

Toxic metals in cereals and derivatives consumed in Cape Verde: a risk assessment study

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Abstract

Cereals and their derivatives are the basis of human nutrition. However, cereals also contribute to the dietary exposure to toxic metals that may pose a risk. Strengthening food security and nutrition information is a high priority challenge for the Cape Verde Government. The toxic metals content (Cr, Ni, Sr, Al, Cd, Pb) has been determined in 126 samples of cereals and derivatives (rice, corn gofio, corn flour, wheat flour, corn, wheat) consumed in Cape Verde. Wheat flour samples stand out for registering the highest Sr (1.60 mg/kg), Ni (0.25 mg/kg) and Cr (0.13 mg/kg). The results show relevant Al levels (1.17 – 13.4 mg/kg) with its highest levels in corn gofio. The mean Pb average content in the cereals is 0.03 – 0.08 mg/kg with the highest level observed in corn gofio. The Al and Pb levels are lower in cereals without husks. A consumption of 100 g/day of corn gofio provide an intake of 1.34 mg Al/day (13.7% of the tolerable weekly intake established at 1 mg/kg bw/week) and 8 µg Pb/day (20% of the BMDL set at 0.63 µg/kg bw/day for nephrotoxic effects). The minimization of the dietary exposure of

the Cape Verdean population to toxic metals is through the importation of higher quality cereals.

Keywords

Cape Verde, cereals, metals, dietary intake, risk evaluation

Introduction

The archipelago of Cabo Verde is located on the West African coast, 500 km from Senegal and is made up of 10 islands, which 9 are inhabited and one is uninhabited. The Cape Verdean diet is characterized by an important consumption of cereals and cereal-based products. Preliminary data from the National Survey on Food and Nutritional Vulnerability of Families indicate that cereals represent 95.3% of the food consumed. According to the preliminary results of the IDRF 2015 the ingestion of cereals occupy the first line of annual per capita consumption expenditure (about 11,611\$), compared to other food products consumed. However, the internal cereal production satisfies only 6.9% of the population's consumption needs, contributing to the highly vulnerable of the country from the standpoint of food security. Food security of Cabo Verde is also affected by agroclimatic variation and external market fluctuation. The 2019 national cereal production was estimated at about 1,000 tons, almost 70 % below the mean average of the previous five years [1]. Therefore, about 85% of the domestic cereal demand (mostly rice and wheat for human consumption) is covered by imports. The cereal import requirements in the 2019/20 marketing year (November/October) were forecast at an above-average level of 87,000 tons [1]. From 2016 to 2020, the cereals import reached a total of 419,749.30 tons, with emphasis on corn (159,979.30 tons), rice (144,799.33 tons), wheat grain (91,623.39 tons). The supply of the market in cereals is done both in form of food aid through cooperative relations with development partners and commercial import [2]. The current corn production does not meet the internal demand, and the cereal must be

imported for food and fodder [3]. Moreover, the main drivers of the food insecurity in Cabo Verde are the effects of dry weather events (drought) and pest attacks on cereal and fodder production [1]. As mentioned above, the food insecurity in Cabo Verde has a structural and multi-factorial nature and concerns the structural deficit of the national food production and the strong dependence on the international market, and the economic accessibility weaknesses. Strengthening the food security and nutrition information system (FSNIS) is an important challenge for Cabo Verde Government [4].

According to FAO, in 2017, about 13% of the population were under-nourished. The data available indicate that 20% of rural families lived in a situation of food insecurity with 13% in a moderate position and 7% in a severe position [2]. Although Cape Verde is in a nutritional transition period characterized by the consumption of high fat, refined carbohydrates, cholesterol, sugar and low consumption of fruit and vegetables causing a rapid and significant increase in the prevalence of being overweight and obese [5], the consumption of cereals and cereal-based products is still relevant, and representing 47% of the total food energy intake. In Cabo Verde, the cereal balance for 2002/03 estimated a cereal consumption of 242 kg/year per person comprising 123 kg of corn (337 g/day), 67 kg of rice (184 g/day) and 52 kg of wheat (142 g/day).

Although the nutritional contribution of cereals is noteworthy, they may contain elements that are harmful to health [6, 7], as is the case of elements such as Al, Cd, Cr, Ni, Pb or Sr. All of these elements have maximum daily or weekly intake values set by reference bodies in food safety such as the EFSA (European Food Safety Authority) or the World Health Organization (WHO) (Table 1).

Table 1. Reference intakes of the analyzed elements

Element	Parameter	Guideline value	References
Cr(III)		0.3 mg/kg bw/day	[8]
Ni	TDI	13 µg/kg bw/day	[9]
Sr		0.13 mg/kg bw/day	[10]
Al	TWI	1 mg/kg bw/week	[11]
Cd		2.5 µg/kg bw/week	[12]
Pb	BMDL	0.63 ¹ µg/kg bw/day	[13]
		1.50 ² µg/kg bw/day	

TDI, tolerable daily intake; TWI, tolerable weekly intake; BMDL, benchmark dose level; bw, body weight; Nephrotoxicity¹ and cardiovascular effects²

Al is a neurotoxic metal with no function in the human body [14]. Prolonged exposures to Al are related to neurodegenerative diseases such as Alzheimer's, and the estimation of its dietary exposure has been the subject of previous studies [15-17]. The "Safety of aluminum from dietary intake" prepared by EFSA states that the estimated dietary intake of Al in the European population is between 0.2 - 1.5 mg/kg of body weight per week in an adult weighing 60 kg. Similarly, it concludes that cereals and cereal derivatives are one of the main foods that contribute the most to the dietary intake of Al in the general population [18].

Cd, a toxic element with a high half-life and a tendency to bioaccumulate [19], is found in cultivation soils, which favors its accumulation in cereals [20]. Cd competes in the body with other essential divalent cations, and affects the renal system, causing irreversible damage to the renal tubules [21, 22]. Rubio et al. [23] determined the dietary exposure to Cd in a population of the Canary Islands (Spain), the intake of Cd from cereals at 1.065 µg/day, and cereals are one of the groups that most contributes to the dietary intake of Cd. In addition, the EFSA scientific report entitled "Cadmium dietary exposure in the European population" establishes that grains and grain-based products constitute one of the foods that most contribute to the dietary intake of Cd in the European population [24].

Cr is mainly found in the trivalent ion form in food. Although oral Cr (III) is not particularly toxic [25], high Cr intakes can trigger chronic kidney failure, dermatitis, bronchitis or asthma [26, 27]. A study by Filippini et al. [28] concluded that beverages, cereals and meat provided the highest dietary contributions of Cr in a northern Italian population.

Ni is essential for plants [29] and grains and grain-based products have been identified as the most important contributors to the chronic dietary Ni exposure in Europe even though Ni is the only regulated element in drinking water and is often studied [9]. Individuals with hypersensitivity to Ni or with kidney disease are susceptible to damage from a high dietary intake of Ni [26].

Sr is an element that is found in food although, to date, no cases of food poisoning by strontium have been reported. However, Sr competes with essential elements such as phosphorus [30] and recent studies in experimental animals reported associated hepatotoxic effects with Sr [31].

Pb is a neurotoxic metal that accumulates in the body causing serious damage to the central nervous system (CNS) as well as kidney disease, gastrointestinal tract disorders and Alzheimer's [13]. Pb can be found in traces in large quantities of food and in drinking water [32, 33], especially in fruits, vegetables and cereals due to the deposit of Pb particles from the atmosphere. In the scientific report of EFSA entitled "Lead dietary exposure in the European population", the food category that contributes mostly to Pb exposure was bread and rolls (8.5%), tea (6.2%) and tap water (6.1%), among others [34].

Food risk surveillance and food safety strategies encourage monitoring the content of metals in each of the food groups consumed by the different populations. The aim of the present study is the determination of Al, Cd, Cr, Ni, Pb or Sr in commonly consumed cereals and cereal-based products in the Cape Verde Islands and their subsequent risk assessment.

Material and Methods

Samples

A total of 126 samples of cereals (rice, corn and wheat) and cereal-based products (corn flour, wheat flour and corn gofio) (Table 2) marketed and consumed in Cape Verde were acquired in two different islands of the Cape Verde archipelago, specifically on the islands of Santiago and São Vicente (Fig. 1). The population of the island of Santiago is approximately 260,000 inhabitants while that of the island of São Vicente is 76,000. Gofio is a traditional artisan food derived from cereals, mainly corn, that is made by first roasting the cereal with its husks and then grinding it until a powder similar to flour is obtained [35-37].



Figure 1. Map of the Cape Verde Islands showing the sampling areas (São Vicente and Santiago)

Table 2. Analyzed cereal and derived product samples

Type	No. Samples	Origin
Rice	56	Santiago
	5	São Vicente
Corn gofio	6	Santiago
	1	São Vicente
Corn flour	10	Santiago
	1	São Vicente

Wheat flour	17	Santiago
	2	São Vicente
Corn	13	Santiago
	2	São Vicente
Wheat	2	Santiago
	11	São Vicente

The sampling were carried out in the period from 2017 to 2019, at the level of importing and retailing establishments of cereal, in Santiago and São Vicente islands.

Sample treatment

One gram of each sample was weighed into Teflon tubes, previously washed with laboratory detergent and Milli-Q quality distilled water. Four mL of 65% nitric acid (Sigma Aldrich, Germany) and 2 mL of hydrogen peroxide (Sigma Aldrich, Germany) were added to the samples. The Teflon tubes were closed and placed in a microwave oven (Multiwave Go, Anton Paar, Austria) for subsequent digestion according to the conditions described in Table 3. After the samples had been digested, they were transferred to 10 mL volumetric flasks and made up with Milli-Q quality distilled water. Finally, they were transferred to airtight jars with a lid for later measurement.

Table 3. Instrumental conditions of the microwave digestion process

No.	Ramp (min)	Temperature (°C)	Time (min)
1	15'00''	50	5'00''
2	5'00''	60	4'00''
3	5'00''	70	3'00''
4	3'00''	90	2'00''
5	20'00''	180	10'00''

Limit temperature: 200°C

Cooling temperature: 50°C

Analytical method

The determination of the metal content was conducted by Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-OES) model ICAP 6300 Duo Thermo Scientific (Waltham, MA, United States) with an Auto Sampler automatic sampler (CETAX model ASX-520).

The instrumental conditions of the method were the following: RF power of 1150 W; gas flow (nebulizer gas flow, make up gas flow) of 0.5 L/min; injection of the sample to the 50 rpm flow pump; stabilization time of 0 s [38, 39]. Instrumental wavelengths (nm) of the analyzed elements were: Al (167.0), Cd (226.5), Cr (267.7), Ni (231.6), Pb (220.3), and Sr (407.7).

The quantification limits of the toxic metals, calculated as 10 times the standard deviation (SD) resulting from the analysis of 15 targets under reproducibility conditions [40], were: 0.012 mg/L (Al), 0.001 mg/L (Cd), 0.008 mg/L (Co), 0.003 mg/L (Ni), 0.001 mg/L (Pb), 0.003 mg/L (Sr).

The quality control of the method (Table 4) was based on the recovery percentage obtained with reference material (SRM 1515 Apple Leaves, SRM 1548a Typical Diet, SRM 1567a Wheat Flour) under reproducible conditions. The recovery percentages obtained with the reference material were above 94% in all cases. The statistical analysis did not detect significant differences ($p < 0.05$) between the certified concentrations and the concentrations obtained.

Table 4. Recovery study results and reference materials used

Metal	Material	Concentration found (mg/kg)	Certified concentration (mg/kg)	R (%)
Al	SRM 1515 Apple Leaves	286±9	285.1±26	99.7
Sr		25.0±2.0	24.6±4.0	98.3
Cr		0.29±0.03	0.30±0.00	97.8
Ni	SRM 1548a	0.37±0.02	0.38±0.04	102.3
Pb	Typical Diet	0.044±0.000	0.044±0.013	98.9
Cd	SRM 1567a Wheat Flour	0.026±0.002	0.026±0.008	98.4

The IBM Statistics SPSS 24.0 computer software for Windows TM was used for statistical analysis. The data was checked for normality using the Kolmogorov-Smirnov and Shapiro-Wilk test, and the Levene Homogeneity of Variances test. Since the data followed a non-normal distribution, non-parametric tests were applied, in this case, the Kruskal-Wallis test [41]. The statistical analysis was performed to verify the existence of significant differences ($p < 0.05$) between raw materials (cereals), corn and cereal-based products (gofio, flour and roasted corn) and flours.

Calculation of dietary intake

The assessment of dietary exposure was based on the calculation of the estimated daily intake (EDI) and the subsequent obtained percentage contribution to the reference value of each of the metals under study.

$$\text{EDI (mg/day)} = \text{Mean consumption (kg/day)} \times \text{Element concentration (mg/kg fresh weight)}$$

$$\text{Contribution (\%)} = [\text{EDI} / \text{Guideline value}] \times 100$$

$$\text{EDI (mg/day)} = \text{Average consumption (kg/day)} \times \text{Element concentration (mg/kg fresh weight)}$$

$$\text{Contribution (\%)} = [\text{EDI} / \text{Guideline value}] \times 100$$

Results and Discussion

Table 5 shows the mean concentrations (mg/kg fresh weight) and standard deviations (SD) of the metals analyzed for each of the types of sample under study.

Table 5. Mean concentrations and standard deviations (mg/kg of fresh weight \pm SD) of the analyzed metals by cereal type and cereal derived product type

Element	Cereals and cereal-based products					
	Rice	Corn	Corn flour	Wheat flour	Corn gofio	Wheat
Al	1.43 \pm 3.96	2.41 \pm 1.87	1.17 \pm 1.08	2.74 \pm 1.38	13.4 \pm 12.7	4.85 \pm 3.75

Cd	0.01±0.02	0.007±0.008	0.005±0.01	0.02±0.01	0.003±0.005	0.01±0.01
Cr	0.02±0.06	0.08±0.16	0.01±0.03	0.02±0.04	0.09±0.07	0.13±0.08
Ni	0.19±0.09	0.15±0.08	0.08±0.05	0.12±0.07	0.22±0.09	0.25±0.08
Pb	0.03±0.02	0.03±0.02	0.03±0.02	0.03±0.01	0.08±0.05	0.07±0.06
Sr	0.21±0.35	0.40±0.59	0.14±0.19	0.77±0.67	0.77±0.65	1.60±1.16

Al was found in the highest concentrations in all the analyzed cereal samples, most clearly in corn gofio where it reached a mean average concentration of 13.4 ± 12.7 mg/kg fresh weight. This concentration differs significantly from the rest of the cereals ($p < 0.05$). Liu et al. [42] concluded that cereal husks contain higher concentrations of metals than the grain. Based on this, the differences in the Al content recorded here in corn gofio may be due to the use of the whole cereal, including the husk, in the manufacture of this product derived from corn [35], which may explain a higher Al content. However, despite the toxicological considerations of this neurotoxic element, current European legislation does not include maximum levels of Al in food.

The wheat flour samples are worth mentioning for presenting the highest levels of Sr (1.60 mg/kg fresh weight), Ni (0.25 mg/kg fresh weight) and Cr (0.13 mg/kg fresh weight). The *Second French Total Diet* had a mean level of Sr in breakfast cereals of 0.842 mg/kg fresh weight [43], this value was lower than the level obtained in the wheat samples of the present study. In addition, Cubadda et al. [44] reported lower Ni levels in flour and wheat (0.035 mg/kg) than those observed in this study. However, Mathebula et al. [45] observed a mean Cr level in wheat of 2.629 mg/kg fresh weight, higher than the mean level recorded in this study.

As observed for Sr, Ni and Cr, the wheat flour samples also presented the highest mean concentration of Cd (0.02 ± 0.01 mg/kg fresh weight). Tejera et al. [46] recorded mean Cd

concentrations of 0.027 mg/kg fresh weight in wheat flour, values similar to those recorded in the present study. Regarding wheat grain, Škrbić et al. [47] observed, however, Cd levels in wheat from Serbia of between 2.4 - 252 µg/kg fresh weight, higher than those registered in the wheat analyzed here (0.01 ± 0.01 mg/kg fresh weight).

As for Pb, the highest mean level was observed in the corn gofio samples, with a mean concentration of 0.08 ± 0.05 mg/kg fresh weight. Furthermore, this concentration may indicate that Pb tends to accumulate in the husk of cereals, since, in the cereal-based products manufactured without the husk, the Pb levels were lower. A study conducted by Bilo et al. [48] in rice and rice husks, concluded that rice husks accumulated higher concentrations of toxic metals than rice. This suggests that gofio, being a derivative produced from whole-grain cereal, including the husk, may have higher Pb levels than flours produced from dehusked cereal.

The statistical analysis showed significant differences ($p < 0.05$) in the Pb content between wheat and the rest of the samples, in the Al content between the rice and wheat samples and in the Sr and Ni content of the rice and corn samples when compared to the wheat samples.

Table 6 shows the concentrations (mg/kg fresh weight), standard deviations (SD), maximums and minimums of the toxic metals analyzed by geographical areas of acquisition and consumption. The samples from São Vicente had the highest mean concentrations of Al, Cd, Cr, Ni, Sr and Pb. Considering that these differences may be due to multiple factors such as intrinsic characteristics of the plant, characteristics of the cultivation soil, as well as climatic factors [47, 49] it is suggested that future risk assessment studies correlate the metal levels with the origin (country or region of the world) and area of cultivation of cereals. The consignments of cereals consumed in São Vicente probably differ from those consumed in Santiago. Minimizing the dietary exposure of the Cape Verdean population to metals of toxicological relevance involves the importation of higher quality cereals and with lower concentrations of

Al, Cd, Cr, Ni, Sr and Pb. Monitoring the levels of these metals on arrival in Cape Verde is a recommended risk management and minimization strategy. In addition, those cereals with higher levels of metals such as Pb and Al should not be used for the production of cereal-based products containing the husk but rather used in the manufacture of flours after being dehusked.

Table 6. Concentrations (mg/kg fresh weight), standard deviations (SD), maximum and minimum concentration values of the metals analyzed according to consumer population

	Al	Cd	Cr	Ni	Pb	Sr	Area
Mean	2.49	0.01	0.03	0.17	0.04	0.41	Santiago
SD	5.13	0.02	0.09	0.09	0.03	0.60	
Min.	0.26	0.00	0.00	0.00	0.00	0.00	
Max.	39.0	0.08	0.64	0.56	0.25	2.81	
Mean	3.93	0.01	0.07	0.20	0.05	0.87	São Vicente
SD	4.44	0.01	0.08	0.09	0.03	1.03	
Min.	0.23	0.00	0.00	0.05	0.01	0.00	
Max.	18.0	0.03	0.32	0.46	0.13	3.15	

In Cape Verde, the cereal balance for 2002/03 estimated a cereal consumption of 242 kg/year per person made up of 123 kg of maize (337 g/day), 67 kg of rice (184 g/day) and 52 kg of wheat (142 g/day). However, since there are no more current data on the consumption habits of cereals and cereal-based products, the estimations here of the dietary exposure (Estimated Daily Intake, EDI) of the Cape Verdean population to the metals under study were performed considering a mean ration of 100 g/day of each cereal and its derivatives (Table 7). The European reference limits were used for the evaluation of the EDI of the Cape Verde population. The tolerable or acceptable daily/weekly intake (TDI, tolerable daily intake; TWI, tolerable weekly intake; BMDL, benchmark dose level) and a mean average weight of an adult individual, similar to that of the Spanish population, of 68.48 [50] were used.

Table 7. Metal dietary intake assessment and evaluation

Element	EDI	%	EDI	%	EDI	%	EDI	%	EDI	%	EDI	%
	(mg/day)	Contribution	(mg/day)	Contribution	(mg/day)	Contribution	(mg/day)	Contribution	(mg/day)	Contribution	(mg/day)	Contribution
	Rice		Corn		Corn flour		Wheat flour		Corn gofio		Wheat	
Cr	0.002	0.01	0.008	0.04	0.001	0.005	0.002	0.01	0.009	0.04	0.01	0.05
Ni	0.02	10.00	0.02	7.89	0.008	4.21	0.01	6.32	0.02	11.6	0.03	13.2
Sr	0.02	0.24	0.04	0.45	0.01	0.16	0.08	0.87	0.08	0.87	0.16	1.80
Al	0.14	1.46	0.24	2.46	0.12	1.20	0.27	2.80	1.34	13.7	0.49	4.96
Cd	0.001	4.09	0.0007	2.86	0.0005	2.04	0.002	8.18	0.0003	1.23	0.001	4.09
Pb	0.003	7.50-3.00	0.003	7.50-3.00	0.003	7.50-3.00	0.003	7.50-3.00	0.008	20.0-8.00	0.007	17.5-7.00

EDI, estimated daily intake when consuming 100 g/day; Percentage of contribution (%) when consuming 100 g/day.

Considering a mean average weight of an adult of 68.48 kg [50].

Thus, the consumption of 100 g/day of corn gofio was found to provide a contribution percentage of 20% of the European BMDL of Pb set at 0.63 $\mu\text{g}/\text{kg}$ bw/day for nephrotoxic effects [13]. This percentage may represent a high contribution to the total intake of Pb with the consequent risk to health. Similarly, the consumption of 100 g/day (700 g/week) of corn gofio contributes 13.7% of the TWI (tolerable weekly intake) of Al set in Europe at 1 mg/kg bw/week [11]. The consumption of 100 g/day of wheat represents contribution percentages of 13.2% to the TDI (tolerable daily intake) of Ni of 13 $\mu\text{g}/\text{kg}$ bw/day. In the case of sensitive individuals or people with kidney problems, a high intake of Ni may harm such a person's health [9].

As for the Al levels detected in the corn gofio differently for Santiago and São Vicente islands, in the case of São Vicente (39 mg Al/kg fresh weight) the consumption of 100 g/day with an Al content of 39 mg/kg fresh weight would mean an intake of 3.9 mg Al/day from this food alone, that is, almost 39.9% of the TWI for Al.

Assuming that food risk management needs to be accompanied by a communication plan, the authors believe that the nutritional re-education campaigns and actions provided for in the PERVEMAC II Project could contribute to communicating and disseminating the knowledge of the Cape Verdean population and its authorities on the risk associated with dietary exposure to these toxic metals. Previous studies carried out in Cape Verde [51] have pointed to the success of involving women in health promotion because of their decision-making power, their multi-dimensional role in purchasing, processing and preparing food as the pillar of familial food security and also their contribution via non-formal economic activities for their families. Focus group discussions and the intensive fieldwork reinforced the higher participation of residents of the informal unit and women in all stages, suggesting the practicability of health promotion campaigns, taking into account the potential of the social capital of the informal settlements and the role of the woman in the family and society in Cape Verde [51].

Conclusions

The existence of significant differences in the content of the elements analyzed between the different cereals is confirmed, which reaffirms the need for continuous monitoring of locally produced cereals as well as imported ones. The reduction of the population's exposure to toxic metals would be by the consumption of less contaminated raw materials, and as such food safety strategies should focus on the importation and control of less contaminated cereals to produce safer cereal-based products. Monitoring the levels of these metals on arrival in Cape Verde is one risk management and minimization strategy. The cereals with higher levels of metals such as Pb and Al should not be used with the husk for the production of cereal-based products, but rather used in the manufacture of flours after removing the husk. In the case of Al, it would be advisable to invite the food safety authorities to set a maximum limit for this element in cereals and cereal-based products that allows quality control and minimization of the population's exposure to this neurotoxic element. The evaluation of the dietary exposure to the metals of toxicological interest studied here in cereals and their cereal-based products should, without a doubt, be complemented with future studies for other groups of basic foods in the diet of the Cape Verde population.

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