

ORIGINAL ARTICLE

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## Characterization of air emissions and residual ash from open burning of electronic wastes during simulated rudimentary recycling operations

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**Abstract** Air emissions and residual ash samples were collected and analyzed during experiments of open, uncontrolled combustion of electronic waste (e-waste), simulating practices associated with rudimentary e-waste recycling operations. Circuit boards and insulated wires were handled separately to simulate processes associated with metal recovery. The average emissions of polychlorinated dibenzodioxins and dibenzofurans (PCDD/PCDFs) were 92 ng toxic equivalency (TEQ)/kg [ $n = 2$ , relative standard deviation (RSD) = 98%] and 11 900 ng TEQ/kg ( $n = 3$ , RSD = 50%) of the initial mass of the circuit boards and insulated wire, respectively. The value for the insulated wire is about 100 times higher than that for backyard barrel burning of domestic waste. The emission concentrations of polybrominated dibenzodioxins and dibenzofurans (PBDD/PBDFs) from the combustion of circuit boards were 100 times higher than for their polychlorinated counterparts. Particulate matter (PM) sampling of the fly ash emissions indicated PM emission factors of approximately 15 and 17 g/kg of the initial mass for the circuit boards and insulated wire, respectively. Fly ash samples from both types of e-waste contained considerable amounts of several metallic elements and halogens; lead concentrations were more than 200 times the United States regulatory limits for municipal waste combustors and 20 times those for secondary lead smelters. Leaching tests of the residual bottom ash showed that lead concentrations exceeded U.S. Environmental Protection Agency landfill limits, designating this ash as a hazardous waste.

**Key words** Electronic waste · Emissions · Ash · Recycling · PCDD · PCDF · PBDD · PBDF · Metals · TCLP · Flame retardants

### Introduction

The fate of electronic wastes (e-wastes) is a growing worldwide environmental issue. The decreasing costs and increasing availability of electronic products of all kinds, including cellular telephones, audio and video equipment, and personal computers and their accessories, coupled with advances in technology that rapidly make these products obsolete, foretell a growing disposal problem. Government estimates project that nearly 250 million computers in the United States became obsolete by 2005 and that this amount will double by 2007.<sup>1</sup> While most of these e-wastes currently enter the solid waste stream and end up in municipal landfills or municipal waste combustors, a small but growing portion is directed for recycling. Recycling is carried out by a small number of domestic recycling programs, as well as an undocumented number of foreign operations, most located in developing nations, including China, India, and Pakistan.<sup>2</sup> China reportedly prohibits the import of used electronic products, although a number of illegal shipments have been documented.<sup>3</sup> Of the obsolete computers from the USA that are recycled, it is estimated that 50% to 80% are exported for processing in developing nations by rudimentary recycling operations.<sup>2</sup>

These low-technology recycling practices typically consist of labor-intensive steps to separate component parts, including plastic cases, metal chassis, cathode ray tubes (CRTs), circuit boards, wires, and printer toner cartridges. Cases and chassis can be sold for their scrap value, while the CRTs, circuit boards, and wires are further processed to recover reusable components, copper, tin, and trace amounts of precious metals. Toner cartridges are opened to recover the remaining toner. CRTs are broken to recover the copper contained in the electron gun. Obsolete electronic chips are removed from circuit boards and dissolved in strong acidic

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solutions from which gold, silver, palladium, and platinum can be recovered. Wire is often burned in the open to remove the plastic insulation to facilitate copper recovery. Circuit boards are also often burned with supplemental fuels in outdoor fires to reduce them to metals. Subsequently, these metals (primarily copper and tin) are separated from the ash by water floatation. The remaining waste and unusable materials, including the CRT glass, acid solutions, and miscellaneous component parts, are either dumped or burned. These practices often take place with little, if any, regard for worker safety or environmental degradation.<sup>4</sup> A detailed description of these various rudimentary recycling processes is presented by Puckett et al.<sup>4</sup>

The unmonitored rudimentary recycling operations described above have largely unmeasured impacts on the local and regional environments. E-waste contains significant levels of toxic elements, including antimony, lead, nickel, tin, and zinc, and exposure to these elements can result in significant health effects and environmental degradation. Further, e-waste contains high levels of halogens. Circuit boards, in particular, contain high concentrations of brominated flame retardants, while wires are often insulated with polyvinylchloride (PVC) plastic. Combustion of e-waste not only has the potential of vaporizing large quantities of semivolatile toxic elements, but also has the potential to produce toxic halogenated organic pollutants, including polychlorinated and polybrominated dibenzo-*p*-dioxins and furans.<sup>5</sup> These organic compounds are persistent; bio-accumulate in the environment; and have toxic effects on reproductive, developmental, and immunological functions.<sup>6,7</sup> A study examining polychlorinated dibenzodioxin and polychlorinated dibenzofuran (PCDD/PCDF) concentrations in sediment, ash, and human hair from the town of Guiyu, China, a site of e-waste recycling, indicated environmental contamination and human exposure as a result of these practices. Analysis of human hair collected from barber shops near Guiyu found PCDD/PCDF concentrations at least 16 times higher than have been previously reported for hair samples.<sup>8</sup> Environmental sampling of ash, soil, and sediments from this area shows considerably elevated levels of many metals as well as identification of a broad array of chlorinated and brominated aromatics,<sup>9</sup> including polybrominated diphenyl ethers (PBDEs), similar to the technical formulations of fire retardants.<sup>10</sup> It is clear that these rudimentary recycling operations may often be conducted under conditions of considerable risk to the recyclers and to the environment.

There are few reports of PCDD/PCDF emission factors for open burning of electronic waste in the literature. Harnly et al.<sup>11</sup> report an analysis of ash/soil matrices from scrap wire open-burn sites, but their concentration values in this matrix cannot be extrapolated for inventory purposes (emissions per mass of waste wire), nor is it apparent how well the ground deposition reflected the emissions as a whole. As a recourse to this dearth of open burning data, reports for emissions from commercial scrap wire recovery facilities cite values of 15.8 ng international toxic equivalents (I-TEQ)/kg<sup>12</sup> and 7.4–14 ng I-TEQ/kg for PVC-containing materials, 40 ng I-TEQ/kg for industrial cable burning,

2280 ng I-TEQ/kg for lead cable, and 3.3 ng I-TEQ/kg when burning scrap motors for wire recovery.<sup>13</sup> For unregulated, open burning of power cable scrap, Bremmer et al.<sup>13</sup> used estimates of 500 ng I-TEQ/kg for inventories in the Netherlands. However, given the significantly higher PCDD/PCDF emission factors and variability obtained during open, uncontrolled burning of household waste<sup>14,15</sup> compared to well-controlled commercial municipal solid waste incinerators, it is not clear how relevant these reported values from commercial activities are to the open burning of e-waste.

Emissions of polybrominated dibenzo-*p*-dioxins and furans (PBDDs/PBDFs) from controlled combustion of materials containing brominated flame retardants, including electronic products, have been measured<sup>16</sup> at levels between 3000 and 130 000 µg/kg. Other tests have examined emissions from accidental fire simulations of electronic products.<sup>17,18</sup> In these references, measurable levels of brominated and chlorinated DDs/DFs were observed, as well as mixed brominated/chlorinated DDs/DFs (when analytically targeted).

We could find no previously published study that reported air emissions of metals from the open burning of e-waste. However, studies by Lemieux et al.<sup>19</sup> and Stewart and Lemieux<sup>20</sup> describe the behavior of metals during the controlled incineration of e-wastes. These studies were performed in a pilot-scale rotary kiln incinerator with an afterburner to examine process parameters that would fully oxidize the organic constituents but allow recovery of the metallic components in the ash residues. They burned batch charges of chipped circuit boards at kiln temperatures ranging from 570° to 880°C with afterburner temperatures between 760° and 815°C, and reported notable emissions of copper, lead, and antimony (1000–8000 µg/m<sup>3</sup>) and lesser emissions of cadmium, manganese, nickel, barium, arsenic, chromium, cobalt, and beryllium. The kiln residual ash did retain much of the metallic components, although closure of the mass balance was not possible. Both the fly ash and residual ash failed a toxicity characteristic leach profile (TCLP) leaching test because of soluble lead. These researchers also reported very low PCDD/PCDF emissions, although they did note emissions of four brominated organics. PBDD/PBDF emissions were not determined, and it is unclear if these species may be formed preferentially to PCDDs/PCDFs. Total bromine (Br<sup>-</sup> + Br<sub>2</sub>) and chlorine (Cl<sup>-</sup> + Cl<sub>2</sub>) emissions were 30 and 8 mg/m<sup>3</sup>, respectively.

The objectives of this research were to provide an initial assessment of the air emissions and ash leaching characteristics from laboratory simulations of open burning practices that often accompany rudimentary e-waste recycling operations. Of particular interest are air emissions of toxic metals and PCDDs/PCDFs from the open burning of circuit boards and PVC-insulated wire as well as PBDDs/PBDFs emissions from the open burning of circuit boards. Because the scope of the tests was only to provide preliminary characterization of emissions, only a limited number of samples, two e-waste compositions, and noncomprehensive sampling regimes were employed. Therefore, the preliminary emission factors reported here for particles, elements, PCDDs/PCDFs, and PBDDs/PBDFs from the open burning of

circuit boards and insulated wire should be supplemented by additional comprehensive sampling to characterize a larger set of emitted pollutants and variations of e-waste composition and burning practices.

## Experimental

### Test facilities

Tests were conducted in a fully enclosed shed 3.0 m wide  $\times$  2.8 m deep  $\times$  2.1 m high, modified to serve as an open burn test facility. This arrangement has been used previously to examine emissions from the open burning of residential waste,<sup>14,15</sup> wheat straw,<sup>21</sup> forest biomass,<sup>22</sup> and chromated copper arsenate (CCA)-treated wood.<sup>23</sup> The facility details are documented more fully in the above articles. To ensure adequate simulation of open burning, oxygen depletion and excess temperatures were avoided by the supply of exterior dilution air (approximately one volume change every 2 min). Fans and flow deflectors were positioned within the shed to enhance internal air circulation, while preventing the incoming air from impinging directly on the burning waste. The inner walls of the facility were aluminum-foil-covered gypsum board attached to the metal frame of a metal-walled structure. The aluminum foil was certified clean (ASTM B-479) and was changed between different experiments to prevent cross-contamination. The floor was made of poured concrete. From the shed, all the dilution air and combustion emissions were removed by an induced-draft fan through a 20.3-cm-diameter transfer duct and through a MAC 72RT21 (Kansas City, MO, USA) baghouse with 18.2 m<sup>2</sup> of singed Dacron polyester bags before being discharged to the atmosphere. The total flow exiting the facility was metered within the transfer duct and a number of thermocouples were used to measure temperatures within the test facility. Extractive samples to characterize the gas and particle emissions were taken from the transfer duct at an average temperature of 33°C.

### E-waste

Two types of e-waste were collected from obsolete personal computers and burned separately. These two types were printed circuit boards (with the integrated circuit chips removed) and insulated wire (double insulated power cables with plugs attached). Table 1 presents the elemental composition of these two e-wastes using two methods. To perform this analysis, several circuit boards were cut into small pieces (<4 cm), mixed, ground into a powder, mixed again, and a portion digested in a nitric acid/hydrofluoric acid (HNO<sub>3</sub>/HF) solution prior to analysis by inductively coupled argon plasma (ICP) spectroscopy (U.S. EPA M-6020<sup>24</sup>). A length of insulated wire was cut into small pieces (<1 cm) and also digested in HNO<sub>3</sub>/HF prior to ICP analysis. Additional analyses were conducted using a wavelength-dispersive X-ray fluorescence (WD-XRF) spectrometer (Philips Model 2404, Panalytical, Natick, MA, USA). Flat

portions of circuit board, without electronic chips, were analyzed. The insulated wire was dissected, excluding the plugs, and its composition determined gravimetrically and by WD-XRF. The WD-XRF data were collected by Panalytical's SuperQ software and analyzed by UniQuant 5 (Omega Data Systems, Veldhoven, The Netherlands) using the AnySample calibration.

Samples were prepared in duplicate with average concentrations reported in Table 1. As expected, the insulated wire had a large copper content (35.2%) and small quantities of other metals. The circuit board contained large quantities of copper (15.2%) as well as significant quantities of bromine (1.7%), tin (2.7%), lead (1.4%), zinc (2.0%), chlorine (0.2%), and nickel (0.2%). The circuit board samples also contained notable quantities of antimony, barium, and manganese.

Table 2, taken from Lemieux et al.,<sup>19</sup> presents a proximate and ultimate analysis of similar circuit board e-waste, typically performed to characterize fuels. Unlike the insulated wire that was composed primarily of copper (35% by mass) and PVC-based insulation (65% by mass, composed of PVC at 18 100 kJ/kg plus inorganic fillers),<sup>25</sup> the materials composing the circuit board waste were much less well characterized. Evident from Table 2 is that the circuit board e-waste had a relatively small carbon content (18%) and a large ash component (66%). This is consistent with the low measured heat of combustion (7000 kJ/kg), which is less than half that of PVC. In comparison, common fuels, including bituminous coals and No. 2 fuel oils, have heats of

**Table 1.** Elemental composition ( $\mu\text{g/g}$ ) of circuit board and insulated wire e-waste

Element	Circuit board <sup>a</sup>	Insulated wire <sup>a</sup>
Al	1500	160
Sb	824 <sup>b</sup>	840
As	8.12 <sup>b</sup>	ND <sup>c</sup>
Ba	530 <sup>b</sup>	ND
Br	17000	ND
Ca	1300	26600
Cl	1900	88400
Cr	4.58 <sup>b</sup>	ND
Cu	152000 <sup>b</sup>	352000
Fe	90.5 <sup>b</sup>	110
Ge	<1 <sup>b</sup>	ND
Pb	13900 <sup>b</sup>	4000
Mg	139 <sup>b</sup>	8600
Mn	604 <sup>b</sup>	ND
Ni	2160 <sup>b</sup>	50
Se	18.40 <sup>b</sup>	ND
Si	39900	7700
Sr	195 <sup>b</sup>	ND
S	17500	240
Sn	27200 <sup>b</sup>	ND
Zn	19500 <sup>b</sup>	ND
Zr	1.99 <sup>b</sup>	ND

<sup>a</sup> Flat portions of circuit board were measured by wavelength-dispersive X-ray fluorescence (WD-XRF) spectrometry. Wire was dissected and components determined gravimetrically and by WD-XRF. Wire analysis does not include the insulated plugs

<sup>b</sup> Composition of ground circuit board determined using HNO<sub>3</sub>/HF digestion and inductively coupled plasma mass spectrometry (ICP-MS) analysis (U.S. EPA<sup>33</sup>, U.S. EPA<sup>23</sup>)

<sup>c</sup> ND, not detected

combustion of approximately 30500 and 45500kJ/kg, respectively.<sup>26</sup> Although not measured in these analyses, most circuit boards (96%) contain tetrabromobisphenol-A (TBBA) fire retardants,<sup>27</sup> and these may have contributed to the volatile materials. With their halogen contents and low heats of combustion, neither the circuit board nor the insulated wire exhibit characteristics of high-quality fuels, and were in fact, difficult to ignite and sustain combustion without an external ignition and fuel source.

## Procedure

After several trials, it was decided that the e-waste should be burned with sufficient quantities of charcoal to sustain combustion and to mimic the observed practice<sup>4</sup> in which honeycomb coal (coal mixed with organic-rich clay-like sediments) is used to initiate and maintain the fires. Charcoal was chosen because it burns fairly cleanly without contributing significant quantities of halogens or elements of interest to the experiment. Charcoal combustion blanks were tested, so that its contributions to the e-waste emissions data could be

**Table 2.** Proximate and ultimate analysis of circuit board e-waste (adapted from Lemieux et al.<sup>19</sup>)

Proximate analysis (wt %)	
Moisture	0.74
Volatile matter	31.38
Fixed carbon	1.58
Ash	66.3
Ultimate analysis (wt %)	
Carbon	18.27
Hydrogen	1.63
Nitrogen	0.51
Sulfur	0.09
Oxygen <sup>a</sup>	12.71
Ash	66.79
Heat of combustion (kJ/kg)	6960

<sup>a</sup>Oxygen determined by difference

determined. Table 3 summarizes ten combustion experiments, including two charcoal combustion blanks, three circuit board experiments, and four insulated wire experiments. A 24-h facility blank (air only, no combustion) was also performed. The e-wastes and charcoal supplemental fuel were burned within a 112 × 112 × 15 cm sand-filled pan supported on an electronic balance for burn rate (mass loss) determination. After the charcoal was ignited (with commercial charcoal lighter fluid, a mixture of petroleum distillates, primarily light and medium aliphatic compounds) the facility door was closed and sampling begun. The charcoal charges were sufficient to maintain combustion bed temperatures over the duration of sampling.

## Emission measurements

Continuous gas analyzers were used for measurement of carbon monoxide (CO), carbon dioxide (CO<sub>2</sub>), oxygen (O<sub>2</sub>), and total hydrocarbon (THC). The sampled gas was transported from the facility interior through a transfer duct to a continuous emission monitor (CEM) cabinet via heated (120°C) Teflon tubing and was filtered using a heated (120°C) quartz filter. The emission gases were dried with a refrigerated air drier and silica desiccant prior to CO, CO<sub>2</sub>, and O<sub>2</sub> measurement. The THC analyzer used a separate heated line to convey the sample for analysis. CEMs were calibrated with compressed gases before and after sampling, including range midpoints, similar to the procedures outlined in EPA Method 6c<sup>28</sup> (M-6c). Potential bias due to losses in the sample transfer line was monitored by injecting the calibration gases both at the point of sampling and at the inlet of the gas analyzers. Temperature measurements were taken with Type K thermocouples.

PCDD/PCDF samples were taken for four experiments within the facility via a modified ambient sampling procedure, EPA Method TO-9<sup>29</sup> (TO-9), using a Graseby PS-1

**Table 3.** Experiment and sample summary

Sample	Description	Charcoal charge (kg)	E-waste charge (kg)	Chamber flow rate (dsm <sup>3</sup> /min)	Sample methods <sup>a</sup>	Sample time (min)
011504	Charcoal blank	4.36	0	14.4	M-29	120
011604	Charcoal blank	9.37	0	16.3	MOUDI	90
020904	Circuit board	4.79	2.29	16.6	M-29	90
011304	Circuit board	4.76	2.31	15.8	TO-9	210
					MOUDI	21
022004	Circuit board	4.49	2.50	17.0	TO-9	180
					M-23	180
021004	Insulated wire	4.85	2.43	16.0	M-29	60
011404	Insulated wire	4.70	4.81	12.0	TO-9	180
					MOUDI	15
022404	Insulated wire	4.08	2.77	16.9	TO-9	180
					M-23	180
020805	Insulated wire	2.54	0.91	28.0	TO-9 <sup>b</sup>	211

dsm<sup>3</sup>, dry standard cubic meter

<sup>a</sup>Sampling methods/instruments used include: M-29, EPA Method 29 (U.S. EPA<sup>33</sup>); M-23, EPA Method 23 (U.S. EPA<sup>31</sup>); TO-9, EPA Toxic Organic-9 (U.S. EPA<sup>29</sup>); and MOUDI, micro orifice uniform deposition impactor

<sup>b</sup>Sample collected at the transfer duct with a high-volume sampler

sampler (see Table 3). A fifth TO-9 sample was taken from the transfer duct using a recently-acquired high volume PS-1 sampler through an extractive probe. All the PCDD/PCDF samples were analyzed via EPA Method 8290<sup>30</sup> (M-8290). One circuit board experiment was additionally sampled from the transfer duct via a modified EPA Method 23 procedure<sup>31</sup> (M-23) for PBDD/PBDF compounds (described below). The TO-9 sampling trains consisted of an open-faced filter holder followed by an XAD-2 bed vapor trap sandwiched between two polyurethane foam (PUF) plugs. The sample flow rates for the regular TO-9 sample trains were between 35 and 55 l/min [all flow rates cited are at dry, standard (0°C, 1 atm) conditions], while the sample flow rate for the high volume TO-9 sample was 220 l/min. The nominal sample volume flow rate for the M-23 train was 20 l/min. Sampling for PXDD/PXDF (X-halogen) compounds was performed for periods lasting between 3 and 4 h. Sampling was stopped when the CO, CO<sub>2</sub>, and THC concentrations returned to the preburn conditions in the facility.

The PUF/XAD-2/PUF cartridge, including filters, were Soxhlet-extracted in toluene using Dean–Stark apparatus to remove the low-boiling solvents that were left over from equipment rinses. The resulting extracts were dark brown when concentrated to 1 ml in toluene. Half of the extract was solvent-exchanged into hexane and then run through a Fluid Management System's Power-Prep clean-up system (Waltham, MA, USA). The resulting toluene sample was concentrated to 100 µl and then analyzed by high-resolution mass spectroscopy (HRMS) for PCDDs/PCDFs (60-m DB-5MS column; MicroMass Ultima AutoSpec, Analytical Perspectives, Wilmington, NC, USA). All PCDD/PCDF values are reported with potential nondetected (ND) isomers as zero. The ratio of the sampled volume to that of the facility air volume input provided the total mass of emitted PCDD/PCDF. The units presented in this work are mass PCDD/PCDF (tetra- to octachlorinated) per mass of initial e-waste or, alternatively, mass toxic equivalent (TEQ<sub>WHO98</sub><sup>32</sup>) per mass of initial e-waste.

The circuit board extract was analyzed by low-resolution mass spectroscopy (LRMS, Hewlett Packard 5971, Palo Alto, CA, USA) for the mono- to tri-BDDs/BDFs only [due to atomic mass unit (amu) range limitations on the mass spectrometer] using a single-point standard calibration. The calibration used one mono-BrDD isomer, a di-BrDD, and a di-BrBDF. No tribrominated compounds were used. The mono-BDFs were quantified as if they had the same response as the mono-BDD, the tri-BDF as the di-BDF, and the tri-BDD as the di-BDD. In each case, the single isomer was used as the calibration for all the isomers in the homologue. Identifications were made via isotopic patterns, expected fragmentation patterns, and general retention time comparisons.

Noncombustion, blank samples (24 h) from the facility operating with airflow input ensured that the sampling and analysis methods, potential facility contamination, and ambient feed air concentrations showed nondetectable PCDDs/PCDFs and the absence of sample bias. Emission testing of the charcoal combustion alone was also performed

and its PCDD/PCDF emissions were insignificant compared to the final measured levels from the e-waste (three orders of magnitude lower).

Particles were sampled isokinetically from the transfer duct by EPA Method 29<sup>33</sup> (M-29). The M-29 samples, collected at a nominal flow rate of 20 l/min on “as received” quartz filters, provided information on total particulate matter (PM) emissions and elemental concentrations. M-29 sampling was conducted until visible emissions had ceased: 60 and 90 min into the burn for insulated wires and circuit boards, respectively.

Additional samples were directed to a micro-orifice uniform deposition impactor (MOUDI Model 110, MSP, Minneapolis, MN, USA). The MOUDI samples, collected on polycarbonate membrane substrates, were used for additional size-dependent analyses. One M-29 and one MOUDI sample were collected for each blank, circuit board, and insulated wire combustion experiment. The MOUDI is a 10-stage, 30 l/min, inertial cascade impactor capable of measuring particle size distributions (PSDs) between 0.056 to 10 µm. To help deter particle bounce, the polycarbonate substrates were coated with Apiezon type L vacuum grease (Apiezon, Rochester, NY, USA) diluted 20 to 1 with hexane and applied with a nitrogen gas airbrush. After coating, the substrates were placed in an oven (85°C) for 1 h, cooled, and placed in vacuum desiccators for 24 h. M-29 filters and MOUDI polycarbonate substrates were weighed according to EPA Method 5 procedures<sup>34</sup> (M-5) including a 24 h and 6 h desiccation between successive weights. The 6 h desiccation step was repeated until the weights agreed within 0.5 mg. MOUDI samples were collected over a period of 15–20 min to prevent excessive substrate loading and measurement bias. Longer sampling times with dilution were not attempted due to the scoping nature of these tests. These sampling times are notably shorter than those for the TO-9, M-23, and M-29 samples and, unlike the organic and particulate samples, represent only the first portion of the e-waste burn cycle. Thus, the size-classified particulate samples from the MOUDI may be less representative of the overall combustion event than the M-29 samples, since the latter smoldering portions of the combustion cycle were not sampled representatively.

The M-29 and MOUDI samples were analyzed for elemental concentrations using WD-XRF. The MOUDI polycarbonate substrates were covered with 4-µm Prolene film (Chemplex Industries, Stuart, FL, USA) and mounted in polyethylene holders prior to WD-XRF analysis. Two 41-mm-diameter punches cut from each of the 100-mm M-29 quartz filter samples were analyzed lying flat in stainless steel sample holders by WD-XRF.

#### Emission factor calculations

Emission factors in this work were calculated using the initial mass of e-waste as the basis. A common alternative basis, the actual mass of e-waste consumed during combustion, could not be reliably distinguished from the mass loss of the charcoal supplementary fuel.

## Ash characterization

Residual ash samples from both the circuit board and insulated wire tests were analyzed by Oxford Laboratories (Wilmington, NC, USA) for soluble chloride by EPA Method 325.2<sup>35</sup> and for a number of other elements using inductively coupled plasma mass spectroscopy (ICP-MS) (Perkin Elmer, Model Elan 6000, Boston, MA, USA). Samples prepared for ICP-MS were digested in HNO<sub>3</sub>/HF and analyzed following EPA M-6020.<sup>24</sup> Copper, iron, and magnesium were analyzed by atomic absorption spectroscopy (AAS) using EPA Method 7000A.<sup>36</sup> These samples unavoidably included portions of charcoal ash and sand with the e-waste ash. In addition, one combined sample of circuit board and insulated wire ash was analyzed to determine if it would be characterized as toxic hazardous waste under federal regulations<sup>37</sup> using the toxicity characteristic leaching procedure (TCLP) EPA Method 1311<sup>38</sup> (M-1311). The TCLP procedure consists of exposing a quantity of the ash to a prescribed acidic leaching solution for 18 h, and then quantifying the leachate for the presence of a number of toxic compounds, including eight elements (arsenic, barium, cadmium, chromium, lead, mercury, selenium, and silver).

## Results and discussion

Temporal measurements of CO, CO<sub>2</sub>, O<sub>2</sub>, and THC CEM data and e-waste/charcoal bed temperatures were unremarkable. CO and THC emissions from both types of e-waste rose quickly to approximately 1000 ppm and 500 ppm, respectively, as volatile species were liberated, and then fell quickly after approximately 5 min to steady state concentrations of approximately 150 ppm and 125 ppm, respectively. THC emissions then fell steadily over the next 60 to 90 min to values of less than 10 ppm. CO emissions fell very slowly to values between 80 to 100 ppm over 3 h. O<sub>2</sub> and CO<sub>2</sub>

profiles also peaked between 0 and 5 min, but were always greater than 19% and always less than 1%, respectively, indicating adequate simulation of O<sub>2</sub>-available open burning. The bed temperature rose over the course of 10 to 30 min to approximately 700° to 900°C, and these temperatures were maintained for over 3 h. Little distinction was observed between the CEM profiles for the circuit boards and insulated wire tests, likely indicating that the combustion characteristics for the two e-wastes were dominated by the charcoal fuel. Unburned e-waste components (copper wire and metal connectors and brackets) were manually recovered from the residual ash 24 h after the completion of each test. Comparison of the initial and unburned mass from each test indicated that 45% ± 7% and 26% ± 1% of the circuit boards and insulated wire, respectively, were consumed.

## PM and metal emissions

Table 4 presents PM and elemental emission concentrations and emission factors determined from the M-29 filter measurements, sample and facility flow rates, and initial mass of e-waste charged. It should be noted that these emission factors are based on the first 60 or 90 min (see Table 3) of the e-waste burn cycle, and not necessarily over the entire burn cycle. However, based on visual observations and the CEM profiles, described above, emissions during these times likely represent the majority of the e-waste consumed. Thus, the PM and element emissions data presented in Table 4 are likely to be reasonable approximations to the full burn emissions, but if anything, they may under-represent the actual emission factors due to the limiting of the sample collection to a portion of the e-waste burn cycle.

PM emissions from the charcoal blank were only approximately 1% of those from the two e-wastes with charcoal (Table 4), so this contribution was ignored. In addition, the charcoal blank yielded nondetectable levels of most of the

**Table 4.** Particulate matter (PM) emissions and elemental composition of combustion fly ash, determined from M-29 filter measurements

Element	Charcoal blank Sample 011504 Conc (µg/g) <sup>b</sup>	Circuit board Sample 020904		Insulated wire Sample 021004	
		Conc (µg/g) <sup>b</sup>	EF (mg/kg) <sup>a</sup>	Conc (µg/g) <sup>b</sup>	EF (mg/kg) <sup>a</sup>
Sb	ND <sup>c</sup>	4850	75.4	8000	140
As	ND	5120	79.6	ND	–
Br	ND	209850	3264	9815	171
Cl	95800	598	9.30	44950	785
Cu	ND	45600	709	6070	106
Pb	ND	75850	1180	55200	964
K	71350	4395	68.4	1465	25.6
Na	46150	3130	48.7	2455	42.9
S	18900	1690	26.3	173	3.02
Sn	ND	8600	134	4650	81.2
Zn	ND	5185	80.6	5620	98.2
Unidentified (assumed carbon)	587100	575550	8952	843900	14740
PM emission concentration and emission factor	0.30 (mg/m <sup>3</sup> )	23.8 (mg/m <sup>3</sup> )	15600	44.2 (mg/m <sup>3</sup> )	17500

Conc, concentration; EF, emission factor

<sup>a</sup>Emission factor: mass of analyte per mass of initial e-waste

<sup>b</sup>Elemental analysis performed by WD-XRF. Concentration in units of mass of analyte per mass of collected particles

<sup>c</sup>ND, non-detect

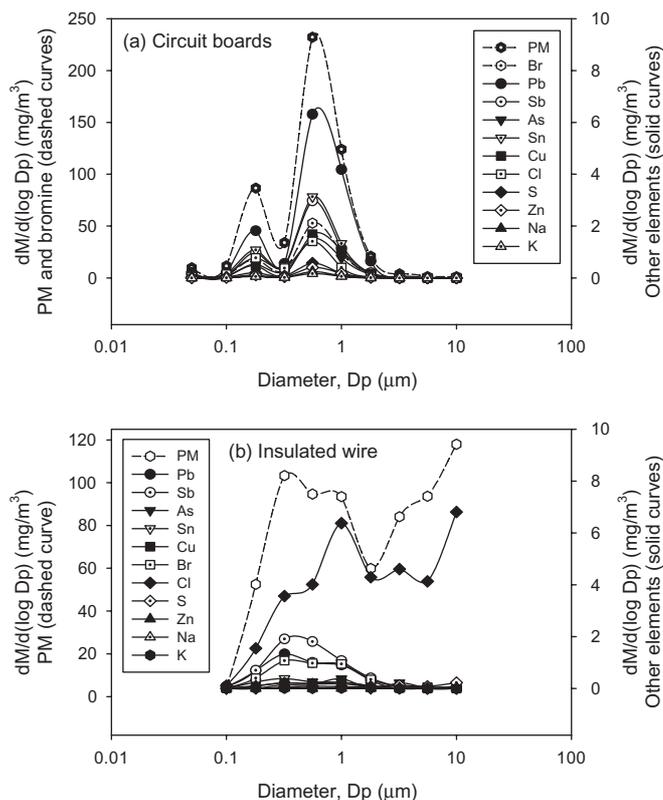
elements of interest, but did indicate measurable emissions of common biomass elements (e.g., sodium, potassium, chlorine, and sulfur). However, the particulate chlorine emissions from the charcoal ( $0.003\text{ mg/m}^3$ ) were a small fraction (0.15%) of those from the insulated wire tests ( $2.0\text{ mg/m}^3$ ).

Calculated PM emission factors for the circuit board and insulated wire were approximately 15 and  $17\text{ g/kg}$  of initial e-waste charged, respectively. WD-XRF analysis of the circuit board fly ash indicated high levels of bromine (21%), lead (8%), copper (5%), and tin (1%) and lesser ( $\sim 0.5\%$ ) contributions of arsenic, zinc, and antimony. Similar analysis of the insulated wire tests indicated high levels of chlorine (4%) and lead (6%), and lesser concentrations ( $<1\%$ ) of bromine, copper, tin, zinc, and antimony. Both fly ash samples included large portions (58% and 84%) of unidentified material (assumed to be carbon). The lead emission concentrations for the circuit boards and insulated wire (Table 4), respectively, were over 350 and 250 times greater than the  $0.20\text{ mg/m}^3$  allowed for new U.S. municipal waste combustors processing more than  $225\text{ Mg/day}$  of waste<sup>39</sup> and 25 to 35 times greater than the  $2.0\text{ mg/m}^3$  allowed for secondary lead smelters.<sup>40</sup> It is interesting that even though copper is a major component of the insulated wire, it is a minor component of the fly ash PM, and this is likely due to the relatively nonvolatile nature of copper at these temperatures (boiling point of  $2567^\circ\text{C}$ ). In contrast, copper emissions from the circuit boards were much higher, and may have been due to differences in the occurrence of copper in these two materials. In circuit boards, copper is used in numerous thin connections between circuit components that offer high surface areas to the combustion environment. In contrast, copper used in wire is braided into strands with a relatively low exposed surface area. This available surface area on circuit boards is likely to promote copper vaporization. It has also been shown that halogens promote the vaporization of a number of otherwise non-volatile metals.<sup>41</sup>

Figure 1 presents the elemental particle size distributions (PSDs) and metal concentrations by size determined from the MOUDI samples. Data for the circuit board test (upper panel) indicate a predominantly submicrometer PSD. Note that PM and bromine mass are presented on the left scale while the other elements are on the right scale. These data are consistent with the M-29 data (Table 4), indicating significant contributions of bromine, lead, copper, tin, antimony, and arsenic. The bimodal behavior may be an artifact of vapor nucleation within the sampling probe.<sup>42</sup> Elemental PSDs for the insulated wire (bottom panel) indicate the presence of both sub- and supermicrometer particle modes, with chlorine contributing to both. However, other elements, including antimony, lead, and bromine, contribute to the submicrometer mode only.

### Residual ash and leaching characteristics

Elemental analyses of the residual ash from the circuit board ( $\sim 70\%$  of the initial mass) and insulated wire ( $\sim 60\%$



**Fig. 1.** Particulate matter (PM) and elemental particle size distributions of fly ash emissions for **a** circuit boards and **b** insulated wire.  $M$ , mass;  $D_p$ , particle diameter

**Table 5.** Elemental composition ( $\mu\text{g/g}$ ) of e-waste residual ash

Element	Circuit board <sup>a</sup>	Insulated wire <sup>a</sup>
Sb	273	883
As	15.9	2.32
Br	2120	366
Cl	210	293 000
Cu	13 900	47 000
Cr	272	47.9
Ge	1.21	<1
Au	23.5	<1
Fe	6 780	730
Pb	3 630	16 900
Mg	2 950	1 250
Mn	45.3	50.1
Ni	279	26.3
Se	4.65	1.03
Sr	200	NM <sup>b</sup>
Sn	2 150	217
Ti	2 260	<1
V	50.1	6.55
Zn	180	764
Zr	46.6	NM

<sup>a</sup>Compositions determined using  $\text{HNO}_3/\text{HF}$  digestion and ICP-MS analysis using U.S. EPA M-6020 (U.S. EPA<sup>23</sup>). Mass of analyte per mass of hand-segregated residual e-waste ash

<sup>b</sup>NM, not measured

of the initial mass, along with unavoidable charcoal ash and sand) are presented in Table 5. These analyses did not include obviously unburned components, connectors, and wire, which were recovered manually. The insulated wire

residual ash contained very high concentrations of chlorine (29%) and notable concentrations of copper (4.7%) and lead (1.7%). The circuit board residual ash was much more varied, containing concentrations of copper (1.4%), iron (0.7%), lead (0.4%), magnesium (0.3%), bromine (0.2%), tin (0.2%), and titanium (0.2%). This is consistent with other e-waste and emissions measurements<sup>19,20</sup> that indicate that circuit boards have a much more varied elemental composition than insulated wire.

Table 6 presents the results of the TCLP analysis of the combined e-waste residual ash and compares these results to federal solid waste standards.<sup>38</sup> The combined e-waste leachate failed the EPA Maximum Contaminant Level Standard for soluble lead. This ash, therefore, would be characterized as hazardous waste under federal regulations.

#### PXDD/PXDF measurements

The calculated PCDD/PCDF emission factors for the two circuit board TO-9 tests were 28 and 155 ng TEQ/kg (see Table 7). These two values fall within the range of values reported for tests of uncontrolled barrel burning of residential waste.<sup>15</sup> The high relative standard deviation (RSD = 98%) between the two samples is not unexpected for these analytes in combustion tests involving relatively small sample sizes. The emissions for the insulated wires were 12400, 18100, and 5400 ng TEQ/kg (RSD = 50%). These exceptionally high emissions were likely exacerbated by the high concentration of chlorine-containing PVC insulation

**Table 6.** Comparison of element concentrations (mg/l) measured in e-waste residual ash toxicity characteristic leach profile (TCLP) leachate with the Federal standards

Element	TCLP leachate concentration <sup>a</sup>	EPA maximum contaminant level
As	<0.005	5.0
Ba	4.17	100.0
Cd	0.683	1.0
Cr	<0.01	5.0
Pb	14.3	5.0
Hg	<0.0005	0.2
Se	0.035	1.0
Ag	0.047	5.0

<sup>a</sup>TCLP leachate measured using U.S. EPA M-1311 (U.S. EPA<sup>37</sup>) on a composite sample of residual ash from one circuit board (011304) and one insulated wire (011404) test

on the wires, as well as by other factors related to incomplete combustion. These values are much higher than those reported for similar or related sources (e.g., Bremmer et al.<sup>13</sup>) and may be the highest reported for any source. The United Nations tool kit<sup>43</sup> uses a value of 5000 ng TEQ/kg for open burning of cable. These high emissions suggest that open burning of insulated wire, a practice typically discontinued in most countries, may have had a significant impact on historical source PCDD/PCDF inventories, but this point requires further study. The average PCDD:PCDF ratios for the circuit boards and wires were 0.24:1 and 0.36:1, respectively, consistent with most combustion samples that typically indicate PCDF concentrations greater than those of PCDDs.

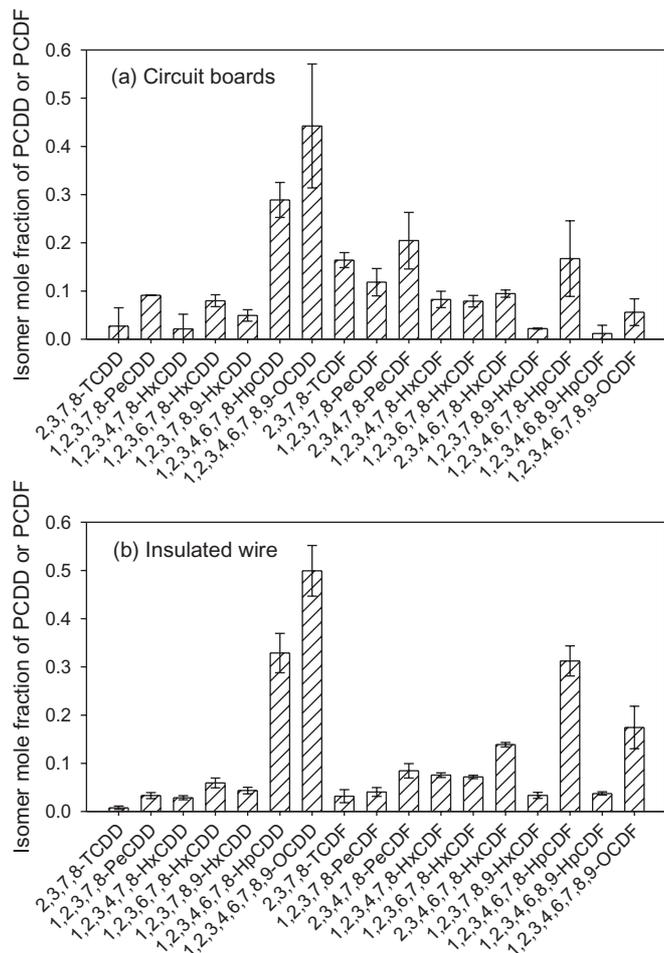
A prominent distinction between the isomer patterns of the circuit boards (Fig. 2a) and insulated wire (Fig. 2b) is the relatively higher molar fraction of the 1,2,3,7,8-Penta CDD (PeCDD), the 2,3,7,8-Tetra CDF (TeCDF), and the two PeCDF isomers in the circuit boards. The dominant 1,2,3,4,6,7,8-Hepta CDD (HpCDD) and Octa CDD (OCDD) dibenzodioxin isomers and the dominant 1,2,3,4,6,7,8-HpCDF isomer in both the circuit boards and the insulated wire are consistent with fly ash sampled at a copper and aluminum metal recovery facility,<sup>11</sup> although their observed PCDD:PCDF ratio was less than 0.1:1. In general, the circuit boards showed fairly distinctive isomers patterns compared to myriad sources published in Cleverly et al.<sup>44</sup> The insulated wire, however, had patterns quite similar to those for medical and hazardous waste incineration sources. The homologue profiles for the circuit boards, presented in Fig. 3a, are similarly distinctive from those of the insulated wire, presented in Fig. 3b, showing a greater molar fraction in the lower-chlorinated homologues, particularly the TeCDD and TeCDF homologues, than in the higher-chlorinated homologues.

The circuit board burn emissions were analyzed for PBDD/PBDF due to the high concentration of brominated flame retardants in circuit boards and the expectation of chemical conversion to this form.<sup>5,45</sup> Selective ion monitoring confirmed the presence of mono- to penta-BDD/F (MW < 550 amu). The observed BDFs were generally several times higher in concentration than the BDDs. Circuit board emissions of comparable homologues for the brominated and chlorinated targets (TeCDD and TeBDD, PeCDD and PeBDD, PeCDF and PeBDF) from Figs. 4 and 3, respectively, show that the brominated homologues were about 50 to 500 times more abundant than their chlorinated counterparts, likely due to the high Br/Cl mass ratio (>300) in

**Table 7.** Polychlorinated dibenzodioxin and dibenzofuran (PCDD/F) emission factors

Sample	Circuit boards		Insulated wire		
	011304	022004	011404	022404	020805
Total PCDD (ng/kg)	320	5200	210000	310000	38000
Total PCDF (ng/kg)	2600	14000	480000	710000	200000
PCDD/F (ng TEQ/kg)	28	155	12400	18100	5400
Average PCDD/F (ng TEQ/kg), RSD (%)	92 (98)		11900 (50)		

TEQ, toxic equivalency; RSD, relative standard deviation

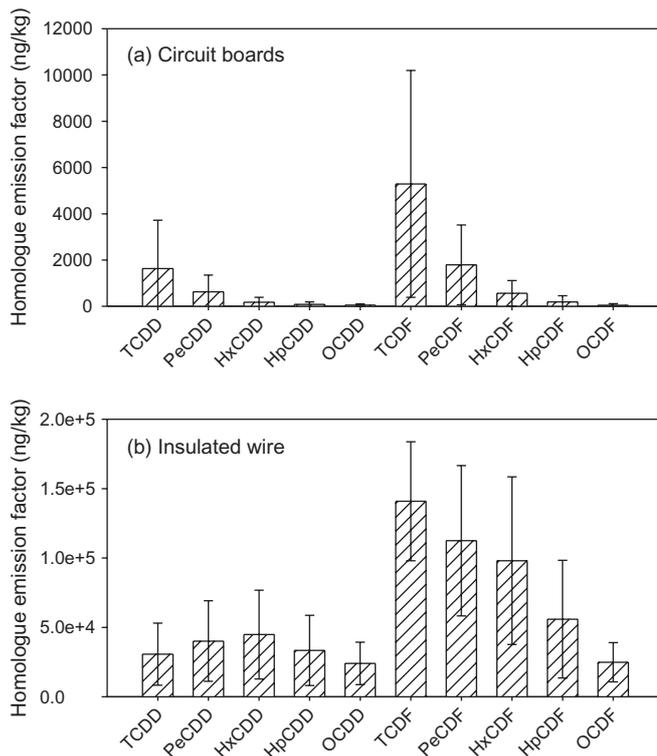


**Fig. 2.** 2,3,7,8-Cl-substituted congener patterns (molar fractions) for **a** circuit boards and **b** insulated wire. PCDD, dibenzodioxins; PCDF, dibenzofurans; T, tetra; Pe, penta; Hx, hexa; Hp, hepta; O, octa

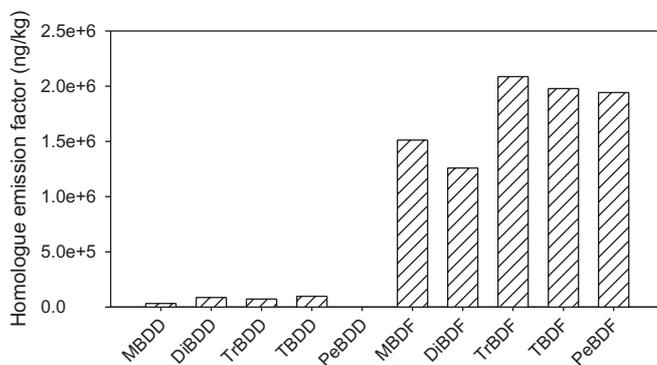
circuit boards. While our efforts focused on PXDD/PXDF compounds, others have also noted the presence of bromobenzenes and mono- to tribromomethanes.<sup>20</sup> We also did not analyze for mixed brominated/chlorinated compounds, which have been found by others<sup>45</sup> to be more prevalent than the unmixed compounds. The insulated wire sample in this work had very low levels of BDDs/BDFs compared to the computer boards, consistent with the minor concentration of Br in the insulated wire (Table 1).

## Conclusions

Combustion of two kinds of e-waste was performed in an open burn simulation facility to provide a preliminary characterization of emissions, including PCDDs/PCDFs, PBDDs/PBDFs, fly ash PM and metals, and residual ash, from rudimentary recycling operations. PM emission factors from the circuit board and insulated wire e-wastes were approximately 15 and 17 g/kg of initial e-waste charged, respectively. In the case of circuit boards, these PM emissions were largely submicrometer in size with notable con-



**Fig. 3.** PCDD/F homologue emission factors for **a** circuit boards and **b** insulated wire



**Fig. 4.** Polybrominated dibenzodioxin and dibenzofuran (PBDD/PBDF) homologue emission factors for circuit boards. M, mono; Di, di; Tr, tri

centrations of bromine, lead, tin, copper, antimony, and arsenic. Lead emission concentrations exceeded U.S. municipal waste combustor limits by over 200 times. For insulated wire, the PM emissions had both submicrometer and supermicrometer components and were composed of chlorine and lead, with smaller amounts of antimony and bromine. Fly ash emissions for both e-wastes contained significant fractions of unidentified mass and this was assumed to be unburned carbon moieties. An exceptionally high PCDD/PCDF emission factor from open burning of insulated wire (~12000 ng TEQ/kg) was likely due to the uncontrolled nature of the fire as well as to the high chlorine content. The PCDD/PCDF emission factor from the circuit

boards (~100 ng TEQ/kg) was also relatively high compared to other sources, such as residential waste<sup>14,15,46</sup> and biomass.<sup>21,22,46,47</sup> Very high PBDD/PBDF emissions from the circuit board confirmed the anticipated conversion of brominated flame retardants to these chemical forms and nominated combustion of flame-retardant-containing waste as a potential source of environmental PBDD/PBDF contamination. Additional organic emissions, such as polycyclic aromatic hydrocarbons (PAHs) and phthalates, were not analyzed. These results suggest that significant health and environmental hazards could result from rudimentary recycling operations that include the open burning of e-wastes. The growing e-waste disposal problem could exacerbate these practices; preventive measures, such as export controls, additional domestic recycling and disposal options, and regulatory and enforcement scrutiny, may ameliorate these environmental impacts.

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