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Toxic footprint and materials profile of electronic components in printed circuit boards

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ABSTRACT

Waste printed circuit boards (WPCBs) contain valuable material resources and hazardous substances, thereby posing a challenge for sustainable resource recovery and environmental protection initiatives. Overcoming this challenge will require mapping the toxic footprint of WPCBs to specific materials and substances used in manufacturing electronic components (ECs). Therefore, this work collected 50 EC specimens from WPCBs in five ubiquitous consumer products, such as television, refrigerator, air conditioner, washing machine and computer. The work extracted and analyzed metal contents and used leachability assessments based on tests adopted by the regulatory policies from China and the United States. The work found that copper and iron are the most abundant constituents in ECs, with concentrations ranging 5.90-796.62 g/kg and 0-831.53 g/kg, respectively; whereas abundance of precious metal content is in the order of silver > gold > palladium > platinum, with silver concentration ranging 15–5290 mg/kg. The content of marginally-regulated toxic substance arsenic ranged 0–9700 mg/kg; whereas fully regulated toxic metals such as chromium, lead and mercury did not exceed the thresholds set by China and US standards. The work found new toxic threats from arsenic and selenium leached from 20 of 50 ECs exceeding regulatory standards. These results will aid manufacturers and recyclers in protecting workers' health and environmental quality from arsenic and selenium pollution, and should initiate discussion about regulating these toxic components as part of a comprehensive program to reduce the toxic footprint of electronic products.

1. Introduction

The active use period of electrical and electronic equipment (EEE) is increasingly brief because of the rapid pace of technical innovation, which in turn leads to rapid accumulation of waste electric and electronic equipment (WEEE, or E-waste) (Calgaro et al., 2015; Mesquita et al., 2018). According to *The global E-waste monitor 2020*, the total amount of WEEE was 53.6 million tons in 2019 (Forti et al., 2020), of which China accounted for 7.5 million tons, more than any other

country in the world. With an average annual growth of 4%, global WEEE is predicted to exceed 74 million tons by 2030. Numerous studies have confirmed that WEEE contains several toxic substances (Evangel-opoulos et al., 2019; Suresh et al., 2018; Tsai, 2020) and valuable metals (Marra et al., 2018; Zeng et al., 2016). Thus, they are not only a waste but also an important urban resource, but pose a risk to human health and the environment if not properly managed through a sustainable circular economy (Awasthi et al., 2019).

Waste Printed Circuit Boards (WPCBs) account for - 4% of WEEE

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Table 1 ECs classification.

	Category		Sub-category
Electronicelements (EEs)	Resistors	6	MFRs $\ SMD-Rs \ CRs \ TRs \ DRs \ CFRs$
	Capacitors	8	$ETCs \setminus FCs \setminus PCs \setminus SCs \setminus CCs \setminus MLCCs \setminus HVCCs \setminus CECs$
	Inductors	6	$CIs \setminus FIs \setminus HPIs \setminus SMD-PIs \setminus AIs \setminus TIs$
	Crystal oscillators (COs)	2	$CCOs \setminus PCOs$
	Electric filters (EFs)	1	-
	Switches	2	$PSs \setminus TSs$
	Linkers	2	connectors \ ports
	Relays	1	-
	Transformers	1	-
Electronicdevices (EDs)	Discrete devices (DDs)	9	$ZDs \setminus SMD\text{-}Ds \setminus RDs \setminus STs \setminus SMD\text{-}Ts \setminus ILTs \setminus PTs \setminus FETs \setminus thyristors$
	Photoelectric Semiconductors (PhSs)	3	$OCs \setminus LEDs \setminus digitrons$
	Logic ICs	2	CPUs; chips
	Analog ICs	1	-
	Buzzers	1	-
	Fuses	1	-
	Rectifier bridges (RBs)	1	-
	Heat sinks (HSs)	1	-
	Tuners	1	-
	Line output transformers (LOTs)	1	-

Note: All abbreviations and pictures of ECs are listed in SI Tables S8 and S9.

and represent its most significant potential economic value, subject to advances in resource recovery technology development and regulatory policies for pollution prevention (Chen et al., 2016; Kousaiti et al., 2020; Wang et al., 2016; Zhu et al., 2012). WPCBs typically contain various types of Printed Wire Boards (PWBs) and Electronic Components (ECs), and the two categories vary widely in materials composition and concentration (Hao et al., 2020). Best available technology for resource recovery includes removal of mounted ECs from WPCBs (Wang and Xu, 2015). Researchers have focused on strategies such as machinery (Huang et al., 2021a, 2021b), hydrometallurgy, pyrometallurgy and biotechnology to recover valuable metals from PWBs (30-40%), such as copper (-16%), aluminum (-5%), tin (-4%), iron (-3%), nickel (-2%) and zinc (-1%), and also precious metals, such as gold (-0.03%), silver (-0.05%) (Chen et al., 2013; Holzer et al., 2021; Karal et al., 2021; Lahtela et al., 2019; Park and Fray, 2009; Wang and Xu, 2015; Yin et al., 2018).

The research is sparse on the efficient recovery of valuable metals and precious metals from specific types of ECs. Wang et al. reported an integrated process to recover aluminum and iron from electrolytic capacitors with a efficiency of 96.52% and 98.68%, respectively (Wang and Xu, 2017). Slurry electrolysis was applied to recover copper and gold from CPU sockets (Li et al., 2019b). Ag, Au, Pd and Pt are recovered from waste integrated circuits (ICs) with a process that combined physical beneficiation and hydrometallurgy (Panda et al., 2021). There are also few studies on the toxicity of ECs constituents. Lim Seong-Rin et al. found that only the low-intensity red LED have excessive Pb under the U.S. regulations, and Pb was not detected in other color and intensity LEDs (Lim et al., 2011). Indeed, different metals are distributed in kinds of ECs, showing diverse resource and toxicity properties (given in supporting information, SI Table S1). However, these results address specific ECs, without a comprehensive understanding of generalizable EC characteristics.

Therefore, the authors selected 50 representative ECs, analyzed their metallic contents and assessed their leaching toxicity based on standardized test leaching procedures according to Chinese and US regulatory. The results should guide future work on resources recovery from ECs as well as contributing to minimize environmental pollution.

2. Materials and methods

2.1. Specimen collection and preparation

Currently, there are 109 authorized WEEE recycling factories

distributed in China, and they are authorized to collect 5 categories of WEEE for government subsidy: TVs, refrigerators, air conditioners, washing machines and computers. To ensure representativeness of specimen collection, 5 of the 109 factories were randomly selected. Then, WPCBs were selected from the five categories of the WEEE on each of the five recycling factories. All the ECs mounted on each of the WPCBs were dissembled in the laboratory. Details for this process and the number of the obtained ECs from each WPCBs are given in SI, Tables S2–S7.

According to the production process, ECs that have not been modified in material composition and structure are recognized as electronic elements (EEs); otherwise, they are called as electronic devices (EDs). Then, according to their functions in the electronic circuit, EEs are then further classified into 9 sub-categories: capacitors, inductors, resistors, crystal oscillators, filters, switches, relays, transformers and linkers. Electronic devices were separated to 10 sub-categories: discrete devices (DDs), photoelectric semiconductors (PhSs), logic integrated circuits (ICs), analog integrated circuits (ICs), buzzers, fuses, rectifier bridges, heat sinks, tuners and line-output transformers (LOTs). Among these subcategories, based on differences in materials, some were secondarily classified, for example, capacitors were classified as electrolytic capacitors (ETCs), film capacitors (FCs), polyester capacitors (PCs), safety capacitors (SCs), ceramic capacitors (CCs), multi-layer ceramic capacitors (MLCCs), high voltage ceramic capacitors (HVCCs) and chip electrolytic capacitors (CECs). Inductors were classified as choke inductors (CIs), filter inductors (FIs), high power inductors (HPIs), SMD power inductors (SMD-PIs), axial inductors (AIs) and toroidal inductors. The final tally resulted in 29 different electronic elements and 21 electronic devices (Table 1).

To achieve comprehensive analysis, specimens were selected from each of the categories and sub-categories, representing a total of 50 specimens. Each specimen was crushed and ground separately into 9.5 mm undersize material for Toxicity Characteristic Leaching Procedure (TCLP) by a cutting mill (SM-2000, Retsch, Germany).

2.2. Metal content and leaching toxicity

1 g of the 50 powdered specimens were taken for digested by HNO₃ - HClO₄ - HF system (Güngör and Elik, 2007) and then analyzed by an Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES, Perkin Elmer, Optima 8300, USA) for metal content analysis. In this work, 24 metals were analyzed and they were categorized as ordinary metals: Co, Cd, Cr, Cu, Fe, Ni, Pb, Sb, Sn, Tl, V, Zn, Hg, Al, Ba, Be, Mg;

	ng/kg					g/kg										
	Hg Cr	Be	Λ	Co	Cd	П	Mg	Sb	Ba	Pb	Sn	Ni	AI	Zn	Cu	Fe
Resistors	VD 585	1 22	70	1280	113	1.86	2.03	1.61	09.60	0.97	24.55	10.25	1.80	103.14	185.12	305.11
	, O)	~ 0) ~	$\sim 0)$	$(50 \sim$	$\sim 0)$	~ 0)	$\sim 0)$	$\sim 0)$	$(0.05 \sim$	$\sim 0)$	$(3.05 \sim$	$(0.34 \sim$	$(0.19 \sim$	$\sim 0)$	$(6.78 \sim$	$(0.42 \sim$
	326	(0) 130)	130)	7180)	290)	4.48)	10.13)	7.88)	46.23)	3.57)	102.41)	53.95)	7.49)	595.82)	384.14)	502.70)
Capacitors	UN DN	818	273	720	2508	4.39	10.43	35.13	60.20	4.55	18.98	33.63	153.88	9.20	193.85	92.42
	1	$\sim 0)$	\sim (30 \sim	$\sim 0)$	\sim 0)	~ 0)	$\sim 0)$	$\sim 0)$	$(0.03 \sim$	$\sim 0)$	$\sim 0)$	$(0.46 \sim$	$(1.16 \sim$	$\sim 0)$	$(13.28 \sim$	$(0.84 \sim$
		6370)	1750)	5000)	17760)	8.46)	60.77)	268.08)	294.96)	20.44)	75.15)	241.97)	734.28)	58.77)	811.18)	311.49)
Inductors	UN UN	35	143	152	222	4.12	2.28	2.72	0.30	2.95	8.54	9.29	2.26	24.23	494.60	190.39
	1	\sim 0)	$\sim 09)$	$\sim 0)$	\sim 0)	$(0.3 \sim$	$(0.16 \sim$	$(0.40 \sim$	$(0.03 \sim$	$(0.13 \sim$	\sim 0)	$(0.19 \sim$	$(0.42 \sim$	$(1.08 \sim$	$(242.0 \sim$	$(46.52 \sim$
		210)	350)	430)	530)	9.29)	6.47)	5.68)	0.57)	8.53)	31.20)	37.64)	6.84)	86.07)	803.42)	340.42)
COS	UN UN	85	70	2060	670	QN	1.49	2.14	0.03	59.19	29.47	55.46	0.82	27.96	482.86	83.82
EFs	UN UN	ND	120	110	ND	QN	ŊŊ	0.47	0.02	ND	DN	18.05	0.63	1.23	19.27	831.53
Switches	UN UN	ND	85	50	330	5.55	18.31	3.33	0.26	ND	7.02	0.88	24.92	19.85	57.17	163.60
Linkers	UN UN	ND	40	20	460	1.55	1.0	3.97	0.17	5.55	19.04	1.89	2.08	105.18	317.54	0.51
Relays	UN UN	ND	80	20	ND	2.35	0.61	4.70	0.16	0.24	ND	8.40	9.88	0.12	193.31	695.83
Transformers	UN UN	ND	110	50	260	6.89	1.59	0.14	0.16	ND	0.38	0.18	0.66	19.73	78.49	154.52
Average	- 65	107	110	496	507	2.97	4.19	6.02	7.88	8.16	12.0	15.33	21.88	34.52	224.69	337.63

Table 3 Ordinary r

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Oruniary III	clai cui	n siliai	I ELECTION	ne vices.													
	mg/kg					g/kg											
	Hg	Ŀ	Be	Λ	Co	Cd	Ba	Mg	П	Pb	Sb	Ni	Zn	Sn	AI	Fe	Cu
DDs	ND	QN	16	63	128	0.26	0.07	0.60	1.67	6.95	4.08	15.63	4.75	12.34	1.32	51.61	663.35
	I	I	$\sim 0)$	$\sim (30 \sim$	$(20 \sim$	\sim 0)	\sim 0)	$(0\sim2.46)$	$(0 \sim 7.75)$	$(0.05 \sim$	$\sim (0.67 \sim$	$(0.19 \sim$	$(0\sim 33.01)$	$(0\sim 88.57)$	$(0.01 \sim$	$\sim 0)$	$(306.20 \sim$
			130)	130)	830)	0.66)	0.39)			12.79)	12.65)	104.70)			5.12)	319.51)	937.06)
PhSs	ND	Ŋ	523	367	250	4.09	1.24	6.11	8.22	ND	32.14	15.07	73.82	43.38	21.36	8.19	533.19
	I	I	$\sim 0)$	$(200 \sim$	$(100 \sim$	$(0.77 \sim$	$\sim 90.0)$	$(0.22 \sim$	$(1.70 \sim$	I	$(9.62 \sim$	$(0.61 \sim$	$(1.10 \sim$	$(4.38 \sim$	$\sim 0)$	$\sim 0)$	$(488.80\sim$
			1170)	450)	550)	6.45)	3.35)	16.25)	12.95)		55.80)	37.60)	219.00)	112.05)	43.35)	12.30)	555.76)
Logic ICs	ND	Ŋ	55	55	120	0.23	15.38	0.69	3.20	2.48	4.50	8.08	0.57	57.76	8.90	1.96	608.06
Analog	ND	Ŋ	QN	60	20	0.45	0.01	0.27	6.42	1.08	1.84	0.19	ND	7.41	ND	ND	784.04
ICs																	
Buzzers	ND	Ŋ	QN	60	60	0.51	0.28	4.33	ND	45.20	0.30	1.55	74.95	1.65	8.03	2.64	276.60
Fuses	ND	Ŋ	ND	30	60	0.21	ND	DN	6.69	126.42	4.66	9.32	89.40	23.41	ND	Ŋ	761.98
RBs	ND	Ŋ	ND	50	30	0.30	0.01	DN	2.05	7.02	6.36	0.23	ND	ND	ND	Ŋ	796.62
HSs	ND	Ŋ	QN	1450	100	2.90	ND	105.25	35.30	DN	ND	3.45	0.05	2.45	625.0	5.05	5.90
Tuners	ND	Ŋ	ND	1050	8750	0.55	4.40	QN	19.85	3.70	9.45	16.60	7.20	139.95	67.30	525.0	203.50
LOTs	ND	Ŋ	ND	1050	800	3.40	3.75	20.80	ND	ND	36.25	69.15	67.05	7.20	62.40	500.50	254.0
Average	ND	Ŋ	119	232	430	1.27	3.62	5.88	6.03	8.18	10.41	11.14	23.78	30.00	31.74	46.79	594.35
Note: ND: n	ot detec	cted; th	ie figures	in brackets	indicate th	e range of n	netal content	t in similar EC	S.								



Fig. 1. The content of precious metals in electronic components (both EEs and EDs).



Fig. 2. The content of metalloids in electronic components (both EEs and EDs).

precious metals: Au, Ag, Pt, Pd; and metalloids: As, B, Se.

Leaching toxicity of the 50 ECs were further investigated according to regulatory standards for China and U.S. procedures using sulfuric acid & nitric acid method (HJ/T299-2007, China) and Toxicity Characteristic Leaching Procedure (TCLP, Method 1311; 40 CFR §261.24). The leachable concentrations of Cu, Zn, Cd, Pb, Cr, Hg, Be, Ba, Ni, Ag, As and Se were measured according to HJ/T299-2007 and that of As, Ba, Cd, Cr, Pb, Se, Ag and Hg were tested based on TCLP by ICP-OES. All the experiments were taken for 3 replicates, and data adopted were average.

3. Results and discussion

3.1. Ordinary metals of EEs

Table 2 presents the results of ordinary metals contents of 9 subcategory electronic elements (EEs), and the detailed ordinary metals contents of 29 EEs are presented in SI Table S10. The concentrations of ordinary metals detected in these EEs are iron (337.63 g/kg), copper (224.69 g/kg), zinc (34.52 g/kg), aluminum (21.88 g/kg), nickel (15.33 g/kg), tin (12.0 g/kg), lead (8.16 g/kg), barium (7.88 g/kg), antimony

(6.02 g/kg), magnesium (4.19 g/kg), thallium (2.97 g/kg). Other metals detected at very low levels are cadmium (507 mg/kg), cobalt (496 mg/ kg), vanadium (110 mg/kg) beryllium (107 mg/kg), chromium (65 mg/ kg). Mercury was not detected in any of the analyzed EEs. The most abundant metal in the 9 EEs is Fe (ranging 0.51-831.53 g/kg), usually used for heat dissipation. EFs contains the highest Fe content (831.53 g/ kg) because of its steel shell and yttrium iron garnet (Kosai et al., 2020); followed by relays (695.83 g/kg) because of the internal frame (including core, yoke, and armature, SI Fig. S1). Linkers contain the lowest Fe (0.51 g/kg), a small amount existed in the pin (SI Fig. S2). Cu is also abundant in the EEs (6.78 g/kg - 811.18 g/kg), which is widely used in printed circuit boards and ECs because of its excellent electrical conductivity. The highest concentration of copper is found in the CECs (811.18 g/kg); followed by CIs (803.42 g/kg) because of the copper wires. COs also contain a high concentration of copper (482.86 g/kg in average), especially PCOs (638.61 g/kg), because the shell is made of brass, SI Fig. S3. The least copper of EEs is EFs (19.27 g/kg). Copper and iron account for about 83% of the metallic content of EEs.

For Zn, Al, Ni and Sn, their contents are one order of magnitude lower than copper and iron. Zn, ranging 0–595.82 g/kg, is mainly from the galvanized steel (Woo et al., 2016). Linkers contain a high concentration of zinc (105.18 g/kg), mainly from its pins, SI Fig. S2. Among linkers zinc in ports is the higher (210.08 g/kg, SI Table S10, SI Fig. S4). Resistors ranked the second for Zn, and DRs contain the highest zinc (595.82 g/kg, SI Table S10) among resistors because of its ZnO varistor (Niu et al., 2021). Al is also a common metal in EEs (0-734.28 g/kg). Al presents in large quantities in capacitors (1.05-734.28 g/kg), especially in ETCs (734.28 g/kg) because the shell and foil of the ETCs are made of aluminum (SI Fig. S5) (Wang and Xu, 2017). The contents of Ni in the 9 EEs range from 0.16 to 241.97 g/kg, widely applied in alloy due to the excellent thermal stability and resistance. MLCCs and PCOs contain high concentrations of Ni (241.97 g/kg and 110.57 g/kg, respectively). Ni of MLCCs is mainly from the electrode (Albertsen, 2004), and Ni of PCOs mainly exists in the plating (SI Fig. S3). The concentration of Sn is in the range of 0-102.41 g/kg. SMD-R contains the highest Sn because of its outer termination (Safonov and Choba, 2022).

As for Pb (0–59.19 g/kg), Ba (0.02–60.20 g/kg), Sb (0.14–35.13 g/kg), Mg (0–18.31 g/kg) and Tl (0–6.89 g/kg), their contents are at least two orders of magnitude lower than copper and iron. Cd (0–2508 mg/kg), Co (20–2060 mg/kg), Be (0–818 mg/kg) and V (40–273 mg/kg) are lower than other metals contents presented above. Cr was only detected in one kind of EEs, DRs (3260 mg/kg).

3.2. Ordinary metals of EDs

Table 3 presents the ordinary metals contents of 10 sub-category electronic devices (EDs), and the detailed ordinary metals contents of 21 EDs is shown in SI Table S11. On average, Cu is the most abundant metal in the 10 sub-categories, 594.35 g/kg. Contents of Fe (46.79 g/ kg), Al (31.74 g/kg), Sn (30.0 g/kg), Zn (23.78 g/kg), Ni (11.14 g/kg) and Sb (10.41 g/kg) are one order of magnitude lower than Cu. Pb (8.18 g/kg), Tl (6.03 g/kg), Mg (5.88 g/kg), Ba (3.62 g/kg) and Cd (1.27 g/kg) are two orders of magnitude lower than Cu. Other metals including Co (430 mg/kg), V (232 mg/kg) and Be (119 mg/kg) are very low. Hg and Cr were not detected in any analyzed EDs. Cu concentrations in the 10 EDs is ranging 5.90 g/kg to 796.62 g/kg, accounting for approximately 77% of the total metal content. Cu is generally found in high concentrations in EDs (except HSs) due to its superior performance as an electrical conductor (Fu et al., 2020). HSs contain a small amount of copper relative to other EDs, 5.9 g/kg, mainly from a small amount of copper doped in the aluminum alloy (Akopyan et al., 2021). For Fe (0-525 g/kg), tuners and LOTs contain high concentrations, 525 g/kg and 500.50 g/kg, respectively. The iron contents of other EDs are at least one order of magnitude lower than tuners and LOTs. For tuners, iron is mainly from the shell and internal printed circuit board, SI Fig. S6. Tuners also contain a high concentration of Sn (139.95 g/kg, SI

Table 4

Chemical composition of leachates from HJ/T299-2007 leaching test for EEs, mg/L.

	Threshold	Capacit	ors							Inductor	rs					
		ECs	FCs	PCs	SCs	CCs	MLCCs	HVCCs	CECs	CIs	FIs	HPIs		SMD-PIs	AIs	TIs
Cu	100	0.12	0.13	0.06	0.11	0.28	0.10	0.10	0.26	0.07	0.31	7.32		0.24	0.24	0.04
Zn	100	0.89	1.33	ND	3.12	0.48	0.26	ND	0.45	0.98	0.23	0.14		0.16	0.60	ND
Cd	1	0.02	0.04	0.08	0.05	0.09	ND	0.04	0.06	0.08	0.09	0.03		0.08	ND	0.04
Pb	5	0.78	ND	0.15	0.09	0.17	0.51	ND	0.66	1.20	ND	0.14		0.02	0.05	ND
Cr	5	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND		ND	ND	ND
Hg	0.1	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND		ND	ND	ND
Ве	0.02	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND		ND	ND	ND
Ba	100	0.04	0.03	0.08	0.01	0.06	0.07	0.01	0.28	0.01	0.04	0.02		0.02	0.02	0.10
Ni	5	0.06	0.07	0.10	0.06	0.08	0.08	0.04	0.45	0.11	0.05	0.37		0.23	0.05	0.01
Ag	5	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND		ND	ND	ND
As	5	ND	ND	ND	0.63	ND	0.50	ND	ND	ND	ND	ND		ND	ND	0.60
Se	1	ND	0.04	ND	ND	0.01	0.02	ND	ND	0.12	ND	ND		ND	ND	ND
	Threshold	Resistor	s					Crystal o	scillators	Filters	Switch	es	Relays	Transformers	Connecto	rs
		MFRs	SMD-Rs	CRs	TRs	DRs	CFRs	CCOs	PCOs		PSs	TSs			Linkers	Ports
Cu	100	0.33	0.23	0.25	0.92	0.17	0.42	0.16	0.46	0.70	0.17	0.30	0.60	0.32	0.06	6.60
Zn	100	0.23	1.34	14.17	ND	0.11	0.51	0.16	0.37	0.18	7.42	1.79	0.31	0.07	1.32	18.19
Cd	1	ND	ND	0.11	ND	0.03	0.08	0.58	ND	ND	0.04	0.06	ND	0.05	0.09	0.08
Pb	5	ND	0.43	ND	4.51	2.35	2.28	0.04	0.71	ND	ND	ND	0.06	ND	0.08	ND
Cr	5	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Hg	0.1	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Be	0.02	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Ba	100	0.020	0.07	0.15	0.020	0.01	0.03	ND	0.02	0.01	0.02	0.03	0.09	0.02	0.05	0.13
Ni	5	0.04	2.81	0.10	0.120	0.05	0.08	0.02	5.31	17.54	0.41	0.08	3.77	0.13	0.20	0.09
Ag	5	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
As	5	ND	ND	0.07	ND	ND	ND	ND	0.50	ND	0.01	ND	0.26	ND	ND	ND
Se	1	ND	0.23	0.25	0.92	0.17	0.420	0.16	0.50	0.70	0.17	0.30	0.56	0.32	0.06	0.60

Note: ND: not detected; concentrations in bold are above the regulatory limit.

Table 5

Chemical composition of leachates from TCLP leaching test for EEs, mg/L.

	Threshold	Capacit	ors								Induct	ors				
		ECs	FCs	PCs	SCs	CCs	MLCCs	HVCCs	CECs		CIs	FIs	HPIs	SMD-PIs	AIs	TIs
As	5	ND	ND	ND	ND	2.79	ND	2.27	ND		ND	ND	ND	ND	ND	ND
Ba	100	ND	ND	0.05	ND	ND	0.98	ND	0.33		ND	ND	ND	ND	ND	ND
Cd	1	0.03	0.05	0.46	0.18	0.08	ND	0.14	0.12		0.13	0.04	0.07	0.02	ND	0.15
Cr	5	ND	ND	ND	ND	ND	ND	ND	ND		ND	ND	ND	ND	ND	ND
Pb	5	0.32	ND	0.21	0.17	0.37	ND	ND	0.02		ND	0.29	ND	ND	ND	ND
Se	1	0.56	4.13	0.72	0.22	17.26	ND	0.45	0.89		1.87	0.40	4.65	16.63	ND	ND
Ag	5	ND	ND	ND	ND	0.55	0.57	ND	ND		ND	ND	ND	0.57	ND	ND
Hg	0.2	ND	ND	ND	ND	ND	ND	ND	ND		ND	ND	ND	ND	ND	ND
	Threshold	Resistor	rs					Crystal o	scillators	Filters	Switch	es	Relays	Transformers	Connecto	rs
		MFRs	SMD-Rs	CRs	TRs	DRs	CFRs	CCOs	PCOs		PSs	TSs			Linkers	Ports
As	5	ND	1.67	ND	ND	5.68	4.69	2.88	ND	ND	3.16	ND	0.05	ND	ND	ND
Ba	100	ND	ND	ND	ND	ND	0.21	0.01	ND	ND	ND	ND	ND	ND	ND	ND
Cd	1	ND	ND	ND	ND	0.11	0.22	0.37	ND	ND	ND	0.26	ND	ND	0.06	0.10
Cr	5	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Pb	5	ND	2.99	ND	ND	ND	0.39	0.21	ND	ND	ND	ND	ND	ND	ND	ND
Se	1	ND	ND	ND	0.81	4.39	ND	0.77	1.80	ND	ND	6.02	0.54	ND	9.62	2.54
٨a																
лg	5	0.56	0.58	ND	ND	0.56	ND	ND	1.12	ND	ND	1.14	ND	ND	1.10	ND

Note: ND: not detected; concentrations in bold are above the regulatory limit.

Table S11), which is from solder (Qiao et al., 2021). Sn (0–139.95 g/kg) is usually used as solder to install ECs on WPCBs. HSs contain the highest Al (625.0 g/kg), Mg (105.25 g/kg), and Tl (35.30 g/kg), mainly because the raw material is aluminum alloy, SI Fig. S7. Zn and Sb have the highest contents in LEDs, 219.0 g/kg and 55.80 g/kg, respectively. For fuses, Pb (126.42 g/kg) present the highest concentration among all the investigated metals, since it contains leaded glass (Besisa et al., 2021).

3.3. Precious metals

different categories of ECs (including EEs and EDs, a total of 19 groups). The detailed concentrations of precious metals in each ECs are shown in SI Tables S12 and S13. On average, the contents of precious metals in ECs ranged from the highest level of silver (992 mg/kg) > gold (303 mg/kg) > palladium (88 mg/kg) > platinum (3 mg/kg). Due to their excellent electrical conductivity, high corrosion resistance, the demand for precious metals in the manufacture of electronic products is increasing (Panda et al., 2021). Ag was detected in 11 groups of ECs, ranging 15–5290 mg/kg, which is much higher than the other three precious metals, because of its high conduction (Li et al., 2019a). Au was detected in all the ECs groups, with a large difference in content ranging

Fig. 1 shows the average concentrations of Pt, Pd, Au, and Ag in

Table 6
Chemical composition of leachates for EDs, mg/L.

Threshold Discrete device (DDs) Photoelectric semiconductor (PhSs) Logic ICs Analog ICs Buzzers Fuses RBs ZDs SMD-Ds RDs STs SMD-Ts ILTs PTs FETs Thyristors OCs LEDs Digitrons CPUs chips TPRs Fuses RBs	ISs Tuners LOTs .08 0.10 6.50 .57 4.94 0.61 .06 0.09 0.07 D 0.01 D
ZDs SMD-Ds RDs STs SMD-Ts ILTs PTs FETs Thyristors OCs LEDs Digitrons CPUs chips TPRs	0.08 0.10 6.50 .57 4.94 0.61 .06 0.09 0.07 UD 0.01 UD
	0.08 0.10 6.50 0.57 4.94 0.61 0.06 0.09 0.07
Cu 100 0.27 0.81 0.44 0.88 0.91 0.14 0.23 0.76 0.16 4.09 1.55 0.76 1.92 0.90 0.44 0.32 0.30 0.76	0.57 4.94 0.61 0.06 0.09 0.07
Zn 100 0.55 0.42 0.82 ND 0.22 2.44 9.28 0.16 13.32 1.29 0.78 0.95 ND 0.59 0.82 0.39 4.32 ND	.06 0.09 0.07
Cd 1 ND ND 0.06 ND 0.05 0.05 0.04 0.35 ND 0.13 0.05 0.03 0.06 0.08 0.06 0.07 ND 0.03	ID 0.01 ND
Pb 5 3.47 1.72 3.65 ND 2.68 4.25 4.32 ND 0.01 ND ND ND 2.67 ND 3.65 1.65 0.47 2.50	ID 0.01 ND
Cr 5 ND	ID ND ND
Hg 0.1 ND	ID ND ND
Be 0.02 ND	ID ND ND
Ba 100 0.02 ND 0.08 0.04 0.05 0.01 0.03 ND ND 0.05 0.05 0.09 0.12 ND 0.08 0.02 ND 0.01	D 0.06 0.18
Ni 5 0.33 3.02 0.10 2.36 0.05 0.09 0.05 0.02 0.01 0.15 0.44 0.25 0.17 0.13 0.10 0.05 35.39 0.06	.11 0.32 0.08
Ag 5 ND ND ND ND ND ND 1.42 ND	JD ND ND
As 5 ND ND 0.39 0.18 ND ND ND ND 0.04 ND ND ND ND ND ND 0.39 0.46 ND ND	D ND 0.15
Se 1 0.01 ND ND 0.05 ND ND ND 0.23 ND 0.04 0.07 ND ND ND ND ND ND ND ND ND	JD ND 0.01
TCLP	
Threshold Discrete device (DDs) Photoelectric semiconductor (PhSs) Logic ICs Analog ICs Buzzers Fuses RBs	ISs Tuners LOTs
ZDs SMD-Ds RDs STs SMD-Ts ILTs PTs FETs Thyristors OCs LEDs Digitrons CPUs chips TPRs	
Ar 5 ND 0.61 7.49 3.11 ND ND ND ND 4.04 ND ND 2.54 5.62 ND 14.22 0.56 ND 5.22	
עם אין ער 10, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1,	JD ND ND
Ba 100	14 072 020
C_{11} C_{12} C	JD ND ND
G_{4} , G_{5} , G_{5	
Co 1 ND 11.02 5.82 ND 0.30 3.00 ND ND 0.10 13.38 20.78 ND ND 5.6 ND 0.07 0.63 ND	214 0.84 ND
A 0 5 0 5 1 10 0 5 ND 0 5 6 0 5 110 ND 114 ND ND 114 112 112 ND ND 114	JD ND ND
Ho 0.2 ND	JD ND ND

Note: ND: not detected; concentrations in bold are above the regulatory limit.

10–665 mg/kg. Gold has been found in many electrical and electronic industries due to its excellent electrical conductivity and excellent corrosion resistance (Fan et al., 2020). For ECs separated from the WPCBs of computer, the metallic pins are covered with thin layers of nickel and gold (Mesquita et al., 2018). Pd was detected in all the ECs groups with contents of 5–502 mg/kg, mainly from electrode (Liu et al., 2022). The content of Pd is one order of magnitude higher than Pt. For Pt, only eleven groups of ECs contain extremely small amounts, ranging 1–42 mg/kg. The maximum content of platinum detected is from tactile switchs, 85 mg/kg, SI Table S12. Pt is rare in both ECs and WPCBs, only special use in space electronics and some electronics components like hard drives (D'Adamo et al., 2019).

3.4. Metalloids

Fig. 2 presents the average concentrations of metalloids, including B, Se, and As. The detailed metalloids contents of ECs are shown in SI Tables S14 and S15. Small amounts of these elements are used as semiconductor materials in ECs (Miao et al., 2017; Servanton and Pantel, 2010; Sivaprakash et al., 2021). Twelve groups of ECs have detected boron (0-6405 mg/kg), with a total average content of 1988 mg/kg, relatively abundant than the other two metalloids. Boron exists in the material in form of different compounds to meet various functional requirements. Boron present in semiconductors in form of boron nitride, exhibiting excellent mobility, optics, bandgap, stability, and other properties (Brasse et al., 2010; Ding et al., 2009). Zirconium boride exists in high-temperature ceramic materials and has high thermal conductivity and excellent heat resistance (Chevykalova et al., 2014). Boron phosphate is deposited in form of a thin film on the silicon dioxide substrate of the Schottky diode (Dalui and Pal, 2008). The maximum content of boron is found in tact switches (9560 mg/kg, Table S14). The contents of Se in different ECs varies greatly (150-1500 mg/kg), with a total average content of 681 mg/kg. Switchs contain the highest concentrations of Se (4785 mg/kg), followed by crystal oscillators (3370 mg/kg). For As, eleven groups of ECs contain it, of which the arsenic contents of 4 groups are lower than 500 mg/kg, and the rest are higher than 1000 mg/kg. Arsenic content in the relay is the highest, reaching 3300 mg/kg.

3.5. Metal content comparison between ECs and WPCBs

The metals contents in ECs and printed wiring boards (PWBs, i.e., WPCBs without ECs) vary greatly. On average, all metals accounted for approximately 67.65% for the ECs, higher than that in PWBs (40%) (Duan et al., 2011). Meanwhile, copper contents in ECs fluctuates greatly according to different functions performed. For ECs, the most abundant is copper, with the contents ranging in 5.9-937.06 g/kg. For PWBs, the most abundant metal is also copper (-16%), followed by metals such as aluminum, iron, tin, lead, and zinc, with mass proportions ranging 1% to 5% (Park and Fray, 2009; Wang and Xu, 2015). The contents of iron in ECs are 0–70%, which is much higher than PWBs (5%). Other metals such as aluminum, tin, lead, and zinc have maximum contents ranging 12% to 75%, which is higher than the corresponding metals contents in PWBs. For precious metals, there are very little content in PWBs (mobile phone PWBs are not discussed here) While in ECs, the contents are relatively high, such as silver (0-0.529%), gold (0-0.067%), palladium (0-0.05%), platinum (0-0.004%).

It can be found that the contents of ordinary metals or rare precious metals in ECs are generally higher than those in PWBs. For these valuable metals, it is very difficult and high costly if they are processed collectively and recycled separately, and even the mixture of various substances will likely generate toxic substances (Wang and Xu, 2015). The stepwise sorting and recycling of PWBs and ECs will effectively improve the separation efficiency of various metals. The notorious toxic metals, Pb, Hg, Cd, Cr^{6+} have been restricted in the RoHS 3 (EU Directive 2015/863). The threshold for cadmium is 100 mg/kg and the

others are 1000 mg/kg. In this research, Hg was not detected in any ECs. For Pb, the concentrations in most ECs exceeds the threshold.

3.6. Hazardous waste assessment of EEs

The details of EEs leaching toxicity are listed in Tables 4 and 5 including HJ/T299-2007 and TCLP method, respectively. In the HJ/ T299-2007 method, the immigrant ratio of heavy metals is low, except nickel in passive crystal oscillator (5.309 mg/L) and filter (17.54 mg/L), of which the leachate concentration exceeds the threshold (5.0 mg/L). Because nickel is used as the surface electroplate of EEs, which is stable in the air and has excellent polish ability and hardness; in TCLP, there are more EEs with excessive arsenic and selenium. The only one kind of EEs with excessive arsenic (5.0 mg/L) is the DRs (5.68 mg/L). The EEs with excessive selenium (1.0 mg/L) have FCs (4.13 mg/L), CCs (17.26 mg/L), CIs (1.87 mg/L), HPIs (4.65 mg/L), SMD-PIs (16.63 mg/L), DRs (4.39 mg/L), PCOs (1.8 mg/L), TSs (6.02 mg/L), connectors (9.62 mg/L) and ports (2.54 mg/L).

Comparing the two methods HJ/T299-2007 and TCLP for EEs, Cr and Hg were not detected in both methods. Ba, Cd and Ag have low contents below the threshold. According to China regulation, only PCOs and EFs in EEs are hazardous waste because of excessive Ni. However, Ni is not the focus of TCLP. For the United States regulation, DRs with excessive As and EEs with excessive Se including capacitors (FCs, CCs), inductors (CIs, HPIs, SMD-PIs), DRs, PCOs, TSs and linkers (connectors, ports) are hazardous waste.

3.7. Hazardous waste assessment of EDs

The results from the toxicity characteristic leaching tests of EDs, respectively HJ/T299-2007 and TCLP method, are presented in Table 6. For HJ/T299-2007, fuse have excessive Ni (35.39 mg/L), because nickel is used as electroplating on the copper caps at both ends. For TCLP, there are more kind of EDs with excessive As and Se, and only one kind of EDs with excessive Pb. RBs is the only kind of EDs with excessive Pb (8.94 mg/L), which are composed of 4 rectifier diodes. The EDs with excessive arsenic (5.0 mg/L) are RDs (7.48 mg/L), CPUs (6.62 mg/L), TPRs (14.32 mg/L) and RBs (5.22 mg/L). For RBs and RDs, the N-type semiconductor is usually silicon or gallium arsenide, and the pin plating is made of leadtin alloy (Özdemir et al., 2003). Integrated circuits, such as CPUs and TPRs, use monocrystalline silicon or gallium arsenide monocrystalline as the substrate to make many transistors and resistors, and integrate the ECs into a complete electronic circuit (Jameel et al., 2015). The EDs whose selenium exceeds the threshold (1.0 mg/L) have SMD-Ds (11.02 mg/L), RDs (5.82 mg/L), ILTs (3.0 mg/L), OCs (13.38 mg/L), LEDs (20.78 mg/L), chips (5.56 mg/L) and HSs (2.14 mg/L). Selenium, with its photosensitive and semiconductor properties, is often used to manufacture photocells, photosensitive devices, laser devices, infrared controllers, phototubes, photoresistors, optical instruments, photometers, rectifiers, and so on in the electronic industry. For all EDs, in both HJ/T299-2007 and TCLP methods, Hg and Cr were not detected. Under China's regulatory policies, fuses should be categorized as hazardous waste because of excessive Ni. Under the U.S. regulatory policies, EDs with excessive As including RDs, CPUs, TPRs and RBs, and excessive Se including SMD-Ds, RDs, ILTs, OCs, LEDs, chips and HSs, are hazardous waste.

4. Conclusion

Through this research, ECs both EEs and EDs, including all types of ECs in home appliances that the Chinese WEEE market currently addresses were surveyed. ECs contain abundant metal resources, especially compared to PWBs, and it is recommended to separately recycle ECs and PWBs from WPCBs. Moreover, it is recommended to sort and recycle ECs according to the most abundant metal contents. On the other hand, with increasingly strict environmental regulations and policies, toxic pollutants such as chromium and mercury were found to not exceed the standard thresholds established by regulatory policies in China and the U.S. However, new toxic threats were emerged, mainly from arsenic and selenium because their leachable concentrations in 20 of 50 ECs exceeded the regulatory standard. These results call for alternatives assessment studies to identify new materials with similar function but less toxic as replacements for these metalloids in electronics. This research contributes results to bridge major knowledge gaps in WPCBs recycling in a way that protects environmental quality and human health.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.wasman.2022.01.019.

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J. Huang et al.

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