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Heavy metal content and health risk assessment of urban road dust from the historical center of Havana, Cuba

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Abstract

Street road dust samples were collected at 33 stations of the Historical center of Old Havana. Samples were analyzed for the determination of some (Co, Ni, Cu, Zn and Pb) heavy metal concentrations using an X-ray fluorescence (XRF) instrument. The mean concentrations of the elements are Zn (548.7 mg/kg), Pb (60.8 mg/kg), Co (7.0 mg/kg) Ni (60.6 mg/kg) and Cu (73.6 mg/kg) were Zn and Pb showed higher values of the background. The study of the Enrichment Factor (*EF*), the Pollution Index (*IP*) and the Integral Pollution Index (*IPI*), showed some stations with high values of contamination close to construction places. Zn was identified as the major pollutant followed by the Pb. Nevertheless, the Hazard Index (*HI*) and Carcinogenic Risks (*CR*) values do not show risk of getting non-carcinogenic or carcinogenic diseases associated to the analyzed pollutants.

Keywords

Heavy metals, XRF, urban road dust, pollution, enrichment factor, health assessment

Ethics approval and consent to participate

Not Applicable

Consent for publication

Not applicable

Availability of data and materials

The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

Competing interests

The authors declare that they have no competing interests

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Authors' contributions

All authors read and approved the final manuscript. The authors contributed to the present work as follow:

ODR (Leader, Data Analysis, Writting manuscript)

AOCD (Data Analysis, Writting manuscript)

AGTR (Data Collection, Data Analysis)

DRL (Data Collection, Data Analysis)

Introduction

Cities are the centers with the greatest amount of anthropogenic activities. Due to rapid urbanization and industrialization, about half of the world population now lives in cities, where the population density, the automotive traffic, industry and economic activities, cause large amounts of pollutants that are discharged into the urban environment. So a lot of environmental problems are arising from air, soil and water pollution. Heavy metal contamination is a major problem, especially in urban atmospheric particles, soil and dust (Lu et al. 2014). The main pollutants associated with urbanized areas are: Copper (Cu), Lead (Pb) and Zinc (Zn) (Rissler et al. 2012), together with these can be found: Nickel (Ni), Cadmium (Cd) and Chromium (Cr), which appear as a product of industrial activities (Garcia et al. 2014), vehicle emissions, particularly near areas with high traffic (Ordóñez et al. 2015, Charlesworth et al. 2011), construction and restoration processes as well as natural geochemical processes.

Heavy metals in street dust can have a direct influence in public health because they can easily enter the human body by ingestion, skin contact, or breathing. (Maltby et al. 1995; Sutherland and Tolosa, 2000; Wei and Yang, 2010). Some of these metals, like Pb, Co, Cd, Cu and Cr are considered dangerous pollutants that accumulate in the human body with a relatively high half-life (Salt et al. 1995). Furthermore, some types of Cadmium, Chromium and Copper may be associated with health effects from dermatitis to various types of cancer (Das et al. 1997; Onder and Dursun, 2006). Consequently, the geochemical characterization of soil and dust contamination is an important contribution to the characterization of the urban environment and the potential risks associated with health.

The Historical Center of Havana city (HCH), capital of Cuba, is characterized by the wide development of the restoration activities of its dissimilar historical sites, monuments and tourist facilities, including some demolitions, especially before the 500th anniversary of the city in 2019. The last implies the use of large amounts of construction materials and machinery representing important sources of heavy metal emissions. Furthermore, the HCH is surrounded by high traffic avenues. In this sense, the goal of the present study was to determine the heavy metal content in urban dust in order to evaluate its impact to the HCH environmental pollution as well as health risk impact to the local population.

Methods and Materials

The HCH is nearly the 50% of the Old Havana municipality area although the 66.1% of its habitants lives there with 70658 habitants (around 33018 habitants.km⁻²). At the north it's limited by the see, at the east by the Havana bay while at the south and west by the rest of the municipality's territory.

Dust samples were collected at 33 stations homogeneously distributed through the studied area (Figure 1). Dust samples (around 100g) were collected by gently sweeping an area of about 16 m² in selected stations homogeneously distributed trying to be close to parks, high traffic avenues, schools, constructions, residential areas and shops. All samples were transferred to a clean, self-sealed polyethylene bag. In the laboratory, large rock, metallic and plastic pieces and organic debris were removed before sieving at a fraction smaller than 63 µm. Then the powder samples were dried at 35 °C until obtaining a constant weight.



Figure 1. Location of the sampling stations in Historical Center of Havana

The heavy metal concentrations were estimated by X-Ray Fluorescence Analysis (XRF) using the Certified Reference Materials (CRM) IAEA-Soil-7, MAG-1 "Marine sediment", IAEA SL-1 "Lake sediment", IAEA Soil-5, IAEA-356 "Polluted marine sediment", BCSS-1 "Marine sediment", BCR-2 "Basalt Columbia River" and SGR-1 "Green River Shale". All samples and CRM were mixed with cellulose (analytical quality) in proportion 4:1 and pressed at 15 tons into the pellets of 25 mm diameter and 4-5 mm height. All pellets were measured using Canberra Si(Li) detector (150 eV energy resolution at 5.9 keV, Be window thickness = 12.0 μm) coupled to MCA. A ^{238}Pu (1.1 GBq) excitation source with ring geometry was used. All spectra were processed with WinAxil code (WinAxil, 2005).

The accuracy of the quantification was verified using the McFarrel criterion (Quevauviller and Marrier 1995) in which an SR parameter is defined as $= \frac{|C_{exp} - C_w| + 2\sigma}{C_w} * 100\%$, where, C_{exp} is the experimentally determined concentration, C_w is the concentration reported for the measured CRM and σ is the standard deviation of C_{exp} . Based on this criterion, the similarity between the reported value and the data analytical concentration, obtained by a particular method, is divided into three categories: if $SR \leq 25\%$, the method is considered excellent for analysis; $25\% \leq SR \leq 50\%$, it is considered acceptable and for $SR \text{ values} > 50\%$, the method it is considered unacceptable for quantification. The XRF analysis of CRM IES-951 (Table 1), shows an excellent quality with $SR < 25\%$ for all the analyzed metals and the obtained results shows a very good correlation between certified and measured values.

Table 1. Analysis of the CRM IES-951 using ED-XRF (Concentration \pm SD, mg.kg^{-1} , except indicated), SR values and Detection Limits (Díaz Rizo et al. 2011).

Element	Calculated Concentration	Reported Concentration	SR (%)	L_D (mg Kg^{-1})
Fe (%)	3.13 ± 0.11	2.97 ± 0.09	12	9
Co	12.5 ± 1.42	12.5 ± 1.44	23	6
Ni	37.3 ± 3.2	37.9 ± 2.82	19	7
Cu	22.7 ± 0.86	20.1 ± 2.31	22	6
Zn	102.5 ± 5.5	96.1 ± 5.8	18	5
Pb	33.3 ± 2.55	36.9 ± 4.01	24	4

Spatial distribution maps of heavy metal concentration of interest present in the urban dusts of the HCH, were built by the Surfer graphing and calculation software (Surfer, version 10.1.561, 1993-2011) using the Kriging algorithm.

The degree of contamination present in dust samples was evaluated using the dimensionless variable denominated Enrichment Factor (EF). EF is estimated as: $EF = (C_n / C_{ref})_{sample} / (C_n / C_{ref})_{background}$,

where C_n is the concentration of the potentially enriched metal and C_{ref} is the reference metal concentration. An $EF > 1$ means the existence of a non-natural contribution to the concentration determined of an element. The degree of enrichment depending on the EF value according to Sutherland (2000) is: $EF < 2$, minimal enrichment; $2 < EF < 5$, moderate enrichment; $5 < EF < 20$, significant enrichment; $20 < EF < 40$, very high enrichment; and $EF > 40$, extremely high enrichment.

Generally, EF less than 2 it is not considered significant, since a small value of EF may be due to fluctuations relative to the used natural background, or for the accuracy and precision of the analytical technique used (Sinem Atgin et al. 2000). In this research, iron was used as reference metal for the normalization as it is hardly influenced by anthropogenic sources due to the high natural levels of this element (Villares et al. 2003).

Additionally, the dust contamination level was evaluated using the Integral Pollution Index (IPI) (Cheng et al. 2005; Sun et al. 2010), which is defined as the average value of all the values of the Pollution indexes for the metals considered $IPI = \frac{1}{N} \sum_{n=1}^N PI_n$, where N is the number of metals considered in the study. The Pollution Index (PI) is defined as the ratio of the concentration of the heavy metal in the studied dust and the geometric mean of the metal concentration of soil taken as background $PI_n = C_n/B_n$, where C is the concentration of the metal to be determined and B the concentration in the soil taken as background (Cheng et al. 2005). The classifications of pollution for these indexes according to Hakanson (1980) are: ($PI < 1$; $IPI \leq 1.0$) Slightly contaminated; ($1 \leq PI < 3$; $1 < IPI \leq 2$) Moderately contaminated; ($3 \leq PI < 6$; $2 < IPI \leq 5$) Considerably contaminated; ($6 \leq PI$; $5 < IPI$) Highly contaminated.

In both cases, EF and PI indexes, the heavy metal content reported for non-urbanized soils of Havana city (Díaz Rizo, et al. 2011) was used as background metal content.

The calculation of the health risk of exposure to heavy metals is based on the studies carried out by the United States Environmental Protection Agency (USEPA 1986; USEPA 1989; USEPA 2002). Health risks can be divided into two categories: risk of acquiring a carcinogenic disease and risks of acquiring a non-carcinogenic disease. For the non- carcinogenic it is used the Average Daily exposure Doses (ADD , $\text{mg.kg}^{-1}.\text{day}^{-1}$) received estimated through the three ways of incorporation to which people are exposed to heavy metals from urban dust (inhalation, ingestion and dermal contact).

$$ADD_{ing} = C \times \frac{R_{ing} \times FE \times ED}{BW \times T}; ADD_{der} = C \times \frac{SA \times SL \times ABS \times FE \times ED}{BW \times T}; ADD_{inh} = C \times \frac{R_{inh} \times FE \times ED}{PEF \times BW \times T} \text{ and } LADD_{inh} = \frac{C \times FE}{PEF \times T} \left(\left(\frac{R_{inh} \times ED}{BW} \right)_{child} + \left(\frac{R_{inh} \times ED}{BW} \right)_{adult} \right)$$

where, C (mg.kg^{-1}) is the concentration of the metal in the studied dust; R_{ing} is the ingestion rate: 200 mg/day for adults and 100 mg/day for children (USEPA 2002); R_{inh} is the inhalation rate: 20 m^3/day for adults and 7.6 m^3/day for children (Zheng et al. 2010; USEPA 2002); ED is the exposure duration: 24 years for adults and 6 years for children; PEF is the particle emission factor, that is, the relationship between the concentration of a pollutant in the dust with the concentration of particles of these pollutants that are suspended in the air we breathe: $1.36 \times 10^9 \text{ m}^3\text{kg}^{-1}$; SA it is the exposed skin area: 5700 cm^2 for adults and 2800 cm^2 for children; SL is the skin adhesion factor: 0.7 $\text{mg.cm}^{-2}.\text{day}^{-1}$ for adults and 0.2 $\text{mg.cm}^{-2}.\text{day}^{-1}$ for children; ABS is the dermal absorption factor, is a dimensionless value and is 0.001 for all items considered; BW is the average body weight: 70 kg for adults and 15 kg for children; FE is the exposure frequency: 350 days.year^{-1} and T , the average time, is the period of time from which the dose is averaged: $ED \times 365$ days for risks associated with non-carcinogenic diseases and 70×365 days for risks associated with carcinogenic diseases (USEPA 2002). $LADD_{inh}$ is the Lifetime Average Daily Dose used for cancer risk calculations.

The hazard quotient (HQ), which is defined as $HQ = ADD/RfD$ of a chemical element, it is used to calculate the non-carcinogenic risk. To determine the effect of the three forms of exposure for each metal, the Hazard Index (HI) (USEPA 1986) is calculated as $HI = HQ_{ing} + HQ_{inh} + HQ_{der}$. If $HI < 1$, then there will be no risk of acquiring a non-carcinogenic disease, if $HI > 1$, then there is a certain probability of acquiring a non-carcinogenic disease, which tends to increase with the HI value. The used RfD values for each metal are present in Table 2.

Table 2. RfD ($\text{mg.kg}^{-1}.\text{d}^{-1}$) values for non-carcinogenic metals and Slope factors ($(\text{mg.kg}^{-1}.\text{d}^{-1})^{-1}$) for carcinogenic metals (Zhang et al. 2018)

	Co	Ni	Cu	Zn	Pb
RfD_{ing}	0.02	0.02	0.04	0.3	0.0035
RfD_{inh}	0.0000571	0.0206	0.012	0.3	0.00352
RfD_{der}	0.016	0.0008	0.04	0.06	0.000525
SF_{inh}	9.8	0.84	-	-	-

The Ni and Co have the potential to pose Carcinogenic Risk (CR) through respiratory exposure. This is the probability that an individual develops some type of cancer during the exposure time to a carcinogenic risk. It is calculated as $CR_{\text{metal}} = LADD_{\text{inh}} \times SF_{\text{inh}}$ where SF is the slope factor with values are also presented in Table 2. An acceptable or tolerable value of $CR = \sum CR_{\text{metal}}$ is in the range between (10^{-6} - 10^{-4}).

Results and Discussion

The means of the metal (Fe, Co, Ni, Cu, Zn and Pb) concentrations determined by X-ray fluorescence analysis of urban road dusts from the HCH in the different stations are shown in Table 3, as well as the standard deviations, medians, maximums, minimums, background values and the reported values from the calculation of concentration of heavy metals in the upper crust (Hans 1995).

Table 3. Statistical results of the concentration (in mg.kg^{-1} , except the indicated) of analyzed metals, background used and upper crust. (^a (Díaz Rizo et al. 2011) ^b (Hans 1995))

	Fe (%)	Co	Ni	Cu	Zn	Pb
Average	1.2	7.0	60.6	73.6	548.7	60.8
SD	0.3	1.1	218.5	262.8	578.3	70.5
Minimum	0.7	6.0	11.0	16.0	124.0	22.0
Maximum	2.2	10.4	1276.0	1535.0	3012.0	422.0
Median	1.2	6.6	20.0	24.0	358.0	39.0
Average/Median	1.0	1.1	3.0	3.1	1.5	1.6
Background ^a	4.7	14.8	58	83	151	28
Upper Crust ^b	4.32	24	56	25	65	14.8

Standard deviations show large values respect de mean values due to outliers at some stations. Station 24 and 23 show the highest values (3012 and 1750 mg.kg^{-1} respectively) for Zn, this could be because between these stations, it is the largest parking lot in the municipality where there was a workshop where they used to do sheet metal work (using zinc plates), and a punch bowl (ZnO is a rubber compound). It used to be another punch bowl at the same street of station 24. There are also some constructions around where new buildings were made. Zn is used for roofing and as a component of cement and paints. Stations 19 (1170 mg.kg^{-1}) and 6 (1461 mg.kg^{-1}) have also outliers for Zn, presumably due to the large construction sites next to them. In the case of station 6 has also the highest values for Ni and Cu because this construction is a museum in capital restauration where big pieces of bronze (containing Cu), stainless steel (containing Ni) and galvanized steel pipes (containing Ni and Zn) received maintenance.

The highest Pb-content (422 mg.kg^{-1}) was found at station 2. At this place there is small seaport for fisher boats that use oil as fuel and it is the main exit of all the vehicles that cross the Havana bay tunnel. The average / median ratios for Fe and Co are close to 1, so there are not outliers and it can be stated that its origins must be predominantly natural. Figure 2 shows the spatial distribution for the determined heavy metals.

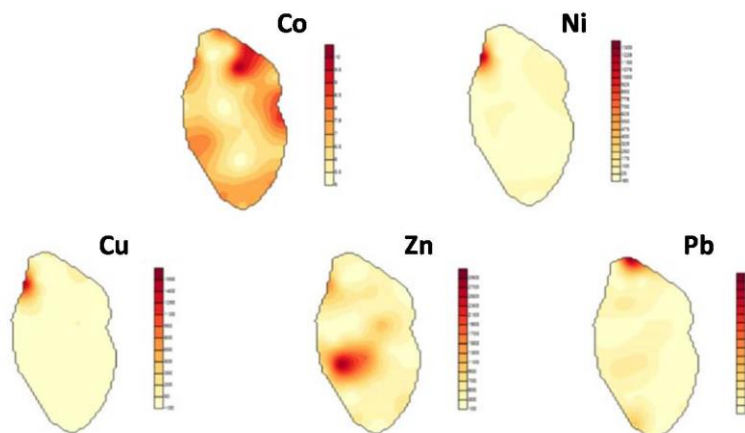


Figure 2. Spatial distribution of the determined heavy metals contents analyzed in the HCH (mg.kg⁻¹)

The average concentration values show a decrease in the form Zn > Cu > Pb > Ni > Co. The determined average/background contents show 3.6 and 2.2 for Zn and Pb, respectively. Referent to the earth's upper crust, Zn and Pb contents show a remarkable excess, while Ni and Cu are closer to the corresponding reference values (Hans 1995). The Pearson correlation of the metal contents determined in urban road dusts of the HCH (Table 4) shows a high Fe-Co correlation indicating some probability of having a common origin. The other metals show a lower correlation.

Table 4. Correlation between determined metals.

	Fe	Co	Ni	Cu	Zn	Pb
Fe	1					
Co	0.74**	1				
Ni	0.23**	0.19**	1			
Cu	0.23**	0.33**	0.06	1		
Zn	0.06	0.03	-0.03	-0.02	1	
Pb	0.25**	0.07	0.01	0.14*	0.25**	1
Signification levels *p<0.05 , **p<0.01						

Table 5 shows the comparison between the metal content determined in the urban dusts from the HCH with contents reported for other Cuban cities, as well as contents reported for historical centers worldwide. Zn concentration levels are most notable for above the other places, although for the rest of the metals these values are generally in the order of the reported concentrations. On the other hand, when comparing them with the soil quality reference values (QRV) recommended for Cuba (Alfaro et al.2015) it is observed that the concentrations of Co, Ni and Cu are lower, Pb content it is slightly higher, while Zn content significantly exceeds the Zn QRV value at 6.4 fold.

Table 5. Average metal concentrations (in mg.kg⁻¹, except the indicated) in the HCH road dust and its comparison with similar studies in other Cuban and worldwide cities. (^aPresent work, ^b(Díaz Rizo et al. 2015), ^c(Díaz Rizo et al. 2019), ^d(Wei et al. 2015), ^e(Bretzel et al.2014), ^f(Elik 2003), ^g(Christoforidis & Stamatis, 2009), ^h(Divrikli et al. 2003), ⁱ(Bourliva et al.2016), ^j(Alfaro et al. 2015))

City	Co	Ni	Cu	Zn	Pb
HCH, Cuba ^a	7.0±1.1	60.6±218.5	73.6±262.8	548.7±578.3	60.8±70.5
Camaguey, Cuba ^b	15±4	66±36	36±12	222±84	63±13
Havana, Cuba ^c	NR	68±131	65±152	369±375	88±114
Beijing, China ^d	NR	26±6	86	272	235
Pisa, Italy ^e	NR	91.9±1.3	107±2	327±1	207±4
Sivas, Turkey ^f	NR	81±4	107±6	284±12	325±10
Kavala, Greece ^g	NR	68±48	172±88	355±115	387±245

Yozgat, Turkey ^h	24±4	51±9	38±3	NR	69±13
Thessaloniki, Greece ⁱ	NR	89.4±44.6	662±348	453±224	209±91
QRV ^j	25	170	83	86	50

The EF values for the analyzed metals are presented in Figure 3. The average enrichments resulted: $EF_{Co} = 1.9$; $EF_{Ni} = 3.5$; $EF_{Cu} = 3.0$; $EF_{Zn} = 13.7$ and $EF_{Pb} = 8.5$. According to the Sutherland (2000), Co presents a minor enrichment. Ni and Cu are found in the moderate enrichment level with an outlier at station 6 that move the average to this region. Pb and Zn have significant enrichments. The highest value of $EF_{Zn}=61.6$ was determined at station 24 as expected due to his high Zn concentration at this station close to the former car bodyshop, punchbowls and constructions. In Cuba, leaded fuels are not used since the end of the 90s (Montero et al. 2017). However, the use of Pb-doped fuels for many years, must be the reason of the significant EF level for Pb in the city since lead exhausts could have been accumulated in parks, walls and streets for years.

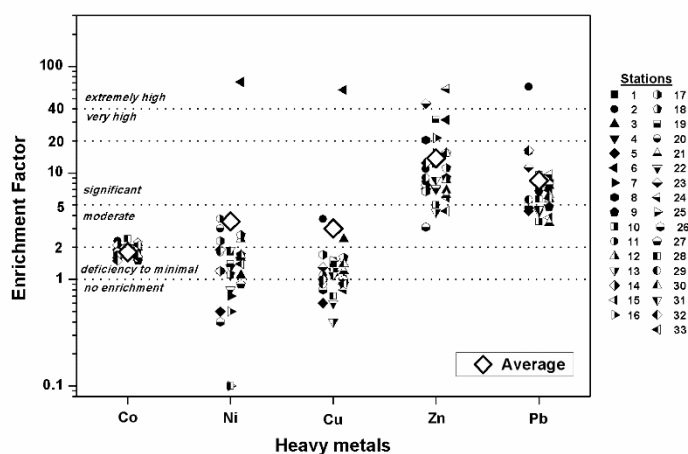


Figure 3. Enrichment Factor values for the metals determined in the urban dust of the HCH.

The average value, over all the stations, of the PI and the IPI is presented in Figure 4. The $PI_{Co} = 0.5$ and $PI_{Cu} = 0.9$ indicate a low contamination, $PI_{Ni} = 1$ and $PI_{Pb} = 2.2$ are at a moderate level and $PI_{Zn} = 3.6$ presents considerable contamination. However, there are some stations with high pollution, as in the case of station 6 ($PI_{Ni} = 22$, $PI_{Cu} = 18.5$), 23 ($PI_{Zn} = 11.6$), 24 ($PI_{Zn} = 19.9$) and 2 with $PI_{Pb} = 15.5$. According to the IPI , all the stations show low to moderate contamination except stations 2, 19, 23 and 24 with considerably contamination and station 6 with extremely high contamination.

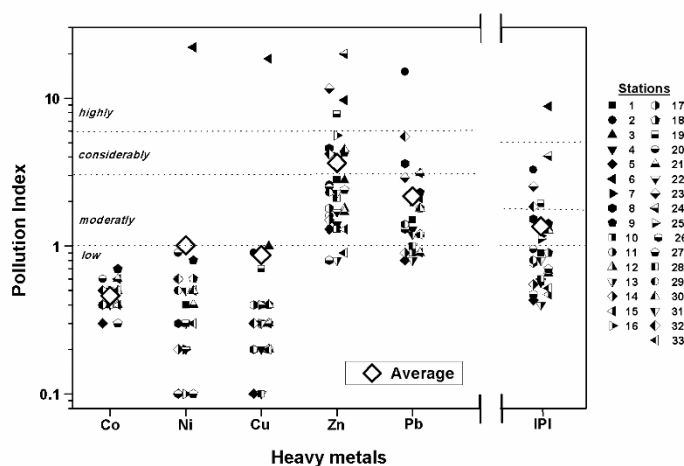


Figure 4. Pollution Index values for the metals determined in the urban dust of the HCH and the Integrated Pollution Index.

Independently of the mentioned high metal enrichments, the calculated metal Hazard Indexes for children and adults for all studied stations are less than unity (Figure 5), indicating no risk for non-carcinogenic diseases. Additionally, the probability to develop carcinogenic disease for each carcinogenic metal as well as the total are under tolerable range, indicating no risk of getting a carcinogenic disease. These values indicate that the heavy metals analyzed do not represent a health problem.

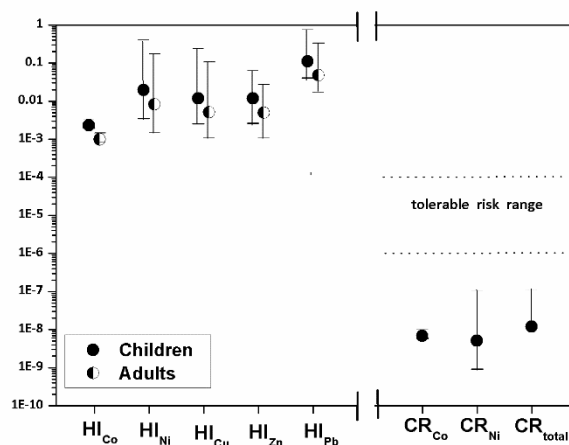


Figure 5. Hazard Index averages and ranges of values for children and adults and the Carcinogenic Risk

Conclusions and Recommendations

Heavy metal content in urban road dust from the historical center of Havana show the behavior $Zn > Cu > Pb > Ni > Co$, with Zn and Pb averages higher than quality reference values recommended for Cuba.

The *EF* and *PI* show the same behavior ($Zn > Pb > Ni > Cu > Co$) with almost all the stations in the range of moderate or less enrichment and pollution for Co, Ni and Cu. Pb has a significant enrichment but only three stations show considerably or more pollution index. Zn shows a significant or higher enrichment in almost all the stations and in correspondence the pollution index goes from moderately to highly pollution.

Health risk assessment shows that *HI* and *CR* are in safe ranges. That confirm that the HCH city is a safe place for visitors and habitants independently the great reconstruction movement performed in the last years.

However, taking into account that the reconstruction movement will continue in the next years, the check of the heavy metal content will be continued as well as the health risks assessments in order to guaranteed safe levels.

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Figures



Figure 1

Location of the sampling stations in Historical Center of Havana Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

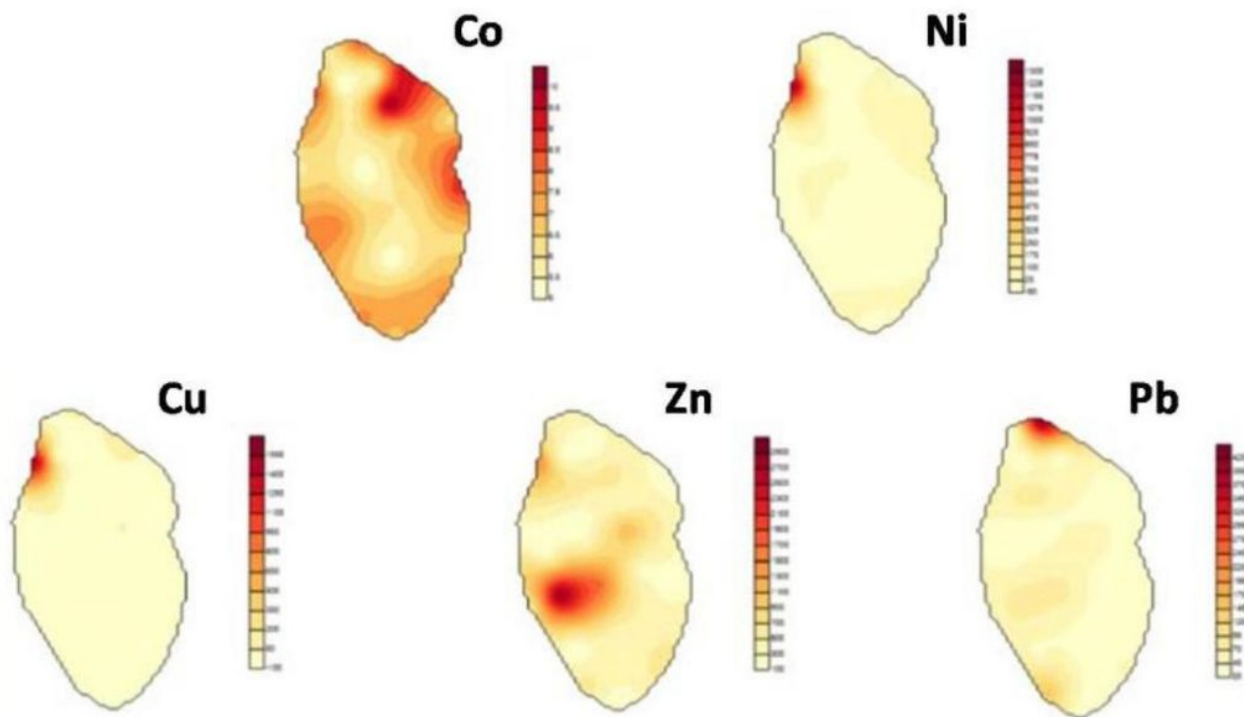


Figure 2

Spatial distribution of the determined heavy metals contents analyzed in the HCH (mg.kg⁻¹)

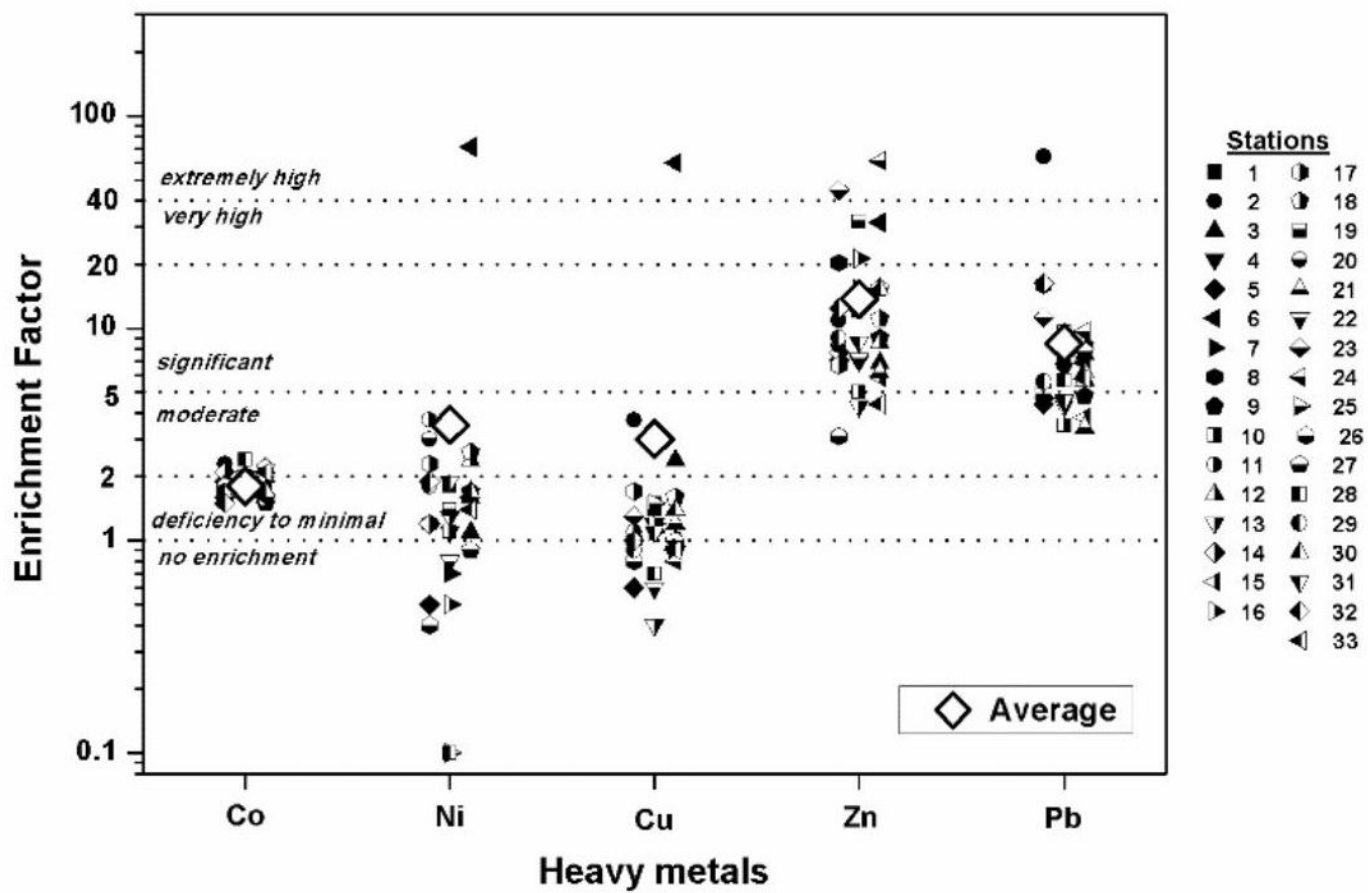


Figure 3

Enrichment Factor values for the metals determined in the urban dust of the HCH.

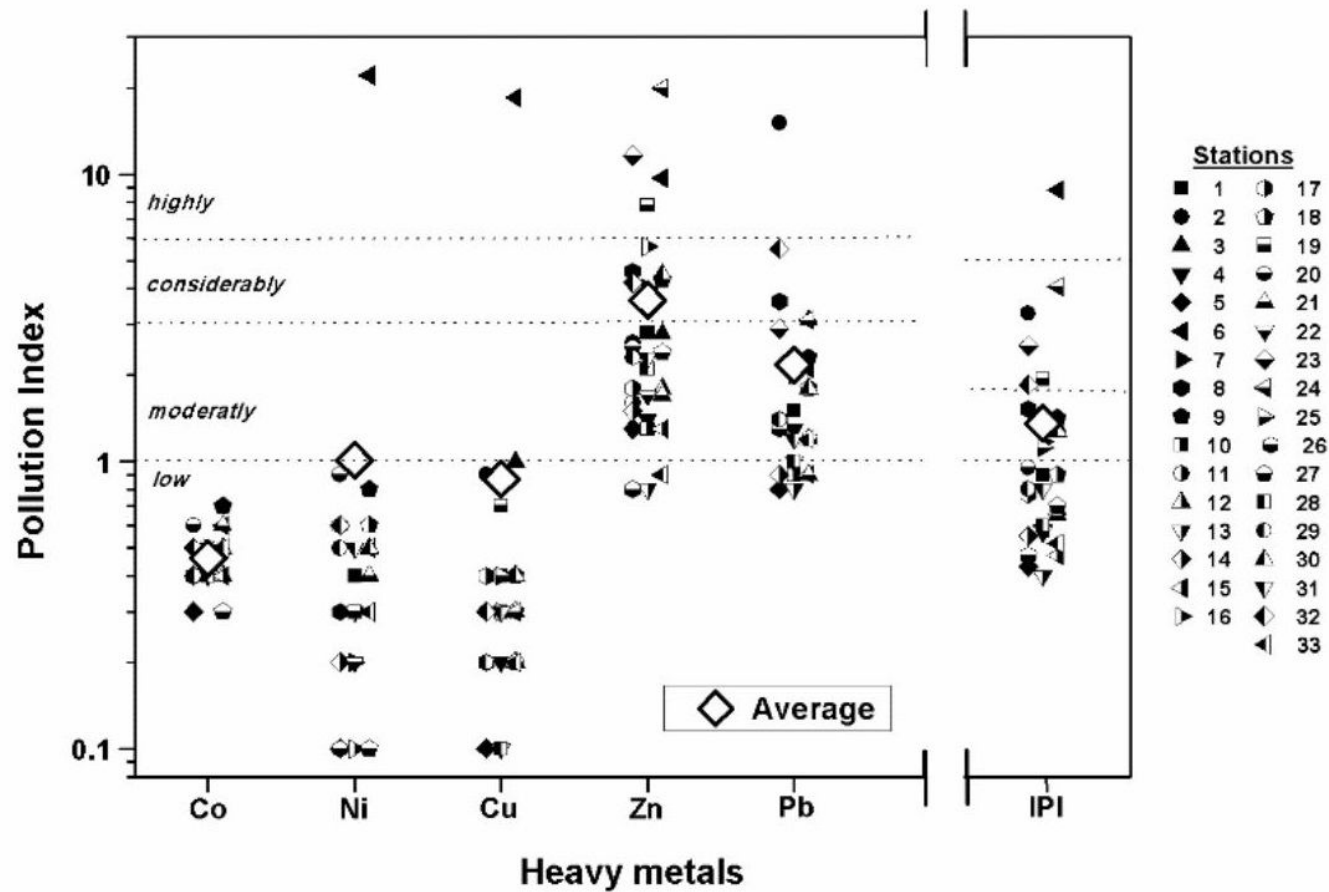


Figure 4

Pollution Index values for the metals determined in the urban dust of the HCH and the Integrated Pollution Index.

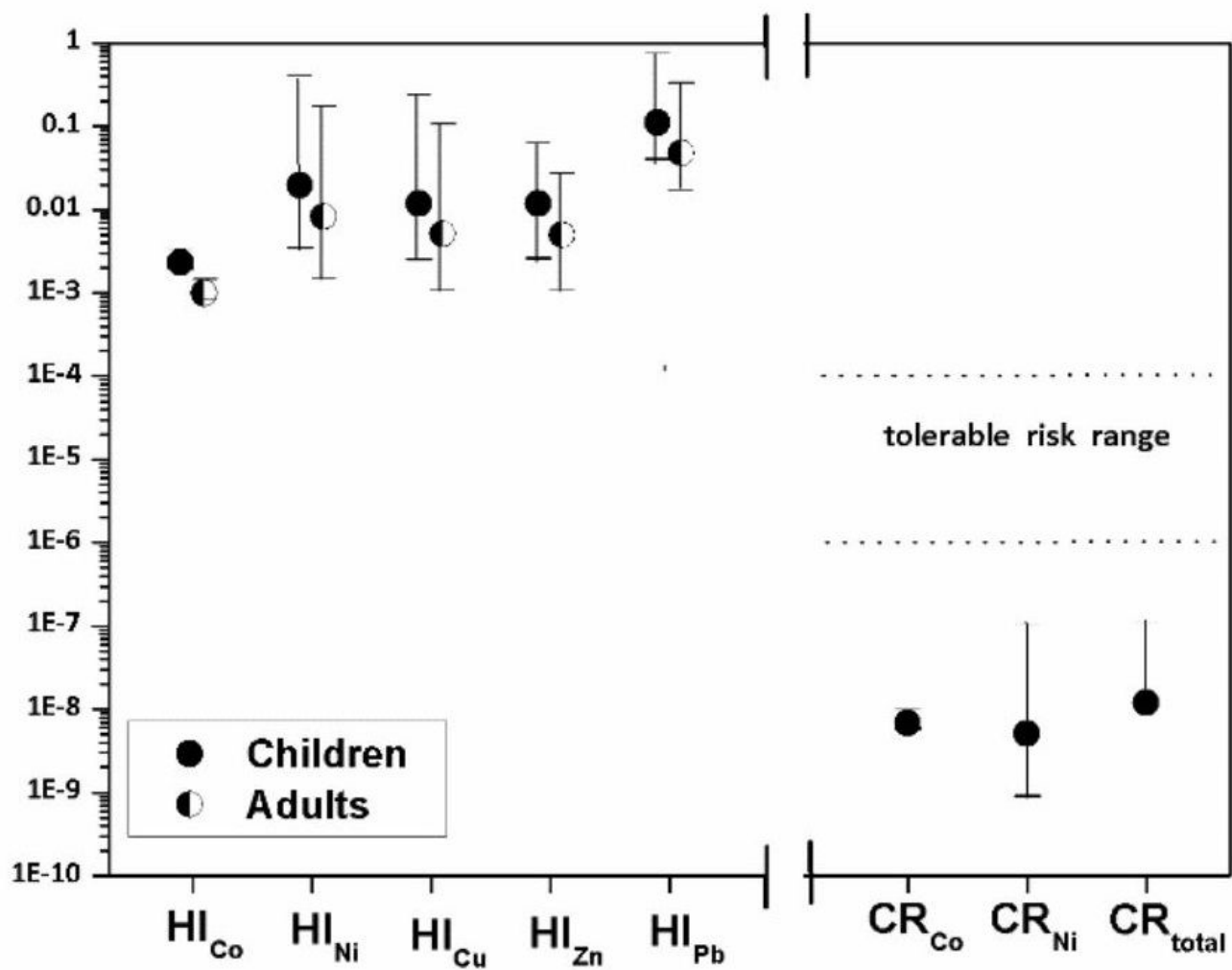


Figure 5

Hazard Index averages and ranges of values for children and adults and the Carcinogenic Risk