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Research article

Comprehensive evaluation of dioxins and dioxin-like compounds in surface soils and river sediments from e-waste-processing sites in a village in northern Vietnam: Heading towards the environmentally sound management of e-waste

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ABSTRACT

The management of electronic waste (e-waste), which can be a source of both useful materials and toxic substances, depending on the processing method, is important for promoting material cycling. In this study, we used the dioxin-responsive chemical-activated luciferase gene expression (DR-CALUX) assay combined with gas chromatography–high-resolution mass spectrometry to evaluate the levels of dioxin-like compounds in surface soils and river sediments collected in and around an e-waste-processing village in northern Vietnam. The WHO-TEQs (Toxic equivalents) of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), coplanar polychlorinated biphenyls (Co-PCBs), and polybrominated dibenzo-*p*-dioxins and dibenzofurans (PBDD/Fs) in soils collected in January 2012 ranged from 0.29 to 310 pg/g (median 2.9 pg/g, $n = 32$), and the WHO-TEQs in sediments ranged from 0.96 to 58 pg/g (median 4.4 pg/g, $n = 8$). Dioxin-like activities (CALUX-TEQs [2,3,7,8-TCDD equivalent]) in soils collected in January 2012, 2013, and 2014 ranged from <30 to 4300 pg/g (median <30 pg/g, $n = 96$), and the activities in sediments ranged from <30 to 4000 pg/g (median 33 pg/g, $n = 24$). Dioxin-like compounds accumulated in samples collected around e-waste-processing areas such as open-burning sites and e-waste-processing workshops, and the compounds may be transported from their sources to surrounding areas over the course of several years. Some of the CALUX-TEQs, but not WHO-TEQs, values were higher than the maximum acceptable WHO-TEQs promulgated by various authorities, indicating that all dioxin-like compounds should be evaluated in samples collected from e-waste-processing areas. Our findings suggest that open burning and open storage of e-waste should be prohibited and that wastewater treatment should be implemented at each workshop to reduce contamination by dioxin-like compounds from e-waste.

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1. Introduction

Waste from obsolete electronic devices and appliances such as personal computers, television sets, mobile phones, printers, and refrigerators is generated at a global rate of approximately 41.8 million tons per year, according to a press release by United

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Nations University [1]. Currently, large quantities of this waste, called e-waste, are recycled in both developed and developing countries because it contains considerable quantities of valuable and reusable metals, plastics, glass, and other materials. However, the use of certain e-waste-processing methods, such as open burning, especially in the developing world, has become an important issue in the last decade because of the adverse human health impacts of these methods [2–5]. Although several developing countries, including China, have enacted legislation focusing on the environmental effects of e-waste disposal and processing, the legislation enacted to date does not effectively regulate e-waste processing [6]. Furthermore, a recent study estimated that by 2018, developing countries will be disposing of more old computers than developed countries [7]. Up to now, the main approach to mitigating the impacts of undesirable e-waste processing has focused on reducing the amount of e-waste that developed countries export to developing ones, but in response to anticipated changes in the flow and generation of e-waste, new approaches will have to be explored.

Numerous studies have targeted contamination hot spots resulting from intensive e-waste-processing activities such as open burning and smelting and acid leaching to retrieve metals (reviewed by Man et al. [4] and Chan and Wong [5]). Many researchers have concluded that intensive e-waste processing is harmful to the environment and to human health. Therefore, our motivation in this study was to provide data for environmental samples collected not only from areas where open burning, an intensive processing activity, is conducted but also from areas where nonintensive processing activities such as collection, storage, and manual dismantling of e-waste are conducted. Since January 2012, our research group has been investigating e-waste-processing activities in Bui Dau, a village in My Hao district, Hung Yen Province, northern Vietnam, to elucidate the current levels of contaminants associated with various types of e-waste processing and to monitor the 3-year temporal trends not only of chemicals that may be initially present in e-waste, such as brominated and chlorinated flame retardants, phosphorus-containing flame retardants, and heavy metals but also of hazardous chemicals that may be generated during e-waste processing, such as chlorinated and brominated dioxins and dioxin-like compounds.

Specifically, we evaluated persistent dioxin-like compounds in surface soils and river sediments collected from the village. The levels of flame retardants such as polybrominated diphenylethers (PBDEs) and possible alternatives have already been reported by Matsukami et al. [8] and Someya et al. [9], and data for heavy metals will be reported elsewhere in the near future. In this study, we used gas chromatography-high resolution mass spectrometry (GC-HRMS) for measurement of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), coplanar polychlorinated biphenyls (Co-PCBs), and polybrominated dibenzo-*p*-dioxins and dibenzofurans (PBDD/Fs), and the dioxin-responsive chemical-activated luciferase gene expression (DR-CALUX) assay for all the dioxin-like compounds. First, we evaluated the levels of PCDD/Fs, Co-PCBs, PBDD/Fs, and emerging dioxin-like compounds in samples collected in January 2012, in order to elucidate the source of these contaminants and possible relationships among them. By using DR-CALUX assay, we also evaluated the levels of all the dioxin-like compounds in samples collected in January 2012, 2013, and 2014 to investigate the time-course of the levels of contaminants related to e-waste-processing activities. Finally, we tried to interpret the obtained results for identification of critical processes of contaminant formation and possible countermeasures for environmentally sound management of dioxin-like compounds derived from e-waste processing and to investigate the validity of using the DR-CALUX assay in combination with GC-HRMS to assess the risks associated with e-waste processing activities.

2. Materials and methods

2.1. Sampling location and sample collection

The study location was an e-waste-processing site in the village of Bui Dau, My Hao district, Hung Yen Province, northern Vietnam. Detailed information about this area has been reported elsewhere [8,10]. The sampling area, which includes living areas and rice fields, covered a surface area of about 4.2 km^2. The total population of the village reached approximately 3000 persons by 2015, according to a survey conducted for this study. The population and area of the village are much smaller than those of Guiyu (population, 150,000; land area, 52.4 km^2) and Taizhou (population, 400,000; land area, 274 km^2), which are well-known e-waste-processing sites in China [4]. E-waste processing in Bui Dau started in the 2000s [10], whereas Guiyu and Taizhou started processing e-waste in the late 1980s and 1970s, respectively. Hence, Bui Dau is a small-scale processing location and has a short history of e-waste processing; therefore, we expected that the degree of chemical contamination derived from processing activities would be low. A map of the specific sites in Bui Dau from which surface soils and river sediments were collected is shown in Fig. 1 (reproduced from Ref. [8]).

During each of the three sampling surveys in January 2012, 2013 and 2014, surface soils (depth 0–5 cm) were collected from 32 fixed locations including footpaths in rice paddies around the village ($n = 19$: SS-1 to SS-19), wires and cables open-burning area ($n = 3$: SS-20 to SS-22), and areas adjacent to e-waste-processing workshops ($n = 10$: SS-23 to SS-32), as shown in Fig. S1. River sediments were collected from eight locations including upstream of the e-waste-processing area (RS-1), the e-waste-processing area ($n = 3$: RS-2 to RS-4), and downstream of the e-waste-processing area ($n = 4$: RS-5 to RS-8). The above indicated surface soil and river sediment samples were collected in three sampling campaigns, respectively January 2012, 2013, and 2014; therefore, a total of 96 soil samples and 24 sediment samples were collected. Each sample was composed of five subsamples, which were collected with a stainless steel shovel from an area of approximately 10 m^2 and were placed in re-sealable polyethylene zipper storage bags. All samples were kept in a freezer in the Research Centre for Environmental Technology and Sustainable Development (CETASD), Hanoi University of Science, until they were transported from Vietnam to Japan with permission from the Ministry of Agriculture, Forestry, and Fisheries. All the imported samples were stored at −20 °C until analysis.

2.2. Sample pretreatment and extraction

After removal of pebbles, weeds, and twigs, the samples were air-dried and manually homogenized with a wooden hammer. Each air-dried sample was transferred to a stainless steel sieve (<2.0 mm), which was then covered with a steel lid and shaken manually. The sieved material was collected and stored in amber glass bottles at −20 °C until extraction.

Approximately 15 g of each sieved sample was extracted by means of a rapid thermal solvent extractor (SE100, Mitsubishi Chemical Analytech Co., Kanagawa, Japan) first at 35 °C for 40 min with 1:1 (v/v) acetone:*n*-hexane at a flow rate of 2 mL/min and then at 80 °C for 40 min with toluene at 2 mL/min. The combined extracts were subjected to solvent evaporation to a volume of 10 mL, and the resulting crude extracts were stored at 4 °C until WHO-TEQ and CALUX-TEQ determinations.