Bio-surveillance of environmental pollutants in the population of Kinshasa, Democratic Republic of Congo (DRC): A small pilot study

Trésor Bayebila (✉ baytresor_pharma@yahoo.fr)

University of Kinshasa: Universite de Kinshasa   https://orcid.org/0000-0002-2734-3148

Patrice Dufour

University of Liege: Universite de Liege

Catherine Pirard

University of Liege: Universite de Liege

Jean Nsangu

University of Kinshasa: Universite de Kinshasa

Jean-Pierre Mufusama

University of Kinshasa: Universite de Kinshasa

Jérémie Mbinze

University of Kinshasa: Universite de Kinshasa

Roland Marini

University of Liege: Universite de Liege

Corinne Charlier

University of Liege: Universite de Liege

Abstract

Background

Environmental pollutants are known to be ubiquitous and may present toxic effects (endocrine-disruption properties, carcinogenicity…). Therefore, they represent a real threat to human health. The aim of the present study was to assess the content of environmental pollutants (inorganic, persistent, and non-persistent pollutants) in biological samples (urine, serum, and whole blood), collected from volunteers in Kinshasa, capital of Democratic Republic of Congo, in order to estimate the exposure level in the population of Kinshasa to environmental pollutants.

Methods

From randomly selected 15 volunteers living in Kinshasa, aged from 25 to 66 years, including 10 men and 5 women, urine, whole blood, and serum samples were used in this study to estimate the contents in these environmental pollutants, using Inductively Coupled Plasma Mass Spectrometry, Gas Chromatography coupled to Mass Spectrometry, and Liquid Chromatography coupled to Mass Spectrometry.

Results

When compared to data nationally and internationally available, the preliminary outcomes of this study indicated a high level of exposure to environmental pollutants in the population of Kinshasa, especially for arsenic, cadmium, lead, benzophenone-3, methyl-paraben, propyl-paraben, triclosan, mono-ethyl phthalate, mono-n-butyl phthalate, 3-phenoxybenzoic acid, cis-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropane-1-carboxylic acid, and for 3,5,6-trichloro-2-pyridinol. The levels of 4,4'dichloro-diphenyl-dichloro-ethylene and glyphosate were also significant although some heavily exposed populations showed higher level of contamination. In contrast, the investigated population of Kinshasa was found to weakly exposed to other pollutants like bisphenol A, dialkyl phosphates, polychlorinated biphenyls, brominated flame retardants, phenolic organo-halogens, and perfluoroalkyl substances.

Conclusion

Although the biologic fluids were collected from a limited number of volunteers (n = 15), the results of the present report clearly indicate that the population of Kinshasa is not spared by the investigated environmental pollutants.

Trial registration: this study was retrospectively registered by the national health ethics committee in the Congo under the series number of 159/CNES/BN/PMMF/2020.

Background

Environmental protection is a key to the sustainable development. For decades, the environment has indeed been threatened by different human activities due to industrialization, including progress in agriculture, growing use of plastic materials, fire management products, pharmaceuticals and cosmetics… [1]. Since decades, the correlation between the increase in chemical production and that of the chronic diseases' prevalence has suggested that some chemicals may be responsible of endocrine disruptions, carcinogenicity or other toxic effects [2–4]. As these compounds are ubiquitous and can operate at low concentrations, their release in the environment poses a potential threat to human health [5, 6]. The actual systemic exposure of an individual to environmental pollutants can be evaluated by the quantification of these compounds or their metabolites in biological fluids [7].

Although being a controversy topic, many bio-surveillance studies have reported harmful effects of environmental pollutants in humans. Among compounds presumed to have health-threatening properties, on one hand, persistent organic pollutants (POPs) are organic compounds with remarkable resistance to degradation into environment and among which there are organochlorine pesticides (OCP), polychlorinated biphenyls (PCB), brominated flame retardants (BFR), phenolic organo-halogens (POH), and perfluoroalkyl substances (PFAS). Among the health concerns associated to these compounds we could mention diseases affecting the central nervous system, metabolic diseases and birth weight alteration [8]. On the other hand, inorganic pollutants (IP), especially arsenic, cadmium, lead, cobalt, tin, etc., are mineral compounds and many of them are likely used in several industrial activities. Some of them are alleged of being neurotoxic and of having damaging effects to noble organs like kidney, liver, heart, etc [9]. Furthermore, non-persistent organic pollutants (nPOPs) like pyrethroids, alkyl-phosphates, bisphenols, triclosan, phthalates, parabens, glyphosate, and benzophenone, are organic compounds with fast degradation into environment but industrially produced in large amounts and are suspected to be responsible of several dysfunctions of the hormonal systems (reproductive or thyroid system), central nervous system, and in the occurrence of metabolic and chronic diseases [10].

With an area of 9,965 km^2 and an estimated population of 14.3 Million of inhabitants in 2020, Kinshasa is the capital, the most populated and the biggest city of the Democratic Republic of the Congo (DRC). Besides an important demographic increase, there is also an increase in morbidity and mortality rates due to chronic and metabolic diseases [11–13]. Moreover, Tuakuila J. et al. 2015 [14] reported a lack of data on bio-surveillance of environmental pollutants in the population of Sub-Saharan African countries in general and particularly in DRC. This lack of data poses a big limitation in the surveillance of exponentially growing pathologies.

The aim of the present study was to assess the content of environmental pollutants (inorganic, persistent, and non-persistent pollutants) in biological samples (urine, serum, and whole blood), collected from 15 volunteers aging from 25 to 66 years, belonging to various business sectors, and living in Kinshasa, in order to estimate the exposure level in the population of Kinshasa to environmental pollutants. To the best of our knowledge, this is the first study reporting on bio-surveillance data of organic persistent and non-persistent pollutants in the population of Kinshasa as well as in that of DRC.

Methods

Sample collection

Biological fluids were collected from 15 volunteers recruited in the population of Kinshasa with ages ranging from 25 to 66 years, including 10 men and 5 women. For this small pilot bio-surveillance study, with the aim to conduct the first exploration of pollutant contamination in general population of Kinshasa and covering various exposition profiles, volunteers were selected among business sectors, including market gardeners, pesticide vendors, plastic manufacturers, aluminum utensil makers, mechanics, traders, students, lawyers, painters, drivers, teachers, polices, students, fitters, and sanitation technicians. Each sector, a volunteer was randomly chosen throughout the city. Prior to be enrolled, volunteers were informed about the study merits and were submitted to a questionnaire to record their age, business activity, commonly handled products, and duration of exposure.

Early in the morning, after breakfast, each volunteer was requested to give about 10 mL of whole blood, kept in a plastic tube (without gel but with heparin), 10 mL of whole blood to prepare the serum, kept in a plastic tube (without gel and without heparin), and 50 mL of urine, kept in a polypropylene vial, all together 45 samples for analysis. These samples were collected between March and April 2019 and placed immediately in a dry ice enclosure, while ensuring that tubes with whole blood were not in direct contact with the ice, to avoid the risk of hemolysis and facilitating their transport, for proper storage, to the Clinical Biology Laboratory of the Faculty of Pharmaceutical Sciences at the University of Kinshasa. Tubes with whole blood for the preparation of the serum samples were centrifuged for 5 minutes at 3000 rpm and kept, together with urine samples, in a freezer at -20°C, while tubes of heparinized whole blood were stored in a fridge at 4°C. Toxicological analyses were carried out in the Laboratory of Clinical, Forensic and Environmental Toxicology, at the University of Liege, in Belgium. For a proper transport to Belgium, all samples were stored in a hermetically sealed enclosure with dry ice, while ensuring that the whole blood tubes were not in direct contact with the ice. The current study was approved by the national health ethics committee in the Congo under the series number of 159/CNES/BN/PMMF/2020.

Analytical procedures

Analysis of metals and metalloids in urine

The inorganic compounds (namely, As, Bi, Cd, Co, Cr, Cu, Mn, Mo, Ni, Sb, Se, Sn, Tl, V and Zn) were analyzed using ICP-MS. Briefly, internal standard (containing Rh, Sc and Ge) was added, at the same time, to the sample, to the quality control sample, and to the standard calibration sample. This mixture was then diluted with an aqueous solution of nitric acid 0.5% before being injected into ICP-MS. For detailed analytical methodology, see supplementary information.

Analysis of glyphosate in urine

Urinary content in glyphosate was investigated following the procedure extensively described in the supplementary information. Briefly, urinary glyphosate in sample, quality control sample, and standard calibration sample was derivatized with fluorenylmethoxycarbonyl chloride (FMOC). A first liquid-liquid extraction was then performed to eliminate residual FMOC and apolar compounds. A second liquid-liquid extraction was performed after acidification to extract the analyte. After evaporation and reconstitution in vial, the sample was analyzed by LC-MS/MS.

Analysis of pyrethroids and organophosphate chlorpyrifos metabolites in urine

Five pyrethroids metabolites (namely, cis- and trans-3-(2,2-dichlorovinyl)-2.2-dimethylcyclopropane carboxylic acid (c- and t-DCCA), 3-phenoxybenzoic acid (3-PBA), 4-fluoro-3-phenoxybenzoic acid (4F-3-PBA) and 3-(2,2-dibromovinyl)-2.2-dimethylcyclopropane carboxylic acid (DBCA)) and one chlorpyrifos metabolite (namely 3,5,6-trichloro-2-pyridinol (TCPY)) were analyzed according to the methodology detailed in Pirard et al. 2020 [15]. Briefly, urinary sample, quality control sample, and standard calibration sample were extracted with diethyl ether. The organic layer was evaporated to dryness and the residue was derivatized with N-tert-butyldimethylsilyl-N-methyltrifluoroacetamide (MTBSTFA). The derivatized extract was then analyzed by GC-MS/MS.

Analysis of alkylphosphates in urine

Five dialkylphosphates (DAPs) (nonspecific metabolites of organophosphate pesticides) (namely dimethylthiophosphate (DMTP), dimethyldithiophosphate (DMDTP), diethylphosphate (DEP), diethylthiophosphate (DETP) and diethyldithiophosphate (DEDTP)) were quantified in urine samples according to the methodology described in Pirard et al., 2020 [15]. In summary, urine sample, quality control sample, and standard calibration sample were extracted on solid phase extraction (SPE) cartridge. The eluate was evaporated to dryness and then derivatized with chloroiodopropane. The derivatized extract was then analyzed by GC-MS/MS.

Analysis of phthalate metabolites, parabens and benzophenone-3 in urine

The urinary concentrations of 7 phthalate metabolites (namely, monoethyl phthalate (MEP), mono-iso-butyl phthalate (MiBP), mono-n-butyl phthalate (MnBP), monobenzyl phthalate (MBzP), mono-2-ethylhexyl phthalate (MEHP), mono-2-ethyl-5-hydroxyhexyl phthalate (5-OH-MEHP) and mono-2-ethyl-5-oxohexyl phthalate (5-oxo-MEHP)), 4 parabens (namely, methylparaben (MeP), ethylparaben (EP), n-propylparaben (PrP) and n-butylparaben (BP)) and benzophenone-3 were determined according to the methodology developed by Dewalque et al. 2014 [16]. Briefly, urine sample, quality control sample, and standard calibration sample were submitted to an enzymatic hydrolysis, then an extraction was performed using SPE cartridge and finally the extract was analyzed on a LC-MS/MS apparatus.

Analysis of triclosan and bisphenols in urine

The levels of triclosan and 7 bisphenols (BP) (namely, BPA, BPAF, BPF, BPZ, BPAP, BPP and BPS) in urine samples were measured by using the methodology detailed in the supplementary materials. In summary, the sample, quality control sample, and standard calibration sample were submitted to an enzymatic hydrolysis followed by an extraction on a SPE cartridge. This first extraction was followed by a liquid-liquid extraction and then by a derivatization. The derivatizated extract was then analyzed by a GC-MS/MS [17].

Analysis of lead in whole blood

Lead was quantified in whole blood. Samples, quality control samples, and standard calibration samples were mixed with internal standard and diluted with a mixture of nitric acid (0.5%), n-butanol (0.2%) and triton (0.1%) in water. The lead content was determined by using an ICP-MS. The procedure has been detailed in supplementary information.

Analysis of polychlorobiphényls (PCBs) and organochlorine pesticides in serum

Fifteen organochlorine pesticides or metabolites, (namely alpha-, beta-and gamma-HCH (α-, β- and γ-HCH), hexachlorobenzene (HCB), aldrin, dieldrin, endrin, trans-chlordane, oxychlordane, trans-heptachlor ep-oxide, cis- and trans-nonachlor, 2,4'- and 4,4'-dichlorodiphenyl-dichloroethylene (DDE), beta-endosulfan) and 3 PCBs (−138, −153, and −180) were quantified in serum. The analytical procedure was extensively detailed in Pirard et al. 2018 [1]. Briefly, sample, quality control sample, and standard

calibration sample were denaturized with acetonitrile and a saturated potassium carbonate solution. The mixture was then extracted twice with hexane-acetone mixture (9/1, v/v). The organic phase was cleaned on a SPE cartridge and then evaporated and reconstituted in nonane. The extract was analyzed on a GC-MS/MS apparatus.

Analysis of BFRs in serum

The methodology to quantify 8 polybrominated diphenylethers (PBDEs) (namely, BDE-28, -47, -99, -100, -153, -154, -183 and -209) has been described in Pirard and Charlier, 2018 [18]. In summary, serum sample, quality control sample, and standard calibration sample were denaturized with a glacial acetic acid/water mixture (3/7, v/v) and then extracted twice with a mixture of hexane and acetone (95/5, v/v). The organic phase was then cleaned on a PHREE cartridge, then evaporated and transferred into nonane. The quantification was performed using a GC-MS/MS.

Analysis of perfluorinated alkyl subtances (PFAS) in serum

The quantification of the serum content in 7 PFASs (namely, perfluoro-octane sulfonic (PFOS), perfluoroctanoic acid (PFOA), perfluorohexane sulfonate (PFHxS), perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), perfluoroheptanoic acid (PFHpA) and perfluoroundecanoic acid (PFUdA)) was performed according to the methodology described in Dufour et al. 2018 [8]. Briefly, serum sample, quality control sample, and standard calibration sample were denaturized with formic acid/water mixture (1/1, v/v). Then the sample was extracted on a weak anionic exchange SPE cartridge, the eluate was evaporated to dryness and then reconstituted in 80 µL of a mixture of mobile phases. The extract was then analyzed using a LC-MS/MS apparatus.

Analysis of phenolic organohalogens (POHs) in serum

POHs (namely, pentachlorophenol (PCP), tetrabromobisphenol A (TBBPA), 2,4,6-tribromophenol (2,4,6-TBP), 2,3,6-tribromophenol (2,3,6-TBP), 2,4,5-tribromophenol (2,4,5-TBP), 2,3,4,6-tetrabromophenol (2,3,4,6-TeBP), 6-hydroxy-polybromodiphenylether 47 (6-OH-BDE 47), 5-hydroxy-polybromodiphenylether 47 (5-OH-BDE 47), 5'-hydroxy-polybromodiphenylether 99 (5'−OH−BDE 99), 4-hydroxy-polychlorinated biphenyl 107 (4-OH-CB 107), 3-hydroxy-polychlorinated biphenyl 138 (4-OH-CB 138), 4-hydroxy-

polychlorinated biphenyl 146 (4-OH-CB 146), 3-hydroxy-polychlorinated biphenyl 153 (3-OH-CB 153), 4-hydroxy-polychlorinated biphenyl 172 (4-OH-CB 172), 3-hydroxy-polychlorinated biphenyl 180 (3-OH-CB 180) and 4-hydroxy-polychlorinated biphenyl 187 (4-OH-CB 187)) were analyzed according to the method described in Dufour et al. 2016 [19]. In summary, the serum sample, quality control sample, and standard calibration sample were denaturized with a mixture of water/formic acid/2-propanol (50/40/10, v/v) and then extracted on a strong anionic exchange SPE cartridge. The eluate is then extracted with hexane; hexane phase was evaporated to dryness and then derivatized with trimethylsilyldiazomethane. The derivatized extract was then analyzed using a GC-MS/MS.

Analysis of creatinine in urine samples

Adjustment to creatinine was used to normalize pollutant contents in urine samples. In this study, urinary creatinine was evaluated on an ARCHITECT Ci 4100 automate from ABBOTT (Illinois, USA), using an immunoassay.

Quality assurance and statistical analysis

To ensure the results quality, all analyses were covered by internal or external quality control sample and for each analysis, a specific internal standard was used as a recovery indicator and a correction factor. A specific calibration curve was applied for each analysis. All statistical analyses were performed using R programming software (version 3.6.3., CRAN) and Microsoft Excel 2013 (Microsoft, Redmond, WA). For analyses with results lower than the limit of quantification (LOQ), a correction was made by multiplying the LOQ by the detection frequency (DF), in order to valorize the investigation outcomes.

Results

All searched ions and molecules in the current study are incorporated in the supplementary information. Table 2 contains most significant ions and molecules and the discussion section of this study turn around this. Results obtained from urine samples analyses were presented in mg/L, and in mg/g of creatinine in parenthesis. Analyses of serum and whole blood samples are expressed in mg/L or in pg/mL (different unities were used to facilitate study comparison).

Table 1 shows the limit of qualification (LOQ) of the used methods for each pollutant as well as their detection frequencies, respectively in pg/mL and in percentage (%). For the most significant compounds under investigation, majority of them were detected in the biological fluids, except for β-HCH, 2,4,6 TBP, 6-OH BDE 47 and BPZ.

Table 2 presents arithmetic and geometric means, medians, minimum and maximum contents in urine, whole blood, and serum samples of some pollutants or their corresponding biomarkers. Being robust at extreme values, the median concentrations were considered as reference points.

The investigated population showed a real exposition to environmental pollutants: for example, MeP was the predominant parabens (445.39 µg/L of median value), followed by PrP (31.35 µg/L of median value) and by EP (0.47 µg/L of median value). For Phthalate metabolites, MnBP presented the highest median value (145.19 µg/L), followed by MEP (108.56 µg/L), MiBP (26.52 µg/L), and MEHP (9.03 µg/L). Median values of As, TCS, Lead and 4,4'DDE were respectively 70.91 µg/L, 40.13 µg/L, 53.57 µg/L, and 1.46 µg/L. For all most significant ions and molecules here reported, only median values of PCP, PBDE 47, and PBDE 153 were smaller than their corresponding LOQ methods and therefore not reported.

Discussion

Many bio-surveillance studies have suggested that only considering the expression of concentration adjusted by creatinine could lead to a bias when comparing different populations, pollutants contents in urine samples in comparative studies were only reported in mg/L [20, 21]. In this study, comparison was also performed in a national and international scale, to have an idea on the exposure level in the population of Kinshasa.

Metals in urine

Except for Mn (detected at 20%), Sn and Sb (detected each at 47%), and Cd (detected at 93%), all other inorganic compounds, investigated in urine were detected in all samples. Detected in most of the samples and with a median concentration of 70.91 µg/L, the population of Kinshasa is clearly more exposed to arsenic than populations in two other studies conducted in mining and industrial areas in Spain (Huelva) and in the DRC (Lubumbashi) (Table 3). Collected from 261 students, the first study reported a median content of 1.17 µg/L, while, the second study, based on 39 pregnant women (control population), stated a median concentration 3 times lower than here reported, probably due to an important food intake in Kinshasa, arising the necessity to figure out the exposure source and to identify the vulnerable population [22, 23]. With a median concentration of 0.61 mg/L, the cadmium exposure here reported seemed to be similar to those measured in the above-mentioned study (control population) from Lubumbashi [23], but two fold higher than the one reported in Spain, with a median content of 0.29 mg/L [22], but also than that stated in two Belgian studies, the first involving a population of 52 men of Ath with a geometric mean of 0.21 mg/L [24], and the second exploring 125 Belgian mothers reporting a median value of 0.22 mg/L [7] (Table 3).

Glyphosate

Used in agriculture for crop protection, glyphosate, a widely used organophosphorus herbicide worldwide, is currently classified in Category 2A "probably carcinogenic to humans" by the International Agency for Research on Cancer (IARC) [25].

The glyphosate median outcome (0.23 mg/L) of the current study was far lower than that stated on 50 Irish adults with a median concentration of 0.87 mg/L [25]. But the study from Conrad A. et al. 2017 [26] on 40 German adults reported a lower median content (0.11 mg/L) than that stated here (Table 3). This result shows a clear glyphosate exposure in the investigated population of Kinshasa, due to a probable usage of this powerful herbicide by farmers or its possible presence in the various imported food products.

Lead

Human exposure to lead (Pb) is caused by various industrial activities such as metallurgy, printing, ammunition, paintings, batteries of accumulators, etc. Atmospheric lead, which is largely responsible for lead body burden, is usually derived from gasoline products where it is used as an anti-detonator [14].

When compared to international studies conducted on 156 women in Beijing [27] and on 52 men in Belgium [24], with respective median content values of 17.6 mg/L and 31.7 mg/L, the median concentration here presented was higher (53.6 mg/L) (Table 3). These differences could probably be explained by a more important lead exposure source in Kinshasa, reinforcing the need to search for exposure source and to identify the vulnerable population.

In a national level, the observed median content was higher to the one of the control population from the 39 women study in Lubumbashi [23], reporting a median concentration of 50.8 mg/L, although the study of Tuakuila J. et al. 2013 [28] stated a higher median content of 86 mg/L on a population of 100 children recruited in 2011 in Kinshasa and aged from 1 to 5 years (Table 3). Based on a chronologic order of these publications, one can think of a possible lead exposure reduction, namely due to the sale suppression of leaded petrol throughout the country since 2009. But it is always important to maintain bio-surveillance studies as some parallel leaded petrol markets may still exist in Kinshasa. Moreover, the observed lead whole blood contents here reported could also be due to a possible release of lead into the blood from internal storage, indeed it is an element known to be accumulated in bones, soft tissues, and blood.

Phthalates

For more than half a century, phthalates have been present in a wide range of daily products, so they are used as plasticizer, especially in polyvinyl chloride (PVC) and in cosmetics [20]. In the current study, median contents in MnBP and MEP, respectively 145.19 mg/L and 108.56 mg/L, were higher than those measured in 261 Belgian adults (33.3 mg/L of MnBP and 34.3 mg/L of MEP) [20], than those determined in 279 French pregnant women (35.7 mg/L of MnBP and 43.5 mg/L of MEP) [29], in 145 Danish women (20 mg/L of MnBP and 29 mg/L of MEP) [30], and in 99 Taiwanese women (52.39 mg/L of MnBP) [31]. This observation could probably due to a strong presence of phthalates in plastic packaging used in Kinshasa, increasing the need for further investigations on the exposure source and the identification of susceptible population. A slightly lower median concentration in MiBP was observed when compared to the French and Danish studies as well as the median content in MEHP was slightly lower than those reported in the French and Taiwanese studies (Fig.1).

Parabens

With bactericidal and fungicidal properties, parabens are largely used in cosmetics, food, and pharmaceuticals as preservatives. Detected in all investigated samples, median concentrations of MeP (445.39 µg/L) and PrP (31.35 µg/L) were clearly higher than those reported in 34 Tunisian women (34.94 µg/L of MeP and 3.06 µg/L of PrP) [32], 215 young Spanish (17 µg/L of MeP and 0.7 µg/L of PrP) [33], 145 Danish women (14 µg/L of MeP and 1.7 µg/L of PrP) [30], and 261 Belgian adults (16.1 µg/L of MeP and 1.2 µg/L of PrP) [20], probably due to a greater use of these preservatives in cosmetics, food and pharmaceutical products marketed in Kinshasa. Thereby, further investigations are needed to detect products containing parabens, their corresponding quantities and identify the vulnerable population. In contrast, this study has found a lower median concentration in EP (0.47 µg/L) (Table 4).

Benzophenone-3 (BP3)

Added in cosmetics and food packaging, benzophenones have ultraviolet filters properties, reducing their deleterious effects on the skin or food. Detected in all investigated samples, the current study has found a median concentration in BP3 (5 µg/L) higher than those reported in the Belgian adult population (1.3 µg/L) [20], in Tunisian women (1.73 µg/L) [32], and in Danish women (3.7 µg/L) [30], probably due to the presence of this UV filter in cosmetics or on food packaging, reinforcing the need to identify the source, used concentrations, and the vulnerable population (Table 3).

Pyrethroids and dialkylphosphate pesticides

Pyrethroids and dialkyl-phosphates (DAPs) are among pesticides largely used in crop culture for the protection of harvests and in public health for the fight against diseases vectors. Associated with weak enzyme baggage, the immaturity in the development of some organs makes children around the world among vulnerable populations to environmental pollution [34].

In the current study, median contents in TCPY (4.4 mg/L), in 3-PBA (2.23 mg/L), and in c-DCCA (0.47 mg/L) were higher than those measured in 240 Belgian children (with 1 mg/L of 3-PBA and 3.9 mg/L of TCPY) [15], in 1149 Chinese pregnant women (with 1.01 mg/L of 3-PBA and 0.44 mg/L of c-DCCA) [35], and in 1077 French pregnant women (with 0.36 mg/L of 3-PBA and 0.16 mg/L of c-DCCA) [36]. Only the median concentration in t-DCCA was lower than those on the two first studies. The high concentration observed in the present study was probably due to a very high use of pyrethroid insecticides in mosquito nets, insecticide sprays and other products for agricultural use (Fig.2). With a detection frequency of 53% for DEP and 47% for DETP with respective median contents values of 0.87 mg/L and <0.5 mg/L, this study has presented a slightly lower DAPs exposure when compared to studies on 240 Belgian children (with 1.8 mg/L of DEP) [15], on 136 Thai farmers (with DEP median value <LOQ and 1.2 mg/L of DETP) [37], and on 273 pregnant women in Jerusalem (with 2.72 mg/L of DEP and 0.55 mg/L of DETP) [38] (Table 4).

Bisphenols and triclosan

Used as epoxy resin monomers, bisphenols are aromatic organic compounds used in the manufacture of plastics and poly epoxides. Added in toothpastes and cosmetics for its antibacterial properties, triclosan is likewise suspected to be endocrine disruptor for instance, it is suspected to be associated with the decrease of some biomarkers of thyroid function [39].

The median content in triclosan here observed (40.13 mg/L) was one or two orders of magnitude higher than those observed in a Belgian population with 131 participants (2.24 mg/L) [17], in 1870 Korian adults (1.53 mg/L) [40], and on 145 Danish women (0.64 mg/L) [30]. Conversely, the investigated population of Kinshasa presented a lower median concentration in bisphenol A (1.36 mg/L) than those reported in the above studies and that on 215 young Spanish of Murcia (2.3 mg/L) [41], but higher than that on 34 Tunisian women (0.35 mg/L) [32] (Table 4). The current study showed that the population of Kinshasa was also exposed to the new bisphenols, especially to bisphenol F (detected in 67% of the samples) and bisphenol S (detected in 80% of the samples) (Table 1). The exposure in the population of Kinshasa to these pollutants is probably due to the presence of triclosan in cosmetic products, toothpastes and that of bisphenols in cans and plastics sold in the city.

Chlorinated pesticides

Previously used in the fight against pests in agriculture, the p,p'-dichlorodiphenyltrichloroethane (DDT) is still used in many tropical and subtropical regions to fight against mosquitoes vector of malaria [42]. In DRC, the use of organochlorine compounds has been banned since March 2005 but Nuapia Y. et al. 2016 [43] found these compounds in raw foods (beans, cabbage, fish and beef) sold in Kinshasa.

Main metabolite of DDT in the environment and in the living organism, p,p'-dichlorodiphenyldichlorethylene (p,p'-DDE) is considered as a marker of previous exposure [44]. Detected in 87% of the samples with a median content value of 1.46 mg/L, the investigated population in Kinshasa presented a concentration largely higher than those on 124 Belgian women (median = 0.41 mg/L) [1] and on Lebanese population with 314 participants (median = 0.13 mg/L) [45], probably either due to a remobilization of DDT accumulated in the soil, a late ban of DDT in DRC compared to other countries or the existence of an illegal DDT market in Kinshasa. Studies on 733 South African pregnant women and on 24 Bolivian women farmers reported a higher median concentration [46, 42] (Table 3).

Polychlorinated biphenyls

Used as insulating fluid in electrical equipment and additives in putty, polychlorinated biphenyls (PCB) are POPs as they also remain intact in the environment for many years. Comparing median contents in PCB 153 of studies performed on 251 Belgians (0.36 mg/L) [1], 314 Lebanese (0.12 mg/L) [45], and on 353 Afro-Americans (1.42 mg/L) [47], the investigated population in Kinshasa was weakly exposed to these compounds (median concentration in the present study = 0.081), probably due to their low use in the Kinshasan market (Table 3).

Perfluoroalkyl substances

Surfactants with high thermal stability, perfluoroalkyl substances (PFAS) are found in various everyday products (kitchen utensils, stoves, microwave packaging, raincoats, sportswear, etc.) thanks to their waterproofing and non-stick properties [8].

In the current study, a low contamination in PFAS (for instance, median PFOS = 0.5 ng/mL and median PFOA = 0.48 ng/mL) was observed in the population comparatively to studies on 237 Belgians (with median concentration of 3.61 mg/L for PFOS and 1.6 mg/L for PFOA) [48], 118 American teenagers (median PFOS = 3.72 ng/mL and median PFOA = 1.8 ng/mL) [49], 141 Chinese pregnant women (median PFOS = 4.31 ng/mL and median PFOA = 3.95 ng/mL) [50], and on 300 Czech adults (median PFOS = 2.43 ng/mL and median PFOA = 0.76 ng/mL) [51], reflecting a probable low presence of these compounds in the market of Kinshasa (Table 4).

Phenolic organohalogens

Contamination by phenolic organohalogens (POH) seemed to be weak in the investigated population: pentachlorophenol, a pesticide used namely in the protection of timber, was only detected in 14% of the population, while it was present in 100% of 272 Belgian volunteers as reported by Dufour et al. 2017 [52], with a median content value of 593 pg/mL (both populations were explored with the same analytical method). Likewise, weak was the contamination by OH-CBs, metabolites of PCBs. Only 4-OH-CB 187 was found in more than 50% of individuals (57%), with a median concentration of 2.4 pg/mL while Dufour et al. 2017 [52] highlighted a detection frequency of 100% and a median level of 39.4 pg/mL in the Belgian population, reflecting a reduced contamination observed for PCBs in the investigated population of Kinshasa.

Brominated flame retardants

Finally, brominated flame retardants (BFR) are compounds incorporated in many materials (especially plastics) to increase their fire resistance and to help reducing the risk of fire. They are therefore found in many everyday products, including vehicles, clothes, furniture, electronic equipment, etc. The investigated population of Kinshasa seemed to escape to these compounds as the most frequent substances, PBDE 47 and 153 were only detected in 27% of the samples, with maximum concentrations of 15 and 13 pg/mL, respectively. This situation is close to that observed in Belgium, with detection frequencies lower than 40% for all PBDEs highlighted with the same analytical method [48], but largely far from the contamination observed in the USA, where diverse studies showed median levels in total PBDEs ranging from 10 to values higher than 40 ng/g in lipids or approximatively 73.5 pg/mL and 294 pg/mL [53]. All comparison results at both national and international scale are presented in tables 3, 4 and figures 1, 2.

Conclusion

The current study provides data on environmental pollutants, including inorganic (Cd, Co, Pb, Zn, Cu, Ni, As, etc.), persistent organic (PCB, PFC, POH, BFR, OCP) and non-persistent organic ones (phthalates, parabens, triclosan, bisphenols, glyphosate, etc.) in serum, whole blood, and urine samples collected from volunteers in the population of Kinshasa, in 2019. These were the first bio-surveillance data on persistent and non-persistent organic pollutants in the city as well as in the country, partly filling the gap of insufficient bio-surveillance data in the populations of the Sub-Saharan African countries.

A comparison with nationally and internationally available data permitted to estimate the exposure level in the population of Kinshasa. Compared to other populations assessed around the world, high contents were observed with respect to arsenic, cadmium, lead, triclosan, methylparaben, propylparaben, mono-ethyl phthalate, mono-n-butyl phthalate, benzophenone-3, trichloro-pyridinol, cis-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropane-1-carboxylic acid (c-DCCA) and 3-phenoxybenzoic acid. The 4,4'dichloro-diphenyl-dichloro-ethylene (4,4'DDE) and glyphosate median values were also significant. In contrast, bisphenol-A, dialkyl-phosphates, PCB 153, perfluorinated compounds and organohalogenated phenolics were found at low concentrations compared to the situation highlighted in other countries. Finally, there was a similarity in median contents of brominated flame retardants with those from a Belgian study. Hypotheses were made to explain the observed differences.

Although the biologic fluids were collected from a limited number of volunteers (n= 15), the results of the present report clearly indicate that the population of Kinshasa is not spared by the investigated environmental pollutants. Therefore, it is of paramount importance to scale-up and validate this study to a larger population of Kinshasa to obtain a database on pollutants and identify potential hot spots of exposure, in order to establish relationship between certain socio-demographic characteristics (age, sex, food, smoking, professional activity, etc.) and the level of exposure to pollutants, to investigate certain possible source of exposure and to explore potential associations between the contamination and the prevalence of some chronic diseases in the population of Kinshasa.

List Of Abbreviations

DRC : Democratic Republic of Congo; POPs: persistent organic pollutants; OCP: organochlorine pesticides; PCB: polychlorinated biphenyls ; BFR: brominated flame retardants ; POH: phenolic organo-halogens ; PFAS: perfluoroalkyl substances ; FMOC : fluorenylmethoxycarbonyl chloride ; c- and t-DCCA : cis- and trans-3-(2,2-dichlorovinyl)-2.2-dimethylcyclopropane carboxylic acid ; 3-PBA: 3-phenoxybenzoic acid ; 4F-3-PBA: 4-fluoro-3-phenoxybenzoic acid ; DBCA : 3-(2,2-dibromovinyl)-2.2-dimethylcyclopropane carboxylic acid ; TCPY: 3,5,6-trichloro-2-pyridinol ; MTBSTFA: N-tert-butyldimethylsilyl-N-methyltrifluoroacetamide ; DAPs : dialkylphosphates ; DMTP : dimethylthiophosphate ; DMDTP : dimethyldithiophosphate ; DEP : diethylphosphate ; DETP : diethylthiophosphate ; DEDTP : diethyldithiophosphate ; SPE : solid phase extraction ; MEP : monoethyl phthalate ; MiBP : mono-iso-butyl phthalate ; MnBP : mono-n-butyl phthalate ; MBzP : monobenzyl phthalate ; MEHP : mono-2-ethylhexyl phthalate ; 5-OH-MEHP : mono-2-ethyl-5-hydroxyhexyl phthalate ; 5-oxo-MEHP : mono-2-ethyl-5-oxohexyl phthalate ; MeP : methylparaben ; EP : ethylparaben ; PrP : n-propylparaben ; BP : n-butylparaben ; BP-3 :

benzophenone-3 ; TCS : triclosan ; BP : bisphenols ; HCB : hexachlorobenzene ; 4 ;4'-DDE : 4,4'-dichlorodiphenyl-dichloroethylene ; PBDEs : polybrominated diphenylethers ; PFOS : perfluoro-octane sulfonic ; PFOA : perfluoroctanoic acid ; PFHxS : perfluorohexane sulfonate ; PFNA : perfluorononanoic acid ; PFDA : perfluorodecanoic acid ; PFHpA : perfluoroheptanoic acid ; PFUdA : perfluoroundecanoic acid ; PCP : pentachlorophenol ; TBBPA : tetrabromobisphenol A ; 2,4,6-TBP : 2,4,6-tribromophenol ; 2,3,6-TBP : 2,3,6-tribromophenol ; 2,4,5-TBP :2,4,5-tribromophenol ;2,3,4,6-TeBP :2,3,4,6-tetrabromophenol ; 6-OH-BDE 47 : 6-hydroxy-polybromodiphenylether 47 ; 5-OH-BDE 47 :5-hydroxy-polybromodiphenylether 47 ; 5'−OH−BDE 99 : 5'-hydroxy-polybromodiphenylether 99 ; 4-OH-CB 107 : 4-hydroxy-polychlorinated biphenyl 107 ; 3-OH-CB 138 : 3-hydroxy-polychlorinated biphenyl 138 ; 4-OH-CB 146 : 4-hydroxy-polychlorinated biphenyl 146 ; 3-OH-CB 153 : 3-hydroxy-polychlorinated biphenyl 153 ; 4-OH-CB 172 : 4-hydroxy-polychlorinated biphenyl 172 ; 3-OH-CB 180 : 3-hydroxy-polychlorinated biphenyl 180 ; 4-OH-CB 187 : 4-hydroxy-polychlorinated biphenyl 187 ; LOQ : Limit of Quantification ; DF : Detection Frequency ; IARC : International Agency for Research on Cancer.

Declarations

Ethics approval and consent to participate

Ethics approval was obtained from the national health ethics committee in the Democratic Republic of Congo. Firstly, participants were informed about the study interest and then, written informed consent was obtained from each participant during data collection. Participants were free to refuse to take part in the study as well as to withdraw any time along the study. Study results were strictly confident according to the declaration of Helsinki.

Consent for publication

Not applicable

Availability of data and materials

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

Competing interest

The authors declare that there is no competing interest regarding the publication of this paper.

Funding

ARES-CCD (Academie de Recherche et d'Enseignement Supérieur-Commission de la Coopération au Développement) was the funding agent of this study.

Authors' contributions

TB and PD drafted the manuscript. All authors read, commented the draft versions, and approved the final manuscript.

Acknowledgments

Author details

a. Faculty of Pharmaceutical Sciences, University of Kinshasa, D.R. Congob. Laboratory of Clinical, Forensic and Environmental Toxicology, University of Liege (ULiege), CHU (B35), 4000 Liege, Belgiumc. Center for Interdisciplinary Research on Medicines (C.I.R.M), University of Liege (ULiege), CHU (B35), 4000 Liege, Belgium

References

1. Pirard C, Compere S, Firquet K, Charlier C. The current environmental levels of endocrine disruptors (mercury, cadmium, organochlorine pesticides and PCBs) in a Belgian adult population and their predictors of exposure. International Journal of Hygiene Environmental Health 1 mars. 2018;221(2):21122. https://doi.org/10.1016/j.ijheh.2017.10.010.

2. UNEP/WHO. *State of the science of endocrine disrupting chemicals*. Geneva, Switzerland, United Nations Environment Programme/World Health Organization, 2012.

3. Cohn BA, Cirillo PM, Christianson RE. Prenatal DDT exposure and testicular cancer: a nested case control study. Arch Environ Occup Health Sept. 2010;65(3):12734. https://doi.org/10.1080/19338241003730887.

4. Wagner-Schuman M, Richardson JR, Auinger P, Braun JM, Lanphear BP, Epstein JN, et al. Association of pyrethroid pesticide exposure with attention-deficit/hyperactivity disorder in a nationally representative sample of U.S. children. Environ Health 28 mai. 2015;14:44. https://doi.org/10.1186/s12940-015-0030-y.

5. Sabatier P, Poulenard J, Fanget B, Reyss J-L, Develle A-L, Wilhelm B, et al. Long-term relationships among pesticide applications, mobility, and soil erosion in a vineyard watershed. Proc Natl Acad Sci USA 4 nov. 2014;111(44):1564752. https://doi.org/10.1073/pnas.1411512111.

6. Marotta V, Russo G, Gambardella C, Grasso M, La Sala D, Chiofalo MG, et al. Human exposure to bisphenol AF and diethylhexylphthalate increases susceptibility to develop differentiated thyroid cancer in patients with thyroid nodules. Chemosphere 1 mars. 2019;218:88594. https://doi.org/10.1016/j.chemosphere.2018.11.084.

7. Pirard C, Koppen G, De Cremer K, Van Overmeire I, Govarts E, Dewolf M-C, et al. Hair mercury and urinary cadmium levels in Belgian children and their mothers within the framework of the

COPHES/DEMOCOPHES projects. Sci Total Environ 15 févr. 2014;472:73040. https://doi.org/10.1016/j.scitotenv.2013.11.028.

8. Dufour P, Pirard C, Seghaye M-C, Charlier C. Association between organohalogenated pollutants in cord blood and thyroid function in newborns and mothers from Belgian population. Environmental Pollution 1 juill. 2018;238:38996. https://doi.org/10.1016/j.envpol.2018.03.058.

9. Åkesson A, Lundh T, Vahter M, Bjellerup P, Lidfeldt J, Nerbrand C, et al. Tubular and glomerular kidney effects in Swedish women with low environmental cadmium exposure. Environ Health Perspect Nov. 2005;113(11):162731. https://doi.org/10.1289/ehp.8033.

10. Duan Y, Yao Y, Wang B, Han L, Wang L, Sun H, et al. Association of urinary concentrations of bisphenols with type 2 diabetes mellitus: A case-control study. Environmental Pollution 1 déc. 2018;243:171926. https://doi.org/10.1016/j.envpol.2018.09.093.

11. Punga-Maole AM-L, Moningo DM, Kayembe PK, Tshikuela ML, Kabongo J-MM. Étude de dépistage du cancer de la prostate au sein d'une population d'employés d'une entreprise de Kinshasa en république démocratique du Congo: Taux de détection et facteurs de risques nutritionnels et géographiques. Progrès en Urologie 1 sept. 2008;18:512–8. https://doi.org/10.1016/j.purol.2008.04.009.

12. Mashinda KD, Kayembe KP, Mapatano MA. Prévalence du cancer en République Démocratique du Congo: données anatomopathologiques recueillies aux Cliniques Universitaires et à l'Hôpital Général de Référence de Kinshasa. Annales africaines de médecine 2012.

13. Tshimpi A, Ndarabu T, Batumona B, Tambwe F, Kayembe JMN. Cancer in Democratic Republic of Congo on 2016. Annales africaines de médecine 2016.

14. Tuakuila J, Kabamba M, Mata H, Mbuyi F. Tentative reference values for environmental pollutants in blood or urine from the children of Kinshasa. Chemosphere 1 Nov. 2015;139:32633. https://doi.org/10.1016/j.chemosphere.2015.06.039.

15. Pirard C, Remy S, Giusti A, Champon L, Charlier C. Assessment of children's exposure to currently used pesticides in Wallonia, Belgium. Toxicol Lett. 2020;329:1–11.

16. Dewalque L, Pirard C, Dubois N, Charlier C. Simultaneous determination of some phthalate metabolites, parabens and benzophenone-3 in urine by ultra high pressure liquid chromatography tandem mass spectrometry. Journal of Chromatography B 15 févr. 2014;949950:3747. https://doi.org/10.1016/j.jchromb.2014.01.002.

17. Pirard C, Sagot C, Deville M, Dubois N, Charlier C. Urinary levels of bisphenol A, triclosan and 4-nonylphenol in a general Belgian population. Environment International 1 Nov. 2012;48:7883. https://doi.org/10.1016/j.envint.2012.07.003.

18. Pirard C, Charlier C. Simple and fast method for the measurement of legacy and novel brominated flame retardants in human serum. Chemosphere 1 Nov. 2018;211:91825. https://doi.org/10.1016/j.chemosphere.2018.08.012.

19. Dufour P, Pirard C, Charlier C. Validation of a novel and rapid method for the simultaneous determination of some phenolic organohalogens in human serum by GC–MS. Journal of

ChromatographyB.15nov2016;10361037:6675. https://doi.org/10.1016/j.jchromb.2016.10.002.

20. Dewalque L, Pirard C, Charlier C. Measurement of Urinary Biomarkers of Parabens, Benzophenone-3, and Phthalates in a Belgian Population. Biomed Res Int. 2014. https://doi.org/10.1155/2014/649314.

21. Schulz C, Butte W. Revised reference value for pentachlorophenol in morning urine. International Journal of Hygiene Environmental Health 3 Déc. 2007;210(6):7414. https://doi.org/10.1016/j.ijheh.2006.11.004.

22. Molina-Villalba I, Lacasaña M, Rodríguez-Barranco M, Hernández AF, Gonzalez-Alzaga B, Aguilar-Garduño C, et al. Biomonitoring of arsenic, cadmium, lead, manganese and mercury in urine and hair of children living near mining and industrial areas. Chemosphere 1 Avr. 2015;124:8391. https://doi.org/10.1016/j.chemosphere.2014.11.016.

23. Musa Obadia P, Kayembe-Kitenge T, Haufroid V, Banza Lubaba Nkulu C, Nemery B. Preeclampsia and blood lead (and other metals) in Lubumbashi, DR Congo. Environ Res. 2018;167:46871. https://doi.org/10.1016/j.envres.2018.07.032.

24. Fierens S, Rebolledo J, Versporten A, Brits E, Haufroid V, De Plaen P, et al. Human biomonitoring of heavy metals in the vicinity of non-ferrous metal plants in Ath, Belgium. Arch Public Health. 2016;74:42. https://doi.org/10.1186/s13690-016-0154-8.

25. Connolly A, Leahy M, Jones K, Kenny L, Coggins MA. Glyphosate in Irish adults – A pilot study in 2017. Environmental Research 1 Août. 2018;165:2356. https://doi.org/10.1016/j.envres.2018.04.025.

26. Conrad A, Schröter-Kermani C, Hoppe H-W, Rüther M, Pieper S, Kolossa-Gehring M. Glyphosate in German adults – Time trend (2001 to 2015) of human exposure to a widely used herbicide. International Journal of Hygiene Environmental Health 1 Janv. 2017;220(1):816. https://doi.org/10.1016/j.ijheh.2016.09.016.

27. Li A, Zhuang T, Shi J, Liang Y, Song M. Heavy metals in maternal and cord blood in Beijing and their efficiency of placental transfer. J Environ Sci (China) Juin. 2019;80:99106. https://doi.org/10.1016/j.jes.2018.11.004.

28. Tuakuila J, Kabamba M, Mata H, Mata G. Blood lead levels in children after phase-out of leaded gasoline in Kinshasa, the capital of Democratic Republic of Congo (DRC). Arch Public Health 4 Avr. 2013;71(1):5. https://doi.org/10.1186/0778-7367-71-5.

29. Zeman FA, Boudet C, Tack K, Floch Barneaud A, Brochot C, Péry ARR, et al. Exposure assessment of phthalates in French pregnant women: Results of the ELFE pilot study. International Journal of Hygiene Environmental Health 1 Juin. 2013;216(3):2719. https://doi.org/10.1016/j.ijheh.2012.12.005.

30. Frederiksen H, Nielsen JKS, Mørck TA, Hansen PW, Jensen JF, Nielsen O, et al. Urinary excretion of phthalate metabolites, phenols and parabens in rural and urban Danish mother–child pairs. International Journal of Hygiene Environmental Health 1 Nov. 2013;216(6):77283. https://doi.org/10.1016/j.ijheh.2013.02.006.

31. Lin S, Ku H-Y, Su P-H, Chen J-W, Huang P-C, Angerer J, et al. Phthalate exposure in pregnant women and their children in central Taiwan. Chemosphere 1 févr. 2011;82(7):94755. https://doi.org/10.1016/j.chemosphere.2010.10.073.

32. Jiménez-Díaz I, Artacho-Cordón F, Vela-Soria F, Belhassen H, Arrebola JP, Fernández MF, et al. Urinary levels of bisphenol A, benzophenones and parabens in Tunisian women: A pilot study. Science of The Total Environment 15 août. 2016;562:818. https://doi.org/10.1016/j.scitotenv.2016.03.203.

33. Adoamnei E, Mendiola J, Moñino-García M, Vela-Soria F, Iribarne-Durán LM, Fernández MF, et al. Urinary concentrations of parabens and reproductive parameters in young men. Science of The Total Environment. 15 avr 2018; 621:2019. https://doi.org/10.1016/j.scitotenv.2017.11.256.

34. Wu C, Feng C, Qi X, Wang G, Zheng M, Chang X, et al. Urinary metabolite levels of pyrethroid insecticides in infants living in an agricultural area of the Province of Jiangsu in China. Chemosphere Mars. 2013;90(11):270513. https://doi.org/10.1016/j.chemosphere.2012.11.050.

35. Qi X, Zheng M, Wu C, Wang G, Feng C, Zhou Z. Urinary pyrethroid metabolites among pregnant women in an agricultural area of the Province of Jiangsu, China. International Journal of Hygiene Environmental Health 1 sept. 2012;215(5):48795. https://doi.org/10.1016/j.ijheh.2011.12.003.

36. Dereumeaux C, Saoudi A, Goria S, Wagner V, De Crouy-Chanel P, Pecheux M, et al. Urinary levels of pyrethroid pesticides and determinants in pregnant French women from the Elfe cohort. Environment International 1 oct. 2018;119:8999. https://doi.org/10.1016/j.envint.2018.04.042.

37. Panuwet P, Prapamontol T, Chantara S, Thavornyuthikarn P, Montesano MA, Whitehead RD, et al. Concentrations of urinary pesticide metabolites in small-scale farmers in Chiang Mai Province, Thailand. Science of The Total Environment 15 déc. 2008;407(1):65568. https://doi.org/10.1016/j.scitotenv.2008.08.044.

38. Ein-Mor E, Ergaz-Shaltiel Z, Berman T, Göen T, Natsheh J, Ben-Chetrit A, et al. Decreasing urinary organophosphate pesticide metabolites among pregnant women and their offspring in Jerusalem: Impact of regulatory restrictions on agricultural organophosphate pesticides use? International Journal of Hygiene Environmental Health 1 Juin. 2018;221(5):77581. https://doi.org/10.1016/j.ijheh.2018.03.013.

39. Skarha J, Mínguez-Alarcón L, Williams PL, Korevaar TIM, de Poortere RA, Broeren MAC, et al. Cross-sectional associations between urinary triclosan and serum thyroid function biomarker concentrations in women. Environment International 1 janv. 2019;122:25662. https://doi.org/10.1016/j.envint.2018.11.015.

40. Kim K, Park H, Yang W, Lee JH. Urinary concentrations of bisphenol A and triclosan and associations with demographic factors in the Korean population. Environmental Research 1 nov. 2011;111(8):12805. https://doi.org/10.1016/j.envres.2011.09.003.

41. Adoamnei E, Mendiola J, Vela-Soria F, Fernández MF, Olea N, Jørgensen N, et al. Urinary bisphenol A concentrations are associated with reproductive parameters in young men. Environmental Research 1 févr. 2018;161:1228. https://doi.org/10.1016/j.envres.2017.11.002.

42. Mercado LA, Freille SM, Vaca-Pereira JS, Cuellar M, Flores L, Mutch E, et al. Serum concentrations of p,p'-dichlorodiphenyltrichloroethane (p,p'-DDE) in a sample of agricultural workers from Bolivia. Chemosphere Juin. 2013;91(10):13815. https://doi.org/10.1016/j.chemosphere.2012.12.023.

43. Nuapia Y, Chimuka L, Cukrowska E. Assessment of organochlorine pesticide residues in raw food samples from open markets in two African cities. Chemosphere 1 déc. 2016;164:4807. https://doi.org/10.1016/j.chemosphere.2016.08.055.

44. Charlier C, Plomteux G. Endocrine disruption and organochlorines residues. Acta Clinica Belgica, supplement 2002-1.

45. Helou K, Harmouche-Karaki M, Karake S, Narbonne J-F. A review of organochlorine pesticides and polychlorinated biphenyls in Lebanon: Environmental and human contaminants. Chemosphere 1 sept. 2019;231:35768. https://doi.org/10.1016/j.chemosphere.2019.05.109.

46. Murray J, Eskenazi B, Bornman R, Gaspar FW, Crause M, Obida M, et al. Exposure to DDT and hypertensive disorders of pregnancy among South African women from an indoor residual spraying region: The VHEMBE study. Environ Res. 2018;162:4954. https://doi.org/10.1016/j.envres.2017.12.006.

47. Pavuk M, Olson JR, Sjödin A, Wolff P, Turner WE, Shelton C, et al. Serum concentrations of polychlorinated biphenyls (PCBs) in participants of the Anniston Community Health Survey. Science of The Total Environment 1 mars. 2014;473474:28697. https://doi.org/10.1016/j.scitotenv.2013.12.041.

48. Dufour P, Pirard C, Petrossians P, Beckers A, Charlier C. Association between mixture of persistent organic pollutants and thyroid pathologies in a Belgian population. Environmental Research 1 févr. 2020;181:108922. https://doi.org/10.1016/j.envres.2019.108922.

49. Gaylord A, Berger KI, Naidu M, Attina TM, Gilbert J, Koshy TT, et al. Serum perfluoroalkyl substances and lung function in adolescents exposed to the World Trade Center disaster. Environmental Research 1 mai. 2019;172:26672. https://doi.org/10.1016/j.envres.2019.02.024.

50. Jiang W, Zhang Y, Zhu L, Deng J. Serum levels of perfluoroalkyl acids (PFAAs) with isomer analysis and their associations with medical parameters in Chinese pregnant women. Environment International 1 mars. 2014;64:407. https://doi.org/10.1016/j.envint.2013.12.001.

51. Sochorová L, Hanzlíková L, Černá M, Drgáčová A, Fialová A, Švarcová A, et al. Perfluorinated alkylated substances and brominated flame retardants in serum of the Czech adult population. International Journal of Hygiene Environmental Health 1 mars 2017; 220(2, Part A):23543. https://doi.org/10.1016/j.ijheh.2016.09.003.

52. Dufour P, Pirard C, Charlier C. Determination of phenolic organohalogens in human serum from a Belgian population and assessment of parameters affecting the human contamination. Science of The Total Environment 1 déc. 2017;599600:185666. https://doi.org/10.1016/j.scitotenv.2017.05.157.

53. Fromme H, Becher G, Hilger B, Völkel W. Brominated flame retardants – Exposure and risk assessment for the general population. Int J Hyg Environ Health. 2016;219:1–23. https://doi.org/10.1016/j.ijheh.2015.08.004.

Tables

Table 1 : LOQ of methods (pg/mL) and DF (%) of environmental pollutants in Kinshasa

matrix	pollutant	LOQ	DF	matrix	pollutant	LOQ	DF
Inorganics Pollutants				*nPOPs*			
Urine	Vanadium	180	73	*Urine*	*Parabens*		
	Chrome	230	100		MeP	790	100
	Manganese	890	20		EP	300	67
	Cobalt	130	100		PrP	360	100
	Nickel	1630	100		*Phthalates*		
	Cadmium	120	93		MEP	940	100
	Tin	730	47		MEHP	620	100
	Antimony	140	47		MnBP	990	100
	Thalium	90	100		MiBP	1230	100
	Bismuth	120	100		MBzP	610	87
	Copper	1600	100		*Benzophenone*		
	Zinc	15000	100		BP3	670	100
	Arsenic	140	100		*Pyrethroid pesticides*		
	Selenium	7900	100		c-DCCA	150	87
	Molybdene	5000	100		t-DCCA	150	93
Blood	Lead	500	100		TCPY	80	100
POPs					DBCA	300	87
Serum	*Organochlorine pesticides*				FPBA	110	7
	4,4'-DDE	400	87		3-PBA	90	100
	HCB	80	7		*Triclosan*		
	β-HCH	50	0		TCS	200	100
	Polychlorinated biphenyls				*Bisphenols*		
	PCB 153	70	60		BPF	70	67
	PCB 138	150	7		BPA	290	100
	PCB 180	50	53		BPZ	60	0
	Perfluoroalkyl substances				BPS	90	80
	PFOA	250	100		*Organophosphate pesticide*		
	PFOS	500	47		DEP	500	53
	PFHxS	150	60		DETP	500	47
	PFNA	100	60		*Glyphosate*		
	Brominated flame retardants				Glyphosate	80	93
	PBDE 47	3.7	27				
	PBDE 153	4.2	27				
	Phenolic organohalogens						
	PCP	44.6	14				
	2,4,6 TBP	49.6	0				
	4-OH CB 146	2.2	29				
	4-OH CB 187	2	57				
	6-OH BDE 47	2.5	0				

LOQ : Limit of Quantification ,

DF : Detection Frequency

Table 2: Mean, geometric mean, median and range concentrations in urine [µg/L (µg/g creatinine)] and at both blood and serum in µg/L.

Pollutant	Mean	Geometric mean	Median	Minimum	Maximum
Urinary					
Cobalt	1.0 (0.31)	0.57 (0.24)	0.43 (0.21)	0.16 (0.089)	6.11 (0.87)
Cadmium	1.15 (0.33)	0.66 (0.28)	0.61 (0.26)	‹LOQ (‹LOQ)	6.12 (0.87)
Arsenic	81.74 (32.33)	69.73 (29.79)	70.91 (30.86)	32.12 (11.97)	215.32 (57.28)
Glyphosate	0.22 (0.095)	0.19 (0.083)	0.23 (0.098)	0.09 (0.05)	0.40 (0.18)
MeP	699.98 (335.48)	216.12 (92.32)	445.39 (121.96)	15.06 (9.94)	4467.5 (1386.5)
EP	11.94 (2.96)	0.73 (0.31)	0.47 (0.25)	‹LOQ (‹LOQ)	86.5 (26.83)
PrP	290.62 (157.53)	38.96 (16.64)	31.35 (5.35)	0.97 (0.65)	2509.15 (778.75)
MEP	309.94 (96.09)	113.09 (48.31)	108.56 (41.96)	13.96 (9.45)	2366.9 (734.6)
MEHP	16.47 (6.26)	8.53 (3.64)	9.03 (3.25)	0.97 (0.84)	62.45 (15.75)
MnBP	229.67 (92.86)	176.42 (75.36)	145.19 (60.72)	32.05 (31.79)	638.11 (292.3)
MiBP	31.39 (13.42)	25.26 (10.79)	26.52 (9.32)	7.43 (4.1)	72.37 (27.97)
BP3	6.76 (2.59)	4.97 (2.12)	5.00 (1.93)	0.76 (0.65)	23.31 (6.67)
c-DCCA	0.81 (0.28)	0.41 (0.18)	0.47 (0.23)	‹LOQ (‹LOQ)	3.9 (0.69)
t-DCCA	1.34 (0.47)	0.72 (0.31)	0.59 (0.31)	‹LOQ (‹LOQ)	0.17 (0.12)
TCPY	37.68 (13.52)	9.19 (3.92)	4.43 (2.19)	0.40 (0.44)	123.36 (54.6)
3-PBA	16.88 (4.84)	3.36 (1.44)	2.25 (1.22)	0.29 (0.15)	173.20 (48.50)
TCS	90.44 (44.45)	40.88 (17.46)	40.13 (17.76)	4.48 (0.64)	277.82 (184.70)
BPA	1.96 (0.88)	1.62 (0.69)	1.36 (0.76)	0.52 (0.38)	5.40 (2.08)
DEP	4.30 (1.24)	0.30 (0.13)	0.87 (0.19)	‹LOQ (‹LOQ)	32.70 (9.18)
DETP	1.95 (0.63)	0.76 (0.33)	0.35 (0.24)	‹LOQ (‹LOQ)	9.23 (2.34)
Blood					
Lead	62.96	53.69	53.57	22.34	156.96
Serum					
4,4'DDE	3.02	1.69	1.46	‹LOQ	9.20
PCB 153	0.09	0.08	0.08	‹LOQ	0.20
PFOA	0.49	0.47	0.48	0.25	0.85
PFOS	0.58	‹LOQ	0.50	‹LOQ	1.54
PCP	‹LOQ	‹LOQ	‹LOQ	‹LOQ	102.4[a]
4-OH CB187	7.48[a]	‹LOQ	2.44[a]	‹LOQ	20.4[a]
PBDE47	‹LOQ	‹LOQ	‹LOQ	‹LOQ	15.06[a]
PBDE 153	‹LOQ	‹LOQ	‹LOQ	‹LOQ	13.45[a]

a : concentration in pg/mL

LOQ : Limit of Quantification

Table 3: Pollutants median values (µg/L) measured in urine, blood and serum from different worldwide populations.

Pollutant	matrix	Year of collection	Population	Median	Reference
Arsenic	*Urine*				
		2012	Huelva (Spain), school children, N=261	1.17	[22]
		2012-2013	Lubumbashi (DRC), pregnant women (control), N=39	23.60[a]	[23]
		2019	**Kinshasa (DRC), population, N=15**	**70.90**	**Current study**
Cobalt	*Urine*				
		2009	Ath-Belgium, men population, N=52	0.16[a]	[24]
		2012-2013	Lubumbashi (DRC), pregnant women (control), N=39	6.97[a]	[23]
		2019	**Kinshasa (DRC), population, N=15**	**0.43**	**Current study**
Cadmium	*Urine*				
		2009	Ath-Belgium, men population, N=52	0.21[a]	[24]
		2011-2012	Belgium, mother, N=125	0.22	[7]
		2012	Huelva (Spain), school children, N=261	0.29	[22]
		2012-2013	Lubumbashi (DRC), pregnant women (control), N=39	0.60[a]	[23]
		2019	**Kinshasa (DRC), population, N=15**	**0.61**	**Current study**
Lead	*Blood*				
		2009	Ath-Belgium, men population, N=52	31.70[a]	[24]
		2011	Kinshasa (DRC), children 1-5 years, N=100	86.00	[28]
		2012-2013	Lubumbashi (DRC), pregnant women (control), N=39	50.80[a]	[23]
		2015-2016	Beijing, maternal blood, N=156	17.60	[27]
		2019	**Kinshasa (DRC), population, N=15**	**53.60**	**Current study**
Glyphosate	*Urine*				
		2012	German, adult population, N=40	0.11	[26]
		2017	Irish, adult population, N=50	0.87	[25]
		2019	**Kinshasa (DRC), population, N=15**	**0.23**	**Current study**
BP3	*Urine*				
		2011	Danish, mother, N=145	3.70	[30]
		2012	Tunisia, women, N=34	1.73	[32]
		2013	Belgium, adult population, N=261	1.30	[20]
		2019	**Kinshasa (DRC), population, N=15**	**5.00**	**Current study**
4,4'DDE	*Serum*				
		2010-2011	Bolivian, women agriculture, N=24	9.34	[42]
		2012-2013	South-Africa, pregnant women, N=733	1.75[b]	[46]
		2013-2015	Lebanon, population, N=314	0.13[b]	[45]
		2015	Belgium, adult population (women), N=124	0.41	[1]
		2019	**Kinshasa (DRC), population, N=15**	**1.46**	**Current study**
PCB 153	*Serum*				
		2005-2007	Anniston Community (USA), African-American, N=353	1.42[b]	[47]
		2013-2015	Lebanon, population, N=314	0.12[b]	[45]
		2015	Belgium, adult population, N=251	0.36	[1]
		2019	**Kinshasa (DRC), population, N=15**	**0.081**	**Current study**

[a] geometric mean

[b] ng /lipid weight concentrations converted to µg / L after multiplation by mean body lipid concentration (0.00735)

Table 4: pollutants median values (µg/L) measured in urine and serum from different worldwide populations.

Year of collection	Population	Parabens in urine			Perfluoroalkyl substances in serum		Alkylphosphates in urine		BPA in urine	TCS in urine	Reference
		MeP	EP	PrP	PFOS	PFOA	DEP	DETP			
2013	Belgium,adult,population,N=261	16.1	1.7	1.2	-	-	-	-	-	-	[20]
2011	Danish, mother, N=145	14	0.89	1.7	-	-	-	-	-	-	[30]
2010-2011	Spain, Young men, N=215	17	1.8	0.7	-	-	-	-	-	-	[33]
2012	Tunisia, women,N=34	34.94	1.77	3.06	-	-	-	-	-	-	[32]
2015-2018	Belgium, population, N=237	-	-	-	3.61	1.6	-	-	-	-	[48]
2014-2016	USA,adolescents exposed,N=118	-	-	-	3.72	1.8	-	-	-	-	[49]
2012	China, pregnant women, N=141	-	-	-	4.31	3.95	-	-	-	-	[50]
2015	Czech,adult population, N=300	-	-	-	2.43	0.76	-	-	-	-	[51]
2015	Belgium, children, N=240	-	-	-	-	-	1.8	-	-	-	[15]
2006	Thailand, famers, N=136	-	-	-	-	-	<LOQ	1.2	-	-	[37]
2012-2016	Jerusalem, Pregnant women, N= 273	-	-	-	-	-	2.72	0.55	-	-	[38]
2011	Belgium, population, N=131	-	-	-	-	-	-	-	2.46	2.24	[17]
2009	Korea, adult population, N=1870	-	-	-	-	-	-	-	2.07	1.53	[40]
2010-2011	Murcia (Spain),Young men, N=215*	-	-	-	-	-	-	-	2.3	-	[41]
2012	Tunisia, women,N=34	-	-	-	-	-	-	-	0.35	-	[32]
2011	Danish, mother, N=145	-	-	-	-	-	-	-	2.1	0.64	[30]
2019	Kinshasa (DRC), population, N=15	445.39	0.47	31.35	0.5	0.482	0.87	<LOQ	1.36	40.13	Current study

BPA: Bisphenol A; TCS: Triclosan; MeP: methylparaben; EP: ethylparaben; PrP: n-propylparaben; PFOS: perfluoro-octane sulfonic; PFOA: perfluoroctanoic acid; DEP: diethylphosphate; DETP: diethylthiophosphate.

Figures

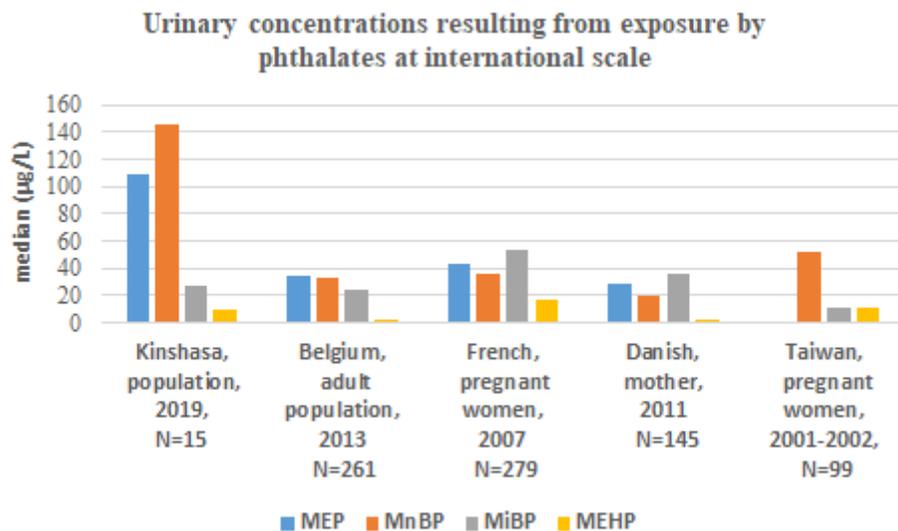Urinary concentrations resulting from exposure by phthalates at international scale

Figure 1

Urinary concentrations resulting from exposure by phthalates at international scale MEP: Monoethyl Phthalate; MnBP: mono-n-butyl phthalate; MiBP: mono-iso-butyl phthalate; MEHP: mono-2-ethylhexyl phthalate.
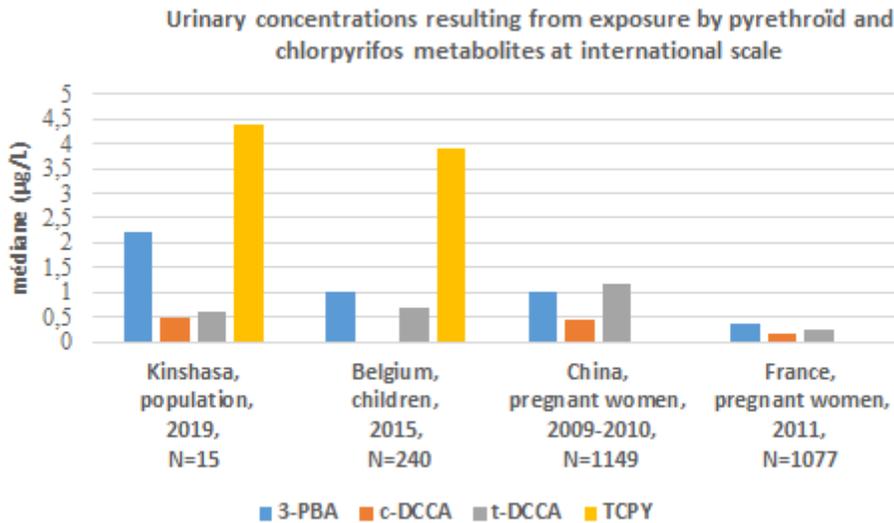

Urinary concentrations resulting from exposure by pyrethroïd and chlorpyrifos metabolites at international scale

Figure 2

Urinary concentrations resulting from exposure by pyrethroïd and chlorpyrifos metabolites at international scale 3-PBA : 3-phenoxybenzoic acid ; c-DCCA : cis-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropane-1-carboxylic acid ; t-DCCA : trans-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropane-1-carboxylic acid ; TCPY : 3,5,6-trichloro-2-pyridinol.

Supplementary Files

This is a list of supplementary files associated with this preprint. Click to download.

- Supplementaryinformation.docx