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Short Communication

Occurrence of organophosphorus flame retardants and plasticizers in wild insects from a former e-waste recycling site in the Guangdong province, South China

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HIGHLIGHTS

- Large quantities of e-waste are generated worldwide and often inappropriately dismantled.
- Occurrence of PFRs was investigated in wild insects from an e-waste recycling site (China).
- TEHP was the most abundant PFR, followed by TPHP, TCIPP, TCEP, EHDPHP and TCP.
- *Odonata* were more contaminated, followed by *Lepidoptera*, *Orthoptera*, *Hemiptera*, *Coleoptera*.
- PFR contamination patterns could be explained by insect habitats and feeding habits.

GRAPHICAL ABSTRACT

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ABSTRACT

Due to the fast growth of the electronic industry, a large quantity of electronic waste (e-waste) is generated worldwide and then often inappropriately dismantled and disposed of. In a pilot study, the occurrence of organophosphorus flame retardants and plasticizers (PFRs) was investigated for the first time in several wild insect species collected from a former e-waste recycling site in the Guangdong province, South China. TEHP was the most abundant PFR (average concentration of 5.8 ng/g ww), followed by TPHP (2.5 ng/g ww), TCIPP (2.2 ng/g ww), TCEP (0.8 ng/g ww), EHDPHP and TCP (both 0.1 ng/g ww). Dragonfly nymphs were the most contaminated insects, with total PFR concentrations of 68 ng/g ww, followed by moth adults (26 ng/g ww) and terrestrial stinkbug (17 ng/g ww). The different contamination patterns observed in the analyzed insects could be explained by their different habitats and feeding habits. This study shows that e-waste recycling areas can be an important local source of contamination with PFRs, mainly caused by inadequate recycling activities.

1. Introduction

After the phase out of certain brominated flame retardants (BFRs) and their addition to the POP list due to toxicity concerns, organophosphorus flame retardants (PFRs) have been implemented as the foremost

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alternatives for BFRs (van der Veen and de Boer, 2012). They are used as flame retardants and plasticizers in a wide variety of industrial applications, from plastics and electronics, to polyurethane foams (PUFs), PVC, textiles and food-wrapping film (Wei et al., 2015).

Due to the fast growth of the electronic industry, a large quantity of electronic waste (so called e-waste), such as computers, mobile phones, televisions, etc., is generated worldwide (Bi et al., 2010). Most of the globally produced e-waste has been imported illegally into China to be dismantled and disposed of, causing major environmental concerns (Bi et al., 2010). For example, in Longtang, one of the former largest e-waste recycling sites in Qingyuan city (Guangdong province, South China), primitive and low-tech recycling activities in family-run workshops have resulted in the extensive and severe release of persistent organic pollutants (POPs) and halogenated flame retardants (HFRs) to the local environment (Huang et al., 2018; Liu et al., 2018). To mitigate e-waste contamination, regulations to control e-waste importation and disposal have been recently implemented in Qingyuan city (Huang et al., 2018). Since 2011, family-run e-waste workshops have been banned and recycling activities were gradually moved to local e-waste disposal centers (Huang et al., 2018). However, leftovers of the dismantling activities are still present and visible in the dumping area (Fig. 1). Since PFRs are not chemically linked with the polymeric material and can leach out from the products (van der Veen and de Boer, 2012; Wei et al., 2015), the inadequate treatment and disposal of e-waste might be a significant source of PFR contamination, posing serious adverse effects to the environment and humans around e-waste areas. For this reason, the shortage of data on the occurrence of PFRs in e-waste dismantling sites, such as Longtang, is still considered a research gap and should be investigated (Wei et al., 2015; Zheng et al., 2015).

In addition, only few studies currently report on PFRs in biota, since recent literature has mostly focused on their occurrence in abiotic matrices (e.g. dust, air, sediments, food) (Poma et al., 2018; Wei et al., 2015). Insects are a dominant component of biodiversity in both aquatic and terrestrial ecosystems and, as such, they are an important link in food webs, playing a key role in the initial steps of bioaccumulation of organic pollutants from the environment (Liu et al., 2018).

In this pilot study, the occurrence of 12 PFRs, (e.g. tris(1-chloro-2-propyl) phosphate (TCIPP), tris-(1,3-dichloro-2-propyl) phosphate (TDCIPP), tris(2-chloroethyl) phosphate (TCEP), tri-*n*-butyl phosphate (TnBP), triphenyl phosphate (TPHP), 2-ethylhexyldiphenyl phosphate (EHDPHP), tri-*n*-propyl phosphate (TnPP), tris(2-ethylhexyl) phosphate (TEHP), tricresyl phosphate (TCP), tris(2-isopropylphenyl) phosphate (T2IPPP), tris(3,5-dimethylphenyl) phosphate (T35DMPP), and tris(*p-tert*-butylphenyl) phosphate (TBPP)) was investigated for the first time in several wild insect species collected in the Longtang former e-waste recycling site (South China).

2. Materials and methods

2.1. Sample collection

Nine wild insect species belonging to five orders (*Odonata*, *Orthoptera*, *Hemiptera*, *Coleoptera* and *Lepidoptera*) were collected around a pond and from the surrounding farmland region (within 50 m range from the pond), area heavily polluted with chemicals associated with e-waste, in Longtang, Guangdong province, between September 2015 and November 2016 (Liu et al., 2018) (Fig. 1). A total of 13 insect samples were considered for the PFR analysis; for dragonfly, grasshopper and moth, both nymph/larval and adult stage were collected and analyzed (Table 1). Dragonfly nymphs and aquatic insects were collected using a macro-invertebrate dip net from the pond and/or from a ditch in the surrounding corn fields, while adults and terrestrial insects were captured using sweep nets from the trees and fields around the pond (Liu et al., 2018). Due to their small individual size, multiple insects from the same species were pooled for analysis into composite samples, freeze-dried, homogenized, and stored at −20 °C until analysis. Full details concerning insect collection and habitat are reported in Liu et al. (2018).

2.2. Sample preparation and instrumental analysis

Insect samples were extracted in October–December 2017 and analyzed according to the in-house validated method described in Poma et al. (2018). Briefly, approximately 200 mg of lyophilized sample was spiked with an internal standard (IS) mixture (containing TPHP-d_{15}, TDCIPP-d_{15}, TCEP-d_{12}, and triamyl phosphate (TAP)) and extracted with acetonitrile. The extract was then centrifuged, the pooled supernatant was evaporated under a gentle nitrogen stream and cleaned-up

Fig. 1. Insect sampling site and e-waste dumping area in Longtang, Guangdong province, South China.