Chemosphere 81 (2010) 1045–1051

Contents lists available at ScienceDirect

Chemosphere

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

Characteristic accumulation and soil penetration of polychlorinated biphenyls and polybrominated diphenyl ethers in wastewater irrigated farmlands

Thanh Wang, Yawei Wang, Jianjie Fu, Pu Wang, Yingming Li, Qinghua Zhang, Guibin Jiang *

State Key Laboratory of Environmental Chemistry and Ecotoxicology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, P.O. Box 2871, Beijing 100085, China

ARTICLE INFO

Article history:
Received 4 May 2010
Received in revised form 29 June 2010
Accepted 18 July 2010
Available online 16 September 2010

Keywords:
PCBs
PBDEs
Wastewater irrigation
Farm soil

ABSTRACT

The impact of wastewater irrigation on the distribution of two groups of persistent organic pollutants (POPs), polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs), in farm soil was investigated in this study. The concentrations of total analyzed PCBs were in the range 256–2140 pg g^{-1} on dry weight basis in surface soils. There was a higher accumulation of PCBs in farms irrigated by wastewater, with decachlorobiphenyl (CB-209) as the predominant congener. The spatial distributions of PBDEs were similar although not as obvious as that of PCBs, and BDE-209 was the predominant congener at 2040–496 000 pg g^{-1} dw, accounting for >96% of the total analyzed PBDEs. However, no significant correlations could be found between PCB and PBDE concentrations in the topsoil samples, and also with soil organic content between the different sites. On the other hand, soil vertical profiles showed significant relationship with soil organic content in cores taken from farms irrigated with wastewater. The vertical distribution was quite uniform at the topsoil, corresponding to the plowed layer, and decreased thereafter exponentially. Furthermore, the soil vertical distribution was found to be congener specific for PCBs, where less chlorinated congeners were able to penetrate deeper into the soil while heavier congeners were more restricted in their movement. This fractionation process was however not found for PBDEs. Also, the prevalence and high relative concentrations of CB-11 and CB-209 suggests that these PCB congeners should more often be included in routine environmental analysis in order to identifying unusual contamination sources.

1. Introduction

Persistent organic pollutants (POPs) have been of great environmental and human health concerns for several decades. These pollutants are persistent in the environment, can travel over long distances, able to bioaccumulate through the food web and are toxic. Polychlorinated biphenyls (PCBs) are a group of POPs that theoretically consists of 209 congeners, and has been targeted by the Stockholm Convention on Persistent Organic Pollutants for final elimination. A structural similar group of compounds, polybrominated diphenyl ethers (PBDEs), have been used in large quantities as flame retardants in upholstery and electronics. Due to increasing levels in the environment and wildlife, two technical formulations (penta and octa commercial mixtures) of PBDEs have recently been listed into the Stockholm Convention for prohibition of production and usage. Despite intensive research, there are still deficiencies in the understanding of the environmental fate of these POPs. For example, the extent and significance of the soil compartment to act as a repository or sink to the global cycling of POPs is still not fully understood (Meijier et al., 2003; Ockenden et al., 2003). Many previous studies focus on the role of soil in forests or grasslands, which have been found to be able to retain large amounts of POPs (Ockenden et al., 2003; Moeckel et al., 2008). In farmlands, POPs that are not intended as pesticides usually enter the soil compartment through atmospheric deposition or unintentionally through contaminated biosolids or irrigation sources (Alcock et al., 1996; Chen et al., 2005). Several previous studies have detected PCBs and PBDEs in municipal treatment plants (North, 2004; Guo et al., 2009), and it has also been found that spray field irrigation of treated municipal wastewater can be potential atmospheric sources of PBDEs to the surrounding region (Goel et al., 2006). Due to factors such as limited water supply, high nutrition load and economic reasons, some farmers in peri-urban regions utilize untreated or partially treated wastewater from contaminated rivers to irrigate their farmlands. Crude estimations imply that the area under wastewater irrigation might reach as high as 20 million hectares globally, with a major part occurring at peri-urban regions in developing countries (Scott et al., 2004). The proximity to urban and industrial areas can therefore lead to contamination of certain chemicals and pathogens to these irrigation sources which further can be transferred to farm soils, crops

* Corresponding author. Tel.: +86 10 6284 9179; fax: +86 10 6284 9334.
E-mail address: gbjiang@rcees.ac.cn (G. Jiang).

0045-6535/$ - see front matter © 2010 Elsevier Ltd. All rights reserved.
doi:10.1016/j.chemosphere.2010.07.045

and eventually to humans. Since urban areas have been considered as contemporary sources of POPs to the environment, it is important to assess the potential transfer of POPs to farm soils in peri-urban farmlands.

This paper examines and compares the distribution of legacy POPs (PCBs) with that of the currently used PBDEs in agricultural soils at an urban–rural fringe in order to discern potential contamination by wastewater irrigation. We are also interested in comparing the downward movements of these two POPs in farm soils since previous reports mainly focus on the distribution of PCBs at the plowed surface layer (During and Gath, 2002; Armitage et al., 2006) or the effect of biosolids and sewage sludge amendment (Beck et al., 1996; Wilson et al., 1996; Matscheko et al., 2002). Also, few field studies have been instigated to assess the vertical distribution of emerging POPs such as PBDEs in wastewater irrigated farms. It is hoped that the results from this study will provide useful information on the transfer of these POPs from wastewater irrigation sources to farms and their distribution in agricultural soil.

2. Method section

The Tongzhou district (TZ) is located in southeast Beijing and situates both industrial and agricultural activities (Fig. SI-1). The Liangshui river (LSR) flows through the district and is utilized as a recipient for wastewater from both treated/untreated industrial and local municipal wastes through a complex network of smaller connecting streams and ditches. Besides from these, the LSR also receives treated effluents from a large wastewater treatment plant (WWTP) located around 15 km north of the river. The predominant soil types in this area are loamy soils, and winter wheat and summer maize is the main crop rotation practice. The farms at the main sampling sites were categorized as wastewater irrigated only (S sites), or mixed irrigated (M sites) which is defined as farms that have been irrigated with both groundwater and wastewater at some point in recent time. The wastewater sources at M sites do not necessarily come directly from the LSR but could also be from local municipal and industrial wastewater effluents. Additional soil samples were collected from farm plots irrigated solely by groundwater (G sites). Samples were also collected at a grass lawn at the north of urban Beijing (site R) in order to distinguish potential urban–rural differences. Surface soils (0–20 cm) were collected as composite samples during three sampling campaigns; July (site M1, M3, S3, R) and November 2008 (M1, M2, M4, S5, G) and May 2009 (M1, S1, S2, S4, S6, S7, S8) (Fig. 1). Vertical soil profiles were collected using in-house built steel core probes (1 m in length and 15 cm in diameter) at penetration depths of approximately 70 cm (M2, S2, S3, S6, R). The core samples were sectioned into 2–4 cm layers in order to obtain high vertical resolution. Water samples (W0, W1, W2) were collected from the LSR at a depth of approximately 20 cm below surface using amber glass bottles pre-cleaned with hexane.

Qualitative and quantitative determination of target analytes was based on an established isotope dilution method, and has been described in our previous works (Wang et al., 2007). The PCB congeners reported in this study include 12 dioxin-like (DL) PCBs (CB-77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169 and 189), six indicator congeners (CB-28, 52, 101, 138, 153, and 180), and eight other congeners (CB-3, 11, 15, 19, 202, 205, 208, and 209). Separate analysis was also performed for 14 PBDE congeners (BDE-17, 28, 47, 66, 71, 85, 99, 100, 138, 153, 154, 183, 190 and 209). All soil samples were reported on a dry weight (dw) and blank corrected basis unless otherwise mentioned.

Detailed information on the sampling sites, extraction, analysis and QA/QC can be found in Supplementary Material.

3. Results and discussion

3.1. Spatial distribution and congener composition of PCBs and PBDEs

Numerous studies have been carried out in order to investigate the contamination of PCBs and PBDEs in agricultural soils due to

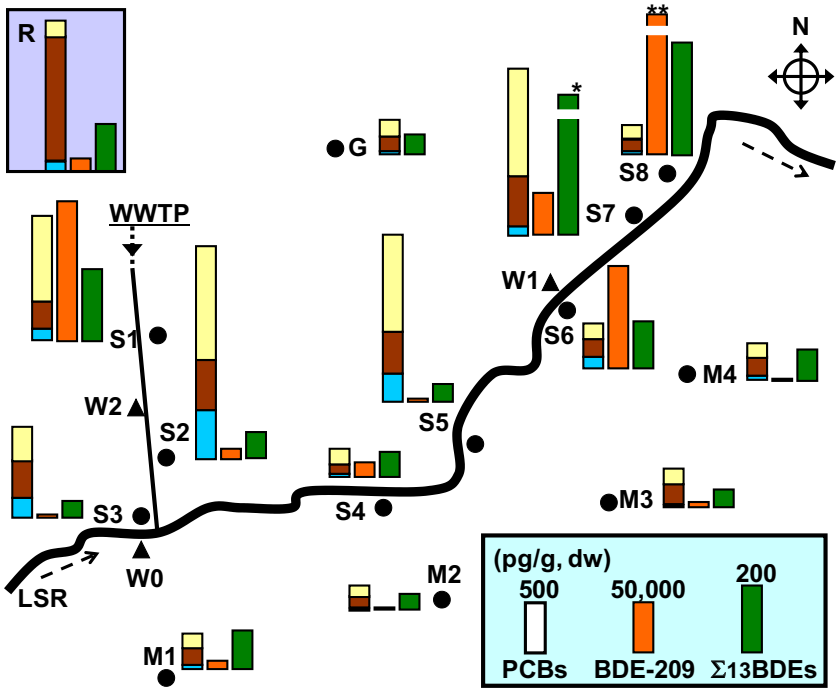

Fig. 1. Sampling locations around the Liangshui River (LSR) and the corresponding soil levels of PCBs and PBDEs (pg g^{-1} dw). Columns for Σ_{26}PCBs depict cumulative levels of dioxin-like PCBs (blue), six indicator PCBs (brown) and other congeners (yellow). BDE-209 (orange) could not be analyzed for site G due to difficulties encountered during sample preparation. (*) Extreme level for Σ_{13}PBDEs (green) at site S7; 770 pg g^{-1}. (**) Extreme level for BDE-209 at site S8; 496 000 pg g^{-1}. Concentrations for water samples (W) are shown in SI. Note that the columns for each group of compounds are depicted at different concentration ranges, and visual inter-comparison is thus not directly feasible. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)