



Urinary Pb levels in schoolchildren from the largest coal mining area in Brazil and its associated factors: a cross-sectional study

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Abstract

Candiota region has the largest coal reserve in Brazil, and previous studies have shown moderate-high levels of metals in the environment, including Pb. On the other hand, there are no studies investigating the factors associated with high levels of urinary Pb in children in the region. To investigate this issue, a cross-sectional study was conducted with 92 schoolchildren from 7 cities comprising this coal-mining region. Socioeconomic and demographic information and parental life habits and children's health information were collected using a semi-structured questionnaire. Diurnal urine samples were collected to quantify Pb levels ($\mu\text{g/g}$ creatinine). Bi- and multivariate Poisson regression with a robust estimator was used to assess factors associated with high levels of Pb. Urinary Pb levels in children in the region vary from not detected to 21.6 $\mu\text{g/g}$ creatinine. The spatial distribution of urinary Pb levels in children indicated an influence of proximity to mining areas, and this factor was confirmed by Poisson regression analysis (bivariate). Other factors associated with high levels of urinary Pb were non-white mothers, paternal occupational exposure, and low BMI. The findings of this study reveal that the Candiota region is a hotspot for high levels of urinary Pb in children (geometric mean: 3.82 $\mu\text{g/g}$ creatinine) and that, in addition to the proximity to mining areas, factors socioeconomic and health conditions may be associated with these high levels.

Keywords Candiota · Children · Lead · Biomonitoring · Urine · Coal

Introduction

Activities involving the extraction of ores have been studied for decades due to their impacts on the environment and human health. A recent report prepared by Greenpeace Southeast Asia (GSA) in partnership with the Center for Research on Energy and Clean Air (CRECA) estimated that pollution generated, mainly by burning fossil fuels, is responsible for approximately 4.5 million premature deaths worldwide each year. This problem occurs because air

pollution contributes to the increased incidence of chronic and acute respiratory diseases (Farrow et al. 2020).

World coal reserves are estimated at 847.5 billion tons, with most of these reserves located in the USA, Russia, and China, which hold about 60% of the world's reserves. According to the Companhia de Pesquisa de Recursos Minerais (CPRM), Brazil's coal reserves are mostly located among the states of the southern region of the country, with the Candiota deposit, in the Rio Grande do Sul state, comprising 38% of all coal national territory (CPRM 2014).

In coal mining and processing activities, particulate matter and toxic gases can be released into the atmosphere with the potential to pollute the environment and compromise human health (Bian et al. 2010). From these activities, Pb stands out due to its relative abundance in coal mining areas (He et al. 2020) and its high toxicity (Rahman and Singh 2019; Kumar et al. 2020). According to the World Health Organization (WHO), Pb is among the ten chemicals of greatest concern to public health and highlights that the organization's Member States need to promote actions to

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protect the health of workers, children, and women of reproductive age (WHO 2021).

Human exposure to Pb can occur in several ways: industrial processes such as lead smelting and coal combustion, gasoline, lead-based paints, and battery recycling, among others (Flora et al. 2012). According to the Center for Disease Control and Prevention (CDC), until the 1980s, the main source of exposure for most of the US population came from aerosols produced by vehicles that used gasoline containing Pb in their composition. However, since its elimination from gasoline, Pb contamination for the adult population has been mainly restricted to occupational activities, while for children, the main source of contamination is lead-based paints and soil contamination (CDC 2021a).

Recent studies in Candiota have shown that in regions close to mining areas, the concentration of Pb in the soil and in the particulate matter (PM10) is close to the limits established in national legislation (da Silva Júnior et al. 2019; Bigliardi et al. 2021). In addition, the study by da Silva Bonifácio (2021) showed that Pb levels in surface waters near the region are above the limits recommended by Brazilian legislation ($10 \mu\text{g l}^{-1}$). Among the negative human health outcomes are DNA damage (Pinto et al. 2017), impaired intellectual development of children (Dupont-Soares et al. 2021), and altered lung function (Bigliardi et al. 2022) and blood biochemical marker (Bigliardi et al. 2021) in adults. A previous study of biomonitoring of urinary levels of metals has already been conducted in the region, but without concern to investigating the spatial distribution and factors associated with high levels of metals (Dos Santos et al 2018). Therefore, the present study aims to (1) spatialize urinary Pb levels according to the place of residence of children living in the Candiota coalfield region and (2) identify the factors associated with high Pb levels in these children.

Materials and methods

Ethical aspects

The ethical aspects established by Resolution 466/12 of the National Health Council, of the Ministry of Health, which regulates research with human beings in Brazil, were respected. This study was approved by the Research Ethics Committee in the Health Area of the Federal University of Rio Grande (no. 36/2013). The children's parents or guardians were asked to sign the Free and Informed Consent term (Supplementary Information) in advance. Schoolchildren were asked to sign the Free and Informed Assent term. Both documents accompanied the questionnaire, and only those who agreed to the terms and submitted the documents in writing were included in the study. In addition, the exams were returned to the participants.

Sample and study design

This is a cross-sectional study with school-age children from 6 to 11 years old who live in Candiota and neighboring municipalities. The sample size was calculated from the universe of 778 schoolchildren in the region in the chosen age of the study, considering a confidence level of 95% and a margin of error of 10%. The ideal sample size for the study was 86 schoolchildren. The sample number ($n=92$) consisted of children from public schools from each location and was randomly selected. The list was provided by the education department of each municipality, and the collections were carried out in 2013.

Study area

The area covered by the study includes seven municipalities located in a coal region in southern Brazil, in the state of Rio Grande do Sul (Aceguá, Bagé, Candiota, Herval, Hulha Negra, Pedras Altas, and Pinheiro Machado) (Fig. 1). In Candiota, there are two coal-fired power plants in operation and two coal mining areas (minor coal mine $31^{\circ} 26' 49.7'' \text{ S}, 53^{\circ} 42' 26.4'' \text{ W}$ and major coal mine $31^{\circ} 34' 59.7'' \text{ S}, 53^{\circ} 43' 15.6'' \text{ W}$). In addition, the influence of a copper mining area located outside the limits of Candiota was also considered, since two municipalities included in the study border the municipality where the copper mine is located.

Demographic and socioeconomic data

A semi-structured questionnaire was applied with socioeconomic and demographic information on the family, parents' habits, and child health information. The main information extracted from the questionnaire was selected and considered for the theoretical model of Poisson regression analysis.

Urine collection and analysis

Urine samples were collected only once in the morning in sterilized plastic containers, identified, packed in a box at 4°C , and analyzed in a laboratory with accreditation to ISO/IEC 17,025 (ToxiLab, Porto Alegre, Brazil). Before collection, the containers were kept in a 10% nitric acid solution for at least 24 h and then rinsed with deionized water for decontamination. Pb concentration was determined from graphite furnace atomic absorption spectrophotometry (GF-AAS) and normalized to creatinine

Fig. 1 Study area highlighting the investigated municipalities (Pinto et al. 2017)



concentration. The limit of quantification (LoQ) was $0.5 \mu\text{g/g}$ of creatinine. Details of the collection and analysis methodology can be consulted in dos Santos et al. (2018).

Spatial distribution

Information on the geographic coordinates of each participant's residence was collected, and then, the concentration of urinary Pb was spatialized. QGIS 3.5.1 was used to make the map. The geographic coordinates of mining areas and coal-fired power plants were also plotted on the map.

Data analysis

A univariate analysis was performed to describe the data obtained through the questionnaires followed by bi- and multivariate analyses using Poisson regression with a robust estimator. In the Poisson regression, the dependent variable (urinary Pb level) was dichotomized into values equal to or below $1.14 \mu\text{g/g}$ of creatinine and values above this threshold. The cutoff point was defined from data obtained from the National Report on Human Exposure to Environmental Chemicals, considering the 95th percentile and the age

group from 6 to 11 years in the 2015–2016 cycle of the National Health and Nutrition Examination Survey (CDC 2021b). The most recent update to this manual is dated March 2021. The independent variables that were part of the theoretical model for the determination of risk were separated into four levels, described below:

Level 1 — Socioeconomic and demographical variables Mother's education (completed elementary school or less, incomplete high school or more), biological mother's color (non-white, white), mother's marital status (single/married), economic class according to the Brazilian Association of Research Companies (ABEP) (poorest/richest), and per capita family income about the minimum wage (MW) ($> 1 \text{ MW}$ or $\geq 1 \text{ MW}$).

Level 2 — Living and working conditions Distance to the source of pollution — two coal-fired power plants, two coal mines, and one copper mine — closest to the residence (less than or equal to 2 km, between 2 and 10 km, and greater than 10 km), mother's occupational exposure (mechanics, hairdressers, agriculture or livestock), father's occupational exposure (coal-fired power plant, agriculture or livestock), current job (mother and father).

Level 3 — Life habits and exposure of the child Smoking mother, smokers in the household (including the mother), and heating with biomass burning (wood stove or fireplace).

Level 4 — Characteristics of the child Gender (male, female), skin color (non-white, white), asthma (yes or not), and Z score for BMI, considering whether the child was under ideal weight (percentile < 5), above ideal weight — overweight and obese — (percentile ≥ 85) or within the ideal weight range (percentile between 5 and 85).

Results

Information on urinary Pb levels in schoolchildren is described in Table 1. The geometric mean of the sample was 3.82 µg/g creatinine, with 3.99 µg/g creatinine among boys and 3.67 µg/g creatinine among girls. A Mann–Whitney test was performed, but significant differences between the sexes were not found. About a quarter of the children had urinary Pb levels below the limit of quantification and the highest value was 21.6 µg/g of creatinine. Furthermore, while for our study the median was 3.50 µg/g of creatinine, the median for the same age group according to the CDC in 2013 was 0.31 µg/g of creatinine (CDC 2021b).

Table 1 Descriptive data on urinary Pb levels in the population studied (n=92)

	N	%	Urinary lead levels (µg/g creatinine)								
			Geometric mean ^a	Mean	DP	Min	Max	10th percentile	50th percentile	90th percentile	95th percentile
Total	92	100	3.82	4.42	4.47	<LOQ ^b	21.6	<LOQ ^b	3.50	10.09	11.13
Boys	47	51.09	3.99	4.59	5.32	<LOQ ^b	21.6	<LOQ ^b	3.20	10.98	16.75
Girls	45	48.91	3.67	4.24	3.41	<LOQ ^b	10.9	<LOQ ^b	3.70	9.74	10.08

^aNull value was disregarded in the calculation of the geometric mean

^bValues below the limit of quantification (0.5 µg/g creatinine). A total of 26.1% of the samples were below the quantification limit

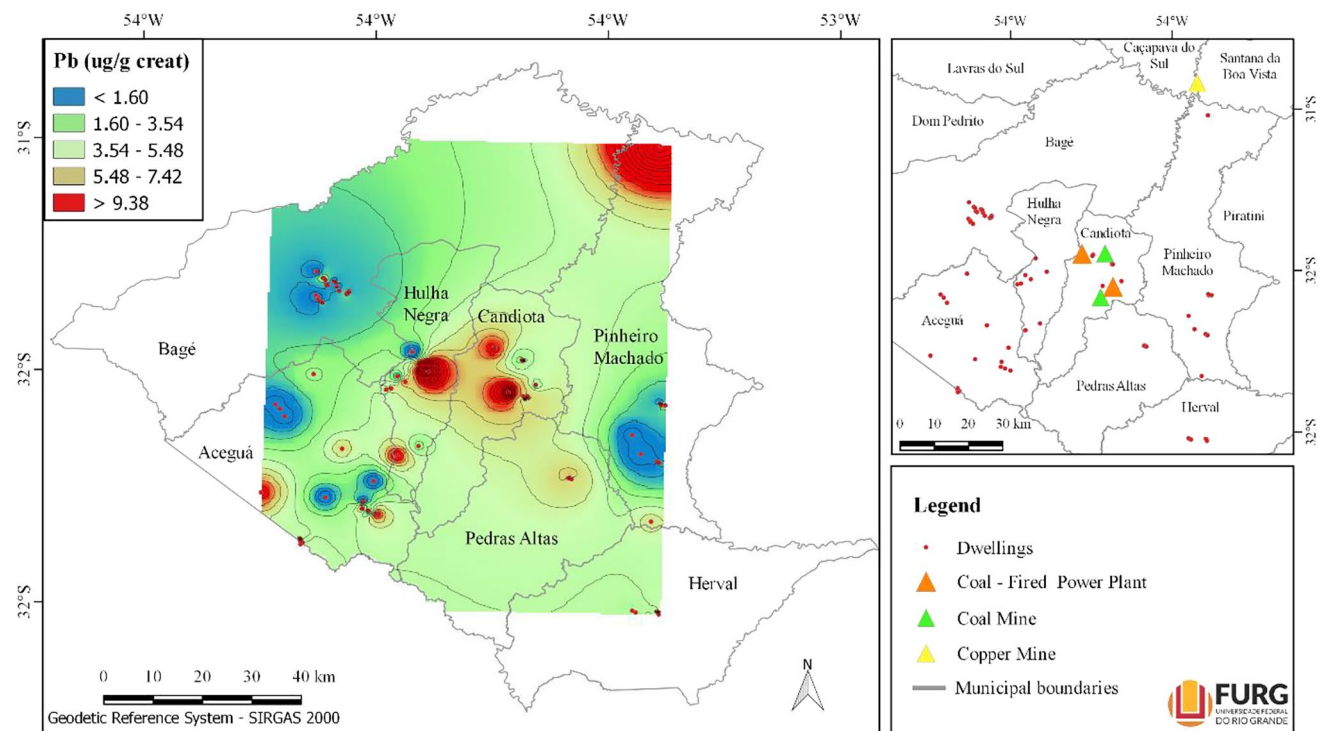


Fig. 2 Sources of pollution and spatial distribution of urinary Pb levels of the 92 children evaluated in the 7 cities of the study

Table 2 Descriptive analysis of socioeconomic and demographic characteristics of children in Candiota and region ($n=92$)

Variables	<i>N</i>	%
Mother's socioeconomic conditions		
Years of schooling		
< 12	62	68.13
≥ 12	29	31.87
Skin color		
Non-white	41	45.05
White	50	54.95
Marital status		
Single	24	26.09
Married	68	73.91
Economic class		
Poorest (C1, C2, and DE)	70	76.09
Richest (A, B1, and B2)	22	23.91
Per capita income*		
< Minimum wage	81	88.04
≥ Minimum wage	11	11.96
Occupational and housing conditions		
Municipality		
Candiota	19	20.65
Other municipalities	73	79.35
Distance from the nearest source of pollution		
< 2 km	7	7.61
≥ 2 km and < 10 km	12	13.04
≥ 10 km	73	79.35
Occupational exposure (mother)		
Yes	14	15.22
No	78	84.78
Occupational exposure (father)		
Yes	29	38.16
No	47	61.84
Mother currently working		
Yes	50	54.35
No	42	45.65
Father currently working		
Yes	68	88.31
No	9	11.69
Life habits		
Smoking mother		
Yes	30	32.97
No	61	67.03
Smokers in the house		
Yes	46	50.00
No	46	50.00
Heating with biomass burning		
Yes	46	50.00
No	46	50.00
Child characteristics		
Gender		
Male	47	51.09

Table 2 (continued)

Variables	<i>N</i>	%
Female	45	48.91
Skin color		
Non-white	37	40.22
White	55	59.78
Asthma		
Yes	30	32.61
No	62	67.39
Z score for BMI		
Overweight	30	32.61
Underweight	7	7.61
Ideal weight	55	59.78

*Minimum wage in the year of the survey (2013) was R \$678.00 (or approximately US \$289.74 in August 2013)

Regarding the spatial distribution of urinary Pb levels based on the place of residence of the students, the highest levels are located close to the main sources of pollution in the region (thermoelectric plants, coal, and copper mines) (Fig. 2).

Table 2 shows the socioeconomic and demographic variables, housing conditions, life habits, and child characteristics that were the basis for the theoretical model of Poisson regression. Approximately 55% of the children had white mothers, 73.91% of the mothers lived with a partner, 68.13% had mothers with 12 or more years of schooling, less than 24% were in the upper strata of economic class, and almost 90% had family income below 1 minimum wage. Regarding living and working conditions, almost 80% of children lived more than 10 km away from some source of pollution, almost 85% of children had mothers without occupational exposure, and this percentage was almost 62% for fathers. A total of almost 33% of the children had a smoking mother, but 50% of the children lived with an active smoker. Half of the children had indoor biomass burning. Regarding the characteristics of the children, 51% were boys, almost 60% of the children were white, 32.61% had a history of asthma, and almost 60% had an adequate BMI.

Crude and adjusted Poisson regression are shown in Table 3. Crude analysis showed that living close to potential sources of pollution (< 2 km) is a risk factor for high urinary Pb levels among children in the PR region = 1.46 (95% CI 1.23–1.73). Furthermore, living between 2 and 10 km also significantly increases the risk of high urinary Pb levels (PR = 1.33, 95% CI 1.03–1.71). Another significant factor in the bivariate analysis (crude) was BMI. Children with a low BMI had a prevalence ratio of 1.33 (95% CI 1.12–1.58).

According to the adjusted analysis, the variable mother's skin color showed that non-white mothers were associated with high urinary Pb levels (PR 1.35; 95% CI 1.01–1.79). In

Table 3 Crude and adjusted analyses of risk factors for increased concentration of urinary Pb levels in children from Candiota

Level	Variables	Crude				Adjusted			
		Prevalence ratio	95% CI Min	95% CI Max	<i>p</i> -values	Prevalence ratio	95% CI Min	95% CI Max	<i>p</i> -values
	Mother's socioeconomic conditions								
1	Skin color				0.06				0.03
	Non-white	1.30	0.98	1.71		1.35	1.01	1.79	
	White	1				1			
	Occupational and housing conditions								
2	Distance from the nearest source of pollution				<0.01				0.14
	<2000 m	1.46	1.23	1.73		1.27	0.92	1.75	
	≥2000 and <10,000	1.33	1.03	1.71		1.25	0.94	1.67	
	>10,000	1				1			
2	Occupational exposure (father)				0.09				0.02
	Yes	1.39	1.08	1.79		1.33	1.03	1.73	
	No	1				1			
	Child characteristics								
4	Z score for BMI				<0.01				0.15
	Overweight	0.83	0.58	1.18		0.78	0.55	1.12	
	Underweight	1.33	1.12	1.58		1.10	0.86	1.41	
	Ideal weight	1				1			

addition, it was also observed that the father's occupational exposure increases the risk of the high urinary Pb levels in these children (PR 1.33; 95% CI 1.03–1.73).

Discussion

The impacts of coal mining activities are recurrently discussed in the literature, and it is not uncommon for Pb to stand out among the elements that pose the greatest risk to human health (Ishtiaq et al. 2018; da Silva Bonifácio et al. 2021). Exposure of children living in risk areas may be higher because they explore environments through manual activities (such as bringing their hands to their mouths, for example) (Penteado et al. 2021) or due to exposure to house dust (Lanphear et al. 2016). Regarding peripheral nervous system damage, adults tend to be more susceptible, whereas damage to children — with the nervous system still developing — is more evident and occurs in the central nervous system (Bellinger 2004). It has already been seen that for school-age children, even low concentrations of Pb in the blood can be associated with a decrease in intellectual capacity, using the intelligence quotient as an evaluative parameter (Lanphear et al. 2016; Wu et al. 2017; Pan et al. 2018).

In our study, we observed that Pb values in children's urine ranged from values close to 0 to 21.6 µg/g creatinine for boys and 10.9 µg/g creatinine for girls. These levels

could represent isolated cases; however, the 90th percentile (10.98 µg/g creatinine for boys and 9.74 µg/g creatinine for girls) indicates that there is a considerable number of samples that presented high levels of Pb in the urine of these children. Comparatively, the value we used as a reference according to the CDC (1.14 µg/g creatinine) is almost 10 times lower than that found in the study area. While for the CDC the 90th percentile of children of both sexes aged between 6 and 11 years considered not exposed is 0.85 µg/g creatinine (2015–2016 NHANES cycle), the 90th percentile including boys and girls from Candiota and region considering the same age group was almost 11 times higher (10.09 µg/g creatinine) (CDC 2021b).

According to CDC data, urinary levels of Pb in children have decreased over the years, possibly as a result of public policies aimed at protecting the health of the population. Despite this, our study shows that potentially polluted locations provide more hazardous exposure scenarios than those encountered more than two decades ago (CDC 2021b).

The environmental exposure of children in areas under the influence of Pb contamination can raise the levels of the metal in the body. A study in Bangladesh points out that all children evaluated ($n = 69$) who lived near lead-acid battery recycling sites had blood Pb levels above 5 µg/dL, that is, above the limit established by the World Organizations of health. The authors also point out that Pb concentrations were confined to the topsoil layer (1–2 cm) (Chowdhury et al. 2021). In addition, a study in China evaluating the

levels of Pb in the blood between 1997 and 2015 found that the concentration of Pb in the body has been falling, but is still high. The authors highlight that soil and poor air quality are important pathways for Pb exposure in children (Han et al. 2018).

The spatialization shows that children with higher urinary Pb levels reside in Candiota and highlights that the copper mine may influence these children's urinary Pb levels. Previous studies in the region reveal that the concentration of Pb in soils decreases as they move away from the thermoelectric plant within a radius of up to 10 km. However, soil Pb concentration undergoes an apparent increase in directions that coincide with coal mines (da Silva Júnior et al. 2019).

Although Candiota concentrates most of the sources of pollution evaluated in the study, some factors may contribute to the contamination of neighboring cities. The Candiota region has low/moderate concentrations of potentially hazardous elements in soil samples near the thermoelectric plants in the region; however, there is a concern regarding the influence of winds on the dispersion of contaminants (da Silva Júnior et al. 2019). In addition, it was observed that levels of potentially hazardous elements at wastewater discharge points from mining activities may be lower at different points in a river (mainly downstream), showing that the impact of these activities is not necessarily local and that contamination can reach regions far away from sources of pollution (Ali et al. 2017).

Urinary Pb levels around 1.50 $\mu\text{g/g}$ of creatinine may be positively associated with several risk factors for cardiovascular disease, including blood pressure (Lin et al. 2020). In addition, in this same study, it was also observed that the Z score for BMI was positively associated with a high concentration of Pb in the urine in underweight children and negatively associated with overweight children of their age, which is in line with our findings. This may be related to the fact that overweight children accumulate the metal in the tissues, while underweight children excrete the metal because they do not favor the accumulation of the metal in the body. Additionally, it has already been reported in the literature that animal models exposed to Pb had a significant increase in weight, triglycerides, hepatic lipid accumulation, and alteration in DNA methylation (Sun et al. 2017). A recent study has observed that some substances may have protective effects against the effects caused by exposure to Pb. In an experiment with Wistar rats, α -lipoic acid was shown to protect neurological, immunological, and reproductive organs against biochemical changes caused by Pb and ZnO nanoparticles (Deore et al. 2021).

Socioeconomic factors may also be associated with high levels of Pb in the urine. Studies in China and India evaluated children (boys and girls) and observed that factors such as age, the place where children played (inside or outside), smoking parents, father education, mother education,

drinking water (tap or bottled), family income, and housing conditions were related to higher levels of urinary Pb (Rashid et al. 2019; Martínez-Hernanz et al. 2020). In our study, only the mother's skin color was associated with the children's urinary Pb levels ($p = 0.03$). Despite not being considered significant in the multivariate analysis, the per capita family income for 81 families (88.04%) was below the minimum wage at the time, reinforcing that social vulnerability can become an aggravating factor for those people who are environmentally exposed in the region's mining area.

The origin of drinking water was not associated with the concentration of Pb in children's urine; however, da Silva Bonifácio (2021) observed that in some points of Candiota, the concentration of Pb in water bodies exceeds the values allowed by Brazilian legislation ($10 \mu\text{g l}^{-1}$) and is very close to the value considered risk according to the Human Health Risk Assessment methodology of the US Environmental Protection Agency. This information must be taken into account for the implementation of public policies to preserve the health of families, especially those that do not consume treated water.

Despite the important findings of our study, some limitations should be highlighted. Due to the lack of information, some variables could not be incorporated into the statistical analysis. For example, information on the origin and destination of sewage or the source of water supply was not well understood by the study population. We also understand that evaluating other biomarkers (blood pressure, DNA damage, or neurological damage, for example) in addition to urinary Pb levels in children would make our study more robust; however, it was not possible to perform such parameters. Finally, some associated factors must be acting together to increase the risk of high Pb levels (for example, living close to the source of pollution and paternal occupational exposure) and, in this case, the individual contribution of each factor may be influenced by other factors.

Conclusion

In the present study, different aspects related to the exposure of school-age children were evaluated in the Candiota region, where the largest mineral coal reserve in Brazil is located. Faced with many contaminants that can be studied in these areas, Pb stood out for presenting much higher urinary levels (4.42, 3.50, and 11.13 $\mu\text{g/g}$ creatinine for mean, 50th percentile, and 95th percentile, respectively) of those considered benchmarks by the Centers for Disease Control and Prevention. It was also seen that the highest concentrations of Pb in the urine of children are concentrated in residents of the municipality of Candiota and residing less than 2000 m from some source of pollution.

Among the factors that may contribute to these high Pb levels are the mother's socioeconomic characteristics, such as skin color, and the father's occupational exposure. Due to this problem, governments and those responsible for public health policies should spare no effort to face the issues that permeate the vulnerability of these children, once they are socially and environmentally exposed.

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Author contribution All authors read and approved the final version of the article. RLB was responsible for reviewing the data, analyzing, interpreting the data, and writing the text. UJSJ was responsible for collecting and interpreting the data and preparing the maps. ALMB was responsible for correcting the text and discussing the results. FMRSJ was the study supervisor.

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Data availability All relevant data and material are visible in the manuscript/supplementary material.

Code availability Not applicable.

Declarations

Ethics approval This study was approved by the Research Ethics Committee in the Health Area of the Federal University of Rio Grande (Nº 36/2013).

Consent to participate The children's parents or guardians were asked to sign the Free and Informed Consent term in advance. Schoolchildren were asked to sign the Free and Informed Assent term.

Consent for publication The manuscript is reviewed and approved by all authors.

Conflict of interest The authors declare no competing interests.

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