



Short communication

Occurrence and distribution of organophosphorus esters in soils and wheat plants in a plastic waste treatment area in China[☆]Weining Wan^a, Shuzhen Zhang^{a,*}, Honglin Huang^a, Tong Wu^b^a 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^b School of Environmental Science and Engineering, Hebei University of Science and Technology, Hebei, 050018, China

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ABSTRACT

This study for the first time reported the occurrence, distribution and concentrations of organophosphate esters (OPEs) in soils caused by plastic waste treatment, as well as their influence on OPE accumulation in wheat (*Triticum aestivum* L.). Eight OPEs were detected with the total concentrations of 38–1250 ng/g dry weight in the soils from the treatment sites, and tributoxyethyl phosphate and tri(2-chloroethyl) phosphate present as the dominant OPEs. There were similar distribution patterns of OPEs and significant correlations between the total OPE concentrations in the soils from the plastic waste treatment sites with those in the nearby farmlands ($P < 0.005$), indicating that plastic waste treatment caused the OPE contamination of farmland soils. The uptake and translocation of OPEs by wheat were determined, with OPEs of high hydrophobicity more easily taken up from soils and OPEs with low hydrophobicity more liable to be translocated acropetally.

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1. Introduction

Organophosphate esters (OPEs) are widely used as flame retardants and plasticizers in various consumer products (Bacaloni et al., 2008), particularly after the implementation of regulations phasing out the production and usage of polybrominated diphenyl ethers in many countries in recent years (Wei et al., 2015). OPEs are mainly used as non-reactive additives in the products, and therefore easily leached out to the environment (Marklund et al., 2003). So, the occurrence and fate of OPEs in the environment have attracted increasing attention from both the public and researchers (Van der Veen and De Boer, 2012).

The recent studies on OPEs have been mainly focused on their occurrence in various environments such as air (Castro-Jimenez et al., 2014), waters (Andresen et al., 2004; Marklund et al., 2005) and sediments (Cristale and Lacorte, 2013), and accumulation in living organisms (Kim et al., 2011) and human blood, milk and urine (Shah et al., 2006; Kim et al., 2014). However, the recent studies on OPEs have been mainly limited to their behavior in aquatic environments, and little research has been conducted on their behavior

in the terrestrial environment. To date only two reports on the analysis of OPEs in soils are available (Mihajlovic and Fries, 2012; Matsukami et al., 2015). Plant uptake and accumulation are the significant steps in the transfer of contaminants into the food chain as well as an important process in their global cycling (Shone and Wood, 1974). Nevertheless, there has been no research addressing plant uptake of OPEs. Furthermore, different OPEs possess diverse molecular structures and substituent groups, and thus may behave differently in plant uptake and accumulation. It is thus necessary to clarify how the chemical properties affect the uptake and translocation of OPEs in plants.

Plastic wastes generated by production and consumption in industries and households have grown considerably (Thompson et al., 2009). Disposal and treatment without appropriate control have led to many primitive and crude plastic waste treatment activities (Nnorom and Osibanjo, 2008; Al-Salem et al., 2009), which may cause the release of OPEs to the surrounding environments, and soils likely represent the major sink. Therefore, in the present study, a plastic waste treatment area in Hebei Province in northern China was selected as the research area to investigate the occurrence and distributions of OPEs in the soils. Potential contamination of farmland soils and crop plants in the surrounding area were further investigated. To our knowledge, this is the first study to report soil contamination of OPEs as the result of disposal and

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treatment of plastic wastes, as well as the behavior of OPEs in the soil-plant system. The results of this study will help in enhancing our awareness of soil contamination of OPEs and their potential to contaminate the food chain.

2. Materials and methods

2.1. Chemicals

OPE standards, including tri(2-chloroethyl) phosphate (TCEP), tri(2-chloropropyl) phosphate (TCPP), tri(2, 3-dichloropropyl) phosphate (TDCPP), tributoxylethyl phosphate (TBEP), tri-*n*-butyl phosphate (TnBP), triphenyl phosphate (TPhP), tricresyl phosphate (TCP) and 2-ethylhexyl diphenyl phosphate (EHDPP), and surrogate standards of tri-*n*-butyl phosphate-d27 (d27-TnBP) and triphenyl phosphate-d15 (d15-TPhP) were purchased from Wellington Laboratories, Inc. (Guelph, Ontario, Canada). Standard of tri-*iso*-butyl phosphate (TiBP) was obtained from AccuStandard (New Haven, USA). Major information about these OPEs are provided in Table S1 of the Supplementary material. Solvents, including dichloromethane, *n*-hexane, ethyl acetate, acetone and toluene, were of HPLC grade (Thermo fisher, MA, USA). Ultra-pure water was prepared by using a Milli-Q advantage water purification system (US Millipore, Bedford, MA, USA). All other chemicals used were of analytical grade (Sinopharm Chemical Reagent Co., Ltd, BJ, China).

2.2. Sampling

Sampling sites are located in Xinle County and Dingzhou County in Hebei Province known as the major area for plastic waste recycling in northern China. Intensive plastic waste recycling processes were previously performed using crude techniques on a small scale in family workshops and had lasted for more than a decade. Soil and wheat samples were collected in June 2014 and sampling locations are shown in Fig. S1. In details, four soil samples (approximately 1 kg) were collected at a depth of 0–15 cm with a cleaned stainless-steel shovel from an area of approximately 1 m² for each sample site. There were a total of 28 soil samples, of which 19 were from the plastic waste treatment sites (A to S) and 9 from the nearby farmlands (H to Q, except M). Whole wheat plants were collected in 4 replicates from each of the corresponding farmlands and divided into root and aboveground tissues. All the samples were wrapped with aluminum foil, immediately stored in a portable refrigerator at 4 °C, and then transported to the laboratory to store in a refrigerator at –20 °C. In laboratory, the soil samples were ground and sieved (<0.25 mm). Plant roots and aboveground tissues were first rinsed carefully with tap water, and then washed thoroughly with deionized water, blotted with filter paper and weighed. Both the soil and wheat samples were then frozen at –50 °C overnight, freeze-dried for 48 h in a lyophilizer (FD-1, Beijing Boyikang Instrument Ltd, Beijing, China) and weighted. The dried plant samples were finely chopped. All the samples were stored in glass containers at –20 °C before chemical analysis.

2.3. Sample extraction and analysis

Briefly, ten grams of soil or 2 g of plant samples were accurately weighted to submit Soxhlet extraction with a *n*-hexane/DCM mixture [1:1 (v/v)]. D27-TnBP and d15-TPhP were added as surrogate standards to the samples prior to extraction. The extracts were cleaned-up according to the method by Möller et al. (2011) with some minor modifications. An Agilent 7890 chromatography–mass spectrometry system (5975 inert) (Agilent, Palo Alto, USA) and a HP-5MS column (30 m × 0.25 mm × 0.25 μm) (Agilent J&W Scientific, USA) were used for the analysis. Further details of sample

extraction and analysis are provided in the Supplementary material and parameters are available in Table S2. Both separation and detection of the target OPEs were satisfied for the analyses of the soil and plant samples. The gas chromatograms for OPEs in a leaves together with procedural blank are displayed in Fig. S2 as examples.

2.4. Quality control/quality assurance

Quality control included regular analysis of the procedural blanks, blind duplicate samples, and random injection of solvent blanks and standards. Quality assurance was guaranteed by the addition of surrogate standards and the standards of OPEs individually to the blank soil and plant samples, which were free of OPEs and collected from the experimental field at Beijing Academy of Agriculture and Forest Sciences in Beijing, China. Recoveries were 77.4–86.5% for d27-TnBP and 79.7–89.6% for d15-TPhP, and 72.2–87.1% for OPEs in soil samples as well as 75.6–90.0% for plant tissues, respectively. The limits of detection (LOD) for OPEs, defined as a signal-to-noise ratio (S/N) of 3, were in the range of 6–200 pg/g in the soil and plant samples (n = 4). The details are provided in the Supplementary material.

2.5. Data analysis

For samples with contaminant concentrations below LOD, zero was used for the calculations. Because there was no OPE detected in the blanks, concentrations reported are not blank corrected. All of the results are expressed on a dry weight basis. Means and standard deviation were calculated from triplicates. Statistical analysis was performed using Microsoft Excel 2010 and Origin 8.0 (OriginLab Corporation). Statistical significance between datasets was tested by analysis of variance (ANOVA), and all significance tests were two sided, using P < 0.05 as the level of significance.

3. Results and discussion

3.1. Levels and distribution of OPEs in the soils from the plastic waste treatment sites

Eight of nine OPEs, with the exception of TDCPP, were detected in the 19 soil samples collected from the plastic waste treatment sites, with the total OPEs (Σ OPEs) concentrations ranging from 38 to 1250 ng/g dry weight (dw) and mean value of 398 ng/g dw (Fig. 1A). High concentrations of the Σ OPEs were concentrated in the areas of sites H to Q with the mean value of 631 ng/g dw, approximately 3.5 fold higher than the concentrations (139 ng/g dw) in the soils from the sites A to G, and sites R to S. Indeed, we observed more obvious open deposits of plastic debris and combustion residues in the areas of sampling sites H to Q than the other sites. TBEP exhibited the highest detected frequency (90%) and concentration (varying from 20 to 592 ng/g dw, mean 200 ng/g dw) among all the OPEs. This is corresponded to the evidence that TBEP is the most common OPE used as plasticizers in plastics or rubber stoppers (Andresen et al., 2004; Van der Veen and De Boer, 2012). TiBP, another non-chlorinated OPE, had a similar occurrence (84%) but much lower concentration (mean 47 ng/g dw) than TBEP. TCP had the second highest mean concentration (119 ng/g dw), but was detected only in 37% of the soil samples. TnBP, TPhP and EHDPP were detected in over 60% of the soil samples with the mean concentrations of 22, 26 and 11 ng/g dw, respectively. Although non-halogenated OPEs are primarily employed as plasticizers (Reemtsma et al., 2008), chlorinated OPEs such as TCEP and TCPP were also detected in 90% and 79% of the soil samples, with concentrations in the range of 7–436 ng/g dw (mean 92 ng/g dw) and 4–52 ng/g dw (21 ng/g dw), respectively. Up to date there are only

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