Science of the Total Environment 637–638 (2018) 50–57

Contents lists available at ScienceDirect

Science of the Total Environment

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

Environmental behaviour of polychlorinated biphenyls in a paddy field: Impact factors and canopy effects

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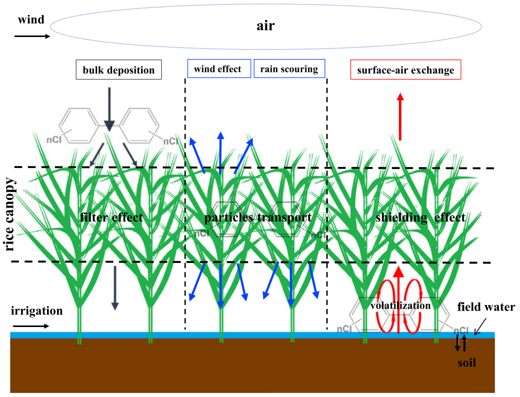
HIGHLIGHTS

- PCB bulk deposition fluxes at top and bottom of canopy presented slight variations.
- PCBs associated with particles showed slightly higher proportions in deposition at top of canopy.
- Atmospheric PCB concentrations at bottom of canopy were higher than top.
- Rice canopy has potential effects on the transport of PCBs.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 27 December 2017
Received in revised form 22 April 2018
Accepted 22 April 2018
Available online xxxx

Editor: Adrian Covaci

Keywords:
Polychlorinated biphenyls
Paddy field
Impact factor
Filter effect
Shielding effect

ABSTRACT

Paddy fields play an important role in the transport of persistent organic pollutants (POPs) due to the filter effects of canopy and their wide distribution. Thus, most studies have been focusing on the filter effects of canopy for POPs. However, shielding effects of canopy might also influence transport and portion of POPs between top and bottom. To investigate these two important processes, our study involved 30 polychlorinated biphenyls (PCBs) in a paddy field. Samples of bulk depositions, surface water, and air were taken to investigate the occurrence and the behaviour of PCBs. We found that rice canopy has potentially crucial effects on the transport of PCBs. The results showed slightly higher abundances for most of high chlorine PCBs (81.0%) at the top of the canopy, indicating that the high chlorine PCBs were intercepted by the rice leaves. Moreover, our study showed that the PCBs in surface water and soil tended to escape into air according to air, water, and soil fugacity. And we found higher atmospheric PCB levels (103 pg m^{-3}) at the bottom of the canopy than top (88.9 pg m^{-3}), indicating canopy shielding effects on escaped PCBs. In addition, the study showed that the PCBs intercepted by the rice canopy may occur in surface water and soil due to air movement and precipitation. These results suggest that paddy fields can enrich POPs, and effects of the environmental factors on POPs transport need to be investigated further.

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https://doi.org/10.1016/j.scitotenv.2018.04.319
0048-9697/Published by Elsevier B.V.

1. Introduction

The fate and transport of persistent organic pollutants (POPs) have drawn continuous scientific interests since they are wide spread in remote areas, particularly, in Arctic and Antarctic regions (Tatton and Ruzicka, 1967). Due to their volatility and high persistence, POPs can be transferred among atmospheric, hydrosphere, and terrestrial environments via physical processes, such as volatilisation and deposition (Cabrerizo et al., 2011; Jones and de Voogt, 1999; Tatton and Ruzicka, 1967; Wania and Mackay, 1993), indicating that soil, water, and atmosphere can be sources or sinks for POPs. Transfer of POPs between different phases might also influence their environmental fate at regional and global scales (Jones and de Voogt, 1999). Polychlorinated biphenyls (PCBs), one important class of POPs, have been used widely for different purposes, such as transformers, capacitors, sealing materials, and paints (Klees et al., 2017). Due to their extensive application (1.324 million tons between 1930 and 1993) as well as the high persistency (Breivik et al., 2002; Nellier et al., 2015), PCBs were still detected very high concentrations in environmental compartments (dust: 19–330 mg/kg; spruce needles: 20–535 μg/kg; Moscow river: 180.7 ng/L) although they have been banned for three decades (Eremina et al., 2016; Klees et al., 2017). Therefore, understanding PCBs environmental behaviour is of primary importance.

Plant ecosystems play an important role in distribution, transport, and fate of semi-volatile organic compounds (SVOCs) (Terzaghi et al., 2013). Due to the fact that plant ecosystems cover large areas, and the canopy has the high biomass content and carbon turnover rates, they might significantly influence the partition of POPs between air and land surfaces (Nizzetto et al., 2008). Several studies have examined canopy effects, such as forest filter effects and air–canopy exchange on different organic pollutants (Liu et al., 2014; McLachlan and Horstmann, 1998; Nizzetto and Perlinger, 2012; Staelens et al., 2008; Su et al., 2007; Terzaghi et al., 2013). Specifically, Su and Su and Wania (2005) reported that the forest filter effect may reduce PCB concentrations in air, ocean, and freshwater at the expense of increased concentrations in forest soils. Nizzetto et al. (2006) found that forest structures could influence PCB deposition in soil. Schaubroeck et al. (2014) studied the behaviours of particulate matter on plant surfaces. However, there is no study focusing on canopy effects on POPs transport at the top and bottom of canopy, which can influence occurrence and fate of POP in plant ecosystem.

Paddy fields also have canopy effects on POPs, since the leaves can filter or capture atmospheric POPs (Wang et al., 2015b). Specially, rice fields are regularly planted in dense, and they are short plants, reducing air exchange between the top and bottom of the canopy. In addition, flooding of paddy fields causes variations on temperature, and influences the volatilisation and deposition of POPs. Since the rice fields cover a large cultivation area and represent the important source for consumed staple food, have received enormous attention recently (Zhu et al., 2017). Several recent studies have studied paddy fields (Gao et al., 2010; Li et al., 2009; Luo et al., 2012; Ma et al., 2012), but most studies have focused on the remediation of the contaminated soils and few have been concerned with the environmental behaviour of pollutants in paddy ecosystems (Li et al., 2017; Lu et al., 2012; Tiwari et al., 2017). For instance, Wang et al. found rice canopy can act like a chamber to influence the partitioning of polycyclic aromatic hydrocarbons (Wang et al., 2015a), and they also reported that the rice cultivation has complex effects on environmental fate of polycyclic aromatic hydrocarbons in paddy ecosystems (Wang et al., 2015b). Li et al. (2017) found that atmospheric PCBs in the paddy field may be from Dongguan, Qingyuan, and Conghua.

China is the largest producer of rice in the world (Peng et al., 2009), accounting for ~19% of world's rice production. In Guangzhou, a typical subtropical city in China, growing of two rice crops a year could be achieved. The relatively high annual mean temperature in the subtropical region results in the frequent transport of pollutants, including volatilisation and deposition, which plays an important role in the fate and behaviour of pollutants (Wania and MacKay, 1996). This study was carried out on farmland in Guangzhou. Air, water, and bulk deposition samples were collected in an experimental paddy field. The purpose was to study the occurrence and the environmental behaviour of PCBs, and canopy effects on transport of PCBs in a paddy field.

2. Materials and methods

2.1. Sample collection

The experiment was conducted on a suburban farmland in Guangzhou, southern China (23°9′59″ N, 113°22′7″ E), which yields two rice crops annually. Detailed site information in previous research can be found in Wang et al. (2015a). Bulk deposition ($n = 54$), field water ($n = 4$) and irrigation water ($n = 4$), and air ($n = 10$) samples were collected during different periods in two seasons from May to December 2012 (specific periods were shown in Table S1). The bulk deposition and air samples were collected at the top and bottom of rice canopy, and the water samples were collected from the surface of field water and outfall of irrigation water. More sampling information is given in S1.1.

2.2. Extraction and analysis

The extraction protocols for target compounds are described in S1.2. Three selected surrogate standards were used including PCB-30, −198, and − 209. Analyses of 30 PCBs (PCB-28, −37, −52, −44, −74, −70, −66, −60, −77, −101, −99, −87, −82, −118, −114, −105, −126, −153, −138, 158, −166, −128, −156, −169, −179, −187, −183, −180, −170, and − 189) were performed using an Agilent 7890GC-5975MS with a BD5-MS column (30 m × 0.25 mm × 0.25 μm; Agilent Technologies Inc., USA). The temperature programme was as follows: hold at 150 °C for 3 min, then increase to 278 °C at a rate of 4 °C min^{-1} and hold for 5 min. A splitless injection mode was used with an injection volume of 1 μL. The congeners of PCBs were identified in the light of three fragment ions with electron impact spectrometry in the selected ion monitoring mode. The mass selective detector source temperature was 230 °C and the quadruple temperature was 150 °C.

2.3. Quality assurance and quality control

We included three parallel samples for bulk deposition and two parallel samples for water during each sampling period, field, and procedural blanks to represent potential contamination. The target compounds were not detected in the blanks. The method detection limits for PCBs were 0.004–0.02 ng/L for water samples, 0.30–6.2 pg g^{-1} for particles, and 0.18–2.15 pg m^{-3} for air samples. The average recoveries from all samples were 86.1 ± 14.5%, 91.5 ± 11.4%, and 92.1 ± 10.3%, respectively, and the recoveries for three surrogate standards are shown in Table S2. Reported values are not corrected for recovery rates.

2.4. Fugacity calculations

Fugacity model is used frequently to measure potential compound escape from one phase to another. In this study, we calculated the fugacity fractions of PCB congeners to describe the trends in air–water and air–soil exchange during flooding periods and dry periods, respectively. The fugacity of water was based on PCB concentrations in field water during flooding periods in two seasons; the fugacity of soil was based on PCBs in soil samples during dry periods, according to previous research (Li et al., 2015); and the fugacity of air was based on atmospheric PCBs collected by passive sampling at the bottom of the canopy in two seasons. According to previous studies (Li et al., 2010; Pandit et al.,