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Sulfonamides removal under different redox conditions and microbial response to sulfonamides stress during riverbank filtration: A laboratory column study

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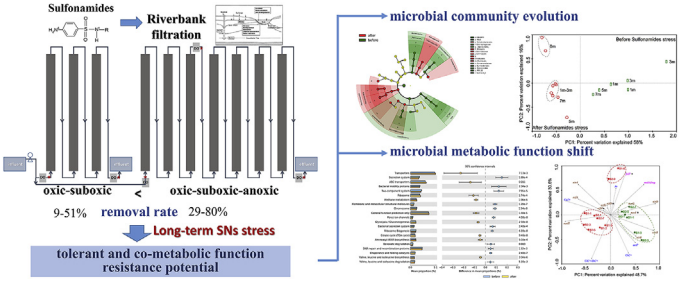
HIGHLIGHTS

- Longer residence time and anoxic condition will benefit sulfonamides (SNs) attenuation in RBF.
- Phylogenetic diversity of bacterial community increased slightly under SNs stress.
- SNs resistance developed in specific bacteria under selective pressure.
- Activated transport function in bacterial community was developed under SNs stress.

GRAPHICAL ABSTRACT



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ABSTRACT

Riverbank filtration (RBF) as a barrier of pathogenic microorganisms and organic micropollutants recently has been proven capable of removing sulfonamides. However, the study about the effect of redox conditions on biodegradation of common and persistent sulfonamides in RBF is limited and the response of microbial communities to sulfonamides stress during RBF is unknown. In this study, two column set-ups (with residence time 5 days and 11 days respectively), simulating different redox conditions of riverbank filtration systems, were operated for seven months to investigate 1) the long-term effect of redox conditions on ng·L^{-1} level sulfonamides (sulfapyridine, sulfadiazine, sulfamethoxazole, sulfamethazine, sulfaquinoxaline) removal, and 2) the microbial community evolution represented by the phylogenetic and metabolic function shift under non-lethal selective pressures of sulfonamides. The results showed that sulfonamides were more degradable under anoxic conditions than oxic and suboxic conditions. In the sulfonamides stressed community, the phylogenetic diversity increased slightly. Relative abundance of an intrinsic sulfonamides resistant bacteria *Bacillus* spp. increased, suggesting that sulfonamide resistance developed in specific bacteria under sulfonamides contamination pressure in RBF systems. At the same time, an activated transport function in the stressed microbial community was noticed. The predicted relative abundance of gene *folP*, which encodes dihydropteroate synthase, also increased significantly, indicating a detoxification mechanism and sulfonamides resistance potential under non-lethal selective pressures of sulfonamides in RBF systems.

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1. Introduction

During years, sulfonamides (SNs) have been used as veterinary antibiotics (Touraud, 2008), taking part of 2.3% global annual consumption in USA and 23% in European countries (Sarmah et al., 2006). Unfortunately, they are not completely absorbed by the organisms and up to 75%–90% are released in the environment (Sarmah et al., 2006). As most of the SNs are polar organic chemicals and water-soluble, they are highly mobile and have been detected in a lot of environmental compartments including soil, water and sediment (Hirsch et al., 1999; Battaglin et al., 2000; Hao et al., 2006; Li et al., 2012), posing potential risk on ecosystems and drinking water resources. The persistence may be intensified by application of contaminated manure on farmland and discharge of slurry, wastewater or sewage sludge (Lai et al., 2011). Concern is growing about water quality deterioration affected by the discharge of sulfonamides, the accelerated evolution of sulfonamides resistant bacteria, as well as the accumulation and dissemination of antibiotic resistance genes (Schauss et al., 2009; Heuer et al., 2011; Deng et al., 2016; Yang et al., 2017).

Riverbank filtration (RBF) has long been recognized as an effective and sustainable treatment technique for pathogenic microorganisms and organic micropollutants (OMPs) removal (Tufenkji et al., 2002). In RBF systems, as residence time increases along the subsurface water flow path, the dissolved oxygen (DO) is consumed continuously and the redox condition gradually changes from oxic zones, via suboxic zones, to anoxic zones during RBF (Bertelkamp, 2015) (Bertelkamp et al., 2016b). In RBF systems, redox condition is an important factor for microbial abundance, activity, function and community composition, which correspondingly affect the biodegradation of antibiotics (Roling et al., 2001; García-Galán et al., 2008). A number of studies have focused on the removal of OMPs under different redox conditions in RBF systems (Storck et al., 2012; Regnery et al., 2015), including SNs. Sulfamethoxazole, the most persistent SNs, was reported more effectively degraded under oxic than anoxic condition (Baumgarten et al., 2011a), while another study gave an opposite conclusion of more rapid and efficiently (99%) elimination under anoxic condition (Heberer et al., 2008). Degradation rate of p-TSA in groundwater, one of the metabolite of SNs, increased due to the microbial adaption to the change of redox conditions from anoxic to oxic (Richter et al., 2009; Meffe et al., 2012).

It is imperative to further identify the factors governing the fate of SNs during RBF and to understand the effect they will have on microbial community (Mohatt et al., 2011). Microbial community structure and its succession under OMPs pressure in RBF systems are essential for identifying microbial geochemical processes and OMPs biodegradation mechanisms. However, the studies about microbial community structure and the community succession in RBF systems are quite limited, only a few bacteria species have been identified in specific RBF sites and experimental set-ups. A study reported the presence of Fe-oxidising bacteria (*Rhodobacter* spp.), S-oxidising bacteria (*Sulfuricurvum* spp.), NO_3^--reducing bacteria (*Ferribacterium*), SO_4^{2-}-reducing bacteria (*Acidovorax* spp.), and Fe(III)-reducing bacteria (*Desulfobrio* sp.) in a RBF site (Medihala et al., 2012). *Proteobacteria, bacteriodetes, actinobacteria* and *firmicutes* were identified as major phylum present in a soil aquifer treatment column set-up (Onesios-Barry et al., 2014). Taken together, no study has focused on the microbial community function and structure in response to continuous pressure of SNs in RBF systems. Although SNs' effect on microbial communities have been frequently studied during wastewater treatment (Collado et al., 2013; An and Qin, 2018), the interaction between SNs and microbial community structure at sub-inhibitory (non-lethal) concentrations in typical drinking water treatment process still need more elaboration.

In the present study, two laboratory-scale column set-ups (residence time 5 days and 11 days respectively) simulating different redox conditions of RBF systems were operated for seven months to investigate 1) the attenuation dynamics of $ng \cdot L^{-1}$ level sulfonamides mixtures in RBF systems, and the long-term effect of redox condition on the removal ratio, 2) the microbial community succession represented by the phylogenetic and metabolic function shift due to non-lethal selective pressures of sulfonamides in RBF systems.

2. Materials and methods

2.1. Experimental set-up and sampling

Two independent laboratory scale RBF sand column set-ups were used. Set-up B1 consisted of 3 stainless steel columns ($L = 1$ m, $D = 36$ mm) in series to simulate the oxic and suboxic zones of RBF systems. Set-up B2 consisted of 7 same stainless steel columns in series to simulate the oxic, suboxic and anoxic zones of RBF systems. Each column was filled with natural RBF sand collected from the filtration site of Dunea drinking water company (The Hague, the Netherlands) (Wang et al., 2016). A bottom-to-top flow was maintained to prevent air entrapment and the RBF systems were operated in a dark climate room with a controlled temperature of $11.5 \pm 5\,^{\circ}C$, corresponding to the natural aquifer temperature (Bertelkamp et al., 2016a). The applied hydraulic loading rate was 27 mL/h, which equals a filtration rate of 0.64 m/d, with a residence time of 4.66 d and 10.88 d for RBF systems B1 and B2 respectively. Pore velocity and porosity in the pilot was determined by tracer experiment using deuterium (^2H) with a DLT-100 Liquid-Water Isotope Analyser (Los Gatos Research, USA). The experimental set-up is shown in Fig. 1.

The RBF systems B1 and B2 were operated for seven months in total, including adaptation period of 2 months (16th Feb to 16th Apr, 2016) and formal experimental period of 5 month (16th Apr to 14th Sep, 2016). SNs antibiotics, sulfapyridine, sulfadiazine, sulfamethoxazole, sulfamethazine and sulfaquinoxaline (Sigma, St Louis, MO, United States), were dosed to the RBF systems B1 and B2 in the formal experimental period. Approximately 0.5 mg/L working solutions of each SNs compound were prepared by dissolving all the material directly into ultra-pure water. It was reported that selection and enrichment for antibiotic resistant bacteria is often a consequence of non-lethal selective pressures in anthropogenically drug-polluted natural environments (Andersson and Hughes, 2012) and the minimal selective concentration (MSC) of antibiotics ranged from 100 ng/L (ciprofloxacin) to 1 mg/L (streptomycin) (Gullberg et al., 2011). Thus, the dosed concentration in this study was set 2.5 μg/L (500 ng/L each) based on the MSC and antibiotic levels detected in European surface waters (Kümmerer, 2009; (WHO, 2011); Straub, 2013; Larsson, 2014; Petrie et al., 2015).

Water samples were collected in duplication at 0 days, 5 days and 10 days after every influent refreshing from influent, effluent B1 and B2 respectively. Before SNs addition, sand samples (2 g) were taken at depth of 1 m, 3 m, 5 m and 7 m (B1-1, B1-3 for B1; B2-1, B2-3, B2-5, B2-7 for B2). After the 5 months operation, sand samples (2 g) were taken from the influent side (B1-0 and B2-0 for B1 and B2 respectively), as well as at depth of 1 m, 2 m, 3 m, 5 m and 7 m (B1-1, B1-2, B1-3 for B1; B2-1, B2-3, B2-5, B2-7 for B2). All sand sample were stored at $-20\,^{\circ}C$ for DNA extraction.

2.2. Water quality and soil parameters

DO, pH and temperature of the water samples were measured using a multimeter (SenTix® 940 IDS probe, Multi 340i, WTW, Germany). Dissolved organic carbon (DOC) was measured with a

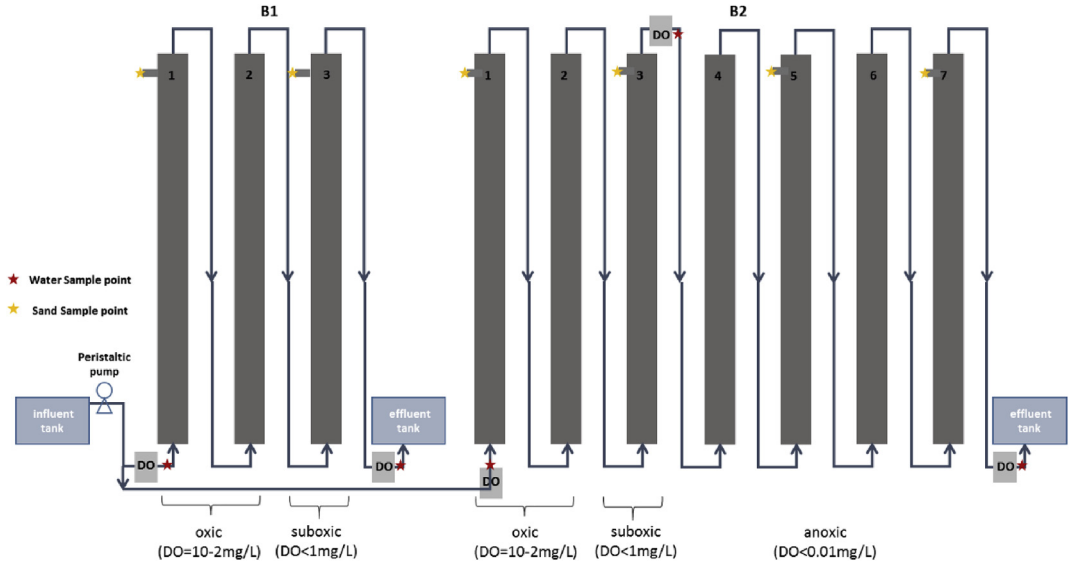

Fig. 1. RBF column set-ups.

Shimadzu TOC analyser. Ion concentrations (Cl^-, NO_3^-, NO_2^-, PO_4^-, SO_4^{-2}, Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}) were measured by a ProfIC 15–AnCat ion chromatograph (Metrohm 881 anion (suppressed) and 883 cation system, Metrohm, Switzerland). Adenosine triphosphate (ATP), as parameter to determine the microbial activity in the RBF systems, was measured using a Quench Gone Aqueous test kit and a LB9509 luminometer (both Aqua tools, France).

Sand characteristics were determined with laser diffraction (Helium-Neon Laser Optical System, KR). The inorganic constituents of the sand were determined by X-ray diffraction analysis (Department of Materials Science and Engineering, TU Delft). Organic matter (OM) content of sand was determined by 550 °C lose-on- ignition (LOI) method (Heiri et al., 2001). Soil pH was measured by multimeter (SenTix 41 probe, Multi 340i, WTW, Germany).

All the parameters were measured in duplication for quality control.

2.3. Water sample pretreatment and sulfonamides quantification

Water samples (1000 mL) were filtered through 0.45 μm glass fiber filter (15 cm, Merck Millipore), acidified to pH 3, added 0.5 g/L Na_2EDTA, then captured and purified by solid-phase extraction (SPE) using Oasis HLB cartridges (6 mL/500 mg/30 μm, Waters, USA). An automatic SPE-system (GX-2754 Aspec, Gilson, USA) were used in this process. Sulfonamides were then quantified with a private method using UPLC-MS/MS (XEVO, Waters, USA). For the calibration, we use a combined standard solution of each sulfonamide component and also an internal (control) standard. The details was showed in supplementary material (Tables S2–S5).

2.4. Pyrosequencing and taxonomic assignment

Total genomic DNA extraction of soil samples was performed using Ultraclean Water DNA Kit (MoBio Laboratories, Inc.) according to the manufacturer's protocol. The extracted DNA was purified using the Power Clean DNA Clean-Up Kit (Mo Bio laboratories). The quality and concentration of the purified DNA was determined by spectrophotometer analysis and 1.5% agarose gel electrophoresis. DNA samples were stored at −20 °C for sequencing. The V4 region of the bacteria 16S rRNA was amplified using region-specific primers for paired-ends sequencing on Illumina MiSeq platform (BGI, Wuhan, China). The sequences displaying more than 97% identity with each other were grouped into one operational taxonomic unit (OTU) using software QIIME (version 1.9.0, USA) based on greengene database. The MiSeq preparation and sequencing protocol is described in former research (Wan et al., 2017). The OTU lists of samples were submitted to the LEfSe pipeline (LDA Effect size, http://huttenhower.sph.harvard.edu/galaxy/) to identify significant differential features of microbial community.

2.5. Metagenome predictions from 16S data

Sequencing data of 16S rRNA were used to predict metagenome of microbes by PICRUSt (phylogenetic investigation of communities by reconstruction of unobserved states), which uses an extended ancestral-state reconstruction algorithm to predict which gene families are present and then combines gene families to estimate the composite metagenome (Langille et al., 2013). The resulting sequences were assembled and annotated based on KEGG database and 3.2G metabolic related genes were obtained from the samples. All the analysis were made by PICRUSt 1.1.1 script (http://picrust.github.io/picrust/). The figures were made by STAMP v2.1.3.

2.6. Statistical analysis

The student's T test ($p < 0.01$) of sulfonamides removal ratio were conducted using SPSS version 21. In the statistical analysis between relative abundance of bacterial taxa and predicted functional gene in community before and after sulfonamides pressure, significant difference were identified with a confidence interval of 95%. Principle component analysis (PCA) of microbial taxonomy and predicted KEGG pathways were conducted by Canoco version 4.5 to evaluate the diversity of microbial community structure and function. Redundancy analysis (RDA) was also conducted to reveal the influence of environmental factors on microbial community alternation.

3. Results

3.1. Sand characteristics, dissolved oxygen and redox conditions

The sand composition, organic carbon content and pH in B1 and

B2 are shown in Tables S6 and S7. It is worth noting that the Fe_2O_3, MnO and organic matter content of the column sand were 1.74, 0.04 and 0.42 respectively. The average DO in the influent water was about 12 mg/L and dropped to less than 1 mg/L after a residence time of 4.66 days in B1, indicating the development of axic condition and suboxic condition. DO was almost completely removed after a residence time of 10.88 days in B2 (Fig. S1), indicating the development of oxic, suboxic and anoxic conditions. The DOC removal ratio of both systems were stable after two months of operation, implying that the expected steady biomass activity was obtained (Fig. S2). Besides, the higher DOC removal in system B2 is likely a combined outcome of redox condition and longer residence time (Regnery et al., 2017).

The average NO_3^- concentration in the influent of both B1 and B2 is about 13.91 mg/L, and the average NO_3^- concentration in effluent B1 and B2 are 6.34 mg/L and 0.99 mg/L respectively (Fig. 2). The NO_3^- removal ratio of RBF system B2 reached almost 100% after one month of adaptation, while that of RBF system B2 fluctuated from 2% to 97% with most of the values between 20% and 60%. The above results confirmed the occurrence of oxic and suboxic (partial NO_3^- removal) zone in B1, and oxic, suboxic and anoxic (complete NO_3^- removal) zone in B2.

3.2. Sulfonamides degradation ratio

A statistically significant difference in SNs removal between oxic-suboxic RBF system B1 and the oxic-suboxic-anoxic RBF system B2 was observed ($p \leq 0.05$). The removal ratio of five SNs compounds are shown in Fig. 3. The average removal ratio of sulfapyridine, sulfadiazine, sulfamethoxazole, sulfamethazine and sulfaquinoxaline were 50.9%, 29.6%, 32.3%, 9.0% and 39.2% in B1, and 73.2%, 41.5%, 79.6%, 28.5% and 49.1% in B2. It can be seen that SNs removal ratio of B2 exceeded that of B1 almost in all cases except for the first sampling.

3.3. Evolution of microbial community under SNs pressure

Before SNs contamination, over 52.0% of the microbial community diversity was represented by *Gemmatimonadetes* (5.2%), *Comamonadaceae* (4.7%), *Acidobacteria-6* (3.9%), *Syntrophobacteraceae* (3.2%), *Nitrospira* (2.4%), *Chloracidobacteria* (2.3%), *Flavobacterium* (2.2%) and *Anaerolineae* (2.2%). 2.7% concerned unassigned sequences while the remaining 45.7% was represented by less abundant populations (individually <1%). In contrast, the community after under SNs contamination was dominated by *Acidobacteria-6* (5.2%), *Gemmatimonadetes* (4.5%), *Candidatus Nitrososphaera* (3.4%), *Syntrophobacteraceae* (3.3%), *Nitrospira* (3.2%), *Pirellulaceae* (2.5%), *Anaerolineae* (2.2%), which represented over 43.1% of the populations (individually >1%). Shannon index (Table S8) for the 16S amplicons also indicate that the community under SNs contamination was more diverse. There was an obvious abundance difference in the phylum of *Actinobacteria, Planctomycetes, Chloroflexi, Nitrospirae* (increase) and *Proteobacteria* (decrease) under SNs pressure (Fig. 4). In the decreased taxa, *Comamonadaceae, Xanthomonadaceae* and *Oxalobacteraceae* are most significant (Table S9). In Fig. 4, it can be noticed that the

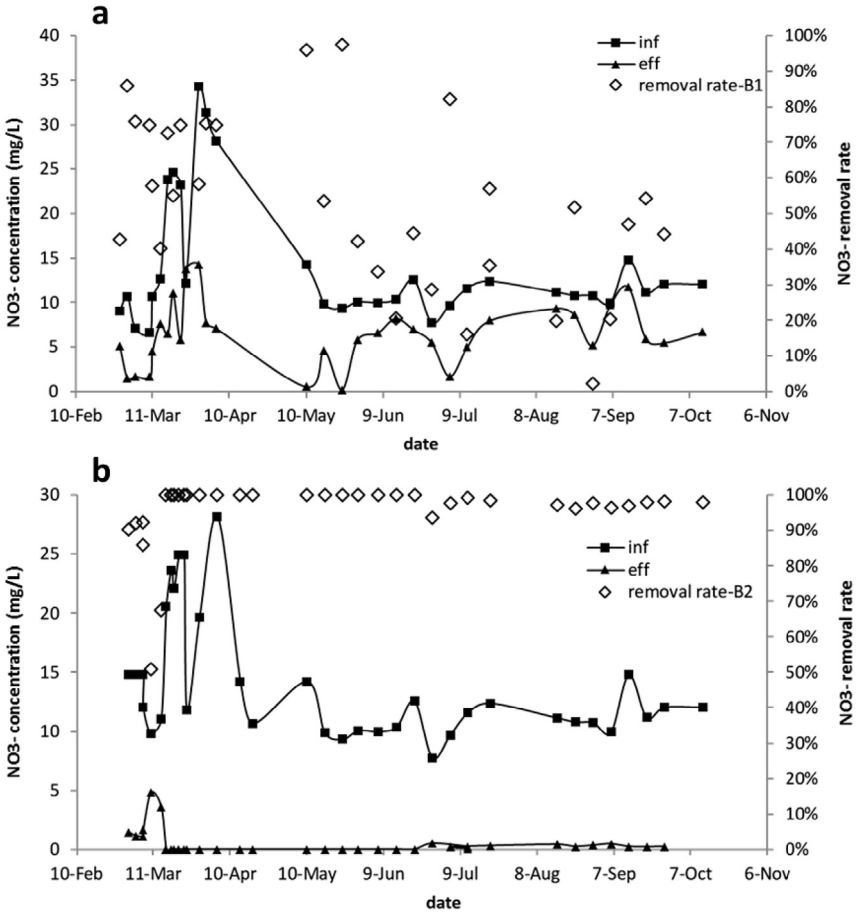

Fig. 2. Nitrate removal ratio in RBF systems B1 (a) and B2 (b).

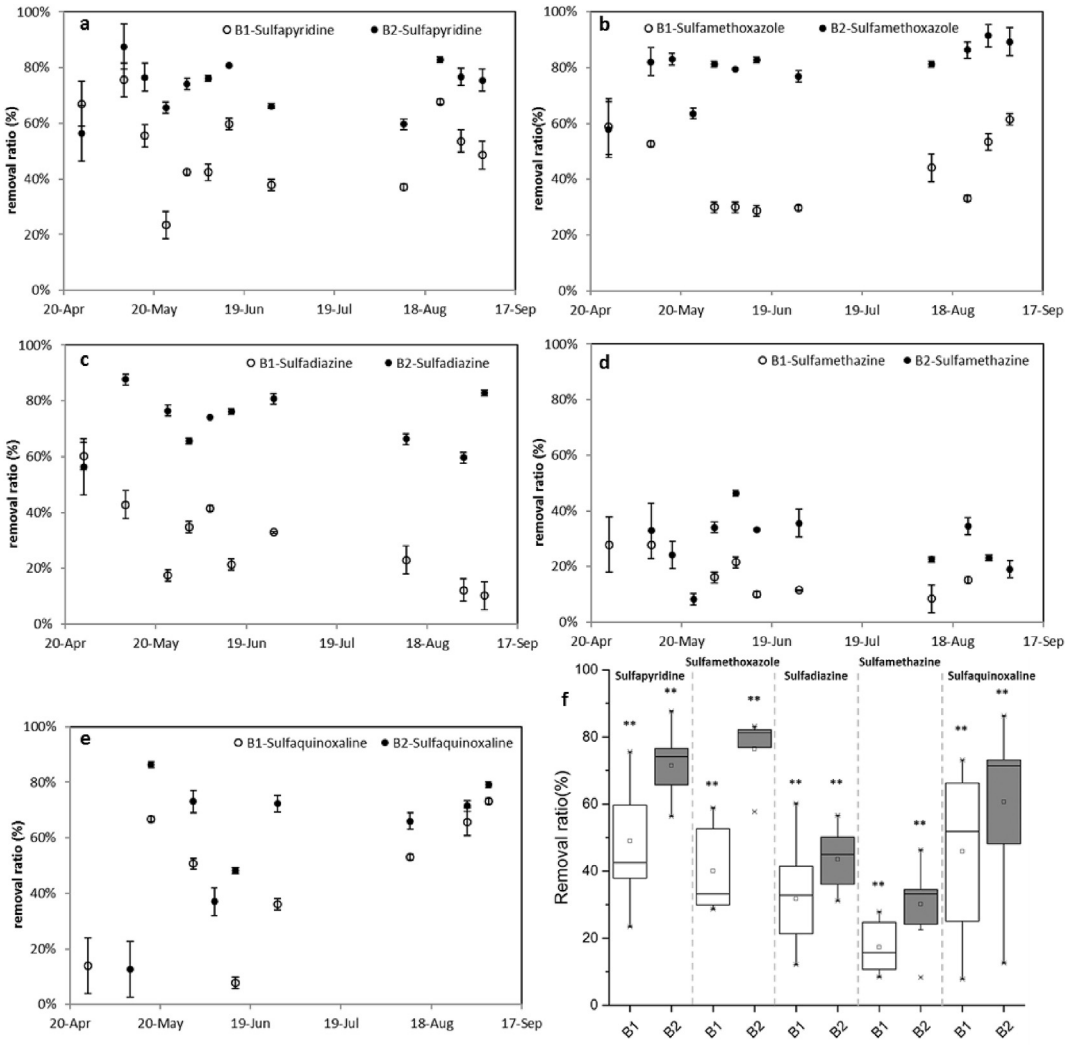

Fig. 3. Removal ratio of sulfapyridine (a), sulfadiazine (b), sulfamethoxazole (c), sulfamethazine (d), sulfaquinoxaline (e) and statistical summary (f) (N = 12, $p < 0.01$).

relative abundance of *Comamonadaceae* decreased significantly from 4.7% to 0.5% after SNs pressure, while *Solirubrobacterales*, *Pirellulales, Gemmataceae*, *A4b, GCA004*, and *Nitrospira, 0319-6A21* went through a slight increase after SNs pressure, which was consistent with the trend at phylum level.

For the microbial community under SNs pressure, the main species also differ with increase of residence time and change of redox condition. It can be noticed that the relative abundance of *acidobacteria-6* at oxic zones (B1-0 and B2-0) was higher than that of further reducing condition. The relative abundance of *Gemmataceae* decreased with the increase of residence time. The increased taxa from the beginning (B2-1) to the end (B2-7) of B2 included *Peptococcaceae*, *Thermodesulfovibrionaceae*, *Geobacteraceae* and *Desulfobulbaceae* (Table S10).

It can be seen from the PCA that the microbial community structure before and after SNs pressure differed significantly at principal component 1 (explained 58% of the variation), revealing a strong influence of antibiotic contamination on microbial community structure in RBF system (Fig. 5). At principal component 2 (explained 16% of the variation), samples showed a variation between shorter residence time (0m-3m, 0 days—4 days) and longer ones (5m-7m, 7 days—10 days).

LEfSe analysis revealed that the microbial community structure went through an intense evolution with *Gemmataceae* spp., *Nitrospira* spp., *Solirubrobacterales* and *Nitrososphaeraceae* spp. more abundant in community under sulfonamides stress, while *Chitinophagaceae* spp., *Ellin6075 spp.* and *PRR-10 spp.* showed the opposite trend (Fig. 6). Further statistical analysis showed that the main taxa with significant abundance increase ($p < 0.05$) belong to family *Bacillaceae*. The taxa with significant abundance decrease ($p < 0.05$) belong to family *Comamonadaceae, Xanthomonadaceae and Oxalobacteraceae* (Table S9). It was also observed that the microbial community structure changed with residence time and redox conditions (Fig. 4). From oxic zones (1 m) to the anoxic zones (7 m), the main taxa with significant abundance increase ($p < 0.05$) included bacteria family *Peptococcaceae, Thermodesulfovibrionaceae, Geobacteraceae* and *Desulfobulbaceae*. The taxa with significant abundance decrease ($p < 0.05$) was *Planctomycetaceae* (Table S10).

The redundancy analysis showed that taxa *Acidobacteria-6, Rhodospirillales, Gennataceae, Betaproteobacteria, Solirubrobacterales, Nitrospira, Syntrophobacterales* and *Pirellulales* were all positively sensitive to SNs concentration and DO, and negatively sensitive to residence time (Fig. 7). The positive correlation of SNs and DO also indicates that the hypothetical redox dependent degradation of SNs, revealing the possible important role of these species in biodegradation of SNs. *0319-6A21* was positive correlated to NO_3^-, while *Sinobacteraceae, Myxococcales* and *A4b* were more positively sensitive to residence time.

Fig. 4. Relative abundance of main bacteria taxa (>1% relative abundance, 100% occupancy) at the identified level before and after SNs stress (sample B1-1 represent RBF system B1 at the depth of 1 m).

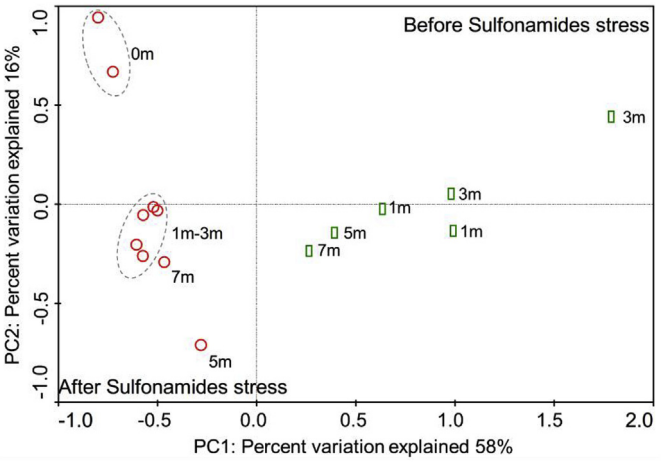

Fig. 5. PCA result of microbial taxonomy in B1 and B2 (Green rectangular and red dot represent sample collected before and after SNs addition respectively. X m represent the flow depth). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

3.4. Predicted metagenome functional profiling of microbial communities

In general, the community under sulfonamides pressure showed a significant higher metabolic potential in category of transporters, ABC transporters, Ribosome (difference in mean proportions < -0.1%), while the metabolic potential of secretion system, bacterial motility proteins and two-component system decreased (difference in mean proportions > 0.1%) under sulfonamides pressure (Fig. 8). Besides, difference in the residence time and the redox condition also contributed to the metabolic function evolution under SNs pressure. Proportion of transporters and ABC transporters significantly increased ($p < 0.05$) among the 20 highest average abundance KEGG pathways in RBF system B2 under SNs pressure (Fig. S3).

It was observed in the PCA result that samples before and after the addition of SNs clustered respectively along axis of principal components 1 (explain 50% of the variation), and with a distinguished separation of influent side (0 m) and other sample points along axis of principal components 2 (explain 26% of the variation), indicating a significant difference of related metabolic pathways influence by SNs pressure and residence time (Fig. 9). Gene *narH*, *nirD*, *nasA* relating to nitrogen metabolism and *folP* relating to SNs metabolism have significant differences before and after SNs addition ($p < 0.05$) (Table S12). Gene *nifD* and *nifK* involving nitrogen fixation and *nirA* involving NO_3^- reduction have a higher abundance in sample 0 m and longer residence time (with longer projection on each gene vector). The abundance of *narG, nirD, nirB* involving whole DNRA (Dissimilatory nitrate reduction to ammonium) process and *norB* involving nitric oxide to nitrous oxide step in denitrification decreased under SNs pressure.

4. Discussion

4.1. Impact of residence time and redox conditions on SNs degradation

The influence of sulfonamides sorption was found to be negligible in RBF systems, indicating a mobile behavior during soil passage processes (Bertelkamp et al., 2014). Besides, photolysis of OMPs is expected to be small and biotransformation was the main removal mechanism in RBF systems (Nödler et al., 2012; Bertelkamp, 2015). Therefore, we only considered the sulfonamides biodegradation in this study. RBF sites are characterized by different redox zones which are created by oxidation-reduction reactions during different residence time. Thus, the residence time essentially impacts redox condition and OMPs removal efficiency.

In this study, the removal ratio of SNs showed statistically significant difference between oxic-suboxic zones (RBF system B1) and the oxic-suboxic-anoxic zones (RBF system B2). All tested sulfonamides were removed more efficiently under anoxic condition than oxic condition, while the optimal redox condition for removal of sulfonamides and other persistent organic micropollutants is disputed in the literature. In a study, sulfamethoxazole, sulfamethazine and sulfapyridine were treated by a detrital aquifer from wastewater effluent, and the removal ratio under anoxic condition (>80%, DO < 1 mg/L) exceeded the removal ratio under oxic condition (around 50%, DO = 10 mg/L) (Candela et al., 2016). The removal of sulfamexthoxazole under Fe/Mn reducing

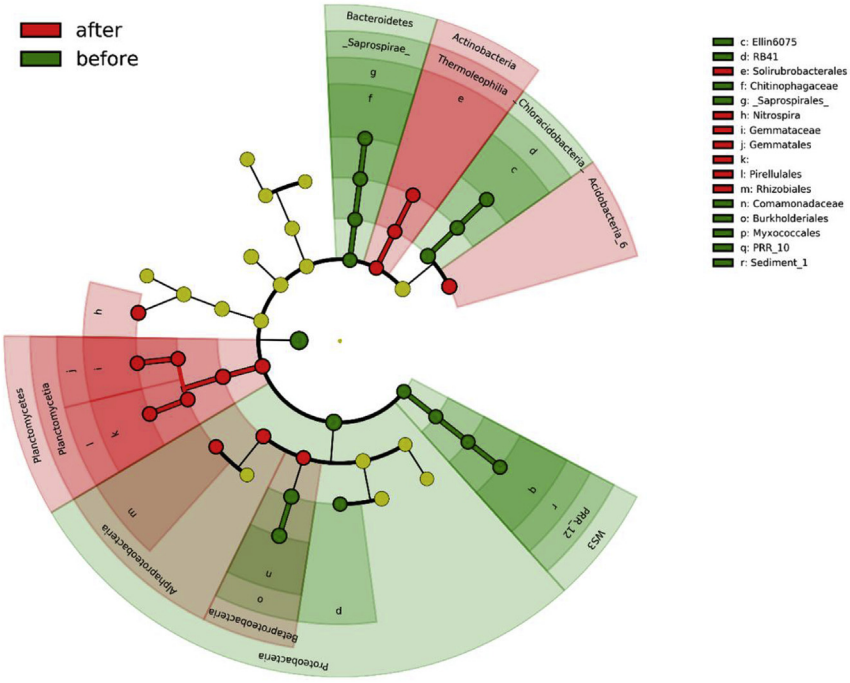

Fig. 6. LEfSe analysis results (The green and red circle represent the enriched taxa in community before and after sulfonamides pressure, yellow circles represent taxa with no significant differences between two communities). The phylum, class, order, family, and genus levels are listed from inside to outside. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

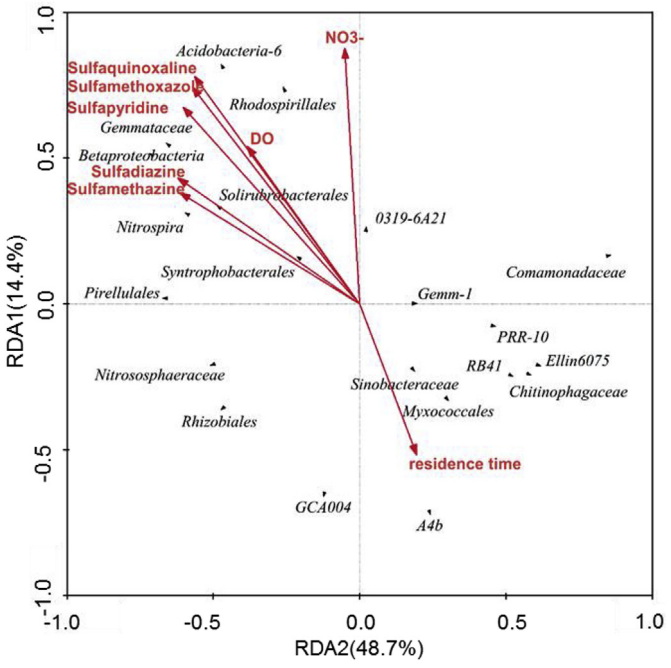

Fig. 7. Redundancy analysis of environmental factors with microbial species in RBF systems.

conditions (anoxic) was 74% and 89% respectively, much higher compared to O_2 (oxic, 41%) and NO_3^- reducing (suboxic) conditions, which could be partially attributed to a longer residence time and more effective removal processes (Wiese et al., 2011). Based on some field studies, it was also suggested that sulfonamides are more degradable under anoxic (Schmidt et al., 2004) and anoxic (Grunheid et al., 2005; Heberer et al., 2008; Jiménez Cisneros and Asano, 2008) conditions than under oxic conditions.

In contrast, some studies (Baumgarten et al., 2011b; Regnery et al., 2015, 2016) reported a higher removal efficiency under oxic conditions in RBF or related treatment processes. It should be pointed out that lab-scale artificial redox condition (N_2 purged) adopted in these studies might not well represent the real situation characterized by redox conditions formed by DOC attenuation and series of microbial assistant reactions (Baumgarten et al., 2011b; Abel et al., 2012; Hoppe-Jones et al., 2012). In this study, a naturally reduced redox condition was constructed and sulfonamides removal ratio was tested separately in two similar RBF systems in a column set-up to simulate the actual situation with subsequent oxic, suboxic and anoxic zones, therefore, the results in this study is convincing. The contradiction in removal ratio under different redox conditions might come from the differences in experimental conditions.

It was also noticed that the increase of start concentration of sulfamethoxazole (0.25 μg/L to 4.5 μg/L) could improve its removal ratio (60%–90%) under oxic condition (Baumgarten et al., 2011b), and it was hypothesized that sulfamethoxazole degradation would only take place above a threshold concentration (Gruenheid et al., 2008). At ng/L level of sulfonamides contamination, the removal ratio under anoxic condition was comparable with that under anoxic conditions in this study. To summarize, comparable contamination levels and similar redox conditions as in the field site are essential to obtain realistic results with lab-scale column experiments.

In conclusion, the studies focusing on the removal of SNs in RBF are limited, and have not come to a consistent conclusion about which redox condition is more beneficial and how much the impact can be. There are already former researcher to explore the pathways and genomic information of single sulfonamide degradation under fixed redox condition by specific bacterial strains (Deng et al., 2016). Further researches are needed to explain the chemical and biological basis and kinetics of sulfonamides degradation under

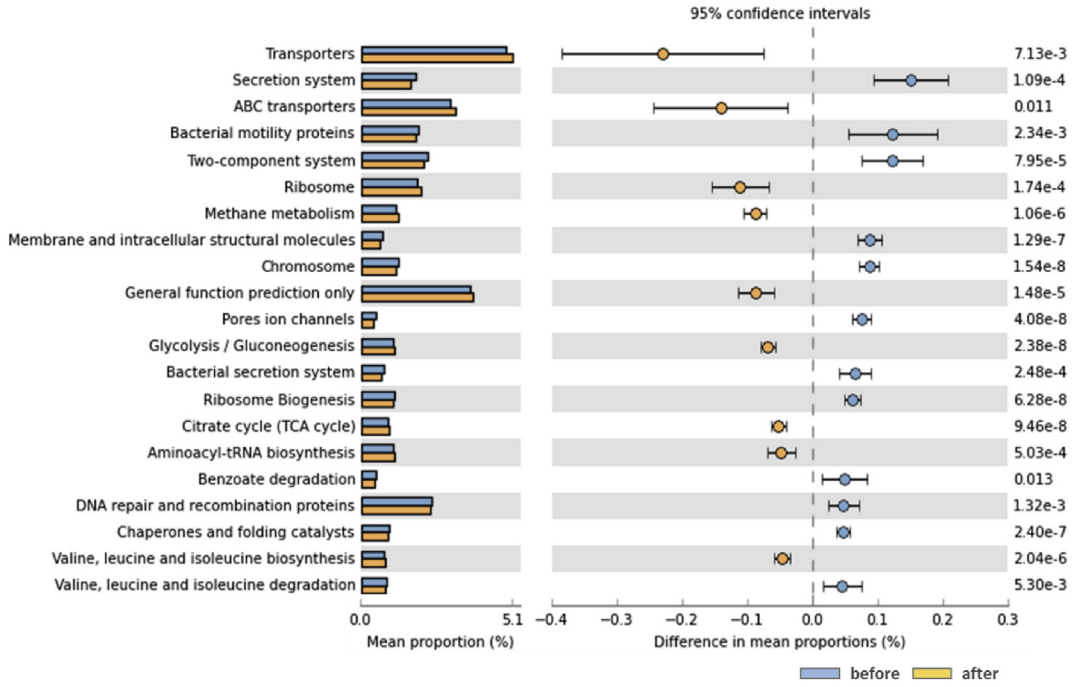

Fig. 8. Metabolism function proportion and diversity before and after SNs pressure ($p < 0.05$).

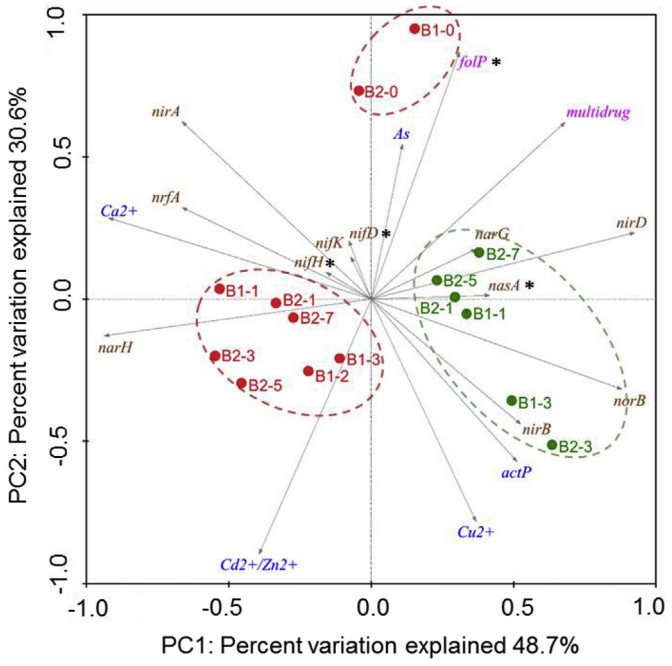

Fig. 9. PCA result of predicted functional genes related to nitrogen (brown), metal (blue) and sulfonamide (pink) metabolism and transport in RBF before (green dots) and after (red dots) sulfonamides stress. Relative abundance with significant difference were labeled with * ($p < 0.05$). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

specific environment, which will facilitate the conclusion drawing in more complex ones such as RBF system.

4.2. Microbial community response to sulfonamides stress in RBF systems

It was observed that more microbial species and a slightly higher community diversity were stimulated after the infiltration of SNs in RBF systems. Specially, the dominant phylum *Proteobacteria* decreased under sulfonamides stress from average 35.7%–25.1%, in which *Comamonadaceae, Oxalobacteraceae and Xanthomonadaceae* were with the highest abundance (Fig. 6). *Comamonadaceae*, a major member of *β-Proteobacteria*, are primary PHBV (solid bacterial polyester) degrading denitrifiers in activated sludge (Khan et al., 2002) and have the surfer transformation ability in rhizosphere communities (Schmalenberger et al., 2008). *Comamonadaceae Oxalobacteraceae and Xanthomonadaceae* were also found to decrease in soil after repeated application of manure spiked with 10 mg/kg sulfadiazine (Berg et al., 2014). Similarly, *Proteobacteria* (α, β and γ) and *Firmicutes* were reported to decrease in soil exposure to sulfamethazine contaminated pig manure (Marina Islas-Espinoza et al., 2012), which was in agreement with this study. This phenomenon indicates that the presence of SNs in soil microbial community might exert a negative effect on these taxa.

The relative abundance of *Bacillus* spp., frequently known as aerobic spore-bearers and with most of the species as saprophytes, was also noticed to increase after sulfonamides stress (Fig. 6, Table S10) ($p < 0.05$). *Bacillus* spp. endospores are not only resistant to hostile physical and chemical conditions, but also able to survive or thrive in harsh environments (Claus and Berkeley, 1986). It has also been previously reported as intrinsic sulfonamides resistant bacteria (Danyun Ou et al., 2015). This result suggests that the community in RBF system evolves to a more tolerant condition, and sulfonamide resistance can be developed in specific bacteria under considerable contamination pressure in RBF system.

One of the dominant phylum *Planctomyces* reported to be capable for anaerobic ammonium oxidation (Anammox), was also found more abundant (5.7%–9.3%) in the sulfonamides stressed community, which showed a decreasing trend with increasing residence time (Table S10). Anammox is a process found in wastewater treatment systems and rarely observed in subsurface environments (Kuenen, 2008). In consistence, detected anammox bacteria *Candidatus Brocadia* spp. and *Candidatus Jettenia* spp. (Shu et al., 2016) in this RBF system were with very low abundance

(<0.05%), indicating anammox not as the dominant ammonia oxidation process.

4.3. Impact of contaminants pressure on microbial metabolic pathways in RBF systems

In general, the community under sulfonamides stress showed a higher metabolic potential indicated by the higher relative abundance of metabolic pathways. Analysis between communities with and without sulfonamides contaminants suggested that both were primarily heterotrophic (with relative abundance of carbon fixation in prokaryotes around two times of that in photosynthetic organisms). The carbohydrate metabolic pathways and carbon fixation in the community without contaminants went through an increasing trend from oxic zones (1 m) to anoxic zones (7 m) (subtotal relative abundance of carbohydrate metabolic pathways from 9.97% to 12.36%, and carbon fixation pathways from 1.24% to 1.55%), which suggested the development of a more robust community and more active metabolic function along with the enhanced reduction condition (Table S11). Since sulfonamides pressure was added to the RBF system for 5 month, the heterotrophic carbon fixation relative abundance increased from 1.23% to 1.48% at 1 m, 1.28%–1.66% at 3 m, 1.32%–2.09% at 5 m, 1.55%–1.67% at 7 m respectively. In contrast, the relative abundance of carbohydrate metabolic pathways decreased significantly in the first meter from 16.44% to 12.06% in RBF system B1 and 17.01%–13.29% in RBF system B2. In RBF system B2 the relative abundance of carbohydrate metabolic pathways peaked at 5 m (16.85%) and then dropped back to about the same value as that at 1 m from the effluent site (13.43%). This indicates a deterioration of carbohydrate metabolism in the sulfonamides stressed microbial community.

Since DOC concentration is usually a few mg/L and organic pollutants are present in concentrations six orders of magnitude lower (ng/L), co-metabolic degradation of organic pollutants is the more likely mechanism in RBF as suggested in a number of studies (Rauch-Williams et al., 2010; Maeng et al., 2011; Alidina et al., 2014). The relative abundance of organic pollutants degradation related pathways in the original community was 2.02% (1 m flow depth) to 2.51% (7 m flow depth), and 2.40% (1 m flow depth) to 3.26% (0 m flow depth) in the sulfonamides stressed community.

In this research, among the KEGG pathways categories that significantly increased after sulfonamides stress, mean proportions of transporters and ABC transports before and after sulfonamides addition showed the highest differences (>0.1%, $p < 0.05$) (Fig. 8), which suggests the activated transport of a wide variety of substrates through the microbe cell membrane in unfavorable environment, including ions, sugars, lipids, sterols, peptides, proteins, and drugs (Table S11). The high number of different transport systems facilitates the acquisition of a broad range of substrate categories, which suggests an advantage in complex environments and adaptation to oligotrophic conditions (Kielak et al., 2016). This is an evidence of the inferred motivated substrate consumption efficiency and detoxification mechanism of a contaminants stressed microbial community (Giovanna Ferro, 1986). Further evidence can be found in the shift of nitrogen metabolism and transport pathways, as well as sulfonamides metabolism pathways before and after sulfonamides stress (Table S12).

Sulfonamide resistance in some bacteria is associated with mutations in the chromosomal gene *folP*, which encodes dihydropteroate synthase (Fiebelkorn et al., 2005). In this research, the predicted relative abundance of *folP* gene showed a significant increasing trend under sulfonamides stress, indicating the resistance potential in the microbial community of RBF system.

5. Conclusions

- In the present study, the removal behavior of 5 sulfonamides at ng/L level was observed continuously for 6 months. A more degradable behavior under anoxic condition and longer flow path was observed.
- Under long-term non-lethal selective pressures of sulfonamides, microbial community structure in RBF systems changed significantly and the phylogenic diversity increased slightly. A more diverse, tolerant and robustness co-metabolic function of bacterial community were observed. A detoxification and an ecological self-update characteristic of microbial ecological communities were developed in RBF systems indicated by an activated transport of a wide variety of substrates through the microbe cell membrane. The predicted relative abundance of *folP* gene increased, and resistant strains were found, indicating an elevated sulfonamides resistance potential of the microbial community in RBF system.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2018.12.167.

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