



# The effects of rice canopy on the air–soil exchange of polycyclic aromatic hydrocarbons and organochlorine pesticides using paired passive air samplers



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## ABSTRACT

The rice canopy in paddy fields can influence the air–soil exchange of organic chemicals. We used paired passive air samplers to assess the exchange of polycyclic aromatic hydrocarbons (PAHs) and organochlorine pesticides (OCPs) in a paddy field, South China. Levels of OCPs and light PAHs were generally higher under the canopy than above it. We found that the rice canopy can physically obstruct the evaporation of most OCPs and light PAHs, and can also act as a barrier to the gaseous deposition of *p,p'*-DDT and heavy PAHs. Paddy fields can behave as a secondary source of OCPs and light PAHs. The homolog patterns of these two types of chemical varied slightly between the air below and above the rice canopy, implying contributions of different sources. Paired passive air samplers can be used effectively to assess the *in situ* air–soil exchange of PAHs and OCPs in subtropical paddy fields.

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## 1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) and organochlorine pesticides (OCPs) are groups of semi-volatile organic compounds (SVOCs) which are of great environmental concern for their persistence, toxicity and bioaccumulation. Airborne SVOCs can participate in air–surface exchange with various matrices including soil, water and vegetation (Nizzetto et al., 2014), which may act as a sink or a reservoir for those compounds in the environment. Understanding these exchange processes is crucial for pollution control and risk management.

The environmental fate of SVOCs can be significantly influenced by vegetation via root exudate dissolution, root uptake, degradation, foliage adsorption and capture (Wania and McLachlan, 2001; Wegmann et al., 2004; Wang et al., 2014). Plant uptake and degradation of SVOCs have been well characterized, especially for

vegetables (Mueller et al., 2006; Huang et al., 2010) and crops (Lin et al., 2007; Su and Zhu, 2008). Plants can also act as filters of atmospheric SVOCs due to the efficient capture of both the vapor and particle phases (Nizzetto et al., 2006; Su et al., 2007; Choi et al., 2008; Li et al., 2009; Moeckel et al., 2009b). However, this “filter effect” has been well characterized only in forest ecosystems (Terzaghi et al., 2013). First, forest canopies can absorb vapor-phase SVOCs through the partitioning with air due to the extensive organic surface of the leaves. Choi et al. (2008) observed the rapid uptake of gaseous PAHs by a forest canopy, especially during bud break in early spring. Second, forests can sequester particle-bound SVOCs via foliage capture, but, because only a small proportion of the low molecular weight SVOCs is particle-bound in the air, aerosol deposition onto the canopy is assumed to be negligible (Horstmann and McLachlan, 1998). Thus, gaseous partitioning is more important for the accumulation of SVOCs by vegetation. Plant leaves can also form a dense canopy, which may obstruct the convection and diffusion of SVOCs between the atmosphere and soil. However, there is still a lack of information regarding the physical obstruction of the air–soil exchange of SVOCs by the plant

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canopy, especially in farmland.

Rice is one of the most widely cultivated and highly produced crops in the world, and has attracted considerable attention for its food safety aspects (e.g., uptake of toxic elements and organic chemicals), as well as the effects of its unique flooding conditions on contaminant transformation (Wang et al., 2015). Compact canopies formed by the high density of rice leaves can impede the convection and diffusive exchange of SVOCs in paddy fields from the elongation stage to the rice harvest. Very few studies have been conducted on the “blocking effect” of rice canopy, probably due to the lack of appropriate sampling methodologies.

Polyurethane foam passive air samplers (PUF-PAS) have been deployed at various geographic scales to assess the air concentration and distribution of SVOCs (Harner et al., 2004; Pozo et al., 2006; Hogarh et al., 2012). PUF-PAS can enable a power-free and cost-effective collection of seasonally integrated air sample, and thus, it can reveal the average concentrations and long-term trends of SVOCs in the atmosphere, which are useful for long-term air quality assessment. We deployed pairs of PASs below and above the rice canopy, respectively. The purpose of this research was to investigate the long-term influence of the rice canopy on the air–soil exchange of selected PAHs and OCPs using *in situ* paired passive samplers in a subtropical paddy field, and to improve our understanding of the effects of rice growth on the environmental fate of these organic compounds.

## 2. Materials and methods

### 2.1. Sample collection

The studied paddy field was located in a small farm in a suburban area of Guangzhou City [23°9′59″N, 113°22′7″E], Guangdong Province, South China, where two crops of rice, of different varieties, are cultivated annually. Pre-cleaned PUF disks (diameter, 14.0 cm; thickness, 1.30 cm; mass, 3.40 g; density, 0.0170 g/cm<sup>3</sup>) were spiked with depuration compounds (DCs, <sup>13</sup>C-PCB28, 52, 101, 138, 153, 180 and 209) 3 days before deployment (Moeckel et al., 2009a; Wang et al., 2012). Paired passive air samplers were deployed simultaneously directly under and above the rice canopy (Fig. 1), from the jointing stage until the rice harvest for both growing seasons (first season: May 29 to July 9, 2012; second

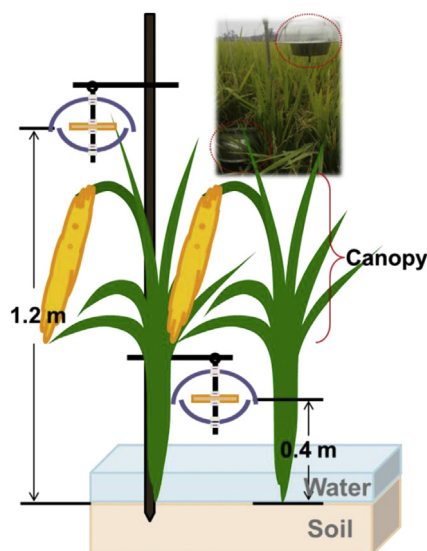


Fig. 1. The details of the paired passive air samplers.

season: September 25 to November 8, 2012). Within each growing season, three duplicated paired-samplers were deployed at an interval of >2 m to reduce random errors. After deployment, the PUF disks were wrapped in aluminum foil, placed in polyethylene Zip-lock bags and stored at −20 °C until analysis.

### 2.2. Sample extraction and analysis

PUF samples were spiked with surrogate standards (naphthalene-D<sub>8</sub>, acenaphthene-D<sub>10</sub>, phenanthrene-D<sub>10</sub>, chrysene-D<sub>12</sub>, perylene-D<sub>12</sub>, TCMX, PCB30, PCB198 and PCB209) and then Soxhlet extracted with dichloromethane (DCM) and activated copper for 24 h. Following solvent exchange with hexane, the extract was first cleaned by passing through a multilayer silica gel/alumina column containing anhydrous sodium sulfate, neutral silica gel (3% deactivated) and neutral alumina (3% deactivated) from the top to the bottom using an eluent of 20 mL hexane/DCM (1:1, v/v) for PAH analysis. The extract was further purified on a column containing 50% (w/w) sulfuric acid–silica gel and neutral silica gel (3% deactivated) using an eluent of 15-mL hexane/DCM (1:1, v/v) for PCB and OCP analysis.

An Agilent 7890GC-5975MS equipped with a DB5-MS capillary column (30 m × 0.25 mm × 0.25 μm) was used for the analysis of 13 PAHs, including naphthalene (NAP), acenaphthylene (ACY), acenaphthene (ACE), fluorene (FLU), phenanthrene (PHE), anthracene (ANT), fluoranthene (FLA), pyrene (PYR), benzo[a]anthracene (BaA), chrysene (CHR), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF) and benzo[a]pyrene (BaP). An Agilent 7890GC-7000A triple-quadrupole mass spectrometer equipped with a CP-Sil 8CB capillary column (30 m × 0.25 mm × 0.25 μm) was used for the determination of the seven <sup>13</sup>C labeled PCBs (DCs), five hexachlorocyclohexanes (HCHs, including α-HCH, β-HCH, γ-HCH, δ-HCH and ε-HCH) and six DDTs (including *o,p'*-DDE, *p,p'*-DDE, *o,p'*-DDD, *p,p'*-DDD, *o,p'*-DDT and *p,p'*-DDT). Helium was used as the carrier gas at 1 mL/min. The initial oven temperature was set at 60 °C for 1 min, raised to 290 °C at the rate of 4 °C/min.

### 2.3. QA/QC

Procedural and field blanks were run with the PUF samples to assess potential contamination during deployment and analysis. Except NAP (8–10 ng/PUF), no target compounds were detected in the blanks. The average recoveries of the surrogate NAP-D<sub>8</sub>, ACE-D<sub>10</sub>, PHE-D<sub>10</sub>, CHR-D<sub>12</sub>, perylene-D<sub>12</sub>, TCMX, PCB30, PCB198 and PCB209 in the PUF samples were 66 ± 17%, 75 ± 12%, 94 ± 7%, 102 ± 12%, 96 ± 18%, 71 ± 15%, 81 ± 9%, 101 ± 14% and 97 ± 16%, respectively. The reported results in this paper were corrected by the blanks, but not by the surrogate recoveries.

### 2.4. Sampling volume calculation

Calculation of sampling rate (*R*) and effective sampling volume (*V*<sub>eff</sub>) was conducted according to the method established by Moeckel et al. (2009a), using a stable DC<sub>Stable</sub>, that can barely volatilize from the PUF to correct for the recoveries of DCs:

$$R = \frac{-\ln(\epsilon') \cdot K_{PUF-A} \cdot \rho_{PUF} \cdot V_{PUF}}{t} \quad (1)$$

$$\begin{aligned} V_{eff} &= K_{PUF-A} \cdot \rho_{PUF} \cdot V_{PUF} \cdot \left( 1 - \exp \left[ \frac{-R \cdot t}{K_{PUF-A} \cdot \rho_{PUF} \cdot V_{PUF}} \right] \right) \\ &= K_{PUF-A} \cdot \rho_{PUF} \cdot V_{PUF} \cdot (1 - \epsilon') \end{aligned} \quad (2)$$

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