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Comparative effectiveness of technical and regulatory innovations to reduce the burden of electronic waste

Shuyuan Chen [a], Rong Wang [b], Jiqin Wang [a], Jiancheng Shu [a], Mengjun Chen [a,*], Oladele A. Ogunseitan [c,*]

[a] Key Laboratory of Solid Waste Treatment and Resource Recycle (SWUST), Ministry of Education, Southwest University of Science and Technology, 59 Qinglong Road, Mianyang, 621010, China
[b] School of National Defense Science and Technology, Southwest University of Science and Technology, Mianyang 621010, China
[c] Department of Population Health and Disease Prevention, University of California, Irvine, CA 92697-3957, United States

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ABSTRACT

Waste Electrical and Electronic Equipment (WEEE) contain toxic metals and organic compounds, posing significant threats to human health and environmental quality. The risks vary according to equipment types and components that are influenced by innovations in technical configuration and regulatory policy. To disentangle the relative impacts of these influences in protecting health and environment, we investigated 13 WEEE and 14 components using a life cycle impact assessment procedure. Additionally, we analyzed Waste Mobile Phones (WMPs) and Waste Printed Circuit Boards (WPCBs) from computers manufactured between 2002 and 2010 to identify trends of toxic chemicals and potential adverse impacts associated with technological configurations and regulatory policies. The results show that, among WEEE, Cathode Ray Tube TV presented the highest carcinogenicity and non-carcinogenicity toxicity potential. Waste Air Conditioner posed the highest potential for ecotoxicity. Among electronic components, Waste Organic Light Emitting Diode displays posed the highest potential risk for carcinogenesis, whereas WPCBs from laptop computers posed the highest potential risk for non-cancer diseases. Solid State Drives posed the highest risks for ecotoxicity. Chromium was associated with risk of carcinogenesis and non-cancer diseases; whereas Al and Fe posed the highest potential for ecotoxicological impacts. During the period covered by the study, innovations in technological configurations and regulatory policies demonstrably reduced the potential toxicity risks posed by E-waste, attributable to reduction in the concentrations of toxic organic chemicals in WMPs/WPCBs. These results advance current understanding of strategies to reduce the risks posed by WEEE through coordination of technological innovations and regulatory policies.

1. Introduction

The demand for frequent innovative updates to the technical capabilities of electronic devices is linked to the increasing generation of waste electrical and electronic equipment (WEEE or E-waste) because of shorter use-phase periods before outdated products are discarded (Arduin et al., 2019; Bovea et al., 2018; Chen et al., 2016; Kumar et al., 2017; Martinho et al., 2017; Schoenung et al., 2004). Improper end-of-life management of E-waste through processes such as incineration and landfilling releases hazardous chemicals including Pb, Cd, Hg, and brominated flame retardants (BFRs) to the environment with adverse impacts on environmental quality and human health (Fu et al., 2018; Ismail and Hanafiah, 2020).

Life Cycle Assessment of the potential impacts of materials used in electronic products has provided insights on the need to develop safer alternatives to problematic materials deemed too toxic to be sustainable (Hibbert and Ogunseitan, 2014; Kang et al., 2013; Lim et al., 2013; Lim and Schoenung, 2009, 2010; Son et al., 2016; Woo et al., 2016). WEEE composition varies substantially across electronic device functions, capabilities, components, and models in terms of period of manufacture (Babich et al., 2020; Liu et al., 2020; Xiao et al., 2020). For example, the cathode materials used in the original generation of Lithium Ion Batteries (LIBs) is $LiCoO_2$ whereas recent generations use $LiNi_{1-x-y}Co_xMn_yO_2$, $LiFePO_4$, or Li polymer batteries (Liu et al., 2019; Xiao et al., 2020). In

* Corresponding author.
E-mail addresses: kyling@swust.edu.cn (M. Chen), oladele.ogunseitan@uci.edu (O.A. Ogunseitan).

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some cases, the quantity of a specific chemicals changes considerably, for example, copper content of waste printed circuit boards (WPCBs) used in Cathode Ray Tube (CRT) televisions, desk computers, and mobile electronic devices vary widely, from 72 000, to 200 000, and 330 000 mg/kg, respectively (Oguchi et al., 2013). Consumers have come to expect annual technological updates to software and hardware, which in turn are linked to decline in WEEE content of precious and semi-precious metals such as gold (Chen et al., 2016).

Major technical innovation is typically accompanied by changes in material constituents, particularly the presence and levels of toxic and precious metals, which may increase; conversely, minor technological advances may decrease quantities of toxic and precious metals per unit product (Singh et al., 2019, 2018). A technologically innovative breakthrough may also provoke consideration of new policy regulations, for example in the use of nanoparticles, especially when it relates to human exposure assessments and environmental fate. Conversely, regulatory policies on environmental protection may spark development of technological side-steps that change material composition of products, for example with respect to the replacement of toxic lead in Sn-Pb solders and other restrictions of Cr^{6+}, Hg, Cd, polybrominated biphenyls and polybrominated diphenyl ethers prompted by European Union's RoHS directive (Adie et al., 2019; Alassali et al., 2020; Chen et al., 2018; Deubzer, 2019). Life Cycle Impact Assessment (LCIA) tools have been used to investigate the potential impacts of specific products or components, but not typically the influence of convergent trends of regulatory policies and technological innovation across electronic product types and components (Lim et al., 2013),(Kang et al., 2013),(Lim and Schoenung, 2009). Therefore, this study aimed to (1) conduct a comparative assessment of toxic metal constituents in WEEE and electronic components, and (2) compare the toxicity evolution trend between WMPs and WPCBs produced and marketed during the period 2002 to 2010.

2. Materials and methods

2.1. Metal contents for WEEE and components by collection

Our review of articles retrieved from the Web of Science, and regulatory policies primarily in China, but extended to other countries informed our selection of 13 WEEE for this research (Andrae and Andersenl, 2010; Bae, 2016; Barba-Gutierrez et al., 2008; Chen et al., 2018; Chung et al., 2011; Control, 2004; Cucchiella et al., 2015; Dhiego Raphael Rodrigues Araujo et al., 2017; Kumar et al., 2019; Li et al., 2008; Lim et al., 2013; Matsuto et al., 2004; Minxi Wang, 2018; Oguchi et al., 2011, 2013; Puneet et al., 2005; Renteria et al., 2011; Song et al., 2015; Taghipour et al., 2014; Tunsu et al., 2014; Yamasue et al., 2009; Zamprogno Rebello et al., 2020; Zeng et al., 2016). We included Incandescent Bulb (IB), Compact Fluorescent Lighting (CFL), Light Emitting Diode (LED) Bulb, Mobile Phone (MP), Desktop Computer (DC), Laptop Computer (LC), Liquid Crystal Display (LCD) Monitor, CRT TV, Plasma Display Panel (PDP) TV, LCD TV, Washing Machine (WM), Refrigerator (RF), Air Conditioner (AC). We also included 14 electronic components, namely, Lithium Battery (LB), Hard Disk Drive (HDD), Solid State Drive (SSD), LCD, Organic Light-emitting Diode (OLED) display, MP WPCB, DC WPCB, LC WPCB, CRT TV WPCB, PDP TV WPCB, LCD TV WPCB, WM WPCB, RF WPCB, and AC WPCB (Bahaloo-Horeh et al., 2018; Buechler et al., 2020; Chen et al., 2016; Cucchiella et al., 2015; Kang et al., 2013; Keith et al., 2008; Kim et al., 2019; Minxi Wang, 2018; Oguchi et al., 2011, 2013; Priya and Hait, 2018a, b; Qi et al., 2020; Sahan et al., 2019; Yamane et al., 2011; Yeom et al., 2018). To compare different WEEE and components, we focused on data for metals because organic pollutants were not uniformly investigated across the two categories.

2.2. Organic pollutants in WMPs and WPCBs

We sampled WMPs and WPCBs from desktop computers manufactured between 2002 and 2010 (Chen et al., 2016 and 2018). To address limitations imposed by sample collection, we categorized WMPs into group A (completed) and group B (without battery or back shell); whereas all WPCBs collected were intact. Information about the samples are presented in Table S1 and S2. Table S3, S4, S5 also includes the concentrations of metals and organic pollutants namely 16 polycyclic aromatic hydrocarbons, polychlorinated biphenyls (monochlorobiphenyl to decachlorobiphenyl), brominated flame retardants (monobromobiphenyl to decabromobiphenyl, monobromodiphenyl ether to decabromodiphenyl ether) and tetrabromobisphenol A. The data were obtained from published research (Chen et al., 2016),(Chen et al., 2018), or analyzed as documented in Table SI.

2.3. Toxicity potential assessment—USEtox™ model

We used USEtox™ (Version 2.1.2, https://usetox.org/) to assess potential risks to ecosystems and human health caused by the release of chemicals from E-waste. Model details and summation characterization factors for substances selected are presented in SI.

3. Results and discussion

3.1. Metal constituents in various WEEE and electronic components

3.1.1. Metal content

Data on the content of metals in 13 WEEE are presented in Table S6. For WEEE (except IB), Cu is abundant in WEEE mainly due to its performance as electrical conductor (Fu et al., 2020) which is an essential property of cables and printed wiring boards, which together account for 4–7% of WEEE (Li et al., 2018; Oguchi et al., 2013). AC contains the highest Cu content (158 965 mg/kg), because its performance in thermal conductivity (Bhanushali et al., 2017); followed by CFL (111 000 mg/kg) where copper is a part of Printed Wring Boards (PWBs) and coil drives (Lim et al., 2013). IB contains the lowest content of Cu, 942 mg/kg since it does not contain PWBs, lighting by a tungsten filament (Li et al., 2018; Lim et al., 2013; Rebello et al., 2020). Iron (Fe) is also abundant (12 800–468 167 mg/kg) in the investigated WEEE except IB (372 mg/kg) and it is mainly from steel and PWBs (Kosai et al., 2020). The contents of Al in 13 WEEE range from 13 883 to 586 125 mg/kg, widely used in WEEE due to its light weight (Chandra et al., 2020). The highest content of Al is found in LED bulb at 586 125 mg/kg, about 50%−60% of the whole LED bulbs (Hendrickson et al., 2010), where the metal is needed for lighting performance (Singh et al., 2020).

Zn concentrations in the 13 WEEE is in the range of 0 to 34 500 mg/kg. Zn is mainly found in the galvanized steel (Woo et al., 2016). For the 13 WEEE, Pb ranges from 7 to 39 941 mg/kg, mainly found in solders (Rawat et al., 2020). For CRT TV, Pb is mainly found in CRT glass (Wang et al., 2019), for preventing radiation (Bhargavi et al., 2015). This is also the reason why CRT TV holds the highest content of Pb and Ba (20 670 mg/kg), while the content of Ba in the other 12 WEEE is 0.4 to 4745 mg/kg. The contents of Cr are low (0.1–284 mg/kg) except MP, used in steel to prevent corrosion or improve hardness (Rawat et al., 2020). Cr in MP is the highest, 22 112 mg/kg, which is consisted with Mejame's study, which showed that the contents of Cr in MPs (not including the batteries) were in the range of 7620–13 323 mg/kg (Mejame et al., 2018). However, it is noted that Cr content in MPs is only 111 to 1384 mg/kg when the battery and cover case were removed (Singh et al., 2019). This means that the biggest sources of Cr in MP should be the cover cases. MP also holds the highest contents of Ni (stainless steel), Co (LBs), Sn (PWBs) and Ag (PWBs and electrical and electronic components), respectively at 16 915, 13 394, 5137, 1304 mg/kg (Ashiq et al., 2018; Kang et al., 2013; Li et al., 2018). Ni (60–1612 mg/kg), Co (0–5389 mg/kg), Ag (2.4–136 mg/kg) in the other 12 WEEE are at least

one order of magnitude lower. For other 12 WEEE, Sn is also mainly from PWBs, ranging from 184 to 4800 mg/kg. In the 13 WEEE, Sb, V, Mo, As and Tl range from 0 to 871, 0 to 342, 0 to 186, 0 to 71 and 0 to 45 mg/kg, respectively. The highest contents of these five are also found in MP. The contents of Hg (0–11 mg/kg), Cd (0–40 mg/kg), Be (0–3.8 mg/kg) are low in the 13 WEEE.

Table S7 provides information on the metal content in the 14 components, which vary significantly, especially for Co (9–190 981 mg/kg), Fe (85–155 000 mg/kg), Cu (14 600–291 025 mg/kg), Al (462–808 750 mg/kg) because they include different electronic components. For LB, Co is present at the highest concentrations among all the investigated metals, 190 981 mg/kg, since its cathode materials is mainly $LiCoO_2$ or $LiNi_{1-x-y}Co_xMn_yO_2$ (Kang et al., 2013). Meanwhile, Co in LB is also present at the highest concentrations for the 14 components. For the other 13 components, Co levels are considerably low (9–307 mg/kg). In LB, Mn is the second (93 923 mg/kg), followed by Al (82 020 mg/kg) and Cu (58 631 mg/kg). Mn concentrations in LB is also the highest among the 14 investigated components. Mn concentrations in other components is at least one magnitude lower. Al and Cu are used as current conductors in LB (Paulino et al., 2008).Occurrence of Al in LB ranks the third highest for the 14 components, just behind HDD (358 752 mg/kg) and SSD (808 750 mg/kg). Al in other 11 components are 1–3 orders of magnitude lower. For electronic drives, the most abundant metal is Al which is used as disk substrates in HDD or as shells for good dissipation in SSD (Han et al., 2020; Ma, 2018). Cu concentrations follow Al concentrations at 89 229 mg/kg in HDD and 97 150 mg/kg in SSD. For the nine WPCBs, except WM WPCB (47 778 mg/kg), the most abundant metal is Cu (61 367–291 025 mg/kg). For screens, LCD and OLED display show the same metal characteristic. When compared with LB, drives and WPCBs, the contents of Cu, Al and Pb in LCD and OLED is much lower, which are respectively 14 600, 15 200 mg/kg for Cu, 462, 3390 mg/kg for Al and 16, 0 mg/kg for Pb.

For the 14 components, HDD contains the highest concentrations of Ba at 18 785 mg/kg, followed by MP WPCB (10 100 mg/kg). HDD uses barium ferrite as the magnetic material (Jing et al., 2014). While the contents of Ba in the rest components range from 94 to 4932 mg/kg. For OLED display, Fe is the most abundant metal, 130 000 mg/kg since it uses iron phthalocyanine as the semiconducting layer for good electrical performance (Boileau et al., 2019). Fe in OLED display is the second highest in the 14 investigated components, just second to SSD (155 000 mg/kg), about 1–4 orders of magnitude higher than other components. OLED display also contains the highest content of Cr among the 14 components, 33 100 mg/kg (auxiliary electrode), followed by HDD which is 17 095 mg/kg (read and write head), 1–3 orders of magnitude higher than other components (Park et al., 2011; Tanvar et al., 2020). For LCD, its most abundant metal is Cu, 14 600 mg/kg. For the 13 components except WPCBs, Cu contents are significantly low.

MP WPCB warrants special consideration: in 9 WPCBs, we discovered the highest concentrations of Cr (958 mg/kg), Ba (10 100 mg/kg), Co (164 mg/kg), As (48 mg/kg) and Mo (75 mg/kg). We also discovered the highest concentration of Cu (291 025 mg/kg), Zn (23 738 mg/kg), Ni (18 133 mg/kg) and Ag (2504 mg/kg) among the 14 components. MP WPCB belongs to flex-rigid board, using metalized holes to connect multi-layers of Cu (Li et al., 2018). WM WPCB is different from other WPCBs. Its most abundant metal is Fe (95 000 mg/kg), followed by Cu (47 778 mg/kg) because some electrical and electronic components (ECs) are be made of Fe in WM WPCB. And WM WPCB only contains a single Cu layer (Li et al., 2018). And this kind of WPCBs technology may be also used in CRT TV and AC. Thus, Cu in WPCBs of WM, CRT TV and AC is one order magnitude lower than it in other WPCBs. For WM WPCB, the contents of Ag, Al, Co, Cr, Sn, Zn are all low. In the 9 WPCBs, LCD TV WPCB contained the highest concentrations of Al (63 000 mg/kg). Furthermore, Pb (52 550 mg/kg) in LC WPCB, Sb (3200 mg/kg) in CRT TV WPCB, Sn (83 000 mg/kg) in RF WPCB are the highest for the 14 components.

Compared with WEEE, the concentrations of a particular metal may be high in a specific component, for example Co in LB, Al in HDD and SSD, and Cr in OLED display. While Cu, Fe, Al are abundant in the 13 WEEE except IB. Most WEEE are integrated by different components like WPCBs and plastics (Oguchi et al., 2013). For toxic metals, Pb, Hg, Cd, Cr^{6+} have been limited in homogeneous materials of EEE in RoHS law (2002/95/EC). The limited content of Cd is 100 mg/kg, while others is 1000 mg/kg. Pb in 6 WEEE (CRT TV, PDP TV, LC, DC, CFL, LCD Monitor, LCD TV) and 11 components (LC WPCB, DC WPCB, AC WPCB, MP WPCB, WM WPCB, RF WPCB, LCD TV WPCB, CRT TV WPCB, PDP TV WPCB, HDD, SSD) exceed the threshold.

However, RoHS has some limitations in that not all the WEEE and components are subject to regulation. RoHS includes exemption of the specific applications of these toxic metals, for examples, Pb used in alloy of steel, aluminum, copper, solders for storage, and CRT glass are exempted. The reason is that technological specifications could not reduce toxic metals like Pb in CRT glass. Also, Hg in CFL should not exceed 5 mg per CFL in exemption. But Hg in the sample of CFL is 11 mg/kg. Though Pb in CRT glass and Hg in CFL are exempted, there still is a suggestion to use more safer e-product with new technology like LCD TV and LED bulb. Cd in the WEEE do not exceed its threshold, while the contents of Cd in RF WPCB, WM WPCB, MP WPCB, DC WPCB, AC WPCB and LC WPCB exceed. Cd is also exempted in electrical contacts and plating. In this study, Cr^{6+} is not considered. However, total Cr could be released in environment and convert into Cr^{6+}in the recycling process of e-waste (Xu et al., 2015). For MP, SSD, HDD and OLED display, the contents of Cr exceed 1000 mg/kg.

3.1.2. Comparative assessment of toxicity potential among WEEE and electronic components

In Fig. 1, we present the results from USEtox™ model assessment of potential toxicity associated with WEEE. The toxicity values on a logarithmic scale on the y axis are shown in panels a, b, c; and the proportion of specific metals relative to toxicity potential of all metals are presented in panels d, e, f. Additional details are presented in Table S8-S10. The relative carcinogenicity potentials, presented in increasing order of magnitude are IB, CFL, RF, WM, LED bulb, AC, LC, LCD Monitor, LCD TV, DC, MP, PDP TV, CRT TV, ranging from 4.07×10^{-8} to 4.67×10^{-4} cases. For the potential to cause or promote non-cancer diseases, the sequence is difference, from CRT TV, PDP TV, DC, LCD TV, AC, RF, LCD Monitor, LC, WM, MP, CFL, LED bulb and IB, in the range of 6.53×10^{-7} to 1.58×10^{-1}cases. Ecotoxicity potentials, ranging from 7.53×10^{3} to 5.30×10^{7} PAF•m^3•day, are in the line of AC, RF, PDP TV, DC, WM, LCD TV, CRT TV, LC, LCD Monitor, LED bulb, MP, CFL, IB. For the 13 WEEE, there are 4–6 orders of magnitude toxicity potential difference. The toxicity potential values are mainly associated with the concentrations of metals with CF. For toxicity contributors, metals with a higher CF are usually considerably higher than those metals with a lower CF.

For potential carcinogenicity, Pb poses the most significant contribution on toxicity potential in CRT TV (94.71%), RF (72.76%). Cr exhibits the highest toxicity potential contribution in MP, LED bulb, WM, LCD Monitor, AC, IB, LC, PDP TV varying from 57.33% to 98.19%. Cr (43.75%) and Ni (43.68%) together pose the most significant role in DC. For LCD TV, Cr (38.78%) and Ni (30.80%) are the most important contributors. For CFL, Hg accounting for 31.99% cancer toxicity potential, followed by Pb (30.56%) and Cd (24.13%). For the potential to cause or promote non-cancer diseases, Cr presents the most significant impact (72.31%) in MP. In AC, Cu consists of 57.20% non-cancer toxicity potential. Zn exhibits the highest non-cancer toxicity potential in LED bulb, CFL, IB, RF, from 63.26% to 81.20%. Pb is associated with the highest toxicity potential ratios in CRT TV, LCD Monitor, PDP TV, LC, DC, LCD TV, WM, ranging from 44.69% to 98.33%. Regarding ecotoxicity potential, Al poses the most significant impact in IB, LED bulb, CFL, DC, LC, MP, PDP TV, ranging from 56.78% to 99.61%. For the other WEEE, Fe content is primarily responsible for ecotoxicity potential with contributions ranging from 51.56% to 78.03%. In addition to Al and Fe, Cu and Zn also contribute significantly to ecotoxicity. The

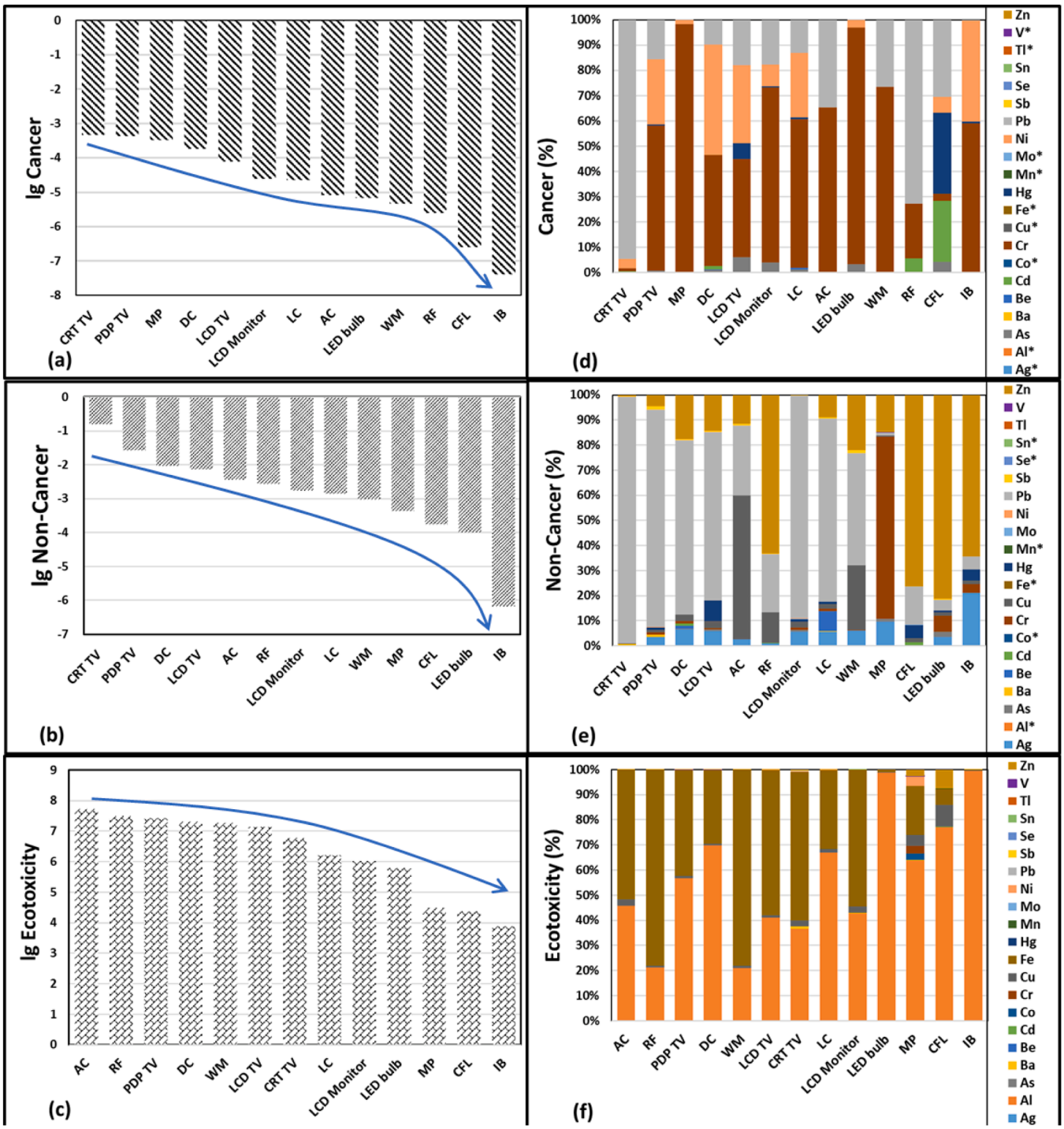

Fig. 1. USEtox™ Results of WEEE toxicity (a, cancer; b, non-cancer; c, ecotoxicity) and proportions of individual metal to the total (d, cancer; e, non-cancer; f, ecotoxicity).

concentrations of As is linked to carcinogenicity potential, and Be, Hg and Ag are associated with the potential for causing or promoting non-cancer diseases; whereas Cu and Zn concentrations are primarily associated with ecotoxicity potential.

Similar WEEE categories exhibit similar toxicity potentials. For example, the computers DC and LC show similarities in the constituent of chemicals associated with carcinogenesis (Cr, highest, followed by Ni and then Pb) and with causation or promotion of non-cancer diseases (Pb highest, followed by Zn). In computer-tablet category of WEEE, Zn and Ag pose the highest potential for causing or promoting non-cancer diseases (Son et al., 2016) due to Pb (73 mg/kg) and Zn (2090 mg/kg). For the three kinds of light bulbs, Zn is the most important metal in terms of causing or promoting non-cancer diseases. In addition, the three bulbs show a considerably low level of chemicals associated with carcinogenesis, non-cancer diseases, and ecotoxicity impacts. The more technically advanced light bulbs, CFL and LED bulbs, show higher toxicity potentials than IB presumably because of the higher concentrations of various metals in CFL and LED bulbs (Lim et al., 2013).

We note that the main potential contributors to non-cancer diseases, carcinogenesis and ecotoxicity are the same for all 13 WEEE categories,

mainly due to the higher contents of metal with a higher CF. For example, the highest potential contributors to carcinogenesis and non-cancer diseases for CRT TV are both Pb, also the highest in terms of carcinogenicity and non-cancer diseases among the 13 WEEE. Similarly, MP also have the same chemical potentiator of cancers and non-cancer diseases, namely, Cr.

The concentration of Pb in CRT TV is associated with the highest potential cancer cases and non-cancer diseases, and the phasing of Pb out of technically more advanced TVs like PDP TVs, LCD TVs has led to a reduction in risk of potential damage to human health (Yao et al., 2018). According to the toxicity ranking, LCD TVs are preferred over PDP TVs due to much lower toxicity potential in cancer, non-cancer diseases and ecotoxicity. This is an evidence that technology innovation could significantly reduce the environmental risk. LCD TV now predominates the market not only because of the potentially lower risk to health and environmental quality, but also due to lighter weight and lower energy consumption (Lehmberg et al., 2019). Whereas, for MP, technological innovation did not change the impact of chromium as a toxicity contributor (Chen et al., 2018). Other researchers have shown that Ni posed the most significant impact on potential carcinogenicity rather than Cr when the sampled MPs excluded the batteries and plastic cover cases (Singh et al., 2019).

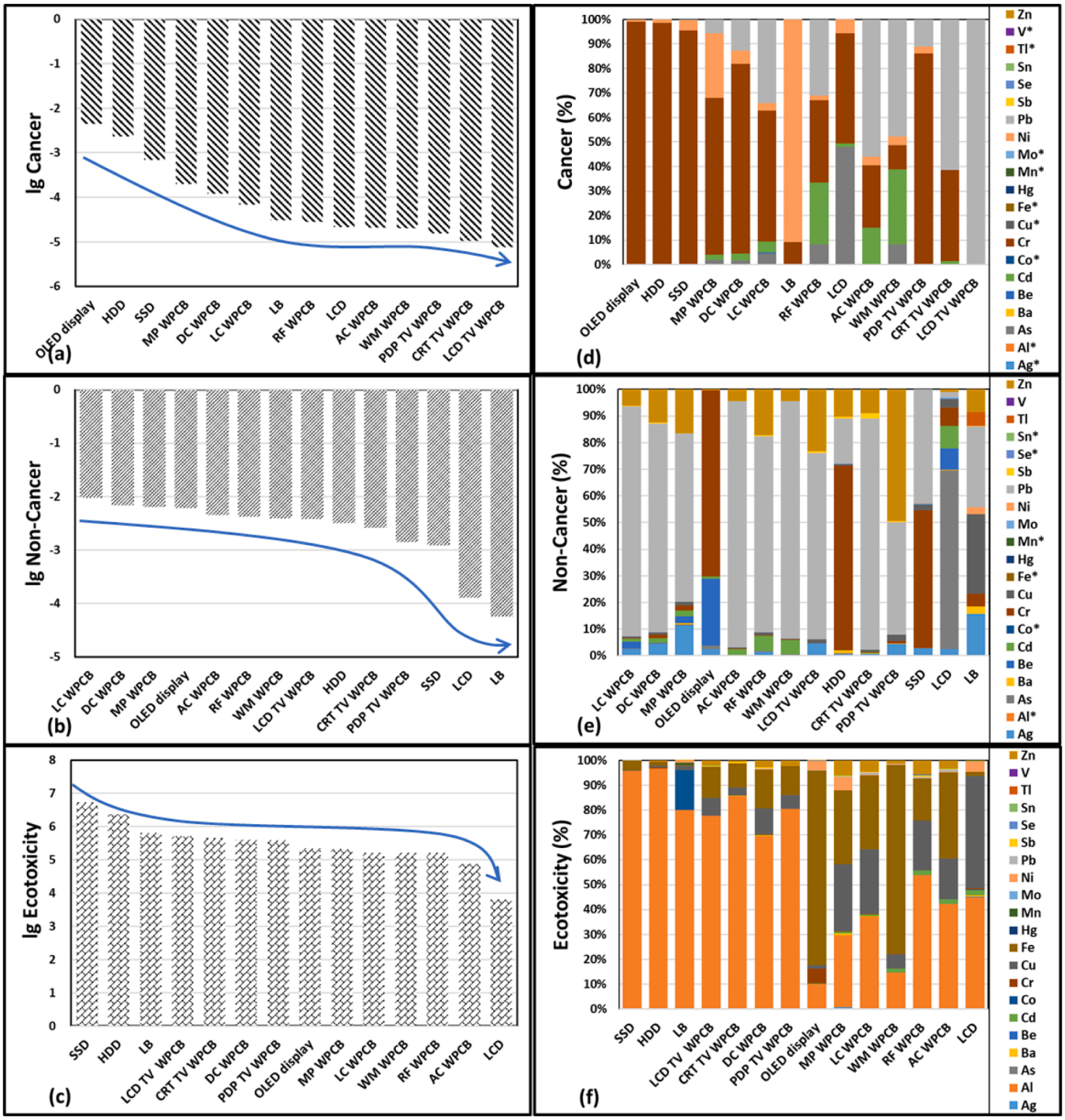

Fig. 2. USEtox™ Results of components toxicity (a, cancer; b, non-cancer; c, ecotoxicity) and proportions of individual metal to the total (d, cancer; e, non-cancer; f, ecotoxicity).

In Fig. 2, we present the USEtox™ modeling results for the relative toxicity contributions of electronic components (the toxicity values on logarithmic scale are shown on the y axis in panels a, b, c; and the proportion of a specific metals with respect to toxicity of all metals are shown in panels d, e, f. Additional data are presented in Table S8-S10 in SI. The carcinogenicity potentials range from 7.54×10^{-6} to 4.48×10^{-3} cancer cases in decreasing sequential order: OLED display, HDD, SSD, MP WPCB, DC WPCB, LC WPCB, LB, RF WPCB, LCD, AC WPCB, WM WPCB, PDP TV WPCB, CRT TV WPCB, and LCD TV WPCB. The decreasing order of potential to cause or promote non-cancer diseases, ranging from 5.69×10^{-5} to 9.47×10^{-3} cases is LC WPCB, DC WPCB, MP WPCB, OLED display, AC WPCB, RF WPCB, WM WPCB, LCD TV WPCB, HDD, CRT TV WPCB, PDP TV WPCB, SSD, LCD, LB. For potential ecotoxicity impacts ranging from 6.45×10^{3} to 5.27×10^{6} PAF•m^3•day, the decreasing sequential order of magnitude is SSD, HDD, LB, LCD TV WPCB, CRT TV WPCB, DC WPCB, PDP TV WPCB, OLED display, MP WPCB, LC WPCB, WM WPCB, RF WPCB, AC WPCB, LCD.

For potential carcinogenicity, As (47.88%) and Cr (44.84%) pose the most threat for LCD. Ni content is associated with up to 90.62% carcinogenicity potential for LB. Pb poses the most significant impact on potential carcinogenicity in WM WPCB (47.84%), AC WPCB (55.93%), CRT TV WPCB (61.41%), LCD TV WPCB (100%). Cr levels ranging from 53.25% to 98.83%. are associated with the highest carcinogenicity potential in OLED display, HDD, SSD, PDP TV WPCB, DC WPCB, MP WPCB, and LC WPCB. In RF WPCB, Cr (33.57%) and Pb (31.04%) together pose the most important impact on carcinogenicity potential. For non-cancer diseases, Pb (30.33%) and Cu (29.84%) together are associated with the most toxicity potential for LB. For LCD, As (67.06%) accounts for the highest non-cancer toxicity potential. Cr is associated with the potential to cause or promote non-cancer diseases in SSD (51.85%), HDD (69.15%), OLED display (69.75%). In AC WPCB, WM WPCB, CRT TV WPCB, LC WPCB, DC WPCB, RF WPCB, LCD TV WPCB, and MP WPCB. Pb is associated with the highest potential for causing or promoting non-cancer diseases, from 63.18% to 92.49%; whereas Zn (49.51%) and Pb (42.25%) together pose the most significant impact on PDP TV WPCB. For ecotoxicity, Fe is associated with the highest level of toxicity in OLED display (78.00%), WM WPCB (75.99%). For LCD, Cu (45.24%) and Al (44.67%) together hold the highest ratios on ecotoxicity. Al (29.02%), Cu (27.00%), Fe (29.54%) together pose the most significant role in MP WPCB. For the rest WEEE, Al is most associated with high ecotoxicity potential and the contribution ranged from 36.70% to 96.69%.

The ecotoxicity of SSD is higher than HDD, both caused by Al. However, SSD is nearly one order of magnitude lower than HDD in cancer and non-cancer toxicity potential. Meanwhile, SSDs are replacing HDDs because of higher speed, smaller sizes, more durability, and low power consumption (Bennato et al., 2019). This is another evidence for technology innovation that it shows a positive influence on reducing human health risk. The same as WEEE, one could also find that adverse effect of technology innovation on risk reduce, i.e., screens. Compared with LCD, OLED display, as a new type of screen, does not need backlights and it has better resolution (Lee et al., 2019), which show a higher carcinogenicity potential, non-cancer illnesses, and ecotoxicity potentials than LCD. OLEDs also presents the highest carcinogenicity potential among the investigated 14 components, about two orders of magnitude higher than that of LCD since the content of Cr in OLED is three orders of magnitude higher than that of LCD. For OLED display, Be should also be noted since it provides 25.11% non-cancer toxicity potentials, secondly to Cr (69.75%).

For the 9 WPCBs, though used in different WEEE, they exhibited the same toxicity characteristics: Al, Cu, Fe are the most influential metals, exhibiting the highest potentials for ecotoxicity. Regarding potential to cause or promote non-cancer diseases, Pb poses the most significant influence, except for PDP TV WPCB. For carcinogenicity potential, Cr or Pb is most influential. In 9 WPCBs, MP WPCB presented the highest carcinogenicity potential because of the highest concentrations of Cr. LC WPCB presented the highest potential for causing or promoting non-cancer diseases because of the highest concentrations of Pb. LCD TV WPCB presented the highest ecotoxicity potential because of the concentrations of Al.

The outcome of the overall assessments of WEEE and electronic components is that decreases in toxicity potentials can be achieved by manufacturing EEE and components with reduced concentrations of Pb, Cr, Ni, Al, Fe. In particularly, Pb, Cr and Ni are associated with the highest levels of potential carcinogenicity; while Pb and Cr are associated with the causation or promotion of non-cancer diseases; and Al and Fe are the most influential in driving potentials for ecotoxicity.

3.2. Influence of technical innovation and regulatory policies

3.2.1. Metal and organic content of WMPs and WPCBs

Data on the metal contents in WMPs and WPCBs from 2002 to 2010 are shown in Table S3. For WMPs, Fe is the most abundant metal (20 817 to 46 844 mg/kg, average of 34 736 mg/kg), representing ~ 21% of the total metallic content. Next in sequential decreasing order are Cu (21 899–33 743 mg/kg), Al (18 591–34 644 mg/kg), Cr (4955–41 820 mg/kg), Ni (3219–28 979 mg/kg) and Zn (184–54 557 mg/kg), representing average values respectively of 27 678, 26 495, 21 191, 16 607, 15 587 mg/kg; and 17%, 16%, 13%, 10% and 9% of the metallic content. Mg (1302–15 028 mg/kg, average of 7170 mg/kg), Co (311–29 461 mg/kg, average of 7098 mg/kg), Sn (2472–6995 mg/kg, average of 4640 mg/kg), Ba (957–4059 mg/kg, average of 2035 mg/kg) represent 1%−4% of its total metallic content. While Ag, Sb, Pb, V, Mo, As, Pd, Tl, Se, Au are at very low concentrations, together approximately 1% of the total metal content. Be and Cd were not detected in WMPs.

Some metals contained in WMPs are highly influenced by technical innovations. Before 2007, WMPs, including slide phones, rotating phones, bar phones, clamshell phones, all contained keyboards (Table S1 in SI). In 2007, full screen touched phones were developed without keyboards (Chen et al., 2018). After that, MPs were totally changed by this new technology. More metals were integrated into electronic devices during this period, leading to the observed increasing trend. After 2007, metal concentrations in electronic devices may have reduced because of miniaturization, cost control and implementation of design for environment initiatives. For example, Zn content is at 6125 mg/kg in 2004, which increased to 15 793 mg/kg in 2005, 48 817 mg/kg in 2006, 54 557 mg/kg in 2007, and after 2007, but the level of Zn eventually returns to a lower level (728–9415 mg/kg). At the same time, single-function MPs evolved with more functions, leading to a sharp increase in battery capacity to support long-term operation. Therefore, Co concentrations increased considerably.

The concentrations of metals in electronic devices may also be influenced by regulatory policies or volatile economic costs. For example, Pb content declined as a result of regulatory policies such as RoHS (adopted in 2003, effective in 2006). At the time that RoHS became effective, Pb-free soldering technology was already being developed, and adoption of Pb-free solders increased rapidly, to be replaced by increases in tin, silver zinc, bismuth, and other soldering metals (Li et al., 2018). Sn-Zn solder is more likely to be used to replace Pb-contained solder rather than Sn-Ag solder because of the economic cost of Ag. This is why Ag concentrations show an overall declining trend during the period covered by this study. Other precious metals, such as Au and Pd, were also discovered to follow a downward trend in concentrations.

For WPCBs, Cu is the most abundant metal, ranging from 177 000 to 243 000 mg/kg with an average of 213 667 mg/kg, representing about 49% of the total metallic content. Fe (ranging from 14 200 to 149 000 mg/kg, average of 65 633 mg/kg), Al (ranging from 33 600 to 81 600 mg/kg, average of 55 111 mg/kg), and Sn (ranging from 32 300 to 61 500 mg/kg, average of 42 156 mg/kg) respectively accounted for 15%, 13%, 10% of the total metallic contents. Zn concentrations ranged from 18 600 to 54 500 mg/kg, with an average of 32 189 mg/kg, accounting

for 7%. Pb (185–26 000 mg/kg, average of 9944 mg/kg), Ni (3030–10 500 mg/kg, average of 4714 mg/kg), Sb (1360–8450 mg/kg, average of 4387 mg/kg), Cr (158–12 800 mg/kg, average of 3462 mg/kg) and Ba (240–4090 mg/kg, average of 2403 mg/kg) are at lower level, together representing about 6% of the total metal. Mg, Ag, Co, Au, V, Mo, Cd, As are at a very low level. Be, Pd, Se, Tl, Hg were not detected in any samples of WPCBs.

For WPCBs manufactured between 2002 and 2010, regulatory policies such as RoHS considerably influenced the concentrations of metals. The phasing out of Sn-Pb solders caused Pb concentrations to decrease precipitously from 26 000 in 2004 to 483 mg/kg in 2005. Meanwhile, Sn concentrations also decreased from 51 400 to 32 300 mg/kg. Due to adoption of Pb-free solders, Ag concentrations increased from 118 in 2004 to 503 mg/kg in 2005. In addition, from 2005 to 2006, the concentrations of Zn increase from 19 000 to 30 200 mg/kg. After that time, from 2006 to 2009, Pb is present at very low levels (185 to 525 mg/kg). The results also show the impact of RoHS on Sb concentrations, which increased from 4900 mg/kg in 2004 to 8450 mg/kg in 2005. We attribute this to RoHS limitations on the use of BFRs (Ayvaz and Görener, 2020) leading to the increase in the use of Sb_2O_3 for flame retardation (Chen et al., 2018). We also discovered possible examples of technical innovation driving the relative concentrations of metal in WPCBs. For example, Cu concentrations decreased from 240 000 to 179 000 mg/kg (2003–2005), and then increased from 177 000 to 243 000 mg/kg (2006–2007), possibly reflecting miniaturization at first, and then increasing device sizes as additional functions are packaged into each device. However, Fe (14 200–149 000 mg/kg), Cr (828–12 800 mg/kg), Ba (240–4090 mg/kg) show an overall increasing trend with small fluctuations. Au concentrations (12–85 mg/kg) showed a decreasing trend, possibly reflecting the combined influences of miniaturization and economic costs for the metal.

In SI Table S4, we present the concentration of hazardous organic chemicals tested in WMPs and WPCBs from 2002 to 2010, including tetrabromobisphenol A, polybrominated diphenyl ethers (monobromodiphenyl ether to decabromodiphenyl ether), polychlorinated biphenyls (monochlorobiphenyl to decachlorobiphenyl), polybrominated biphenyls (monobromobiphenyl to decabromobiphenyl) and polycyclic aromatic hydrocarbons. In Table S5, we present the distribution polybrominated diphenyl ethers, polybrominated biphenyls, and polycyclic aromatic hydrocarbons among WMPs and WPCBs. Only nine bromodiphenyl ether and decabromodiphenyl ether were detected in WMPs of 2002 and 2005, and total polybrominated diphenyl ethers were respectively 1470, 1685 μg/kg. Total polycyclic aromatic hydrocarbons in WMPs stabilized in the range of 184–272 μg/kg, except the year of 2002 (950 μg/kg), due mainly to naphthalene found at concentrations of ~ 860 μg/kg. For WMPs manufactured between 2003 and 2010, naphthalene and phenanthrene are the main polycyclic aromatic hydrocarbon compounds found, respectively at 70–154 μg/kg and 82–133 μg/kg. We also detected fluorene, fluoranthene, pyrene, benzo[*g,h,i*]perylene in WMPs. Tetrabromobisphenol A, polychlorinated biphenyls, polybrominated biphenyls could not be detected in any WMPs. Total polybrominated diphenyl ethers in WPCBs are in a range of 750–99 260 μg/kg, with an average of 25 808 μg/kg, mainly decabromodiphenyl ether though hexabromodiphenyl ether to decabromodiphenyl ether could be found. Total polybrominated biphenyls in WPCBs ranges from 0 to 820 μg/kg, with an average of 273 μg/kg, and only nine-bromobiphenyl and decabromobiphenyl could be detected. Sixteen polycyclic aromatic hydrocarbons were detected in WPCB samples, mainly naphthalene or acenaphthylene and polycyclic aromatic hydrocarbons present at ~ 262–2000 μg/kg. We did not detect acenaphthene and dibenzo[*a,h*]anthracene. Tetrabromobisphenol A was detected at low levels in WPCBs manufactured in 2009 and 2010. Polychlorinated biphenyls were not detected in any WPCBs.

Regulatory policies may limit the concentrations of toxic organics chemicals such as polychlorinated biphenyls which are not detected both in WMPs or WPCBs because they were banned in the 1980s (Westin et al., 1981). RoHS limits the use of polybrominated biphenyls and polybrominated diphenyl ethers in homogeneous materials of EEE (Ayvaz and Görener, 2020). Polybrominated biphenyls were not detected in WMPs and they were in a low level in WPCBs, no more than the permitted levels. Pentabromodiphenyl ether, octabromodiphenyl ether, decabromodiphenyl ether are commonly used as BFRs in electronic devices (Earnshaw et al., 2013). Pentabromodiphenyl ether and octabromodiphenyl ether were banned in August 2004 (Siddiqi et al., 2003). Decabromodiphenyl ether was added to the list of exemptions of RoHS by the European Union, but was banned after July 2008 (Earnshaw et al., 2013). This may be the reason why Decabromodiphenyl ether was abundant especially in early period covered by this research, and the concentrations of polybrominated diphenyl ethers decreased from 82 110 μg/kg in 2005 to 1030 μg/kg in 2006. Higher concentrations of specific BFRs were identified in WPCBs and a wider variety of BFRs were found. This phenomenon may be attributed to safety considerations because consumers typically carry MPs, resulting in lower potential risk. Therefore, manufacturers use newer brominated flame retardants such as decabromodiphenylethane to lower the risk (Li et al., 2017). Polycyclic aromatic hydrocarbons in WPCBs and WMPs are similar with respect to BFRs in WPCBs and WMPs, although BFRs may reduce the generation of chemical radicals which could impact the generation of polycyclic aromatic hydrocarbon compounds (Moltó et al., 2011).

3.2.2. *Toxicity evolution of WMPs and WPCBs*

In Fig. 3, we present a summary of the of metallic toxicity potential in WPCBs and WMPs from 2002 to 2010 (the toxicity values on logarithmic scale on the y axis are shown in panels a, b, c; and the proportion of a specific metals respect to toxicity of all metals are shown in panels d, e, f). Additional data are presented in Table S11-S13 in SI. For carcinogenicity potential, both WPCBs (1.95×10^{-5}–6.78×10^{-4} cases) and WMPs (3.50×10^{-5}–7.08×10^{-4} cases) show an overall increasing trend from 2002 to 2010. The carcinogenicity potential of WPCBs in 2002 and 2003 is due mainly to Cr concentrations, respectively at 52.31% and 50.59%. After 2003, the contributions of carcinogenicity potential attributable to Cr concentrations increased to more than 90%. For WMPs, Cr also posed the most significant impact on carcinogenicity potential for WMPs, representing ~ 95.04% to 98.83%. The potential to cause or promote the incidence of non-cancer diseases associated with WPCBs declined from 2002 to 2010 (from 7.00×10^{-4} cases to 3.72×10^{-3} cases). The most significant contributor to the potential toxicity changed from Pb to Zn (increasing from 17.73% to 73.43%). While for WMPs, the potential to cause or promote non-cancer diseases increased (from 4.52×10^{-5} cases to 7.87×10^{-4} cases), mainly due to the concentrations of Cr and Zn. The ecotoxicity potentials of WMPs (8.81×10^{3}–3.61×10^{4} PAF•m^3•day) and WPCBs (1.74×10^{5}–3.72×10^{5} PAF•m^3•day) seemed stable and Al posed the most important influence for both WPCBs (53.18%−79.85%) and WMPs (51.39%−79.91%), followed by Fe and Cu.

In Fig. 4, we present the summary of the evolution of toxicity potentials for organic chemicals in WPCBs and WMPs from 2002 to 2010 (only 16 polycyclic aromatic hydrocarbons, decabromodiphenyl ether and polychlorinated biphenyls because of the database limitation of USEtox™). The toxicity values on logarithmic scale on the y axis are shown in panels a, b, c; and the proportion of a specific metals relative to the toxicity of all metals are shown in panels d, e, f. Additional data are presented in Table S14 - S16 in SI. For carcinogenicity potential of organic compounds, WPCBs are relatively stable during the period investigated (1.47×10^{-10}–7.40×10^{-9} cases). The concentrations of acenaphthylene posed the most significant impact on potential carcinogenicity in WPCBs of 2002, 2004 and 2006, while for the other years, benzo[*a*]pyrene presented the most significant risk of carcinogenicity. The carcinogenicity potential attributable to organic compounds in WMPs between 2002 and 2010 ranged from 4.86×10^{-12} to 4.61×10^{-11} cases. Naphthalene, phenanthrene, benzo[*g,h,i*]perylene] posed the most significant impacts. For the potential to cause or promote non-

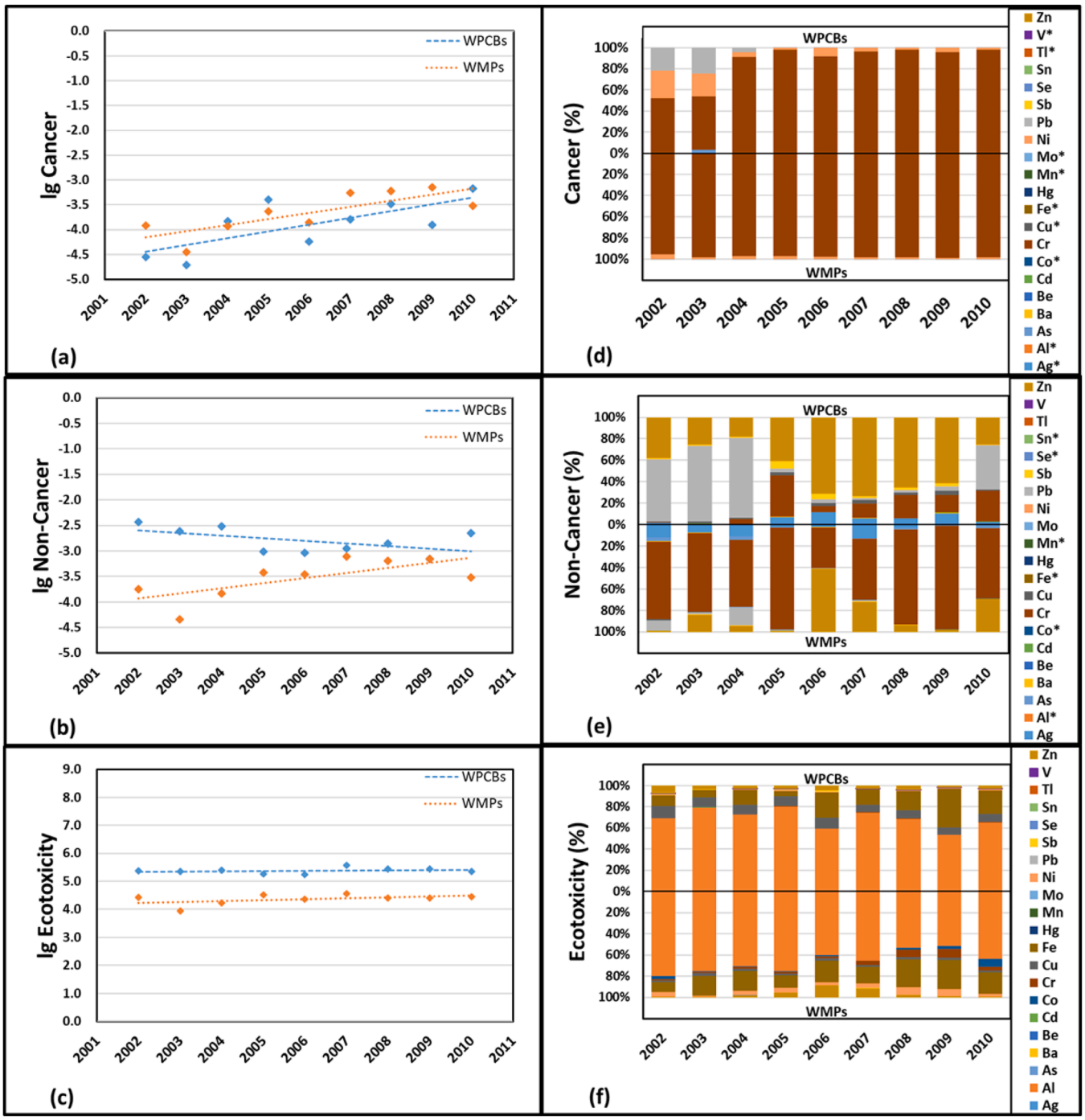

Fig. 3. USEtox™ Results of toxicity trend caused by metals in WMPs and WPCBs from 2002 to 2010 (a, cancer; b, non-cancer; c, ecotoxicity) and proportions of individual metal to the total (d, cancer; e, non-cancer; f, ecotoxicity).

cancer diseases associated with exposure to organic chemicals, WPCBs (1.58×10^{-10} –2.34×10^{-8} cases) and WMPs (1.21×10^{-12}–1.51×10^{-10} cases) both show a decreasing trend. For WPCBs, decabromodiphenyl ether exhibited the highest potential to cause or promote non-cancer diseases and it is the same for WMPs in the year of 2002 and 2005. For the other WMPs, naphthalene concentrations were associated with the highest potential to cause or promote non-cancer diseases. For potential ecotoxicity, WPCBs (1.09×10^{-4} to 7.09×10^{-2} PAF•m^3•day) and WMPs (1.32×10^{-4} to 3.85×10^{-3} PAF•m^3•day) both show an upward trend, with naphthalene concentrations being the most associated with ecotoxicity risk in 2002 and 2004 whereas pyrene was the most important for all the other years of device manufacture, except 2006.

4. Conclusions

This research provides comprehensive new perspective on the relative toxicity of various categories of WEEE and electronic components with emphasis on the influence of technical innovation and regulatory policies designed to attract consumers while protecting human health and environmental quality. Differences across WEEE product categories could account for 4–6 orders of magnitude toxicity potential differences, and 2–3 orders of magnitude in toxicity associated with various electronic components. These differences are mainly associated with

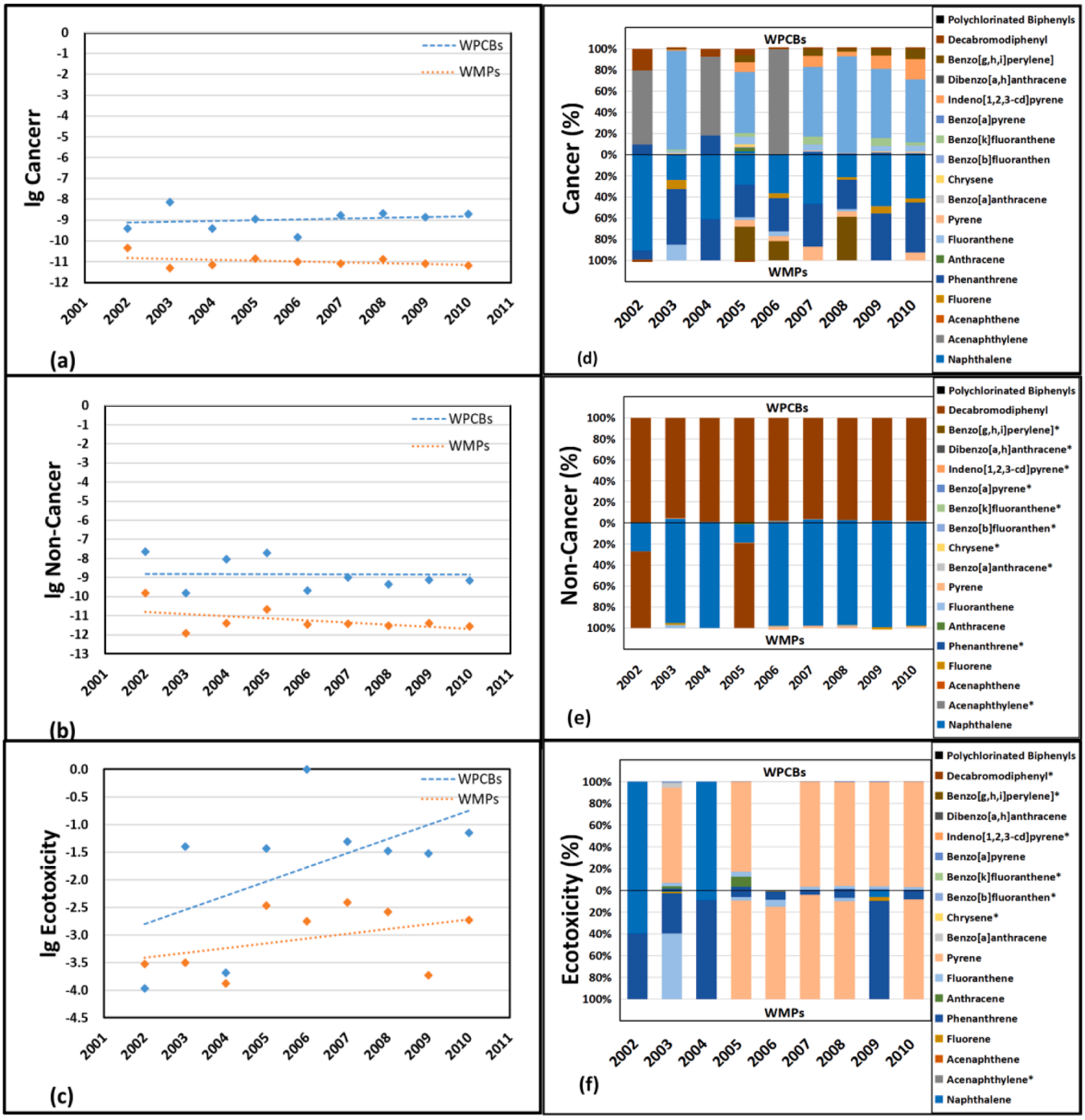

Fig. 4. USEtox™ Results of toxicity trend by organics in WMPs and WPCBs from 2002 to 2010 (a, cancer; b, non-cancer; c, ecotoxicity) and proportions of individual organics to the total (d, cancer; e, non-cancer; f, ecotoxicity).

concentrations of Cr, Pb, Ni, Zn, Hg, Cd, Cu, Al, Fe in WEEE; and of Cr, Ni, As, Pb, Cu, Al, Fe in electronic components. Technological innovation and regulatory policies show both coincidental and discordant but selective effects on the trends of toxic materials in WMPs and WPCBs. Some of the effects may be regional in impact, depending on which electronic devices and components consumers purchase, use and discard. Other effects are global where the innovations and regulatory policies are global. For example, the phaseout of Pb from solders used in manufacturing electronic products. Technical innovation introduces new materials for which there are uncertainties about toxicity, for example, nanomaterials, and innovations in energy storage batteries and flame retardance.

Author statement

S.C. performed the experiments, analyzed the data and wrote the manuscript. R.W. contributed to the statistical analysis. J.W. analyzed the data and revised the manuscript. M.C. designed the experiments, analyzed the data and revised the manuscript. J.S. revised the manuscript. Oladele Ogunseitan serves as co-director. All the authors discussed the results and commented on the manuscript.

Declaration of Competing Interest

We declare that we do not have any commercial or associative

interest that represents a conflict of interest in connection with the work submitted.

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Supplementary materials

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