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Inhibitory effects of metal ions on reductive dechlorination of polychlorinated biphenyls and perchloroethene in distinct organohalide-respiring bacteria

Qihong Lu[a,b], Xueqi Zou[a], Jinting Liu[a], Zhiwei Liang[a], Hojae Shim[d], Rongliang Qiu[a,c,*], Shanquan Wang[a,b,c,*]

[a] School of Environmental Science and Engineering, Sun Yat-Sen University, Guangzhou 510275, China
[b] Environmental Microbiome Research Center, Southern Marine Science and Engineering Guangdong Laboratory (Zhuhai), Sun Yat-Sen University, Guangzhou 510275, China
[c] Guangdong Provincial Key Laboratory of Environmental Pollution Control and Remediation Technology, Guangzhou 510275, China
[d] Department of Civil and Environmental Engineering, Faculty of Science and Technology, University of Macau, 999078, Macau

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ABSTRACT

Bioremediation of sites co-contaminated with organohalides and metal pollutants may have unsatisfactory performance, since metal ions can potentially inhibit organohalide respiration. To understand the detailed impact of metals on organohalide respiration, we tested the effects of four metal ions (i.e., Cu^{2+}, Cd^{2+}, Cr^{3+} and Pb^{2+}), as well as their mixtures, on reductive dechlorination of perchloroethene (PCE) and polychlorinated biphenyls (PCBs) in three different cultures, including a pure culture of *Dehalococcoides mccartyi* CG1, a *Dehalococcoides*-containing microcosm and a *Dehalococcoides-Geobacter* coculture. Results showed that the inhibitive impact on organohalide respiration depended on both the type and concentration of metal ions. Interestingly, the metal ions might indirectly inhibit organohalide respiration through affecting non-dechlorinating populations in the *Dehalococcoides*-containing microcosm. Nonetheless, compared to the CG1 pure culture, the *Dehalococcoides*-containing microcosm had higher tolerance to the individual metal ions. In addition, no synergistic inhibition was observed for reductive dechlorination of PCE and PCBs in cultures amended with metal ion mixtures. These results provide insights into the impact of metal ions on organohalide respiration, which may be helpful for future *in situ* bioremediation of organohalide-metal co-contaminated sites.

1. Introduction

Halogenated organic compounds or so-called organohalides are globally widespread, recalcitrant and toxic environmental pollutants, which mainly accumulate in anoxic sediments and soils (Zanaroli et al., 2015). Microbial reductive dehalogenation has been proposed as an effective and inexpensive strategy for *in situ* remediation of organohalide pollution (Nijenhuis and Kuntze, 2016; Wang et al., 2016; Atashgahi et al., 2018). Nonetheless, at contaminated sites, organohalides may co-exist with metal pollutants or other organohalides, which can have inhibitory impact on the microbial reductive dehalogenation (Sandrin and Maier, 2003; Zhang et al., 2012; Arjoon et al., 2013). For instance, electronic waste (e-waste) dumping sites are generally co-contaminated by polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs) and a variety of metals (e.g., Cu, Pb, Cd and Cr), specifically in forms of metal ions (Wuana and Okieimen, 2011; Zhang et al., 2012; Deng et al., 2018; Wu et al., 2019). Consequently, it is imperative to understand how co-existing organohalides and metals affect the microbial reductive dehalogenation.

Different co-contaminates may affect the microbial conversion of a specific pollutant in very different ways (Sandrin and Maier, 2003). Take the co-existence of multiple organohalides for example, inhibitive dehalogenation was generally observed, due to inhibitory effects of co-existing organohalides on the organohalide-respiring bacteria (OHRB) and their syntrophic populations (Kuo and Genthner, 1996; Pardue et al., 1996; Kong, 1998; Chan et al., 2011; Weathers et al., 2015; Mayer-Blackwell et al, 2016; Peng et al., 2019). Mayer-Blackwell et al. (2016) found that *cis*-dichloroethene (cDCE) strongly inhibited dechlorination of 1,2-dichloroethane (1,2-DCA) by changing *Dehalococcoides*-containing microbial community composition. Nonetheless,

* Corresponding authors at: School of Environmental Science and Engineering, Sun Yat-Sen University, Guangzhou 510275, China.
E-mail addresses: eesqrl@mail.sysu.edu.cn (R. Qiu), wangshanquan@mail.sysu.edu.cn (S. Wang).

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exceptions were observed on PCB dechlorination, in which tetrachloroethene (PCE) and polybrominated biphenyls (PBBs) could stimulate, rather than inhibit, the chlorine removal from PCBs (Bedard et al., 1998; Wu et al., 1999; Xu et al., 2019). One possible mechanistic reason for the biostimulation is that the PCB-dechlorinating bacteria employ bifunctional reductive dehalogenases (RDases, e.g. PcbA1, PcbA4 or PcbA5) to catalyze the chlorine removal from both PCBs and PCE (Wang et al., 2014; Chen and He, 2018; Xu et al., 2019). As such, the presence of PCE and PBBs as priming compounds could stimulate the growth of fastidious PCB-dechlorinating bacteria and consequently stimulate PCB dechlorination (Wang et al., 2014; Wu et al., 1999; Xu et al., 2019). In contrast to the extensive studies on the impacts of co-existing organohalides on reductive dehalogenation, effects of metal ions on microbial reductive dehalogenation, specifically the organohalide respiration, remain elusive.

Metal ions have been proven to inhibit microbial processes through different interactions with enzymes involved in biodegradation or general metabolism (Hu et al., 2004; Li and Fang, 2007; Altas, 2009; Oyetibo et al., 2013; Berg, 2014; Roane et al., 2015; Zhu et al., 2019): (1) metal ions may substitute for physiologically essential cations within an enzyme, e.g., Cd^{2+} substitutes for Zn^{2+} (Sandrin and Maier, 2003); (2) metal ions (e.g., Cd^{2+}) are capable of binding to DNA bases to break single-stranded DNA, or binding to sulfhydryl (-SH) groups of enzymes (McMurray and Tainer, 2003; Roane et al., 2015). At the microbial community level, negative impact of metal ions on cytoplasmic biodegradation have been observed in three patterns (Sandrin and Maier, 2003): (1) inhibition increases progressively when increasing the concentration of metal ions (Amor et al., 2001; Gopinath et al., 2011); (2) low concentrations of metal ions stimulate biodegradation, whereas high concentrations are inhibitive (Kuo and Genthner, 1996; Arjoon et al., 2015); (3) low metal concentrations inhibit biodegradation, whereas high concentrations inhibit less (Sandrin et al., 2000). Accordingly, microorganisms have evolved to employ different strategies to resist toxic metals, including metal reduction, metal efflux pump, and formation of metal chelates (Roane et al., 2015). Therefore, due to the varied toxicity and impact of metal ions, biological systems may have different tolerance to these metal ions. For example, Kuo and Genthner (1996) investigated the effects of gradient concentrations (0.01–100 mg/L) of Hg^{2+}, Cu^{2+}, Cd^{2+} and Cr^{6+} on fermentative degradation of benzoate, 3-chlorobenzoate (3-CB), phenol and 2-chlorophenol (2-CP) in anaerobic microcosms. Results showed that 3-CB biodegradation was most sensitive to Cd^{2+} or Cr^{6+} with inhibitive concentrations over than 0.5 mg/L, whereas biodegradation of 2-CP was inhibited most by Cd^{2+} or Cu^{2+}. Notably, these studies were conducted in complex mixed cultures, in which organic compounds (e.g., benzoate, phenol, 3-CB and 2-CP) served as both electron donors and carbon sources. For the microbial reductive dehalogenation mediated by OHRB, organohalides are electron acceptors in their membrane-bound respiratory electron transport chains (Wang et al., 2018). Therefore, compared to above-mentioned biodegradation processes, metals may have different impact on the microbial reductive dehalogenation in OHRB, which still awaits experimental evidence.

In this study, we investigated the impact of gradient concentrations of Cd^{2+}, Cu^{2+}, Cr^{3+}, Pb^{2+} and their mixtures on reductive dechlorination of PCE and PCBs in three different cultures: (1) a pure culture of *Dehalococcoides mccartyi* CG1; (2) a coculture of *D. mccartyi* CG1 and *Geobacter lovleyi* LYY, and (3) a *Dehalococcoides*-containing microcosm established with urban river sediment. In the mixed culture study, changes of microbial community composition upon metal ion addition were analyzed with 16S rRNA gene amplicon sequencing. Knowledges generated in this study might be helpful to *in situ* bioremediation of organohalide-metal co-contaminated sites by providing insights into impact of metal ions on microbial reductive dehalogenation in OHRB.

2. Materials and methods

2.1. Culture setup and cultivation

Two pure cultures, i.e., *Dehalococcoides mccartyi* CG1 (CP006949.1) and *Geobacter lovleyi* LYY (MK850090.1), were used in this study, which were isolated from PCB contaminated soil and black-odorous urban river sediment, respectively. A *Dehalococcoides*-containing sediment sample was collected from black-odorous urban river sediment in Guangzhou, China (23°07′53″N; 113°22′19″E). All cultures were cultivated in defined anaerobic mineral medium as previously described (Wang and He, 2013, Wang et al., 2014). Briefly, 95 mL of bicarbonate-buffered mineral medium with 10 mM acetate (for pure cultures) or lactate (for sediment microcosm) as the carbon source was dispensed into 160 mL serum bottles which sealed with black butyl rubber septa (Geo-Microbial Technologies, Ochelata, OK, U.S.) and secured with aluminum crimp caps. To achieve obligate anaerobic conditions, L-cysteine (24 mg/L) and Na_2S (48 mg/L) were spiked as reducing agents. For pure culture transfer, hydrogen as the electron donor (5×10^4 Pa) was spiked into the described 100 mL medium (with 5% inoculum). On the other hand, lactate was amended into the sediment microcosms as the fermentative organics to produce carbon source (i.e., acetate) and electron donor (i.e., H_2) needed by OHRB. Depending on the experiments, defined concentration of PCE (41.5 mg/L) or PCB180 (1 mg/L) were amended into the serum bottles as the electron acceptor by using HPLC-grade pure solution and 5 g/L isooctane-dissolved stock solution, respectively. Single metal ions (i.e., Cu^{2+}, Cd^{2+}, Cr^{3+}, Pb^{2+}) was added into the three different cultures (i.e., pure culture, sediment microcosms and *Dehalococcoides-Geobacter* coculture) at following concentrations: (1) 1, 5, 10, 20 and 50 mg/L of Cu^{2+}, 1, 2, 5, 10, and 20 mg/L of Cd^{2+}, Cr^{3+} and Pb^{2+} for PCE dechlorination tests; (2) 1, 10 and 50 mg/L Cu^{2+}, 1, 5, 20 mg/L of Cd^{2+}, Cr^{3+} and Pb^{2+} for PCB dechlorination tests (Table 1) by using the following salts: $CuCl_2{\cdot}2H_2O$, $CdCl_2$, $CrCl_3$ and $PbCl_2$. Later, three concentration levels of mixed metals were used to further investigate the effects of metal ion mixture on dechlorination process based on the results from single metal ion experiments: mix-1, 1 mg/L Cu^{2+}, 1 mg/L Cd^{2+}, 1 mg/L Cr^{3+} and 1 mg/L Pb^{2+}; mix-2, 10 mg/L Cu^{2+}, 2 mg/L Cd^{2+}, 5 mg/L Cr^{3+} and 2 mg/L Pb^{2+}; mix-3, 50 mg/L Cu^{2+}, 20 mg/L Cd^{2+}, 20 mg/L Cr^{3+} and 20 mg/L Pb^{2+}. Inoculum with PCE or PCB180 and no metals served as positive controls, while negative controls were amended with target organohalides and metals but without inoculation. The bottles were incubated in the dark at 30 ± 1 °C without shaking. All experiments were set up in triplicate unless otherwise stated.

2.2. Analytical and quantitative methods

Chloroethenes and PCB180 were analyzed as previously described (Wang and He, 2013; Yu et al., 2018). Briefly, headspace samples of chloroethenes were injected manually with a gastight, luer lock syringe (Hamilton, Reno, NV, U.S.) into a gas chromatograph (GC; Agilent 7890B) equipped with a flame ionization detector (FID) and a GS-GasPro column (30 m × 0.32 mm; Agilent J&W Scientific, Folsom, CA, U.S.). The oven temperature was initially held at 80 °C for 0.2 min, increased at 45 °C min^{-1} to 190 °C, and held for 1 min. Injector and detector temperature were 220 and 250 °C, respectively. For PCB180 analysis, 1 mL mixed liquid sample was firstly taken from the homogenized serum bottle and subjected to a liquid–liquid extraction with an equal volume of isooctane. Then, the extracted and centrifuged solvent phase of 0.5 mL sample was transferred to a 2 mL amber glass vial for subsequent GC analysis (Wang and He, 2013). PCB180 was quantified by the same model GC equipped with an electron capture detector (ECD) and an HP-5 capillary column (30 m × 0.32 mm × 0.25 mm film thickness; Agilent J&W Scientific, Folsom, CA, U.S.). The temperature program was initially held at 170 °C for 5 min, increased at 2.5 °C min^{-1} to 260 °C and held for 10 min. Injector and detector

Table 1
Comparison of dechlorination rates (k_1) of PCE and PCB180 in the three cultures amended with gradient concentrations of metal ions.

Metals and Concentration (mg/L)		V = (k_1/k_0)·100 %[a]					
		PCE			PCB180		
		CG1	Microcosm	Co-culture	CG1	Microcosm	Co-culture
Cu^{2+}	0	100	100	100	100	100	100
	1	120	96	96	73	87	94
	2	108	N.A.[b]	98	N.A.	N.A.	N.A.
	10	102	105	100	104	79	93
	20	98	N.A.	94	N.A.	N.A.	N.A.
	50	6	8	5	0	0	0
Cd^{2+}	0	100	100	100	100	100	100
	1	114	93	80	62	116	50
	2	110	71	81	N.A.	N.A.	N.A.
	5	83	73	53	40	62	48
	10	82	70	5	N.A.	N.A.	N.A.
	20	36	69	3	18	45	24
Cr^{3+}	0	100	100	N.A.	100	100	N.A.
	1	99	101	N.A.	107	87	N.A.
	2	89	98	N.A.	N.A.	N.A.	N.A.
	5	85	104	N.A.	107	85	N.A.
	10	61	90	N.A.	N.A.	N.A.	N.A.
	20	40	69	N.A.	92	79	N.A.
Pb^{2+}	0	100	100	N.A.	100	100	N.A.
	1	107	106	N.A.	102	92	N.A.
	2	109	89	N.A.	N.A.	N.A.	N.A.
	5	109	76	N.A.	110	101	N.A.
	10	27	79	N.A.	N.A.	N.A.	N.A.
	20	28	72	N.A.	103	67	N.A.
Mix	0	100	100	N.A.	100	100	N.A.
	L1	102	78	N.A.	81	82	N.A.
	L2	91	44	N.A.	52	85	N.A.
	L3	4	5	N.A.	0	0	N.A.

temperature were set at 250 °C and 300 °C, respectively. Nitrogen was used as the carrier gas as described previously (Wang and He, 2013).

Chloroethenes and PCBs were quantified by using a customized calibration standard. Mole percent values for each organohalides was calculated as described (Bedard et al., 1998; Wang and He, 2013). Dechlorination rate coefficient, k_1, was calculated from plots of the natural logarithm of organohalides concentration versus time according to the integrated first-order rate equation (Said and Lewis, 1991):

$$\mathrm{Ln}\left(\frac{C_t}{C_0}\right) = -kt \quad (1)$$

where C_0 and C_t are the concentration of organohalides at time 0 and t, respectively. Half-lives ($t_{1/2}$) were calculated as follow (Said and Lewis, 1991):

$$t_{1/2} = 0.693/k \quad (2)$$

2.3. DNA extraction, PCR and sequencing

Sediment microcosm samples for genomic DNA (gDNA) extraction and subsequent microbial community analyses were collected at the end of each experiment, and their community gDNA was extracted using the FastDNA Spin Kit for Soil according to the manufacturer's instruction (MP Biomedicals, Carlsbad, CA, United States). The 16S rRNA gene (V4-V5 region) was amplified with the U515F(5′-GTGCCAGCMGCCGCGGTAA-3′) forward primer and U909R (5′- CCCCGYCAATTCMTTTRAGT-3′) reverse primer (Narihiro et al., 2015). Illumina Hiseq sequencing (Illumina, San Diego, CA, United States) service was provided by BGI (Shenzhen, China). The pair-end (2 × 250 nd) demultiplexed sequences were assembled and filtered using Mothur v.1.33 (Schloss et al., 2009), and subsequent processing and downstream analysis was performed with Quantitative Insights Into Microbial Ecology (QIIME, v1.8.0) (Caporaso et al., 2010). The assignment of obtained operational taxonomic units (OTUs) was carried out using the SILVA132 database (https://www.arb-silva.de/). Raw Illumina Miseq sequencing reads were deposited into European Nucleotide Archive (ENA) with accession No. PRJEB33960.

3. Results

3.1. Impact of metal ions on reductive dechlorination of PCE and PCB180 by D. mccartyi CG1

The impact of gradient concentrations of metal ions (i.e., 0–50 mg/L Cu^{2+}, or 0–20 mg/L Cd^{2+}, Cr^{3+}, Pb^{2+}) on organohalide respiration was first tested with the PCE/PCB-dechlorinating pure culture of *D. mccartyi* CG1 (Fig. 1). In PCE-fed cultures amended with the metal ions, PCE was gradually dechlorinated to dichloroethenes (DCEs) via trichloroethene (TCE) within 15 days of incubation, in which the PCE-to-TCE dechlorination was more susceptible to metal ions compared to its subsequent TCE-to-DCEs dechlorination (Fig. S1). The dechlorination kinetics followed the first-order equation (Table 1; Table S1). None of the abiotic controls showed dechlorination activity. Overall, the PCE dechlorination rates decreased progressively with increasing metal ion concentrations in CG1 cultures (Fig. 1; Table 1), in line with their decreasing dechlorination rate constants (k_1) (Table 1) and increasing half-live ($t_{1/2}$) values (Table S1). The PCE dechlorination kinetic data suggested remarkable inhibition in CG1 cultures amended with 50 mg/L Cu^{2+}, 20 mg/L Cd^{2+}, 10 mg/L Cr^{3+}, or 10 mg/L Pb^{2+} (Table 1). Notably, no inhibitive dechlorination was observed in CG1 cultures amended with 0–20 mg/L Cu^{2+} (Fig. 1A) or 0–5 mg/L Pb^{2+} (Fig. 1D). By contrast, Cd^{2+} and Cr^{3+} started to show inhibitory effects at low concentrations, and inhibition increased proportionally with their concentrations.

PCB180 (2345-245-CB), as a major congener in PCB commercial mixture Aroclor 1260 and in weathered PCBs from contaminated sites (Adrian et al., 2009; Payne et al., 2001), was employed to investigate

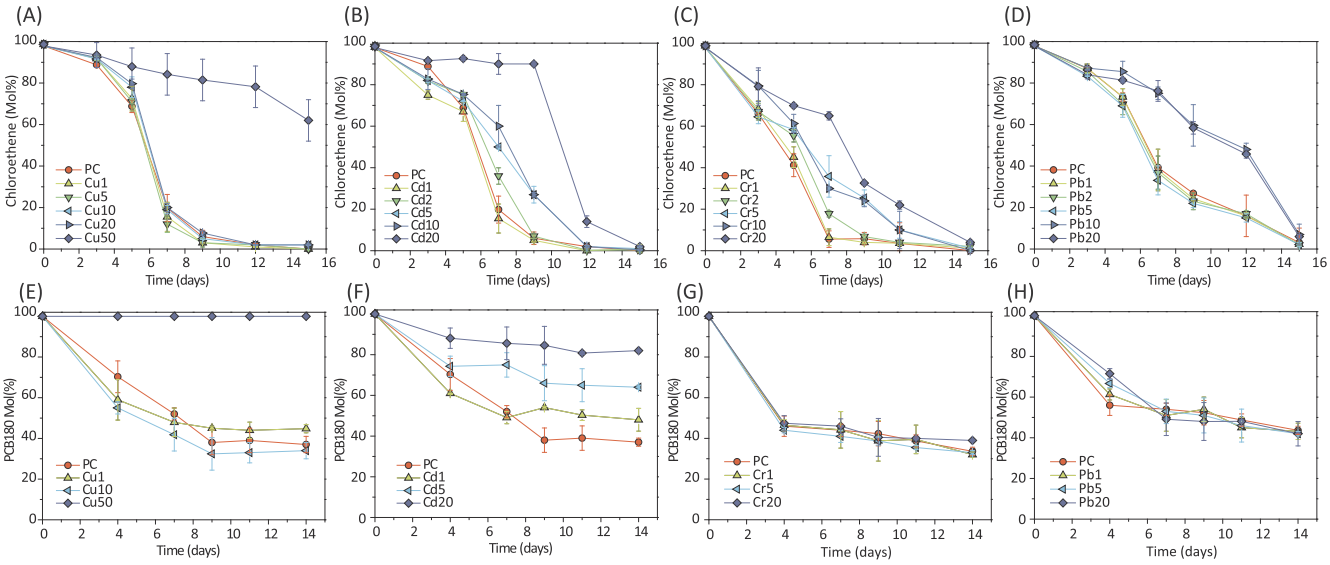

Fig. 1. Effects of metal ions on dechlorination of PCE (A-D) and PCB180 (E-H) by CG1 pure cultures amended with gradient concentrations of Cu^{2+} (A and E), Cd^{2+} (B and F), Cr^{3+} (C and G) or Pb^{2+} (D and H). PC represents biotic controls without metal ion amendment.

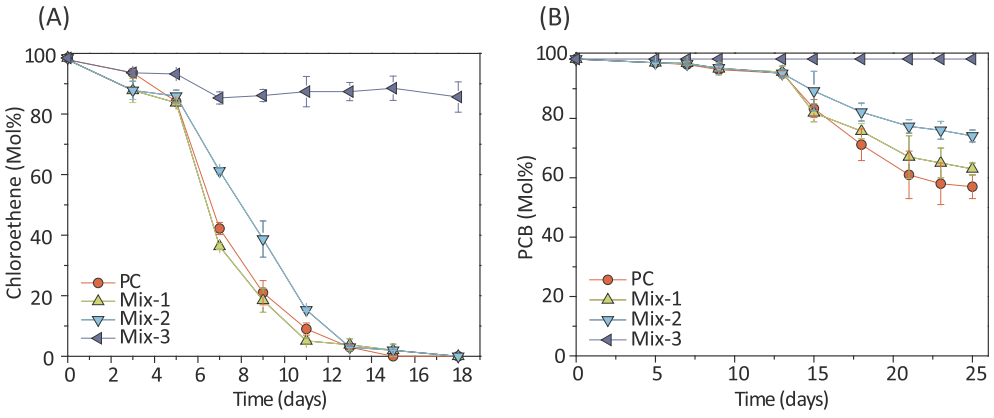

Fig. 2. Effects of metal ion mixtures on dechlorination of PCE (A) and PCB180 (B) by CG1 pure cultures. PC represents biotic controls without metal ion amendment. Mix-1: 1 mg/L Cu^{2+}, 1 mg/L Cd^{2+}, 1 mg/L Cr^{3+} and 1 mg/L Pb^{2+}; Mix-2, 10 mg/L Cu^{2+}, 2 mg/L Cd^{2+}, 5 mg/L Cr^{3+} and 2 mg/L Pb^{2+}; Mix-3, 50 mg/L Cu^{2+}, 20 mg/L Cd^{2+}, 20 mg/L Cr^{3+} and 20 mg/L Pb^{2+}.

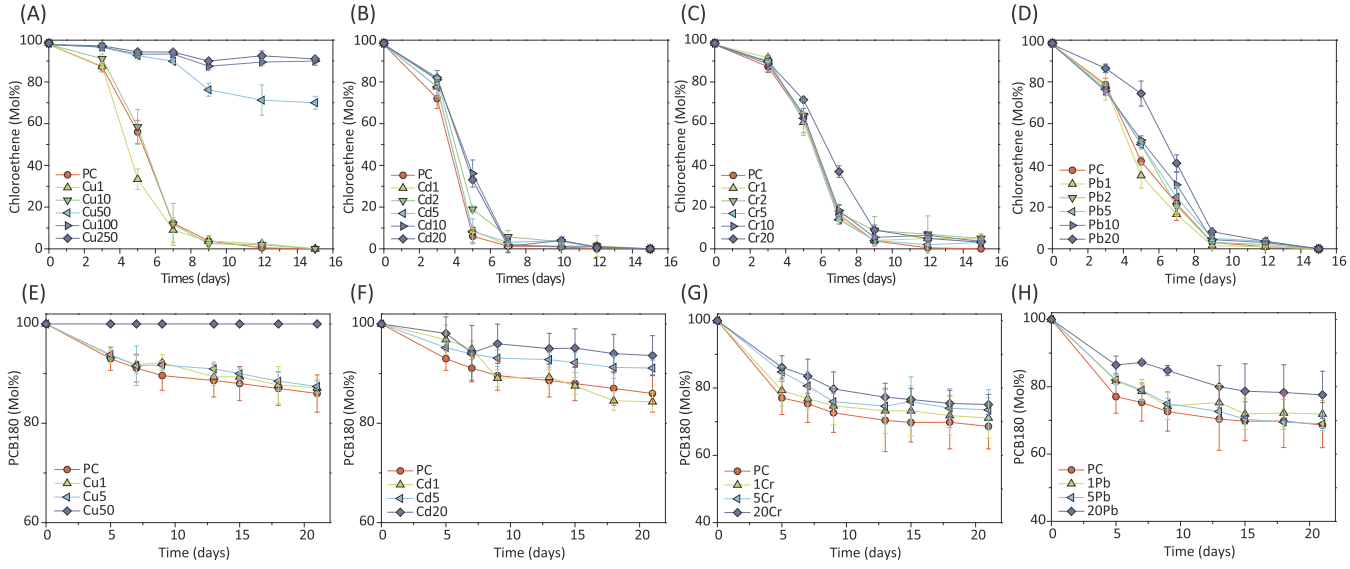

Fig. 3. Effects of metal ions on dechlorination of PCE (A-D) and PCB180 (E-H) by sediment microcosms amended with gradient concentrations of Cu^{2+} (A and E), Cd^{2+} (B and F), Cr^{3+} (C and G) or Pb^{2+} (D and H). PC represents biotic controls without metal ion amendment.

the impact of metal ions on reductive dechlorination of persistent organohalides. Three gradient concentrations of metal ions (i.e., 1, 5 and 20 mg/L for Cd^{2+}, Cr^{3+} and Pb^{2+}; 1, 10 and 50 mg/L for Cu^{2+}) were selected in the PCB dechlorination test based on above-described PCE dechlorination kinetics. In the PCB-fed CG1, PCB180 was partially dechlorinated to PCB153 (245-245-CB) through removing double flanked meta-chlorines within 10 days (Fig. 1 and Fig. S2). Interestingly, compared to PCE dechlorination, the four metal ions affected PCB dechlorination in different ways. For example, no obvious inhibition was observed in PCB-fed CG1 amended with 0–10 mg/L Cu^{2+} (Fig. 1E; Table 1), 0–20 mg/L Cr^{3+}, or 0–20 mg/L Pb^{2+} (Fig. 2G and H; Table 1). These results suggested the comparatively higher tolerance of PCB-fed CG1 to these metal ions, compared to PCE-fed CG1. By contrast, inhibition on PCB dechlorination increased progressively when increasing Cd^{2+} concentrations from 0 to 20 mg/L (Fig. 1F; Table 1). Moreover, increasing concentrations of Cd^{2+} not only affected the PCB dechlorination rate constant (k_1) but the percentage of PCB180 dechlorination (Fig. 1F; Table S2).

At contaminated sites, multiple metal ions might co-exist with each other and have synergistic effects on indigenous microbial metabolic processes (Sandrin and Maier, 2003; Arjoon et al., 2013). In order to simulate this situation, we mixed the four metal ions and prepared three mixtures to investigate their inhibitory effects on PCE and PCB dechlorination in CG1 (Fig. 2). Interestingly, the PCE- and PCB180-dechlorination in the presence of metal ion mixtures were similar to the results obtained in cultures amended with single metal ions, i.e., similar to Cu^{2+} inhibition in PCE-fed CG1 (Fig. 1A; Fig. 2A) and to Cd^{2+} inhibition in PCB-fed CG1 (Fig. 2F; Fig. 3B). Consequently, no synergistic inhibition of metal ion mixtures on reductive dechlorination of PCE and PCB180 was observed in CG1 (Fig. 2; Table 1).

3.2. Impacts of metal ions on dechlorination of PCE and PCB180 by a river sediment microcosm

At contaminated sites, dechlorinating bacteria (e.g., *Dehalococcoides*) need to work closely with other beneficial microorganisms (e.g., *Desulfovibrio* and *Methanosarcina*), which together form a core community for interspecies carbon- and electron-transfers to facilitate organohalide respiration (Chau et al., 2018; Wang et al., 2019). Consequently, compared to dechlorinating pure cultures, impact of metal ions on organohalide respiration in the synergistic dechlorinating communities could be different. To elucidate the detailed effects, sediment microcosms containing *Dehalococcoides* were setup with contaminated urban river sediment for reductive dechlorination of PCE and PCB180 with gradient concentrations of the four metal ions (Fig. 3; Fig. 4). In contrast to PCE-to-DCEs dechlorination in CG1 (Fig. S1), PCE could be completely dechlorinated to non-toxic ethene via TCE, DCEs and vinyl chlorine (VC) in the sediment microcosms (Fig. S3), possibly mediated by multiple OHRB. Compared to CG1 pure cultures, the similar concentrations of metal ions showed less inhibition on PCE dechlorination in sediment microcosms (Fig. 3), suggesting improved metal ion tolerance of the *Dehalococcoides*-containing microcosms. Take microcosms amended with 20 mg/L of Cd^{2+}, Cr^{3+} or Pb^{2+} for example, slight inhibition (with $< 30\%$ k_1 reduction) on PCE dechlorination was observed in these sediment microcosms (Fig. 3B, C and D), compared to their $> 60\%$ k_1 reduction in CG1 pure cultures (Table 1). By contrast, PCE dechlorination in sediment microcosms were shown to be more sensitive to metal ion mixtures compared to CG1 pure cultures (Fig. 4A). For example, PCE dechlorination rate constant (k_1) in the sediment microcosm amended with mix-2 decreased around 55%, compared to $< 10\%$ decrease in CG1 spiked with the same mixture of metal ions (Table 1).

In PCB-fed sediment microcosms, less than 30% of PCB180 was dechlorinated to PCB146 (235–245-CB) in the presence of either single metal ions or metal ion mixtures (Fig. 3; Fig. 4B; Fig. S4), suggesting the presence of different PCB-dechlorinating microorganisms in sediment microcosms rather than the PCB180-to-PCB153 dechlorinating *D. mccartyi* CG1. Different from PCE dechlorination, but similar to PCB dechlorination by CG1, inhibition on PCB180 dechlorination in sediment microcosms became more significant when increasing metal ion concentrations (Fig. 3). Consequently, similar to CG1 pure cultures, PCB dechlorination in sediment microcosms was more sensitive to the co-existing metal ions compared to PCE dechlorination. Notably, of the four tested metal ions, both PCE and PCB dechlorination in the sediment microcosms was most sensitive to Cd^{2+} (Fig. 3; Fig. 4). Accumulation of *cis*-DCE as a major dechlorination intermediate (Fig. S1), as well as the decreased k_1 (Table 1), further corroborated the comparatively higher toxicity of Cd^{2+}.

3.3. Impacts of metal ions on dechlorination of PCE and PCB180 by a Dehalococcoides-Geobacter coculture

Obligate OHRB (e.g., *Dehalococcoides* and *Dehalogenimonas*) and non-obligate OHRB (e.g., *Geobacter* and *Desulfitobacterium*) frequently coexist in organohalide contaminated sites (Wang and He, 2013; Huang et al., 2019). Therefore, we prepared a coculture consisting of *D. mccartyi* CG1 and *G. lovleyi* LYY to investigate the impact of both the least and most toxic metal ions (i.e., Cu^{2+} and Cd^{2+}) on reductive dechlorination, in which the *G. lovleyi* LYY is capable of dechlorinating PCE to *cis*-DCE with acetate as both a carbon source and electron donor (Fig. S2). For PCE dechlorination, *D. mccartyi* CG1 and *G. lovleyi* LYY in the coculture need to compete for the carbon source (i.e., acetate) and electron acceptor (i.e., PCE). In all PCE-fed cocultures, PCE was rapidly dechlorinated to *cis*-DCE without *trans*-DCE production, suggesting the phase-out of *trans*-DCE producing CG1 in PCE dechlorination (Fig. 5A and B; Fig. S5). This phenomenon might be due to the higher PCE dechlorination rate of strain LYY compared to strain CG1. Therefore, impact of Cu^{2+} and Cd^{2+} on PCE dechlorination in the coculture

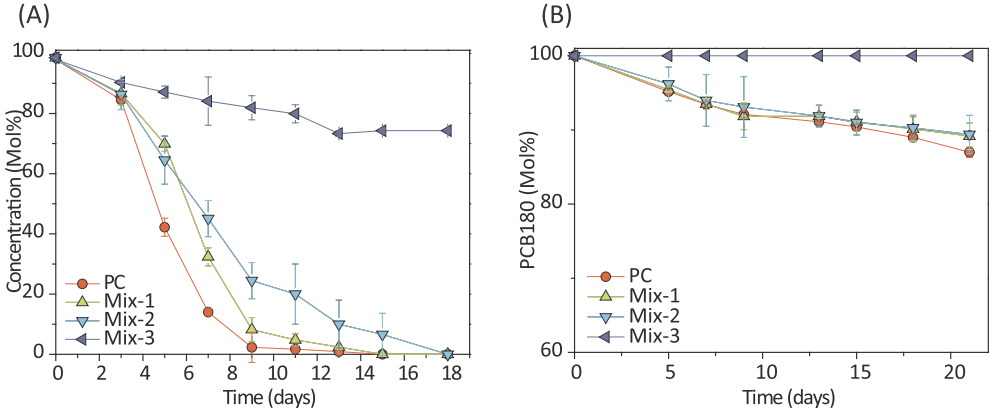

Fig. 4. Effects of metal ion mixtures on dechlorination of PCE (A) and PCB180 (B) by sediment microcosms. PC represents biotic controls without metal ion amendment. Mix-1: 1 mg/L Cu^{2+}, 1 mg/L Cd^{2+}, 1 mg/L Cr^{3+} and 1 mg/L Pb^{2+}; Mix-2, 10 mg/L Cu^{2+}, 2 mg/L Cd^{2+}, 5 mg/L Cr^{3+} and 2 mg/L Pb^{2+}; Mix-3, 50 mg/L Cu^{2+}, 20 mg/L Cd^{2+}, 20 mg/L Cr^{3+} and 20 mg/L Pb^{2+}.

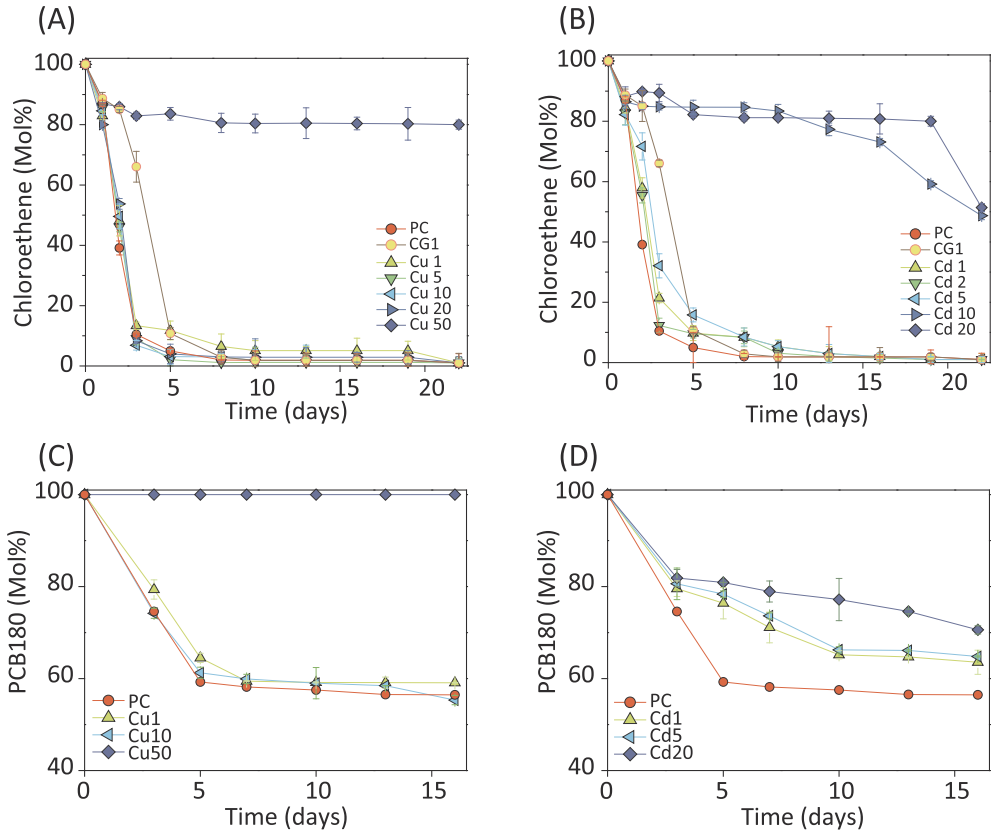

Fig. 5. Effects of metal ions on dechlorination of PCE (A and B) and PCB180 (C and D) by *Dehalococcoides-Geobacter* cocultures amended with gradient concentrations of Cu^{2+} (A and C) or Cd^{2+} (B and D). PC represents biotic controls without metal ion amendment.

represent their impact on strain LYY, which was further verified with experiments by using the *G. lovleyi* LYY pure culture (Fig. S6). Interestingly, inhibition trends of the two metal ions on the coculture were similar to them in CG1 pure cultures with an exception that the coculture was sensitive to 10 and 20 mg/L Cd^{2+} (Fig. 1B and Fig. 5B), suggesting the comparatively higher sensitivity of *Geobacter* to Cd^{2+}. In terms of PCB dechlorination, only CG1 in the coculture can dechlorinate PCB180 (Fig. 5C and D). Consequently, the impacts of metal ions on PCB dechlorination in *Dehalococcoides-Geobacter* cocultures (Fig. 5C and D) showed similar trends with CG1 pure cultures (Fig. 1E and D).

3.4. Microbial community analysis

To evaluate the effects of metal ions on the microbial community profiles of *Dehalococcoides*-containing microcosms, samples were collected from the PCE/PCB-fed cultures amended with gradient concentrations of single metal ions or their mixtures for 16S rRNA gene amplicon sequencing analyses. Previously proposed core microbial community consisting of *Dehalococcoides*, *Desulfovibrio* and *Methanosarcina* for PCB dechlorination (Wang et al., 2019) was observed in all of these PCB- and PCE-dechlorinating microcosms (Fig. 6). Interestingly, both organohalides (as electron acceptors) and metal ions were showed to be primary factors shaping the microbial community structures, but in distinctly different ways (Fig. 6; Fig. 7). For instance, bacteria of *Sphaerochaeta*, Rikenellaceae and *Lentimicrobium* were likely to be dominant fermenters in the PCE-fed microcosms, whereas populations of *Desulfovibrio* and Desulfobacteraceae were enriched in the PCB-fed cultures (Fig. 6). Principal coordinated analysis (PCoA) showed the microbial community clustering based on the metal ion amendment (Fig. 7B and C). Specifically, relatively low abundance of *Methanosarcina* (< 1%) was observed in both PCE- and PCB-fed microcosms, compared to their relatively high abundance (> 10%) in the presence of other metal ions. Moreover, the enrichment of other populations could be helpful to maintain the availability and balance of acetate and H_2 for organohalide respiration of OHRB, e.g., Rikenellaceae and *Lentimicrobium* in PCE-fed cultures, and *Youngiibacter*, Soehngenia and Tyzzerella in PCB-fed cultures (Fig. 6). Cr^{3+} enriched similar fermentative lineages with Pb^{2+}, but had less impact on methanogens (e.g., *Methanosarcina*). Surprisingly, the presence of metal ions did not obviously affect the relative abundance of *Dehalococcoides* in PCE-fed cultures (Fig. 6; Fig. S7), consistent with observations that metal ions had minimal impact on PCE dechlorination in sediment microcosms. Compared to PCE-fed cultures, the PCB-dechlorinating communities were more susceptible to the metal ions (Fig. 3; Fig. S7). Experimental evidences suggested that the higher-taxon diversity in PCE-fed cultures might provide improved resistance to metal ions (Fig. 3; Fig. S7A). Nonetheless, another possible reason for the improved resistance might be that the PCE-dechlorinating OHRBs were more resistant to metal ions, compared to PCB-dechlorinating OHRBs. Notably, the increasing concentrations of Pb^{2+} and Cu^{2+} remarkably lowered the alpha diversity richness of the sediment microcosms (Fig. S7A), resulting in subsequent enrichment of *Dehalococcoides* (Fig. S7B). This observation might be due to the high concentrations of the two metal ions being able to preferentially inhibit non-dechlorinating lineages in the syntrophic community. Therefore, Pb^{2+} and Cu^{2+} might be employed to enrich and isolate OHRB. By contrast, decrease in relative abundance of *Dehalococcoides* was observed in sediment microcosms fed with increased concentrations of Cd^{2+} and Cr^{3+} (Fig. S7).

4. Discussion

Effective remediation of environmental sites co-contaminated with metal pollutants and organohalides requires information on the

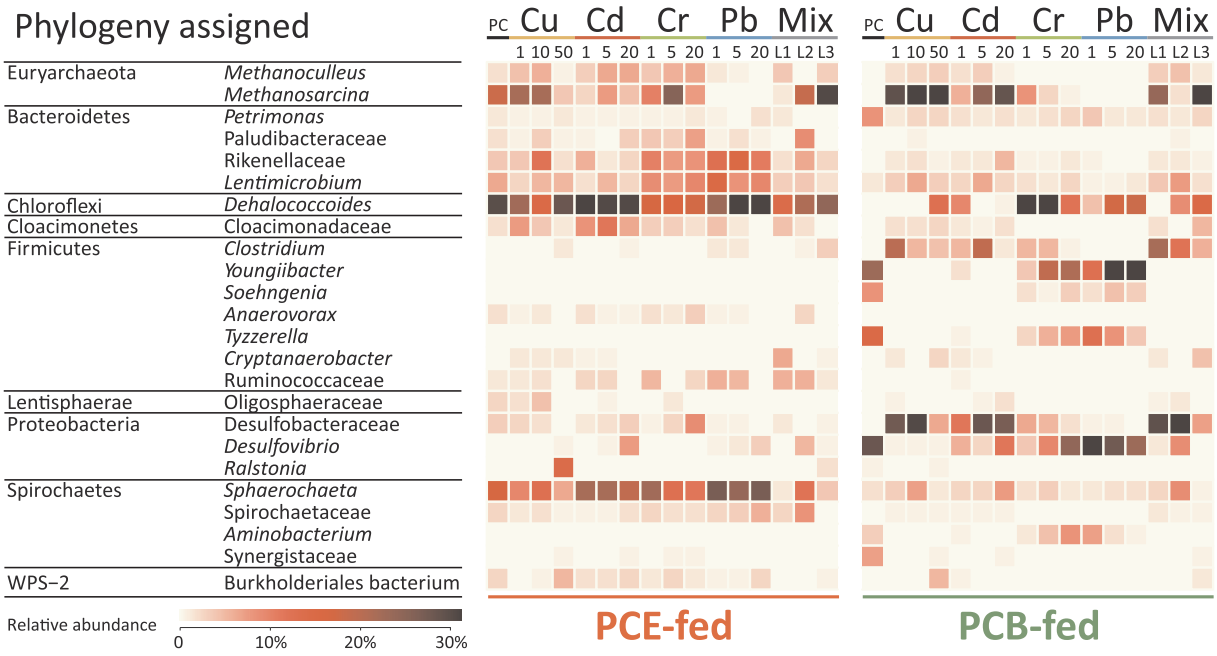

Fig. 6. Microbial community profiling of PCE- and PCB-fed sediment microcosms amended with gradient concentrations of metal ions. The microbial lineages with relative abundance over than 0.5% in at least one culture were shown here.

inhibitory impact of metal ions on OHRB and their mediated dehalogenation processes. Our study revealed how gradient concentrations of four typical metal ions affected PCE and PCB dechlorination by three different cultures, i.e., *D. mccartyi* CG1, *Dehalococcoides*-containing microcosm and *Dehalococcoides-Geobacter* coculture. The experimental evidences suggested that the detailed impact of metal ions on PCE/PCB dechlorination in all of the three organohalide-respiring cultures depended on both the type and concentration of metal ions, consistent with previous observations (Kuo and Genthner, 1996; Kong, 1998, Sandrin and Maier, 2003). Nonetheless, compared to other biodegradation processes reported previously, OHRB seemed to have comparatively higher tolerance to metal ions, i.e., 50, 5, 10 and 10 mg/L for Cu^{2+}, Cd^{2+}, Cr^{3+} and Pb^{2+}, respectively (Fig. S8). For instance, 1 mg/L Cd^{2+} was shown to inhibit the dioxygenase- and monooxygenase-catalyzing degradation of phenanthrene and naphenalene (Malakul et al., 1998; Maslin and Maier, 2000). 10 ug/L Cu^{2+} could significantly reduce phenol degradation activity of *Acinetobacter calcoaceticus* AH (Nakamura and Sawada, 2000), which employed the dioxygenase to catalyze the phenol degradation (Nesvera et al., 2015). These studies had shown that the biodegradation processes were generally inhibited at comparatively lower concentrations of metal ions (Fig. S8). The relative higher metal resistance of OHRBs may benefit the *in situ* bioremediation application of microbial reductive dehalogenation process at sites co-contaminated with organohalides and metal pollutants.

In *Dehalococcoides*-containing communities, *Dehalococcoides* could form a syntrophic metabolic network with other beneficial populations, e.g., *Desulfovibrio* and *Methanosarcina*, for interspecies carbon- and electron-transfers (Men et al., 2012; Men et al., 2014; Wang et al., 2019). Consequently, metal ions inhibiting fermentative and methanogenic microorganisms, as well as acetogenic syntrophs, might indirectly affect organohalide respiration. In this study, PCE dechlorination in sediment microcosms was more sensitive to mixed metal ions compared to CG1 pure cultures, indicating the indirect impact of metal ions on organohalide-respiration through affecting the syntrophic metabolic network. Due to the different toxicities of metal ions, the metal ion mixtures possibly resulted in negative impact on different microbial linages and finally inhibited organohalide-respiration (Kuo and Genthner, 1996). Nonetheless, the *Dehalococcoides*-containing microcosms had higher tolerance to the induvial metal ions, compared to CG1 pure culture. Possible reasons for the improved tolerance in the mixed cultures included: (1) The slow-growing OHRB proceeded organohalide respiration with low concentrations of acetate and H_2, e.g., < 1 nM H_2

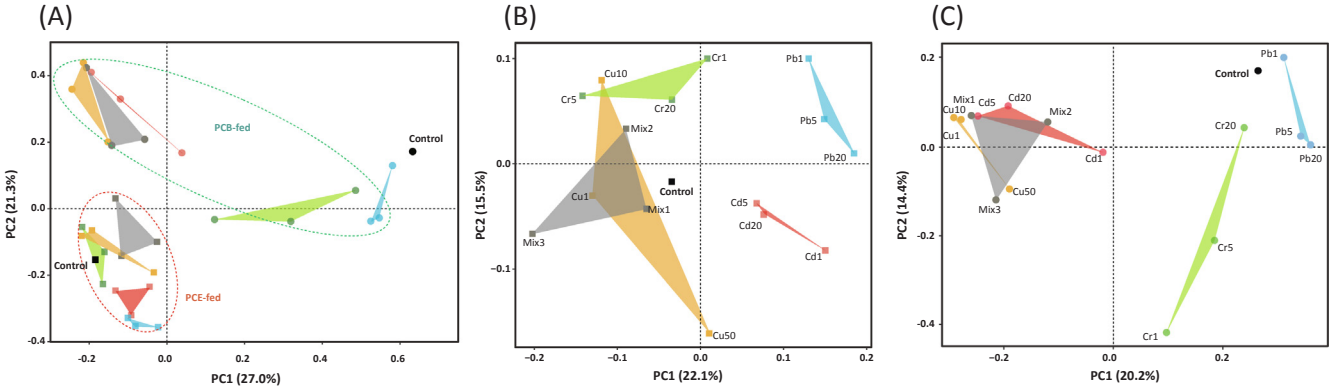

Fig. 7. Principal coordinates analysis (PCoA) of the microbial community composition in PCE- and PCB-fed sediment microcosms amended with gradient concentrations of metal ions: (A) PCE- and PCB-fed microcosms; (B) PCE-fed microcosms; (C) PCB-fed microcosms.

for *Dehalococcoides* (Löffler et al., 1999; Löffler et al., 2013; Dolfing, 2016). Therefore, incomplete inhibition of metal ions on acetate- and hydrogen-producing lineages will not obviously inhibit obligate OHRB. (2) Many anaerobic microorganisms in the sediment microcosms could be resistant to or capable of transforming metals (Nies, 2003; Goulhen et al., 2006), alleviating inhibitory effects of metal ions on organohalide respiration.

Experimental evidences suggested significant difference in tolerance of organohalide respiration to different metal ions, e.g., the higher tolerance to Cu^{2+} compared to the other three metal ions (nonessential metals for biological functions) (Hughes and Poole, 1989; Bruins et al., 2000; Roane et al., 2015). These different inhibitory effects might be due to their distinct interactions with microbial cells and cellular macromolecules (Nies, 1999; Roane et al., 2015). For example, toxic effects of the well-characterized Cd^{2+} included the function of thiol-binding, protein denaturation, interaction with calcium/zinc metabolism, membrane damage and loss of a protective function (Nies, 1999; Roane et al., 2015). The inhibitive concentration of Cd^{2+} on trichloroaniline degradation could be as low as 0.01 mg/L in a mineral-dominated soil (Pardue et al., 1996). Also, the order of metal inhibition for the anaerobic transformation of 2-/3-chlorophenols was identified to be $Cd^{2+} > Cr^{3+} \geq Cu^{2+}$ (Kong, 1998), in line with data obtained in our study. In addition, Pb^{2+} could deplete glutathione and protein-bound sulfhydryl groups, resulting in reactive oxygen species as superoxide ions to enhance lipid peroxidation, DNA damage and sulfhydryl homeostasis (Arjoon et al., 2013). For example, 2.8 and 1.4 mg/L Pb^{2+} could significantly inhibit degradation of crude oil by *Pseudomonas* and *Micrococcus*, respectively (Benka-Coker and Ekundayo, 1998). By contrast, Cr^{3+} was usually considered to be less toxic compare to Cr^{6+} which presented as an oxidizing agent (Kotas and Stasicka, 2000). Furthermore, Cr^{3+} caused adverse effects on microorganisms through the inhibition of a metallo-enzyme system (Kotas and Stasicka, 2000). For example, Said and Lewis (1991) reported that 13 mg/L Cr^{3+} started to inhibit biodegradation of 2,4-dichloro-phenoxyacetic, consistent with observations in our study. The different impact of metal ions on microorganisms were further corroborated by our subsequent microbial community analyses, which changed the microbial community composition in distinct ways. In addition, both obligate and non-obligate OHRB might have different sensitivities to metal ions due to their different cellular structure and metabolism (Roane et al., 2015). Moreover, notably, synergistic inhibition was barely observed when metal ion mixtures were introduced to dehalogenation microcosms, that is different from previously studies that mixed metal ions generally exerted synergistic inhibitions (Volland et al., 2014; Umoren and Solomon, 2017).

In all, our study suggested: (1) organohalide respiration had relatively high tolerance to metal ions, among which Cd^{2+} was identified to be the most toxic metal ion; (2) metal ion mixtures have no synergistic inhibition on microbial reductive dechlorination of PCE and PCBs; (3) both organohalides and metal ions could shape microbial community structures; (4) metal ions may have indirect inhibition on organohalide respiration through affecting non-dechlorinating populations of the synergistic dechlorination communities; (5) Pb^{2+} and Cu^{2+} might be employed to enrich OHRB.

CRediT authorship contribution statement

Qihong Lu: Conceptualization, Methodology, Software, Investigation, Visualization. **Xueqi Zou:** Investigation, Validation. **Jinting Liu:** Investigation. **Zhiwei Liang:** Formal analysis, Data curation. **Hojae Shim:** . **Rongliang Qiu:** Supervision, Project administration, Funding acquisition. **Shanquan Wang:** Conceptualization, Resources, Supervision, Visualization, Funding acquisition.

Declaration of Competing Interest

We declare that we have no conflict of interest.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2019.105373.

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