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ABSTRACT BOOK

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Human Health Risk Assessment Due to Agricultural Activities and Crop Consumption in the Surroundings of an Industrial Area

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The aim of the present work was to integrate data of potentially toxic elements (PTE) (As, Cd, Cr, Cu, Hg, Ni, Pb and Zn) from different environmental compartments (soil and crops) with data of PTE in human hair in order to assess the quality of agricultural soil and crop plants and the risks to human health from different exposure routes, and to verify the possible relationship between environmental data and biomonitoring data. The gardens housing of the surrounding fields of the industrial chemical complex of Estarreja (ECC) municipality, central Portugal, were selected as the study area. The analysis of agricultural soil, tomato (*Lycopersicon esculentum Mill*.), potato (*Solanum tuberosum*, L.) and cabbage (*Brassica oleracea*, L.), and the estimation of cancer and non-cancer risks showed high health risks for the local population, particularly for the children, due to the exposure to As and Hg. The higher PTE levels in soils and horticultural crops are mainly related to historical industrial activities, mostly from arsenopyrite roasting and a chloralkali plant. Agricultural soils in the ECC surrounding presented high concentrations of As, Cu, Hg and Pb. The assessment of risks to human health for the ECC surrounding residents (children 4-8 years old; elderly adults >55 years old) showed that agricultural soil-dust ingestion induces a high non-carcinogenic risk for As, Pb and Hg and carcinogenic risk for As. Exposure through consumption of tomatoes and potatoes grown in the study soils does not present a high health hazard. However, exposure to As through consumption of cabbage present both carcinogenic and non-carcinogenic health risks for both studied age groups. It is likely that hair As and Hg concentrations, increased either in children and adults, can be related to the ingestion of agricultural soil and cabbage and inhalation or dermal contact with contaminated soil. Yet, this assumption requires further investigation, including on other potential sources of PTE for the local population. Hair Cr content in the adult group of residents showed maximum values above the normal range for non-exposed individuals, as well as high mean and median values, which may be related to the high Cr content in the studied foods. The results of the present study are consistent with the results of other similar studies regarding As and Hg concentrations in the hair of children and adults. Thus, the present study corroborates the use of As and Hg content in hair as biomarkers of local environmental exposure.

Human health risk assessment due to agricultural activities and crop consumption in the surroundings of an industrial area

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Aims

- To integrate potentially toxic elements (PTE: As, Cd, Cr, Cu, Hg, Ni, Pb and Zn) data from different environmental compartments (soil, crops) and human hair in order to assess agricultural soil and vegetable quality and the risks to human health arising from different exposures pathways.
- To check for eventual relationship between environmental data and biomonitoring data.

Overview of the study area

The study area is located in the surroundings of the Estarreja Chemical complex (ECC) - Figure 1A and B), which is a chemical-industrial complex located about 3 km to the north of the city of Estarreja.

Surrounding populations historically rely on groundwater as a source of water supply for human and agricultural uses.

The ECC has been in operation since the 1930s. Nowadays the main companies within the ECC produce the following contaminants: polyvinyl chloride (CIRES); nitrobenzene, aniline, cyclohexylamine, cyclohexanol, sulphuric acid, nitric acid and sulfanilic acid (CUF), polymeric methylene diphenyl diisocyanate (DOW CHEM); pure and mixed gases for industrial and medical uses (Air Liquide); and, aluminium sulphates and polyaluminum chlorides in liquid form (AQP).

The production of these chemicals within the ECC requires the use of a long list of potentially hazardous substances, which have left their environmental legacy in the surrounding environment. In the past, liquid effluents containing aniline, benzene compounds, NO_3, Cl, Al, As, Cd, Cr, Hg, Ni, Zn and Pb were discharged without any previous treatment into manmade permeable water channels, contaminating agricultural fields, surface water and groundwater. Solid wastes comprised sludges containing pyrite, calcium hydroxide, Hg and As were deposited in permeable areas with no pre-treatment.

Despite the rehabilitation actions taken during the last 20 years, the footprint of the ECC industrial activity may still be identified in impacted air, soils, sediments, surface and groundwater quality around the ECC.

Materials and methods

Sampling and analysis of soils and crops: A total of 26 composite soil samples, as well as 25 tomato plants, (*Lycopersicon esculentum Mill.*) 4 potato plants of (*Solanum tuberosum, L.*) and 26 cabbage plants (*Brassica oleracea, L.*) were collected (Figure 1B) in kitchen gardens/small farms, in the vicinity of the ECC. Soil samples were air-dried, sieved < 2 mm (for soil quality assessment) and < 0.063 mm (for human risk assessment) and PTE were analysed for these two grain size. The determination of PTE in both soil and plant samples was performed by Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-AES) and Inductively Coupled Plasma-Mass Spectrometry (ICP-MS).

Hair sampling and analysis: The biomonitoring program was conducted in two groups of permanent residents: > 55 years old, n=88 subjects; 4-8 years old, n= 39 subjects) living in or around the city of Estarreja. Hair trace elements levels were determined through ICP-MS technique using a Thermo Fisher Scientific iCAP™ Q instrument (Thermo Fisher Scientific, Walthman, MA, USA) at the Trace Elements Analysis unit (Laboratory of Applied Chemistry), Faculty of Pharmacy, University of Porto.

Human Health Risk assessment: The non-carcinogenic and carcinogenic risks were estimated according to the United States Environmental Protection Agency (USEPA 2011) methodology. The human health risk assessment was calculated assuming that both children (4-8 years old) and adult (> 55 years old) groups were directly exposed to PTE by soil (particle size < 63 µm) and through crop consumption (tomato, potato and cabbage).

The HI (the chronic hazard index) was calculated as the sum of the hazard quotients for multiple exposure pathways. When HI values >1, there is a chance that non-carcinogenic risk may occur; otherwise, i.e., if HI < 1 the individuals are exposed to concentrations that do not present a hazard. The carcinogenic risks (CR) for As, Cd, Cr and Ni exposure of the studied groups were calculated according to the Exposure Factors Handbook (USEPA 2001, 2011) and using the Slope Factors according to USDE (2013). The acceptable values for human cancer risk proposed by USEPA (2011) range between 1×10^{-4} to 1×10^{-6}.

Results

1. Agricultural soils in the ECC surrounding showed high concentrations of some PTE, such as As, Cu, Pb and Hg (Table 1).
2. As, Cd and Hg contents in some cabbage samples were higher than the maximum limits (ML) established in the EU Commission Regulation (CE) No 1881/2006 and by the Ministry of Health of the People's Republic of China (2005) (Table 2).
3. Except for Hg and As, the PTE mean contents in adult hair samples obtained in this study (with an environmentally exposed population) were within the range of values reported for non-exposed people. For Hg and As, both adult and children showed hair levels higher than the maximum values reported in other studies (Table 3).
4. The assessment of the health risks for the ECC surrounding residents (children with 4-8 years old and adults >55 years) showed that agricultural soil/dust ingestion is an important exposure pathway for both age groups (Table 4).
5. Direct exposure to the studied soils presents a high non-carcinogenic risk for As, Pb and Hg and carcinogenic risk for As. Exposure through ingestion of tomato and potato grown in the study soils does not present a high health hazard. However, exposure to As through consumption of cabbage presents either carcinogenic and non-carcinogenic health risks for both age groups (Table 4).
6. These results are in accordance with the high levels of As in adults and children hair (Table 3).
7. The assessment of health risks for ECC surrounding residents showed that the elements of most concern among those studied are As and Hg, regarding both carcinogenic and non-carcinogenic risk.

Conclusions

- The inter-disciplinary approach applied in this study was successful in identifying links between different datasets.
- The exposure study results are in agreement with As and Hg concentrations in both children and adults hair and validate it as a biomarker of As and Hg local environmental exposure.
- Nevertheless, further studies should be carried out with other local crops that may be sources of exposure.
- The prevention from contamination to avert the negative effects on human health is necessary. The development of site-specific, farmer-driven strategies along with policy support for effective soil decontamination in the most critical locations should be considered.

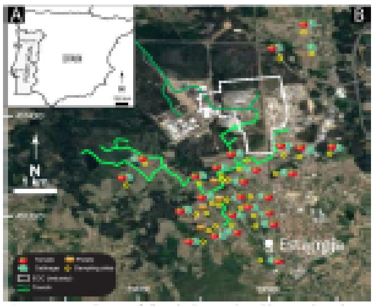

Figure 1 – Study area (A) including soil and horticultural crops sampling locations (B)

Table 1 – Descriptive statistics for PTE content (mg/kg) in soil samples (F1 fraction = grain size < 63 µm; F2 fraction = grain size < 2 mm).

[illegible]	[illegible]	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
F1	Minimum	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
	Mean	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
	Median	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
	Maximum	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
	SD	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
F2	Minimum	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
	Mean	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
	Median	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
	Maximum	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
	SD	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
	PRV	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
	CEQG	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
	% samples > PRV	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
	% samples > CEQG	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]

PRV – Portuguese Reference Values for soils (median value) [illegible]. CEQG – Canadian Environmental Quality Guidelines (mg/kg) for agricultural land use [illegible], for grain size < 2 mm.

Table 2 – Descriptive statistics for PTE content (µg/g) in fresh weight tomato, potato and cabbage.

[illegible]	[illegible]	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Tomato: Fresh weight (µg/g)	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
ML		[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
% of samples > ML		[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
Potato: Fresh weight (µg/g)	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
ML		[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
% of samples > ML		[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
Cabbage: Fresh weight (µg/g)	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
ML		[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
% of samples > ML		[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]

[illegible]

Table 3 – Descriptive statistics for PTE content (µg/g) in adults (n= 88) and children (n=39) hair and Rodushkin and Axelsson (2000) data.

	This study (adults)				This study (children)				Rodushkin and Axelsson (2000)			
	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
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[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]

Table 4 – Range (minimum and maximum) of Hazard Indexes (HI) for the different PTE and Cancer Risks (CR) due to agricultural soil exposure and due to cabbage consumption.

	HI (soil)		CR (soil)		HI (cabbage)		CR (cabbage)	
	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]		
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]		
[illegible]	[illegible]	[illegible]			[illegible]	[illegible]		
[illegible]	[illegible]	[illegible]			[illegible]	[illegible]		
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]		
[illegible]	[illegible]	[illegible]			[illegible]	[illegible]		
[illegible]	[illegible]	[illegible]			[illegible]	[illegible]		

Note: Maximum values that are above the target values are in bold

References: Canadian guideline: Minister of the Environment (Canada). Soil, Ground Water and Sediment Standards for Use under Part XV.1 of the Environmental Protection Act. ¶ Inácio M, Neves O, Pereira V, Ferreira da Silva E (2014) Levels of selected potential harmful elements (PHEs) in soils and vegetables used in diet of the population living in the surroundings of the Estarreja Chemical Complex (Portugal). Appl Geochem. 44: 38-44. ¶ Ministry of Health of the People's Republic of China (2005) the maximum levels of contaminants in foods (GB 2762-2005). Maximum Levels of Contaminants in Foods. _Beijing_China. ¶ USEPA (2011) United States Environmental Protection Agency. Exposure factors handbook 2011 edition. ¶ Rodushkin I, Axelsson MD (2000) Application of double focusing sector field ICP-MS for multielemental characterization of human hair and nails. Part II. A study of the inhabitants of northern Sweden. Sci Total Environ 262(1-2): 21-36.

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