Soil Biology & Biochemistry 78 (2014) 307–315

Contents lists available at ScienceDirect

Soil Biology & Biochemistry

journal homepage: www.elsevier.com/locate/soilbio

Influence of redox conditions on the microbial degradation of polychlorinated biphenyls in different niches of rice paddy fields

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ARTICLE INFO

Article history:
Received 17 March 2014
Received in revised form
14 August 2014
Accepted 16 August 2014
Available online 6 September 2014

Keywords:
Paddy field
Polychlorinated biphenyls (PCBs)
Microzones
Phospholipid fatty acids (PLFAs)
Biodegradation
Bio-dechlorination
Rice (*Oryza sativa*)

ABSTRACT

The biotransformation of PCBs in three niches with contrasting redox conditions (water–soil interface, rhizosphere and non-rhizosphere subsoil) was studied using rhizoboxes subjected to the sequential flooding and draining conditions associated with rice cultivation in paddy fields. Rice cultivation favored PCB dechlorination and further transformation of the degradation products. Microbial dechlorination activity not only began earlier but was also 50% greater in the rhizosphere in comparison with bulk soil. Distinct profiles of both phospholipid fatty acids (PLFAs) and PCB congeners arose in each niche. Signature PLFAs attributed to Gram-positive (G+) bacteria were significantly correlated with the dissipation of PCB congeners in paddy field soils, especially in the rhizosphere.

1. Introduction

Polychlorinated biphenyls (PCBs) are a family of 209 congeners in which chlorine atoms are attached in different positions to biphenyl. Due to their low water solubility and high octanol–water partition coefficients (Hawker and Connel, 1988), PCBs adsorb strongly to organic materials and are therefore associated with solid phases, rather than the aqueous phase, in soils and aquatic systems. PCBs also readily partition into lipids; consequently, within organisms they are found in fatty storage tissues characterized by a slow metabolism rate (Jones and de Voogt, 1999). In addition to persisting within biota, PCBs also bioaccumulate, becoming concentrated in the higher trophic levels of food webs where they have many harmful effects, including effects on human health (Borja et al., 2005). The release of not just PCBs but also heavy metals and other persistent organic pollutants to the environment during the storage and crude recycling of waste electrical equipment (i.e., capacitors and transformers) is a serious environmental problem in China (Yang et al., 2013; Zhang et al., 2014). One of the largest centers for the disposal of electrical equipment occupies more than 17.8 km^2 and is located in the city of Taizhou. For nearly 35 years, electrical equipment has been transported to numerous recycling sites near farmhouses, farmlands or riversides (Tang et al., 2010), resulting in the contamination of soil by PCBs (Shen et al., 2008, 2009; Zhang et al., 2014). Although the concentration of PCBs is comparatively low, pollution is widespread as a consequence of direct input from household workshops and indirect input from atmospheric deposition, as well as polluted irrigation water (Tang et al., 2010). Accordingly, the behavior and environmental fate of PCBs in farmland are of special significance in terms of food safety and human health.

The natural attenuation rate of PCBs determined in the field is usually 1 to 2 orders of magnitude lower than values determined in a laboratory setting, which is most likely due to the lower concentrations and more limited bioavailability of aged pollutants in the field (Gomes et al., 2013). Microbial degradation of PCBs is known to occur via two main routes: anaerobic and aerobic. Highly chlorinated PCB congeners can be dechlorinated under anaerobic conditions to form less chlorinated congeners, which are more susceptible to aerobic degradation (Abramowicz, 1995; Furukawa and Fujihara, 2008). Because complete mineralization of PCB congeners is limited in many environments, researchers have proposed

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http://dx.doi.org/10.1016/j.soilbio.2014.08.014
0038-0717/

the coupling of two or more processes, such as sequential exposure to anaerobic and aerobic conditions, to increase the efficiency of remediation strategies (Master et al., 2001; Meade and D'Angelo, 2005). For example, compared to no net PCB loss in either aerobic or anaerobic treatments, Master et al. (2001) observed a significant decrease in the amount of PCB residues in soils after sequential anaerobic–aerobic treatment during the same time period.

The paddy field system of farming wetlands, whereby anoxic conditions prevail during the period of plant growth and oxic conditions prevail during the fallow period, accounts for a large proportion of the farmland in our study area in Taizhou (Liesack et al., 2000; Tang et al., 2010). During the rice growing season, O_2 diffusion/advection through the water column and O_2 transport through above-ground plant tissues into the subsoil leads to the formation of adjacent aerobic and anaerobic niches at the soil–water interface and in the rhizosphere (Brune et al., 2000). It has been previously demonstrated that many biogeochemical processes are accelerated by the prevalence of adjacent aerobic and anaerobic niches in wetlands (Reddy et al., 1989). Beyond that, a greater extent of readily available carbon in the form of root exudation, as well as ameliorated Eh, pH and aeration conditions in the rice rhizosphere, is also likely to promote the transformation of PCBs (Walker et al., 2003). In particular, it should be noted that based on the log K_{ow} and water solubility of PCB congeners, significant uptake by plants and translocation in the transpiration stream is unlikely (Liu and Schnoor, 2008; Liu et al., 2009), especially for rice (Bi et al., 2002). Therefore, the cultivation of rice in paddy fields could favor the degradation of PCBs with minimal ecological risk.

To date, the great potential of paddy field conditions in general and the specific redox conditions of distinct niches to augment the natural attenuation of PCBs have rarely been studied (Baba et al., 2007). Furthermore, little information is available about the impacts of micro-environmental variances on soil microbial biomass and communities responsible for PCB attenuation in paddy fields. In a previous study, we compared the differences in the characteristics of PCB dissipation between rice-planted and non-planted paddy soil (Chen et al., 2014). Hence, the aim of this study was to investigate the planted system further and determine the differences in PCB dissipation in different niches during the remediation process of a paddy soil contaminated with weathered PCBs. It was hypothesized that sequential reductive dechlorination-aerobic degradation of PCBs would be accelerated in the rice rhizosphere with established anoxic-oxic microzones and nutrition supplements in the form of exudates. Niche-specific differences in PCB transformation dynamics would also be associated with changes in the microbial community size and composition. The results from this study will improve our knowledge regarding the fate and behavior of PCBs in farmland and guide further work in choosing cultivation patterns that could reduce the risk of pollution.

2. Materials and methods

2.1. Rhizobox experiment

For the experiments, we used the Xiuyou 5 variety of rice seed (*Oryza sativa*). Seeds were surface-sterilized with a 10% (v/v) hydrogen peroxide solution for 30 min and germinated on moist non-woven gauze in Petri dishes before transfer to a rhizobox that contained 2.0 kg soil and two seedlings per box.

Soil contaminated with a mixture of commercial PCBs (0–20 cm depth) was collected from a cultivated paddy field close to the waste electrical equipment recycling operations of Taizhou. According to particle size analysis, the soil had a silty sandy loam texture. Soil pH and total organic C were 6.85 and 2.86 g kg^{-1}, respectively.

Each rhizobox was split into three compartments. The middle compartment (30 mm wide) contained the roots of the rice plants, and on either side there were 55 mm wide compartments containing soil only. The overall size of the individual rhizoboxes was 140 mm × 140 mm × 200 mm (length × width × height). Rhizosphere soil was considered to be the 5-mm zones of soil in the outer two compartments that were adjacent to the middle compartment. Non-rhizosphere soil was the soil in the outermost 5 mm of the rhizobox. Surface soil, at the water–soil interface, was the uppermost 1–2 cm of soil (Fig. S1). The rhizoboxes were placed at random on the same bench in a greenhouse with temperature control (25 °C) at 60% relative humidity and with additional illumination (with a light intensity of 250 μmol m^{-2} s^{-1}, under a 14/10 h-light/dark cycle).

To imitate the typical cycle of flooding and drainage associated with rice production, sufficient water was added to the rhizobox to maintain a water layer of 5 cm above the soil surface, and no mineral nutrients were added to the soil throughout the experimental period. The plants were harvested after 180 days. The shoots were removed by cutting above the soil surface, and the soil moisture content was then adjusted to 60% gravimetric water content for another 120 days. The soil for sterilized treatment was autoclaved for 30 min (121 °C, 15 psi). To make the soil completely sterile, autoclaving was repeated twice over the next 2 days. The sterilized control was exposed to the same water management. Three replicates were performed for each treatment.

Samples of the rhizosphere and the bulk soil were collected near the bottom of the rhizobox using a sterile cork borer. Soil samples were taken 50, 100 and 180 days after seedling transplantation and 60 and 120 days after rice harvest to correspond with three rice growth stages: tillering, booting and maturation. After harvest, the soil was allowed to dry, with the first 60 days being referred to as drying stage I and the subsequent 60-day period as drying stage II. Soil samples were freeze-dried overnight and stored at 4 °C prior to further chemical analyses.

2.2. Chemical analyses

For PCB analysis, 5.0 g soil was extracted in a Soxhlet apparatus for 24 h with 250 mL of a hexane-acetone (1:1, v/v) mixture. The extracted solutions were subsequently concentrated to 4–5 mL by a rotary evaporator. The extracts were passed through a column packed with layers of florisil and anhydrous sodium sulfate. Each eluate was evaporated to 1 mL prior to further analysis. Congener-specific PCB analysis was performed on an Agilent 7890 gas chromatograph (Agilent, USA) equipped with a ^{63}Ni electron-capture detector (Shen et al., 2009). The elution time of PCB congeners in contaminated soil was determined with PCB mixtures of Aroclor 1242 and Aroclor 1254 from AccuStandard (Frame et al., 1996). All procedures were rigorously monitored for quality control in order to meet USEPA requirements. PCB209 was spiked to determine the recovery efficiency. The recovery efficiencies for surrogate standards ranged between 75 and 105%, so the final concentrations of the compounds of interest were not corrected according to recovery efficiency.

2.3. PLFA analysis

The PLFAs were extracted from freeze-dried soils (3.0 g) and separated from other fractions by a solid-phase column that was described fully by Ding et al. (2009). In brief, the total extractable lipids were recovered in chloroform and then fractionated on silica-bonded phase columns (SPE-Si, Supelco, Poole, UK) into neutral,