

Spatial Distribution of Polybrominated Diphenyl Ethers and Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans in Soil and Combusted Residue at Guiyu, an Electronic Waste Recycling Site in Southeast China

ANNA O. W. LEUNG,[†]
WILLIAM J. LUKSEMBURG,[‡]
ANTHONY S. WONG,[‡] AND
MING H. WONG^{*,†}

Croucher Institute for Environmental Sciences, and Department of Biology, Hong Kong Baptist University, Hong Kong, PR China, and Vista Analytical Laboratory, Inc., 1104 Windfield Way, El Dorado Hills, California 95762

Surface soils and combusted residue from a village located in southeast China, which has been intensely involved in the dismantling and “recycling” of computer parts (e-waste) for the past decade, were analyzed for polybrominated diphenyl ethers (PBDEs) and polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs). Total PBDE concentrations were highest in combusted residue of plastic chips and cables collected from a residential area (33 000–97 400 ng/g, dry wt), in soils from an acid leaching site (2720–4250 ng/g, dry wt), and a printer roller dump site (593–2890 ng/g, dry wt). BDE-209 was the most dominant congener (35–82%) among the study sites indicating the prevalence of commercial Deca-BDE, however signature congeners from commercial Penta- and Octa-BDE were also found. PCDD/F concentrations were also highest in soil from the acid leaching site (12 500–89 800 pg/g, 203–1100 pg WHO-TEQ/g, dry wt) and in combusted residue (13 500–25 300 pg/g, 84.3–174 pg WHO-TEQ/g, dry wt) and were comparable to PCDD/F levels of some open dumping sites in Asian developing countries. Of the e-waste activities, acid leaching and open burning emitted the highest concentrations of PBDEs and PCDD/Fs. This study is among the very few studies dealing with the important issue of pollution generated from crude e-waste recycling. Our results show that the crude processing of e-waste has become one of the major contributors of PBDEs and PCDD/Fs to the terrestrial environment.

Introduction

Electronic-waste (e-waste), a term used to describe obsolete electrical and electronic equipment, is a global 21st century problem. Currently, there are about 50 million tonnes of e-waste produced annually in the world (1). In developed

countries, it is becoming one of the fastest growing waste streams comprising more than 5% of municipal solid waste. Between 1997 and 2007, a forecasted 500 million computers will become obsolete in the United States, adding to the burden. Moreover, e-waste is being transported in massive quantities around the world to developing countries (2). About 80% of computer e-waste is being exported to Asia, and 90% of these exports have been sent to China through illegal imports, for “recycling” (2). During the crude recycling of e-waste, persistent toxic substances (PTS) such as PBDEs and PCDD/Fs may be emitted into the environment. PCDD/Fs are known as Stockholm Convention persistent organic pollutants (POPs), and commercial pentabromodiphenyl ether (Penta-BDE) and octabromodiphenyl ether (Octa-BDE) are chemicals under consideration for inclusion in the Convention (3).

PBDEs are brominated flame retardants that have been widely applied to furniture, textiles, and office equipment. In computers, the three main technical mixtures of PBDEs—Penta-BDE, Octa-BDE, and Deca-BDE—are mainly used in four applications: printed circuit boards, plastics (computer casing), cables, and components such as connectors. Due to health risks, Penta-BDE and Octa-BDE have been banned within the EU since August 2004 (4) and are being phased out in several states of the United States since January 1, 2006 (5). The Directive on the Restriction of the Use of Certain Hazardous Substances in Electrical and Electronic Equipment (RoHS) (Directive 2002/95/EC) prohibits the use of Penta-BDE and Octa-BDE exceeding set maximum concentration values in new electrical and electronic products that enter the EU market after July 1, 2006 (2). Currently, Deca-BDE is the most widely used commercial PBDE flame retardant (6). In 2001, the total world market demand for Deca-, Octa-, and Penta-BDEs was 56 100, 3790, and 7500 metric t, respectively (7).

The combustion of waste containing chlorinated materials is the major source of PCDD/Fs to the global environment (8). Biomass combustion including burning of household refuse in backyard barrels (9) and open burning at landfills and dump sites (10) is one of the largest known, non-regulated sources of dioxins (11). However, the open dumping and burning of e-waste may also significantly contribute to emissions of PCDD/Fs and could be a major component of the global dioxin inventory. PVC-containing materials are PCDD/F precursors, and when burned, they produce PCDD/Fs (12). Studies have shown that copper acts as a catalyst for the formation of PCDD/Fs during the combustion of PVC (13). Therefore, the combustion of e-waste in the presence of copper from printed circuit boards and computer wiring may lead to higher emissions of PCDD/Fs into the environment.

PCDD/Fs have been recognized as chemicals of concern since the 1970s and many studies have been conducted that indicate their ubiquitous occurrence in environmental media and biota (11), whereas PBDEs have only begun receiving attention in the 1990s. However, increasing studies show that PBDEs are globally distributed in the environment, such as in air, sediment, sewage sludge, biota, and in human milk worldwide affecting infant body burden (14). High concentrations of PBDEs have also been found in blood of workers in a Swedish electronic dismantling plant (15). Toxicological studies show that PCDD/Fs and PBDEs can cause adverse human health impacts (16, 17).

The occurrence of recycling of e-waste by environmentally unfriendly, hazardous, and primitive technologies in developing countries has increased over the past years. At present,

* Corresponding author phone: +852-3411-7746; fax: +852-3411-7743; e-mail: mh Wong@hkbu.edu.hk.

[†] Hong Kong Baptist University.

[‡] Vista Analytical Laboratory, Inc.

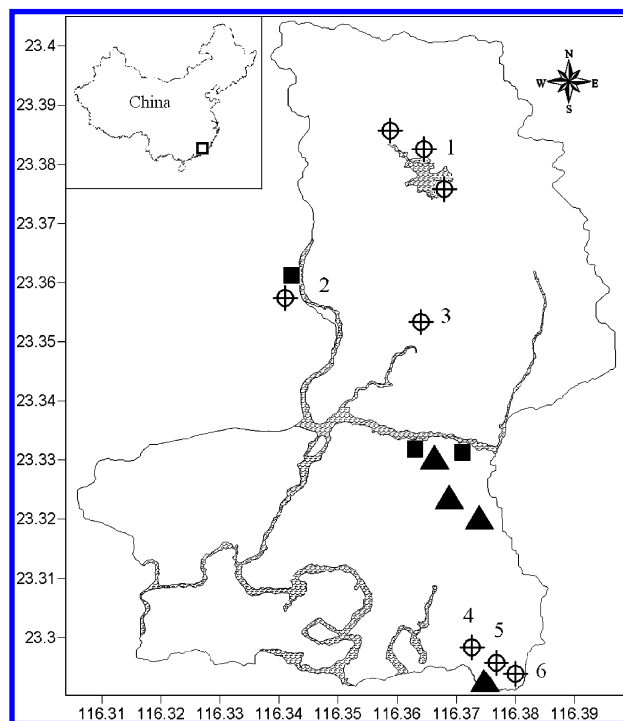


FIGURE 1. Map of Guiyu and sampling locations: (circle with plus sign) sampling location; (▲) e-waste dumping and open burning site; (■) acid leaching of printed circuit boards site.

there is a large data gap that must be urgently filled regarding how the environment and human health are affected by this growing industry. Using Guiyu as a case study for e-waste “recycling”, the major aim of this study is to conduct a reconnaissance survey to investigate and characterize the level and spatial distribution of PBDEs and PCDD/Fs in soil and combusted residue. It is hoped that the information collected will provide a better understanding of the extent of pollution by e-waste and also contribute useful information for further research on the fate and transport of these PTS and their potential environmental and health impacts.

Experimental Section

Study Area. Guiyu, a town in southeast Guangdong Province, China, has a population of 150,000 and has been involved in e-waste “recycling” for approximately 10 years. It occupies an area of 52.4 km² and is approximately 30 km away from Shantou, the closest city, and about 250 km northeast of Hong Kong. Recycling operations consist of scrap sorting, heating printed circuit boards to recover lead solder and electronic components, using concentrated acid solutions to leach and recover aluminum, gold, copper, platinum, and other metals, toner sweeping, dismantling electronic equipment, selling computer monitor yokes to copper recovery operations, plastic chipping and melting, and burning cables to recover copper wires (18). Acid leaching, dumping, and burning of unsalvageable components often take place alongside rivers and rice fields.

Sample Collection and Sample Preparation. Soil samples (3 samples (replicates) from each sampling site) were collected in February 2004 (total 15 samples) using a stainless steel shovel at a depth of 0–10 cm. A composite sampling strategy of 4–5 subsamples was used where possible to obtain representative samples. Combusted residue samples (3 replicates) of e-waste were also collected. All collected samples were wrapped in aluminum foil and stored at –20 °C in the laboratory prior to analysis. The locations of the six sampling sites are shown in Figure 1 and described in Table 1.

Sample Extraction, Cleanup, and Analysis. The soil and combusted residue samples were freeze-dried, sieved (<2 mm) and homogenized. PBDE and PCDD/F concentrations were analyzed by Vista Analytical Laboratory, Inc. (formerly Alta Analytical Laboratory, Inc.) as follows.

PBDEs. An aliquot of each sample (2 g) was spiked with a mixture of 13 ¹³C-labeled internal standards (¹³C₁₂-BDE-3, ¹³C₁₂-BDE-15, ¹³C₁₂-BDE-28, ¹³C₁₂-BDE-77, ¹³C₁₂-BDE-99, ¹³C₁₂-BDE-100, ¹³C₁₂-BDE-138, ¹³C₁₂-BDE-153, ¹³C₁₂-BDE-154, ¹³C₁₂-BDE-183, ¹³C₁₂-BDE-197, ¹³C₁₂-BDE-207, ¹³C₁₂-BDE-209) (Cambridge Isotope Laboratory and Wellington Laboratory) and then Soxhlet extracted (19) with toluene for 16 h. The extracts were spiked with a cleanup recovery standard (¹³C-BDE-126) and then cleaned-up using an acid–base silica gel column (15 mm i.d., packed from bottom to top with silica gel (1 g), basic silica gel (4 g), silica gel (1 g), acid silica gel (8 g), silica gel (2 g), and granular anhydrous sodium sulfate (4 g)) eluted with methylene chloride:hexane (1:1). The eluate was concentrated, exchanged to hexane, and then transferred to an acid alumina column (15 mm i.d., 6 g of acid alumina) for cleanup and eluted with methylene chloride/hexane (1:1). The eluate was concentrated and transferred to a vial containing a recovery standard and nonane for analysis. The extracts were analyzed using an AutoSpec Ultima HRGC/HRMS instrument (splitless injection) with DB-5HT column (30 m × 0.25 mm i.d. × 0.10 μm). The GC temperature program was initiated at 100 °C (held for 1.10 min) and increased to 300 °C at 5 °C/min (held for 5.4 min), and then to 350 °C at 40 °C/min (held for 3.83 min). Forty-three BDE congeners were identified by comparing the GC retention time and ion-abundance ratio of two exact *m/z*s with the corresponding retention time of an authentic standard and the theoretical or acquired ion-abundance ratio of the two exact *m/z*s. Several congeners coeluted and these were reported as a pair (i.e., BDE-28 and BDE-33). Method blanks and ongoing precision and recovery samples were analyzed with each extraction batch and met all of the quality criteria specified by U.S. EPA Draft Method 1614 (20). Average recoveries of the majority of the internal standards and the cleanup recovery standard were monitored to ensure accuracy and ranged from 81 to 116%. Average recoveries of ¹³C₁₂-BDE-3, ¹³C₁₂-BDE-183, and ¹³C₁₂-BDE-209 were 53, 138, and 133%, respectively. No modifications to U.S. EPA Draft Method 1614 were made.

PCDD/Fs. Seventeen 2,3,7,8-substituted PCDD and PCDF congeners and total tetra- through octa- homologue concentrations were measured in compliance with U.S. EPA Method 1613 (Revision B) (21). An aliquot of each sample (5 g) was spiked with a mixture of 16 ¹³C-labeled internal standards (exclusive of ¹³C-1,2,3,7,8,9-HxCDD) (Cambridge Isotope Laboratory and Wellington Laboratory). The samples were Soxhlet extracted with toluene for 16 h followed by acid/base silica gel, acid alumina, and florisil cleanup. ³⁷Cl–TCDD Certified Reference Standard (CRS) was added after extraction, but before cleanup to measure the recovery of the cleanup steps. The extracts were analyzed using an AutoSpec Ultima HRGC/HRMS instrument (splitless injection) with a DB-5HT column (30 m × 0.25 mm i.d. × 0.10 μm). The GC temperature program was initiated at 190 °C (held for 2.13 min) and ramped to 220 °C /min (held for 15.4 min), 290 °C at 7 °C/min (held for 5.14 min), and then to 310 °C at 20 °C/min (7.5 min). Method blanks and ongoing precision and recovery samples were analyzed with each batch and met all of the quality criteria specified by the method. Recovery of internal standards and cleanup recovery standards were monitored to ensure accuracy, and all internal standards met the recovery criteria specified in the method. Average recoveries ranged from 71 to 113%.

Statistical Analysis. Analysis of variance (ANOVA) was performed on all experimental data and means were com-

TABLE 1. Description of Sampling Locations

location	location no.	sample type	description
reservoir (control site)	1	soil	located in the hills in the northern part of Guiyu away from the e-waste center; a water supply plant is currently being constructed to supply tap water from reservoir to residents of certain districts of Guiyu
acid leaching	2	soil	small village, Yaocouwei, located within 2 km from Guiyu; workers involved in acid leaching of shredded printed circuit boards for recovery of precious metals; acid leaching using mixture of concentrated HNO ₃ and HCl carried out along narrow riverbank;
printer roller dump site	3	soil	samples collected from directly underneath acidified shredded printed circuit boards several hundred rubber printer rollers discarded in the Nanyang district of Guiyu; site also served as a storage or dump site for other materials such as metal computer hardware casing, plastic printer casings and computer fans
rice field	4	soil	thick black smoke plumes from e-waste open burning activities were observed from several adjacent rice fields; black smoke had signature irritating acrid smell of burning e-waste; open burning of e-waste in rice fields was a common daily occurrence
riverbank	5	combusted residue	located approximately 10 m from village houses and a family vegetable garden (i.e., spinach); combusted residue of cable wiring and plastic chips;
duck pond	6	soil	samples collected from bank of tributary of the Lianjiang River located beside village houses; over a hundred ducks reared by a farmer; open burning of domestic waste was observed nearby the duck pond in addition to open burning of e-waste in the surrounding rice fields

TABLE 2. Concentration of Selected PBDE Congeners in Soils and Combusted Residue Collected from Guiyu (ng/g, dry wt; n = 3)^a

congener	soil					combusted residue
	acid leaching	printer roller dump site	duck pond	rice field	reservoir	riverbank
BDE-3	8.63	0.040	0.069	0.050	ND ^b	1.06
BDE-7	10.5	0.063	0.068	0.043	0.003	1.30
BDE-15	10.0	0.205	0.120	0.104	0.008	2.34
BDE-28/33	36.3	2.23	0.285	0.282	0.026	7.04
BDE-47	73.6	129	1.78	0.641	0.237	15.4
BDE-49	53.7	8.69	0.854	0.269	0.049	14.8
BDE-66	35.7	8.28	0.426	0.184	0.052	3.36
BDE-71	9.70	0.357	0.057	0.023	ND	1.12
BDE-77	11.3	0.479	0.060	0.028	0.005	1.12
BDE-85	6.10	15.9	0.050	0.014	0.003	1.28
BDE-99	149	333	1.70	0.419	0.199	24.0
BDE-100	22.0	53.1	0.364	0.071	0.039	3.15
BDE-119	13.8	0.628	0.063	0.014	0.012	2.84
BDE-126	5.15	0.133	0.018	0.002	ND	0.445
BDE-138	19.7	5.37	0.211	0.056	ND	75.3
BDE-153	66.2	47.2	3.12	0.558	0.053	1230
BDE-154	53.9	20.6	1.33	0.212	0.035	151
BDE-183	72.2	111	17.3	2.14	0.066	2563
BDE-190	30.9	7.64	0.686	0.139	ND	205
BDE-197	50.7	57.2	8.76	1.04	0.051	3540
BDE-203	109	20.1	3.43	0.35	ND	1230
BDE-207	132	37.4	10.6	1.15	0.102	2300
BDE-209	1270	510	328	37.3	2.76	48600
total PBDEs ^c	3570 ± 781 b	1440 ± 1260 c	398 ± 181 c,d	48.2 ± 19.8 d	3.8 ± 2.2 d	63300 ± 32400 a

^a Detection limits (ng/g): BDE-3 = 0.00958, BDE-71 = 0.00392, BDE-126 = 0.00530, BDE-138 = 0.00991, BDE-190 = 0.0183, BDE-203 = 0.0214
^b ND-not detected under method detection limit. ^c Means having different letters are significantly different at the 5% probability level according to the Duncan's Multiple Range Test.

pared using the Duncan's Multiple Range test with SPSS version 11 software. The significance level was $p \leq 0.05$.

Results and Discussion

Soil Concentrations and Trends. The average total PBDE and PCDD/F concentrations are presented in Tables 2 and 3, respectively. The range of total PBDE concentrations in soil decreased in the following order: acid leaching (2720–4250 ng/g, dry wt) > printer roller dump site (893–2890 ng/g, dry wt) > duck pond (263–604 ng/g, dry wt) > rice field

(34.7–70.9 ng/g, dry wt) > reservoir (2.0–6.2 ng/g, dry wt). The average PBDE concentration at the acid leaching site was greater than that at the reservoir (control site) by over 930 times and was statistically higher ($p < 0.05$) than all of the other sites. Since printed circuit boards contain PBDEs and the soil samples had been collected from directly underneath shredded pieces of printed circuit boards, it may be possible that PBDEs stripped off the printed circuit boards in the acid bath during the acid leaching process, and was then transferred to the soil when spent acid solution was

TABLE 3. Concentration (pg/g and pg WHO-TEQ/g, dry wt) of PCDD/Fs in Guiyu Soils and Combusted Residue from Different Land Uses ($n = 3$)

analyte	TEF (WHO)	soil										combusted residue	
		acid leaching		printer roller dump site		duck pond		rice field		reservoir		riverbank	
		pg/g	pg WHO-TEQ/g	pg/g	pg WHO-TEQ/g	pg/g	pg WHO-TEQ/g	pg/g	pg WHO-TEQ/g	pg/g	pg WHO-TEQ/g	pg/g	pg WHO-TEQ/g
individual congeners													
2,3,7,8-TCDD	1	10.5	10.5	ND ^a	ND	3.58	3.58	1.51	1.51	ND	ND	12.3	12.3
1,2,3,7,8-PeCDD	1	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	30.6	30.6
1,2,3,4,7,8-HxCDD	0.1	ND	ND	0.211	0.021	6.46	0.646	2.88	0.288	ND	ND	16.9	1.69
1,2,3,6,7,8-HxCDD	0.1	32.4	3.24	1.16	0.116	14.1	1.41	5.28	0.528	ND	ND	40.0	4.00
1,2,3,7,8,9-HxCDD	0.1	ND	ND	1.10	0.110	14.8	1.48	6.91	0.691	0.595	0.059	33.4	3.34
1,2,3,4,6,7,8-HpCDD	0.01	240	2.40	8.19	0.082	111	1.11	67.5	0.675	3.70	0.037	195	1.95
OCDD	0.0001	695	0.070	165	0.017	754	0.075	779	0.078	390	0.039	429	0.043
sum		978	16.2	176	0.346	905	8.30	863	3.77	394	0.135	757	53.9
2,3,7,8-TCDF	0.1	459	45.9	4.42	0.442	29.5	2.95	11.7	1.17	0.405	0.041	69.4	6.94
1,2,3,7,8-PeCDF	0.05	641	32.1	3.80	0.190	30.4	1.52	10.0	0.502	0.654	0.033	96.6	4.83
2,3,4,7,8-PeCDF	0.5	498	249	5.24	2.62	26.5	13.2	8.04	4.02	0.707	0.354	79.1	39.5
1,2,3,4,7,8-HxCDF	0.1	464	46.4	4.51	0.451	25.1	2.51	7.37	0.737	0.794	0.079	71.3	7.13
1,2,3,6,7,8-HxCDF	0.1	499	49.9	2.60	0.260	21.7	2.17	6.15	0.615	0.760	0.076	68.0	6.80
2,3,4,6,7,8-HxCDF	0.1	371	37.1	3.38	0.338	19.7	1.97	4.98	0.498	0.674	0.067	62.7	6.27
1,2,3,7,8,9-HxCDF	0.1	196	19.6	0.820	0.082	6.73	0.673	2.65	0.265	ND	ND	19.6	1.96
1,2,3,4,6,7,8-HpCDF	0.01	747	7.47	12.5	0.125	56.5	0.565	13.7	0.137	1.71	0.017	164	1.64
1,2,3,4,7,8,9-HpCDF	0.01	202	2.02	ND	ND	7.20	0.072	1.39	0.014	ND	ND	24.1	0.241
OCDF	0.0001	256	0.026	11.6	0.0012	29.0	0.0029	9.74	9.74E-04	0.350	3.50E-05	78.4	0.0078
sum		4332	489	48.9	4.51	252	25.7	75.8	7.96	6.05	0.667	733	75.4
homologues													
total TCDD		679	10.5	73.0	ND	2344	3.58	625	1.51	12.0	ND	8467	12.3
total PeCDD		596	ND	48.3	ND	1191	ND	287	ND	9.71	ND	3987	30.6
total HxCDD		605	3.24	34.9	0.247	593	3.53	184	1.51	9.02	0.059	1710	9.02
total HpCDD		474	2.40	19.3	0.082	318	1.11	192	0.675	8.82	0.037	452	1.95
total OCDD		695	0.070	165	0.017	754	0.075	779	0.078	390	0.039	429	0.043
total PCDDs		3050	16.2	341	0.346	5200	8.30	2067	3.77	429	0.135	15044	53.9
total TCDF		20243	45.9	87.3	0.442	1157	2.95	396	1.17	15.6	0.041	3283	6.94
total PeCDF		10103	281	62.1	2.81	629	14.8	173	4.52	11.6	0.386	1656	44.4
total HxCDF		4340	153	32.1	1.13	252	7.32	67.6	2.11	6.97	0.223	692	22.2
total HpCDF		1307	9.49	12.5	0.125	91.8	0.637	21.1	0.151	1.71	0.017	265	1.88
total OCDF		256	0.026	11.6	0.0012	29.0	0.0029	9.74	9.74E-04	0.350	3.50E-05	78.4	0.0078
total PCDFs		36250	489	206	4.51	2159	25.7	667	7.96	36.2	0.667	5974	75.4
total PCDD/Fs ^b		39300	506 a	546	4.86 c	7360	34 c	2730	11.7 b	465	0.8 d	21000	129 a
range		12500–89800	203–1100	508–609	2.1–8.2	3870–9700	15–48	2320–3130	10–13	228–834	0.39–1.5	13500–25300	84.3–174

^a ND = not detected under method detection limit. ^b Means having different letters are significantly different at the 5% probability level according to the Duncan's Multiple Range Test.

dumped at the riverbank. The rubber component of printer rollers also contain PBDEs which could have leached out (14) to the soil during warm weather as PBDEs are additive flame retardants and are not chemically bound. However, there was no significant difference found between soil PBDE concentration at the printer roller dump site and at the duck pond, but a significant difference was noted between printer roller dump site and rice field. No significant differences were found among the duck pond, rice field, and reservoir.

The range of average total tetra- to octa-substituted PCDD/F homologues decreased in the following trend: acid leaching > duck pond > rice field > printer roller dump site > reservoir. In terms of PCDD/Fs toxicity equivalency, the order of decreasing toxicity was also the same and ranged from 506 to 0.80 pg WHO-TEQ/g. The concentration at the acid leaching site was significantly higher ($p < 0.05$) than all the other sites with the average concentration being 85 times higher than the reservoir in terms of pg/g and 630 times higher (both $p < 0.05$) in terms of pg WHO-TEQ/g. The elevated PCDD/Fs level at the acid leaching site indicated the proximity of the site to combustion sources. Concentrations at the duck pond and the rice field were higher than those at the printer roller dump site and may have been due to atmospheric transportation and deposition of the pollutants from nearby open burning of e-waste. There were no open burning sites near the printer roller dump site, and the range of concentrations (pg/g) at this site was within the background range found at the reservoir.

Combusted Residue Concentrations and Trends. The total PBDE concentration of the combusted residue was significantly higher ($p < 0.05$) than all of the soil samples and ranged from 33 000 to 97 400 ng/g, dry wt. The average concentration was 18 times and more than 16 000 times greater than at the acid leaching site and the reservoir, respectively. In general, extremely elevated concentrations of PBDEs were found in both combusted residue and in soil from the acid leaching site. The higher PBDE level of the combusted residue was most likely due to the presence of burnt and melted plastic cables and plastic chips treated with PBDEs.

The average PCDD/F concentration (pg/g) of the combusted residue was between 3 and 45 times higher than that of soil at four of the study sites, but interestingly, it was only about two times lower than soil at the acid leaching site. The source of the higher PCDD/Fs level at the acid leaching site was unclear. One suggested reason may be the open burning (ignition by waste paper and cigarette lighter) of dumped and unwanted e-waste along a nearby riverbank. The PCDD/F concentration of the combusted residue was very similar to that reported for combusted residue of soft-PVC plastic resulting from fire accidents at plastic manufacturing or storage facilities (22).

Aside from PCDD/Fs, the burning of electronics containing PBDEs can produce toxic air emissions of brominated [polybrominated dibenzo-*p*-dioxins and furans (PBDD/PBDFs)] and mixed [polybromochlorodibenzodioxins and polybromochlorodibenzofurans (PBCDD/Fs)] substitutions (23). Currently, there is no precise knowledge of their toxicities, however limited studies have shown effects similar to those of TCDD (24).

Homologue Profiles and Congener Concentrations. PBDEs. All the soils and the combusted residue contained congeners belonging to mono- to deca- homologues except for the reservoir soil which did not contain any mono-brominated substitutions (see Figure S1 and Table S1, Supporting Information). Of the PBDE homologues, deca-BDE existed in the highest abundance at all the six sites, accounting for 35–82%, and was statistically higher at the riverbank, acid leaching site, and the printer roller dump site than the three other sites. The respective percentages of

mono-, di-, and tri-BDE homologues did not exceed 5%. The percentage of tetra-BDE homologues at the acid leaching site, printer roller dump site, and the reservoir ranged from 7 to 10.5%. The printer roller dump site contained the highest percentage of penta-BDE (29%). Respective homologues of hexa-BDE to nona-BDE composed between 2 and 11% at all sampling sites.

The printer roller dump site had the highest concentrations of BDE-47, -99, and -100 (Table 2), and the combusted residue had the highest concentrations of higher molecular weight congeners (e.g., -183, -190, -197, -203, -207, and -209). Soil from the acid leaching site also had high concentrations of BDE-47, -99, -100, and high molecular weight congeners.

PCDD/Fs. In general, the dominant 2,3,7,8-substituted PCDD and PCDF congeners were OCDD, 1,2,3,4,7,8-HpCDD, 1,2,3,4,6,7,8-HpCDF, and the tetra- to penta-substituted CDF congeners. 2,3,7,8-TCDD was only detected in soil from the acid leaching, duck pond, and rice field sites, and in combusted residue. The percentage of 2,3,7,8-TCDD compared to total TEQ was highest for the rice field (12.9%) and the duck pond (10.5%). 1,2,3,7,8-PeCDD which also has a toxicity equivalent factor of 1 (25) was below the detection limit at all the sites.

It is important to report congener/homologue-specific data where appropriate because reporting of only the total concentration may mask important differences in source patterns and environmental fate and behavior (26). PCDD/F homologue profiles of the environmental samples can provide information about whether they represent a source or a sink (i.e., collection reservoir) for PCDD/Fs. The PCDD/F homologue profiles (Figure S2, Supporting Information) clearly showed similarities among the printer roller dump site, duck pond, rice field, and the riverbank, whereas the soil profile for reservoir area was distinctly different and dominated by OCDD. Since the reservoir is located away from the e-waste center, it serves as a sink for PCDD/Fs, and therefore any profile that is different from that of the reservoir can generally be regarded as a possible source of PCDD/Fs. The profiles of the samples were also compared to typical PCDD/F homologue profiles (not shown) of (a) municipal waste incinerator emissions (8), (b) 12 different combustion sources (27), and (c) an environmental sink (28). The profile for the acid leaching site resembled the profile of an environmental source (i.e., dominated by tetra- through hexachlorinated furans) because PCDFs accounted for 92% of the total PCDD/Fs concentration. The PCDD/F homologue proportions for printer roller dump site, duck pond, rice field, and riverbank resembled an emission sources profile more than an environmental sink for PCDD/Fs since they had a relatively high percentage of lower chlorinated furans and a decreasing trend from tetra- to heptachlorinated dibenzo-*p*-dioxins. Therefore, there were PCDD/F sources (such as open burning) at these sites or in close proximity. Interestingly, with regard to printer roller dump site and rice field, there was a relatively high percentage of OCDD (30 and 29%, respectively). The duck pond had a lower OCDD percentage (10%). The homologue profile for the reservoir closely resembled the profile of an environmental sink. In this case, OCDD accounted for 84% of the total PCDD/Fs. Its profile was also in agreement with homologue profiles of control soils from India and Vietnam (10). As the reservoir was located in the northern hills of Guiyu, away from e-waste burning, the PCDD/Fs in soil mainly originated from atmospheric deposition. The homologue profiles for the six study areas may provide unique fingerprints for e-waste activities such as open burning and acid leaching thereby identifying the existence of e-waste activities at other locations.

Investigation of Commercial Formulation Profiles at e-Waste Sites. An attempt was made to elucidate the predominant commercial source of PBDEs at the study sites

by comparing the soil and combusted residue PBDE congener profiles to Penta-BDE (i.e., Bromkal 70-5DE), Octa-BDE, and Deca-BDE commercial product profiles since they have major ingredients that are distinct from each other. To elucidate the presence of Bromkal 70-5DE at the study sites, the percentage congener profiles normalized to the sum of the major congeners (BDE-47, -99, -100, -153, and -154) that make up its composition (29) were plotted for each site (Figure S3, Supporting Information). The soil from the reservoir was determined to most resemble the Bromkal 70-5DE profile because its congener percentage compositions were similar. The printer roller dump site also exhibited a similar profile except that the BDE-47:BDE-99 ratio was 1:2.6 instead of 1:1 as in Bromkal 70-5DE (30). Acid leaching site, duck pond, and rice field shared similarities in that BDE-153 was predominant, making up 17, 37, and 29%, respectively, of the concentration of the five congeners found in the commercial product. The combusted residue had a distinctly different profile than the soil samples because BDE-153 and -154 were the major congeners accounting for 82 and 10% of the normalized congener profile, respectively. Therefore, Penta-BDE commercial product was not the major source of PBDE for the combusted residue. Moreover, nearly all of the PBDE congeners in the combusted residue were high molecular weight congeners (hexa- to deca-bromination).

BDE-183 is a marker for the Octa-BDE commercial product (29) because only trace quantities are detected in Penta-BDE and it is not found in Deca-BDE. Therefore, if excluding BDE-209 from the total PBDE concentration, BDE-183 ranged from 3.1 (acid leaching) to 24.6% (duck pond). This suggests that the Octa-BDE formulation is also an important pollutant at Guiyu.

BDE-209 (which makes up 97–98% of the Deca-PBDE commercial product) (31) was approximately 3–5 times higher than the respective Σ (total mono- to nona-BDE) concentrations for the duck pond, rice field, riverbank, and reservoir implying that the technical Deca formula widely exists in the environment of Guiyu and there appeared to be a continual input into the environment. It is uncertain whether deca-BDE debrominated to the lighter molecular weight molecules (i.e., BDE-47, -99), however correlation analysis may provide some information. Analyses by Pearson's correlations showed no significant correlations at the 0.05 and 0.01 probability levels between BDE-209 and BDE-47, and BDE-209 and BDE-99 at the printer roller dump site, rice field, duck pond, and the reservoir suggesting that BDE-47 and -99 did not originate from the decomposition of BDE-209 (32). For the combusted residue, BDE-209 was also not significantly correlated with the penta-BDE formulation.

Comparison With Worldwide PBDE and PCDD/F Levels.

Data on PBDEs in soil are very limited and their reporting in literature has not been consistent (i.e., different congeners) making comparisons among different studies difficult. Where possible, the results of this study were compared with those of other studies using the same congener or homologue basis (see Table S2, Supporting Information).

The reservoir and rice field concentrations of our study were in agreement with background concentrations along a latitudinal transect through UK and Norway grasslands and woodlands (based on 20 congeners which did not include BDE-209, mono-, di-, and octa- and nona-BDE congeners) (29). Comparatively, the average concentration at the printer roller dump site (736 ng/g) was approximately 131 times higher than the highest background concentration (29). The BDE-47 and -99 concentrations at acid leaching site and printer roller dump site were 2–4 and 4–8 times higher, respectively, than concentrations recorded in soil near a United States polyurethane foam manufacturing plant which used Penta-BDE as a flame retardant (33). BDE-209 at the rice field was 19 times greater than an agricultural area in

Japan (34). With regard to e-waste sites, the penta-BDE concentrations in soil from the printer roller dump site and acid leaching site were higher than that reported for soil from an e-waste storage site in Hong Kong (35). In addition, the ratio of BDE-99 and BDE-47 in soil from the printer roller dump site was similar to that of soil from a transformer recycling hotspot in Taizhou, Zhejiang Province, which was probably attributed to commercial Penta-BDE (36). Compared with a soil sample we collected in 2003 from the same printer roller dump site in Guiyu, the congener profiles were similar indicating the same source of PBDEs (37–39). Furthermore, our previous sampling at Guiyu showed that a soil sample from an open dump site for burnt plastic consisted of 1140 ng/g of PBDEs (37), and combusted residue samples consisting of charred plastic, ash, and sand ranged from 8291 to 12 100 ng/g Σ (total mono-BDE to total hepta-BDE homologues) whereby BDE-47 and -99 predominated (40). The PBDE concentrations at Guiyu are some of the highest found in environmental media and in combusted residue in the world.

There are few standards regarding PBDE-contaminated land. The U.S. EPA standard for penta-BDE in residential soil warranting preliminary remediation is 120 mg/kg, and for deca-BDE the standard is 610 mg/kg (41), however, these do not consider potential human health effects. Standards or guidelines for PBDEs which are specific to different land uses (agricultural, playgrounds, commercial) are needed to protect human health.

The background PCDD/Fs toxic equivalency concentration at the reservoir was comparable to background rural soil concentration ranges reported for the United States (0.1–6 pg I-TEQ/g) (42) and in EU Member States (43). In developing Asian countries, the dumping and open burning of municipal waste is contributing an important amount of PCDD/F to the global emission inventory (10). PCDD/Fs concentrations at riverbank, acid leaching site, duck pond, and rice field were comparable to concentrations detected at municipal waste dump sites in Philippines, Cambodia, India, and Vietnam (10). The PCDD/Fs concentration (494 pg WHO-TEQ/g) of an electronic component waste sample collected at Guiyu by Luksemburg et al. (40, 44) consisting of a mixture of combusted residue, soil, and a large percentage of tiny metallic parts was comparable to the concentration of the acid leaching site of this study. Furthermore, three samples of combusted residue had PCDD/F concentrations ranging from 155–14 400 pg WHO-TEQ/g demonstrating the high variability in PCDD/F concentration in combusted residue.

To gain a better understanding of the relative extent of contamination by PCDD/Fs at the sampling sites, the concentrations were compared to international soil guidelines and regulations which specify maximum acceptable levels according to different land use (see Table S3, Supporting Information). China has no soil guidelines for dioxins (45). The concentrations at the sampling sites were below the U.S. guideline (1000 pg TEQ/g), however, 2,3,7,8-TCDD at acid leaching site and at riverbank exceeded the U.S. EPA Regions 6 and 9 risk-based criteria by up to three times (41, 46). PCDD (all sites) and PCDF (all sites except reservoir) levels also exceeded U.S. EPA Region 5 ecological screening levels (47). Exceedence of the Canadian guideline for agricultural land use for rice field was 3-fold (48). 2,3,7,8-TCDD levels at acid leaching site, duck pond, rice field, and riverbank are a cause for concern for wildlife with respect to The Netherlands guideline (49). Overall, concentrations at the sampling sites, except for reservoir, exceeded stricter international guidelines for specific land uses and risk-based criteria.

This study is among the very few studies dealing with the important issue of pollution, with focus on PBDE and PCDD/F

Fs concentrations, generated from crude e-waste recycling. The study has shown that e-waste is a hotspot for PBDEs and PCDD/Fs and investigations with regards to exposure assessment and human health risk assessment are warranted.

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Supporting Information Available

Figures S1, S2, and S3, and Tables S1, S2, and S3. This material is available free of charge via the Internet at <http://pubs.acs.org>.

Literature Cited

- (1) Commission of the European Communities. *Proposal for a directive of the European parliament and of the council on waste electrical and electronic equipment/Proposal for a directive of the European parliament and of the council on the restriction of the use of certain hazardous substances in electrical and electronic equipment*; Presented by the Commission of the European Communities: Brussels, June 13, 2000, COM(2000) 347 final; <http://www.eeb.org/activities/waste/WEEE%20and%20ROS%20ENfinal.pdf>.
- (2) UNEP DEWA/GRID-Europe. E-waste, the hidden side of IT equipment's manufacturing and use; Environment Alert Bulletin 5; UNEP: Nairobi, Kenya, 2005; http://www.grid.unep.ch/product/publication/download/ew_ewaste.en.pdf.
- (3) UNEP. *Report of the Persistent Organic Pollutants Review Committee on the work of its second meeting*; Geneva, November 6–10, 2006; UNEP/POPS/POPRC.2/17.
- (4) Cox, P.; Ethymiou, P. Directive 2003/11/EC of the European parliament and of the council of February 6, 2003 amending for the 24th time Council Directive 76/669/EEC relating to restrictions on the marketing and use of certain dangerous substances and preparations (pentabromodiphenyl ether, octabromodiphenyl ether). *Off. J. Eur. Union OJ L* **2003**, *42*, 45–46.
- (5) de Wit, C. A.; Alae, M.; Muir, D. C. G. Levels and trends of brominated flame retardants in the Arctic. *Chemosphere* **2006**, *64*, 209–233.
- (6) Wenning, R. J. Uncertainties and data needs in risk assessment of three commercial polybrominated diphenyl ethers: probabilistic exposure analysis and comparison with European Commission results. *Chemosphere* **2002**, *46*, 779–796.
- (7) Bromine Science and Environmental Forum. *Major brominated flame retardants volume estimate. Total market demand by region in 2001*; BSEF: Brussels, Belgium, 2003; www.bsef-site.com/bromine/our_industry/.
- (8) Brzuzy, L. P.; Hites, R. A. Global mass balance for polychlorinated dibenzo-*p*-dioxins and dibenzofurans. *Environ. Sci. Technol.* **1996**, *30*, 1797–1804.
- (9) Gullet, B. K.; Lemieux, P. M.; Lutes, C. C.; Winterrowd, C. K.; Winters, D. L. Emissions of PCDD/F from uncontrolled, domestic waste burning. *Chemosphere* **2001**, *43*, 721–725.
- (10) Minh, N. H.; Minh, T. B.; Watanabe, M.; Kunisue, T.; Monirith, I.; Tanabe, S.; Sakai, S.; Subramanian, A.; Sasikumar, K.; Viet, P. H.; Tuyen, B. C.; Tana, T. S.; Prudente, M. S. Open dumping site in Asian developing countries: A potential source of polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans. *Environ. Sci. Technol.* **2003**, *37*, 1493–1502.
- (11) Fiedler, H. Dioxins and furans (PCDD/PCDF). In *Persistent Organic Pollutants, The Handbook of Environmental Chemistry*; Fiedler, H., Ed.; Springer-Verlag: Berlin/Heidelberg, 2003; Vol. 3, Anthropogenic Compounds, Part O, pp 123–201.
- (12) Takasuga, T.; Makino, T.; Umetsu, N.; Senthilkumar, K. Quantitative analysis of toxic compounds formed from combustion of some plastic materials and newspaper. *Organohalogen Compd.* **2003**, *63*, 86–89.
- (13) Shibata, E.; Yamamoto, S.; Kasai, E.; Nakamura, T. Formation behaviour of PCDD/Fs in PVC pyrolysis with copper oxide. *Chemosphere* **2003**, *50*, 1235–1242.
- (14) de Wit, C. A. An overview of brominated flame retardants in the environment. *Chemosphere* **2002**, *46*, 583–624.

- (15) Sjödin, A.; Hagmar, L.; Klasson-Wehler, E.; Kronholm-Diab, K.; Jakobsson, E.; Bergman, Å. Flame Retardant Exposure: Polybrominated Diphenyl Ethers in Blood from Swedish Workers. *Environ. Health Perspect.* **1999**, *107*, 643–648.
- (16) Agency for Toxic Substances and Disease Registry. Toxicological profile for chlorinated dibenzo-*p*-dioxins; ATSDR: Atlanta, GA, 1998.
- (17) Hoque, A.; Sigurdson, A. J.; Burau, K. D.; Humphrey, H. E.; Hess, K. R.; Sweeney, A. M. Cancer among a Michigan cohort exposed to polybrominated biphenyls in 1973. *Epidemiology* **1998**, *9*, 373–378.
- (18) The Basel Action Network and Silicon Valley Toxics Coalition. *Exporting harm: The high-tech trashing of Asia*; BAN and SVTC: San Jose, CA, February 25, 2002.
- (19) U.S. EPA. Method 3540C Revision 3, Soxhlet extraction. In *Test Methods for Evaluating Solid Waste, Physical/Chemical Methods*; SW-846; Washington, DC, 1996; www.epa.gov/epaoswer/hazwaste/test/main.htm.
- (20) U.S. EPA. Draft Method 1614: Brominated diphenyl ethers in water, soil, sediment, and tissue by HRGC/HRMS. In *Test Methods for Evaluating Solid Waste, Physical/Chemical Methods*; SW-846; Washington, DC, August 2003.
- (21) U.S. EPA. Method 1613 (Revision B). Tetra- through octachlorinated dioxins and furans by isotope dilution HRGC/HRMS. In *Test Methods for Evaluating Solid Waste, Physical/Chemical Methods*; SW-846; Washington, DC, September 1997.
- (22) Thiesen, J.; Funcke, W.; Balfanz, E.; König, J. Determination of PCDFs and PCDDs in fire accidents and laboratory combustion tests involving PVC-containing materials. *Chemosphere* **1989**, *19*, 423–428.
- (23) Sakai, S.; Watanabe, J.; Honda, Y.; Takatsuki, H.; Aoki, I.; Futamatsu, M.; Shiozaki, K. Combustion of brominated flame retardants and behaviour of its byproducts. *Chemosphere* **2001**, *42*, 519–531.
- (24) Birnbaum, L. S.; Staskal, D. F.; Diliberto, J. J. Health effects of polybrominated dibenzo-*p*-dioxins (PBDDs) and dibenzofurans (PBDFs). *Environ. Int.* **2003**, *29*, 855–860.
- (25) Van den Berg, M.; Birnbaum, L.; Bosveld, A. T. C.; Brunström, B.; Cook, P.; Feeley, M.; Giesy, J. P.; Hanberg, A.; Hasegawa, R.; Kennedy, S. W.; Kubiak, T.; Larsen, J. C.; van Leewen, F. X. R.; Liem, A. K. D.; Nolt, C.; Peterson, R. E.; Poellinger, L.; Safe, S.; Schrenk, D.; Tillitt, D.; Tysklind, M.; Younes, M.; Wærn, F.; Zacharewski, T. Toxic equivalency factors (TEFs) for PCBs, PCDDs, PCDFs for humans and wildlife. *Environ. Health Perspect.* **1998**, *106*, 775–792.
- (26) Duarte-Davison, R.; Sewart, A.; Alcock, R. E.; Cousins, I. T.; Jones, K. C. Exploring the balance between sources, deposition, and the environmental burden of PCDD/Fs in the U.K. terrestrial environment: an aid to identifying uncertainties and research needs. *Environ. Sci. Technol.* **1997**, *31*, 1–11.
- (27) U.S. EPA. *Database of sources of environmental releases of dioxin-like compounds in the United States*; External Review Draft, EPA/600/P-98/002Ab; The Office of Research and Development, National Center for Environmental Assessment: Washington, DC, April 1998.
- (28) Baker, J. I.; Hites, R. A. Is combustion the major source of polychlorinated dibenzo-*p*-dioxins and dibenzofurans to the environment? A mass balance investigation. *Environ. Sci. Technol.* **2000**, *34*, 2879–2886.
- (29) Hassanin, A.; Breivik, K.; Meijer, S. N.; Steinnes, E.; Thomas, G. O.; Jones, K. C. PBDEs in European background soils: Levels and factors controlling their distribution. *Environ. Sci. Technol.* **2004**, *38*, 738–745.
- (30) Sjödin, A.; Jakobsson, E.; Kierkegaard, A.; Marsh, G.; Sellström, U. *J. Chromatogr. A* **1998**, *822*, 83–89.
- (31) WHO/IPCS. Environmental Health Criteria 162: Brominated diphenyl ethers. World Health Organisation, Geneva, Switzerland, 1994.
- (32) Mai, B. X.; Chen, S. J.; Luo, X. J.; Chen, L. G.; Yang, Q. S.; Sheng, G. Y.; Peng, P. A.; Fu, J. M.; Zeng, E. Y. Distribution of polybrominated diphenyl ethers in sediments of the Pearl River Delta and adjacent South China Sea. *Environ. Sci. Technol.* **2005**, *39*, 3521–3527.
- (33) Hale, R. C.; La Guardia, M. J.; Harvey, E.; Mainor, T. M. Potential role of fire retardant-treated polyurethane foam as a source of brominated diphenyl ethers to the US environment. *Chemosphere* **2002**, *46*, 729–735.
- (34) MOE Japan, (Ministry of the Environment, Office of Environmental Risk Assessment, Environmental Health Department, Japan). *Findings of the fiscal 2000 survey on brominated dioxins*; 2001.

- (35) Hong Kong Environmental Protection Department. *Press Release: Test results of soil samples collected from e-waste workshops*; 2005; http://www.epd.gov.hk/epd/english/news_events/press/press_050421a.html.
- (36) Cai, Z. W.; Jiang, G. B. Determination of polybrominated diphenyl ethers in soil from e-waste recycling site. *Talanta* **2006**, *70*, 88–90.
- (37) Leung, A.; Cai, Z. W.; Wong, M. H. Environmental contamination from electronic waste recycling at Guiyu, southeast China. *J. Mater. Cycles Waste Manage.* **2006**, *8*, 21–33.
- (38) Wang, D. L.; Cai, Z. W.; Jiang, G. B.; Leung, A.; Wong, M. H.; Wong, W. K. Determination of polybrominated diphenyl ethers in soil and sediment from an electronic waste recycling facility. *Chemosphere* **2005**, *60*, 810–816.
- (39) Wang, D. L.; Cai, Z. W.; Jiang, G. B.; Wong, M. H.; Wong, W. K. Gas chromatography/ion trap mass spectrometry applied for the determination of polybrominated diphenyl ether in soil. *Rapid Commun. Mass Spectrom.* **2005**, *19*, 83–89.
- (40) Luksemburg, W. J.; Mitzel, R. S.; Peterson, R. G.; Maier, M. M.; Schuld, M.; Hedin, J. M.; Zhou, H. D.; Wong, A. S. Polychlorinated dibenzodioxins and dibenzofurans (PCDDs/PCDFs) and polybrominated diphenylether (PBDE) levels in environmental and human hair samples around an electronic waste processing site in Guiyu, Guangdong Province, China. Poster presented at 22nd International Symposium on Halogenated Environmental Organic Pollutants and Persistent Organic Pollutants (POPs) (Dioxins 2002); Barcelona, Spain, August 11–16, 2002; Spanish Council for Scientific Research Laboratory for Dioxins.
- (41) U.S. EPA. *Region 9 (The Pacific Southwest): Superfund Preliminary Remediation Goals*; U.S. EPA, Region 9: San Francisco, CA, 2004.
- (42) U.S. EPA. *Exposure and human health reassessment of 2,3,7,8-tetrachlorodibenzo-p-dioxins and related compounds. Part I: Estimating exposure to dioxin-like compounds – Volume 2: Sources of dioxin-like compounds in the United States*; Draft Final Report, EPA/600/P-00/001Bb; National Center for Environmental Assessment, U.S. EPA: Washington, DC, September, 200; www.epa.gov/ncea/pdfs/dioxin/dioxreass.htm.
- (43) Fiedler, H.; Buckley-Golder, D.; Coleman, P.; King, K.; Petersen, A. Compilation of EU dioxin exposure and health data: Environmental levels. *Organohalogen Compd.* **1999**, *43*, 151–154.
- (44) Luksemburg, W. J.; Mitzel, R. S.; Peterson, R. G.; Maier, M. M.; Schuld, M.; Hedin, J. M.; Zhou, H. D.; Wong, A. S. Polychlorinated dibenzodioxins and dibenzofurans (PCDDs/PCDFs) levels in environmental and human hair samples around an electronic waste processing site in Guiyu, Guangdong Province, China. *Organohalogen Compd.* **2002**, *55*, 347–349.
- (45) Zheng, M. H. *Progress and prospects for dioxins analysis in China*; Presented at The China International Symposium on Persistent Toxic Substances; Beijing, China, 2004; Key Laboratory of Environmental Chemistry and Ecotoxicology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences; 2004.
- (46) U.S. EPA. *Region 6 (South Pacific): Human Health Medium-Specific Screening Levels*; U.S. EPA: Washington, DC, 2004.
- (47) U.S. EPA. *Region 5 (Illinois, Indiana, Michigan, Minnesota, Ohio, Wisconsin and 35 Tribes), RCRA Ecological Screening Levels*; U.S. EPA: Chicago, IL, August 22, 2003.
- (48) Canadian Council of Ministers of the Environment. *Canadian Environmental Quality Guidelines, Summary Table, Update*; December 2003.
- (49) Health Council of the Netherlands. *Polychlorinated dibenzo-p-dioxins, dibenzofurans and dioxin-like polychlorinated biphenyls*; Publication No. 1996/10E; Committee on Risk Evaluation of Substances/Dioxins, 1996.

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