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Short Communication

Distribution of polychlorinated biphenyls in an urban riparian zone affected by wastewater treatment plant effluent and the transfer to terrestrial compartment by invertebrates

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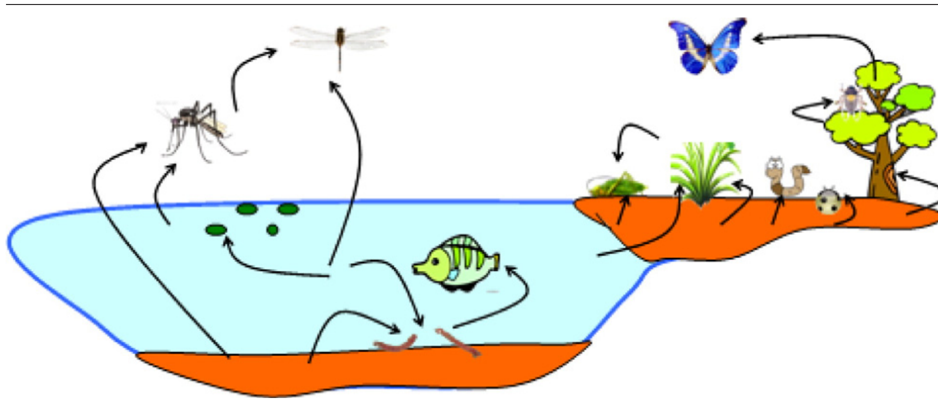
HIGHLIGHTS

- The distribution of PCBs in an urban riparian zone around a wastewater effluent affected river was investigated.
- Relatively high abundances of PCB-11 and PCB-28 were found for most samples.
- Mid-chlorinated congeners (PCB-153 and PCB-138) were more accumulated in chironomids and dragonflies as well as soil dwelling invertebrates.
- Emerging invertebrates can carry waterborne PCBs to the terrestrial compartment.
- The estimated annual flux of PCBs from emerging chironomids ranged from 0.66 to 265ngm−2·y−1.

GRAPHICAL ABSTRACT



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ABSTRACT

In this study, we investigated the distribution of polychlorinated biphenyls (PCBs) in a riparian zone affected by the effluent from a wastewater treatment plant (WWTP). River water, sediment, aquatic invertebrates and samples from the surrounding terrestrial compartment such as soil, reed plants and several land based invertebrates were collected. A relatively narrow range of $\delta^{13}C$ values was found among most invertebrates (except butterflies, grasshoppers), indicating a similar energy source. The highest concentration of total PCBs was observed in zooplankton (151.1 ng/g lipid weight), and soil dwelling invertebrates showed higher concentrations than phytophagous insects at the riparian zone. The endobenthic oligochaete Tubifex tubifex (54.28 ng/g lw) might be a useful bioindicator of WWTP derived PCBs contamination. High bioaccumulation factors (BAFs) were observed in collected aquatic invertebrates, although the biota-sediment/soil accumulation factors (BSAF) remained relatively low. Emerging aquatic insects such as chironomids could carry waterborne PCBs to the terrestrial compartment via their lifecycles. The estimated annual flux of PCBs for chironomids ranged from 0.66 to 265 $ng \cdot m^{-2} \cdot y^{-1}$. Although a high prevalence of PCB-11 and PCB-28 was found for most aquatic based samples in this riparian zone, the mid-chlorinated congeners (e.g. PCB-153 and PCB-138) became predominant among chironomids and dragonflies as well as soil dwelling invertebrates, which might suggest a selective biodriven transfer of different PCB congeners.

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1. Introduction

Polychlorinated biphenyls (PCBs) are persistent organic pollutants (POPs) that consists of 209 possible congeners and have been prohibited for production and application because of their toxicity, bioaccumulation and high persistence (http://chm.pops.int). Wastewater derived from households and industrial activities have been shown to be potential sources of PCBs and can contaminate the surrounding environment through the effluents from wastewater treatment plants (WWTPs) (Blanchard et al., 2004; Wang et al., 2007). In recent years, 3,3-dichlorobiphenyl (PCB-11) has gained increasing interest because this congener is normally not found at significant level in common technical PCB mixtures (Frame et al., 1996). Nevertheless, PCB-11 was found at high concentrations in WWTPs in the New York city area (Litten et al., 2002) and even served as a tracer for wastewater effluents (Rodenburg et al., 2011). Rodenburg et al. (2010) found that PCB-11 was mainly associated with pigment manufacturing. Due to its relatively high volatility, PCB-11 has been prevalently found in Chicago air (Hu et al., 2008) and was even detected at relative high abundance in environmental samples from the Antarctica (P. Wang et al., 2012).

It is generally thought that transport of PCBs is mainly dominated by physical systems such as the atmosphere and ocean currents, but recent studies have revealed that biologically driven transfer through the behaviors and lifecycles of certain animals can also contribute to contaminant movement between compartments (Blais et al., 2007; Walters et al., 2010). Several studies have been carried out to explore the biodriven transfer of aquatic PCBs. For example, spiders which prey on aquatic insects can contribute to the flux of PCBs from sediment to the terrestrial ecosystem (Walters et al., 2010). Mayflies could bioaccumulate PCBs in sediment during the aquatic stage and transfer these to land in large quantities due to high reproduction cycles (Daley et al., 2011). These studies suggested that emerging aquatic insects might be important vectors of contaminants from the aquatic compartment to the terrestrial ecosystem. However, most previous studies focused on sediments contaminated from historical production of PCBs and few studies have shown a direct biomediated transfer of PCBs originated from WWTP effluent in urban areas. Because treated WWTP discharge can provide large amount of nutrients to the recipient environment, there could also be increased presence of invertebrates near by the effluent.

In this study, we collected river water, soil, sediment, vegetation and invertebrates in a riparian zone around the effluent of a large WWTP. We aimed to investigate and compare the levels of PCBs in all collected samples, study the biodriven transfer of PCBs from WWTP effluent to terrestrial compartment by emerging invertebrates and the potential bioaccumulation of PCBs in this urban riparian zone.

2. Materials and methods

2.1. Sample collection

The sampling was conducted around a river in north Beijing. A portion of this river is continuously receiving treated effluent from a WWTP with a wastewater treatment capacity of approximately 400,000 m^3 per day. The river also receives diffuse effluents from households, drainage and small workshops. Sampling was conducted about 0.5 km downstream of the WWTP effluent in the summer of 2010. Several small dams are placed upstream of the WWTP effluent to control the river flow and are sometimes closed off which leads to occasional drying of the river bed upstream of the WWTP. Collection of soil and biota samples in upstream sites was not feasible because the upstream river bank was paved by concrete on both sides, but sediment and common reed samples were collected about 3.5 km upstream of the WWTP effluent. Common reed samples were pooled from about five individual plants. A water sample was collected in the river close to the WWTP effluent and was stored at 2 °C before pretreatment. Composite sediment and tubifex tubifex samples (~100 individuals) were collected from the same spots using a sediment grabber. Zooplankton was sampled using plankton net with mesh size of 200 μm and transferred to a precleaned glass bottle. Individual butterflies (n = 10), cicada (n = 2), large dragonflies (n = 5), small dragonflies (n = 5), large grasshoppers (n = 4) and small grasshoppers (n = 4) were composited to form a single sample for each species. Emerged chironomids were sampled by sweep net and pooled together to obtain sufficient quantity for subsequent analyses. A soil sample was pooled from five subsamples at a grass plain within 2 m from the stream shoreline where influence from the river water is most significant. Composite samples of earthworms (~50) and larvae of scarabs (also called white grubs, ~40) were collected from the soil sampling sites. These two terrestrial invertebrates are mostly buried under the soil surface, where earthworms feed on organic matter whereas white grubs mainly feed on plant roots.

A total of 21 composite samples were collected and labeled as WWTP effluent (WWTP-E), reed stem-leaf upstream (Reed-SLu), reed root upstream (Reed-Ru), reed stem-leaf downstream (Reed-SLd), reed root downstream (Reed-Rd), sediment downstream (Sed-d), sediment upstream (Sed-u), soil downstream (Soil-d), zooplankton (Zooplan), butterfly (Butt), chironomids (Chir), tubifex tubifex (Tubi), cicada (Cica), large dragonfly [Drag(L)], small dragonfly [Drag(S)], small grasshopper [Gras(S)], large grasshopper [Gras(L)], small scarabaeidae larva [Scar(S)], large scarabaeidae larva [Scar(L)], small earthworm [Earth(S)], large earthworm [Earth(L)]. All the biota, soil and sediment samples were wrapped in aluminum foil, sealed in ziplock bags, transported back to the laboratory within the same day. The different worms and larvae were placed on moist filter paper for 24 h in order to evacuate their gut contents. The plants (common reed) were washed with purified water and divided into roots and stem/leaf.

2.2. Chemicals

Native and labeled PCB standards (purity ≥ 98%,isotopic purity ≥ 99%), were purchased from Wellington Laboratories (Ontario, Canada). PCB surrogate standards (68A-LCS) include $^{13}C_{12}$-PCB-1, 3, 4, 15, 19, 37, 54, 81, 77, 104, 105, 114, 126, 155, 156, 157, 169, 188, 189, 202, 205, 206, 208 and 209. The internal standards (68A-IS) comprised of $^{13}C_{12}$-PCB-9, 52,101,138 and 194.

2.3. Sample pretreatment and instrumental analysis

The pretreatment and analysis of the different sample matrices is based on the isotope dilution quantification protocol outlined in the US EPA 1668 methods and has been described in our previous studies (Wang et al., 2007; Wang et al., 2010). Briefly, the solid samples were freeze dried, ground, spiked with labeled surrogate standards, extracted by accelerated solvent extraction (ASE), cleaned up by a multilayer silica column and further spiked with labeled internal standards before instrumental analysis.

Instrumental analysis was performed on a high resolution gas chromatography/high resolution mass spectrometer (HRGC-HRMS). The HRMS was operated in EI mode at a resolution ≥10,000. A total of 26 PCBs including 12 dioxin-like PCBs (PCB-77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169, 189), 6 indicator PCBs (PCB-28, 52, 101, 138, 153, 180) and other 8 PCBs (PCB-3, 11, 15, 19, 202, 205, 208, 209) were quantified.

Stable isotopes in were measured using a Thermo DELTA V Advantage isotope ratio mass spectrometer combined with a Flash EA1112 HT elemental analyzer (Thermo Fisher, Waltham, MA).

More details on the sample pretreatment, analysis and quality assurance and control can be found in the Supplementary Materials.