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Distributions of chlorinated paraffins and the effects on soil microbial community structure in a production plant brownfield site☆

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ABSTRACT

The distributions of chlorinated paraffins (CPs) in soils and their ecological effects attract much attention, while site-scale data are still scarce. In this study, a comprehensive investigation was performed to understand the CP distributions at a CP production plant brownfield site, as well as their effects on soil microbial community. Short-, medium- and long-chain CPs (SCCPs, MCCPs, LCCPs) were detected in most samples with total contents ranging ND-5,090, ND-6,670, and ND-1450 ng g^{-1} (dw), respectively. A CP-hotspot was observed 10 m beneath the synthesis workshop, indicating the downward migration of CPs. The consistence of soil SCCP congener profiles with commercial product CP-52 suggested the leakage of CP products as the contamination source. Besides CPs, petroleum hydrocarbons (PHC) contamination also occurred beneath the synthesis workshop. Soil microbial community composition and diversity were significantly influenced by SCCPs ($p < 0.05$) despite their lower contents compared to other concerned contaminants. Microbial network analysis indicated nonrandom co-occurrence patterns, with *Acinetobacter*, *Brevibacterium*, *Corynebacterium*, *Microbacterium*, *Stenotrophomonas*, and *Variibacter* as the keystone genera. Genera from the same module showed significant ecological links ($p < 0.05$) and were involved in the degradation of PHCs and chlorinated organic contaminants. This study provides the first phylogenetic look at the microbial communities in CP contaminated soils, indicating that the long-term exposure to CPs and PHCs may lead to microbial group assemblages with the potential for degradation.

1. Introduction

Chlorinated paraffins (CPs) have been widely used as metal-working fluids and additives for polymeric materials in the industry (van Mourik et al., 2016). They are semi-volatile and hydrophobic mixtures of polychlorinated *n*-alkanes (Sijm and Sinnige, 1995). According to carbon chain length, CPs can be classified as short-chain CPs (SCCPs, C_{10-13}), medium-chain CPs (MCCPs, C_{14-17}), and long-chain CPs (LCCPs, $C_{>18}$), with chlorine contents ranging 10–72% (Bayen et al., 2006; Pellizzato et al., 2007). In industry, MCCPs and LCCPs are typically the desired constituents in commercial formulae. CP-42 and CP-52, for example, are products containing an average chlorine content of 42% and 52%, respectively, regardless of the congener profiles. However, CPs are directly synthesized via the free radical chlorination of *n*-alkane (paraffin) feedstock from petroleum distillation using molecular chlorine (Fiedler, 2010). Thus, commercial CPs usually contain SCCPs in an amount that depends on the constituents of the feedstock.

Among the CP categories, SCCPs have attracted the most attention due to their persistence, potential for long-distance transportation, carcinogenicity, and bioaccumulation (Wei et al., 2016). SCCPs are under review by the Stockholm Convention as persistent organic pollutants (POPs) and were officially listed in Annex A at the 8th Conference of Parties (UNEP, 2017). Besides, as the properties of MCCPs are similar to SCCPs, concerns on this species also attract attention (Wang et al., 2019a).

CPs among all categories are inevitably released to the

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environment during their life cycle, such as during the production, storage, transport, usage, and disposal stages. In recent decades, SCCPs and MCCPs have been found ubiquitously in the global environment (Diefenbacher et al., 2015; Wei et al., 2016; Jiang et al., 2017). Significant attention has been paid to soils and sediments as CPs are hydrophobic and tend to deposit in soils/sediments (UK Environment Agency, 2007). In the life cycle of CPs, the production step is regarded as a significant source of SCCPs in the environment (European Chemicals Board, 2002). For example, Xu et al. (2016) and Wang et al. (2018a,b) have reported the dispersions of SCCPs and MCCPs in soils from CP production plants. In addition, these contaminants have also been found in the sewage sludges, sediments from industrial areas, urban soils, and farmland soils irrigated with wastewater (Chen et al., 2011; Zeng et al., 2011; Gao et al., 2012; Wang et al., 2019a).

Despite the various studies on CP occurrence and dispersions, only a few of them focused on the vertical distributions, while CPs are of high-density and supposed to migrate downward in soil/sediment matrix (Chen et al., 2011; Zeng et al., 2011). The downward migration of contaminants is an important process in the environment that may influence the dispersion patterns in the total environment and especially the groundwater quality. However, understandings on CP distributions and migrations in soils are still limited.

Aside from CPs, a typical target contaminant in the production site is petroleum hydrocarbons (PHCs), which are the raw materials of CP synthesis and may also release to the environment via the same process as CPs during production. The co-contamination of both CPs and PHCs may be an important characterization of CP production site, which may lead to the indifferent environmental behaviors and ecological effects of both contaminants. Yet so far, the occurrence of co-contamination has not been evidenced.

Compared to the investigations on CP distributions in the environment, even fewer studies try to understand the ecological effects of these contaminants. So far, only a limited number of studies have investigated the toxicity of SCCPs to microbial communities, most of which are at laboratorial scale (Bezchlebová et al., 2007). To investigate the ecological effects of contaminants, the compositions and diversities of microbial communities are reliable indicators. They are sufficiently sensitive to integrative information regarding environmental change (Wu et al., 2017). Previous studies revealed that soil microbial communities shift due to contaminations (Liu et al., 2015; Wu et al., 2017). Typically, contamination could result in a decrease in microbial diversity and enrichment of tolerant species via the environmental filtering process, which, in turn, may affect the functioning of the overall ecosystem (Liu et al., 2015; Jiao et al., 2016). However, there are important gaps in the understanding about how the microbial communities respond to the CPs, especially to the POP-like SCCPs. This is an important issue to address not only because it can provide information on the ecological effect of CPs, but also provide theoretical support for the *in situ* remediation in such contaminated brownfield sites.

To understand the CP dispersions and migration in soils due to CP production, as well as their ecological effects, this study investigated the spatial and vertical distribution of CPs in soils from a production plant brownfield site. PHCs were also studied for comparisons. Special focus was paid to POP-like SCCPs. Furthermore, the composition and diversity of the soil microbial communities were studied to elucidate the effects on soil ecological function. We conducted the investigations to verify the following hypotheses: (1) the downward migration tends to occur with CPs with shorter carbon chain length and less chlorine; (2) the POP-like SCCPs may be an important factor to the soil microbial communities, compared to other CPs and PHCs; (3) the exposure to CPs and PHCs may lead to a self-purification function of soil microbial community based on co-metabolism. To the best of our knowledge, this study is the first to investigate the distribution of SCCPs, MCCPs, LCCPs and PHCs in soils at a CP production plant brownfield site together with the resulting effects on microbial community composition and diversity.

2. Materials and methods

2.1. Sample collection

To investigate the CP dispersions and their effects on microbial communities, we took a CP production plant brownfield site in China as an example. The investigated CP production plant brownfield site is located in Guangzhou City, south China. The plant was established in 1995 and operated for more than 20 years. Since its establishment, the plant has produced CP-42 and CP-52 in a set of reaction facilities. The annual production was approximately 15,000 tons. Commercial products were mainly supplied to plastic manufacturing industries, including plastic track materials and polyvinylchloride (PVC). Soil samples were collected in June 2018, six months after the complete shutdown of the plant. In total, seven soil core columns were collected at the site. Soils were covered by a hardened layer of 50 cm thickness. Fig. 1 shows the locations of the sampling sites. Reference soils (ck1 and ck2) were sampled to compare the influences on microbial communities by contaminations. They were surface soils (0–50 cm) collected in the northwestern and eastern forest lands outside the brownfield site, and might not be influenced by the contamination. As revealed by previous studies that contaminants enrich or migrate in distinct soil layers due to varied soil textures (Zhang et al., 2014), the sampling depth in each core was decided based on the geological profile of each soil layer (Fig. S1, Supporting Information (SI)) according to the technical guidelines for environmental site monitoring (HJ25.2–2014) established by the Chinese Ministry of Environment and Ecology (2014). In total, 33 soil samples were collected from 7 soil cores using a Geoprobe® system. A deepest soil sample was collected 10 m beneath the synthesis workshop. The collected soil samples were freeze-dried and stored at −80 °C before analysis.

2.2. Materials

Standard SCCP (51.5% Cl, 55.5% Cl, and 63% Cl), MCCP (42% Cl, 52% Cl, and 57% Cl), and LCCP (36% Cl and 49% Cl) mixtures were purchased from Dr. Ehrenstorfer (Germany). $^{13}C_{10}$-anti-Dechlorane Plus was purchased from Cambridge Isotop Laboratory Inc. (USA). Pesticide residue-grade methanol, dichloromethane, *n*-hexane, and acetonitrile were purchased from Merck (Germany). Double-distilled water prepared by a Milli-Q Synthesis water purification system (Bedford, USA) was used for all experiments. Granular anhydrous sodium sulfate and aluminum oxide (200–300 mesh) were purchased from Anpel Chemical Reagent Co., Ltd. (Shanghai, China).

2.3. CP detection in soil samples

After spiking with an aliquot of the internal standard ($^{13}C_{10}$-anti-Dechlorane Plus), soil samples were ultrasonically treated in dichloromethane for 30 min. The extracts were then concentrated and re-extracted by *n*-hexane twice. The collected extracts were concentrated again and subjected to cleanup using multi-layer silica gel and an aluminum column. Then, the collected eluents were evaporated under a mild nitrogen flow and re-dissolved by acetonitrile before detection.

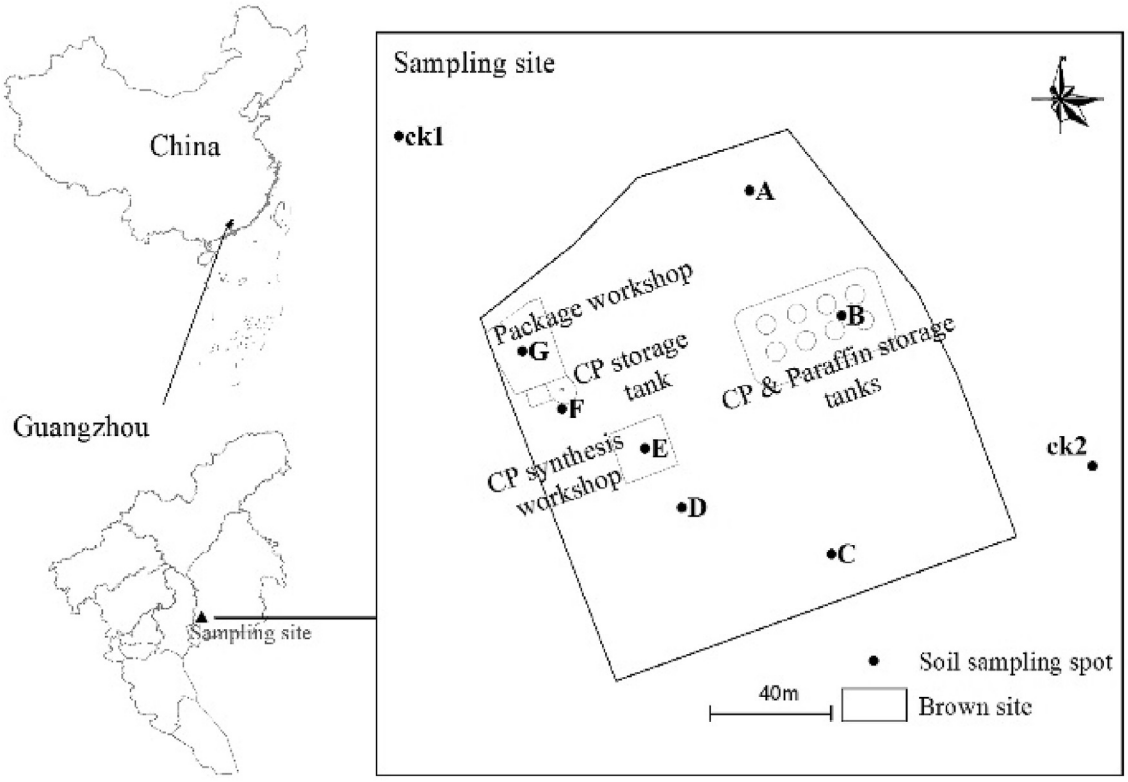

Fig. 1. The location of the studied brownfield site and sampling spots. In particular, core E is located in the middle of the former CP synthesis workshop while cores B and F are located adjacent to the former CP storage tank.

The detection of SCCPs, MCCPs, and LCCPs was conducted using high performance liquid chromatography coupled with an electron spray ionization quadrupole time-of-flight mass spectrometer (HPLC-ESI-QTOF/MS, Agilent1290-6540, USA) following the method reported by Zhou et al. (2019), which was developed based on Li et al. (2017) and Cariou et al. (2016). The method detection limit was 0.1 mg/kg (dry weight, dw) for the SCCPs, MCCPs, and LCCPs in soils. Details for the instrumental parameters and quantification are described in the SI.

For quality assurance and quality control, triplicate analysis was conducted for each of the ten samples. Procedural and solvent blanks were used for each ten samples. CPs were not detected in any of the blanks. A small amount of CPs was spiked in ten random samples to calculate the recoveries, which were 88.9%, 92.9%, and 86.9% for the SCCPs, MCCPs, and LCCPs, respectively. The relative standard deviations for the quantification of the total SCCPs (ΣSCCPs), total MCCPs (ΣMCCPs), and total LCCPs (ΣLCCPs) were <15%.

2.4. PHC detection in soil samples

The contents of petroleum hydrocarbons (PHCs) in the range from C_{10}–C_{40} (referred to as PHCs(sv)) and those in the range from C_6–C_9 (referred to as PHCs(v)) were determined by gas chromatography following the methods reported in ISO 16703:2004 and EPA 8015D, respectively. The analysis of other parameters, including soil physico-chemical parameters and metal concentrations, are described in the SI.

2.5. Characterizations of microbial community structure

In this study, the microbial community composition, diversity and co-occurrence networks of taxa were analyzed to characterize the soil microbial community structures from the brownfield site. In consideration of both the contamination levels and the soil properties (*i.e.* soil type, moisture, depth, pH, etc.), we characterized the microbial communities of 21 soil samples (CK1, CK2, A1, A2, A3, A4, B1, B3, B5, C1, C2, C3, C4, D1, E1, E2, E3, E4, F4, G1, G2, G3, G4). Their properties and contamination levels are shown in Table S1.

A Fast DNA Spin Kit (MOBIO, USA) was used to extract DNA from 0.4 g soil sample. Each DNA sample was amplified in triplicate, using the 515F/806R primer set with a sample-specific 12-bp barcode sequencing on 806R. The amplicons for each sample were pooled for purification using QIA quick Gel Extraction Kit (Qiagen, Chatsworth, CA). A single composite sample was prepared by combining approximately equimolar amounts of purified PCR products from each sample and then sequenced via Illuminate MiSeq sequencing.

Raw pyrosequencing data were processed with the pipeline coupling Mothur (Kozich et al., 2013) and QIIME (Caporaso et al., 2010) software. The raw paired-end sequences were first trimmed using a sickle software to remove any low-quality reads with a q-value < 20 at the 5′ and/or 3’ end. The Flash software was then used to combine the trimmed paired-end reads. The qualified sequences obtained with QIIME were clustered into operational taxonomic units (OTUs) at a 97% similarity level and representative sequences were selected after removing putative chimeras (Edgar, 2010). The taxonomy of the qualified sequence reads was assigned at an 80% identity threshold with the Ribosomal Database Project (RDP) Classifier (version 2.6) (Wang et al., 2007).

The relative abundance of an individual taxon in the community was estimated by comparing the number of sequences assigned to a specific taxon with the number of total sequences obtained. The

alpha diversity (*i.e.* Faith' PD, Chao1, and Shannon) were measured based on a subset of randomly selected sequences from each sample. The relationship between microbial community structure and the selected environmental variables was analyzed using the redundancy analysis (RDA), while RDA with variance partitioning was used to determine the contributions of these selected variables to the variation in community composition. These methods were proved to be reliable by previous studies (Wu et al., 2017; Wu et al., 2019).

Microbial association network was used to explore the co-occurrence pattern of bacterial genera (those with relative abundance > 1% were selected for analysis). A Spearman's correlation between two genera was considered statistically robust when the correlation coefficient (r) was >0.6 ($P < 0.05$). All robust correlations were identified by pairwise comparison of the genera abundance form a correlation network. Each node in the network represents an individual genus and each edge indicates a strong and significant correlation between the nodes ($r > 0.6$, $p < 0.05$). To describe the topology of the resulting networks, measures including the number of nodes and edges, network diameter, average degree, graph density, average path length, clustering coefficient and modularity, were calculated using igraph (Csardi and Nepusz, 2006) packages in R environment. The networks were visualized using Cytoscape (v3.4.0) (Shannon et al., 2003). Ten-thousand Erdöse-Réyni random networks were generated for comparison with the topology of the real network, where each edge has the same probability of assignment to any node (Erdöse and Réyni, 1960).

2.6. Data analysis

Data were processed using ArcGIS version 10.3 (Esri, USA), AutoCAD 2017 (Autodesk, USA), OriginPro 9.1 (OriginLab, USA), Microsoft Excel 2017 (Microsoft, USA), SPSS 18.0 for Windows (SPSS, USA), CANOCO software 4.5 (Microcomputer Power, USA), and R packages (http://www.r-project.org), unless specified elsewhere. The spatial and vertical distributions of SCCPs were visualized by using Earth Volumetric Studio version 2019.6. The correlations among the concerned variables were analyzed by the Spearman's correlation (two-tailed test) using statistical package SPSS 18.0, as well as the stepwise multiple regression analyses. Correlations with $p < 0.05$ were considered significant.

3. Results and discussion

3.1. Spatial and vertical distribution of CPs and PHCs

Contents of CPs and PHCs in each soil sample are presented in Table S1. No CP was detected in the reference soils. SCCPs, MCCPs and PHCs were detected in most of the soil samples collected from the brownfield site while LCCPs were predominantly detected in cores E, F, and G. The smaller distribution area of LCCPs suggests their weaker migration capacity. The concentrations of ΣSCCPs, ΣMCCPs, ΣLCCPs, PHCs(v), and PHCs(sv) ranged from ND-5090 ng g^{-1} (dw), ND-6670 ng g^{-1} (dw), ND-1450 ng g^{-1} (dw), ND-400 ng g^{-1} (dw), and 500-1.2 × 10^6 ng g^{-1} (dw), respectively. Contents of ΣMCCPs were higher than those of ΣSCCPs and ΣLCCPs. This is possibly due to the higher emission volume and greater persistence of MCCPs (main product) compared with SCCPs (byproduct) (Xu et al., 2016). LCCPs, on the other hand, are large molecules that could not easily permeate the hardened layer and, thus, a reduced amount of LCCPs was detected in the soil samples. Along with the CP contamination, PHC(sv) contamination occurred beneath the CP synthesis workshop. The co-occurrence of PHCs suggests that CP synthesis induces co-contamination of PHCs and CPs.

Spatially, a hotspot distribution pattern was observed for both species of contaminants (Fig. 2 and Table S1), i.e., the average concentrations of PHCs and ΣCPs in samples from core E (former CP synthesis workshop) were the highest, followed by those noted for core F (former CP storage tank). These results demonstrate that CP production and storage induced soil contamination of SCCPs and MCCPs. So far, no study has revealed soil contamination due to CP production except for reports by Xu et al. (2016) and Wang et al. (2018a,b). They reported concentrations as high as 554 μg g^{-1} (dw) and 2074.8 ng g^{-1} (dw) for ΣSCCPs and ΣMCCPs, respectively, in surface soils collected from other CP production plants (Xu et al., 2016; Wang et al., 2018a,b). The contamination levels in this study are comparable to these two previous studies and are much higher than those reported for soils sampled from areas in Switzerland (Bogdal et al., 2017), Shanghai (Wang et al., 2014), and Beijing (Zeng et al., 2011). Our results indicate that CP production is an important source of CPs and PHCs in soil environment (see Fig. 3).

Despite the co-occurrence, the correlations among the concentrations of CPs and PHCs suggested their different behaviors in soil environment. On the other hand, SCCPs, MCCPs and LCCPs were similar in environmental behaviors. As shown in Table S2, a significant correlation (Spearman's $r = 0.827$; $p < 0.01$) was observed between the concentrations of ΣSCCPs and ΣMCCPs for all the samples, which agrees with the results reported by Xu et al. (2016). Meanwhile, a lower correlation (Spearman's $r = 0.677$; $p < 0.01$) was observed between the concentrations of ΣSCCPs and ΣLCCPs, as well as between the concentrations of ΣMCCPs and ΣLCCPs (Spearman's $r = 0.656$; $p < 0.01$). These correlations suggest that all CPs originated from the same source while SCCPs and MCCPs were more similar in terms of their environmental fate and migration patterns. However, the concentrations of PHCs were insignificantly related to any species of CPs, which may be due to the different properties of the contaminants, especially the densities.

As for the vertical distributions of contaminants, different cores presented significantly different patterns. In the synthesis workshop, storage tank, and their surroundings (cores B, C, E, and F), the CP concentrations were not the highest in the topsoil but, rather, in the layers below a depth of 4 m. The highest concentrations of ΣSCCPs and ΣLCCPs were both found in sample E5, which was 10 m beneath the synthesis facilities, where the concentration of ΣMCCPs was the second highest among the collected samples (Table S1). In other areas (cores A, D, and G), the CP concentrations were higher in the topsoils. These varying vertical distribution patterns suggest that the CPs had been released to the soils for an extended period due to synthesis and storage, and further migrated to deeper soil layers due to gravity and groundwater motion. Besides the varying vertical distribution patterns, the correlations between CP contents and the content of soil organic matter (SOM) were insignificant except for LCCPs (Spearman's $r = 0.620$, $p < 0.01$; Table S2). This result is inconsistent with the previous studies (Chen et al., 2011; Zeng et al., 2011) reporting that there were significant correlations between CP contents and total organic carbon (TOC) contents in soils and sediments, and TOC might be an important factor influencing the distribution of CPs. This inconsistence could be attributed to the high contents of CPs at the brownfield site compared to the previous studies. As known, commercial CP mixtures are viscous oily liquid. They form disconnected immiscible droplets/ganglia or even dense non-aqueous phase liquids (DNAPLs) at elevated contents in aquifers. In this case, their mixture in soils tend to migrate downward due to gravity, regardless of the soil properties including SOM contents. The highest content of total CPs (ΣSCCPs+ΣMCCPs+ΣLCCPs) detected in this study was as high as 11,810 ng g^{-1} (dw), which is much higher than those reported by

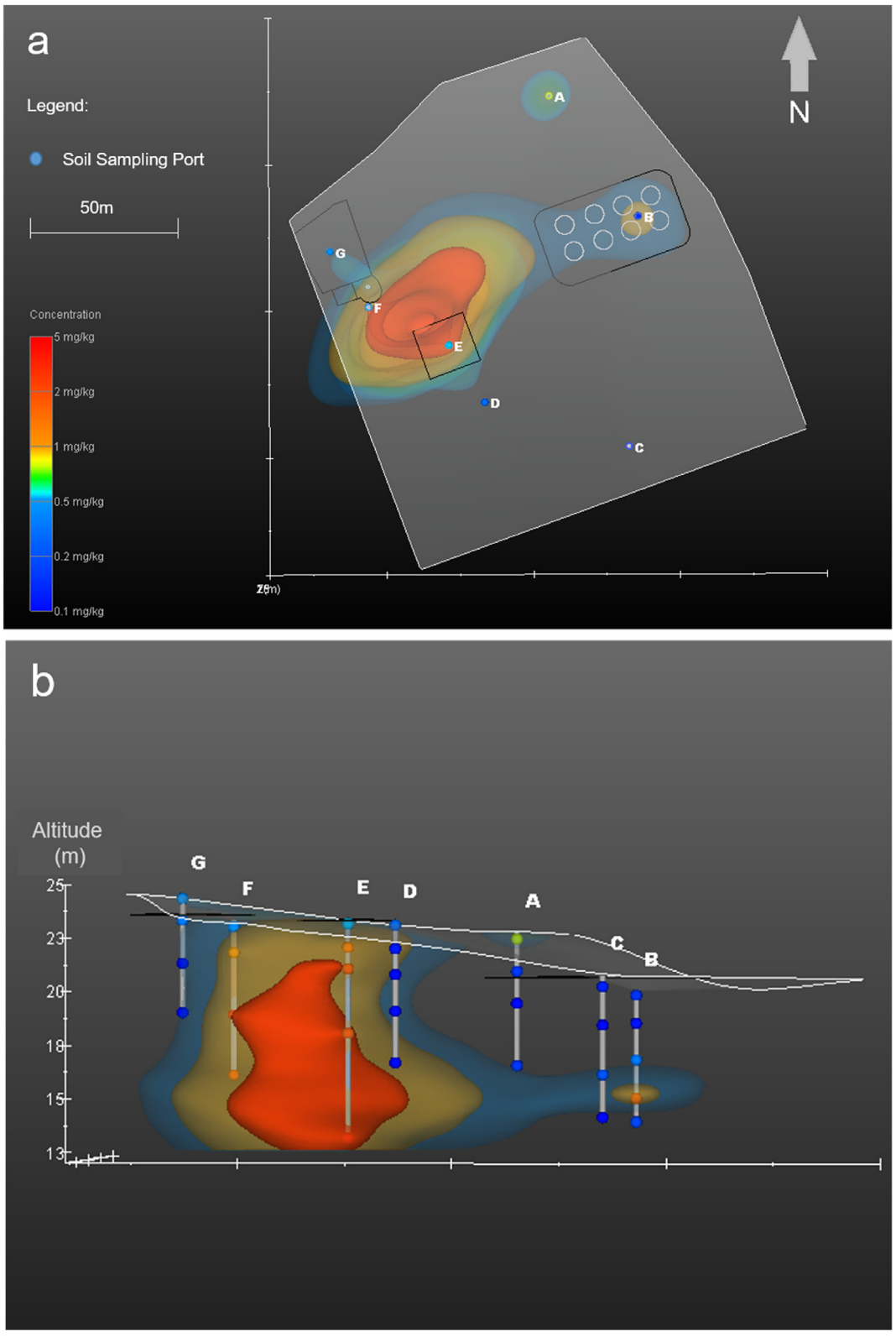

Fig. 2. The distribution of SCCPs at the CP production brownfield site: (a) vertical view; (b) profile view.

Chen et al. (2011) and Zeng et al. (2011). The existence of soil-sorption-free state CP mixtures is probable. To confirm the possibility, we established two groundwater monitoring wells during core sampling, and did observe soil-sorption-free state contaminant in the groundwater (Table S3 and Fig. S3). Thus, the CP distribution in this study appears to have less of a relation with SOM and PHC contents (Table S2). These results suggest that SCCP and MCCP migration to deeper soil layers should not be neglected.

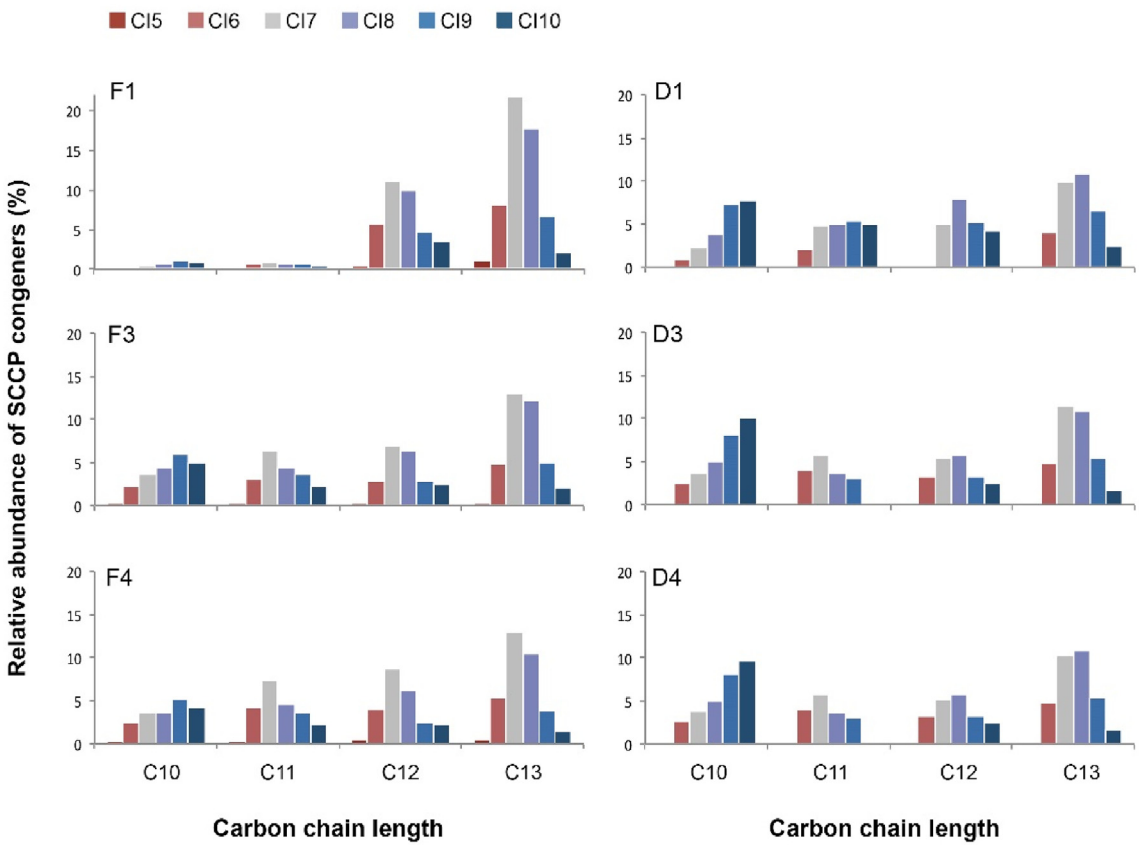

Fig. 3. The SCCP congener profiles in different sampling areas. Site F is located adjacent to the former CP storage tank while site D is near the former CP synthesis workshop.

Downward migration of these contaminants may pose ecological risks to soil animals and microbial communities (Wang et al., 2019b), or cause health hazards for local inhabitants that consume groundwater.

On the other hand, the ΣMCCPs/ΣSCCPs ratios of cores D, E, and F progressively decreased with soil depth, whereas cores A, C, and G were characterized by an opposite trend (Table S1). The different ΣMCCPs/ΣSCCPs ratios of different soil cores may be a combined result of two aspects: (1) SCCPs and MCCPs had varying behaviors and vertical migration patterns due to their different molecular weights and hydrophilicities. The octanol-water partition coefficients ($\log K_{ow}$) of CPs increase with increasing carbon chain length while vapor pressures and subcooled-liquid water solubilities usually decrease (Bettina et al., 2011; Glüge et al., 2013). Thus, SCCPs with a lower $\log K_{ow}$ are supposed to be more potential for vertical migration in soils, compared to MCCPs and (2) MCCPs may degrade into SCCPs via natural/microbial degradation, which, however, has not yet been reported by previous studies and needs further investigation.

3.2. SCCP congener profiles

SCCPs are of priority concern due to their POP-like properties compared to MCCPs and LCCPs. Their congener profiles in the environment can provide information on their sources and migration patterns. In this study, the SCCP congener profiles based on their chloride and carbon atom numbers were examined in four soil cores (B, D, E, and F). As illustrated in Fig. 3 and S2, the relative abundances of C_{13}-SCCPs, Cl_7-SCCPs, and Cl_8-SCCPs were the highest in all samples while the levels of SCCP congeners were nearly uniform among the samples. The congener profiles obtained in this study were consistent with reported SCCP congener profiles of the commercial product CP-52 (Gao et al., 2016) and PVC products (Wang et al., 2018a,b). This suggests that CP products were the main source of SCCPs in soils.

The congener profiles obtained in this study were also consistent with previous results obtained from soils and sediments, for which a higher abundance of SCCP congeners with increased carbon atoms were reported (Chen et al., 2011; Gao et al., 2012; Wang et al., 2014). Our results differ from Xu et al. (2016) and Wang et al. (2018a,b) for the other CP production plants. Xu et al. (2016) reported higher abundances of C_{10}-SCCPs, Cl_8-SCCPs, and Cl_9-SCCPs in surface soils, while Wang et al. (2018a,b) reported the predominance of C_{13}-SCCPs and Cl_{10}-SCCPs in surface soils. These differences may be due to variations in the commercial product compositions and varied congener mobilities. Most of the CP commercial products in China are synthesized without consideration of the carbon chain length or chloride profiles, provided that the average percentage of chlorine fulfills industrial standards. CPs are usually synthesized directly by free radical chlorination of *n*-alkane (paraffin) feedstock through petroleum distillation with molecular chlorine. Thus, the compositions of commercial CPs depend on the constituents of the feedstock. Products from different manufacturers may lead to different CP congener profiles. On the other hand, $\log K_{ow}$ of CPs increases with increasing carbon chain length while vapor pressures and subcooled-liquid water solubilities usually decrease (Bettina et al., 2011; Glüge et al., 2013). Consequently, congeners with shorter carbon chains and lower degrees of chlorination tend to migrate longer distances via atmospheric transport and finally deposit on surface soils. In Xu et al. (2016), the higher abundances of C_{10}-SCCPs compared with C_{13}-SCCPs may be a result of the so-called “fractionation effect” due to

atmospheric transport and deposition, which has been observed for the atmospheric dispersion of polychlorinated biphenyls and organochlorine pesticides from urban areas to the surrounding environment (Harner et al., 2004; Wang et al., 2008). Correspondingly, CP dispersion observed in this study were resulted by the CP production leakage and thus the SCCP congener profiles in soils maintained consistency with the congener profiles of the commercial formulae.

Despite the uniform nature of the SCCP congener profiles, we did observe changes along the soil depth. As shown in Figs. 3 and S2, the abundance of SCCPs with a lower degree of chlorination and those with shorter carbon chains increased with the soil depth. This trend is consistent with Zeng et al. (2011), strongly indicating that SCCP congeners vary in terms of their vertical migration patterns. This phenomenon may be due to the "fractionation effect" along depth as the result of the varied physicochemical properties of SCCP congeners. It may also be due to the degradation or dechlorination of SCCPs. A previous study demonstrated that the co-metabolic dechlorination of CPs is possible using the bacteria isolated from soils (Omori et al., 1987). Furthermore, this finding is consistent with changes noted for the ΣMCCPs/ΣSCCPs ratios along soil depth. MCCPs may also degrade into SCCPs in the soils. Further studies are needed to understand CP degradation in soils.

3.3. Soil microbial community composition and diversity

Soil microbial communities at the CP production plant brownfield site were characterized to understand the ecological effects of contamination. In total, 1,138,184 quality sequences were produced from all samples. The mean number of sequences per sample was 54,454 (ranging from 38,172 to 64,187). Most sequences (~98.86–100%) can be classified as bacteria. A total of 2698 operational taxonomic units (OTUs) was identified (defined by a 97% sequence similarity). Rarefaction analysis revealed that the full extent of microbial diversity was generally high in the reference soils, suggesting the adverse effects of contaminations.

Taxonomic classification further revealed significant differences between the community composition of the samples from the brownfield site and the reference soils. At the genus level, 144 and 221 genera were detected in ck1 and ck2 (i.e., the reference soils), respectively. The number of genera detected in samples from the brownfield site was lower, ranging 78 to 201. As illustrated in Fig. 4(a), in the samples from the brownfield site, members of *Acinetobacter*, *Brevibacterium*, *Brevundimonas*, *Corynebacterium*, *Microbacterium*, and *Stenotrophomonas* dominated across the different communities. In contrast, microbial communities in the reference soils were dominated by *Acinetobacter*, *Candidatus Solibacter*, *Acidibacter*, *Acidothermus*, and *Variibacter*. Microbial alpha diversities (Shannon's index) of the reference soils and soils from the brownfield site showed significant differences (*t*-test, $p = 0.0118$). Furthermore, the results of RDA (Fig. 4(b)) illustrated different distribution patterns for the reference soils and soils from the brownfield site. This result further indicates that the soil microbial community shifted due to contaminations.

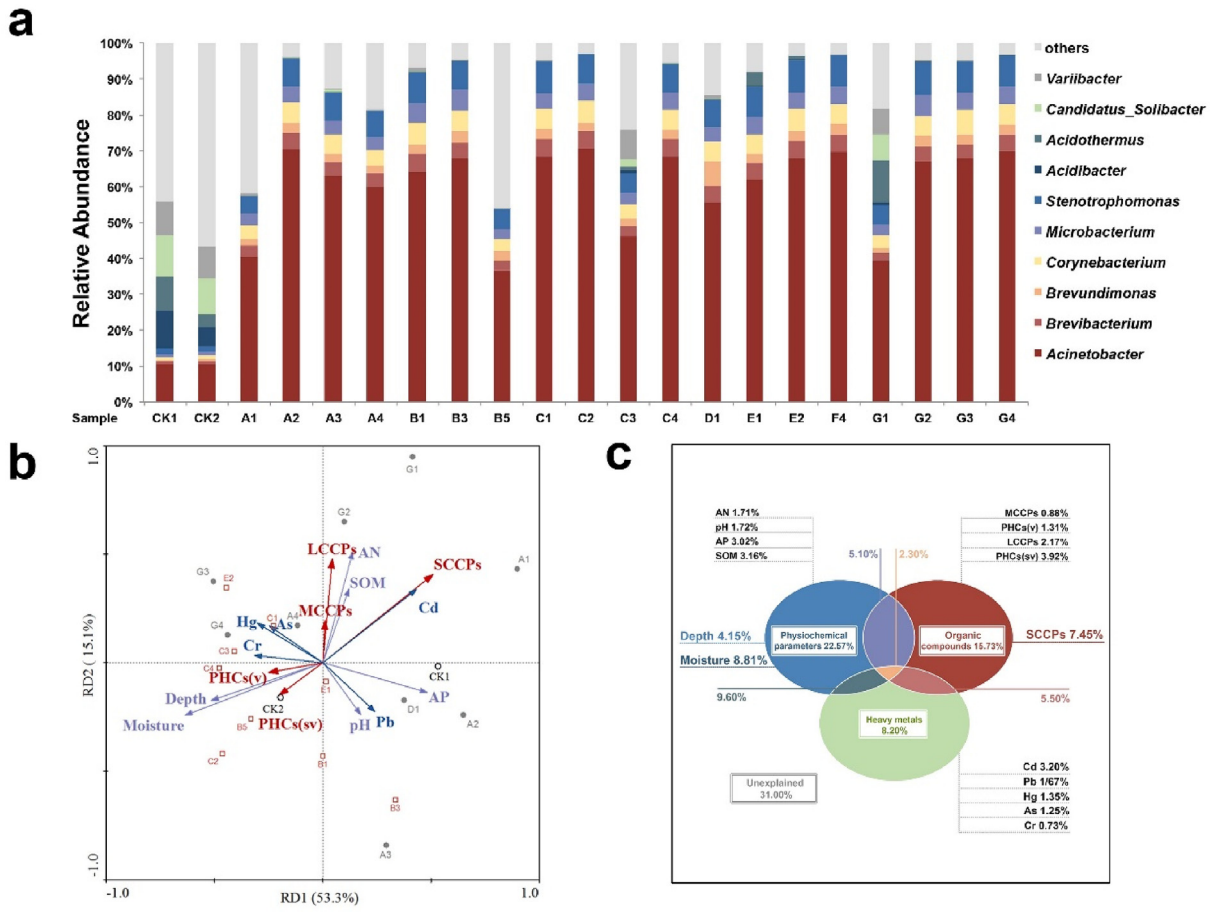

Fig. 4. (a) The relative abundance of dominant microbial taxa across selected analyzed soil samples. (b) A redundancy analysis of microbial data and the subset of sixteen environmental variables: PHCs(sv) are semi-volatile PHCs ranging C_{10}–C_{40} and PHCs(v) are volatile PHCs ranging C_6–C_9. (c) A variation partition analysis of the effects of physiochemical parameters, heavy metals and organic contaminants on microbial community composition.

Six genera of bacteria were predominant in the soil samples from the brownfield site, covering a relative abundance range of 53.8–97.0% (Fig. 4(a)). Samples were similar in terms of their microbial community composition, which may be due to the contamination by CPs and PHCs. Indeed, members of *Acinetobacter* and *Stenotrophomonas*, the two most abundant genera in all soil samples collected from the brownfield site, have the ability to degrade chlorinated organic compounds, including atrazine (Tao et al., 2019), chloroform (Zhang et al., 2019), DDT (Fang et al., 2018), and chlorimuron-ethyl (Zang et al., 2016). These genera can possibly tolerate CPs or even use them as carbon/electron sources. On the other hand, *Brevibacterium, Brevundimonas*, and *Corynebacterium* are reported to be related to the degradation of PHCs in soils (Wang et al., 2016; Zhang et al., 2016; Becarelli et al., 2019; Djahnit et al., 2019). These genera can achieve co-metabolism of PHCs with other carbon sources and, thus, may be able to degrade less toxic CPs via the same metabolic pathway. *Microbacterium* are less related to the degradation of organic compounds than the abovementioned genera. However, members of *Microbacterium* are able to secrete chitosanase, a natural surfactant that may increase the bioavailability of chlorinated contaminants such as perchloroethylene, and facilitate its degradation by other bacteria genera (Sun et al., 2015; Wan et al., 2019). Similar effects might also occur in the studied samples at site scale. The presence of chitosanase or other biosurfactants might enhance the desorption of hydrophobic contaminants from soils and decrease the surface tension between contaminants and bacterial cellular membranes, facilitating the transfer of contaminants from soils to bacterial cells. The biosurfactant secretion ability has also been reported for members of *Stenotrophomonas* (Tiwari et al., 2016) and *Brevundimonas* (Singh et al., 2016). Thus, different microbial genera in the community might "cooperate" in the degradation of CPs and PHCs with different divisions of function.

Further, RDA was performed to quantify the relative influences of the environmental variables on the overall microbial diversity of soils collected from the brownfield site. These variables include soil physiochemical properties, CPs, PHCs, and metal contaminants (data shown in Tables S1 and S4). As shown in Fig. 4(b), the first two axes of the RDA explained 53.3 and 15.1% of the total variation, respectively. Moreover, the sixteen selected variables together can explain 69.00% of the observed variation in the community composition, leaving 31.00% unexplained (Fig. 4(c)). Subsequent variance partitioning analysis revealed that the six soil parameters, five organic contaminant species, and five metal contaminant species explained 22.57, 15.73, and 8.20% of the variation, respectively. The effects of interaction among all the selected variables explained 22.50% of the variation *(Monte Carlo* permutation test, $p < 0.05$). Moreover, the community compositions were clustered in accordance with the CP vertical distribution patterns. In other words, samples from the synthesis workshop, storage tank, and their surroundings (cores B, C, E, and F) were more similar in their soil community compositions while other areas (cores A, D, and G) shared different soil community composition patterns (Fig. 4(a)(b)). These results are consistent with our previous findings that soil properties and contaminations together shape the compositions of the microbial communities (Wu et al., 2017).

Notably, the microbial community was significantly influenced by the soil moisture, SCCPs, and soil depth, in descending order (all $p < 0.05$, Fig. 4(c)). Soil moisture and depth together accounted for 12.96% of the bacterial community variation, while SCCPs alone accounted for 7.45%. Soil moisture and depth were important factors regulating microbial diversity in this study. Spearman's correlation analysis revealed that several predominant genera were significantly correlated with soil moisture or depth (Table S6). Consistent result has been found by previous studies that soil moisture is the primary factor influencing microbial community compositions, regardless of the soil type (Schimel et al., 1999; Liu et al., 2015). In addition, Brockett et al. (2012) found that soil moisture is an important driver of overall microbial activities and is closely related to the microbial biomass, basal respiration, and enzymatic activities. Variation in the soil moisture may alter soil physicochemical properties, including the pH, redox potential, and total nitrogen content, which may lead to a complex effect on the enzymatic activities and microbial community composition. The influence of soil depth, on the other hand, is usually correlated to the changes of other important factors including the carbon availability and composition, pH, redox potential, and temperature (Liu et al., 2018; Zhao et al., 2019). These changes provide different living habitats for the microbial community. The environmental gradient represented by the depth profiles influences the abundance, composition, and functions of soil microbial communities (Stone et al., 2014). In this study, the soil depth showed a significant positive correlation with the soil moisture content (Table S2). Thus, the influence of the soil depth is also related to the moisture.

Among the significant influencing factors, only SCCPs is related to the contamination. It is not only a counteracting variable against the soil moisture and depth, but also a variable with the same influencing direction with cadmium (Fig. 4(b)). These results suggest that SCCPs may influence the microbial community compositions in a pathway similar to cadmium (Abaye et al., 2005; Chen et al., 2016) due to their ecotoxicity. As known, cadmium can cause oxidative stress by generating reactive oxygen species (ROS), which can lead to DNA damage, inhibit the DNA mismatch repair system, and disrupt the synthesis of nucleic acids and proteins (Jin et al., 2003). SCCPs in the soils may also generate ROS and damage bacterial cells in a similar process with cadmium. Indeed, the generation of ROS and the inhibitory effects on cell viability induced by CPs have been identified by cell viability assay and pseudotargeted metabolomics studies on human hepatic cells at laboratorial scale (Ren et al., 2019). Similar toxicity mechanism may occur to microbial cells. So far, only a limited number of studies have investigated the toxicity of SCCPs to microbial communities. Despite that, the significant decrease in microbial respiration in the microbial tests has been reported (Bezchlebová et al., 2007). Moreover, as shown in Fig. 4(c), although present at low contents, SCCPs appeared to exert an important effect on soil microbial community composition, compared to other contaminants such as MCCPs, LCCPs and PHCs. The significant effect on soil microbial community compositions by SCCPs at low contents further suggests their higher ecological risk compared to the other CP species and PHCs. Intensive studies are needed to understand the toxic effect and the underlying mechanism.

3.4. Co-occurrence network analysis

The relative abundance of dominant microbial taxa across soil samples can provide information on the composition and diversity of the soil microbial communities. However, it cannot provide visual information on the microbial interactions. To better understand the topological properties of the microbial community composition, a co-occurrence network was established to describe the complex pattern of inter-relationships among nodes and identify the keystone taxa (commonly understood as species that have a disproportionate deleterious effect on the community upon their removal (Berry and Widder, 2014)).

The microbial network of the soils from the CP production brownfield site is illustrated in Fig. 5. The microbial network consisted of 53 nodes (genera) and 249 edges (with a mean of 4.698 edges per node). Comparison of this network with a random

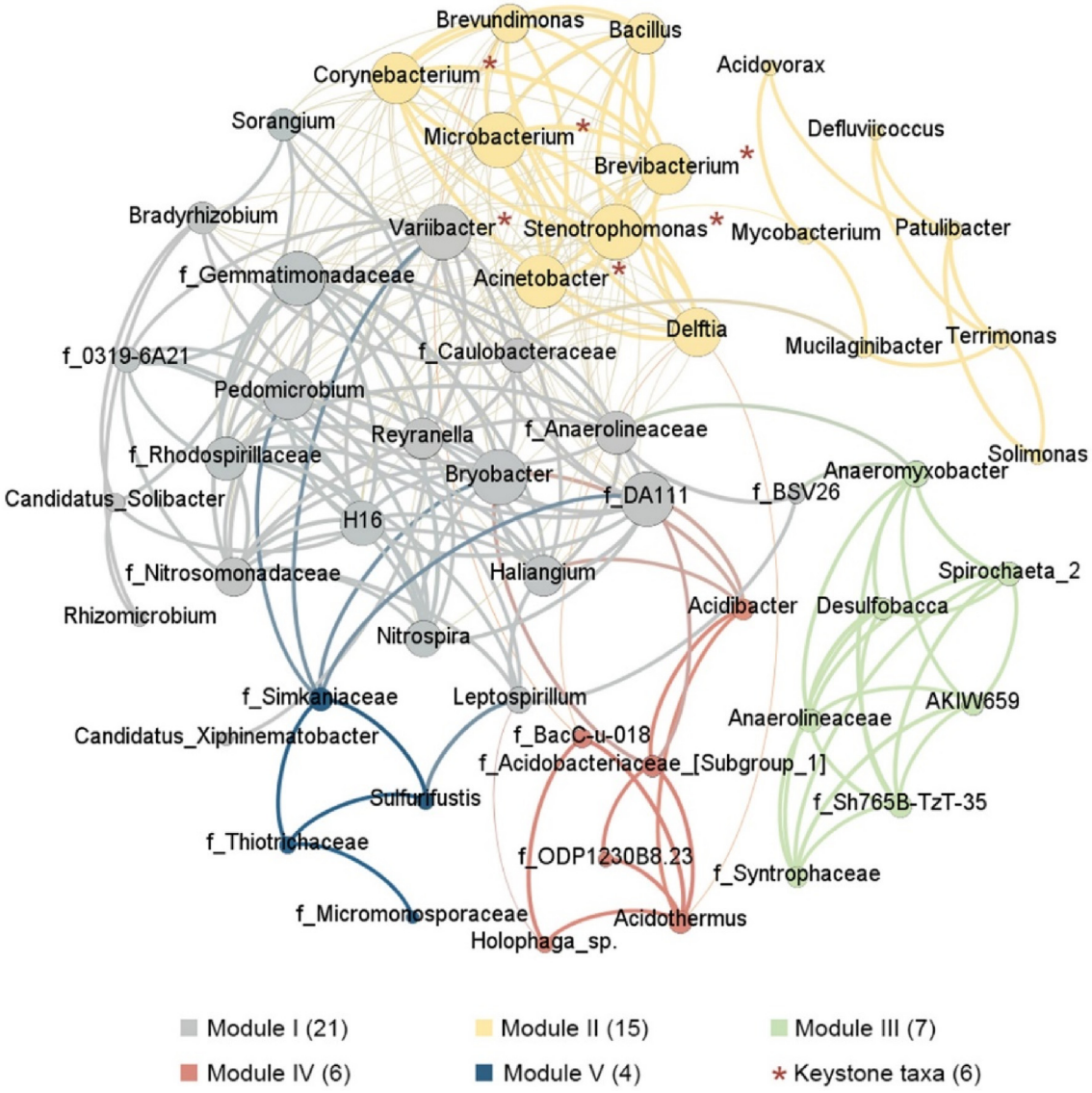

Fig. 5. A co-occurrence network colored by the modularity classes based on a correlation analysis. A connection indicates a strong (Spearman's r > 0.6) and significant (P < 0.05) correlation. The size of each node is proportional to the relative abundance. The thickness of each connection between two nodes (edge) is proportional to the value of the Spearman's correlation coefficients.

network illustrated the role of deterministic processes in the structure of the microbial community (Table S5). A "small world" topology was observed with strong ecological links that manifested as a cluster of robust co-occurrence correlations.

Five modules of bacteria genera with different functions were identified (Fig. 5). Module I is mainly related to the biogeochemical cycles of C and N. For example, *Nitrospira* bacteria are reported to be nitrifiers (Daims et al., 2015). *Bradyrhizobium, Bryobacter, Candidatus Solibacter,* members of *Rhodospirillaceae*, and several other genera are also involved in C- and N- cycles (Anderson et al., 2011). Module II is likely related to the degradation of organic contaminants. Members of this module included *Acinetobacter*, *Brevibacterium*, *Brevundimonas*, *Corynebacterium*, *Microbacterium*, and *Stenotrophomonas*. They are related to the degradation of organic compounds, such as PHCs and chlorinated organics, as discussed in section 3.3. The short distance between several members from modules I and II indicates a high probability of co-occurrence of these genera and suggests that their functions may be correlated. "Cooperation" by genera from these two modules may have led to the co-metabolism processes of organic contaminants. Certain members in module III, such as *Desulfobacca* and *Spirochaeta*, were reported to be related to electron transfer (Jiao et al., 2016). Biological electron transfer is an important process in the microbial degradation of organic contaminants. Hence, module III may take part in the degradation of CPs and PHCs by transferring electrons. The functions and linkages of different modules indicate that members of the soil microbial community "cooperate" to resist the stress from PHCs and CPs. This community may also have the potential to degrade both contaminants.

The keystone genera in the microbial community are *Acinetobacter*, *Brevibacterium*, *Corynebacterium*, *Microbacterium*, *Stenotrophomonas*, and *Variibacter* (Fig. 5). It is worth noted that all the keystone genera except for *Variibacter* are in module II, which is likely related to CP and PHC degradation, suggesting the function of module II may be a crucial process for the community. Moreover, these genera are the most abundant in soils from the brownfield

site (Fig. 4(a)), indicating they are a dominant part of the community. They are adapted to the habitat shaped most significantly by soil moisture, SCCPs and depth, and are candidate indicator microbial groups for the SCCP contamination. These results suggest that the exposure to CPs and PHCs may lead to a self-purification function of soil microbial community based on co-metabolism.

4. Conclusions

In this study, the spatial and vertical distribution of SCCPs, MCCPs, LCCPs and PHCs in soils from a CP production plant brownfield site was investigated. We found that long-term manufacturing and storage led to soil contamination by not only SCCPs, MCCPs and LCCPs, but also PHCs. The downward migration potential of CPs, especially SCCPs, in soil layer was rather significant and a hotspot of SCCPs was found 10 m beneath the synthesis workshop. Soil microbial community composition and diversity were influenced by the contamination by SCCPs, although which was at rather low contents compared to other CPs and PHCs. These results suggest that groundwater in this area may also be contaminated due to CP production, with CPs and PHCs as particular contaminants. Further studies should focus on the combined effect of CPs and PHCs, ecological risk they present to soil organisms and the health risk they pose to local people that consume groundwater. Fortunately, we also found the microbial community inhabiting the contaminated region may offer potential to degrade CPs and PHCs. Further studies are also needed to quantify the possibility and understand the degradation pathway.

CRediT authorship contribution statement

Yingxin Wu: Conceptualization, Methodology, Writing - original draft, Funding acquisition. **Jiahui Wu:** Data curation, Software. **Haijian Tan:** Validation, Writing - review & editing. **Qingmei Song:** Investigation, Methodology. **Jie Zhang:** Investigation. **Xi Zhong:** Data curation, Software, Visualization. **Jingyan Zhou:** Investigation, Visualization. **Wencheng Wu:** Supervision, Conceptualization, Resources, Writing - review & editing. **Xinde Cai:** Project administration, Funding acquisition. **Weihua Zhang:** Funding acquisition. **Xiaowen Liu:** Project administration.

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Appendix B. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2020.114328.

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