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Possible Sources of Exposure to Toxic Elements (As, Cd, Pb) in Kinshasa's Population, Democratic Republic of Congo.

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ABSTRACT

Arsenic (As), Cadmium (Cd) and Lead (Pb) are ubiquitous and some of the most common toxic elements to which Kinshasa's population are exposed. But, the exact sources of this exposure are unclear and/or little known. This study aimed at investigating the possible sources of the exposure to these toxic elements in the population of Kinshasa. In total, 25 samples of drink and 100 samples of soil were collected in the 4 sampling locations. The concentrations of 3 elements were determined by inductively coupled plasma mass spectrometry (ICP-MS). Careful control was applied during collection, handling and analyses of the samples to avoid any contamination. Globally, the results indicate that drink and soil are not significant sources of exposure to As, Cd and Pb in the Kinshasa's population. These new data will serve decision makers and/or health professionals and future research. The emphasis should shift to understand the different sources of As, Cd and Pb emissions in ambient air (low fuel quality, congested traffic; wood, charcoal, crop, residual oil, and waste residues combustion for heating and cooking in open fires, etc.) and to examine the contributions of food sources (vegetables, fish, meat, etc) to urine or blood levels of As, Cd, and Pb in the Kinshasa's population.

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INTRODUCTION

Arsenic (As), cadmium (Cd), and lead (Pb) are elements ranked among the top ten most toxic substances by the US Agency for Toxic Substances and Disease Registry [ATSDR 2017]. Evidence suggests that each of these elements is associated with several adverse outcomes: As exposure may affect all organ systems including skin lesions, cardiovascular, nervous, hepato-billiary, renal, gastro-intestinal, and respiratory systems effects [TCHOUNWOU et *al.*, 2003; TUAKUILA et *al.*, 2012]. The literature has also pointed to significantly higher standardized mortality rates for cancers of the bladder, kidney, skin, and liver in many areas of arsenic pollution. The severity of adverse health effects is related to the chemical form of As, and is also time- and dose-dependent [TCHOUNWOU et *al.*, 2002; YEDJOU et *al.*, 2006; IARC, 2012]. NORDBERG and NORDBERG [1987] and NORDBERG et *al.* [2015] showed that Cd is a toxic metal that accumulates almost irreversibly (half-life > 15 years) in the body and particularly in the renal tubular cells and its prolonged exposure by inhalation or ingestion can cause kidney damage and bone demineralization and fractures. Cd and its compounds have also been classified as human carcinogens that can cause cancer of the lung by inhalation [IARC, 2012]. Pb is a major worldwide public health concern, given the high levels of environmental contaminations and the severe and long term neurotoxic effects [CDC, 1991; LIDSKY and SCHNEIDER, 2003; CANFIELD et *al.*, 2003; LANPHEAR et *al.*, 2005; BELLINGER, 2008; TAYLOR et *al.*, 2011; TUAKUILA et *al.*, 2016].

Findings from previous studies conducted in Kinshasa showed that living in Kinshasa was associated with elevated levels of these elements (As, Cd, Pb) in urine as compared to the reference values in databases involving American, Canadian, French or German populations [N = 220, 6 - 70 years; GM of AsT (total As) = 39.20 µg/L (35.20 - 43.60); GM of Cd = 0.46 µg/L (0.42 - 0.51); GM of Pb = 5.56 µg/L (5.04 - 6.13)]. And these elements reach levels of public health concern [TUAKUILA et *al.*, 2012]. In a case-control study between preeclampsia and toxic metals, ELONGI et *al.* [2016] observed that the urinary concentrations of these elements were substantially higher than reference values for adults from industrially developed countries [GM of As : Control (N = 88) 26.8 µg/L (13.4 - 51.6) vs Preeclamptic women (N = 88) 46.9 µg/L (26.1 - 79.9), $p < 0.001$; GM of Cd : Control (N = 88) 0.53 µg/L(0.29 - 0.68) vs Preeclamptic women (N = 88) 1.78 µg/L(0.71 - 3.83), $p < 0.001$; GM of Pb Control (N = 88) 7.98 µg/L(3.14 - 13.8) vs Preeclamptic women (N = 88) 71.5 µg/L(8.89 - 398), $p < 0.001$]. The daily urinary excretions of these elements were significantly higher in women with preeclampsia than in control women [GM of As : Control (N = 88) 30.5 µg/day (15.0 - 59.3) vs Preeclamptic women (N = 88) 40.0 µg/day (23.3 - 78.7), $p = 0.05$; GM of Cd : Control (N = 88) 0.61 µg/day (0.32 - 0.78) vs Preeclamptic women (N = 88) 1.51 µg/day (0.59 - 2.73), $p < 0.001$; GM of Pb Control (N = 88) 9.09 µg/day (3.15 - 20.5) vs Preeclamptic women (N = 88) 60.9 µg/day (8.83 - 3945), $p < 0.001$]. A significant interaction was found between season and preeclampsia for these elements, with higher urinary excretions in preeclamptic women than controls during the dry season, but not during the rainy season.

Human exposures to these toxic elements may result from diet, air, drinking water, occupational exposures, and/or tobacco use. In Kinshasa, the exact sources of this exposure are unclear and/or little known. Through a study carried out to determine the levels of selected trace elements in the Kinshasa's waters, the authors found lower levels of As (AM < 0.07 µg/L, n = 100), Cd (AM < 0.01 µg/L, n = 100) and Pb (AM = 1.0 µg/L, n = 100) as compared to the WHO drinking water guidelines [TUAKUILA, *al.*, 2016]. In another study, KABAMBA et *al.* [2016] showed elevated levels of As (range: 0.9 - 6.0 ng/m^3), Cd (range: 2.5 - 5.9 ng/m^3) and Pb (range: 166.2 - 1422.5 ng/m^3) as compared to the european guidelines. In order of expanding environmental data about these toxic metals in Kinshasa, the present study aims to investigate background levels of As, Cd and Pb in drinks and residential soils.

MATERIALS AND METHODS

Study area

Kinshasa, the capital of the Democratic Republic of the Congo (DRC), is located in the west of the country, it is divided into 4 districts and has a total area of 9,965 km^2 with more than ten million inhabitants. The City has a tropical climate which is characterized by a high humidity (with 2 seasons, the dry season extends from 15 May to 15 August; the rainy season takes the remaining period), a mean annual rainfall of 1620 mm and temperatures which are uniformly high throughout the year with an average of 25°C [CIA, 2017].The study area comprised 4 sampling locations of Kinshasa: Mont - Amba, Tshangu, Funa and Lukunga.

Sample collection

At each location, soil and drinks were collected at different sampling points between May and June 2010 according to the sampling method previously used by KABAMBA et *al.* [2016]. At each point:

- 100 soil samples were obtained as followed: A sample of 0 - 1 cm surface soil was taken from the perimeter of main roadsides, homogenized and sieved (2 mm) to obtain a coarse fraction (sand), according to method used by BRETIN [2005].
- 25 different drink samples (alcohol and soft) were collected from different local markets of Kinshasa (As these toxic elements are ubiquitous in the environment, they may be absorbed onto drinks that are consumed by many people in Kinshasa).
- All samples were transported in an ice box for analysis at the Louvain Center for Toxicology and Applied Pharmacology (LTAP, Brussels, Belgium).

Sample preparation

The drink samples were maintained at 4°C, acidified to pH < 2 (concentrated HNO_3) and balanced for at least 3 days / analyzed the elements [ROSSITER et *al.*, 2010]. 0.50 mL of drink sample was diluted with 4.50 mL of diluent (1% v/v HNO_3, 0.5 % v/v HCl) and 50 ppb of Sc, Ge, Rh and Ir as internal standard. The method used for the determination of three toxic elements in drinks is ISO 15189 certified. The soil samples were dried at 25°C until the weight remained constant, filtered by nylon subsampling to 2 mm from the mesh; 0.3 g were digested with 5 mL of nitric acid (65% SUPRAPUR, E. MERCK, Darmstadt, Germany) in the Teflon vessel for 3 min (A Paar Multiwave V 3.20.5). After digestion, solutions were filtered and compounded with 25 mL of nanopur water. The soil pH was measured with nanopur water (1:2 ratio, dry wt/v). Precaution was taken to avoid contamination during all stages of collection, transport, and analysis [TUAKUILA et *al.*, 2013b].

Sample Measurements

As in previous publications [TUAKUILA et *al.*, 2012, 2016], the concentrations of these toxic elements (As, Cd, Pb) were simultaneously measured by inductively coupled argon plasma mass spectrometry (ICP-MS, Agilent 7500 ce), using validated and ISO15189 certified procedures, with quality control/quality assurance procedures, as described in literature [TUAKUILA et al., 2012; HOET et *al.*, 2013; ELONGI et *al.*,2016]. Among the

Table 1. Distribution of measured toxic elements in soils. The minimum, mean, maximum and median values are given along with the typical levels found in the literature.

Parameter	Unit	N	LD	AM	Min	Q1	Med	Q3	Max	Typical levels[a]
As	mg/kg	100	0.03	0.62	0.07	0.24	0.5	0.67	3.91	5
Cd	mg/kg	100	0	0.08	0.01	0.03	0.06	0.11	0.28	<1
Pb	mg/kg	100	0.01	26.92	3.81	8.22	15.85	26.63	233.59	25-May

a: WILLIAMS et *al*. [2000].
LD = limit of detection;
Min = minimum value;
Max = maximum value;
Q1-25th percentile;
Med=median -50th percentile,
Q3-75th percentile.
AM= arithmetic mean.

Table 2. Distribution of measured toxic elements in drinks (alcohol, soft). The minimum, mean, maximum and median values are given along with the WHO guideline values.

Parameter	Unit	N	LD	AM	Min	Q1	Med	Q3	Max	WHO guidelines[a]
As	µg/L	25	0.07	4.52	0.28	1.09	4.15	8.37	10.73	10
Cd	µg/L	25	0.01	0.04	0.01	0.02	0.03	0.05	0.14	3
Pb	µg/L	25	0.03	0.03	0.02	0.02	0.02	0.03	0.07	10

a: WHO [2011].
LD = limit of detection;
Min = minimum value;
Max = maximum value;
Q1-25th percentile;
Med = median -50th percentile;
Q3-75th percentile.
AM = arithmetic mean.

Table 3. Median with minimum and maximums valus of toxic elements in different environmental media of Kinshasa.

Parameter	Drinks: alcohol-soft, (µg/L)[a]	Soil, (mg/kg) [a]	Ambient air (ng/m^3) [b]	Water Drinking, (µg/L) [c]
As	4.15 (0.28 - 10.73)	0.50 (0.07 - 3.91)	4.0 (0.9 - 6.0)	<0.07 (<0.07 - 1.90)
Cd	0.03 (0.01 - 0.14)	0.06 (0.01 - 0.28)	3.7 (2.5 - 5.9)	<0.01 (<0.01 - 0.43)
Pb	0.02 (0.02 - 0.07)	15.85 (3.81 - 233.59)	358.3 (166.2 - 1422.5)	0.4 (0.05 - 21.1)

a: Present study
b: KABAMBA et *al*. [2016]
c : TUAKUILA et *al*. [2016]

three elements measured by ICP-MS, Cd (m/z 111) and Pb (m/z 208) were analyzed using mode Standard without using the collision cell and As (m/z 75) was analyzed using helium as collision gas. Vacuum laboratory was prepared using nanopur water and treated in the same way as the samples.

Data presentation and statistical analysis

Data management and statistical analysis were performed in SAS 9.4 (SAS Institute, Cary, USA). Data are reported as minimum, maximum, medians, arithmetic means with 25th, 50th and 75th percentiles. The limit of detection (LOD) divided by 2 was used for imputation of values lower than the LOD.

RESULTS

The results from the trace element analysis (mean, minimum, lower inter-quartile range (Q1), median, higher inter-quartile range (Q3) and maximum values) are displayed in Tables 1 and 2. The number of samples analyzed (N), the limit of detection (LD) and the guidelines or typical levels in the different environmental medias are also presented.Tables1 and 2 also include the typical values from WILLIAMS et *al.* [2000], and the WHO guidelines from WHO [2011], respectively. The soil pH [Med (min - max)] was 8.06 (7.74 - 8.86). Median with minimum and maximum values of toxic elements in different environmental medias of Kinshasa are presented in Table 3. None of the measured parameter was significantly different among the 4 sampling locations investigated (data not shown).

In soil, the results of the toxic elements measurements show that none of the As and Cd mean concentration with maximum was higher than the typical levels, whereas about more than 25% of Pb concentration was higher than this typical level with 233,59 mg/Kg as a maximum. In drink, the values were substantially lower than the WHO guideline values or in line with these values.

DISCUSSION

The first biomonitoring study carried out to assess the exposure to trace elements in urine showed elevated levels of Al, As, Cd, Pb and Hg as compared to other databases; but data on environmental exposure to these elements in Kinshasa are scarce [TUAKUILA et *al*., 2012]. Moreover, in a case-control study between preeclampsia and toxic metals in Kinshasa, ELONGI et *al*., [2016] revealed that women with preeclampsia excrete higher amounts of several toxic metals, especially Pb, than control women. Although the exact sources of this exposure are unknown, these findings underscore the need for preventing environmental exposures to Pb and other toxic metals. The present study originated from that observation and its main objective was to investigate the possible sources of the exposure to trace elements (As, Cd, Pb) in the population. Great care was taken to consider our published data (ambient air and water) and to select representative samples (soil and drink) as sources of exposure to trace elements in the population of Kinshasa but, in the absence of reliable data, it is not possible to assess the exact representativeness of our samples and a bias is not formally excluded. There is, however, no reason to suspect a bias caused by self-selection based on either high or low exposure to these toxic elements.

As levels in water, drinks (alcohol and soft), soil and ambient air

Through a study carried out to investigate background levels of toxic elements in different environmental medias of Kinshasa, TUAKUILA et *al.* [2016] showed that As concentrations were lower than the WHO drinking water guidelines or the typical levels [the level (min - max) ranged from 0.04 to 1.90 µg/L; 0.04 to 0.66µg/L; 0.12 to 1.12µg/L in drinking water, groundwater, and surface water, respectively]. Moreover, the current levels of As in drinks (alcohol and soft) [mean (min - max) : 4.52 µg/L (0.28 - 10.73)] and in soil [mean (min - max) : 0.62 mg/Kg (0.07 - 3.91)] are in line with the WHO [2011] drinking water guidelines and the typical levels proposed by WILLIAMS et *al.* [2000], respectively. Thus, drink and soil do not appear to constitute predominant sources of exposure to As in the Kinshasa's population. However, KABAMBA et *al.* [2016] showed that living in Kinshasa is associated with elevated levels of As [the 24 - h level ranged from 0.90 to 6.0 ng/m^3] as compared to databases from Australia and Europe [WHO, 2000; AAS and BREIVIK, 2008; SA, 2015; ANP 2016]. No industry or landfill known to release As existed near the study places [INS, 2017]. The main sources of inorganic As for general populations worldwide are drinking water and food (rice and grains), seafood (fish, shellfish, and seaweed) being the main source of organic As compounds [FELDMANN and KRUPP, 2011]. In addition to the ambient air source of As in Kinshasa, Fish (''mpiodi'', i.e., *Trachurus trachurus)*, corn and rice are some of the frequently consumed food, whose consumption might also explain the high total As levels observed in the urine of general populations [TUAKUILA et *al.,* 2012] and of women with preeclampsia [ELONGI et *al.,*2016].

Cd levels in water, drinks (alcohol and soft), soil and ambient air

In the present study, drink (alcohol and soft) [mean Cd level (min - max): 0.04 µg/L (0.01 - 0.14)] and soil [mean Cd level (min - max) : 0.08 mg/Kg (0.01 - 0.28)] did not appear to be a significant source of exposure to Cd as compared to the WHO [2011] drinking water guidelines and the typical levels proposed by WILLIAMS et *al.* [2000]. The Similar results were observed in water [the level (min - max) ranged from 0.04 to 1.90 µg/L; 0.04 to 0.66µg/L; 0.12 to 1.12µg/L in drinking water, groundwater, and surface water, respectively [TUAKUILA et *al*,. 2016].Studying some metals in ambient air of Kinshasa, KABAMBA et *al.* [2016] showed that living in Kinshasa is associated with elevated levels of Cd [the 24 - h level ranged from 2.5 to 5.9 ng/m^3] as compared to databases from Australia and Europe [WHO, 2000; AAS and BREIVIK, 2008; VINCENT and PASSANT, 2013; SA, 2015; ANP, 2016].

Kinshasa was not known as a long-standing producer of Cd [CIA, 2017]. Moreover, MBUYI and TUAKUILA [2002] showed that vegetables cultivated along the side of main roads of the city contained high levels of Cd (mean 0.87 mg/kg) as compared to the Cd maximum levels in vegetables (0.05 and 0.2 mg/kg) as determined by a European regulation [EC, 2008]. In addition to vegetables and the ambient air sources of Cd in Kinshasa,

smoking is known to be a major source of Cd in smokers [LAUWERYS and HOET, 2001; TUAKUILA et *al.,* 2012].

Pb levels in water, drinks (alcohol and soft), soil and ambient air

Pb is one of the most widespread toxicants, and although its uses have been progressively prohibited by rules and regulations resulting in a dramatic decline of Pb exposure in many countries, its poisoning remains a matter of public health concern, especially for children [LIDSKY and SCHNEIDER, 2003]. In 2009, Kinshasa completed the removal of Pb from water pipes and introduced lead-free petrol [TUAKUILA et *al*,.2013b]. However, the ambient air, battery manufacture, recycling dust and soil remain important sources of exposure of Pb [TUAKUILA et *al*,. 2012, 2013a,b, 2016; KABAMBA et *al.,* 2016]. Moreover, the current levels of Pb in soil [mean (min - max): 26.92 mg/Kg (3.81 - 233.59)] are higher than the typical levels proposed by WILLIAMS et *al.* [2000]. However, drink (alcohol and soft) [mean Pb level (min - max): 0.03 µg/L (0.02 - 0.07)] does not appear to be a significant source of exposure to Pb as compared to the WHO [2011] drinking water guidelines. Similar results were also observed in water [the level (min - max) ranged from 0.05 to 21.10 µg/L; 0.03 to 0.25µg/L; 0.03 to 183.10µg/L in drinking water, groundwater, and surface water, respectively [TUAKUILA et *al.,* 2016].

CONCLUSION

This study reports the distribution levels of As, Cd, Pb in the drink and soil samples collecting in Kinshasa, which further contributes to delineate exposure levels in Kinshasa's population. These data will serve decision makers and/or health professionals and future research. Globally, the results indicate that the sources of exposure to these toxic elements from drinks and soil are not significant in Kinshasa's population. The emphasis should shift to understand the different sources of As, Cd and Pb emissions in ambient air (low fuel quality, congested traffic; wood, charcoal, crop, residual oil, and waste residues combustion for heating and cooking in open fires, etc.) and to examine the contributions of food sources (vegetables, fish, meat, etc.) to biological As, Cd, and Pb levels.

RESUME

Sources possibles d'exposition aux éléments toxiques (As, Cd, Pb) dans la population de Kinshasa, R.D. Congo.

L'Arsenic (As), le Cadmium (Cd) et le Plomb (Pb) sont omniprésents dans l'environnement et constituent certains des éléments toxiques auxquels les kinois sont exposés. Cependant, les sources exactes de cette exposition sont moins connues. Cette étude avait comme objectif d'investiguer sur les possibles sources d'exposition à ces éléments toxiques dans la population de Kinshasa. Au total, 100 échantillons de sol et 25 échantillons d'eau de boisson étaient collectés à 4 endroits. Les concentrations de ces 3 éléments ont été déterminées par l'ICP-MS (Couplage inductif à plasma et spectrométrie de masse). En vue d'éviter toute contamination, un contrôle minutieux a été appliqué durant la collection, la manipulation et les analyses des échantillons. Globalement, les résultats indiquent que l'eau et le sol ne sont pas des sources significatives d'exposition aux éléments toxiques (As, Cd, Pb). Ces nouvelles données seront utiles pour des décideurs politiques, des professionnels de santé et des futurs chercheurs. L'accent devrait être mis pour comprendre les sources d'émissions de As, Cd et Pb dans l'air ambiant (faible qualité du carburant, congestion dans le trafic routier, combustion de bois, de l'huile résiduelle, feux ouverts, etc.) et examiner la contribution des sources alimentaires (légumes, poissons, viande, etc.) aux niveaux urinaires ou sanguins de As, Cd et Pb dans la population de Kinshasa.

Mots clés

Eléments toxiques (As, Cd, Pb), sources d'exposition, Kinois.

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