

Classification of plastic waste originated from waste electric and electronic equipment based on the concentration of antimony

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Classification of plastic waste originated from waste electric and electronic equipment based on the concentration of antimony

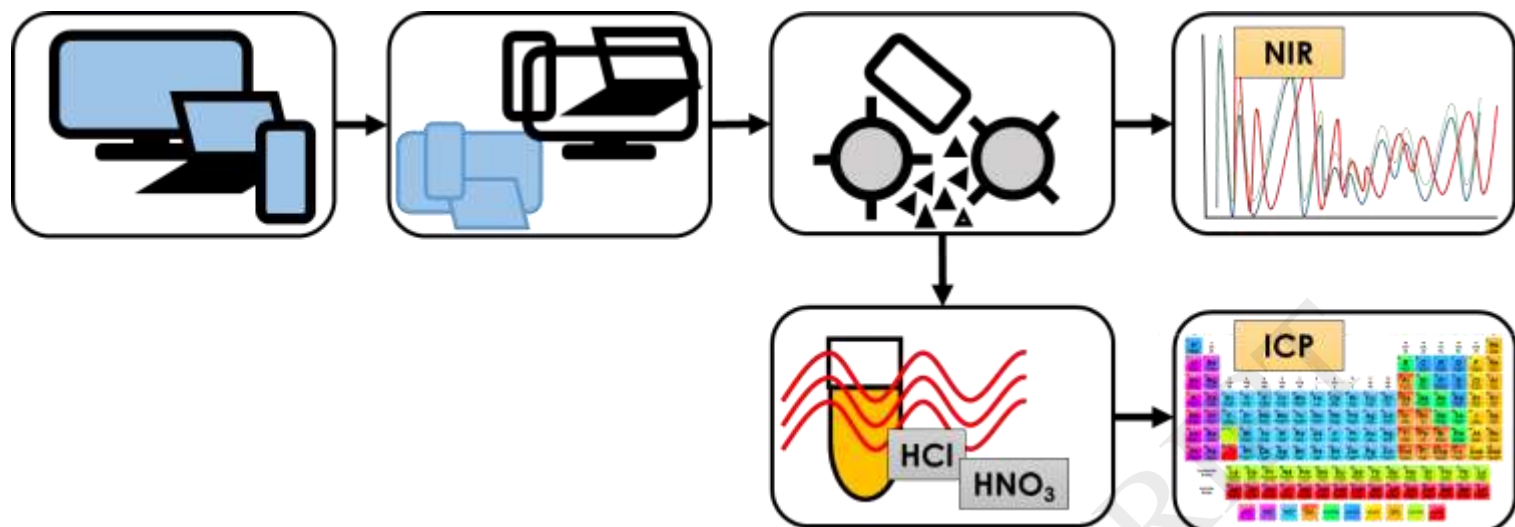
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Graphical Abstract



Highlights

- A screening of antimony in plastic components of different e-waste is presented
- Specific polymers were identified within each plastic fraction
- Antimony was analyzed through acid digestion and leaching tests
- Higher amounts of antimony were found in e-waste with less abundant plastic fractions
- Only limited fractions of Sb content are likely to be released in the environment

Abstract

The aim of this research is a preliminary assessment of antimony concentration in plastic fractions deriving from different e-waste. We considered microwave ovens, desktop computers, laptops, mobile phones, a TV case, a PC monitor and LED lamps (63 items in total). The plastic fraction ranged from 8%-wt in computers and microwave ovens, up to 40%-wt in cell phones and 59%-wt in LED lamps. Specific polymers were identified through Near Infrared spectroscopy. The samples followed three parallel procedures: acid digestion

with aqua regia; conversion into ashes at 600 °C then acid digestion with aqua regia; leaching according to UNI10802 reference procedure. Plastic components with significant amounts of antimony were the ones derived from desktop computers (25-1900 mg/kg) and from microwave ovens (830 mg/kg), yet their relative amount compared to the total weight of the item was limited. Items with larger plastic fractions showed lower concentrations of antimony (1-6 mg/kg in mobile phones cases and 160-640 mg/kg in plastic components of LED lamps). Leaching tests revealed that the analyzed plastic fractions could be mostly admitted in non-hazardous waste landfills. The analysis of ashed samples highlighted the need to further improve the acidic extraction procedure.

Keywords: antimony; e-waste; hazardous; plastic; waste

1. Introduction

Significant amounts of valuable resources are landfilled or incinerated each year, resulting in substantial economic and environmental losses [1]. Due to the growing generation of waste from electric and electronic equipment (WEEE), and as a potential consequence of the predicted rise in environmental contamination by metals mobility, WEEE has been identified by the European Union (EU) as a priority waste stream [2]. The plastic fraction of WEEE contains inorganic hazardous compounds originating from fillers, coloring agents, electrical parts and other components, including base and rare metals and heavy metals [3–6].

Antimony (Sb) is used in plastics, coatings, textiles and electronics as a synergetic flame retardant (in the form of antimony oxide [6]) with halogenated flame retardants, which results in reducing the consumption of halogenated flame retardants [7,8]. In addition, Sb or Sb-compounds are commonly applied in lead - acid batteries, to confer better physical and

chemical resistance to lead; as a decolorizing agent in glass or catalyst in PET production [8]; as color stabilizer in paints (i.e., yellow striping in road pavements) and as lubricant in automotive brakes [9]; in pesticides, ammunition and medicines [10,11]. At present, Sb consumption as a catalyst, flame retardant and heat stabilizer accounts for about 50% of its total production [12,13].

Sb and its compounds were classified by the Directive 67/548/EC as hazardous substances and they were assigned the following risk and safety phrases: Harmful (Xn), environmentally dangerous (N), harmful and poisonous dust and poisonous by skin contact (R51-53) [14]. Attention is being highly paid to Sb due to its toxicity, especially in regions where mining and smelting activities take place [15]. The Agency for Toxic Substances and Disease Registry (ATSDR) reported the consequences of Sb exposure on animals and humans [16]. In a study performed on female rats, it was found that the inhalation of Sb compounds can increase the occurrence of lung tumors [17]. Furthermore, increased menstruation disturbances and spontaneous abortions were reported by women who worked in Sb metallurgical plants [18]. The International Agency for Research on Cancer (IARC) classified antimony trioxide (Sb_2O_3) as carcinogenic for animals [19]. Other studies suggested that Sb—in combination with other compounds—alters cellular defense mechanisms and is therefore carcinogenic [20]. Exposure to Sb can happen by ingestion (water and food), inhalation (air and urban dust) or dermal contact [16], yet drinking water was classified as the most common route [21], and its toxicity is solely dependent on its chemical form [22]. Accordingly, wastes containing Sb were categorized as hazardous under the Basel convention in 1989 [23].

Due to the regulations regarding Sb limits in the environment (i.e. the EU drinking water limit is 5 $\mu\text{g/l}$ [24]), as well as to being included in the EU directive on the subject of acceptance in landfills [25], stricter regulations were put in place. Following a standard batch test (NS-EN 12457), leachate levels of Sb above 0.7 mg/kg requires disposal in a hazardous waste

landfill, while 5 mg/kg Sb concentration is the upper allowable limit for hazardous waste [26]. From an environmental perspective, Sb is a contaminant of concern, due to its potential mobilization from polymeric waste items that are disposed of in landfills, incinerated or that are recycled [27]. Generally, the plastic fraction of WEEE contains a large number of additives and flame retardants [3,28]; however, to our knowledge a systematic quantitative assessment on the occurrence of Sb in WEEE seems limited to few measurements [27]. As was reported by Tostar et. al., 2013 [25], up to 22,700 mg/kg of Sb could be leached from acrylonitrile butadiene styrene (ABS) in discarded computer housing using heated sodium hydrogen tartrate in dimethyl sulfoxide. Similarly, high concentrations of Sb (2,000-18,000 mg/kg) were detected in WEEE polymeric fraction [28]. Accordingly, for the aim of a proper material management, material composition analysis is highly needed to evaluate if the plastic fractions of WEEE are hazardous and eco-toxic [29].

The objective of this work is a preliminary assessment of Sb concentrations within different categories of plastics derived from WEEE, with two main aims: 1. quantifying plastic fractions' hazardousness on the grounds of antimony content; 2. support the identification of antimony-rich waste flows, considering that Sb is a critical raw material according to the EU [13] and that the development of recycling procedures is highly needed. The specific plastic fractions were identified through Near Infrared (NIR) spectroscopy and categorized following the threshold limit assigned by the California Department of Toxic Substance Control (TTLC) (i.e. 500 mg/kg) [3]. This research considered mobile phones (40 items), LED lamps (9 items), microwave ovens (5 items), desktop computers (5 items), laptops (2 items), a TV screen (1 item), and a computer monitor (1 item) for a total of 63 items dismantled and separated into their different plastic components. Our approach hypothesized different waste management perspectives, such as landfilling or treatment of plastic waste (recycling or incineration) [30]. The samples followed three parallel procedures: acid digestion with aqua

regia; conversion into ashes at 600 °C then acid digestion with aqua regia; leaching according to UNI10802 reference procedure.

2. Materials and methods

2.1. Samples origin and types

The samples analyzed in this study were obtained from a WEEE collection point in Hamburg (Germany), except for the LED lamps, which were new items. The research involved WEEE of different brands and only their plastic components were considered. In total, 63 WEEE were considered:

- 40 mobile phones, all put on the market between 2002 and 2010. Only the casings and displays were involved in the research;
- 5 desktop computers, manufactured between 2000 and 2008;
- 2 laptops, put on the market in 2005 and 2010. Only the external casing was involved in the research;
- 5 microwave ovens;
- 1 computer monitor produced in 2002;
- 1 TV screen, produced in 2005. Only the external casing was involved in the research;
- 9 LED lamps, different for technical aspects such as lumen, energetic class, power, shape and composition.

2.2. Plastic fraction identification and preparation

The considered WEEE devices were manually dismantled and their plastic components were sorted by origin and type. The different fractions obtained were qualitatively analyzed by Near Infrared (NIR) spectroscopy using a microPHAZIR™ RX Analyzer. The identified polymers were: Acrylonitrile butadiene styrene (ABS), High Impact Polystyrene (HIPS),

Polybutylene terephthalate (PBT), Polycarbonate (PC), polymers with fiberglass (FG), Polymethylmethacrylate (PMMA), Polyoxymethylene (POM), Polypropylene (PP). Weighing the different fractions separately allowed a material balance assessment. A Sartorius balance (1.0 mg precision) was used for the small components, such as the parts of the LED bulbs. A bench platform scale SFB (1.0-2.0 g precision) was used for the mobile phones, while a platform scale Pfister Waagen GmbH (20.0-50.0 g precision) was used for the heavy parts of computers and microwave ovens. Each of the identified polymers was individually shredded, first using a universal shredding machine (JBF shredding systems), followed by a second shredding stage using a Retsch Cutting Mill SM 300. The samples were shredded to a particle size ranging between 2.0 and 4.0 mm.

2.3. Microwave-assisted aqua regia digestion

Shredded samples underwent acid digestion following the DIN EN 16174 standard. Duplicate samples (0.5-1.0 g) of each polymer were digested in 10 ml aqua regia (37% HCl and 65% HNO₃ in 3:1 ratio) in a CEM Mars 6 microwave oven. The acid digestion happened as follows: heating to 175°C with a ramp time of 15 minutes, 175 °C kept constant for 15 minutes and pressure increased at 800 psi, cooling for 30 minutes. The acid solution obtained from the digestion was vacuum filtered through glass fiber filters with a pore size of 1.2 µm. The filtrate was afterwards diluted with distilled water to a final volume of 0.1 L.

2.4. Samples ashing

Ashing involved plastics derived from the following samples: mobile phone casings, laptop casings, a TV screen casing, and a computer monitor. The ISO standard 3451-1, procedure A [31] allowed to analyze the ash content. The samples were weighed and placed inside porcelain crucibles (half filled), pre-burned with a butane Bunsen burner and finally

converted into ashes in a muffle at 600 °C. Ashing proceeded until a constant mass was obtained (total time below 3 h). The ash samples were afterwards digested as explained in section 2.3.

2.5. Antimony analysis

The analysis of Sb concentration in the filtered acidic digested solution was performed by an Agilent 5100 ICP OES spectrometer. The reading time was set at 5 seconds with a radial view mode. The nebulizer flow was set at 0.7 L/min and the plasma flow at 12 L/min, with a pump speed of 12 rpm. The radio frequency power applied was 1.2 kW. The analysis was done in triplicate.

2.6. Leaching Tests

The leaching tests involved 9 samples of different plastic materials: two from the mobile phones (display and plastic cases), four from desktop computers and three from microwave ovens. The tests followed UNI 10802 reference procedure [32]: 90 g samples (particle size below 4 mm) were inserted in a glass bottle with deionized water with a liquid/solid ratio equal to 10 and shaken in a Heidolph Reax 20 shaker for 24.0 h \pm 0.5 h at room temperature. Afterwards, the eluate was filtered by a 0.45 μ m filter. Metals (As, Cu, Ni, Zn) concentrations in the eluate were analyzed as explained in section 2.5. Chemical Oxygen Demand (COD) and chloride and sulfate concentrations were analyzed according to reference procedures [33].

3. Results and discussion

3.1. Mobile phones

The external plastic case of the considered mobile phones showed the largest share of the

total weight of the plastic fraction (~ 70%), which represented ~ 40% of total unit weight. This fraction was mostly made of ABS, PC and a mix of both. The concentration of Sb in mobile phone covers was negligible (0.79 ± 0.94 mg/kg, see Figure 1). All samples deriving from mobile phones showed negligible antimony concentration values.

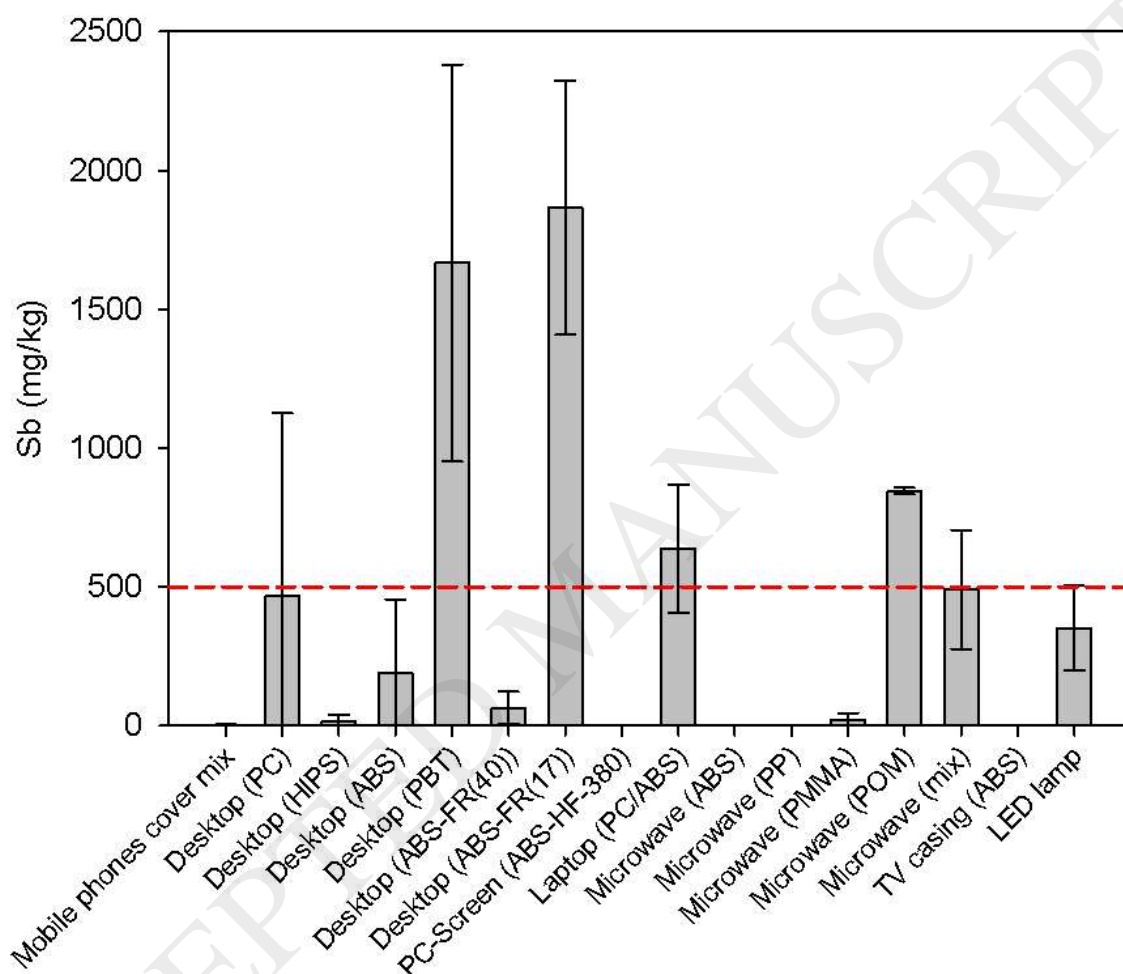


Figure 1. Concentration of Sb (mg/kg) in the considered samples (the dotted line represents the TTLC value)

3.2. Personal Computers

The plastic fraction in the desktop computer represented 4.5 - 12.0% of the total weight of

each unit. Over 50.0% of the plastic fraction was identified as ABS and HIPS (see Figure 2). These polymers were mostly used for the external cases, yet they were also found in the internal structural parts. PBT, found in internal fans, represented the third largest fraction (~ 8.0%-wt). Blends of different polymers were also detected: ABS+PBT, polymers with fiberglass (e.g., PC + 20.0% FG), and polymers with added flame retardants (e.g., ABS + Halogen-free organic phosphorus compounds flame retardant, ABS-FR(40)).

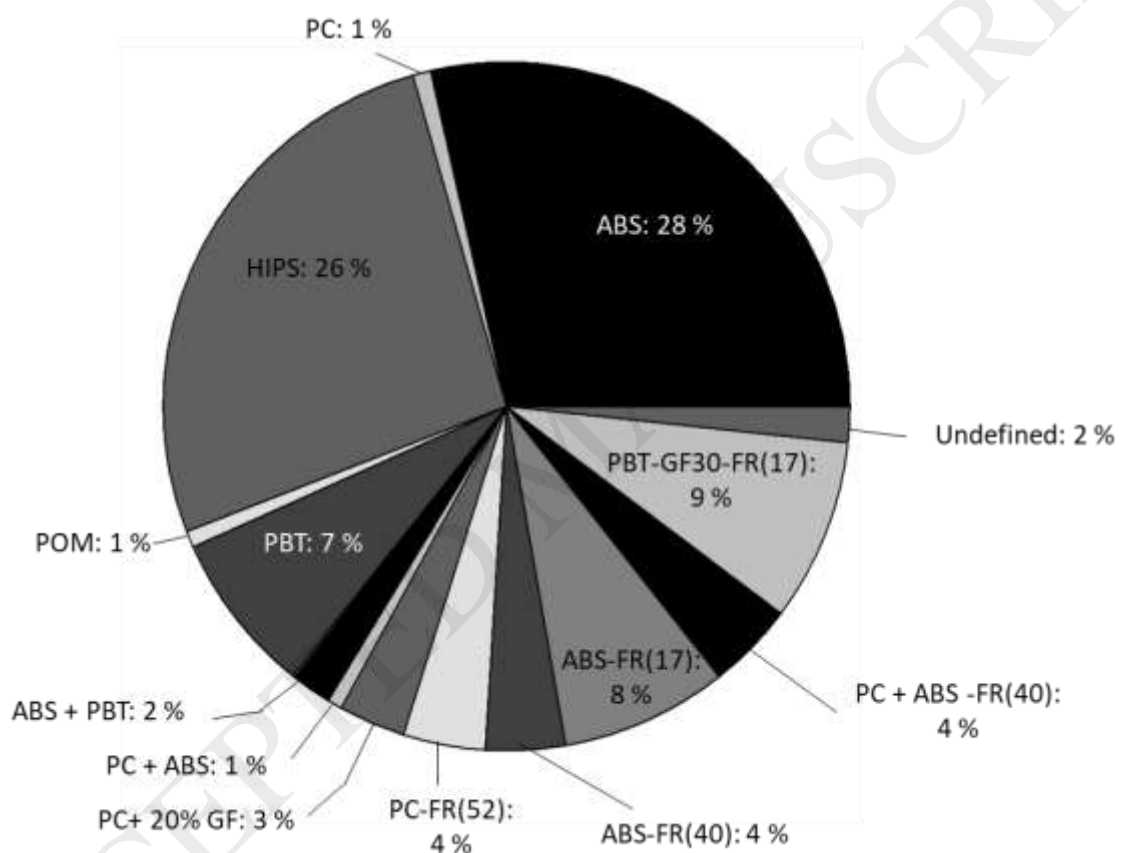


Figure 2. Distribution of the plastic fraction in the analyzed desktop computer units

Only three types of polymers showed concentrations of Sb equal to or exceeding the TTLC limit (see Figure 1): ABS-FR(17) (1865.9 ± 458.0 mg/kg), PBT (1666.8 ± 715.3 mg/kg) and PC (466.7 ± 660.0 mg/kg). In fact, according to the marking standard of Hewlett Packard

plastics © (i.e. the brand of one of the samples), the code FR(17) indicates the application of aromatic brominated compounds in combination with Sb (as flame retardants) [34]. For the other samples, excluding ABS halogen-free organic phosphorus compounds flame retardant (ABS-FR(40)), the type of flame retardants used was not indicated by the producer.

3.3. Laptops

Plastic fractions of the external casings of laptops were a mixture of PC+ABS and ABS, which corresponded to 15% of the total unit weight. Laptop housing is generally built from steel, aluminum, magnesium and plastics (ABS+PC) [35]. Sb concentration values exceeded the TTLC limit reaching 636.5 ± 230.7 mg/kg (see Figure 1). However, the number of the analyzed units was limited to two, hence further analyses are needed to draw conclusions about the materials' hazardousness.

3.4. Microwave ovens

Plastic fraction in the analyzed microwave ovens was equal to about 6.0%-wt; the dominant components were metals. ABS represented the main polymer (over 55.0%-wt), where its main application was observed in the external casings, in frames, and in the operation buttons. PP (around 26.0%-wt) occurred in internal support parts, such as the ventilation system. POM (5.0%-wt) and PMMA (4.0%-wt) were respectively used for the fans of the ventilation systems and for the transparent screen on the front door. Sb was absent in ABS and PP components. Trivial amounts were detected in PMMA (19.2 ± 27.1 mg/kg), yet both POM and the mixed fraction showed significant concentrations (844.7 ± 11.4 mg/kg and 489.9 ± 214.6 mg/kg, respectively).

3.5. TV case and Computer monitor

The TV case was made of 100% -wt ABS, while the computer monitor was made of 100%-wt ABS-HF-380. In both samples Sb amount was negligible, indicating either the application of Sb - free flame retardants or the complete absence of flame retardants [28].

3.6. LED lamps

The plastic fraction in LED lamps was dominant (~ 59.0%-wt) (see Figure 3). The cases were made of plastic (instead of glass) in eight out of nine examined bulbs.

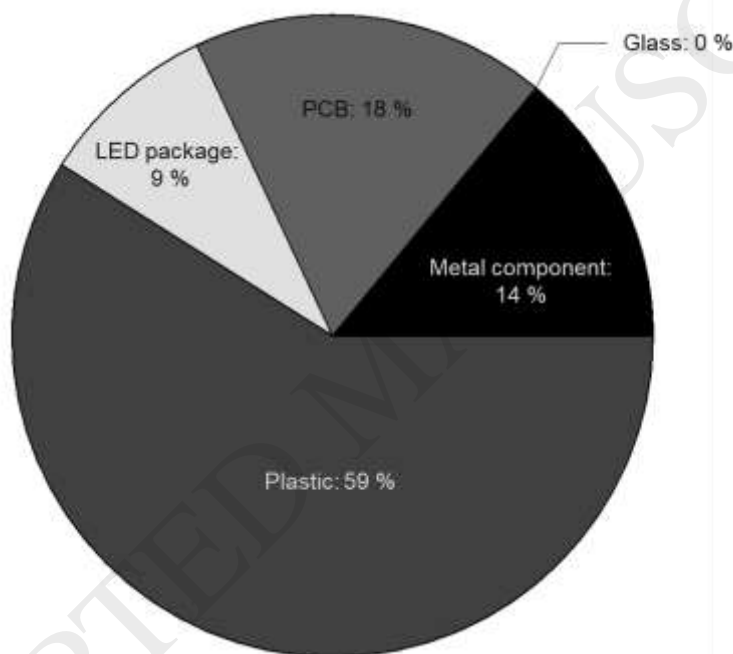


Figure 3. Component analysis of the external lid of the considered LED lamps

In order to understand the distribution in LED lamps, the Sb content of different fractions was analyzed and compared to its content in the plastic fraction. Results (see Figure 4), show that the plastic fraction has the highest concentrations of Sb, followed by circuit boards. A deviation to this was observed in lamp no. 8 (see Figure 4). This is due to the lack of a plastic casings in this specific lamp. The plastic fraction generally showed higher

amounts of Sb (163.0-640.0 mg/kg). LED lamp no. 9 exhibited the highest Sb concentration: over 600.0 mg/kg in the plastic part and more than 500.0 mg/kg in the circuit board. Overall, the plastic fraction of LED lamps contained Sb concentrations exceeding the TTLC limit, which indicates the necessity of special treatment procedures for this kind of WEEE.

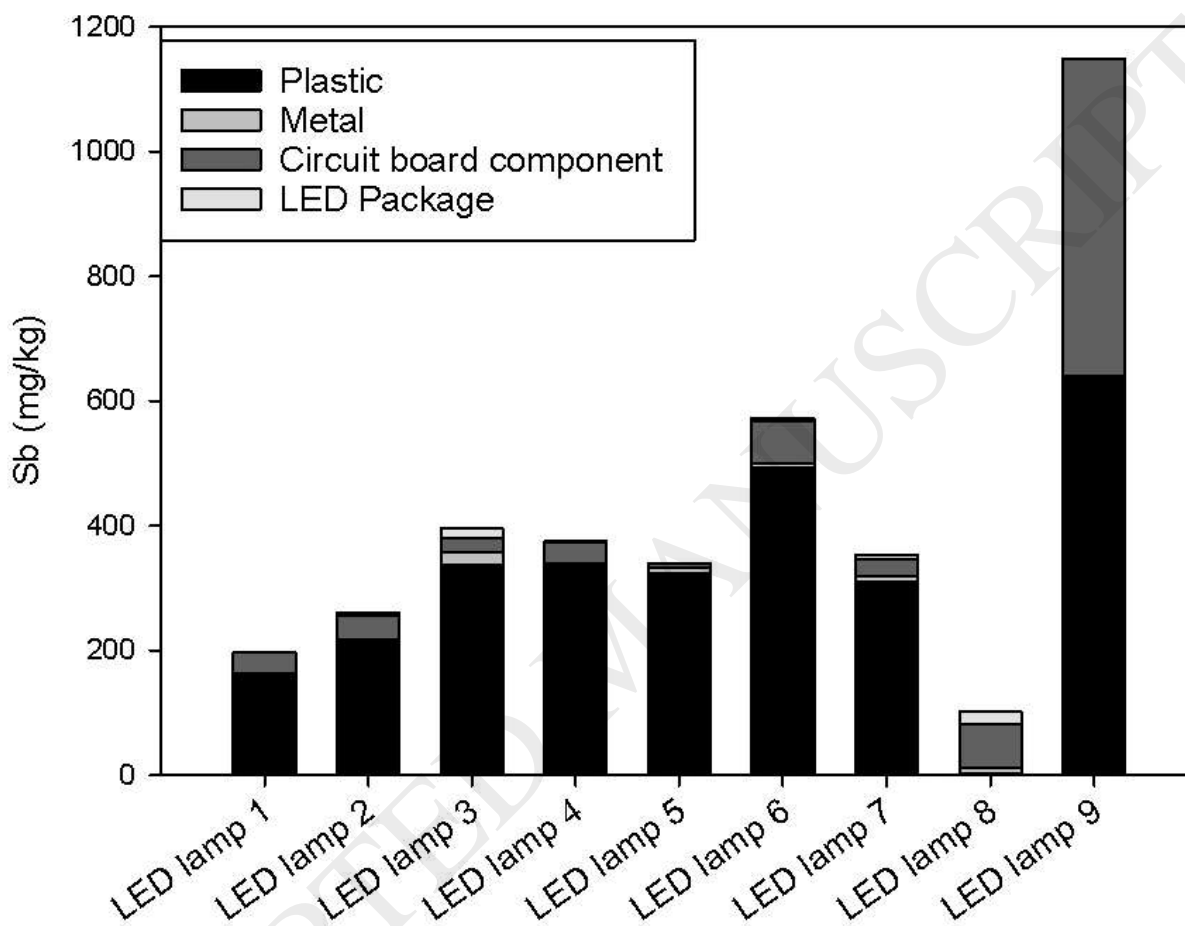


Figure 4. Antimony distribution in different fractions of the considered LED lamps

3.7. Ashed samples

The existing waste management systems in Europe are various, yet there is an increased interest in recycling activities, either through material recovery or energy recovery. In this study plastic samples were ashed to simulate waste incineration and to quantify Sb in the bottom ashes.

The ash content for the tested samples varied, depending on the different inorganic additives in each material: 1.4%-wt in the computer monitor, 0.5%-wt in the TV screen, 4.6%-wt in mobile phone covers and 4.5%-wt in laptop covers. When antimony concentration was back calculated, mobile phone covers had 4.8 ± 0.5 mg/kg, which is slightly higher than what was obtained by acid digestion. This indicates that the digestion was not completely efficient in disrupting the mobile phones plastic fraction. The TV screen and the computer monitor showed concentrations similar to what was obtained by the acid digestion. The ash fraction of laptop casings returned ~ 40% of the Sb (when compared to the values obtained from acid digestion), indicating that 60% of Sb possibly gets lost in the fly ash fraction.

3.8. Leaching test

The choice to add leaching tests in this study was done considering the global situation of WEEE illegal trading. In fact, ~75% of WEEE in Europe is landfilled, incinerated or exported to other countries, that in many cases, do not have strict environmental standards or do not use best waste management technologies [36].

The pH values of the leachate ranged between 5.9 and 8.6. Following the criteria for hazardous waste acceptability in landfills for non-hazardous waste [37], and analyzing COD, chloride, sulphate, As, Cu, Ni, Zn and Sb contents, only two plastic samples were found not suitable to be landfilled in non-hazardous waste landfills (see Figure 5) because of COD and Sb concentration values. The plastics derived from mobile phone cases had COD values exceeding the limits (>800 mg/kg), which could be related to the presence of liquid crystals. However, regarding the metals' concentrations, PBT polymer from microwave ovens leached 5.6 mg Sb/kg (limit value is 0.7 mg Sb/kg material).

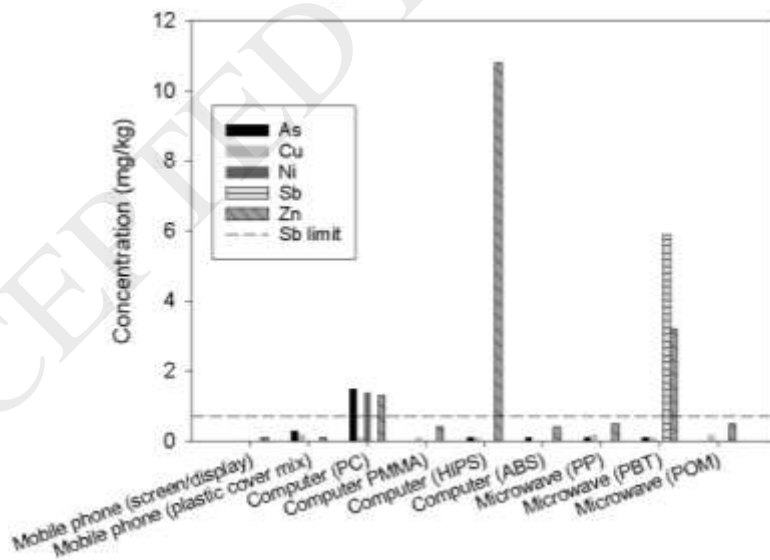
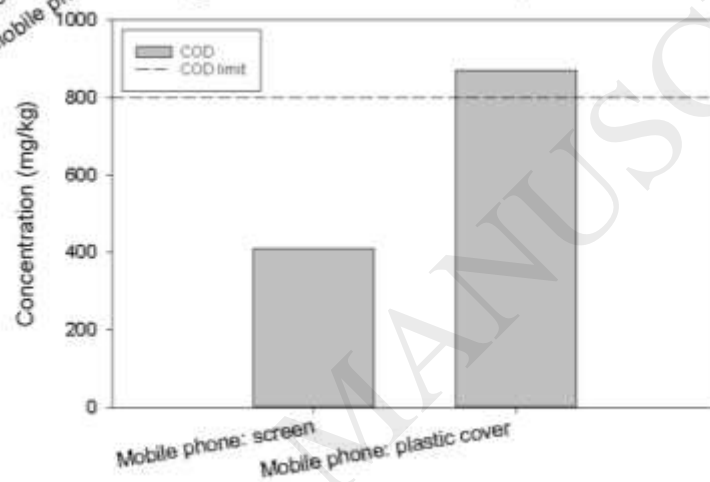
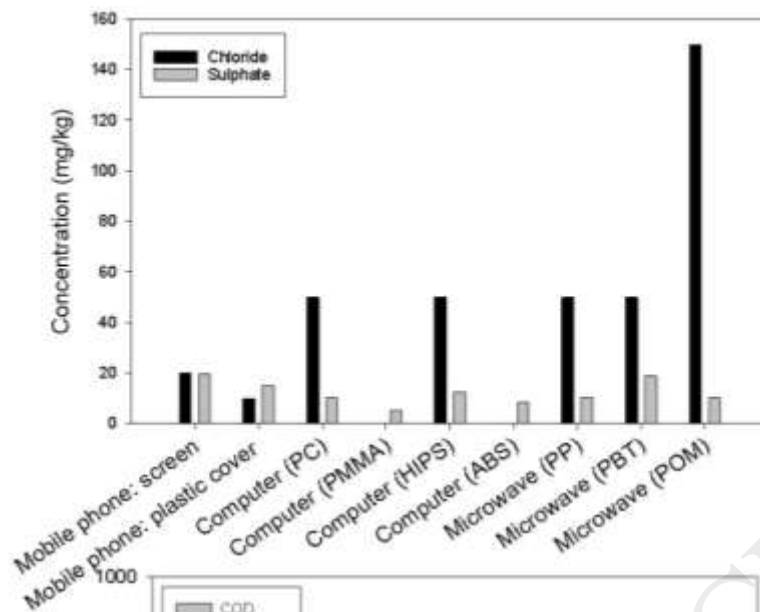


Figure 5. Results of the leaching test (dotted lines represent threshold limits for COD and Sb for acceptability in non-hazardous waste landfills)

3.9. Overall evaluation of Sb content

Figure 1 provides an overview of Sb concentrations analyzed in the different polymers derived from the considered WEEE units. Some of the analyzed resins—PC (from desktops), PBT (from desktops), ABS-FR(17) (from desktops), PC+ABS (from laptops), POM (from microwave ovens), mixed plastic (from microwave ovens) and plastics (from LED lamps) —displayed relevant concentrations of Sb, reaching or surpassing the TTLC threshold value; representing hazardousness. Sb was found in 80% of the analyzed samples, where only 20% indicated concentrations \geq TTLC. Including the standard deviation in the evaluation, up to 40% of the analyzed samples have shown Sb concentrations exceeding the abovementioned limit. Antimony distribution in the samples was analyzed twice, once in regard to the material source (i.e., electrical unit) and a second time in regard to the polymer type (see Figure 6). For both, the Normality Test (Shapiro-Wilk) and the Equal Variance Test (Brown-Forsythe) of Sb concentration according to the tested units failed ($P < 0.050$). Examining the statistical significance of differences in the mean values among the groups indicated that the differences were not great enough to exclude the possibility that the difference is due to random sampling variability; $F(6, 36) = 2.36$, ($P = 0.107$). On the other hand, the Sb concentration in regard to the polymer type does not pass the normality test (Shapiro-Wilk) ($P < 0.050$); yet the data passed the Equal Variance Test (Brown-Forsythe), ($P = 0.908$). The one-way analysis of variance showed that the polymer type had a significant impact on the Sb concentration ($F(8, 34) = 2.22$, $P = 0.029$), indicating that there is a tangible statistical relationship between polymer type and antimony concentration. A post hoc Tukey test, with critical q value at $p < 0.05$ was applied on the polymer type set of data. The null hypothesis was rejected for ABS versus PBT, PC versus PBT, PC/ABS versus PBT, HIPS versus PBT, PP versus PBT and PMMA versus PBT.

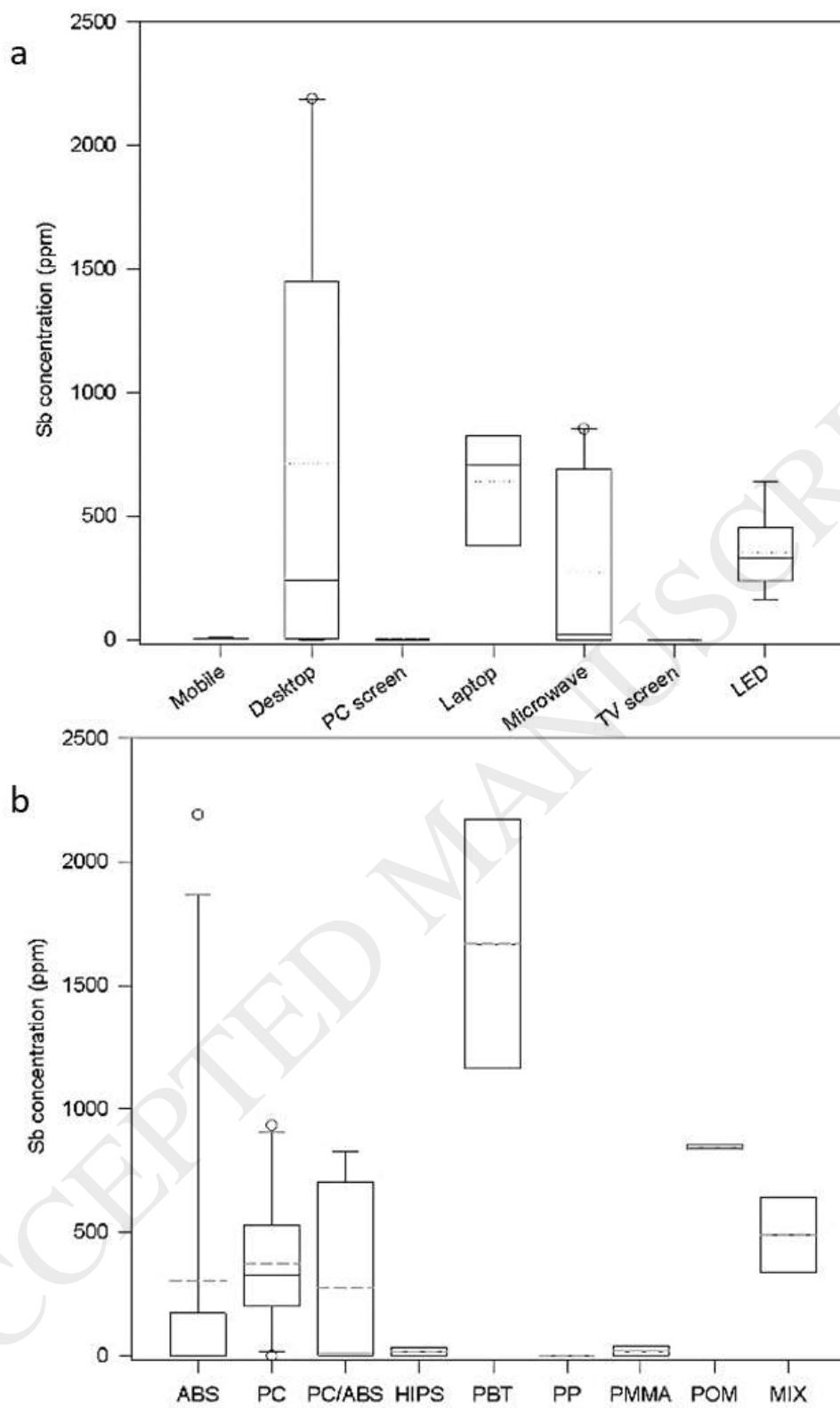


Figure 6. Description of antimony concentrations versus (a) material source and (b) polymer type

4. Conclusions

This research was focused on the investigation of antimony content in the plastic fractions of different WEEE categories. Firstly, the quality and relative abundance of the plastic fractions from different WEEE items (compared to the total weight of the item) were investigated; secondly, the amount of antimony was analyzed in each plastic waste material. The aim was to classify the plastic fractions as hazardous or non-hazardous waste, according to the TTLC value defined for antimony by the California Department of Toxic Substance Control. Antimony's hazardousness is related to its release into the environment, particularly in cases of landfilling or dumping. However, being that antimony is a critical raw material, its detection in specific waste flows could support the development of recycling procedures.

The research showed that some samples (derived from microwave ovens and desktop computers and laptop computers) contained antimony concentrations higher than the threshold limits. Specifically, it was observed that the concentration of Sb is mostly dependent on the type of polymer rather than on the WEEE item of origin. Accordingly, these plastic parts need to be treated separately before being landfilled, incinerated or sent to recycling facilities.

In contrast, the cases of mobile phones and the TV external casing showed no antimony, indicating that those parts may contain other types of flame-retardants. Considering only the plastic fraction of these units, results showed that desktops, laptops and LED lamps exhibited the highest possible antimony amount released. Although the plastic fraction of microwave ovens showed higher amounts of antimony, due to the small plastic portion, the total fraction to be possibly released by inappropriately disposed microwave ovens is low. The results of the leaching tests showed that plastic fractions of the considered WEEE items could mostly be admitted in non-hazardous waste landfills. The overall results of the study

need to be further validated through the optimization of the acid extraction phase and the analysis of more samples, however a preliminary assessment of antimony amounts in plastic fractions from different WEEE was provided.

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