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Assessment of potential human health, radiological and ecological risks around mining areas in northeastern Brazil

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Abstract Mining is responsible for the release of metallic pollutants and radioactive materials into the environment, which have the potential to disrupt ecosystems and pose significant risks to human health. Significant mining activity is concentrated in the municipality of Caetité (northeastern Brazil), where Latin America's only active uranium mine and significant iron ore deposits are located. Although previous studies have shown that the regional soil and water resources are highly contaminated by various toxic elements and that exposure to these elements is

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A. M. F. Fausto · A. W. Mol · F. G. Velasco Center for Research in Radiation Sciences and Technologies (CPqCTR), State University of Santa Cruz, Ilhéus, Bahia, Brazil known to have adverse effects on human health, the health risks in this mining region have never been assessed. The aim of this unprecedented comprehensive investigation was to assess the health, radiological and ecological risks in this mining region, which is home to nearly 100,000 people. To achieve our goal, soil and water samples were collected in the vicinity of the mines and in the main settlements in the region. Fifteen metallic toxic elements were determined using Instrumental Neutron Activation Analysis and Inductively Coupled Plasma Optical Emission Spectrometry. The HERisk code, which follows the main methodological guidelines for risk assessment, was used to quantify human health, radiological and ecological indices. The average values of the total risk and cancer risk indices indicated that region falls into the moderate risk category $(1.0 \le HI_{tot} < 4.0)$. However, 63% of the sites had high risk values, with Fe, Co and As being the metals contributing most to total and cancer risk, respectively. Near the mining areas, the potential ecological risk can be considered extreme (PERI \geq 600). The values of the calculated radiological indices correspond to typical values in natural uranium areas. However, in the communities near the mine, the dose values are slightly above the permissible limit (1 mSv y^{-1}), so they must be continuously monitored, and risk mitigation measures must be taken.

Keywords Mining impacts · Toxic metallic trace elements · Naturally occurring radioactive materials(NORM) · Non-carcinogenic & carcinogenic effects · Radiological hazard

Introduction

Mining has played a prominent role in the development of human society, as it provides raw materials for most industrial processes. However, it is one of the anthropogenic activities that has the most negative impact on the environment and society (Worlanyo & Jiangfeng, 2021). Several studies focused on the impacts of mining on the environment worldwide agree that it is responsible for the release of various pollutants into the environment, which can alter the entire ecosystem and pose significant risks to human health. For this reason, mining areas are considered as regions that are particularly vulnerable to environmental degradation (Enyigwe et al., 2021; Haddaway et al., 2019; Magidi & Hlungwani, 2023; Mundaca, 2024).

The waste generated by mining processes contains significant concentrations of sulphated compounds and metallic species (Fernandes et al., 2008). Discharge of mining wastewater, spills, leakages of hazardous substances, and windblown dust facilitate the migration of these pollutants and increase their concentrations in adjacent soils and water resources, altering their physico-chemical properties (Doležalová et al., 2019; Egbueri & Enyigwe, 2020; Ling et al., 2019). These disturbances can lead to soil degradation and loss of productivity, affect the quality of water resources and destroy aquatic life. They can also threaten biodiversity and cause significant changes in land use, which can lead to food insecurity and a high cost of living (Qin et al., 2021; Worlanyo & Jiangfeng, 2021).

Uranium mining requires special attention, as a naturally occurring radioactive material (NORM) is mined here. The processes of uranium mining alter the secular equilibrium of the natural radioactive chains (²³⁸U, ²³⁵U and ²³²Th) and release the radionuclides that make them up into the environment. In addition, rock wastes contain a large group of longlived radioactive elements (*e.g.* ²³⁴U, ²³⁰Th, ²²⁶Ra, ²¹⁰Pb, etc.), that can be classified as technologically enhanced natural radioactive materials (TENORM) (IAEA, 2005; Fernandes et al., 2008). The contamination of environmental compartments by these radionuclides can last for generations and poses a risk to human health due to their high toxicity and associated radiobiological effects (IAEA, 2005; Bonotto, 2015; Corlin et al., 2016). For example, the release of radon isotopes (^{222,220}Rn) in areas with uranium deposits poses a significant health problem, as inhalation of this gaseous element increases the risk of lung cancer (IAEA, 2004).

In the municipality of Caetité (state of Bahia, NE Brazil), there are several uranium-enriched zones with a total reserve of ca.112,000 metric tons of triuranium octoxide (U_3O_8); it is therefore considered the most important uranium province in Brazil (Lobato et al., 2015). Since 1999, uranium exploitation has been concentrated Caetité, where the only active uranium mine (open-pit mine, area: 1700 ha) in Latin America is located (INB, 2021). In addition, the southern part of the municipality hosts an important iron ore deposit with more than 10 billion tons of iron (CETEM, 2013). The "Pedra de Ferro" mine, built to produce up to 20 million tons of iron per year, would make Bahia the third largest iron-producing state in the country (BAMIN, 2016).

The contamination of water resources and soil in these mining areas has been previously assessed and discussed in Santana et al. (2020) and Montalván-Olivares et al. (2021), respectively. In the first study, eleven metallic elements were determined in surface and groundwater samples. The results indicated significant contamination by Cd, Pb, and U, based on the limits set by Brazilian and international legislation. In Montalván-Olivares et al. (2021) 34 elements were determined in regional natural, agricultural, and urban soils. It was reported that almost 90% of the samples collected had metal contents above the recommended limits of Brazilian legislation, so the region could be classified as highly contaminated. Both studies agreed that, at the regional level, mining and agriculture are mainly responsible for the introduction of pollutants such as toxic trace metals (As, Cd and Pb) into soils and water resources and for the alteration of the content of natural elements such as Al, Fe, K, Th, and U.

It is well known that soil and water pollution are closely linked, as soil is able to accumulate pollutants and then release them into waters bodies through leaching and migration (Kiliç, 2021). Furthermore, soil and water are direct pathways for the transfer of toxic metals to crops and livestock, playing a crucial role in food safety and thus human health (Qin et al., 2021).

As has long been discussed and reported, human exposure to toxic elements and NORM is of great public concern and is associated with various adverse effects on human health (Abba et al., 2024; Järup, 2003). Iron, for example, causes some effects such as nausea, hematemesis, and vomiting, which are very dangerous especially for children under six years old (Yuen & Becker, 2020). The toxic effects of Fe have also been associated with neurological disorders such as Parkinson's disease and suicide (Sane et al., 2018). Continuous exposure to Ba, Co, Cr, Cu, and V can lead to nephrotoxic effects, gastrointestinal complaints, skin irritation as well as reproductive impairment, damage to the fetus, and neurotoxic effects (exacerbation of manic depression) (Yu & Yang, 2019). As, Cd and Pb are linked to hematological disorders (anemia, inhibition of hemoglobin synthesis), toxic effects on the kidneys (renal tubular dysfunction), cardiac arrhythmias, cognitive disorders (especially in children), and high exposures can lead to death. They are also associated with skin, lung and liver cancer, among others (Agbasi et al., 2023; Martin & Griswold, 2009). In addition, exposure to radioactive elements can cause minor burns, cataracts, respiratory irritation, sterility, various types of cancer and congenital diseases (IAEA, 2005).

The proven contamination of water resources and soils with metallic elements in the area of influence of the two mines in the Caetité region represents an unfavorable and high-risk scenario for the local population. Approximately 100,000 people live in this region, most of whom use these natural resources to develop subsistence activities such as agriculture and livestock farming (IBGE, 2021). Despite the potential adverse effects described above, no risk assessment study has yet been carried out in this mining region. Therefore, the aim of this work is to assess the potential risks resulting from exposure to toxic elements and NORM to which the local population is subjected. Based on the concentration values of 15 elements determined in the regional soils and water resources, the risks to human health, radiological and ecological risks were assessed.

This research, unprecedented in the region, highlights the potential impact of mining on human health and the environment, an issue of great public concern. It also provides citizens, local authorities and the international community with solid arguments for the development of risk management strategies to mitigate impacts, restore natural resources and protect the health of the local population.

Materials and methods

Description of the study area

The study was conducted in the areas of influence of the iron and uranium mines of the municipality of Caetité, located in the central-southern region of the state of Bahia (Northeastern Brazil) (Fig. 1). These areas also include communities from the neighboring municipalities of Lagoa Real, Guanambi, and Pindaí. The entire region is characterized by a semiarid climate (Köppen-Geiger classification: Aw) with temperatures between 17°C and 35°C. The average annual precipitation is around 72 mm and rainfall is concentrated in the months of November to January (the summer season in the southern hemisphere).

Geologically, the region belongs to the "Lagoa Real" lithological unit, a granitic-gneissic complex characterized by the presence of alkaline granite bodies, orthogneisses, albite rocks and amphibolites (Montalván-Olivares et al., 2021). The predominant soil types are dystrophic red-yellow latosols, eutrophic cambisols and neosols (arenosols). The former have high permeability (percolation 150 mm h^{-1}), while the latter are extremely susceptible to erosive processes. The study area is characterized by ephemeral rivers that remain dry most of the year, except during the rainy season. The Fundo, São João and Grande Rivers, which flow in the area of influence of the mines, are indirect tributaries of the Contas and São Francisco Rivers, the two main watersheds of the state of Bahia (Santana et al., 2020). In addition, the Grande River flows into the Ceraíma reservoir, the region's main water supply. The geological characteristics of the region favored the formation of fissured rock aquifers, resulting in an area with high permeability and good water storage. For this reason, groundwater is the main source of fresh water supply for the inhabitants of these mining areas (Lamengo et al., 2008).

Fig. 1 Location of the municipality of Caetité in the state of Bahia (Northeast Brazil). The studied mining facilities and the neighboring municipalities of Lagoa Real, Guanambi, and Pindaí are also shown on the right-hand side





There are important settlements in the neighborhood of the mining areas, where about 100,000 people live (IBGE, 2021). The urban areas of the municipalities of Caetité and Lagoa Real are located less than 40 km from the uranium mine and are therefore considered within the Direct Area of Influence (DAI). The districts of Maniaçu (11 km) and São Timóteo (14 km) are located in the Special Area of Influence (SAI), which extends up to 15 km from the mine. In addition, there are small farms and communities of family groups that traditionally inhabit these places, such as Buração (2.5 km), Gameleira (3.5 km), Juazeiro (8 km), Tamanduá (3 km), among others, of which Juazeiro is the most populated with about 800 inhabitants (INB, 2015). Likewise, within the DAI of the iron mine are the districts of Brejinho das Ametistas (15 km), Guirapá (18 km) and Ceraíma (21 km).

It is worth mentioning that, in addition to mining activities, the agricultural sector also plays an important role in the economic dynamics of the region. Livestock, dairy production and food (fruit, coffee, beans, cassava, corn and sorghum) stand out as the main commodities of the region (IBGE, 2021). Sampling procedure and chemical analysis

Soil (agricultural and urban) and water samples from taps, artesian wells, and water bodies (used by the population) were collected in two campaigns (rainy and dry seasons) at 43 locations around the iron and uranium mines and in the main population settlements in the region. Sampling procedure was carried out according to the criteria of the U.S. Environmental Protection Agency (USEPA, 1989a) and the Brazilian environmental authorities (CETESB-ANA, 2011; EMBRAPA, 2014). The soil samples were taken at a depth of 20 cm from the surface layer. Five individual samples were randomly collected in a $5 \times 5 \text{ m}^2$ area, and then combined and homogenized into a single composite sample of approximately 1 kg. The water samples were collected at a depth of up to 30 cm from the water sheet. For proper preservation, they were acidified by adding 3 mL L^{-1} of concentrated nitric acid (HNO₃).

The composition of the soil samples was analyzed by Instrumental Neutron Activation Analysis (INAA) in the research reactor IEA-R1 of the institute for Nuclear and Energy Research (IPEN). Thirty elements were determined by subjecting the samples to short- and long-term irradiations.¹ The soil samples were also analyzed with Inductively Coupled Plasma Optical Emission Spectrometry (ICP OES) to determine Cd, Cu, Ni, and Pb, which have some difficulties when measured with INAA (Montalván-Olivares et al., 2021). The digested soil samples were analyzed with a spectrometer (Varian 710-ES), which had previously been calibrated using Merck standards. Table S1 (Suppl. Material) contains relevant information on the main parameters of the radionuclides measured by the INAA, the spectral lines used for the ICP OES analysis and the limit of quantification (LOQ) for each element. To verify the accuracy and precision of the measurements, five certified rock and soil reference materials $(CRM)^2$ were irradiated (digested) and measured under the same conditions as the samples under study. The Z-score test was used to compare the concentration values obtained with the values given for the CRM. For all elements, the Z-score values were within the accepted range and 90% between [-1,1] (see Figure S1, Suppl. Material).

The water samples were analyzed only by ICP OES with the same spectrometer, and eleven elements (Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, U, V, and Zn) were quantified. The standard procedure proposed by the ISO 11466 (1995) was used for the acid digestion of the water samples. To ensure the reliability of the metal analysis, subsamples of the CRM NIST 1643f (*trace metals in water*) were prepared and measured under the same conditions as the water samples. Also in this case, the Z-score test showed a good agreement between the certified and the measured concentrations (see Table S2, Suppl. Material).

The concentration values of all elements included in the risk assessment determined in soil and water samples collected in the 11 human settlements considered in this study are listed in Tables S3-S5 (Suppl. Material). Readers wishing to delve deeper into the methodological aspects of the chemical analysis of the soil and water samples are invited to consult our previous works Santana et al., 2020 and Montalván-Olivares et al., 2021.

Human health risk assessment

The risks to human health were assessed for 15 trace elements: Al, As, Ba, Br, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, U, V, and Zn. This selection was made on the basis of toxicological and epidemiological studies, linking this elements to potential adverse health effects (cardiovascular, gastrointestinal, reproductive, respiratory, and neurological disorders) (ATSDR, 2008; Jarüp, 2003; Martin & Griswold, 2009; Yu & Yang, 2019). In addition, some of these elements have been included by the International Agency for Research on Cancer (IARC) in the different groups of carcinogenic substances (Group 1: As, Cd, Cr, and U; Group 2A: Co, and Pb; Group 2B: Ni, and V), as they are associated with the occurrence of different types of cancer (IARC, 2021).

In this study, the three main scenarios (agricultural, industrial, and residential) and seven routes of human exposure to these elements were considered. Regarding the exposure to contaminants in the soil, the following pathways were considered: accidental ingestion, inhalation of resuspended particles emitted from the soil, and dermal contact. For the waterrelated exposure pathways (ingestion and dermal contact), water samples from taps or artesian wells were considered. As elements present in soil and water can be transferred to plants and animals that are part of the human food chain, exposure to metal contaminants via dietary intake was also included in the risk assessment. Given the strong presence of agriculture and livestock farming in the region, the main foods considered were animal and plant foods harvested and consumed by the local population (cow's milk, lettuce, banana, carrot, beans, cassava and passion fruit). Based on the habits of the local population, the ingestion of soil and water was considered acute or short-term exposure. In contrast, the other routes were considered as chronic or long-term exposure.

The average daily dose (*ADD*) for each exposure route was calculated using standard USEPA (1989b, 2005, and 2011) and CETESB (2001) equations (see Eqs. S1 to S6, Suppl. Material). In the case of dietary ingestion, the concentrations of metal contaminants in food were estimated using the transport equations proposed by Health Canada (2005) (see Eqs. S7 to

 $^{^1}$ Short irradiations: Time: 10 s, irradiated sample mass: 78.5 ± 0.5 mg.

Long irradiations: Time: 8 h, irradiated sample mass: 108.5 ± 0.5 mg.

² List of CRM: BE-N (basalt), GS-N (granite), JB-1 (basalt), W1 (diabase), and IAEA-Soil-7 (Soil).

S11 Suppl. Material). Specifically, the concentrations determined in agricultural soils and waters used for irrigation and livestock watering were used for these calculations. The nomenclature, units and Brazil-specific values of all parameters appearing in these equations are reported in Tables S6-S9 (Suppl. Material).

The non-carcinogenic and carcinogenic risks were evaluated by calculating all indices for both effects (HI_{agg} , HI_{tot} , CR_{agg} , and CR_{cum} , see Eqs. S13 & S14 Suppl. Material). Non-carcinogenic effects were assessed considering an exposure period of 25 years, which corresponds to the average lifespan of a mine. The carcinogenic effects were calculated for 78 years, which corresponds to average life expectancy in the world. According to USEPA criteria, the non-carcinogenic effects can be classified as negligible ($HI_{tot} < 0.1$), low ($0.1 \le HI_{tot} < 1.0$), moderate $(1.0 \le HI_{tot} \le 4.0)$, and high $(HI_{tot} \ge 4.0)$. Similarly, if CR_{cum} is below 1×10^{-6} , the carcinogenic effects are negligible; if they are within the range 1×10^{-6} - 1×10^{-4} , it poses an acceptable risk to humans, and values above 1×10^{-4} indicate a high risk of cancer development in humans.

Radiological hazard assessment

Since Caetité is a naturally occurring uranium region, well-known radiological indices were included in this study to quantify the radiological hazards to the local population as part of the health risk assessment. These indices were calculated based on the concentrations of natural radionuclides (238 U, 232 Th, and 40 K) determined in the sampled topsoil.

The important quantities that must be evaluated when considering radiation risks to humans are: (1) the radium equivalent activity (Ra_{ea}) , which assumes that 370 Bq kg⁻¹ of 238 U, 259 Bq kg⁻¹ of 232 Th and 4810 Bq kg⁻¹ of 40 K produce the same gamma-ray dose rates and is therefore expressed mathematically as the weighted sum of the activities of these three radionuclides (Eq. S15, ICRP, 1991); (2) the absorbed dose rate in air (D), which is also calculated as a weighted sum (Eq. S16, ICRP, 1991) and expresses the dose absorbed in the open air due to the gamma radiation emitted by these primordial radionuclides; (3) the effective annual equivalent dose (H_{eff}) Eq. S17), which estimates the total dose that a person receives in a year, and for the public, its value should not exceed 1 mSv y^{-1} (UNSCEAR, 2000).

The indices for the external and internal Radiological Hazard Indices (H_{ext} and H_{int} , Eq. S18) allow estimating the radiation hazards for the internal and external organs of the human body that may be affected by direct contact, ingestion, and/or inhalation of these radionuclides and their progeny, in particular the radon isotopes (²²⁰Rn and ²²²Rn). The values of these indices must be less than one for the radiation hazard to be considered negligible (ICRP, 1991). Likewise, the probability of cancer occurrence from exposure to natural radionuclides was estimated by calculating the excess lifetime cancer risk (*ELCR*, Eq. S19, Belyaeva et al., 2019).

Ecological risk assessment

To evaluate the ecological risk in the studied region, the Potential Ecological Risk Index (PERI) was used. It provides a quick and straightforward quantitative value for the potential ecological risk of a given contamination situation. This index is calculated as the sum of the product between the contamination factor (CF_i) and the toxic-response factor (T_r^i) for the *i*-metals under consideration (see Eq. S20). For the CF calculations, the background values reported for this specific region and other semi-arid areas in northeastern Brazil were chosen (Biondi et al., 2011; Da Silva et al., 2015; Machado, 2008). The toxicresponse factor for the metals of interest assumes the following values: As (10), Cd (30), Co (5), Cr (2), Cu (5), Mn (1), Ni (5), Pb (5), V (2) and Zn (1) (Doležalová et al., 2019). According to the PERI values, the sample locations can be classified into four categories: Low Risk (PERI < 150), Moderate Risk $(150 \le PERI < 300)$, High Risk $(300 \le PERI < 600)$, and Extreme Risk (*PERI* \geq 600).

HERisk code

The risk to the health of the local population from exposure to toxic metal elements and NORMs was carried out with the Human, Ecological and Radiological Risks code (HERisk) developed by our working group. This code was created to facilitate risk calculations for a broad group of chemical pollutants and different age groups spanning all stages of human life. It adopts the basic elements of the methodologies recommended by the main leading institutions on this subject (CETESB, Health Canada, IAEA, ICRP, UNSCEAR, USEPA, and WHO). Unlike other risk codes, HERisk has a space–time approach that allows a detailed risk assessment over time for each location.

Briefly, the code calculates average daily doses and the main hazard indices (mentioned in Sect. "Human health risk assessment" and summarized in the Suppl. Material) for non-carcinogenic and carcinogenic effects from the pollutant concentrations determined in the matrices of interest. In the input files, in addition to the concentrations, the user also provides information on the number of locations, the scenarios considered, the routes of exposure and other exposure parameters. The code has updated databases that contain all the necessary reference values for the risk calculations. In the latest version of the code, the routines "Radio_risk" and "Ecol_risk" have been implemented. The first routine estimates the potential health risk associated with exposure to natural radionuclides using the radiological indices described in Sect. "Radiological hazard assessment" and allows verification that the dose rate values are within the limits. The second routine assesses the level of contamination and potential ecological risks in the different environmental compartments of the study region by calculating several quantitative contamination indices. All quantities calculated by the code and their respective uncertainties for each location, pollutant, and age group are summarized in different output files, specific to each routine.

All features of the first version (HHRisk code) and the second version (HERisk code) are described in detail in Neris et al. (2019) and Neris et al. (2021), so readers interested in more detailed information about the code are invited to consult these works. It is worth noting that this code has been successfully applied in real case studies, demonstrating its usefulness in assessing risks to human health. For example, in the study conducted by Neris et al. (2021) in the nickel mining area of the municipality of Itagibá (Bahia, Brazil), the code made it possible to identify that Co was the chemical specie that posed the greatest risk and that the ingestion of contaminated water was the main route of entry of toxic elements into the human body. In addition, the Hazard Index (HI) values indicated that the entire area around the mine should be classified as a high risk for human health. In the work of Agbasi and Egbueri (2022) and Ayejoto et al. (2022), the code was used to quantify oral and skin health risks from the presence of toxic elements

(Cr, Fe, Mn, Ni, Pb and Zn) in water resources in two regions of Nigeria. The application of the code revealed that a high percentage of the water samples analyzed posed high risks to the local population, especially children, with non-carcinogenic and carcinogenic risks exceeding the legal limits.

Finally, it is important to point out that from the values of the main indices calculated for all the points collected in the study area, including those of the communities, maps were created with ArcGIS software using the Kriging spatial interpolation method.

Results and discussion

Human health risks

Non-carcinogenic effects

At the regional level, the mean values of the aggregate risk index (HI_{agg}) decreased in the following order: Fe (3.48)>Co (1.61)>Al (0.40)>V (0.20) > Mn (0.14) > Zn (0.13) > Ni (0.11) > As = Cd(0.10) > Ba (0.06) > U = Pb (0.03) > Cu (0.01) > Br(0.002) > Cr (0.001). Iron was the metal with the highest HI_{agg} values (range: 0.7-6.5) and accounted for about 45% of the average total risk, followed by Co, which contributed 28% (Fig. 2A). Although the HI_{agg} values of Al, As, Cd, Mn, Ni, V, and Zn did not exceed unity, these metals contributed up to 8% of the total risk. In the case of Ba, Br, Cr, Cu, Pb and U, the contributions were less than 1%. Based on the average values obtained, the health risks resulting from exposure to Fe and Co can be considered as moderate $(1 \le HI_{agg} < 4)$, for Al, As, Cd, Mn, Ni, V, and Zn as low $(0.1 \le HI_{agg} < 1)$ and for Ba, Br, Cr, Cu, Pb, and U as not significant ($HI_{agg} < 0.1$).

As shown in Fig. 2A, the highest HI_{agg} values for As, Co, Cu, Mn, and V and high-risk values for Fe were observed in Caetité, the urban center with the largest commercial and industrial presence in the region. In the entire the region, the highest HI_{agg} value for U (0.10) was found in Lagoa Real, the principal city of the municipality of the same name. Uranium was one of the elements that contributed most to the risk to human health in this locality. Similarly high-risk levels were also observed in the communities near the mines. For example, in the district of Brejinho das Ametistas, a HI_{agg} value for Fe equal to



Fig. 2 Aggregate risks (HI_{agg}) in the main human settlements of the study region. A) Average values of HI_{agg} for each element. B) Contributions (in %) of the routes of exposure to the total aggregate risk (HI_{agg})

6.5 was found. Likewise, in Ceraíma and Guirapá, also close to the iron mine, the values obtained were almost on the borderline of a high risk. In Buracão, close to the uranium mine, the highest HI_{agg} values were found for Al and Cd.

Based on the results obtained, it was possible to identify that the main routes of exposure in the study region are related to soil (Fig. 2B). Dermal contact with soil was the riskiest exposure route, representing approximately 60% of the total risk. This finding is consistent with Wahab et al. (2020), who reported that for soils with high iron content, dermal contact is the most important exposure pathway for adults and children. The largest contributions to risk from this route were observed in the two most populous cities in the region: Caetité (65%) and Lagoa Real (67%), as well as in rural settlements near mines such as Maniaçu (62%), Juazeiro (61%) and Guirapá (61%), where high concentrations of pollutants were detected in the soils. Inhalation of particulate matter was the second route that contributed the most to the total risk (~15%), with low-risk values in all localities except in Caetité, Guirapá, and Brejinho, where the risk from this route was moderate ($1 \le HI_{agg} < 4$).

Although ingestion of soil and (tap) water were considered acute-type exposures, they ranked third and fourth among the riskiest routes with mean HI_{agg} values of 0.65 and 0.28, representing 21% of the total risk. Both routes were more significant in rural areas (Fig. 2B). In the communities of Tamboril, Ceraíma, and Brejinho, accidental ingestion of soil contributes about 20% to the total risk. In the community of Buração, which is close to the uranium mine, water ingestion became the second most important route contributing to the risk. It is imperative to highlight that considering these pathways as acute exposure routes (i.e. a single exposure per year) may lead to an underestimation of the risk values, especially in the case of drinking water. It should not be forgotten that tap water is widely used in households and is the main type of water consumed in communities far from urban centers, given the scarcity and lack of continuous water supply in the region. For this reason, the risks associated with water intake may be higher than those presented in this study. Indeed, Shams et al. (2020) and Envigwe et al. (2021) reported that frequent consumption of groundwater from mining areas can lead to non-carcinogenic risk indices greater than one.

In contrast, the risks associated with dermal contact with water and consumption of food (fruit, vegetables, and milk) were low. At the regional level, they did not exceed 2.5%, except in Buracão, where the risk due to dermal contact with water reached 9.5% (Fig. 2B). It is worth noting that the highest risks associated with food consumption were observed in the cities of Caetité and Lagoa Real, which are the main destinations for food produced in rural areas.



Fig. 3 Contributions (in %) of the routes of exposure to the total aggregate risk (HI_{agg}) for each age group

When analyzing the contribution (in %) to the aggregate risk of each element by age group, it was found that the contributions of Fe and Zn are higher in children and adolescents (up to 18 years) and decrease in adulthood (see Fig. S2 Suppl. Material). On the other hand, the opposite was observed for Al, As, Ba, Cd, Co, Ni, and V: The contributions are higher in adulthood (over 18 years old) and reach a maximum for Al (8.9%), As (1.6%), Ba (1.6%), Cd (2.8%), Co (29%), Ni (4.8%) and V (9.8%) in those over 65 years of age. No significant differences were observed between the different age groups for Mn, Pb, and U. However, the most remarkable contributions of these elements to risk were observed in the age group between 16 and 18 years. The contributions of Br, Cr, and Cu to the total risk remained relatively constant for all age groups and were considered negligible (below 0.5%).

The analysis of exposure routes by age groups revealed that dermal contact with soil and inhalation of particulate matter are more representative in adulthood, where they account for up to 64% and 15% of the total risk, respectively (Fig. 3). In contrast, accidental ingestion of soil plays a more decisive role in children up to 11 years of age, where the risk contributions vary between 20 and 31%. This is an expected result, since hand-to-mouth movement is more frequent in children and ingestion of soil is therefore the main route of exposure to toxic contaminants (Ghosh et al., 2023; Kumari & Bhattacharya, 2023; Zhang et al., 2021). The intake of pollutants via water ingestion increases slightly with age and remains the fourth most dangerous route for people over the age of 11, accounting for over 9%. The risk associated with food consumption is higher for children and adolescents with a share of around 5.5%, ranking as the third route with the highest risk for children up to 6 years of age (Fig. 3). In the case of dermal contact with water, the contributions increase slightly with age and reach the maximum value (6.6%) in elderly (>65 years). Overall, considering all exposure routes, children were found to be more vulnerable to noncarcinogenic health risks than adults, as reported in similar studies (Ghosh et al., 2023; Kumari & Bhattacharya, 2023; Mondal & Singh, 2021).

The total risk index (HI_{tot}), which calculates the risk level considering all pollutants and exposure routes, exhibited a mean value equal to 3.6, indicating that the risk of non-carcinogenic effects in the region, can be considered moderate. Figure 4 shows the distribution of HI_{tot} values in the study region. As can be seen, HI_{tot} values for all human settlements are above unity, with 63% of sites having values that fall into the high-risk category (Fig. 4A). The urban center of Caetité and the district of Brejinho das Ametistas stand out with the highest risk values (HI_{tot} \geq 6.5). In addition, the communities near the two mining areas had risk scores above 4, and others, such as Juazeiro $(HI_{tot}=3.9)$, were practically on the threshold of the high-risk category. The lowest risk value was found in the agricultural community of Tamboril de São Sebastião ($HI_{tot} = 1.7$), which is moderate. In general, the risk values within the moderate category (orange tone) predominate on the map (Fig. 4B). However, it is noticeable that the urban region of Caetité and the settlements in the area of influence of both mines are highlighted in red, indicating high-risk values. These results are consistent with the findings of Envigwe et al., 2021, Ghosh et al. (2023), and Zhang et al., 2021, who also reported HI values that are considered highly alarming for the welfare of the human health of the local population living in the mining districts.

Carcinogenic effects

Unlike the effects discussed in the previous section, carcinogenic effects are stochastic and cumulative,



◄Fig. 4 Total risk index for non-carcinogenic effects (HI_{tot}) for the main human settlements of the study region. A) Specific values for each local. The horizontal blue lines indicate the threshold values for the moderate (M) and severe (S) risk categories. B) Interpolated map of the region showing the calculated HI_{tot} values

i.e. they do not necessarily occur as a result of a single exposure. The likelihood of these effects occurring after several years of exposure can be greatly increased. These effects were assessed by calculating the carcinogenic risk index (CR), which is a measure of the probability that an individual will develop cancer given the characteristics of exposure at the study site.

Of the elements considered carcinogenic (As, Cd, Co, Ni, and Pb), As contributed the most ($\geq 94\%$) to the aggregated cancer risk index (CR_{agg}) (Fig. 5). The highest probabilities of cancer occurrence related to the presence of As were observed in Brejinho das Ametistas ($CR_{agg} = 3.7 \times 10^{-5}$) and in the city of Caetité ($CR_{agg} = 2.1 \times 10^{-5}$). In the localities where arsenic was not quantified, Cd and Pb were the main contributors to cancer risk, especially in the districts of Ceraíma and Guirapá, both located near the iron mine, and in the agricultural community of Tamboril de São Sebastião. As reported in several studies, As, Cd and Pb are the metallic elements with the greatest carcinogenic effects on humans (Kumari & Bhattacharya, 2023; Shams et al., 2020). In the case of Co and Ni, the values were low $(10^{-10} < CR_{agg} < 10^{-7})$, so the cancer risk for both elements was considered negligible. For all age groups, As was the predominant carcinogen, with a slight decrease in adults over 65 years of age.

The soil-related exposure routes again contributed the most to the risks (see Fig. S3 Suppl. Material). However, for carcinogenic effects, the average contribution of dermal contact with the soil decreased to 37% of the total CR_{agg} value, while the inhalation of particulate matter showed the highest degree of representativeness (~60%). Indeed, as reported by Kumari and Bhattacharya (2023) and Mondal and Singh (2021), inhalation is a major route of exposure to trace elements such as Cd, Ni and Pb, which increases the potential risk of cancer. Consumption of food harvested in the region ranked third with contributions between 0.1% and 6.3%, while the average contributions of the other routes did not exceed 0.2% and were therefore considered insignificant. The analysis by age group revealed that food-related routes of exposure make a larger contribution in children under 6 years of age and decrease in adulthood. The contributions to cancer risk associated with both dermal contact with soil and inhalation of particulate matter increase with age and peak after 18 years of age. For the other three routes (soil ingestion, water ingestion, and dermal contact with water), the contributions remain practically constant for all age groups and with very low values ($\sim 0.1\%$).

The average cancer risk in the region was 6.6×10^{-6} , indicating a moderate probability of developing cancer in the local population according to the USEPA classification $(1 \times 10^{-6} \le CR_{cum} < 1 \times 10^{-4})$, Taking advantage of the capabilities of the HERisk code, an estimate of the temporal variation of the cumulative carcinogenic index (CR_{cum}) was made in some localities of interest (Fig. 6).

In Fig. 6A can be observed the temporal variation of cumulative cancer risk in some representative human settlements of the region. As can be seen, CR_{cum} values in Buracão, Maniaçu, and São Timóteo exceed 1×10^{-6} after a few years of exposure (>4 years), indicating a moderate probability of cancer risk associated with exposure to these pollutants. The same trend was observed in other communities close to mining areas (Lagoa da Vargem Grande, Tamboril de São Sebastião, Juazeiro, and Guirapá). In the main urban area of Caetité, the cancer risk is moderate from the first year of exposure ($CR_{cum} \ge 1 \times 10^{-6}$). From the seventh year onwards, the probability of a person developing cancer during their lifetime increases to one in 100 thousand people.

The Brejinho das Ametistas district was the only one with a very high probability of occurrence of cancer (Fig. 6B). After three years of exposure, the cancer risk is higher than 10^{-5} , and after 63 years, the CR_{cum} value exceeds 1×10^{-4} , which means that one in 10,000 people from this population may suffer from this disease. It is believed that the cancer risk at this location is related to the high As and Cd concentrations detected in the area, which show some correlation with the iron mining that is carried out only 15 km from this population center.



Fig. 5 Contributions (in %) of elements to total aggregate carcinogenic risk (CR_{$3\sigma\sigma$}) in each location

It is important to point out that neither Cr nor U were included in the assessment of carcinogenic risks.³ For this reason, the results obtained in this work may be underestimated.

Radiological risks

The radiological effects were estimated from the concentrations of the primordial radionuclides: ²³⁸U, ²³²Th, and ⁴⁰K in the soil (see Table S1, Suppl. Material), which are the main contributors to natural radioactivity. Table 1 summarizes the most important values of the indices determined in the region and shows the corresponding global average values.



Fig. 6 Temporal variation of the cumulative risk of cancer (CR_{cum}) during the average life expectancy (78 years). A) Communities of interest in the study region. B) District of Brejinho das Ametistas. The continuous lines indicate the CR_{cum} categories: Moderate $(1 \times 10^{-6} \le CR_{cum} < 1 \times 10^{-4})$ and High (CR_{cum} $\ge 1 \times 10^{-4}$)

The average radium equivalent activity (Ra_{eq}) in the region was lower than the world average. However, the samples collected in Maniaçu and Buracão communities, both located near the uranium mine, exceeded this value. For absorbed dose rate (D), 85% of the locations were above the world average, with the highest values observed in the settlements around the uranium mine (Buracão, Juazeiro, Maniaçu and São Timóteo) (see Fig. S4 Suppl. Material). The effective doses (H_{eff}) for the outdoor and indoor areas were determined from the absorbed dose rate

³ Since we did not perform a speciation study, the reported Cr concentrations refer to total Cr and not to Cr(VI), which is the carcinogen. In the case of U, the USEPA does not provide SF values for any of the routes considered and, therefore, the carcinogenic risks cannot be calculated by the method followed. To avoid misinterpretation of the results, it was decided not to include these elements and thus not to overestimate the carcinogenic effects.

Table 1 Values of radiological indices determined in the study region

	$\operatorname{Ra}_{\operatorname{eq}}\left(Bq\ kg^{-l}\right)$	$D(nGy h^{-1})$	h^{-l}) $H_{\text{eff}}(mSv y^l)$		ELCR (× 10^{-4})		H _{ext}	H _{int}
			Outdoor	Indoor	Outdoor	Indoor		
Interval	124–594	54–264	0.07-0.32	0.27-1.30	3-13	10–51	0.3–1.6	0.4–2.2
Average \pm SD	300 ± 22	133 ± 10	0.16 ± 0.01	0.65 ± 0.05	6.4 ± 0.6	25 ± 2	0.8 ± 0.1	1.1 ± 0.1
World Average*	370	59	0.07	0.05	2.9	-	-	-

^{*}taken fromBelyaeva et al., 2019



Fig. 7 Interpolated map of the region showing the radiological hazard indices values in the study region

(see Eq. S18). The average values of H_{eff} : outdoor (0.16 mSv y⁻¹) and indoor (0.63 mSv y⁻¹) exceed two and thirteen times the global average values for these quantities, respectively (see Table 1). Nevertheless, only in Maniaçu (1.30±0.16 mSv y⁻¹) and Buracão (1.09±0.08 mSv y⁻¹) were the indoor H_{eff} values slightly above the permissible dose limit for the population (1 mSv y⁻¹), suggesting a possible radiological risk in these communities. The high values of these radiological indices can be attributed to the presence of minerals such as monazite, ilmenite, zircon and barite, which have high concentrations of natural radionuclides (Montalván-Olivares et al., 2021). However, due to the impacts of uranium mining, these indicators can reach values more than 10 times the global averages (Li et al., 2024).

The external (H_{ext}) and internal (H_{int}) radiological hazard indices were determined to characterize the risks associated with direct exposure to these radionuclides. In general, the entire north of the municipality



Fig. 8 Interpolated map of the region showing the calculated potential ecological risk index (PERI) values in the study region

of Caetité, which corresponds to the area of natural uranium deposits, can be considered a radiological risk zone (Fig. 7). In all communities located in this region, direct contact with soils and waters containing these radionuclides can induce various adverse health effects such as skin irritation, swelling, mild burns, cataracts and moderate hair loss (IAEA, 2005). Once U and Th decay products enter the body by inhalation and/or ingestion, they can cause hepatotoxic changes that manifest as acute nausea, headache, vomiting, ulcers, respiratory tract irritation, development of paralytic ileus, and kidney and liver damage (Semenova et al., 2020). In addition, decreased levels of sex hormones, sterility and congenital diseases are also signs of U toxicity (Wang et al., 2020).

Excess lifetime cancer risk (ELCR) was determined to assess the carcinogenic risk that may be associated with exposure to radionuclides in soil and waters. In most locations (>70%), outdoor ELCR values exceeded the world average (2.9×10^{-4}) , being up to 10 times higher in the settlements surrounding the uranium mine and more than twice as high in the urban centers of Caetité and Lagoa Real (Fig. S5 Suppl Material). Based on the average value (6.4×10^{-4}) , it can be estimated that the probability of developing cancer due to radiation exposure in the region is about 64 people per 100,000 inhabitants, which is considered high. As Semenova et al. (2020) report, the body sites targeted by U and Th decay products are the epithelium respiratory tract, bone marrow, gastrointestinal tract, kidneys and skin, so that an increased risk of hematopoietic lesions, leukemia and cancer of the lung, stomach and kidneys has been evidenced in the uranium mining regions.

In the study region, PERI values vary between 293 and 1449 (mean: 551), indicating a severe ecological risk ($300 \le PERI \le 600$). As can be noticed in Fig. 8, the ecological risk values in the areas near both mines were extreme (PERI \geq 600). The highest value of PERI was observed near the iron mine, where high concentrations of As, Cd, Cr and Ni were detected. In addition, extreme values of PERI were also observed in the city of Caetité and the rural settlements of Brejinho das Ametistas, Guirapá, Maniaçu, and Tamboril de São Sebastião. These results show that not only mining, but also agricultural, industrial, and domestic activities severely impact the environment. Similar results agree that the toxic elements introduced into the environment by mining, agriculture and other human activities have disturbed the quality of soil and water, posing potential moderate to severe ecological risks (Doležalová et al., 2019; Haddaway et al., 2019; Ling et al., 2019; Zhang et al., 2021).

Conclusions

This study focused on the assessment of risks to human health and the environment due to exposure to toxic elements and NORM in the mining region of Caetité (NE Brazil), where high levels of soil and water contamination had been reported. Based on the average values of non-carcinogenic risks (3.6) and the estimated probability of developing cancer (7 people per million inhabitants), the region can be classified as moderate risk. However, 63% of the sites had values that fall into the high-risk category, with the city center of Caetité and the communities near the mines being the places with the highest risks to human health. The radiological indices showed average values that are within the range of typical values reported for other uranium-producing regions. However, in Maniacu and other communities near the uranium mine, the effective dose limit for the population (1 mSv y^{-1}) was exceeded, which means that the radiological risk cannot be ignored. In fact, the estimated probability of developing cancer due to radiation exposure is about 64 people per 100,000 inhabitants, which is considered high. Likewise, the analysis of PERI values has shown that anthropogenic activities such as mining, industry and agriculture are responsible for severe ecological impacts, which are extreme in mining areas.

It is known that risk assessment has some limitations that do not allow accurately determining the magnitude and extent of a hazardous pollution event in a given region. Factors such as different individual tolerances to the effects of pollutants, large gaps in information regarding the effects of mixtures of pollutants and the interaction between them that may neutralize or amplify their effects on organs and body functions, need to be further investigated. Furthermore, a stochastic approach based on the Monte Carlo method should be integrated into risk models to have more realistic predictions. Although the risk assessment process does not provide a firm final answer, it has a high relative impact on the scientific and decision-making communities, who can gather efforts to reduce environmental-related hazards to the population. This is why studies like this one, which are the first to address the health and ecological risks in this highly polluted region, are of great importance.

Finally, based on our findings it can be concluded that the local population of the Caetité mining area is subject to high-risk conditions due to exposure to toxic metals and natural radionuclides. These pollutants have the potential to cause adverse human health effects, the probability and severity of which could increase in the coming years. Therefore, immediate risk mitigation measures must be taken to stop this continuous increase in risk levels and to minimize both the adverse health effects and the negative impacts on the already severely degraded environment.

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Data availability The data presented in this study can be available upon request to the corresponding author.

Declarations

Conflict of interest The authors declare no conflict of interest. The funders had no role in the design of the study; in the analysis or interpretation of data; or in the decision to publish the results. We note that none of the figures presented in this article make or imply jurisdictional claims.

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