



Toxicity trends in E-Waste: A comparative analysis of metals in discarded mobile phones

Narendra Singh^{a,b,*}, Huabo Duan^{a,*}, Oladele A. Ogunseitan^c, Jinhui Li^d, Yuanyuan Tang^b

^a School of Civil Engineering, Shenzhen University, Shenzhen, 518060, China

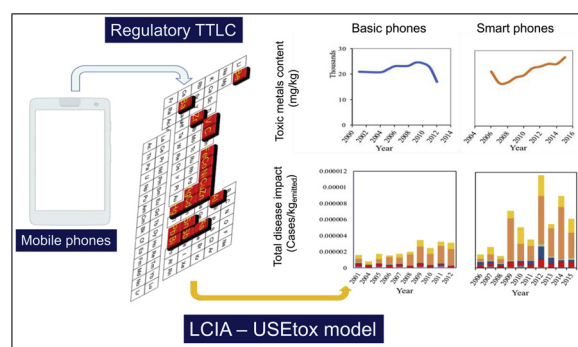
^b School of Environmental Science and Engineering, Southern University of Science and Technology, Shenzhen, 518055, China

^c Department of Population Health & Disease Prevention, Program in Public Health, University of California, Irvine, CA, 92697, USA

^d State Key Joint Laboratory of Environment Simulation and Pollution Control, School of Environment, Tsinghua University, Beijing, 100084, China



GRAPHICAL ABSTRACT



ARTICLE INFO

Editor: Deyi Hou

Keywords:

Mobile phones
Human health
Ecotoxicity
Toxic elements
E-waste

ABSTRACT

Mobile phones and various electronic products contribute to the world's fastest-growing category of hazardous waste with international repercussions. We investigated the trends in potential human health impacts and ecotoxicity of waste mobile phones through quantitative life cycle impact assessment (LCIA) methods and regulatory total threshold limit concentrations. A market-dominant sample of waste basic phones and smart-phones manufactured between 2001 and 2015, were analyzed for toxicity trends based on 19 chemicals. The results of the LCIA (using USEtox model) show an increase in the relative mass of toxic materials over the 15-year period. We found no significant changes in the use of toxic components in basic phones, whereas smart-phones contained a statistically significant increase in the content of toxic materials from 2006 to 2015. Nickel contributed the largest risk for carcinogens in mobile phones, but the contributions of lead and beryllium were also notable. Silver, zinc and copper contents were associated with non-cancer health risks. Copper components at 45,818–77,938 PAF m³/kg dominated ecotoxicity risks in mobile phones. Overall, these results highlight the increasing importance of monitoring trends in materials use for electronic product manufacturing and electronic-waste management processes that should prevent human and environmental exposures to toxic components.

* Corresponding authors at: Room 304, School of Civil Engineering, Shenzhen University, Shenzhen, 518060, China.

E-mail addresses: singhn@sustech.edu.cn (N. Singh), huabo@szu.edu.cn (H. Duan).

<https://doi.org/10.1016/j.jhazmat.2019.120898>

Received 18 October 2018; Received in revised form 24 June 2019; Accepted 13 July 2019

Available online 18 July 2019

0304-3894/ © 2019 Elsevier B.V. All rights reserved.

1. Introduction

Rapid developments in the electrical and electronic equipment's (EEEs), especially in mobile phones and their ever-wider distribution with the increasing current demands and shorter lifespan which is typically less than 2–3 years, has ultimately led to high generation of waste mobile phones (WMPs) (Tansel, 2017; Hira et al., 2018; Tan et al., 2017). During the past three decades, mobile phone technology has had transformative international impacts with an estimated 7 billion users worldwide, and a population-level penetration rate of 97% in 2016 (ITU, 2017). Given the popular demands for innovation in the mobile phone industry, it is likely that the use of “high tech” materials in manufacturing will continue to increase. Many of these materials will be untested in terms of resource depletion, and potential impacts on environmental quality and human health (Ogunseitan et al., 2013; Ogunseitan and Schoenung, 2012). The United Nations reported that approximately 45 million metric tons (Mt) of e-waste was generated globally in 2016, an 8% increase from 2014 level, and predicted further increase to 21% by 2021 (Baldé et al., 2017; Singh et al., 2016a).

The materials composition of WMP is similar to others waste EEEs, including printed circuit boards (PCBs) and Liquid crystal display (LCD) screens, which represent 50% to 80% of mass per unit. The chemical composition of PCBs and LCDs include precious metals (e.g. gold, silver, and palladium), rare or conflict minerals (e.g. coltan), and a variety of potential hazardous substances, including toxic metals such as arsenic, beryllium, cadmium, lead, antimony, nickel, and organic chemicals (e.g. halogenated flame retardants), which could potentially threaten human life and environment quality if improperly managed (Lincoln et al., 2007; Kiddee et al., 2013; Nnorom and Osibanjo, 2009). In most of the developed and developing countries, waste PCBs from WMBs are considered hazardous because of their toxic constituents (Chen et al., 2016; Yadav et al., 2014; Stuhlfarrer et al., 2016). The hazardous substances in e-waste have been regulated by the various government agencies because of their toxicity or persistence in organisms and food webs (Hira et al., 2018; Nnorom and Osibanjo, 2009; Hibbert and Ogunseitan, 2014; Tang et al., 2010; Henriquez-Hernandez et al., 2017). Human exposure to these hazardous substances in e-waste can disrupt important physiological processes resulting in diseases (An et al., 2014; Ceballos and Dong, 2016; Song and Li, 2014).

The most important approaches to manage WMPs in a sustainable way have focused on dismantling, recycling, and material resources recovery (Chen et al., 2016; Singh et al., 2018), but industrial-scale implementation still is a challenge because of the lower economic incentive of processing mixed WMPs components. For example, China produces as many as 450 million mobile phones annually. Disposal of obsolete mobile phones generated in 2015 was 400 million units, of which fewer than 2% was recycled (MIIT, 2016; ECNS, 2017); In the US, only 10% of the obsolete mobile phones are recycled, while the remaining 90% is stored at home by users or disposed in landfills, ultimately resulting the environmental pollution by toxic substances (Chen et al., 2018; Silveira and Chang, 2010).

Since the recognition of e-waste as a major environmental threat, various domestic and international governments or organizations have enacted legislation to curb the use of hazardous substances in EEEs, including “Directive on the restriction of the use of certain hazardous substance in electrical and electronic equipment” (RoHS) and the “Waste Electrical and Electronic Equipment Directive” (WEEE) which are considered among the best examples (RoHS, 2018; European Directive, 2011). However, the indication is that the use of toxic metals such as lead, chromium, cadmium, mercury and others in the EEEs, specifically in the mobile phones, has been changing with the innovation and rapid technological advancement (Charles et al., 2017; Singh et al., 2016b; Schoenung et al., 2004). Manufacturers are motivated by profit-margins, functional reliability, and compliance with regulatory limits, which may not be sufficiently stringent to eliminate potentially toxic exposures (Chen et al., 2018).

In this study, we collected and analyzed the material composition of waste basic phones and smartphones manufactured between 2001 and 2015. After manually dismantling of these waste devices, all components or parts (excluding battery and cover case) were quantified the concentrations of toxic chemicals. We then used a life cycle impact assessment approach- USEtox model-to determine the trends of potential impacts on human health (cancer and non-cancer disease) and ecological toxicity. The results are useful for the eco-design of EEEs and provide a valuable information to guide the development of cross-cutting e-waste management strategies.

2. Materials and methods

2.1. Sample preparation

A total of 20 waste mobile phones including 10 types of basic phones and 10 types of smartphones were collected from the local WMPs recycling company in Shenzhen city- one of the major domestic WMPs recycling facilities in China. The samples were grouped by the physical dimensions and their types (brand of the manufacturer) are shown in Supporting Information (SI) Table S1. In order to separate basic phones and smartphones, devices with the smaller size of display including the keypad were categorized as basic phones which enables only voice messaging. Conversely, the devices with bigger size of display and camera, a high-resolution touch screen display, internet connectivity capability, and ability to accept sophisticated application were categorized as smartphones (Techopedia, 2018). The selection of the manufacturer was based on their popularity and the coverage of market share in the last decades (Wikipedia, 2018a). Each device was weighed with analytical balance and then the plastic cover case was manually removed from the phones. The batteries and plastic cover case were excluded from the study because the batteries and the half plastic cover case had already been removed from the WMPs at the mobile recycling station and sent to a special battery recycling station.

2.2. Toxic elements analysis

Different parts of the mobile phones were cut into small pieces about 1 cm² using a pair of standard scissors, and the small pieces of the WMPs were then combined separately and cooled in liquid nitrogen before milling to provide a reasonable homogenized powder of particle size less than 0.1 mm, sufficient for the complete detection of the toxic metals. The milling process was carried out using cryogenic planetary ball mill systems. 0.2 g samples of the homogenous powder were taken and subjected to microwave-assisted acid digestion. The International Electro-technical Commission (IEC) methods IEC 62321-4:2013 CV-AAS (cold vapor atomic absorption spectrometry), and ICP-OES (inductively coupled plasma optical emission spectrometry); (IEC, 2017) and IEC 62321-5:2013 ICP-MS (inductively coupled plasma mass spectrometry) (IEC, 2013) were used to analyze the concentration of copper (Cu), nickel (Ni), aluminum (Al), barium (Ba), iron (Fe), zinc (Zn), silver (Ag), chromium (Cr), zirconium (Zr), lead (Pb), tin (Sn), beryllium (Be), cobalt (Co), bismuth (Bi), arsenic (As), vanadium (V), antimony (Sb), cadmium (Cd) and mercury (Hg). A complete process of the analysis is described in Fig. S1 of the Supporting Information (SI). The final reading of the samples was taken as the average value of three readings of each sample.

2.3. Human health and environmental impact assessment using USEtox

We used the life cycle impact assessment (LCIA) model - USEtox version 2.0 (<http://www.usetox.org>) - to assess the potential impacts of the toxic chemical in WMPs on human health and environmental quality. USEtox is a life cycle initiative (LCI) model based on the scientific consensus of international researchers and was developed under the guidance of the United Nations Environment Program (UNEP) and

Table 1

The concentration of the metals in the waste basic phones.

Elements	S1	S2	S3	S4	S5	S6	S7	S8	S9	S10	Mean
Cu	370,632	369,187	381,752	409,528	396,281	397,867	423,727	390,371	368,526	276,186	378,406
Ag	411	105	532	1,112	1,105	2,262	2,128	2,541	3,262	2,541	1,600
Fe	2,845	2,424	5,395	6,975	5,734	3,150	4,184	3,984	4,063	3,921	4,268
Al	5,382	3,873	8,537	6,381	15,281	18,320	15,834	11,946	17,493	12,208	11,526
V	2.4	2.4	5.3	6.7	8.3	5.9	4.2	9.2	3.5	7.2	5.5
Sn	39	27	52	350	536	263	481	24	69	110	195
Ni	8,381	7,942	8,352	6,031	8,361	6,705	9,487	7,493	7,846	10,696	8,129
Zn	2,536	2,354	3,735	1,301	3,635	3,235	4,763	7,846	4,645	5,390	3,944
Ba	4,735	5,565	3,652	3,422	5,635	8,429	2,744	9,845	16,498	8,726	6,925
Zr	456	635	231	495	654	293	274	165	2,746	1,204	715
Bi	45.3	68.6	27.4	22.7	6.3	7.6	27.4	8.4	22.4	5.3	24
Co	78.3	32.2	27.4	1,012	148.3	87.4	59.3	41.4	37.4	19.5	167
Be	210	75	325	104	443	ND	323	11	564	ND	206
Cd	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Pb	201	113	47	149	134	127	122	432	345	104	177
Sb	5.1	ND	4.2	0.0	6.3	0.0	3.2	7.2	9.2	0.0	3.5
As	6.7	8.4	8.3	1.9	4.2	3.5	8.2	3.1	6.9	4.9	5.6
Cr	603	494	704	535	663	413	992	1,102	1,384	495	739
Hg	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND

ND = Not detected; Unit = mg/kg.

the Society for Environmental Toxicology and Chemistry (SETAC).¹³ This scientific model was used to characterize the potential impact of toxic chemicals in products on human toxicology and ecotoxicology, and the output of USEtox modeling include, human toxicity criteria related to outcomes such as the incidence of cancers and non-cancer diseases associated with toxic emission concentrations and multi-media exposure assessments, and ecotoxicity in terms of potentially affected fraction (PAF) of species due to change in concentration of toxic emissions (PAF m³ day/kg) (Chen et al., 2018; Hauschild et al., 2008; Kang et al., 2013). In this study, USEtox was used to calculate the potential impacts of toxic elements on human health and environment that we found in WMPs; the calculation was done by the following eq.:

$$IS_x = C_x \cdot M \cdot C_{fx}$$

Where IS represents the impact score of metal x in the WMP; C_x is the concentration of metal x in the WMP (cases/kg); M is the mass of each sample in kg; C_{fx} is the weighting or characterization factor for the corresponding potential of metal x. Data of toxic metals used in the calculation are shown in Tables 1 and 2. The calculated characterization factors for ecotoxicity and human toxicity which includes cancer,

non-cancer and total impacts (cancer and non-cancer) were associated with impacts of toxic metals emitted to urban air, rural air, fresh water, sea water, agricultural soil and natural soil. The detailed calculation values are attached in Tables S2–S19 in SI. The unit of the characterization factor for ecotoxicity was (PAF. m³.day/kg_{emitted}) and for human toxicity was (cases/kg_{emitted}) (Huijbregts et al., 2010).

3. Results

3.1. Metals content in the waste mobile phones

Both basic phones and smartphones have been selected for characterizing the content of toxic metals. Specifically, the evaluation of 19 metals' content in basic phones are presented in Table 1. Copper was the most significant metals in terms of mass ranging from 276 to 424 g/kg, with an average of 378 g/kg; followed by aluminum ranging from 3.9 g/kg to 17.6 g/kg, with an average 11.5 g/kg, and nickel ranging from 6.7 g/kg to 10.7 g/kg, with an average 8.1 g/kg. The use of copper in the main PCBs and flexible PCBs between the screen and keypad to facilitate electrical conductivity in the mobile phones are the main

Table 2

The concentration of the metals in the waste smartphones.

S.no.	S11	S12	S13	S14	S15	S16	S17	S18	S19	S20	Mean
Cu	369,403	268,945	283,930	311,070	330,095	379,362	375,111	413,380	409,679	434,628	357,560
Ag	449	660	549	2,552	1,437	1,924	2,338	2,660	4,323	2,437	1,933
Fe	2,634	5,935	2,138	7,915	5,223	7,364	8,944	3,109	4,693	8,492	5,645
Al	12,748	14,836	17,967	9,754	6,282	10,382	12,168	15,379	12,945	16,284	12,875
V	5.3	4.2	6.2	3.8	6.9	5.3	5.3	4.3	7.2	6.2	5.5
Sn	583	404	622	621	369	603	805	312	594	748	566
Ni	12,645	15,382	10,775	16,596	14,059	13,856	25,374	21,729	18,113	26,935	17,546
Zn	3,936	4,926	2,021	2,683	8,101	4,836	5,691	2,428	5,200	7,452	4,727
Ba	5,037	9,573	8,256	13,102	14,199	12,856	15,419	7,487	10,832	18,475	11,524
Zr	285	396	509	242	833	382	306	446	520	593	451
Bi	52.5	37.4	60.7	61.1	25.2	58.4	21.2	61.6	43.9	83.2	51
Co	83.4	58.4	72.7	70.8	129	273	372	74.5	52.8	372	156
Be	98.3	231.6	ND	49.6	220	123.2	19.7	ND	112.1	90	94
Cd	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Pb	1223	729	349	95	299	983	1309	1155	323	576	704
Sb	5.6	11.2	2.5	ND	ND	8.3	ND	ND	2.1	4.6	3.4
As	17.3	5.2	2.2	8.9	2.7	11.2	4.2	4.1	4.8	3.8	6.4
Cr	704	111	251	314	777	988	336	349	401	821	505
Hg	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND

ND = Not detected; Unit = mg/kg.

Bold = Exceeding the regulatory limits.

reason behind its abundance in the basic phones (Chen et al., 2015). Aluminum and nickel are mostly used in the welding and in the cover case of the screen which provides strong internal support in the mobile phone. Barium and iron were the next in the list, which represented about 6,925 mg/kg and 4,267 mg/kg (on average), followed by zinc, silver and chromium about 3,944 mg/kg, 1,600 mg/kg and 739 mg/kg (on average), respectively. Then the zirconium, beryllium, tin, lead, cobalt, bismuth, arsenic, vanadium and antimony were about 715 mg, 206 mg, 195 mg, 177 mg, 167 mg, 24 mg, 6 mg, 6 mg, and 4 mg in one kg of basic phones, respectively. Cadmium and mercury metals were not detected in all samples.

Similarly, the concentration of 19 metals contained in smartphones are presented in Table 2. Again, copper was the most significant metals ranging from 269 to 435 g/kg, with an average of 358 g/kg; followed by nickel ranging from 10.8 g/kg to 17.5 g/kg, with an average 11.5 g/kg, and aluminum ranging from 6.3 g/kg to 16.3 g/kg, with an average 12.9 g/kg. Similar to the basic phones, the use of copper in the main PCBs and flexible PCBs are the major reason behind its highest amount mass in smartphones. Nickel content was the second highest mass after the copper in the smartphones, the maximum use of nickel in the smartphones as compared to basic phones could be the reason of its lightweight, hardness and ductile nature than aluminum (Wikipedia, 2018b). Barium and iron were about 11,524 mg/kg and 5,645 mg/kg (on average), followed by zinc, silver and chromium about 4,727 mg/kg, 1,600 mg/kg and 739 mg/kg (on average), respectively. Then the lead, tin, zirconium, cobalt, beryllium, bismuth, arsenic, vanadium and antimony were about 705 mg, 566 mg, 451 mg, 156 mg, 54 mg, 51 mg, 6.4 mg, 5.5 mg, and 3.4 mg in one kg of smartphones, respectively. Cadmium and mercury metals were not detected in all samples.

3.2. Metals evolution and hazardous assessment

In terms of the quantification of the content of metals in various types of WMPs, Fig. 1(a) shows a comparative average content of each metal contained in the basic phones and smartphones. The results show that the content of copper, chromium, zirconium, beryllium and antimony in basic phones were higher in mass than smartphones while the content of nickel, aluminum, barium, iron, zinc, silver, lead, tin, cobalt, bismuth, arsenic and vanadium in smartphones were higher in mass than basic phones. The average mass of nickel in the smartphones (17.5 g/kg) was more than double to the mass of the basic phones which was about 8.1 g/kg. This is probably due to that the bigger in size and advanced functions of some key components such as the screen and PCBs which could require more nickel amount (Wikipedia, 2018b). Similarly, the average mass of barium in smartphone (11.5 g/kg) was almost double the concentration in basic phones which were about 6.9 g/kg. The most statistically significant difference among metals was lead metal, the average content mass of lead in smartphone was about 704 mg/kg, which was almost four times higher in mass than basic phones (177 mg/kg). The main use of lead in the basic phones and smartphones are Sn-Pb solders, which provide a strong support in mobile phones and other EEEs products (Hibbert and Ogunseitan, 2014).

Fig. 1(b and c) shows the average total metals content between basic phones and smartphones from 2001 to 2015. The results show that the total mass content of metals in basic phones were in increasing in trend from 2001 to 2009, and from 2009 to 2012. It was observed in declining in trend. However, in smartphones the average mass content of metals decreased sharply from 2006 to 2007, and then from 2007 to 2015, the trend continued from 17.0 g/kg to 27.3 g/kg. Overall, the average metals content mass in basic phones were in declining in trend or the mass of metals content reduced over the period of time, while in smartphones the average metals content mass were in increasing in trend or the mass of metals content increased over the period of time, the detail explanations are attached in SI Fig. S2.

Table 3 shows the average content of the metals in basic phones and smartphones and the total threshold limit concentration (TTL). The

results show that the average metals content mass of copper, nickel, silver and beryllium were higher than the TTL limits in the waste mobile phones while the average metals content mass of copper, nickel, barium, silver and beryllium were higher than the TTL limits. Average content mass of lead in smartphone was way higher than the basic phones but less than the TTL limit, but the sample number S11, S17 and S18 in smartphones were having the lead content mass more than the TTL limit (shown in Table 2), which shows that the lead content mass of smartphones are almost close to TTL limit. Moreover, the content of lead relatively high in smartphones, despite the European Union's Restriction on Hazardous Substances (RoHS) directive restricting lead level in EEEs (Council, 2003).

3.3. Assessment of human health and environmental impacts

Data on metallic mass obtained through chemical analysis of the basic phones and smartphones (shown in Tables 1 and 2) were used to estimate human health impact characterization factors and ecotoxicity characterization factors with the USEtox 2.0 model to develop potential human disease outcomes and ecological impacts due to exposure to WMPs. Fig. 2 shows the eco-toxicological impact results from the USEtox life cycle assessment of waste mobile phones. Fig. 2(a), the results show that among the metals investigated, copper posed the most significant ecotoxicity risks in basic phones, ranging from 32,306- to 61,859 PAF·m³·day·kg⁻¹, followed by aluminum and nickel. Besides, the ecotoxicity evolution trend in basic phones shows that the average risks of ecotoxicity were in declining in trend from 2001 to 2013, except in 2009 which could be presumed as an exception because of the limited number of samples.

Fig. 2(b) shows the ecotoxicity impacts of smartphones, whereby the results show that among the metals identified, copper posed the significant ecotoxicity risks in smartphones, ranging from 43,761-149,388 PAF·m³·day·kg⁻¹, followed by aluminum and nickel (on average about 318 and 119 PAF·m³·day·kg⁻¹). The evolution trend of ecotoxicity in smartphones was seen in increasing in trend from 2006 to 2015. However, the trend was observed in declining tendency from 2009 to 2011, which might have been because of technological innovation or governmental restriction on toxic metals level on EEEs (Chen et al., 2018). Overall smartphones posed the most significant risks of eco-toxicity with all identified metals except beryllium, which shows more than 50% risks in basic phones.

Fig. 3 shows the result of cancer and non-cancer disease on human. Fig. 3(a) shows among all identified metals, nickel posed the most significant risks of producing cancer diseases, in basic phones, followed by lead, beryllium and arsenic. Similarly, the nickel posed the most significant risks of producing cancer diseases, in smartphones; followed by lead, beryllium and arsenic, shown in Fig. 3(b). Fig. 3(c) shows that among the metals identified in basic phones, silver posed the most significant risks of producing non-cancer diseases, followed by zinc and copper; while in smartphones also, silver posed the most significant risks of producing non-cancer diseases, followed by zinc, Lead and copper.

Fig. 4 shows the total human toxicity (cancer and non-cancer) risk results from the USEtox life cycle assessment of waste mobile phones. Fig. 4(a, b) shows that silver posed the most significant risks of producing total human diseases from basic phones and smartphones, followed by zinc, copper and lead. The risks of total human toxicity trend show that the toxicity risks have been changed into higher in level from 2004 to 2012, while from 2001 to 2004, it has been declined in cell phones. Besides, in smartphones, total human toxicity risks evolution shows that there was no clear trend of declining or sharp increment, however, if we compare from 2006 to 2015, the result shows that the overall risks of total human health have been increased a lot and if we compare from 2012, the trend of risks has fallen down. Overall, the comparison between basic phones and smartphones shows that smartphones posed the most significant risks of producing total human

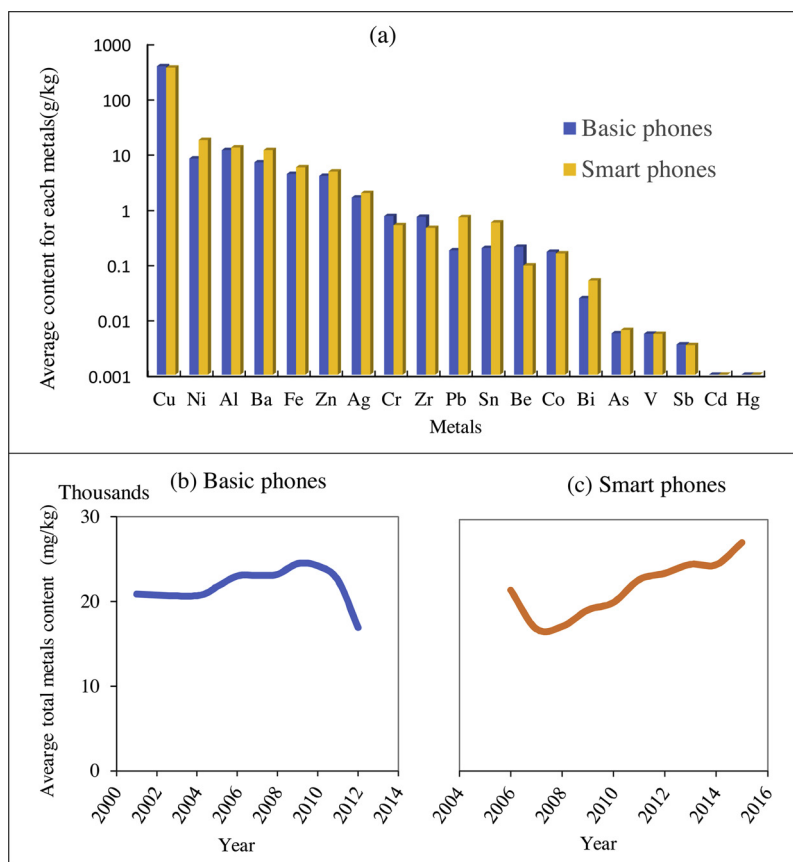


Fig. 1. Toxic metals evolution in the WMPs from 2001 to 2015: (a) average metal content in the basic phones and smartphones; (b) and (c). Average total metal content in the basic phones and smartphones from 2001 to 2015.

Table 3
Average metal content in basic phones and smartphones, and TTCL limit.

Elements	Basic phones mg/kg	Smartphones	TTCL Limit ^a
Cu	438,406	417,560	2,500
Ni	8,129	17,546	2,000
Al	11,526	12,875	*
Ba	6,925	11,524	10,000
Fe	4268	5,645	*
Zn	3,944	4,727	5,000
Ag	1,600	1,933	500
Cr	739	505	2,500
Zr	715	451	*
Pb	177	704	1,000
Sn	195	566	*
Be	206	94	75
Co	167	156	8,000
Bi	24	51	*
As	6	6	500
V	6	6	2,400
Sb	4	3	500
Cd	0	0	100
Hg	0	0	20

* Regulatory limit not set.
^a = Used for California regulated hazardous waste. Source is California Code of Regulations, Title 22, Chapter 11, and Article 3.

diseases as compare to basic phones.

4. Discussion

Mobile phones have become one of the most important and useful electronic devices in modern daily life, and today’s mobile phones have

changed from some large and heavy radio devices into the small and lightweight multimedia products with very advanced functions (B. Convention, 2006; Ylä-Mella et al., 2007). But, the improper treatment of WMPs can result in a loss of valuable materials, which depending on their physical-chemical state and concentrations can create undesirable effects on human health and environmental contamination (Li et al., 2012). The concentration of the toxic metals and its potential impacts on human health and ecotoxicity in WMPs and various parts of the mobile phones have been extensively discussed (Hira et al., 2018; Hibbert and Ogunseitan, 2014; Chen et al., 2018; Kang et al., 2013), and these studies have indicated that the toxic metals content mass in WMPs continue to pose a significant threat to human health and ecosystem. However, the evolution of toxic metals in waste basic phones and smartphones, and this type of comparative analysis was not included in the previous studies.

According to Chen et al. (2018), the pace of technological innovation and regulations development did not show any notable impact on total metal content evolution on WMPs (combined basic phones and smartphones) from 2000 to 2014. However, their finding revealed that the total metal content in the WMPs initially increased from 2002 to 2007 and then decreased in 2013. They also found that the total metals content in the iPhone’s devices were fluctuating on every 2 years of the time period (Chen et al., 2018). These findings proved that there was a big gap in the evolution of total metals content between basic phones and smartphones. In this study, we revealed that the total content in basic phones increased from 2001 to 2009 and then decreased from 2009 to 2012, while in smartphones the total metals content increased from 2007 to 2015.

Various investigators have conducted the chemical analysis of toxic metals in the WMPs, the data obtained by them and the data from this study are shown in Table 4. The previous data of toxic metals in the

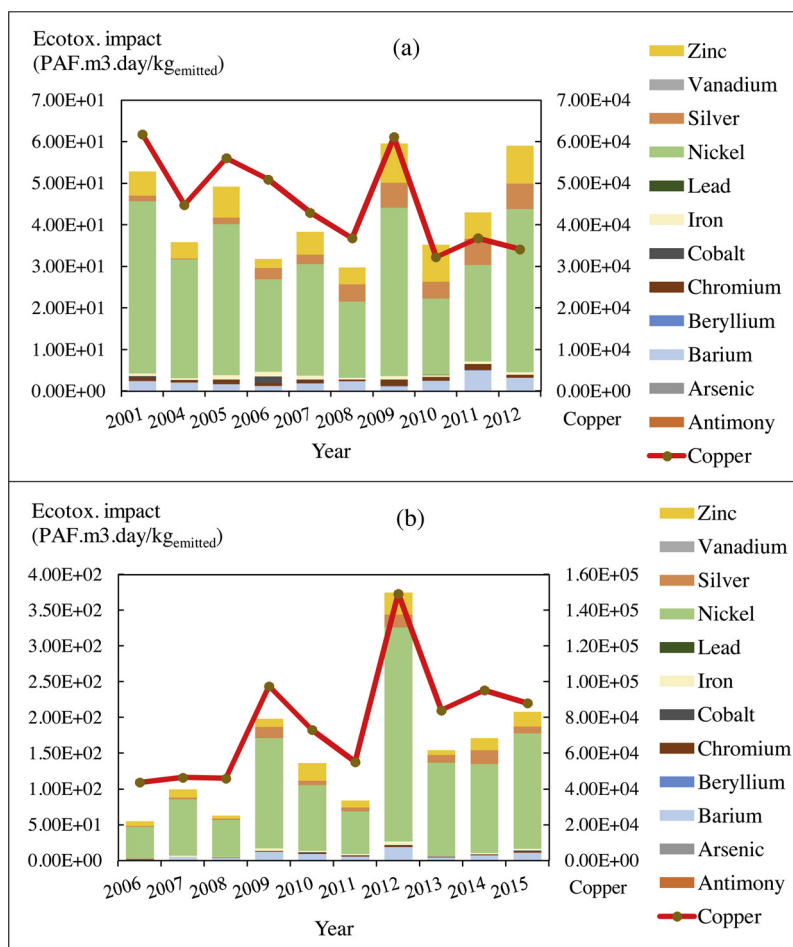


Fig. 2. Eco-toxicological impact results from the USEtox life cycle assessment of waste mobile phones; (a) Basic phones; (b) Smartphones. (Note: aluminum metal is excluded in the figure to make a clearer representation of other minor ecotoxic metals).

WMPs and the data from basic phones and smartphones of this study are compared with the TTLT limit derived from California EPA. While the content of the metals differs between the sources, this table gives a preliminary indication of potentially toxic metals in the WMPs, and basic phones and smartphones. In addition to the metals listed in the WMPs in previous studies in Table 4, results from these studies revealed that the lead, zinc and antimony has surpassed the TTLT limits which were not shown in the present study.

The trend of total toxic metals content in smartphones as compared to basic phones which are waning from the market demand urgently required the telephony industries to rethink about the current design of smartphones and their metals consumption trend. Smartphones recycling centers need to secure a safer mechanism system and take special efforts to protect public health around the disposal centers. Besides, legislators need to strengthen the e-waste regulation to restrict the informal recycling of WMPs and other e-waste which ultimately reduce the exposure of toxic metals identified in this study and potentially prevent adverse impacts on human health and ecosystem.

This study has several constraints and limitations, which should be considered in interpreting the results. As we evaluated the risks of toxic metals evolution in WMPs and comparative analysis between basic phones and smartphones, we did not include organic chemicals such as halogenated flame retardants. We excluded these organics from the study because of the lack of current recovery or recycling strategies for them. Most of the organic substances such as brominated flame retardants (BFRs), dioxins and furans, others are mostly present in various components of plastics and PCBs (Chen et al., 2016; Hibbert and Ogunseitan, 2014). The composition of organic toxic substances in

different parts of WMPs and their content dynamics in the basic phones and smartphones would provide new scope for future studies.

5. Conclusions and policy implications

The results of this research revealed that adverse human health impacts and environmental contaminations are potentially associated with WMPs and the mass of toxic metals content has increased in the past decade especially in the smartphones as compared to basic phones, despite the pace of technical innovation in the mobile phone manufacturing industry. Workers and the general population who are directly exposed to the WMPs, especially the informal recycling sector in developing countries are at high risk for various serious health effects. However, the current approaches for handling WMPs in a sustainable way are metals recovery (especially precious metals), proper dismantling and recycling, but there are very few options that could be used to minimize the risks of WMPs and to assure good management of the toxic metals in WMPs. Thus, the findings of this research support the urgent call for national and international regulation of WMPs, to develop effective management strategies that prevent human exposure and environmental pollution.

In addition, our findings show that the trend of toxic metals in smartphones have been increasing and there were moderate or negligence impacts of recent technological innovations on minimizing the hazardous substances in smartphones. According to various international reports, the possession of smartphones will increase in the coming years worldwide (ITU, 2017; Baldé et al., 2017; ECNS, 2017), which will potentially generate a large amount of obsolete mobile

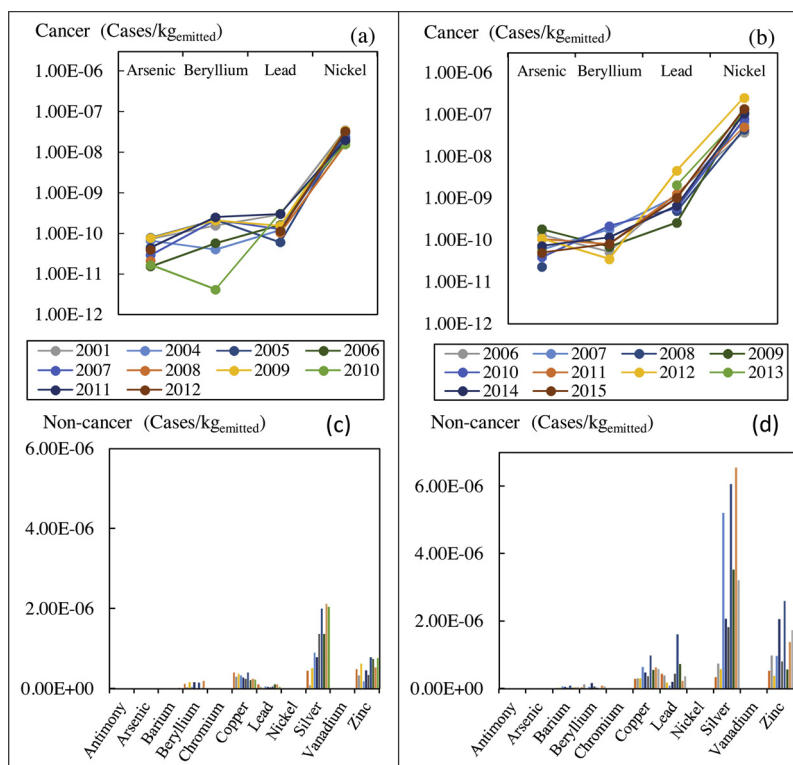


Fig. 3. Cancer and non-cancer disease on human results from USEtox life cycle assessment of waste mobile phones: (a) and (c) basic phones; (b) and (d) Smart phones. (Note: the risks of cancer diseases were associated with only 4 metals-nickel, lead, beryllium and arsenic).

phones. Consequently, these trends of the increasing demand for smartphones and embodied toxic metals in the devices will pose a serious danger to the environment and human health if not properly collected and recycled.

Actually, these results could help inform smartphones manufactures and relevant decision makers (such as Recyclers, government agencies, and designers) around the world to further lessen the use of toxic metals, put forward effective prevention and control measures during the EoL stage, and recover precious metals, specifically in developing countries where the collection and recycling rates are very low (Singh et al., 2018; Borthakur and Govind, 2017; Zeng et al., 2017). Specifically, the growing generation of obsolete mobile phones provides huge potentials of secondary mining or urban mining. For example, a large amount of precious metals (gold, silver, palladium, and platinum) and

other valuable base metals (Cu, Al, Fe, Ni) contained in the discarded mobile phones. Moreover, the increasing amount of toxic metals (Pb, Be, As, Hg) in smartphones should be paid special attention and properly managed to safeguard the environment and human health.

Acknowledgements

This study was supported by the China Postdoctoral Science Foundation (2017M612704), National Natural Science Foundation of China (NSFC21507090, NSFC21507115, NSFC 51575287) and Development of Science and Technology Fund of Macau (FDCT 0011/2018/A).

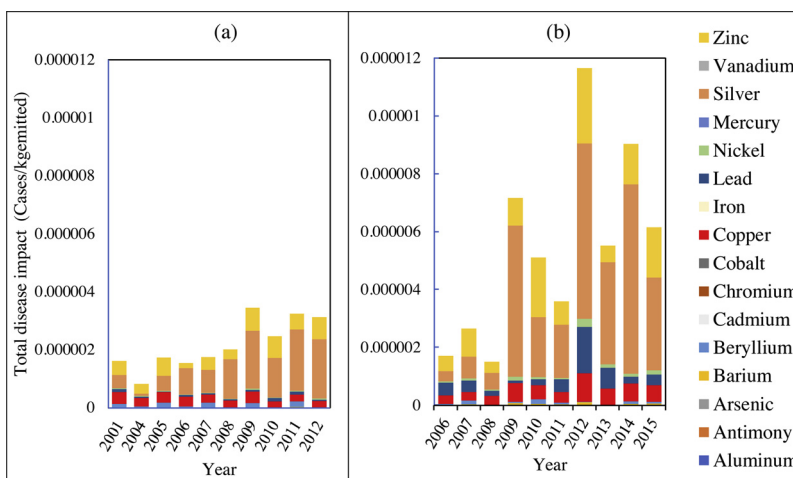


Fig. 4. Total human toxicity (cancer and non-cancer) results from the USEtox life cycle assessment of waste mobile phones: (a) Basic phones; (b) Smartphones (Note: aluminum, cadmium, cobalt and iron metals were not applicable for the human toxicity).

Table 4

The concentrations of the metals in WMPs by the various researchers and this study results in comparison with TTLc limit.

Elements	Hibbert et al., 2014	Chen et al., 2018	Mejame et al., 2018	Lincoln et al., 2007	Kasper et al., 2011	Oguchi et al., 2013	This study, CP	This study, SP	^a TTLc Limit
Cu	393,500	28351	80,133	203,000,0	378,100	330,000	438,406	417,560	2,500
Ni	244,700	16915	11,112	9,247	25400	NT	8,129	17,546	2,000
Al	NT	27567	25,349	NT	6100	15,000	11,526	12,875	*
Ba	24,050	2,385	2,007	5,383	NT	19,000	6,925	11,524	10,000
Fe	NT	34,335	563,29	NT	4850	18,000	4268	5,645	*
Zn	8,650	13,319	14,666	11,007	1820	5,000	3,944	4,727	5,000
Ag	720	608	229	65.9	50	3,800	1,600	1,933	500
Cr	2,635	22,111	11055	958	NT	1,100	739	505	2,500
Zr	NT	NT	NT	NT	NT	NT	715	451	*
Pb	12,200	292	387	10,140	1230	13,000	177	704	1,000
Sn	NT	5,137	6,696	NT	2,550	35,000	195	566	*
Be	43	ND	117	12	NT	21	206	94	75
Co	767	10,778	NT	241	NT	NT	167	156	8,000
Bi	NT	NT	NT	NT	NT	NT	24	51	*
As	73	170	17	36	NT	NT	6	6	500
V	13	352	968	ND	NT	NT	6	6	2,400
Sb	581	426	247	1023	NT	760	4	3	500
Cd	1.3	ND	ND	3	NT	4	ND	ND	100
Hg	0.002	NT	NT	0.79	NT	NT	ND	ND	20

* Regulatory limit not set; ND = Not detected; NT = Not targeted; Unit = mg/kg; CP = Basic phone; SP = Smart phones.

^a = Used for California regulated hazardous waste. Source is California Code of Regulations, Title 22, Chapter 11, and Article 3.

Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:<https://doi.org/10.1016/j.jhazmat.2019.120898>.

References

- An, T., Huang, Y., Li, G., He, Z., Chen, J., Zhang, C., 2014. Pollution profiles and health risk assessment of VOCs emitted during e-waste dismantling processes associated with different dismantling methods. *Environ. Int.* 73, 186–194.
- B. Convention, 2006. Guideline on Material Recovery and Recycling of End-of-Life Mobile Phones. Mobile Phone Partnership Initiative (MPPPI).
- Baldé, C.P., Forti, V., Gray, V., Kuehr, R., Stegmann, P., 2017. The Global E-waste Monitor. United Nations University (UNU), International Telecommunication Union (ITU) & International Solid Waste Association (ISWA), Bonn/Geneva/Vienna.
- Borthakur, A., Govind, M., 2017. Emerging trends in consumers' E-waste disposal behaviour and awareness: a worldwide overview with special focus on India. *Resour. Conserv. Recycl.* 117, 102–113.
- Ceballos, D.M., Dong, Z., 2016. The formal electronic recycling industry: challenges and opportunities in occupational and environmental health research. *Environ. Int.* 95, 157–166.
- Charles, R.G., Douglas, P., Hallin, I.L., Matthews, I., Liversage, G.J.W.M., 2017. An investigation of trends in precious metal and copper content of RAM modules in WEEE: implications for long term recycling potential. *Waste Manag.* 60, 505–520.
- Chen, M., Ogunseitan, O.A., Wang, J., Chen, H., Wang, B., Chen, S., 2016. Evolution of electronic waste toxicity: trends in innovation and regulation. *Environ. Int.* 89–90, 147–154.
- Chen, Y., Chen, M., Li, Y., Wang, B., Chen, S., Xu, Z., 2018. Impact of technological innovation and regulation development on e-waste toxicity: a case study of waste mobile phones. *Sci. Rep.-U. K.* 8, 7100.
- Chen, M., Huang, J., Ogunseitan, O.A., Zhu, N., Wang, Y.-m.J.Wm., 2015. Comparative study on copper leaching from waste printed circuit boards by typical ionic liquid acids. *Waste Manag.* 41, 142–147.
- Council, E., 2003. Directive 2002/95/EC: Restriction of the Use of Certain Hazardous Substances in Electrical and Electronic Equipment. European Union, Brussels, Belgium, pp. 19–23 2003.
- ECNS, 2017. China Faces Mobile Phone Recycling Challenge. <http://www.ecns.cn/2017/02-15/245496.shtml>.
- European Directive, 2011. Directive 2011/65/EU of the European Parliament and of the Council of 8 June 2011, on the Restriction of the Use of Certain Hazardous Substances in Electrical and Electronic Equipment (recast).
- Hauschild, M.Z., Huijbregts, M., Jolliet, O., MacLeod, M., Margni, M., van de Meent, D., Rosenbaum, R.K., McKone, T.E., 2008. Building a Model Based on Scientific Consensus for Life Cycle Impact Assessment of Chemicals: the Search for Harmony and Parsimony. ACS Publications.
- Henriquez-Hernandez, L.A., Boada, L.D., Carranza, C., Perez-Arellano, J.L., Gonzalez-Antuna, A., Camacho, M., Almeida-Gonzalez, M., Zumbado, M., Luzardo, O.P., 2017. Blood levels of toxic metals and rare earth elements commonly found in e-waste may exert subtle effects on hemoglobin concentration in sub-Saharan immigrants. *Environ. Int.* 109, 20–28.
- Hibbert, K., Ogunseitan, O.A., 2014. Risks of toxic ash from artisanal mining of discarded cellphones. *J. Hazard. Mater.* 278, 1–7.
- Hira, M., Yadav, S., Morthekei, P., Linda, A., Kumar, S., Sharma, A., 2018. Mobile phones-an asset or a liability: a study based on characterization and assessment of metals in waste mobile phone components using leaching tests. *J. Hazard. Mater.* 342, 29–40.
- Huijbregts, M., Hauschild, M., Jolliet, O., Margni, M., McKone, T., Rosenbaum, R., Meent, D., 2010. USEtoxTM User Manual v. 1.01. February.
- IEC, 2017. IEC 62321-4:2013+AMD1:2017 CSV Consolidated Version. Determination of Certain Substances in Electrotechnical Products - Part 4: Mercury in Polymers, Metals and Electronics by CV-AAS, CV-AFS, ICP-OES and ICP-MS.
- IEC, 2013. IEC 62321-5:2013. Determination of Certain Substances in Electrotechnical Products - Part 5: Cadmium, Lead and Chromium in Polymers and Electronics and Cadmium and Lead in Metals by AAS, AFS, ICP-OES and ICP-MS.
- ITU, 2017. ICT Facts and Figures. <http://www.itu.int/en/ITUUD/Statistics/Pages/facts/default.aspx>.
- Kang, D.H.P., Chen, M., Ogunseitan, O.A.J.Es., 2013. Potential environmental and human health impacts of rechargeable lithium batteries in electronic waste. *Environ. Sci. Technol.* 47, 5495–5503.
- Kasper, A.C., Berselli, G.B., Freitas, B.D., Tenório, J.A., Bernardes, A.M., Veit, H.M., 2011. Printed wiring boards for mobile phones: characterization and recycling of copper. *Waste Manag.* 31.12, 2536–2545.
- Kiddee, P., Naidu, R., Wong, M.H., 2013. Metals and polybrominated diphenyl ethers leaching from electronic waste in simulated landfills. *J. Hazard. Mater.* 252–253, 243–249.
- Li, X., Ortiz, P., Kuczenski, B., Franklin, D., Chong, F.T., 2012. Mitigating the Environmental Impact of Smartphones with Device Reuse. Sustainable ICTs and Management Systems for Green Computing. IGI Global, pp. 252–282.
- Lincoln, J.D., Ogunseitan, O.A., Shapiro, A.A., Saphores, J.-D.M., 2007. Leaching assessments of hazardous materials in cellular telephones. *Environ. Sci. Technol.* 41, 2572–2578.
- Mejame, P.P.M., Kim, Y.M., Lee, D.S., Lim, S.-R., 2018. Effect of technology development on potential environmental impacts from heavy metals in waste smartphones. *J. Mater. Cycles. Waste* 20, 100–109.
- MIIT, 2016. Statistical Bulletin of Electronic Information Industry. <http://www.miit.gov.cn/n1146312/n1146904/n1648373/c4655602/content.html>.
- Nnorom, I.C., Osibanjo, O., 2009. Toxicity characterization of waste mobile phone plastics. *J. Hazard. Mater.* 161, 183–188.
- Ogunseitan, O.A., Schoenung, J.M.J.Mb., 2012. Human health and ecotoxicological considerations in materials selection for sustainable product development. *Mater. Sustain. Dev.* 37, 356–363.
- Oguchi, M., Sakanakura, H., Terazono, A., 2013. Toxic metals in WEEE: Characterization and substance flow analysis in waste treatment processes. *Sci. Total Environ.* 463, 1124–1132.
- Ogunseitan, O.A., Allgood, J.M., Hammel, S.C., Schoenung, J.M., 2013. Translating the Materials Genome into Safer Consumer Products. ACS Publications.
- RoHS, 2018. RoHS Guide Compliance. <http://www.rohsguide.com/>.
- Schoenung, J.M., Ogunseitan, O.A., Saphores, J.D.M., Shapiro, A.A., 2004. Adopting lead-free electronics: policy differences and knowledge gaps. *J. Ind. Ecol.* 8, 59–85.
- Silveira, G.T., Chang, S.-Y.J.Wm., 2010. Cell phone recycling experiences in the United States and potential recycling options in Brazil. *Waste Manag.* 30, 2278–2291.
- Singh, N., Duan, H., Yin, F., Song, Q., Li, J., 2018. Characterizing the materials composition and recovery potential from waste mobile phones: a comparative evaluation of cellular and smart phones. *ACS Sustain. Chem. Eng.* 6, 13016–13024.
- Singh, N., Li, J., Zeng, X., 2016a. Global responses for recycling waste CRTs in e-waste. *Waste Manag.* 57, 187–197.
- Singh, N., Li, J., Zeng, X., 2016b. Solutions and challenges in recycling waste cathode-ray

- tubes. *J. Clean. Prod.* 133, 188–200.
- Song, Q., Li, J., 2014. A systematic review of the human body burden of e-waste exposure in China. *Environ. Int.* 68, 82–93.
- Stuhlpfarrer, P., Luidold, S., Antrekowitsch, H., 2016. Recycling of waste printed circuit boards with simultaneous enrichment of special metals by using alkaline melts: a green and strategically advantageous solution. *J. Hazard. Mater.* 307, 17–25.
- Tan, Q., Dong, Q., Liu, L., Song, Q., Liang, Y., Li, J., 2017. Potential recycling availability and capacity assessment on typical metals in waste mobile phones: a current research study in China. *J. Clean. Prod.* 148, 509–517.
- Tang, X., Shen, C., Shi, D., Cheema, S.A., Khan, M.I., Zhang, C., Chen, Y., 2010. Heavy metal and persistent organic compound contamination in soil from Wenling: an emerging e-waste recycling city in Taizhou area, China. *J. Hazard. Mater.* 173, 653–660.
- Tansel, B., 2017. From electronic consumer products to e-wastes: global outlook, waste quantities, recycling challenges. *Environ. Int.* 98, 35–45.
- Techopedia, 2018. Smartphone. Definition - What Does Smartphone Mean?.
- Wikipedia, 2018a. Mobile Phone. https://en.wikipedia.org/wiki/Mobile_phone.
- Wikipedia, 2018b. Nickel. <https://en.wikipedia.org/wiki/Nickel>.
- Yadav, S., Yadav, S., Kumar, P., 2014. Metal toxicity assessment of mobile phone parts using Milli Q water. *Waste Manag.* 34, 1274–1278.
- Ylä-Mella, J., Pongrácz, E., Tanskanen, P., Keiski, R.L., 2007. Environmental impact of mobile phones: material content. *Proc. 22nd International Conference on Solid Waste Technology and Management* 1612–1617.
- Zeng, X., Duan, H., Wang, F., Li, J., 2017. Examining environmental management of e-waste: china's experience and lessons. *Renew. Sustain. Energy Rev.* 72, 1076–1082.