotic and biotic conditions (Supplementary information, Fig. S1), degradation rate constants, k, were estimated from slurry incubations under biotic condition (Table 2). While CBZ in both sediment samples exhibited similar rates (0.0035 ± 0.0004 and 0.0029 ± 0.0001), HHCB and OD-PABA exhibited different rates of degradation according the location (e.g. G10 and G14) with a factor around 2 (Table 2). This suggested that ECs degradations were depending on the geochemical parameters of the sediments (e.i. grain size, organic matter content). For instance, a previous study demonstrated that ECs have a strong affinity for particles (Li et al., 2016), as observed with sludge in wastewater treatment plants (Liu et al., 2012), suggesting those parameters and as well as environmental factors have to be taken in consideration to understand the fate of ECs in the ocean. Estimation of the half life time of reaction for these ECs indicated that the UV filter OD-PABA was between 21 ± 1 and 44 ± 4 days (Table 2). These results are similar with another UV filter degradation (i.e. EH-DPAB) estimated in marine sediments microcosm with a t_{1/2} ranging from 18 to 50 days (Volpe et al., 2017). Nevertheless, half life reaction for synthetic musk HHCB and anti-epileptic CBZ reached around 8 months and 7 months, respectively, indicated the high persistence of these contaminants in natural marine sediments compare to the UV filter OD-PABA (Table 2). Lack of data on HHCB and CBZ degradation

Table 2

First-order rate constants k (in days⁻¹) for degradation of HHCB, OD-PABA and CBZ in the slurry sediments incubations under biotic condition (in triplicate). Pearson determination coefficients ranged from 0.76 to 0.98 and p-values < 0.05 for estimation of k. The corresponding half-lives (t_{1/2}, in days) were calculated as ln(2)/k. G10 and G14 correspond to canyon surface sediment and continental shelf sediment, respectively.

Emerging contaminants	Slurry incub.	k ± SD (days ⁻¹)	t _{1/2} ± SD (days)
HHCB	G10	0.0032 ± 0.0005	224 ± 41
	G14	0.0065 ± 0.0029	129 ± 44
OD-PABA	G10	0.0324 ± 0.0014	21 ± 1
	G14	0.0158 ± 0.0014	44 ± 4
CBZ	G10	0.0035 ± 0.0004	199 ± 19
	G14	0.0029 ± 0.0001	231 ± 17

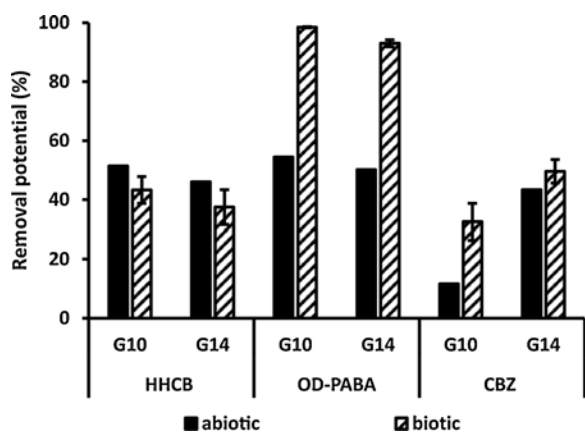


Fig. 3. HHCB, OD-PABA and CBZ removal potential (%) after 110 days of sediment incubation (under biotic and abiotic conditions from Capbreton Canyon sediments (G10, and G14). Initial exposure concentration was 100 ppb. Data are mean \pm SD of three replicates.

potentials in marine sediments does not allow comparison with these present data. Nevertheless, these preliminary results demonstrate the high persistence of these ECs in natural environment. In sediments, ECs may be associated to particulate matter, establishing equilibrium relations in the water-sediment interface, limiting their bioavailability. ECs remobilisation depends both of geochemical characteristics variations of the sediment, the overlying water column and pore water. Thus, ECs persistence in environment depend on coupling of abiotic and biotic factors (Lee Wolfe and Macalady, 1992; Jeon et al., 2016), such as physicochemical properties of the ECs, environmental factors (e.i. temperature (Lange et al., 2015), pH (Perelo, 2010), redox processes (Jeon et al., 2013) photolysis process (Chiron et al., 2006; Vogna et al., 2004)) and the presence of microbial communities able to degrade these ECs (Onesios et al., 2009). It was also demonstrated that bioturbation or resuspension improved oxidation of anoxic sediments leading an increase of lability of ECs which can be more available for degradation processes (Booij et al., 1992).

Additionally, chemical characterization of the collected sediments revealed that the three-targeted ECs were not found at concentration detectable with the analytical method used (Azaroff et al., 2020). The

removal potential was observed under both abiotic and biotic conditions with percentage of removal for each compounds, HHCB, OD-PABA and CBZ, ranging from 37.5%–51.4%, from 50.1%–98.5% and from 11.6%–49.6%, respectively (Fig. 3). Results of HHCB and CBZ slurry incubations suggested that the degradation was mainly driven by an abiotic process (except for G10 exposed with CBZ). Removal potential of OD-PABA under biotic condition was 2 times higher compared to the abiotic condition, suggesting that micro-organisms in these sediments improved the degradation of this UV filter compound (Fig. 3).

3.2. Enrichment in liquid medium

Results of the second enrichment in liquid medium (Fig. 4) demonstrated a higher removal in oxic conditions than under anoxic conditions for the five ECs studies (e.i. HHCB, AHTN, OD-PABA and CBZ). These ECs are well known to be observed in WWTP, where CBZ is frequently detected in the liquid effluents while the other contaminants, due to the very hydrophobicity, occur in sludges or remain through membrane post-treatment (Krzeminski et al., 2017). Although new advanced treatment processes, such as activated carbon adsorption, advanced oxidation processes, reverse osmosis or membrane bioreactors, are developed to increase the micropollutant removal of the WWTPs to struggle against the spread of micropollutants in aquatic environment, they are still observed in the WWTP effluents (Luo et al., 2014). Those results suggested that oxic processes could improve the ECs removal and limit their spread from the WWTP sewage. Moreover, higher removals under abiotic anoxic conditions than biotic were observed. It is likely due to a higher ECs adsorption on cells present in biotic condition leading to trap ECs and reduce the quality of the liquid extraction (Bell and Tsezos, 1987; Tsezos and Bell, 1989). Additionally, the high concentrations (ppm level) used for the selection of strains in these enrichments could inhibit microbial activity, thus could have mask their ECs removal capacities under anoxic condition (Barra Caracciolo et al., 2015).

Under oxic condition, similar removal potentials were observed for the abiotic and biotic condition, except for OC and CBZ (Fig. 4). That may indicate a potential involvement of microorganisms in the OC removal. Contrary to OC, CBZ removal was observed only in abiotic anoxic condition, likely due to the same explanations suggested previously for the removal potential under anoxic condition (e.i. adsorption on cells and/or high exposition concentrations). These

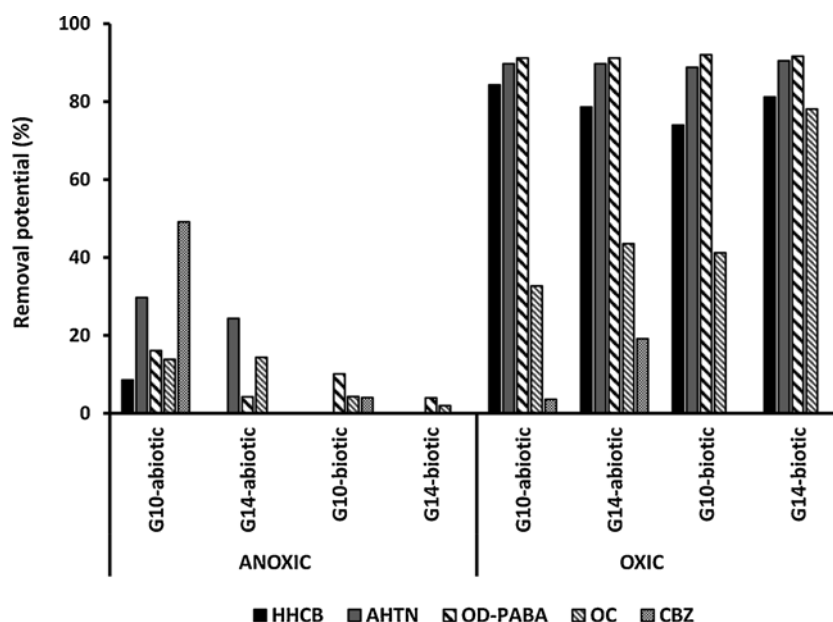


Fig. 4. HHCB, AHTN, OD-PABA, OC and CBZ removal potential (%) after one month on the second liquid enrichment under anoxic and oxic conditions with inoculum from G10 and G14 stations (biotic). Abiotic control was achieved without inoculum.

results led to continue the study of ECs degradation with the isolated strains under oxic conditions.

Nevertheless, even though an efficient removal of ECs under oxic condition was observed, metabolites were not studied. Indeed, degradation of ECs to metabolites have been observed for CBZ (König et al., 2016), HHCB (Vallecillos et al., 2017) and also for others UV filters (Volpe et al., 2017). These results highlighted the complexity of

ECs removal under experimental conditions as well as in WWTPs, where their efficiency are dependant of both physical, chemical and biological processes but also to identify metabolites in further studies.

3.3. Isolated strains involved in ECs degradation

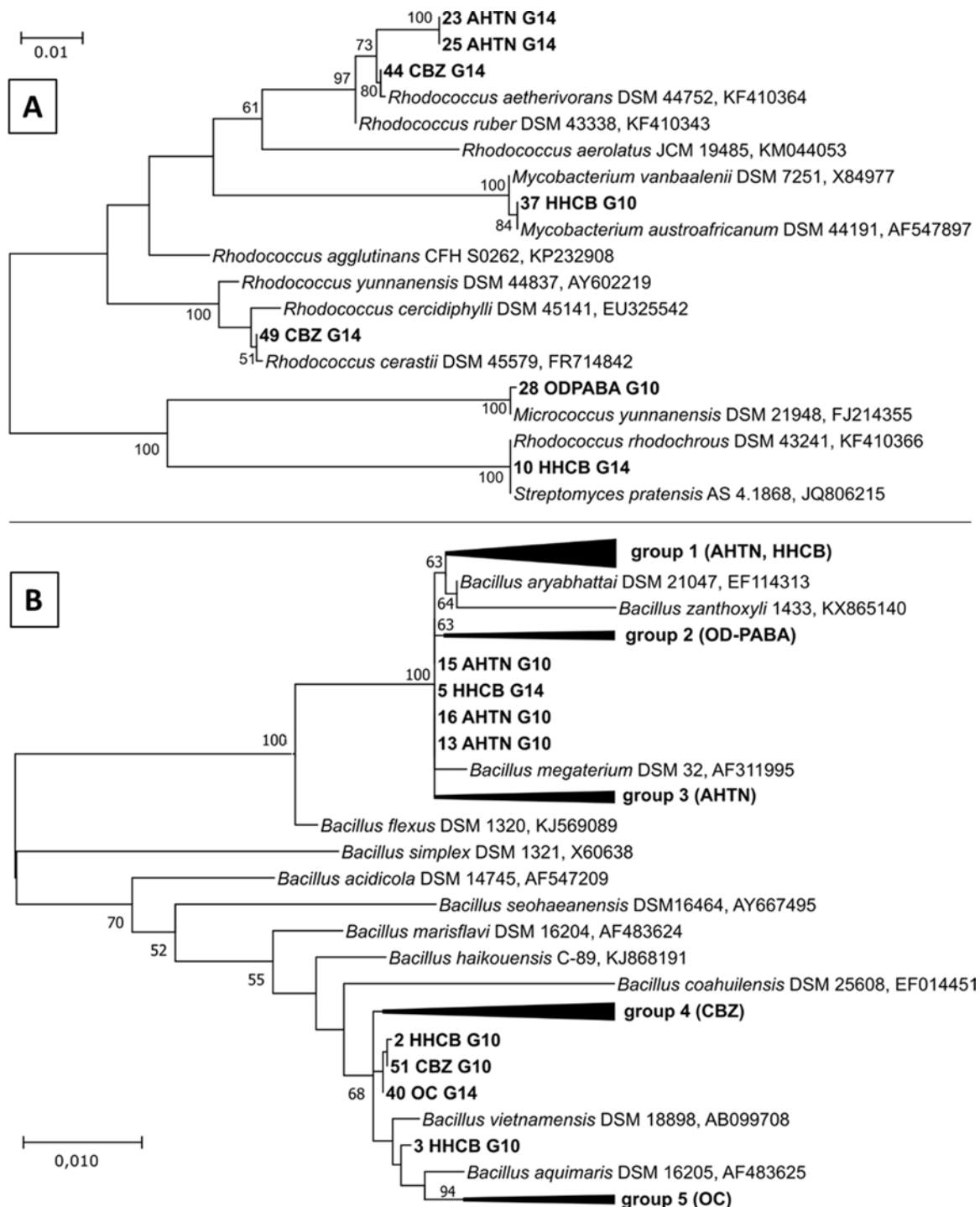


Fig. 5. Phylogenetic trees of strains obtained after the aerobic isolation in solid medium with different ECs (AHTN, HHCB, OC, OD-PABA and CBZ) from sediments collected in the submarine Canyon of Capbreton (stations G10 and G14). Fig.5A and Fig.5B correspond to *Actinobacteria* and *Firmicutes*, respectively. Isolated strains are in bold, reference strains are in italic. The evolutionary history was inferred by using the Maximum Likelihood method and Tamura-Nei model. The bootstrap values are above 50 % are shown next to the branches. The scale indicates nucleotides substitution. Groups 1, 2, 3, 4 and 5 included 10 (20, 21 AHTN-G10; 22, 27 AHTN-G14 and 4, 6, 7, 8, 11, 12 HHCB- G14), 3 (33, 35, 36 ODPABA-G14), 4 (17, 18, 19 AHTN-G10 and 24 AHTN-G14), 6 (45, 46, 47, 48 CBZ-G14 and 50, 52 CBZ-G10) and 3 (41, 42, 43 OC-G10) isolated strains, respectively.

While the degradation of complex organic substances can occur both under anaerobic and aerobic conditions, it is well known that their degradation rates can be higher under aerobic condition (Biel-Maeso et al., 2019; Ying et al., 2007). In accordance with this observation, isolation of strains and degradation test were consequently performed under aerobic condition. Twenty seven strains were isolated with the current method with 6, 8, 4, 4 and 5 strains constrained with HHCB, AHTN, OD-PABA, OC and CBZ, respectively (Supplementary information, Fig. S3). These strains were related to two families, dominated by the *Firmicutes*, followed by the *Actinobacteria* (Fig. 5 A B).

Within the *Actinobacteria*, *Rhodococcus* was the main bacteria genus isolated followed by *Streptomyces*, *Mycobacterium* and *Micrococcus*. Within the *Firmicutes*, only *Bacillus*-related strains were isolated (Fig. 5 A, B). Studies have shown that *Actinobacteria* were involved in degradation of some organic and inorganic compounds. For instance, *Rhodococcus* strains are involved in the removal of pharmaceutical compounds (Ivshina et al., 2018), tetrabromobisphenol A (TBBPA) (Xu et al., 2018) and dominant in naphthalene degrading enrichments (Rhee et al., 2004). It has also been demonstrated that the members of the genus *Streptomyces* produce laccase (i.e. ligninolytic enzymes) that can be involved in the removal of recalcitrant and ECs (Majeau et al., 2010). It has also been demonstrated that members of the genus *Streptomyces* were involved in the CBZ biodegradation (Popa et al., 2014; Popa Ungureanu et al., 2015). Finally, experiments performed from marine sediments demonstrated that *Bacillus thuringiensis* was a novel group participating to the removal of PAHs and pesticides (Ferreira et al., 2016).

Within these isolated strains, only eight exhibited a capacity to degrade contaminants in the conditions used (1 ppm, 5 days, 37 degrees in rich organic medium) with 1, 2, 3 and 2 strains for HHCB, AHTN, OD-PABA and OC, respectively (Table 3). Seven were related to the

Table 3

Removal capacity of isolated strain and their related affiliation based on phylogenetic tree (Fig. 5). % of removal is calculated relatively to the control removal and in triplicate, after 120 h of experimentation at 1 ppm exposition concentration for each emerging contaminants. The strains for which degradation were not determined are not shown (Fig. 5).

Isolated strains	Phyla	Family	% of removal \pm SD
2 HHCB G10	<i>Firmicutes</i>	<i>Bacillus</i>	44 \pm 31
17 AHTN G10	<i>Firmicutes</i>	<i>Bacillus</i>	68 \pm 9
23 AHTN G14	<i>Actinobacteria</i>	<i>Rhodococcus</i>	20 \pm 7
33 ODPABA G14	<i>Firmicutes</i>	<i>Bacillus</i>	34 \pm 19
35 ODPABA G14	<i>Firmicutes</i>	<i>Bacillus</i>	45 \pm 9
36 ODPABA G14	<i>Firmicutes</i>	<i>Bacillus</i>	71 \pm 19
40 OC G14	<i>Firmicutes</i>	<i>Bacillus</i>	17 \pm 17
43 OC G10	<i>Firmicutes</i>	<i>Bacillus</i>	14 \pm 13

genus *Bacillus* and one was related to the genus *Rhodococcus* (23 AHTN G14). While studies have demonstrated high bioremediation capacities by *Actinobacteria*, this study demonstrated high capacity of marine *Bacillus* to degrade ECs. These results suggested that the isolated *Bacillus* were able to remove OD-PABA with a capacity ranging between 34 % \pm 19 % and 71 % \pm 19 % (Table 3). These strains could be involved in the biotic degradation observed in the slurry incubation experiment (Fig. 3). Although CBZ slurry incubations experiments (G10) demonstrated a possible biotic degradation (Fig. 3), isolated strains did not exhibit a remediation for this pollutant under the conditions tested (Supplementary information, Fig. S3). Moreover, the $t_{1/2}$ of CBZ was estimated around 7 months, suggesting that the time should be expanded to observe a remediation capacity. Moreover, it is well known that in nature, the microbial consortia have to be considered to estimate biodegradation pollutant removal process (Wu et al., 2010). Moreover, global microbial communities analysed *in situ*, in G10 and G14 demonstrated a relative abundance of the main phyla similar in both sediments (and similar to the final kinetic time of the slurry incubations, Supplementary information Fig. S2), with a dominance of *Proteobacteria* (relative abundance ranged between 47.9 and 49.4 %), followed by *Bacteroidetes*, *Planctomycetes* and *Acidobacteria* (Fig. 6). Although the *Actinobacteria* and *Firmicutes* were minors *in situ* (Fig. 6), representing less than 1% of the total abundance, phylogenetic trees based the sequences of the isolated strains (Fig. 5 A,B) demonstrated that these isolated strains were related to the *Actinobacteria* and *Firmicutes* families. This suggests a good selection process of these bacterial groups in the presence of these ECs exposition with the isolation method used. In accordance with previous studies, the isolated bacterial strains from the Capbreton Canyon sediments could be good candidate to degrade ECs. Nevertheless, in order to estimate the natural resilience of those ECs, it is crucial also to take account both the biological (e.i. biodegradation, biosorption/adsorption), physical and chemical processes, highlighting further studies are needed to decipher their fate in oceans. It is difficult to extrapolate results based on pure strains to the field. The culture conditions used (medium, temperature, ...) enhance some microorganisms and inhibit others. The role of co-metabolisms in microbial consortia is extremely important as well as the role of the uncultivated prokaryotes. But the objective of this study *in fine* to isolate strains on which we will be able to work on. The isolated strains can be a fantastic tool since they can be used to study the degradation products, to study the isotopic fractionation due to micro-organisms, or to study the genetic determinism of this pathway. These data could then be used as new tools for new environmental studies (compound specific isotopic fractionation, functional genes, ...).

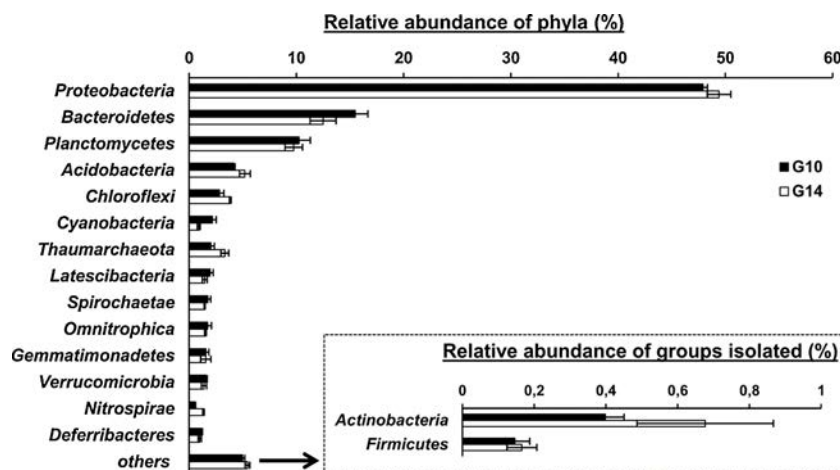


Fig. 6. Relative abundance of prokaryotic 16S rDNA gene sequences (MiSEQ) from sediments of G10 and G14. Data are average percentages (\pm SE). Categories representing $>$ 1% are shown. Relative abundances of phyla from which the isolated strains belonged are also shown.

4. Conclusion

This work characterized the degradation kinetics for ECs (HHCB, OD-PABA and CBZ) from marine sediments under *in situ* conditions (biotic, anaerobic and temperature) thought slurry sediment incubations. The half life times obtained demonstrated that half time live of OD-PABA was lower than HHCB and CBZ, with $t_{1/2}$ ranging from few weeks to 8 months. This underlies that the ECs degradation in marine sediment is a long process and could be a high concern for the environment considering a chronic contamination from the coastal human activities (e.i. WWTPs sewage, agriculture...). Additionally, contrary to CBZ, different half lives were observed between muddy (G10) and sandy (G14) sediments for HHCB and OD-PABA. That indicates ECs degradation depends on the physicochemical properties of both chemicals and sediment.

Moreover, novel synthetic musks and UV filters degrading bacteria were isolated from marine sediment. Based on phylogenetic characteristics, microorganisms were mainly identified as *Bacillus* sp. dominated by *Bacillus megaterium* followed by *Bacillus aquimaris* and *Bacillus vietnamensis*. One other isolated strain which was able to degrade musk was related to *Rhodococcus ruber*. The isolated bacteria here, could have a strong potential to degrade a wide variety of organic pollutants (e.g. priority and emerging), such as synthetic musks and UV filters. Improving the knowledge about the microbial diversity in marine environment may lead to the development of technical tools with degradation abilities and high tolerance in the future but also highlight how microbial remediation contributes to the nature resilience in marine environment.

Author statement

All person who meet authorship criteria are listed as authors, and all authors certify that they have participated sufficiently in the work to take public responsibility for the content, including participation in the concept, design, analysis, writing, or revision of the manuscript. Furthermore, each author certifies that this material or similar material has not been and will not be submitted to or published in any other publication before its appearance in the *Journal of Hazardous Material*

Specific contributions made by each author (list the authors' initials) :

R.G., A.A., designed research; M.M., R.G. acquired funding; A.A., R.G., M.M., C.M., C.G. collected data ; A.A., R.G., C.M., C.G. analyzed data; A.A, R.G. wrote the paper, A.A., R.G., M.M reviewed and edited the paper. All authors have read and agreed with the published version of the manuscript.

Declaration of Competing Interest

The authors declare that they have no conflict of interest.

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Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:<https://doi.org/10.1016/j.jhazmat.2020.123477>.

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