Human and Environmental Impact Producted by E-waste Releases at Guiyu Region (China)

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Abstract

Over the last decades, the amount of electronic waste (e-waste) has increased rapidly in the world. It has become one of the emerging problems of the 21st century. About 50-80% of e-waste from industrialized countries is exported to recycling centers in developing countries such as China, India, Pakistan, and the Philippines because of the lower wages for labor and less strict environmental and safety regulations in these countries. However, China, due to its size and population not only receives enormous quantities of used devices from developed countries but also generates tremendous amounts of domestic e-waste due to its fast consumption rates of electrical and electronic (EE) products. Guiyu, a town in the Guangdong Province, southeast of China, was identified as the largest e-waste site in the world and the second most polluted spot, due to informal recycling processes (acid extraction for metals, open burning of wires to get copper), which release chemicals to the environment, representing a threat for human health, both for "recyclers" and nearby citizens, and the environment. Measured data on environmental concentrations and human health are scarce and scattered. Hence, environmental modeling was applied in order to generate an overview over the distribution of selected hazardous substances due to informal recycling in Guiyu. As all available models have a specific focus and various pros and cons, four models were chosen, which cover different geographical scales and address

different environmental compartment and objectives in order to assess the potential risk of the selected chemicals to humans and the environment.

These models have been applied to different scenarios, mainly for two chemicals, decabromobiphenyl ether (DeBDE) and lead (Pb). Emissions of DeBDE and Pb that represent the input to the models are based on the SFA (Substance Flow Analysis) developed for Guiyu presented in the chapter "Global E-waste flow with focus on China". In this chapter the results of the four models are presented and compared among them. The impact of the selected chemicals for the environment and human health at Guiyu region has been assessed on different scales, i.e., on a global, regional, and local scales.

1. Introduction

As already mentioned in the Chapter "Global E-waste flow with focus on China", China not only generates very high amounts of domestic e-waste due to their fast consumption rates of electrical and electronic (EE) products, but also receives enormous quantities of used devices from developed countries. In particular, Guiyu was shown to be the largest e-waste site in the world and the second most polluted spot, due to informal recycling processes (e.g. acid extraction for metals, open burning of wires to get copper) which release chemicals to the environment, representing a threat for human health, both for "recyclers" and nearby citizens (BAN and SVTC, 2002).

Therefore, having in mind the results of the SFA for the country of China and the Guiyu region, presented in the chapter "Global E-waste flow with focus on China", the main objective of the present chapter is to evaluate the distribution of two electronic device additives, lead (Pb) and decabrominated diphenyl ether (DeBDE), into the different environmental compartments during the e-waste recycling and their possible impact on the environment and workers/habitant's (adults and children) health. In particular, the study is focused on the application of one dynamic fate and transport model (2 FUN TOOL) and three models which comprise steady-state multimedia fate and transport (QWASI) and exposure models (EUSES, USEtox). These models link the results from the SFA on mass fluxes to the environment (see chapter "Global E-waste flow with focus on China") with the distribution and subsequently accumulation of the additives in the respective region, creating a potential exposure of workers/habitants during the e-waste recycling processes and a hazard for human health.

In the present case study a high level of variability and uncertainty has been found when researching data on the e-waste additives topic. Models such as 2-FUN are very useful to treat uncertain values and therefore, they fit to the current necessities. In addition, using not only these two models but also QWASI and USEtox widens the scope of the study due to their capacity to treat different scales and to simulate the distribution of substances in different environmental compartments. With this purpose, 2-FUN and QWASI are used for local scale, EUSES for regional scale and finally, USEtox for global scale. This last model was used as a first overview giving a background view and the results can be used for both life cycle impact assessment and risk assessment. Furthermore, the fact of applying different models allows assessing the strengths and drawbacks of them when applied in this type of studies.

The characteristics of the applied models have been described in detail in the chapters "Environmental Fate Models" and "A revision of current models for environmental and human health impact and risk assessment for application to emerging chemicals" and only a brief overview is given here. Since each model has its own approach, (i.e. QWASI is focused on the aquatic system), the combined results are expected to give a wider view with in-depth-analyses for different aspects compared to just one model with its special characteristics.

The study is divided in two phases: Firstly, the simulation of fate and transport through different pathways that the additives take from their release during recycling processes to the environment, and secondly, the uptake of Pb and DeBDE by local people through different exposure routes. In a first phase, the steady state models QWASI, USETOX and EUSES were applied, using as inputs the results obtained in the substances' fluxes calculated in the Chapter "Global E-waste flow with focus on China", and the obtained results were analyzed. However, the EUSES model was not applied for the inorganic substance Pb as considered not suitable. The USEtox^{TM} model was used to characterize the environmental risk of DeBDE and Pb for the whole China in order to provide certain background information to the case study of Guiyu. The steady state model EUSES was implemented for the analysis of the Region of Guiyu considering the regional scale, whereas the steady state model QWASI was implemented for the Guiyu Town, therefore considering the local scale. In the second phase of the study, the Physiologically Based Pharmacokinetics Model (PBPK), present in the 2 FUN Tool, was used for the Guiyu Town case study to predict the possible Pb concentration in the arterial blood of workers/habitants (adults and children), considering as inputs the results obtained with the steady state model QWASI. Previously, additional runs using as inputs environmental monitoring data from literature were undertaken and the results were lastly compared with the results predicted when using as inputs the outputs of QWASI model. Finally, sensitivity analysis for the dynamic 2 FUN model outputs was performed.

2. Case study framework

2.1. Selection of the study area

A literature review was made in order to identify the main Asiatic countries recycling e-waste and to select the most representative for this study. Twenty five articles were found to be of interest and were analyzed. The country selected was China, which seems to import and recycle 80% of e-waste produced in the US and about 10% of e-waste produced in Europe. An important recycling place in China is the Guiyu town (Fig 1), formed by several villages located in the Chaozhou region of Guangdong Province, 250 km northeast of Hong Kong, and which has a population of 150.000 including 100.000 migrants. At least 50% are labourers, and more than 300 companies and 3000 individual workshops are spread over more than 20 villages of the total 28 villages engaged in e-waste recycling work (Xinhua Net, 2005). This place has been defined by Greenpeace China as the second most polluted place in the world. For example, the report by the Basel Action Network and Silicon Valley Toxics Coalition published in 2002 pointed out that the lead concentration in samples taken in an e-waste recycling location from Guiyu was 2400 times that prescribed in World Health Organization (WHO) Drinking Water Guidelines (WHO, 1993). In December 2001, the levels at the same site were found to be 190 times the threshold WHO level. Furthermore, Leung et al. (Leung, et al. 2006) conducted a detailed study of the Guiyu area to quantify the pollution levels generated from e-waste, and found PBDE levels up to 1.169 mg kg^{-1} (dry weight) in soils near the recycling areas, which were 10–60 times higher than PBDE contamination reported at other locations in the world. In 2007 this study was extended (Leung et al. 2007) to surface soils and combusted residue in the Guiyu area and found total PBDE concentrations were highest in the combustion residue of plastic chips and cables collected from a residential area (33–97.4 mg kg^{-1}, dry wt); in soils from an acid leaching site (2.72–4.25 mg kg^{-1}, dry wt); and a printer roller dump site (0.59–2.89 mg kg^{-1}, dry wt). The authors also found that DeBDE was the dominant congener (35–82%) among the study sites.

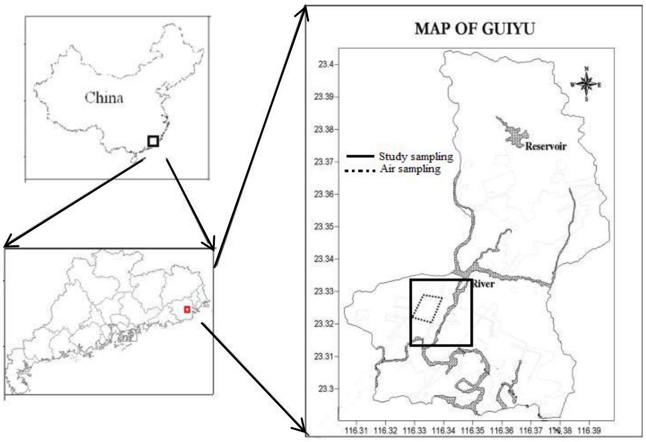

Figure 1. Map of Guiyu Town. The rectangles show the sampling area for determination of Pb in air and water as pointed out by Leung et al. 2008 and Guo et al. 2009.

2.2. Modelled substances

The models were run for two different electronic additives, one metal - lead (Pb) and one organic compound – decabrominated diphenyl ether (DeBDE).

The importance of assessing human and environmental impacts of metals and brominated flame retardants (BFRs) emissions has been growing in recent years (Pizzol et al. 2010, Takasuga et al. 2004). Metals are known for their high toxicity and capacity of accumulation in the environment and in the human body, such that long-term exposure to even low levels of metal contamination can lead to significant impacts on human health. Impacts resulting from chronic lead exposure have been extensively studied and demonstrated; for example, the lowered cognitive and learning abilities in children exposed to Pb during infancy (Canfield et al. 2003, Jusko et al. 2008). Concerning the (BFRs), of which chemical class DeBDE is, their impact on health and the environmental characteristics are generally not well known. However, the acute toxicity of most of the BFRs has proven to be fairly low, but some BFRs have shown similar toxic effects to PCBs and PCDDs and furans (DEPA 1999). The available data suggest, for example, that the lower PBDE congeners (tetra to hexa) are likely to be carcinogens, endocrine disrupters and/or neurodevelopment toxicants (Burreu et al. 2000). DeBDE, which is the major commercial product, is presumed to be a less active congener than the lower BDEs because of its lower bioavailability and poor gastrointestinal adsorption (Hopper and McDonlad 2000).

3. Environmental and Human Health modelling

3.1. Description of models

A detailed description of the four models involved in this study has been provided in the Chapters “Environmental Fate Models” and “A revision of current Models for environmental and human health impact and risk assessment for application to emerging Chemicals”. Models were run preferably for Pb and DeBDE on different geographical scales:

USEtox (Rosembaum, et al., 2008) was used to address the continental scale. It can be applied to assess either eco-toxicity or human toxicity from different pollutants. It calculates characterization factors for human toxicity and freshwater ecotoxicity, taking into account the environmental fate, exposure and effects of the substance.

For regional scale, EUSES, a European Union multimedia environmental model for risk assessment of new and existing substances was applied considering the region where the e-waste recycling sites are.

QWASI, the Quantitative Water, Air Sediment Interaction model by Mackay et al. (1983) is a fugacity III model (Version 3.10, 2007) and it describes the fate of chemicals in aquatic systems, depending on direct discharge, inflow in rivers, and atmospheric deposition. Hence, this model addresses the local scale, as does the 2-FUN Tool.

The 2-FUN Tool is a new integrated software based on an environmental multimedia model (comprising several environmental compartments), physiologically based pharmacokinetic (PBPK) models (to simulate the body burden of toxic chemicals), and associated databases.

In Table 1 a characteristics summary of the applied models is presented. The listed parameters are selected with regard to relevance to this chapter and do not assume completeness.

Table 1 Characteristics summary of the applied models

	USEtox	EUSES	QWASI	2-FUN
Geographical scale addressed in this chapter	Global (Results obtained for China)	Regional (Results obtained for Guiyu region)	Local (results obtained for Guiyu town)	Local (results obtained for Guiyu town)
Environmental compartments addressed	air freshwater coastal	air freshwater sediment	air sediment water	Air water soil

	water natural soil agricultural soil	natural soil agricultural soil industrial soil vegetables, animalproducts and fish		vegetables animal products (milk and beef) fish
Human health related output	Human intake doses	Human and ecosystems risk	-	Arterial blood concentrations in workers/children
Substances addressed in this chapter	DeBDE Pb	DeBDE	DeBDE Pb	Pb

In the following subsections, the inputs required and outputs obtained by these models are presented.

3.2. Inputs to models

Each model requires inputs, some of them are common but there are others that differ, in the following subsections these inputs are explained with more detail.

3.2.1. Background data:

USEtox^{TM}. In order to determine the environmental risk characterization for DeBDEs and Pb in China, USEtox^{TM} requires to define different scales. Since China is such a big country, the whole country has been considered as a continent. Therefore, the continental landscape data is defined by parameters describing this target country. Moreover, due to the nature of the model, two additional scales have to be defined, Global and Urban. To solve this issue, the world is defined as the global scale whereas Guiyu is taken into account as the urban one. However, although two other scales are defined, the final results are obtained for China.

Euses. The present model requires, as the USEtox model, the definition of different scales. In this study the regional scale environment is calibrated for Guiyu region (10^3 km^2), and the continental scale for the country of China. Concerning the data sources, the information for the continental scale has been extracted from GLOBACK. It seems important to highlight that the parameter sets contain the spatially differentiated parameters for the GLOBOX model. Monte Carlo simulation was run using Crystal Ball 2000.2.2, Standard Edition software. Detailed input data, used for the present case study, are available in the Annex X.

QWASI Lacking more detailed knowledge on the actual Guiyu site, it was assumed that emissions take place to a river that is 5 m deep, 60 m wide and 5 km long. Its sediment has an active layer of 0.05 m, and an organic content of 3 %. The burial rate of solids equals the resuspension rate and is assumed to be 0.6 g·m-2·day-1. The flow rate of the river was estimated using the data in Italian rivers, because of the scarcity of data from Chinese rivers. Therefore, based on the previously settled Guiyu river characteristic and the slope of the Guiyu surface (data available from Google Earth) and comparing with the Italian rivers, a flow rate of 40 m^3s^{-1} was considered. An average rain rate of 2.2 m·y-1 is used. Further assumptions are specified in the appendix.

2 FUN Tool. For the development of the scenario, it was only considered the Guiyu area where the e-waste recycling facilities are located, near the main rivers Lianjiang and Nianyang, 10 km^2. Due to the fact that the two rivers are connected it has been considered as one main river. This approach was previously adopted by Leung et al. 2008 and Guo et al. 2009 (Fig.1). The river geometry data, necessary for the scenario development, has been settled based on Google Earth distance measurements using the ruler tool, and was used when running QWASI model as well. The depth of the river was settled at 5 m based on data reported by Chen et al. (2005) which presented the velocity profile of rivers in China. Water flow has been estimated to be 40 m^3·s-1. The weather data of the region, necessary for the development of the scenario (air temperature, wind speed, precipitation) was available from http://www.tutiempo.net/en/Climate/Shantou/05-2010/593160.htm (last online check 24/04/2012) whereas the soil temperature was calculated based on air temperature using the approach described by D. L Nofziger (http://soilphysics.okstate.edu/software/SoilTemperature/document.pdf).
Monitoring data for Pb concentration in water and air found in literature were scarce, therefore, maximum and minimum values were selected, based on two main climatic seasons. A sinusoidal function was applied using the maximum and minimum values in order to create the essential model inputs. The inputs for Pb concentration in air were created based on the data reported by Leung et al. (2008) and Wong et al. (2007) whereas the inputs for Pb concentration in surface water were taken from Guo et al. (2009). Additional regional data as total suspended particles in the atmosphere and global solar radiation were set based on information available in literature (Deng et al. 2006, Liang and Xia 2005). All the input data, used for the present case study, are available in the Annex X.

3.2.2. Substance data:

The substance data required by the models (e.g. physico-chemical parameters, toxicological data, etc.) have been normally extracted from different databases depending of the substance. In the present case, the information for both substances, lead and PBDE were taken from the Riskcycle Database.

3.2.3. Emissions:

USEtox^{TM}. In order to make sure that the results provided by USEtox^{TM} are referred to China, the predicted emissions into the environmental compartments have been calculated using as a basis the information on the e-waste flow and presented in the chapter "Global E-waste flow with focus on China". Therefore these emissions are taken as inputs. Table 2 presents a summary with the main values extracted from the aforementioned chapter and used to run the USEtox^{TM} model.

Table 2.- Emission (to China) input entered to the USEtox^{TM} model

Emission to: ($t \cdot y^{-1}$)	**Air**	**Soil**	**Water**
Pb	0.179	28.6	2.437
DBDE	$2.72 \cdot 10^{-2}$	0.736	$9.38 \cdot 10^{-2}$

Euses. The annually tonnage treated in China was assumed to be $2.5 \cdot 10^{6}$, the percentage treated in Guiyu was assumed to be 50% (with a uniform probability distribution between 40 and 60%). Air emission is the sum of three emission sources (emission to air during burning, migration to dust and air, and loss during the process of dismantling) (see chapter "Global E-waste flow with focus on China"). Soil emission takes into account the concentration of DeBDE in ashes after open burning operation (see chapter "Global E-waste flow with focus on China"). Finally water emission takes into account the leaching in open dumps (see chapter "Global E-waste flow with focus on China"). The values of the emission to air, industrial soil and surface water are presented in Table 3. Whereas in air and soil normal distributions were considered for probabilistic analysis, a uniform probability function in surface water emission was assume due the scarcity of data of DeBDE leachates concentration.

Table 3.- Emission to different environmental compartments.

Emission to: ($t \cdot y^{-1}$)	**Air**	**Industrial Soil**	**Surface water**
DeBDE	$8.7 \cdot 10^{-3} \pm 1.7 \cdot 10^{-3}$	$2.4 \cdot 10^{-1} \pm 5.7 \cdot 10^{-2}$	(Min $5.6 \cdot 10^{-3}$; max $5.6 \cdot 10^{-2}$)

QWASI. As QWASI focusses on aquatic systems, either one or more of the following input data is needed in order to calculate the partitioning of substances between air, water and sediment: Emissions to water, concentration in effluents, or concentration in (emitted) air. For lead, the SFA-study estimated a total of approximately 9020 kg of Pb being emitted to water every year (0.02 $kg \cdot y^{-1}$ from dumped plastics; 0.19 $kg \cdot y^{-1}$from dumped metals, 9019.68 $kg \cdot y^{-1}$ from CRT glass), while 46 $kg \cdot y^{-1}$were emitted to air from burned plastic, applying emission factors from von Oers (2011) and Thomas et al. (1998). For the Pb distribution, the input via atmospheric concentration is assumed to have a significant impact on the distribution of the heavy metal. The derivation of average concentration of Pb in air from the SFA results for burned plastics, however, is difficult. Instead, the sensitivity of Pb partitioning to water and sediment for different air concentrations was investigated (low: $0.44 \cdot 10^{-3}$mg·m^{-3} (Deng et al. 2006); high: $7.47 \cdot 10^{-3}$mg·m^{-3}, Leung et al. 2008 and rain rates (0.021 to 4.38 m·y^{-1}), assuming the emission to water of 9020 kg·y^{-1} to remain constant.

With regard to DeBDE, the SFA suggested 186 t to be deposited around Guiyu as waste every year. Assuming, as done with Pb, that a certain amount of additives, that have been deposited as waste after the recycling process, would leach from the disposal sites to water, would have been more complicated because leaching data for DeBDE are scarce. Accordingly, a range of emission values to water (0 to 30 kg y^{-1}) were used as input data. Resulting concentrations for sediment and water were then compiled from the different simulations.

3.2.4. Human exposure:

2-FUN tool and **EUSES** . The main pathways of human exposure considered in the scenario were inhalation of contaminated air, ingestion of fish from the river, ingestion of several crops (potato, root, leaf) considered as being cultivated in the area and irrigated with the water from the river, and the ingestion of beef and milk of habitant's cows (Fig. 2). For the 2 FUN Tool, the concentration of Pb in the grass used for cows nutrition was necessary and was calculated considering the Pb concentration in the Guiyu soil and grass uptake (Rotkittikhun et al. 2007). The drinking water was not considered as exposure pathway due to the fact that the levels of toxicity of the water bodies in the Guiyu region were high and the local authorities decide to import the drinking water from other regions, as reported in the literature (BAN and SVTC 2002). The values for the ingestion rates by humans of fish and potato were taken from Xing et al. 2009 and FAO 2008 respectively. The values for the ingestion rates for root and leaf crops, beef and milk were settled based on literature values (Li et al. 2011) or taken from GLOBOX. Parameters from GLOBOX were obtained from FAO Statistical Databases. All the values are presented in the Annex X.

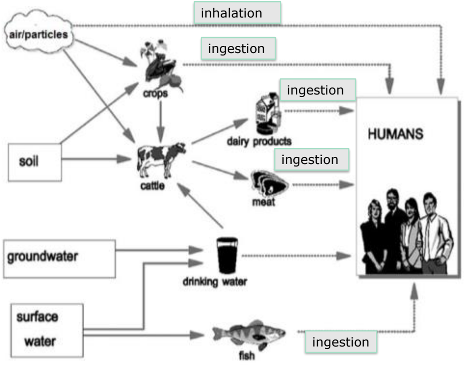

Fig. 2. Lead and PBDE pathways into humans

The key input parameters used in the 2 FUN model were given in the form of probability density function (PDF) to allow the exhaustive probabilistic analysis and sensitivity analysis in terms of simulation outcomes.

As a general comment related to the input data, it is interesting to mention that all the considered models allow a wide modification of the input data but also provide default values when there exists a lack of input data. Furthermore, there has been a consensus to select the data used as a basis for all the four applied models from a set of different databases. These data can be consulted in the Annex 1.

3.3. Models Outputs

In this section a brief definition of the main outputs of each model is presented. Again, these outputs can be either common or different. In any case, the outputs provided by each model are detailed below.

3.3.1. Concentration in the environmental compartments:

USEtox^{TM}. Those concentrations can be obtained for the theoretical case of one kg emitted into the urban air (default USEtox^{TM}) or considering the emissions obtained with the developed scenarios (chapter 1). It is important to highlight that these concentration values are calculated by the model considering processes such as advection, transportation, degradation, etc. among the different scales implemented by USEtox^{TM}.

Euses. In EUSES 2.0 steady-state exposure concentrations at the regional, continental, and global scales are calculated for all environmental compartments (air, water, soil, sediment and air) using the multi-media fate model SimpleBox 3.0.

QWASI. Quasi is a fugacity model and has mainly been developed for substances that have a vapour pressure which drives their distribution in the environment. However also inorganic substances such as lead have been modelled, even though they are not volatile and do not partition significantly into air. A modification of the model, requiring partition coefficients for the different interfaces and calculating the partitioning from water concentration has been used here for lead. Even though speciation is not considered, data are compiled for resulting concentration of additives in sediment, water and air, as well as on their sensitivity regarding emission pathways. The output data are then compared with literature data and provide one basis for human health risk assessment and the environmental risk assessment.

2 FUN Tool. Depending on the complexity of the scenario the 2FUN Tool calculates the concentration in soil, water, vegetables (leaf and root), cereals, fruits, animal products (milk and beef) and fish.

3.3.2. Human intake Fractions:

USEtox^{TM}. The USEtoxTM model provides outputs such as human intake fraction of a certain substance ($kg_{intake} \cdot kg_{emitted}^{-1}$) for different exposure pathways.

EUSES. As in case of Usetox Model, the present model provides outputs such as human intake fraction of a certain substance for different exposure pathways. In the present case study, estimation of the human intake doses for Guiyu was calculated. These results were compared with the incidence and severity of the effects (dose-response assessment).

3.3.3. Human and ecosystem risk evaluation

EUSES. The assessment of potential adverse effects induced by DeBDE exposure in both human and environment targets has been evaluated by means of the Euses model. Toxicological and ecotoxicological values for this contaminant were included as input values and then compared with the results of the exposure for humans and environmental receptors.

Chronic daily intake (CDI), Hazard Index (HI) and Cancer Risk (CR) for carcinogenic effects were calculated and exposures associated with HI<1 and CR<1E-6 were considered negligible.

For the ecological assessment, risk analysis was based on PEC/PNEC ratio (Hazard Quotient) where PEC is the predicted environmental concentration (resulting from Euses results) and PNEC the predicted no-effect concentration.Exposures associated with HQ<1 were considered negligible.

2 FUN Tool. The 2FUN Tool provides outputs such as bioaccumulation of the substances in the target organs or concentrations in the human blood. In the present study the Pb concentration in the arterial blood of children/workers has been assessed.

4. Results

Once the main outputs of each model have been explained, the results of the application of the models to the present case study are presented in this section.

4.1. USEtox^{TM}

The outputs provided by the model can be used to perform a risk assessment if they are compared with reference limit values and literature data in order to determine whether the situation is risky or not. The values obtained by USEtox^{TM} and used for the characterization of the risk in China are presented in the following tables. Table 4 presents the values for the concentration into the environmental compartments of the concerning additives (Pb and DeBDE).

Table 4.- Additive concentration in the environmental compartments calculated by USEtox^{TM}

Concetration in: (mg·m $^{-3}$)	Air	Fresh Water	Coastal Water	Nat. Soil	Agr. Soil
Pb	$8.47 \cdot 10^{-11}$	$9.24 \cdot 10^{-4}$	$4.54 \cdot 10^{-6}$	$1.99 \cdot 10^{1}$	$1.99 \cdot 10^{1}$
DeBDE	$1.29 \cdot 10^{-11}$	$2.20 \cdot 10^{-5}$	$5.06 \cdot 10^{-8}$	$1.12 \cdot 10^{-3}$	$1.12 \cdot 10^{-3}$

Comparing these results with the literature values on maximum concentration permitted (MCP) of additives for different environmental compartments, the following statements can be done:

- The Pb concentration obtained in air with USEtox is much lower ($8.47 \cdot 10^{-11}$ mg·m^{-3}) than the MCP values found in literature ($5 \cdot 10^{-2}$ mg·m^{-3}, OSHA (2012)).
- The Pb concentration obtained in water with USEtox is lower ($9.24 \cdot 10^{-4}$ mg·m^{-3}) than the MCP values found in literature (50 mg·m^{-3}, De Mora and Harrison, 1984).

- The DeBDE concentration obtained in air with USEtox is much lower ($1.29\cdot10^{-11}$ mg·m^{-3}) than other concentration values ($1.24\cdot10^{-5}$ – $9.89\cdot10^{-8}$–$1.24\cdot10^{-5}$ mg·m^{-3}) found in literature (Chen et al., 2011).
- The DeBDE concentration obtained in water with USEtox is lower ($2,20\cdot10^{-5}$ mg·m^{-3}) than concentration values ($3.35\cdot10^{-4}$ – $6.52\cdot10^{-2}$ mg·m^{-3}) found in literature (GuanYu et al., 2007).

Although China is one of the countries with more informal recycling facilities and therefore with a high release of selected additives (Pb and DeBDE) into the environment, the results obtained are lower than the expected ones. This is due to the fact that even if the amount of additive released is high, since China is such a big country, the result concentration is diluted. Moreover, there could be an underestimation of the treated e-waste volume in China.

On the other hand, values of intake doses obtained with USEtox for human beings are presented in

Intake Dose through: (mg·(kg·d)$^{-1}$)	**Air**	**Drinking water**	**Exposed produce**	**Unexposed produce**	**meat**	**Dairy products**	**fish**	**TOTAL**
Pb	3.91·10-11	4.25·10-8	2.74·10-6	2.33·10-7	5.95·10-10	1.65·10-9	8.34·10-8	2.99·10-6
DeBDE	5.94·10-12	7.02·10-11	2.29·10-10	3.64·10-11	1.47·10-10	3.27·10-11	8.86·10-10	1.41·10-9

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Table 5.- Intake doses for human beings provided by the USEtox^{TM} model

Intake Dose through: (mg·(kg·d)$^{-1}$)	**Air**	**Drinking water**	**Exposed produce**	**Unexposed produce**	**meat**	**Dairy products**	**fish**	**TOTAL**
Pb	$3.91\cdot10^{-11}$	$4.25\cdot10^{-8}$	$2.74\cdot10^{-6}$	$2.33\cdot10^{-7}$	$5.95\cdot10^{-10}$	$1.65\cdot10^{-9}$	$8.34\cdot10^{-8}$	$2.99\cdot10^{-6}$
DeBDE	$5.94\cdot10^{-12}$	$7.02\cdot10^{-11}$	$2.29\cdot10^{-10}$	$3.64\cdot10^{-11}$	$1.47\cdot10^{-10}$	$3.27\cdot10^{-11}$	$8.86\cdot10^{-10}$	$1.41\cdot10^{-9}$

After an intensive research on maximum intake dose permitted for human being through different pathways, it has been observed that the most reliable was to consider the total intake since the particular intake pathway data was not clear enough. After comparing these values, the following statements can be made:

- The total intake dose for Pb obtained by USEtox ($2.99 \cdot 10^{-6}$ mg·(kg·d)$^{-1}$) is much lower than the one from literature ($25 \cdot 10^{-3}$ mg·(kg·d)$^{-1}$, JEFCA (2002)).
- The total intake dose for DeBDE obtained by USEtox ($1.41 \cdot 10^{-9}$ mg·(kg·d)$^{-1}$) is much lower than the one from literature (0.1 mg·(kg·d)$^{-4}$, EPA (1984)).

In this case, a similar situation to the previous one of the environmental compartments occurs. The fact that the intake dose is calculated for the whole China population supposes that these intake doses are lower than the expected again. They are calculated taking into account the emissions coming mainly from the polluted spots but considering all the Chinese population as a potential receptor. However, not all the citizens are affected by these intake doses. In addition, as mentioned before the potential underestimation of the treated e-waste volume in China could be the cause of these low values.

4.2. EUSES Model

In EUSES 2.0 steady-state exposure concentrations at regional scale are calculated for all environmental compartments using the multi-media fate model SimpleBox 3.0.

Table 6 shows the result of Predicted environmental concentration in air, soil (natural, agricultural and industrial) fresh water and sediment for regional scale (Guiyu region).

Table 6.- Predicted environmental concentration (PEC) of DeBDE (mean ± standard deviation).

	Air (mg·m^{-3})	Nat. Soil (mg·kg^{-1})	Agr. Soil (mg·kg^{-1})	Ind. Soil (mg·kg^{-1})	Fresh Waterwater (mg·l^{-1})	Sediment (mg·kg^{-1})
PEC Regional	$1.4 \cdot 10^{-8} \pm 2.8 \cdot 10^{-9}$	$1.9 \cdot 10^{-2} \pm 4.0 \cdot 10^{-3}$	$1.6 \cdot 10^{-2} \pm 3.2 \cdot 10^{-3}$	$1.3 \cdot 10^{1} \pm 3.4$	$4.9 \cdot 10^{-6} \pm 1.2 \cdot 10^{-6}$	$7.6 \cdot 10^{-3} \pm 1.8 \cdot 10^{-3}$

These PECs fit well with data from Guiyu town obtained from a literature review (see Annex 1). It can be observed that the most impacted compartments are soil and sediments. The value of total regional intake doses of DeBDE for human beings is 29.7 mg·(kg·d)$^{-1}$ with a range from 7.8 to 72.5 mg·(kg·d) $^{-1}$. The main regional pathway of exposure with more than 99.9% of the total contribution is daily intake of root crop due to the high levels in soils. Other regional daily intake are drinking water, fish intake, meat intake, inhalation intake with values $8.0\cdot10^{-8}$, $1.2\cdot10^{-6}$, $1.1\cdot10^{-6}$ and $4.2\cdot10^{-9}$ mg·(kg·d)$^{-1}$, respectively.

In order to assess the risk for the population living in Guiyu, the daily intake was compared with the oral reference value for DeBDE. According with EPA, the DeBDE daily oral reference and oral slope factor are 7.0E-03 mg·(kg·d)$^{-1}$ and $7.0\cdot10^{-4}$ mg^{-1}·kg·d. Inhalatory reference concentration and inhalatory unit risk have not been established.

Results of the daily intake (oral exposure) in Guiyu are much higher than the oral reference value. Therefore hazard quotient is higher than one. That means that there is a risk of developing other effects than carcinogenic ones due to the dietary exposure. The carcinogenic risk resulted to be $8.9\cdot10^{-3}$ (8.9 cancer cases in 1000 inhabitants) higher than the acceptable value. On the other hand, the risk for the soil ecosystem (worm-eating predators) is high due to the elevated concentrations found in soil.

4.3. QWASI

On the basis of the above-mentioned assumptions, atmospheric concentrations, used as inflows to the model, in the range of $9.37\cdot10^{-9}$ to $2\cdot10^{-6}$ mg·m^{-3} (data not shown) did not have a significant influence on the partitioning of DeBDE. A strong impact could be assigned to the direct discharge and to chemical concentration in inflow waters. There is, however, no information available on concentration of DeBDE in emitted waters. Consequently, different direct discharges from the e-waste sites were used as emission data and the subsequent concentrations for sediment and water concentrations in the river were calculated (Fig. 3).

Applying emission data in the range of 1 to 30 kg·y^{-1}, sediment concentrations ranged from $1.4\cdot10^{-2}$ to $4.32\cdot10^{-1}$ mg·kg^{-1}, water concentrations were calculated to be within $0.7\cdot10^{-6}$ and $2.37\cdot10^{-5}$ mg·L^{-1}

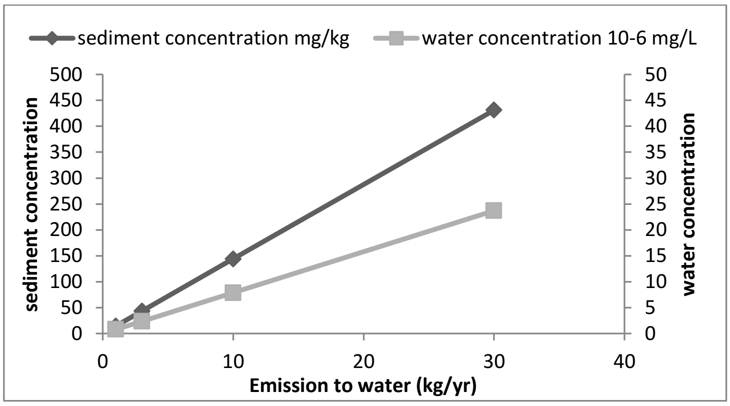


Fig. 3: Relationship between direct discharges of DeBDE to water in kg/yr from a hypothetical e-waste side and the partitioning of the substance between sediment and water phase.

Comparing the result from the EUSES model for the concentration of DeBDE in water ($4.43 \cdot 10^{-6}$ $mg \cdot L^{-1}$), this would refer to approximately 6 kg DeBDE$\cdot y^{-1}$ emitted to water and a concentration in sediment of below 0.1 $mg \cdot kg^{-1}$. With regard to sediment concentration, this is in the same range of magnitude as data published by Luo et al 2007, who measured DeBDE concentrations at 3 different sites in Guiyu of between $1.4 \cdot 10^{-2}$ and $6.2 \cdot 10^{-2}$ $mg \cdot kg^{-1}$.

Comparing the results on direct discharges with the results of the SFA for Guiyu of 186 t of DeBDE accumulating over 1 year in a land disposal, the emission of 6 kg would account for $3 \cdot 10^{-3}$ % leached material within one year, which is well in the range of $3 \cdot 10^{-3}$ to $3 \cdot 10^{-2}$% that have been calcucated by Choi et al (2007) for leaching of DeBDE in contact with dissolved humic substances from TV housing plastics.

Lead:

Table 7 lists Pb concentrations in water (upper table) and sediment (lower table) for different air concentrations and rain rates, using a constant emission of lead to water of 9019 kg per year. Resulting sediment concentrations vary between $0.4 \cdot 10^{3}$ $mg \cdot kg^{-1}$ (low rain rate, low air concentration) and 38 $\cdot 10^{3}$ $mg \cdot kg^{-1}$ (high rain rate, high air concentration. Water concentrations under the same conditions range from $7 \cdot 10^{-3}$ to $63 \cdot 10^{-2}$ $mg \cdot L^{-1}$.

Table 7 : Pb concentrations in water (upper table) and sediment (lower table) as calculated by QWASI

Pb WATER concentration (mg·L^{-1})				
		Rain rate in m·y^{-1}		
		Low 0.021	Average 2.2	High 4.38
Concentration in Air	Low (0.44·10^{-3}mg·m^{-3})	7·10^{-3}	2.5·10^{-2}	4.4·10^{-2}
	High (7.44·10^{-3}mg·m^{-3})	9·10^{-3}	3.2·10^{-1}	6.3·10^{-1}

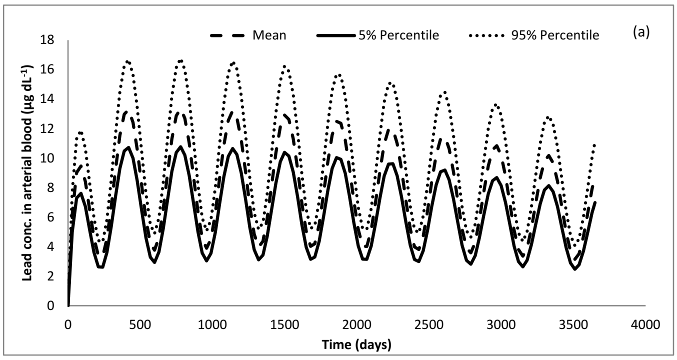
Pb SEDIMENT concentration (mg·kg^{-1})				
		Rain rate in m·y^{-1}		
		Low 0.021	Average 2.2	High 4.38
Concentration in Air	Low (0.44·10^{-3}mg·m^{-3})	0.4·10^{-3}	1.6·10^{-3}	2.7·10^{-3}
	High (7.44·10^{-3}mg·m^{-3})	0.5·10^{-3}	1.9·10^{-2}	3.8·10^{-2}

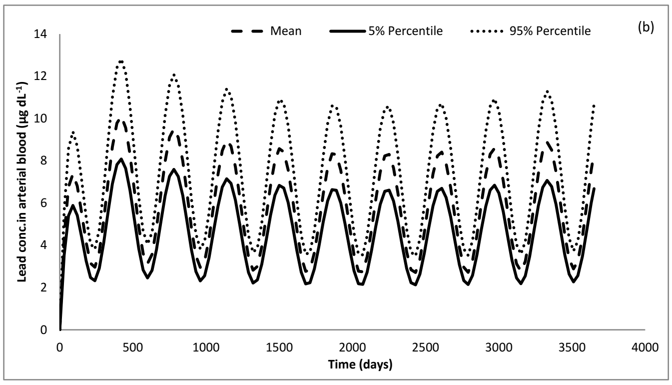
According to Guo et al. 2009, lead concentrations of 6·10^{-3} to 6.1·10^{-2} mg·L^{-1} have been measured in water in Guiyu. This would be in the range of our results assuming a low concentration in air and low to high rain rates, or a high concentration in air and low rain rate. The combination of high air concentration and high rain rate, resulting in as much as 38·10^{3} mg·kg^{-1} in sediment is highly unlikely, as increased precipitation would quickly reduce the lead content in air.

4.4. 2 FUN

Figure 4 presents Pb concentrations in the arterial blood over the simulation period, with values at mean, 5th and 95th percentiles. The inputs data for Pb concentration in air and water of the region, used for the simulations, were created based on monitoring data available in literature. Simulations were performed for 10 years, setting the initial age of population at 2 years (Fig 1a) 10 years (Fig 2b) and 20 years (Fig.2c) respectively. This distribution comes from the fact that many articles report that young children spent a long time in e-waste recycling sites, sometimes to help their parents. It can be assumed that the values at 95th percentile represent "pessimistic" scenarios in the context of health risk assessment. It was found that the Pb concentration in blood is much higher in very young children than in more grown up children whereas the general trend is to find the lowest concentration in adults blood. Looking at the values at mean and 95th percentile for all the cases considered the values of Pb in the arterial blood were higher that the limit established by the Centre of Disease, Control and

Prevention , 10 μg·dL^{-1} (CDC 1991). For the very young children in the first six simulated years even the values at 5 % were higher than the limit of 10 μg·dL^{-1}. These results are in agreement with the monitoring results reported by Huo et al. 2007, who observed that the levels of Pb in blood in 165 children of Guiyu ranged from 4.40 to 32.67 μg·dL^{-1} with a mean of 15.3 μg ·dL^{-1}.





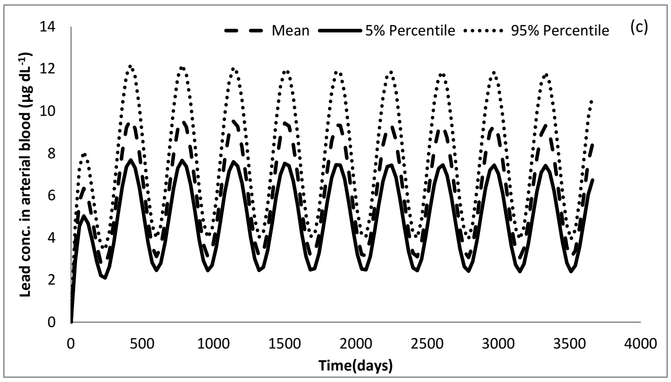


Fig. 6. Lead concentrations in arterial blood (µg dL^{-1}) over 10 years simulation; (a) initial age 2 years, (b) initial age 10 years, (c) initial age 20 years

Concerning the results, using as inputs the concentration of Pb in the river water calculated with the QWASI model, any significant differences on the concentrations of Pb in the arterial blood of children/adults were not observed when these results were compared with the obtained using as inputs the literature values for lead concentration in the river water (data not showed). This was somehow expected considering that the results of QWASI model were in the range with the measurements reported in literature. Furthermore, any significant differences weren't observed for lead concentrations in leaf, root, fish, beef, milk and potato either

4.4.1. Global sensitivity analysis

A global sensitivity analysis was performed for the lead concentration in the arterial blood model (Fig 7) over the simulation period for each parameter. Parameters considered for the sensitivity analysis are listed in Table 1(Annex X). The magnitude of sensitivity is shown by relative sensitivity index. It was observed that the most influential parameter is the porosity of the sediment of the river (phi_sed) followed by the density of the dry grass, the second regression coefficient of the relationship expressed by log(fish_BCF)= α + β×log(C_dis_water) and the density of the dry root. The higher concentration of lead was observed in fish when comparing with the beef, milk and the considered crops (Fig 8).

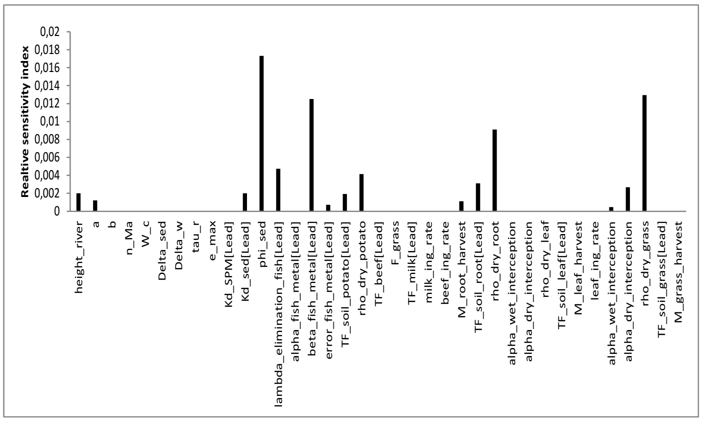


Fig.7. A global sensitivity analysis for lead concentration in arterial blood

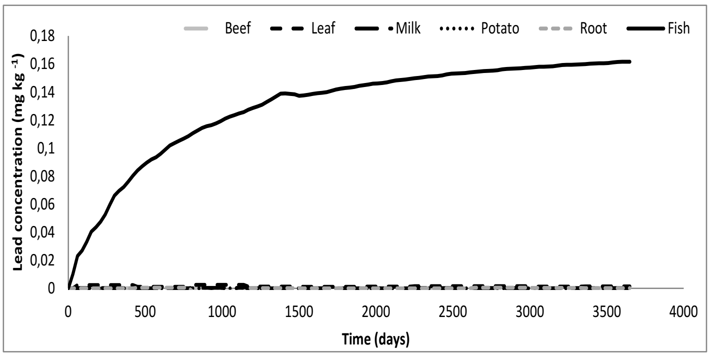


Fig.8. Lead concentration in the considered crops, fish, milk and beef (mg kg^{-1})

5. Discussion

After presenting the results obtained by each model, an integration of all them is done in this section going from the general background information (China) to the most specific case scenario (Guiyu).

The results obtained with USEtox^{TM} by China show that the situation for the Chinese country against the hazards caused by the selected additives is not risky. However, this is due to the fact that all the area of land and fresh water in China have been taken into account whereas there is only a fraction of this area that is really affected as well as that not all the population intakes the obtained fractions.

Obviously, when focussing on the source of pollution, for example the Guiyu Region, these results are higher since, according the present research (Zoeteman et al. (2004) and Lee et al. (2002)), this region deals with practically half of the China e-waste.

It is interesting to mention that USEtox^{TM} is mainly a tool for LCIA studies where characterization factors are obtained for a wide list of substances. However, the model also provides as intermediate outputs parameters (e.g. intake doses, concentrations in environmental compartments, substance exposure) that can be used for risk assessment studies. This has happened in the present case comparing the values from USEtox with reference limit values.

Moreover, this model counts with a substance database for both organic and inorganic substances as well as default values when a parameter is unknown. However, the model is more developed for the organic compounds than for the inorganic ones. Multicalculations can be performed at the same time with different substances.

The results obtained with the EUSES model for the region of Guiyu are in some cases concerning, as already showed by the USEtox results. In particular, it was observed that the daily intake (oral exposure) is much higher than the oral reference value. On the other hand, the carcinogenic risk (8.9 cancer cases in 1000 inhabitants) is higher than the acceptable value. Because the higher dietary levels come from root vegetables, especially from those that grow up in contaminated soil, population should be informed about this potential risk. Regarding environmental risk, there is a high risk situation for soil ecosystem and especially in the case of worm-eating predators ($1.2 \cdot 10^2$) (PEC/PNEC ratio).

Euses is a useful model for chemicals outside the domain of the persistent, non-dissociating substances of intermediate lipophilicity. Due to the high complexity of the model it is lacking in transparency on the other hand, the performance of the model is characterised as a good compromise between complexity and practicability. In order to adapt the model to different assumptions or assess the uncertainty is necessary to use the EU TGD2003 Spreadsheet version 1.24 of April 2008. This spreadsheet aims to represent the algorithms described in the 2003-version of the EU Technical Guidance Document, as implemented in EUSES 2.0.3.

Under the applied QWASI model assumptions, the QWASI results are in the range of measured data reported in literature and thus support that the strongest impact to sediment and water concentrations of DeBDE are from direct emission to water as opposed to atmospheric concentrations. This result points out the high importance of DeBDE-leaching from deposited waste material and a lower meaning of the fraction that is transferred to the atmosphere.

Pb concentration in the environment, on the other site, is strongly influenced by air concentration and rain rate, in addition to direct emissions to water. For this metal the exposure pathway via burning gains a high importance among the different informal recycling processes.

While QWASI is an easy to use multimedia fate modelling tool, it has been originally designed as a fugacity model. Even though an adaptation to ionic substances exist and it has been applied to lead before, it needs to be recognized that it does not take speciation of metals into account. This adds to the overall uncertainty of results.

If the results of the EUSES model show a certain concern for human risk and ecosystems due to DeBDE released during the e-waste recycling operations in the region of Guiyu, the results of the 2 FUN Tool show a real health risk for workers/habitants of the Guiyu town due to the released of the other additive considered in this study, Pb. A higher risk was observed for very young children, with values of Pb in the arterial blood at 5 %, mean and 95 % higher than 10 µg dL^{-1}. The U.S. Centers for Disease Control and Prevention (CDC) defined elevated blood lead levels as those ≥ 10 µg dL^{-1} in children ≤ 6 years of age (CDC 1991). Nevertheless, studies have increasingly shown that low blood Pb concentrations, even < 10 µg/dL, were inversely associated with children's IQ (Inteligent coefficient) scores and academic skills (Canfield et al. 2003; Lanphear et al. 2000, 2005). Therefore, no safety margin at existing exposures were identified (Koller et al. 2004).

Average range between Pb concentrations at 5th and 95th percentile over the 10 years simulation period, for very young children (i.a. 2 years), children (i.a. 10 years) and adults (i.a. 20 years) was 1.3 orders of magnitude. It indicates that the parametric uncertainties and variability contained in input parameter contribute significantly to propagation of such gaps in outputs.

Regarding the global sensitivity analysis, the results indicate that the variation of the model output is highly sensitive to the variations of parameters used in fish and root compartments. The higher concentration of PB in fish than in potato, leaf, root, milk and beef (Fig 8) reflects that the variation of the model output is more sensitive to variations of fish parameters than of potato, leaf, root, milk and beef parameters.

However, as a general observation, this study demonstrated the feasibility of the integrated modelling approach to couple an environmental multimedia and a PBPK models, considering multi-exposure pathways, and thus the potential applicability of the 2-FUN tool for health risk assessment. The global sensitivity analysis effectively discovered which input parameters and exposure pathways were the key drivers of Pb concentrations in the arterial blood of adults and children. This information allows us to focus on predominant input parameters and exposure pathways, and then to improve more efficiently the performance of the modelling tool for the risk assessment.

6. Conclusions

During the development of the present study, the uncertainty has been present in many occasions, especially when gathering data concerning the e-waste amounts, the content of additives in the e-waste inflow and the percentages of additive going to the different environmental compartments. This has implied that some assumptions have to be taken. However, due to the capability of the selected models of treating uncertain values, the results obtained after the models calculations were quite acceptable. Through the comparison of the results of each model with values extracted from literature, in most of the cases it was observed that the predicted values were at the same order of magnitude as the monitored values. This fact reinforces the suitability of the selected models and the validity of the obtained results.

Concerning the risk characterization of the selected additives, it has been observed a clear increase of the risk for the environment and human health when reducing the scale of the study. Therefore when analysing Guiyu city, the most worrying results are obtained. This statement could seem obvious but there exists a strong belief that the whole country of China is suffering the hazard of the e-waste pollution, even if when running the USEtoxTM, to assess the environmental distribution of additives for the country of China, low concentrations of e-waste additives were found. Nevertheless, at a regional scale, the results provided by EUSES model were higher and quite similar to the monitoring data found in literature for the region of Guiyu. Therefore, since the monitored values from literature are pointed out to be higher than the reference limit values, it can be stated that, as expected, the region of Guiyu present a high level of risk both for environment and human health. When analysing the local scale results, a similar situation was observed looking at the data obtained by the QWASI model for the aquatic environment; values on the same range that the monitored values extracted from literature were observed coming up with similar conclusions than the ones obtained for the regional scale. Finally, with the PBPK model of the 2-FUN tool were obtained values for lead concentration in the arterial blood at a higher range than the limit of concern established by the CDP, above all for the very young children. These results were, as in the previous cases, in agreement with the monitoring data reported in the literature. With all this, it seems clear to state that the potential risk caused by the e-waste additives due to the informal recycling in Guiyu is significant.

As a final objective of this study, an assessment of the strengths and weaknesses of the selected models for this type of studies was undertaken. Despite the fact that the USEtoxTM model is actually a tool for LCIA assessment, a risk characterization has been performed with this model in order to provide some background information to the present case study. The EUSES model has presented results quite similar to the monitoring data of the literature, therefore it can be stated that the model can be considered a suitable tool for this type of studies. As aforementioned, application of the QWASI model resulted in an estimation of the most important pathways to water and sediment, which were the emissions to water and – in the case of lead – also to air. As this model is focussing on local scale and the distribution of substances between the compartments air, water and sediment, its results could be more accurate – depending on the data base – but are also limited to the quatic scenario. Finally, the 2-FUN model provided data (lead level in blood) useful to perform a detailed risk assessment, by linking the environmental concentrations to the human body .Bearing in mind the foresaid advantages and drawbacks, it is strongly believed that the combination of all the strengths of the selected models has provided an interesting picture of the case study situation.

7. Acknowledgments

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