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Contamination of water and food crops by trace elements in the African Copperbelt: A collaborative cross-border study in Zambia and the Democratic Republic of Congo

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ABSTRACT

Background. Environmental pollution by trace metals is of concern in the African Copperbelt, a region of intense mining situated on either side of the border between the Democratic Republic of Congo (DRC) and Zambia. Scientists from both countries undertook a collaborative study using the same sampling and analytical methodologies to assess metal contamination of water and food in both countries.

Methods. In six mining areas (Lubumbashi, Likasi, Kipushi in DRC; Chingola, Kitwe, Luanshya in Zambia) samples of water (surface, boreholes, taps) and various crops were collected in two residential sites near mining operations, and in rural villages farther away (controls). Nine elements (Mn, Co, Ni, Cu, Zn, As, Cd, Pb, U) were measured by ICP-MS or ICP-OES.

Results. In drinking water obtained close to mining, the median concentrations (μg/L) of all trace elements, except Zn, were substantially higher in DRC (n=20) than Zambia (n=18), this being most pronounced for Pb (27 vs 0.08), Cd (0.7 vs <0.015) and Co (65 vs 1.3). In plants, most of which were leafy vegetables, the concentrations of all trace elements, except Mn, were consistently higher in DRC (n=145) than Zambia (n=128). Compared to control sites, crops obtained near mining exhibited significantly higher concentrations of Zn and Pb in Zambia, and of Co, Cu, As, Cd, Pb and U in DRC. Cd and Pb exceeded international standards in most DRC crop samples.

Conclusion. This unique cross-border study documented metal contamination of drinking water and locally grown vegetables near mining activities in Zambia and DRC, with the contamination being worse in DRC. The higher pollution observed in DRC compared to Zambia must largely be attributed to differences in environmental governance between the two countries. Efforts are needed to enforce regulations and to mitigate the negative impacts of mining and smelting operations on the environment, population health, and food security in the African Copperbelt.

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Capsule

In a cross-border collaborative study, scientists evaluated mining-related pollution in the African Copperbelt and established that pollution is much worse in the DRC than in Zambia

Introduction

The central African Copperbelt is a geologic region, also known as the Lufilian Arc, situated in the southern part of the former Katanga Province in the Democratic Republic of Congo (DRC) and the Copperbelt and Northwestern Provinces in Zambia. This region constitutes one of the largest mining areas in Africa, stretching over more than 500 km from Kolwezi in the DRC to Luanshya in Zambia (Butler, 2006). From the colonial period until now, ores containing copper (Cu) and various other metals – most notably cobalt (Co), but also zinc (Zn), cadmium (Cd) and uranium (U) – have been extracted, in both countries, from large-scale mines and then further processed and refined in local smelters. From the end of the 20th century onwards, artisanal mining has also contributed to a substantial share of cobalt production in the DRC (Yager, 2012). Mining activities account for a major part of the DRC's and Zambia's current export earnings and they thus contribute to a substantial part of their GDP (Ministry of Mining and Hydrocarbons, 2003; SADC/FANR, 2011; Fraser and Lungu, 2007; Mwitwa and Kabemba, 2007). However, decades of mining activities have also caused considerable pollution of ecosystems, through discharges in surface waters, air pollution from tailing dust and emissions from smelter stacks (Feeney, 2001).

Besides mining, about 70% of the population in both countries depend on agriculture for livelihoods; agriculture is also a significant source of exports and intra-regional trade (SADC/FANR, 2011). The production of food crops is mainly by smallholder farmers in rural areas, who rely on rain-fed and traditional farming dominated mainly by root and tubers, plantains, maize, rice, groundnuts, beans, sorghum, millets and vegetables (SADC/FANR, 2011; Muimba-Kankolongo, 2018). However, food crops are also widely grown in kitchen gardens in urban and suburban areas (Peša, 2020a). In recent years, recurrent and prolonged drought have led to food insecurity in the region (Chapoto et al. 2018; Muimba-Kankolongo, 2018) but, in addition, food security is being compromised, to a hitherto poorly quantified degree, by the degradation of the environment, including farmlands, caused by mineral extraction and operations of metallurgic industries (Mwitwa et al., 2012).

Despite numerous reports on environmental contamination by mining and metallurgic industries, no common research has been conducted between the DRC and Zambia to tackle their common challenges regarding environmental pollution and its related challenges regarding food and nutrition security and public health challenges. The objectives of this first ever collaborative study were to assess levels of metal contamination in water and crops intended for human consumption, with the long-term aim of improving communities' livelihoods and protecting public health. Providing objective data on the degree of mining-related contamination by trace metals should contribute to setting up governance guidelines and arrangements to minimize environmental damage in the Copperbelt of the DRC and Zambia.

The study was designed to test the hypotheses that water and locally cultivated crops would show higher contents of trace elements when sampled close to mining activities than in control sites far from mining activities. However, another important, previously untested hypothesis was that the degree of contamination by trace elements would differ between sites in Zambia and sites in the DRC.

Materials and methods

Study sites

The field studies were carried out between March 29th and April 8th, 2011 in two adjacent provinces with copper-cobalt extraction and processing, namely the Haut-Katanga Province in the DRC and the Copperbelt Province in Zambia. Based on past and current mining activities, as well as reports or complaints about environmental contamination from plant effluents and spills or discharges from tailings, three main locations were selected in each country: the districts of Chingola (CHI), Kitwe (KIT) and Luanshya (LUA) in Zambia, and the cities of Lubumbashi (LUB), Likasi (LIK) and Kipushi (KIP) in the DRC (**Fig. 1**). Within each of these locations, two urban/suburban residential sites in close proximity (from 100 m to a few kilometers) of mining or metallurgic activities were selected for sampling, as well as one or more rural villages far away (more than a kilometer but less than 20 km) from the mines to serve as controls. The selection of the sites was based on accessibility and the observable or presumed degree of pollution by neighbouring mining or metallurgic activities, on the one hand, and for the control sites on the absence of visible sources of industrial pollution, on the other hand. The names and location (determined by GPS) of the sampling sites are provided in Fig. 1. In each of the six locations, the sampling campaigns were done in one day (except two days for Lubumbashi).

The sampling campaigns were authorized by the relevant administrative authorities. In each location, the purpose of the survey was first explained to local community representatives and samples were always obtained without coercion.

Sample collection, preparation, and analysis

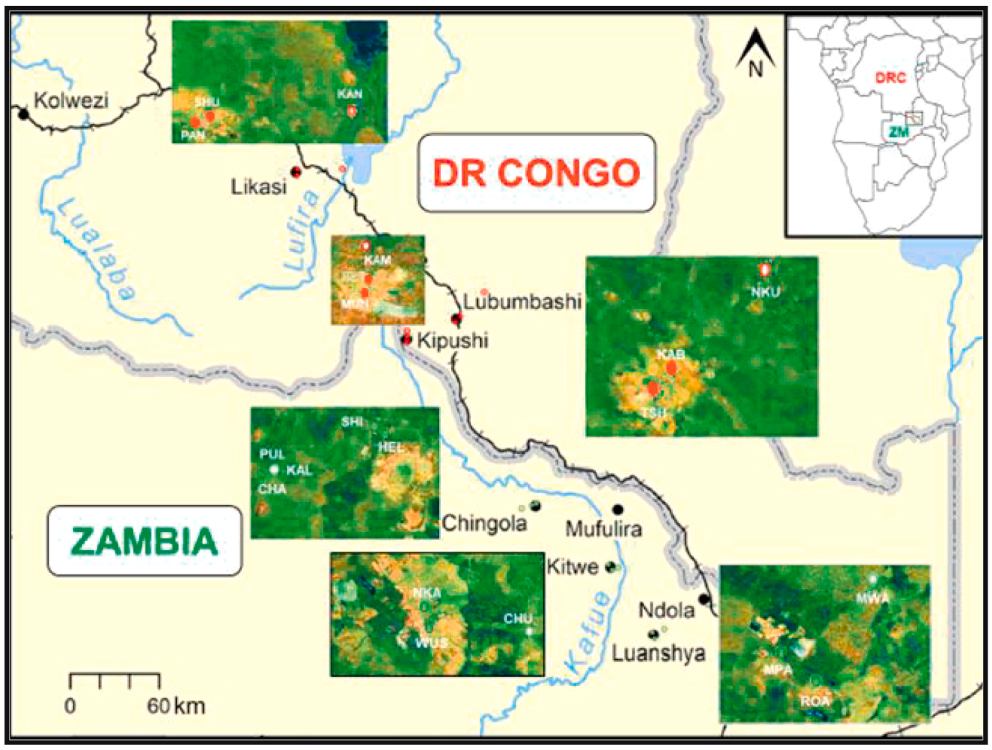
Water samples

Water samples were collected from rivers (17 samples), domestic wells or boreholes (37 samples) and (communal) taps (23 samples), as available in the various sites. Samples from rivers were collected from the water body by pointing open bottles against the current and avoiding floating debris. Adequate water quantities were obtained from a full flow-out of faucets/taps after taps or pumps had been left to run for at least two minutes to clear the line. Water was slowly filled in 40 mL clean polystyrene vessels with leak-proof screw caps (Plastiques Gosselin, Hazebrouck, France) to below the container rim before tightening the lids securely. In the deep dug boreholes or wells, water was obtained using tins cut on one-side or plastic containers connected to long wooden canes with wire or rope and then filled in the polystyrene vessels. Collected samples were then pipetted into 1.5 mL Eppendorf tubes, and transported in cooler boxes to the laboratory to be stored in a freezer at -20°C until shipping in sealed cooler boxes with ice packs for analysis in Belgium.

Samples of water were analyzed in the internationally accredited laboratory of the Louvain Centre for Toxicology and Applied Pharmacology (Université catholique de Louvain, Brussels, Belgium), using validated procedures for the determination of trace metals in urine, as described in Hoet et al. (2013). Water samples were not filtered prior to analysis. Briefly, water specimens (500 μL) were diluted (1+9) with a HNO_3 1%, HCl 0.5% solution containing internal standards (Sc, Ge, Rh, Ir), and 24 elements were quantified by inductively coupled plasma mass spectrometry (ICP-MS) on an Agilent 7500ce instrument (Agilent Technologies), using quality control and quality assurance procedures, as described in Hoet et al. (2013). For the present study, we considered nine elements (Mn, Co, Ni, Cu, Zn, As, Cd, Pb and U). Samples below the Limit of Detection (LOD) were attributed a value of LOD/2, using previously determined LODs (Hoet et al., 2013).

Food crop samples

Village headmen and community members in selected



Samples of water and locally cultivated crops were obtained from six locations, three in Zambia and three in DR Congo; in each main location there were two (sub)urban sites close to mines or smelters (full symbols) and one rural site at some distance (open symbols). Inserts are reproduced from Google Earth with each sampling site labelled by a three-letter acronym, as explained below. Note: the scales are not identical for all inserts. Zambia: KITWE district: Wusakile compound (WUS), Nkana West compound (NKA) and Chankalamu village (CHU); CHINGOLA district: Heleni Kasenji village (HEL), Shimulala village (SHI), Kalipenta village (KAL), Pule village (PUL) and Chama village (CHA); LUANSHYA district: Roan (ROA), Mpatamatu (MPA) and Mwansa village (MWA). DR Congo: LUBUMBASHI: Kabecha (KAB), Tshansansa (TSH) and Nkumanwa village (NKU); LIKASI: Shituru (SHU), Panda (PAN) and Kansalabwe village (KAN); KIPUSHI: Q. Mungotshi (MUN), Q.Betty (BET) and Kampemba (KAM).
Map based on a publication by I. Peša (2020a)

Fig. 1. Map of the study sites showing the different locations of the central African Copperbelt where the survey took place.

neighbourhoods were first informed about the study and requested permission to obtain samples from their farms or gardens. Samples of edible food crops were collected, generally with the assistance of the owners or community members, according to their availability in the backyard homestead gardens or in the fields. Consequently, the numbers and types of collected crop samples varied from one location to another. The most commonly available food crops (Table A in the Supplement) consisted of leafy vegetables [leaves of amaranth (*Amaranthus hybridus*) (50 samples), pumpkins (*Cucurbita maxima*) (50 samples), cassava (*Manihot esculenta*) (41 samples), and sweetpotato (*Ipomea batatas*) (36 samples)], tomato (*Lycopersicon esculentum*) (37 samples) and common beans (*Phaseolus vulgaris*) (30 samples). Samples of maize grains (*Zea mays*) (16 samples), sweetpotato tubers (10 samples) and cassava tubers (3 samples) were also collected. Occasionally, other plants [onions: *Allium cepa* (5 samples), cabbage: *Brassica oleracea* (2 samples)] were also obtained (and analysed) but these results are not included because they were too few to be meaningful.

The collected samples were identified and suitably packaged in paper bags. They were then pre-treated, in the Laboratory of Environmental Engineering of the School of Mines and Mineral Schemes at Copperbelt University in Zambia and the Laboratory of Biology at the 'Institut Supérieur Pédagogique' in Lubumbashi in the DRC, by rinsing in tap water to remove soil particles and then 3-4 times with distilled water, before they were sliced with a clean knife, oven-dried at 70°C to constant weight for 48 hours, and separately homogenized by mechanical grinding using porcelain mortars and pestles. The weight of each sample powder was recorded before packing in sealed minigrip bags which were maintained at ambient temperature and later shipped to Belgium for laboratory analysis.

The specimens were analyzed in the Division of Water and Soil Management of the KU Leuven, as described in Cheyns et al. (2014). Briefly, samples (~50 mg) were digested in 2 mL boiling 65% HNO_3 and analysed for metal content by ICP-MS (Thermo Fisher Scientific, Walton, MA, USA) for As, Co, Cd and Pb, or inductively coupled optical emission spectroscopy (ICP-OES, Perkin-Elmer Optima 3300 DV, Norwalk, CT, USA) for the other elements. Each analytical run included certified reference material NIST SRM-1573 (tomato leaves, National Institute of Standards and Technology, Gaithersburg, MD, USA) and appropriate blank samples for the determination of limits of quantification (twice SD of 6 blanks). For the present study, the following nine elements were considered: Mn, Co, Ni, Cu, Zn, As, Cd, Pb and U.

Statistical analysis

Since data were generally non-normally distributed, results are presented as medians with 5^{th}, 25^{th}, 75^{th} and 95^{th} percentiles, and non-

Table 1
Concentration of trace elements (μg/L) in unfiltered water samples from Zambia and the DRC.

		ZAMBIA (n=42)		DRC (n=35)			
	WHO limit μg/L	P50 [P25-P75] μg/L	>WHO limit n (%)	P50 [P25-P75] μg/L	>WHO limit n (%)	P50 DRC / P50 Zambia	P*
Mn	50	**12.6** [8.00-43.1]	9 (21%)	**79.8** [15.8-2606]	19 (51%)	**6.3**	0.001
Co	NA	**2.04** [0.83-8.61]		**80.0** [21.4-2624]		**39**	<0.001
Ni	70	**1.74** [1.02-6.53]	0	**7.55** [2.16-119]	11 (31%)	**4.3**	<0.001
Cu	2000	**5.09** [1.40-17.8]	1 (2%)	**139** [27.4-3416]	11 (31%)	**27**	<0.001
Zn	5000	**14.6** [4.74-27.8]	1 (2%)	**27.3** [8.31-482]	4 (11%)	**1.9**	0.021
As	10	**0.167** [0.098-0.244]	0	**1.89** [0.504-23.7]	11 (31%)	**11**	<0.001
Cd	3	***0.008*** [*0.008*-0.017]	0	**0.416** [0.049-6.12]	11 (31%)	**55**	<0.001
Pb	10	**0.080** [0.030-0.690]	1 (2%)	**14.2** [1.28-354]	21 (60%)	**177**	<0.001
U	30	**0.033** [*0.004*-0.172]	0	**0.808** [0.131-3.21]	8 (23%)	**24**	<0.001

WHO limit is upper limit for drinking water according to World Health Organization guidelines (not available for Co); P50, P25 and P75 are 50th (median), 25th and 75th percentiles, respectively

* p value for difference between Zambia and DRC (Mann-Whitney test); values in italics are imputed values (half the value of Limit of Detection). See Supplemental Table B for more details

parametric tests (Mann-Whitney and Kruskall-Wallis tests, Spearman correlations) were applied for statistical analyses, using GraphPad Prism version 8.0.0 for Windows (GraphPad Software, San Diego, CA, USA). A level of $p<0.05$ (two-sided) was considered statistically significant.

Results

Contamination of water by trace elements

The concentrations of nine elements in unfiltered water samples taken from Zambia (n=42) and the DRC (n=35) are shown in Table 1 and in Supplemental Table B**.** For all elements, the concentrations spanned several orders of magnitude between the lowest and highest concentrations, e.g. from below 0.1 μg/L to above 100 mg/L for Cu. The concentrations were significantly ($p<0.001$, except for Zn) higher in water samples obtained from the DRC than in those obtained from Zambia. The ratios of median concentrations of values obtained in the DRC over values obtained in Zambia ranged from 2 for Zn ($p<0.05$) to 177 for Pb. Ratios of geometric mean (GM) concentrations ranged from 4.5 for Zn to 222 for Pb. Correspondingly, the proportions of values above WHO guidelines were lower in Zambia than in the DRC. Overall, the concentrations of the nine elements in water were highly correlated (Spearman correlations are reported in Table C in the Supplement). However, the correlations among elements were generally weak or non-existent for samples obtained from Zambia, and high for those obtained from DRC.

The concentrations of trace metals in surface waters, and water from boreholes or taps are depicted by location in Supplemental Fig. A. In surface waters (17 samples), the concentrations of trace elements were highest in Likasi (DRC), where water was sampled almost directly from effluents of metallurgical plants, and lowest in the Chanembo river in the Chingola District in Zambia. Concentrations of trace elements in water from boreholes (37 samples) or taps (23 samples) were consistently higher in the various locations in the DRC than in Zambia. As shown in Fig. 2**,** the concentrations of Co, Ni, Cu, As, Cd, Pb, and U in drinking water (boreholes and taps) obtained in the vicinity of mining sites were significantly and substantially higher in the DRC than in Zambia, this being most pronounced for Pb (335-fold difference for medians; 112-fold difference for GM), Cd (96-fold difference for medians; 50-fold difference for GM), and Co (51-fold difference for medians; 105-fold difference for GM).

Contamination of food crops by trace metals

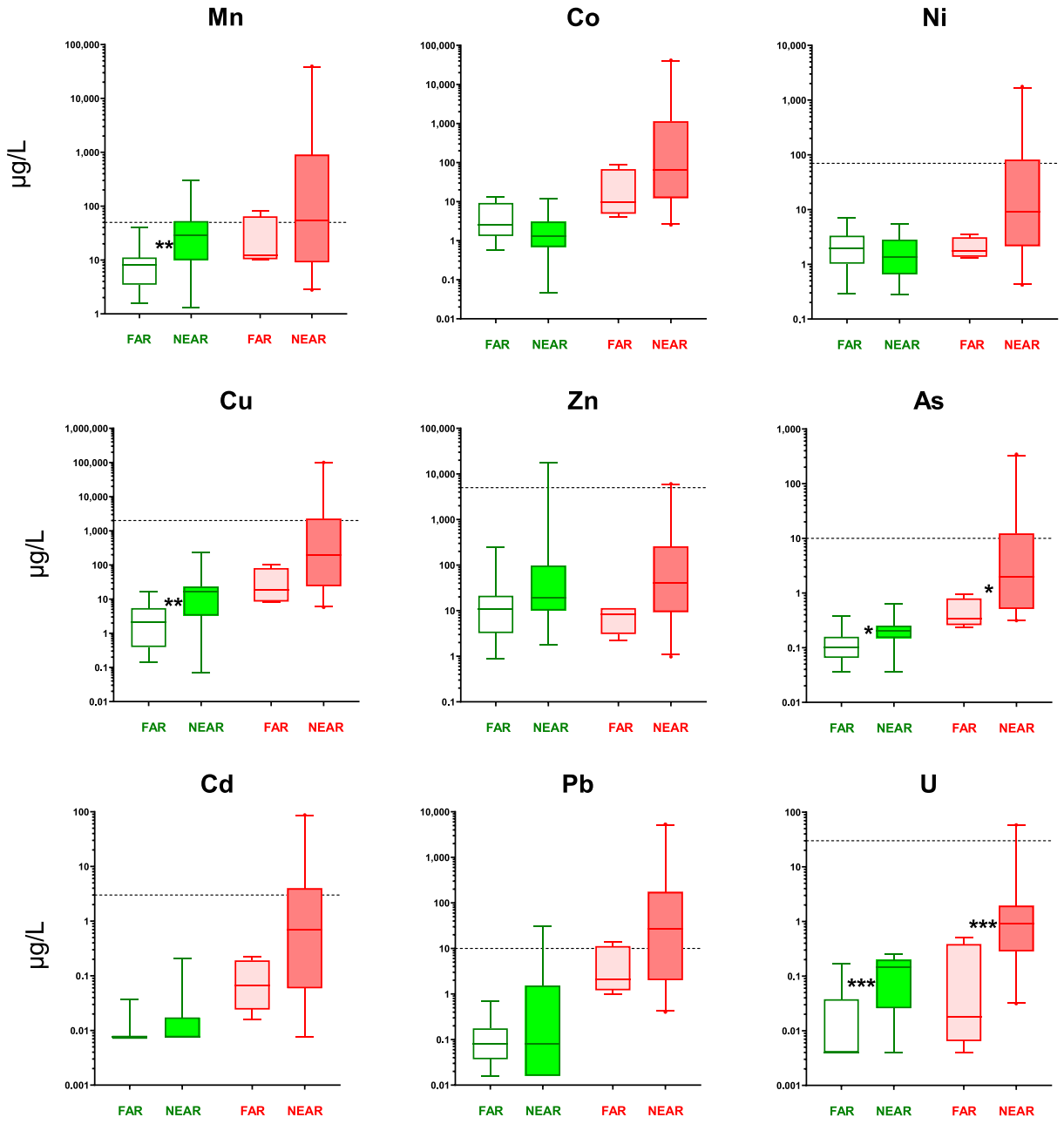
The distributions of the concentrations of nine elements in food crops taken from Zambia (n=128) and the DRC (n=145) are shown in Table 2 and Supplemental Table D. For all elements, the concentrations, again, spanned several orders of magnitude between the lowest and highest concentrations. The concentrations were significantly ($p<0.001$) higher in plant samples obtained from the DRC than in those obtained from Zambia, for all elements except Mn. Ratios of median values for the DRC versus Zambia were below 2 for Ni, Cu, Zn and U, and between 3 and 8 for Co, Cd and Pb. For As, most values were below the limits of quantification (LOQ), but the proportion of samples with quantifiable As was higher in samples from the DRC than in those from Zambia. Overall, the Spearman correlations (Table E in Supplement) between concentrations of the various elements in food crops were significant, except for Mn, the concentrations of which were correlated with none of the other metals except Zn. Correlations were generally stronger for samples from the DRC than for Zambia.

In general, the concentrations of metals were higher (per dry weight) in leafy vegetables (amaranth, pumpkin, sweetpotato and cassava leaves), tomato and beans than in maize and tuberous roots (for which only few samples were available) (Fig. B in the Supplement). The concentrations of trace elements in all food crops are shown, by location, in Supplemental Fig. C and those by village or urban neighborhood in Supplemental Fig. D.

Fig. 3 and Supplemental Table E display the concentrations of trace elements in food crops according to their proximity to mining operations, separately for Zambia and the DRC. Details for 8 separate categories of vegetables are shown in Supplemental Fig. E.

For Mn, significantly lower concentrations (by about half) were found in areas close to mining compared to control sites, both in Zambia and in the DRC. In Zambia, only two metals (Zn and Pb) had significantly higher concentrations – by approximately two-fold – in vegetables collected near mining compared to vegetables from control sites. In the DRC, statistically significant and pronounced contrasts between concentrations in crops growing near mining compared to crops obtained in sites remote from mining were found for six elements (Co, Cu, As, Cd, Pb, U), with GM ratios ranging from about 2 for As to more than 5 for Co. Even control values were significantly and substantially higher in the DRC than in Zambia for Zn (2-fold), Cd (3-fold) and Pb (4-fold), and values near mining were significantly higher in the DRC than in Zambia for all elements, except Mn and Zn, with more than 7-fold higher GMs for Cd and Pb. Excluding maize and tuberous roots did not substantially modify these comparisons (not shown).

The only elements for which WHO-FAO guidelines are available are As, Cd and Pb (FAO/WHO, 2011, FAO/WHO, 2017). For inorganic As, the FAO-WHO *Codex Alimentarius* gives a maximum limit of 0.2 mg/kg fresh weight in rice, i.e. about 1 mg/kg dry weight (assuming a 5/1 ratio for wet/dry weight); this limit was exceeded in 4/128 samples (3%) in Zambia (all close to mining) and 59/145 samples (41%) in the DRC (all but 4 samples close to mining). For Cd, the maximum limits vary according to the type of vegetable (0.05 mg/kg for brassica, bulb and



Data are shown as box plots (median, P25-P75, whiskers P5-P95). Note logarithmic scales.Water samples were obtained from boreholes or taps in three locations in Zambia (left, green) and three locations in the DRC (right, red). In each location, samples were collected NEAR mining operations (n=18 for Zambia, n=20 for DRC) or FAR from mining operations (n=18 for Zambia, n=4 for DRC). Horizontal interrupted lines indicate upper limits for drinking water according to WHO guidelines (not available for Co). Except for Zn, concentrations (NEAR+FAR) are higher in DRC than in Zambia (Mann-Whitney test p<0.001 for Co, Cu, As, Cd, Pb and U; p<0.01 for Mn and Ni); stars indicate significance (p<0.05; ** p<0.01; *** p<0.001) for FAR vs NEAR.*

Fig. 2. Concentrations (µg/L) of 9 trace elements in unfiltered drinking water according to proximity to mining operations, separately for Zambia and the DRC.

fruiting vegetables; 0.1 mg/kg for legume vegetables, pulses, root and tuber vegetables, stalk and stem vegetables, cereal grains; 0.2 mg/kg for leafy vegetables, and wheat). Taking the most liberal limit (0.2 mg/kg) and again a factor of 5 for converting dry weight to fresh weight, 4/128 samples (3%) exceeded 1 mg/kg dry weight in Zambia, whereas nearly half (68/145, 47%) of crop samples exceeded 1 mg/kg dry weight in the DRC [4/33 (12%) in control sites, 64/112 (57%) in mining areas]. All the 20 tomato samples from the DRC contained more than 0.25 mg Cd/kg dry weight (GM 2 mg/kg), against 10/17 (59%) in Zambia (GM 0.24 mg/kg); similarly, the majority (13/18, 72%) of samples of beans collected in the DRC contained more than 0.5 mg Cd/kg (GM 0.82 mg/kg) against none in Zambia (GM 0.089 mg/kg). For Pb, the maximum limits also vary according to the type of vegetables [0.20 mg/kg for cereals, legumes and pulses, 0.10 mg/kg for vegetables (excluding brassica vegetables, leaf vegetables, fresh herbs and fungi), 0.30 mg/kg for brassica vegetables, leaf vegetables and cultivated fungi, 0.10 mg/kg for fruit excluding berries and small fruit, 0.20 mg/kg for berries and small fruit]. Taking a maximum limit of 0.30 mg/kg fresh weight and a conversion factor of 5, it appears again that a large proportion of samples exceeded the limit of Pb content: 23/128 (18%) in Zambia (all but one in mining sites) and 101/145 (70%) in the DRC [9/33 (27%) in control sites and 92/112 (82%) in mining sites].

Discussion

This collaborative study was undertaken by scientists in Zambia and

Table 2
Concentration of trace elements (μg/g dry weight) in samples of food crops from Zambia and the DRC

	ZAMBIA (n=128)	DRC (n=145)		
	P50 [P25-P75] μg/g dry weight	P50 [P25-P75] μg/g dry weight	P50 DRC / P50 Zambia	P^*
Mn	**78.1** [32.9-170]	**66.1** [39.4-128]	**0.8**	0.34
Co	**1.49** [0.80-2.96]	**6.44** [1.63-16.4]	**4.3**	<0.001
Ni	**7.06** [2.50-12.0]	**10.4** [4.90-17.9]	**1.5**	<0.001
Cu	**33.4** [19.9-59.0]	**62.1** [24.8-166]	**1.9**	<0.001
Zn	**72.8** [40.5-137]	**140** [59.5-324]	**1.9**	<0.001
As	***0.800*** [*0.800-0.800*]	***0.800*** [*0.800*-2.60]	**1.0**	<0.001
Cd	**0.156** [0.056-0.373]	**0.867** [0.359-2.25]	**5.4**	<0.001
Pb	**0.464** [*0.150*-0.997]	**3.87** [1.11-10.4]	**8.3**	<0.001
U	**1.96** [0.931-3.69]	**3.09** [1.47-5.79]	**1.6**	<0.001

P50, P25 and P75 are 50th (median), 25th and 75th percentiles, respectively;
* p value for difference between Zambia and DRC (Mann-Whitney test); values in italics are imputed values (half the value of Limit of Quantification). See Supplemental Table D for more details.

the DRC based on each side of the border that separates the central African Copperbelt into its Zambian and Congolese parts, in order to address common concerns about contaminations of water and locally consumed food crops resulting from past and current mining and metallurgic activities. To our knowledge, such cross-border collaborative research using similar procedures and analyses to assess environmental pollution is novel and unique in sub-Saharan Africa.

The study evaluated the relations between proximity of mining and the levels of nine relevant trace elements – manganese (Mn), cobalt (Co), nickel (Ni), copper (Cu), zinc (Zn), arsenic (As), cadmium (Cd), lead (Pb) and uranium (U) – in surface water and drinking water, as well as in widely consumed, locally cultivated food crops. A strength of the study is that it has extended the number of copper-cobalt mining locations investigated – each containing sites close to the presumed source of pollution as well as a more distant control site – and the range of trace elements analyzed beyond what has been done hitherto in either country. In both countries, measured levels of trace elements were consistently higher in samples obtained close to the pollution sources as compared to their control areas, but the degree of contamination was markedly higher in samples from the DRC than in those from Zambia.

The current study corroborates and complements the findings of several prior or later campaigns that assessed environmental pollution by mining activities in either Zambia or the DRC. In Zambia, various surveys performed in the past two decades have shown "heavy" metal contamination of rivers and lakes, groundwater, soil and dusts, trees and wild plants, fish, wildlife and farm animals (Křibek et al., 2010; Ettler et al., 2011; Sracek et al., 2012; Mwitwa et al., 2012; Nakayama et al., 2013; M'kandawire et al., 2017). Many of these studies have concentrated on the severe metal pollution in Kabwe – known as one of the "10 most polluted sites" worldwide (Blacksmith Institute, 2007) – which is situated in Zambia's Central Province, i.e. not in the area investigated in the current study. Various studies have provided data on contamination of edible food crops by trace metals in Zambia (Kapungwe, 2013; Křibek et al., 2014; Kourouma et al., 2019). In the former Katanga Province of the DRC, various publications have addressed mining-related contamination of locally grown food crops (Katemo Manda et al., 2010; Mpundu Mubemba Mulambi et al., 2013; Cheyns et al., 2014; Kashimbo Kalala et al., 2015).

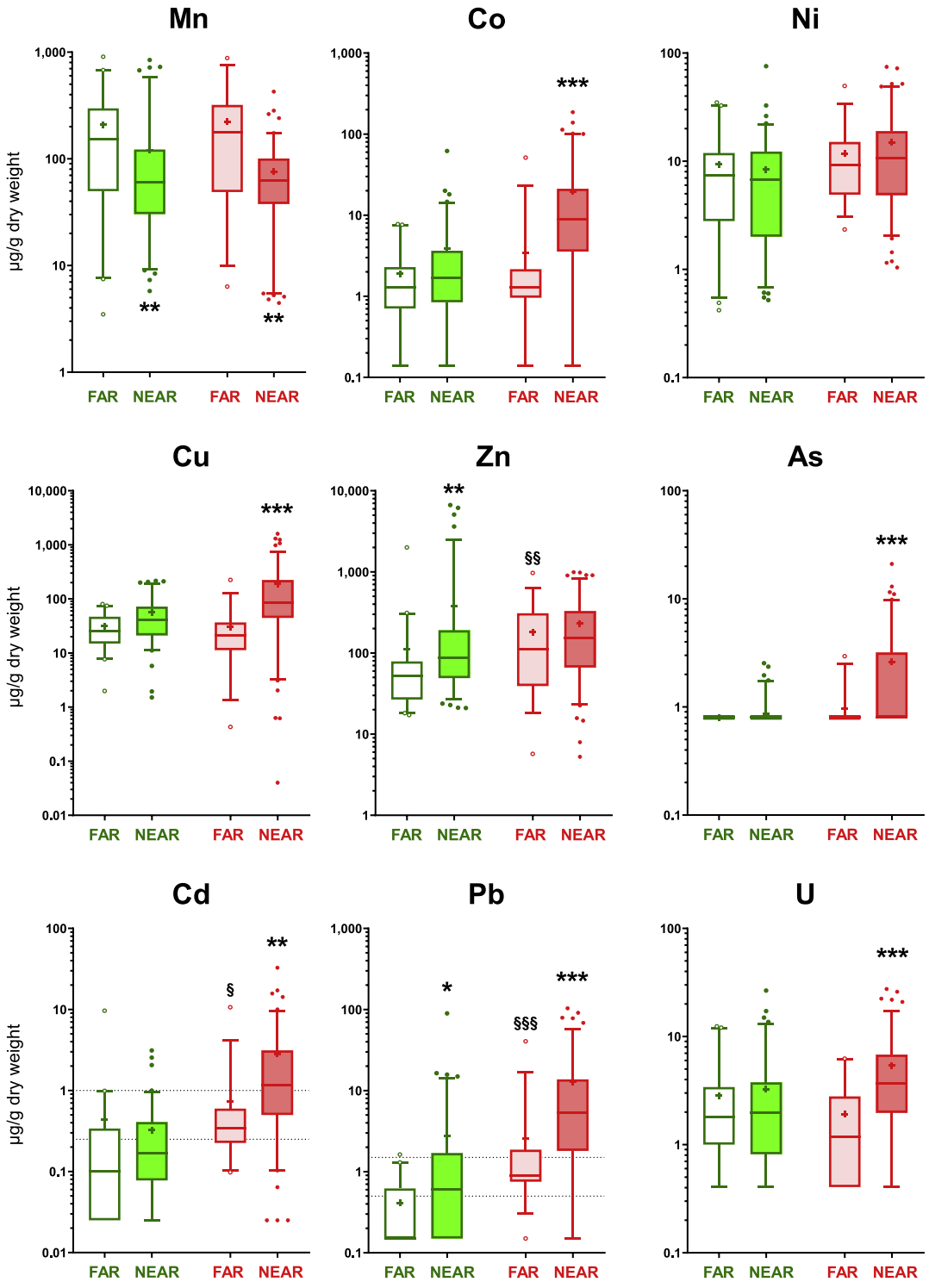
Regarding pollution of river water, the situation appeared to be much worse in the DRC than in Zambia. This is due in part to the choice of sampling sites: in Zambia, river water samples were obtained in two rivers away from mining areas (Chanembo and Mushishima), whereas in the DRC water was obtained close to where effluents from the Likasi hydro-metallurgic industry were discharged into the river, thus explaining the extremely high concentrations found in this location (well above 1,000 μg/L for Mn, Co, Ni, Cu, Zn and Pb; around or above 100 μg/L for As, Cd, and U). This highly polluted water flows further into the Lufira and then contaminates Lake Changalele, where high concentrations of cobalt (Cheyns et al., 2014) and toxic metals have been previously measured in fish (Katemo Manda et al., 2010; Squadrone et al., 2016). However, although the data regarding surface water may not be totally representative for the two countries, the findings regarding *drinking* water from communal taps and boreholes indicate consistently higher metal(loid) contamination in the DRC than in Zambia. In Zambia, nearly all samples were well below WHO limits for drinking water (WHO, 2017), whereas a high proportion of samples from the DRC considerably exceeded these limits, including for the most toxic elements, i.e. As, Cd, and Pb. The situation is particularly worrisome for Pb in tap water in the DRC, where the majority of samples exhibited concentrations above the WHO guideline value of 10 μg/L, with some values well above 100 μg/L. One could criticize that water samples were not filtered before analysis and, hence, the measured metal concentrations could be due in part to the presence of metals in colloids and particles but, on the other hand, this unfiltered water is what people actually consume.

The present study focused on the possible contamination of locally grown food crops. The most frequently obtained crops consisted of leafy vegetables – such as leaves of amaranth, cassava, pumpkins and sweetpotato, all of which are widely consumed as cooked legumes – followed by tomato and common beans. Samples of maize, sweetpotato and cassava tubers contained lower concentrations of most trace metals per dry weight than the leafy vegetables, tomato and common beans. No substantial differences in metal content were seen among the leafy vegetables, a detailed evaluation of this being beyond the scope of the current study. Although the vegetables were rinsed and washed to remove dust and soil particles, it cannot be guaranteed that this procedure completely removed adherent small particles containing metals. However, in practice, vegetables are unlikely to be washed more thoroughly before cooking.

As in the case of water, the concentrations of trace metals in food crops were consistently higher in samples obtained from the DRC than in those from Zambia, except for Mn (we have no explanation for the two-fold *lower* levels of Mn in crops from mining areas than in those from presumably non-polluted areas). The mean levels of all other elements, except Zn, measured in crops from mining areas were much higher in the DRC mining sites than in the Zambian mining areas, this being especially so for Co, Cd and Pb. For the latter two metals, even the "background" concentrations in crops growing at a distance from mining were 3 to 4 times higher in the DRC than in Zambia. In Zambia, levels of trace elements in crops did not differ much between mining and non-mining sites, except for Zn and Pb for which levels were about two-fold higher in mining sites compared to control sites. In contrast, in the DRC, levels of most trace elements were considerably higher in crops obtained close to mining than in control samples (again Mn was lower in mining sites), this being especially pronounced for Co, Cu, Cd, Pb and U. Collectively, these findings indicate that crops cultivated close to mining were more likely to be heavily contaminated by trace elements in the DRC than in Zambia.

In the absence of limit values in edible crops for most of the elements studied here, it is difficult to assess the health risks for the populations concerned. WHO-FAO guidelines are available for As, Cd and Pb only (FAO/WHO, 2011; FAO/WHO, 2017). As indicated in the results, these guidelines were exceeded in crop samples taken in both countries, but much more so in the DRC, where limit values were exceeded in 41%, 57% and 70% of samples for As, Cd and Pb, respectively. Moreover, if one considers the high correlation between metals (Spearman rho 0.74 for Pb and Cd) this means that many samples (3/128 in Zambia, 63/145 in the DRC) exceeded maximal values for both metals (1 mg Cd/kg and 1.5 mg Pb/kg), and also contained high amounts of all the other elements (except Mn).

An estimation of the possible health effects of consuming the contaminated water and home-grown vegetables is not possible without



Data are shown as box plots (median, P25-P75, whiskers P5-P95). Note logarithmic scales. Samples of various types of crops were obtained in three locations in Zambia (left, green) and three locations in the DRC (right, red). In each location, samples were collected NEAR mining operations (n=88 for Zambia, n=112 for DRC) or FAR from mining operations (n=40 for Zambia, n=33 for DRC). (Horizontal dotted lines for Cd and Pb indicate maximum levels according to WHO-FAO (level depends on type of crop). Stars indicate significance (p<0.05; ** p<0.01; *** p<0.001) for NEAR vs FAR from mining; section signs (§) indicate significance (§ p<0.05; §§ p<0.01; §§§ p<0.001) for FAR from mining in DRC vs FAR from mining in Zambia (Kruskal-Wallis tests followed by Dunn's post-hoc test).*

Fig. 3. Concentrations (µg/g dry weight) of 9 trace elements in food crops according to their proximity to mining operations, separately for Zambia and the DRC.

knowledge of the amounts consumed daily by the concerned population. Moreover, such estimation must also consider the provenance, degree of contamination and amount consumed of other food items, such as other staple food and meat or fish (Cheyns et al., 2014). Another important aspect to take into consideration is the ingestion of possibly contaminated dust blown by the wind from industrial operations, tailings, unpaved roads or arid surfaces, particularly in the dry season. This is particularly relevant for young children, who spend most of their time playing close to the ground and who tend to put dirty fingers and objects in their mouths. A recent study, done in Lubumbashi, has shown that the amount of ingested dust is considerably higher than the default values that are commonly assumed (on the basis of studies done in industrially developed countries) to assess health risks of soil and dust contamination (Smolders et al., 2019).

Although human health risks can be estimated on the basis of measurements of trace metal concentrations in water, food or dust, it would be more rewarding to perform targeted biomonitoring studies to assess the *internal* exposure of exposed adults and children. This is because the uptake of metals present in water, food or dust in the human body is determined by their bioavailability and degree of gastrointestinal absorption, which depend not only on the type of the contaminants themselves but also on the overall composition of the food (e.g., calcium and other divalent cations), as well as on individual factors, such as age or health condition (e.g., iron status) (Ali et al., 2019; Riediker et al., 2019). Several biomonitoring studies have been conducted in various mining areas in the DRC, and they have documented high concentrations of multiple trace elements in urine or blood of residents or workers (Banza et al., 2009; Musa Obadia et al., 2018; Nemery and Banza Lubaba Nkulu, 2018). Except for lead in blood in children living in Kabwe (Yabe et al., 2015; Human Rights Watch, 2019), no such studies of human body burdens of trace metals have, to our knowledge, been published in the studied mining areas in Zambia.

Based on the current findings and the known human toxicity of trace metals, the excessive concentrations of lead and cadmium found in drinking water and vegetables are arguably the most serious causes of toxicological concern for the people living close to mines, especially in the DRC. Lead, cadmium and arsenic are well-known for causing adverse effects on several biological systems in humans, especially neurodevelopment, kidney damage and cancer (Skerfving and Bergdahl, 2015; Nordberg et al., 2015; Fowler et al., 2015). Although uranium is often associated with the risk of radioactivity, this is mainly a problem due to the emission of radon, one of its gaseous decay products, in enclosed spaces (such as in poorly ventilated mines or homes) (Keith et al, 2015; WHO, 2010). Nevertheless, uranium is also nephrotoxic and possibly affects also other organs (Ma et al., 2020). Cobalt is an essential element but exposure to high amounts of cobalt is associated with possible toxic effects on the lungs, heart, thyroid and blood (Lison, 2015; Banza et al., 2009). Moreover, the *combined* toxicity of the multiple metals to which people are chronically exposed from an early age in the affected areas is probably of concern for human health. The long-term impact of such high exposure to a "cocktail" of metals on cancer, reproduction and fertility, neurodevelopment, hypertension and cardiovascular disease, endocrine and metabolic disease, and other conditions remains to be evaluated by epidemiological studies. In a case-control study in Lubumbashi, it was found that babies with birth defects were more likely to have been fathered by men with a mining-related job (Van Brusselen et al., 2020). Other studies have shown associations of pre-eclampsia with blood lead levels in women in Lubumbashi (Musa Obadia et al., 2018) and of sperm alterations with exposure to arsenic and cadmium in men living in mining areas (Mukendi et al., 2018).

Although the current collaborative study has several strengths, its limitations must be also acknowledged. The choice of locations was based on objective considerations and the general sampling strategy was uniform across locations, but in the specific sites the sampling depended on various logistic aspects, such as accessibility, availability of crops, willingness of residents, which were not identical across locations. This led to variable numbers and a somewhat unbalanced composition of the water and crop samples. Nevertheless, in view of the general and spatial consistency of our findings, this relative lack of comparability is unlikely to have caused a major bias, let alone to invalidate the main conclusions of the study. Since it has been demonstrated that contamination from point sources may be spread over long distances (Kribek et al., 2010; Ettler et al., 2011), we recognize that the control sites may not have been entirely free from contamination by mining activities. However, the purpose of our study was mainly to document and quantify levels of contamination close to mining activities in comparison with sites without nearby pollution sources. If anything, possible contamination of the control sites would have resulted in some underestimation of the degree of contamination of the sites close to pollution sources. Another relative limitation is that apart from the cultivated crops, no other food items like fish, meat, eggs and several other commercially available foodstuffs were obtained, thus precluding a full assessment of the uptake of trace metals via food by the population. Hence, the current study should be complemented by well-designed biomonitoring research to evaluate the body burden of trace metals in the populations concerned. Although the sampling campaigns were done many years before the current publication, there is no reason to believe that these findings are no longer relevant.

The objective results demonstrating water pollution and contamination of crops are in line with the subjective evaluations gathered from local residents during samples collection. They consistently indicated that both surface and groundwater were not suitable for domestic consumption, a problem they blamed on waste release by mining and metallurgic industrial effluents. During our campaign, vegetation on the fringe of mines was often seen dying through siltation, and surface and well water was often of poor quality, being silted and turbid with a brownish color presumably originating from mine waste, thus making it not potable and unsuitable for human use. Yet, this is the water that is daily used in various communities. In a previous study on mining, forest management and livelihoods in both Zambia and the DRC (Mwitwa et al, 2012), residents were administered a brief questionnaire on their knowledge of safety from mining extraction and metallurgic operations, and on their perceptions of positive and negative impacts on their livelihood. Many residents living close to mining often expressed suffering and anxiety because of industrial exploitation. They pointed to poor environmental conditions, especially caused by emissions of irritant gases, including sulfur dioxide, and dust generated from opencast activities, waste rock stockpiles, tailings disposal facilities, crusher and mineral processing plants where ore is handled, processed or stockpiled, and from road transportation of the ore. Further, they attributed many acute symptoms and chronic diseases to their poor environment and considered that children and adults suffered from numerous sicknesses, including cancer, respiratory disorders (e.g. tuberculosis, coughing, asthma, pneumonia), eye irritation, sore throat and several others like diarrhea, skin tingling and severe fatigue.

Our study revealed that, overall, the degree of environmental pollution by trace metals was higher in the DRC than in Zambia. The exact causes for this situation are probably multiple. Although the locations studied in the two countries all lie within a few hundred kilometers in the same Copperbelt region, geological differences, such as the depths and composition of the extracted copper and cobalt ores, may explain differences in the industrial processing of the ores and the release of toxic contaminants, such as As, Cd, Pb or U, in the environment. However, the apparent differences in mining-related pollution between the DRC and Zambia must probably be attributed to a large extent to differences in environmental governance between the two countries. Such differences possibly already started during colonial times when the DRC and Zambia were under Belgian and British rule, as the Belgian Congo and Northern Rhodesia, respectively. In neither of the two countries did industry or colonial authorities give much priority to environmental issues (Peša, 2020b). After independence, both countries nationalized their mining

industry, but environmental issues still did not feature prominently until the 1990s. However, in Zambia, the environmental protection and pollution act (ZEMA, 2011) and the environmental management policies and institutions (ZEMA, 2017) became quite functional in establishing Zambia's Environmental Council with a mandate to protect the environment, control pollution, and provide for the welfare of humans through the health of the natural environment, and they have not been compromised by personal interests in the mining sector. Moreover, the Zambia Environmental Management Agency (ZEMA) is adequately funded and supported by the Government of the Republic of Zambia to minimize environmental impacts arising from mining operations. Zambia has also benefited from the World Bank's support for environmental safeguards applicable to all projects, mainly in enforcing legislation on the environment, in particular by the extractive industries (World Bank, 2015). This has helped to mitigate the environmental risks related to pollution of soil and groundwater in the areas where copper is produced, and to rehabilitate and expand water and sanitation facilities in five municipalities of the Copperbelt. In contrast, in the DRC, despite numerous revisions of the Mining Code until the current 'Code Minier' amended and completed by decree n° 18/024 of 8th June 2018 (Code Minier, 2018), mining companies have hardly complied with the environmental regulatory requirements and the government has not systematically monitored the industries' implementation of the environment assessment protocols (ASADHO/Katanga, 2007; Global Witness, 2006).

The contrast shown here between the two countries' degree of mining-related environmental pollution reflects their respective positions according to the World Bank's governance indicators: for the past 20 years, Zambia has consistently ranked in the 25-50th percentile range whereas the DRC has ranked in the 0-10th percentile range for the six aggregated indicators (World Bank, 2020).

Conclusion

The findings of this unique collaborative study convincingly document "footprints" of extractive industries in both countries of the African Copperbelt, with salient negative impacts on the quality of surface and underground water, and on locally cultivated food crops, especially in the DRC. The latter is most likely due mainly to deficiencies in environmental governance.

In light of the nature and extent of the contamination, concerted efforts from the private and public sectors, international organizations and both bilateral and multilateral donor communities are needed to reinforce and implement guidelines regulating mining and metallurgic operations for protecting the environment, ensure food and nutrition safety, and preserve population health in the central African Copperbelt.

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Authors Contributions

A. Muimba-Kankolongo: Research proposal and methodology conceptualization, team building, funding acquisition, research administration, field investigation, sample collection and preparation and original draft writing. C. Banza Lubaba Nkulu: Study methodology, field investigation, sample collection and preparation, laboratory sample analysis, data compilation, draft review and editing. J. Mwitwa: Field investigation, sample collection, draft review and editing. F.M. Kampemba: Field investigation, sample collection and preparation. M. Mulele Nabuyanda: Field investigation, sample collection and preparation, draft review and editing. V. Haufroid: Laboratory sample analysis, methodology and draft review and editing; E. Smolders: Laboratory sample analysis, methodology and draft review and editing. B. Nemery: Facilitation of laboratory sample analysis, data compilation and statistical analysis, making map and graphs, reviewing and editing successive drafts.

Declaration of Competing Interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.envadv.2021.100103.

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