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Evaluation of arsenic contamination from mining exploration in the eastern Amazon

Avaliação da contaminação por arsênico oriunda da exploração de minero na Amazônia oriental

Evaluación de la contaminación por arsénico de la exploración minera en la Amazonía oriental

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Abstract: Environmental exposure to Arsenic (As) has been increasing over time, either naturally or through anthropic action, this process causes concern in the population as to health risks. Thus, the present study aimed to list the scientific information regarding arsenic contamination in the city of Santana-AP, through a review of the literature in the Capes Periodic Portal, Scielo, Academic Google and PubMed database. Ten studies were identified and analyzed, the only ones being published in this region for the study of As concentration. After analyzing the results, the concentration of As in the water that the community of Elesbão consumes were within the established parameters of <10 μg.L^{-1}. However, the concentration in the soil (50 mg.kg^{-1}) and hair (1.0 mg.kg^{-1}) of this population were above the reference value. This is justified by the level of As total identified in fish and shrimp, causing a possible biomagnification process. Therefore, it is necessary to study all the possible variables of the route of contamination by As in the focus region of the present study, since the divergences of results raise doubts about the level of contamination. Thus, this review will serve as a prerequisite for the evaluation of publications from other regions that have undergone or suffer anthropogenic actions.

Keywords: Manganese. Arsenic. Metaloid. Bioaccumulation.

Resumo: A exposição ambiental ao Arsênio (As) vem aumentando ao longo do tempo, seja naturalmente ou por ação antrópica, este processo causa preocupação na população quanto aos riscos à saúde. Assim, o presente estudo teve como objetivo elencar as informações científicas sobre a contaminação por arsênio na cidade de Santana-AP, por meio de revisão da literatura no Portal de Periódicos da Capes, Scielo, Google Acadêmico e base de dados PubMed. Foram identificados e analisados 10 trabalhos, sendo os únicos publicados na referida região com objeto de estudo a concentração de As. Após análise dos resultados, a concentração de As na água que a comunidade do Elesbão consome, apresentaram-se dentro dos parâmetros estabelecidos <10 µg/L-1. No entanto, a concentração no solo (50 mg.kg-1) e em cabelos (1,0 mg/kg) da referida população estavam acima do valor de referência. Isso é justificado pelo nível de As total identificado em peixes e camarões, ocasionando um possível processo de biomagnificação. Portanto, são necessários estudos que envolvam todas as variáveis possíveis de rota de contaminação pelo As na região foco do presente trabalho, uma vez que, as divergências de resultados geram dúvidas sobre o nível de contaminação. Assim, a presente revisão servirá como pressuposto para avaliação de publicações de outras regiões que sofreram ou sofrem ações antrópicas.

Palavras-chave: Manganês. Arsênio. Metaloide. Bioacumulação.

Resumen: La exposición ambiental al Arsénico (AS) ha ido aumentando con el tiempo, ya sea de forma natural o por acción antrópica, este proceso genera preocupación en la población en cuanto a los riesgos para la salud. Por lo tanto, el presente estudio tuvo como objetivo enumerar las informaciones científicas sobre la contaminación por arsénico en la ciudad de Santana-AP, a través de una revisión de la literatura en el Portal de Periódicos da Capes, Scielo, Google Scholar y base de datos PubMed. Se identificaron y analizaron diez trabajos, siendo los únicos publicados en esa región con la concentración As como objeto de estudio. Después del análisis de los resultados, la concentración de As en el agua que consume la comunidad de Elesbão estuvo dentro de los parámetros establecidos <10 µg/L-1. Sin embargo, la concentración en suelo (50 mg.kg-1) y en cabello (1,0 mg/kg) de la referida población estuvo por encima del valor de referencia. Esto se justifica por el nivel de As total identificado en peces y camarones, provocando un posible proceso de biomagnificación. Por lo tanto, se requieren estudios que involucren todas las variables posibles de la ruta de contaminación por As en la región de enfoque de este trabajo, ya que las diferencias en los resultados generan dudas sobre el nivel de contaminación. Así, esta revisión servirá de base para evaluar publicaciones de otras regiones que han sufrido o están sufriendo acciones antrópicas.

Palabras clave: manganeso. Arsénico. No metal. Bioacumulación.

1 Introducion

Environmental exposure to Arsenic (As) has raised public concern about the risks it can pose to human health, especially after cases of mass contamination were reported in West Bengal, Bangladesh and Mexico (FIGUEIREDO; BORBA; ANGELICA, 2007). In Brazil, from mining activities in the region of Minas Gerais, contamination by As was identified in water samples from underground gold mines and springs in Ouro Preto and Mariana (BORBA; FIGUEIREDO; CAVALCANTI, 2004). Studies carried out in humans have concluded that after long periods of exposure, high levels of this element can be identified in the skin, hair, nails and bones (MANDAL; OGRA; SUZUKI, 2003). As well, they can cause neuropathy, skin lesions, vascular lesions and cancer (MODI *et al.*, 2006).

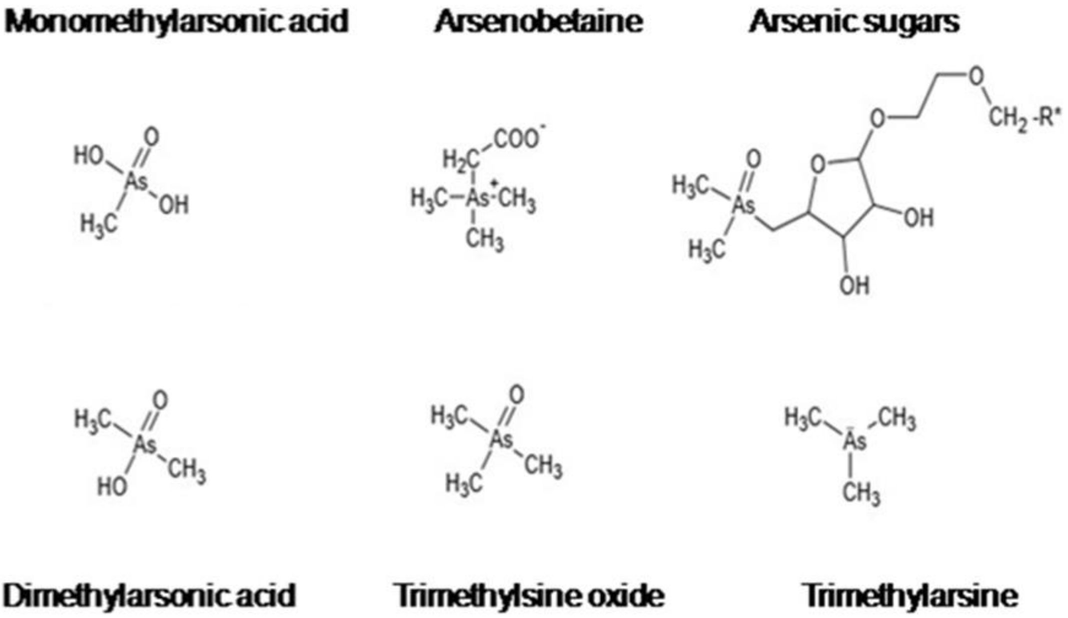
In 2014, a review entitled "arsenic – health: a relationship that requires surveillance" was developed, in which the authors addressed the importance of structuring a health nationally surveillance program, taking into account the relative concentrations of As in foods consumed in Brazil, meantime, this study did not address the contamination of As by anthropic or environmental action (SILVA; BARRIO; MOREIRA, 2014).

However, in several countries, numerous studies are being carried out with the objective of evaluating the relationship between As and the incidence of diseases in humans. However, in Brazil there are few studies, focusing on only four regions: the ferriferous quadrangle region (Minas Gerais) the Ribeira Valley region

(Santa Catarina) (MATSCHULLAT *et al.*, 2000), São Paulo (CUNHA, 2012) and Amapá (LIMA *et al.*, 2007). This is the last focus of the present study, justifying the lack of publications on the subject, besides belonging to the Amazon region. Thus, this work will serve as an alert to the possible negative impacts resulting from the exploitation of natural resources in Amazon, especially after the federal government stimulates the exploitation of this resource (CARDOSO, 2016).

The features of the Amazon River in the study region favored the chemical variations of As, since it is classified as a river class II according to CONAMA resolution 357/05, with a pH range of 6.0 to 9 ,0, in addition, it is characterized as a white water river (muddy) (PEREIRA *et al.*, 2011). As is characterized by being a solid, crystalline, and grayish metal, with chemical valences of 3-,0, 3+ and 5+, varying its chemical form in water according to the pH and environment redox potential (CUNHA, 2012). It occurs naturally in rocks and is obtained as a byproduct of the treatment of copper, lead, cobalt, gold and manganese ores, ie, it is a ubiquitous element, present naturally or by human action, in soil, water, air and food (LIMA *et al.*, 2007; CARDOSO, 2016). In the environment, it appears in four oxidation states: arsenide As(-III), elemental arsenic As (0), arsenite As (III) and arsenate As (V), the organic forms are represented in figure 01.

Figure 01. Chemical Structure of Arsenic Organic Compounds Common



Source: (adapted SHEN *et al.*, 2009; PEREIRA *et al.*, 2011).

The acute and subacute toxicological effects caused by the inorganic As can be relate to various organs including gastrointestinal tract, skin, cardiovascular system and respiratory system. Inorganic As^{3+} is methylated by the hepatocytes in most mammals, as As^{5+} before being methylated is reduced in the blood, so approximately 70% of the As ingested is excreted in the urine, with a half-life of 10 to 30 hours (PEREIRA *et al.*, 2011). The pathologies resulting from As contamination are related to the direct consumption of thiols, in addition to the endogenous formation of reactive oxygen species (ROS), such as superoxide anion, hydrogen peroxide, hydroxyl radical, peroxyl radical and singlet oxygen, in which it affects equilibrium homeostatic, by means of the intracellular consumption of antioxidants, causing harmful effects on proteins, lipids, deoxyribonucleic acid and carbohydrates, thus establishing oxidative stress (SHEN *et al.*, 2009; HARPER; ANTONY; BAYSE, 2014).

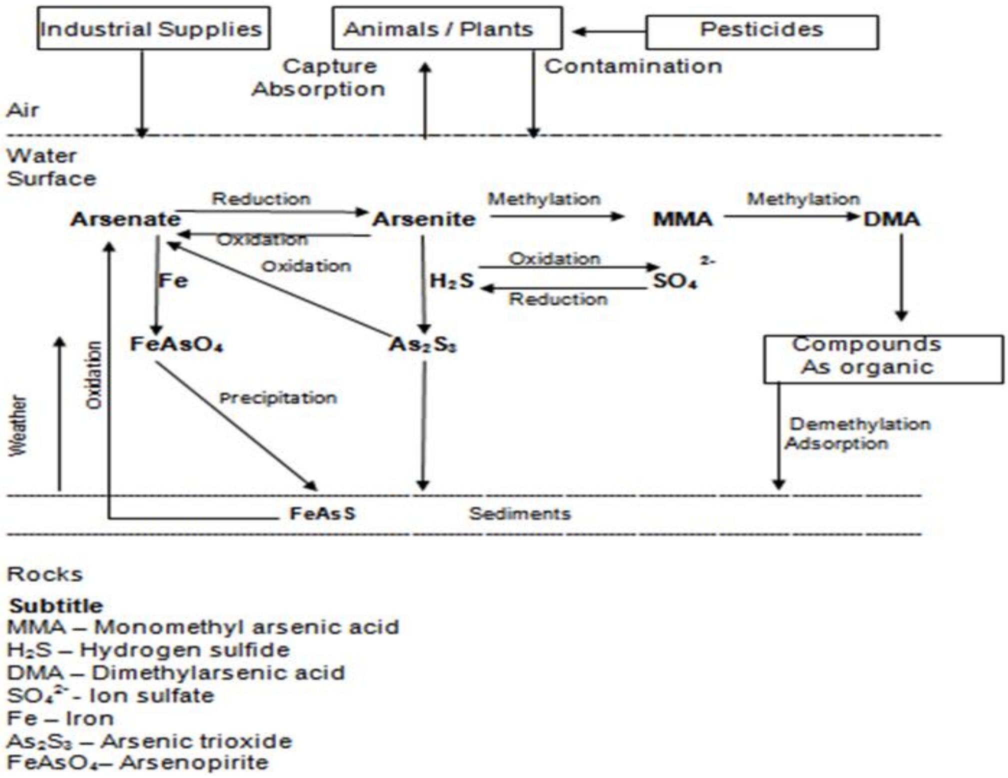
Therefore, it is necessary to study all the possible variables of the route of contamination by As in the focus region of the present study, since the divergences of results raise doubts about the level of contamination. Thus, this review will serve as a prerequisite for the evaluation of publications from other regions that have undergone or suffer anthropogenic actions.

1.1 Arsenic geochemical cycle

Among the anthropogenic sources of As, the mining activity comes first, as well as the metallurgical industry, in addition, have the burning of fossil fuels that is also a source of atmospheric contamination. In agriculture, As was the basis of some pesticides (ESTEVES, 2009).

In this way, the As in the form of AsO_4^{-3} in the presence of iron (Fe) undergoes precipitation in the form of $FeAsO_4$ that can oxidize to AsO_4^{-3}, which can undergo a reduction process, becoming AsO_3^{-3} that in the presence of the H_2S will occur the formation of As_2S_3, being able to oxidize forming again AsO_4^{-3}. However, there is the possibility of methylation of AsO_4^{-3} with the formation of monomethylarsenic acid (MMA), which will result in another methylation transforming into dimethylarsic acid (DMA) that will sediment in rocks (BARRA *et al*., 2000; ROY; SAHA, 2002) (Figure 02).

Figure 02. As geochemical cycle



Source: (adapted ROY; SAHA, 2002; SIMÕES, 2014).

The mechanism of pathogenicity of inorganic As has not been fully elucidated, but studies indicate that it involves the induction of DNA damage due to the formation of reactive oxygen species, such damages can produce genetic and epigenetic effects (SILVA; BARRIO; MOREIRA, 2014), the latter being corroborated by Argos *et al.*, (2015), with evaluation of the association between As exposure (urine and blood sample) and DNA methylation of white blood cells along the epigenome, with a sample of 400 adult participants from rural communities in Bangladesh. The mentioned study observed significant associations between exposure to As and DNA methylation, suggesting that epigenetic modifications may be a pathway to toxicity.

In vitro study with two hamster cell lines demonstrated that As is able to inhibit DNA replication and induce of sister chromatid exchanges, causing chromosomal aberrations (HELLEDAY; NILSSON; JENSSEN, 2000). The same induced human carcinogenicity in prostate epithelial cells (TOKAR; DIWAN; WAALKES, 2010).

In this context, in 50 decade, through the exploitation of manganese in the municipality of Serra do Navio, located at latitude 0º55 north and longitude 52º05 west, on the margin of the Amapari river, situated in the state of Amapá, the company Indústria e Comércio de Minérios SA (ICOMI) was shed manganese for 192km on the railroad linking the municipalities of Serra do Navio and Santana (MONTEIRO; COELHO; SILVA, 2003; SCARPELLI, 2003), during that period of exploration there was environmental contamination by As in the industrial/port area of ICOMI, mainly by the pelletization of As occurred in the municipality of Santana, by heating the manganese at high temperatures, with the objective of beneficiation of the manganese ore, eliminating the low content, consequently the As was released into atmosphere, where it cooled and condensed with rain, so the substances were dragged into the water table (MORIM; PORTO, 2017).

This possible environmental damage became public in 1998 when the useful domain of the ICOMI area for company Amapá Florestal e Celulose (AMCEL) was transferred, in which an environmental audit was carried out by the company JakkoPoyry Engenharia LTDA, which, when evaluating the hydrogeochemical characteristics of the area identified points with anomalous contents for some metals (Manganese, Iron and Arsenic) (CHAGAS, 2010).

Thus, the present study had the objective of evaluating the arsenic contamination from the mining exploration in the Eastern Amazon, through literature review.

2 Material and Methods

This study is a review of As contamination from mining in the Eastern Amazon. In this way, research identified in the Periodicals Capes Portal, Scielo, Academic Google and PudMed database were used, using the descriptors: arsenic Amapá; Santana arsenic; arsenic environment, written in English or Portuguese, published from 2003 through July 2022.

As inclusion method, articles, dissertations, theses and technical reports that had the arsenic issue in the city of Santana-AP as their object of study were included in our research. In total, 67 works were identified, sorted according to the inclusion criteria., and according to the same criteria, 59 who did not meet this criterion and were excluded. In this context, 10 studies were analyzed that addressed the evaluation of As in the municipality of Santana-AP, listed in table 01.

Table 01. Articles and / or dissertations, theses and technical reports used in the bibliographic review

Nº	Title	Reference
1	ICOMI in Amapá: half a century of mineral exploration	(MONTEIRO, 2003)
2	Exposure to mercury and arsenic in Amazonian states: a synthesis of Evandro Chagas Institute studies / FUNASA	(SANTOS *et al*., 2003)
3	Evaluation of total arsenic, traits and bacteriological elements in drinking water in the community of Elesbão, Santana, Amapá state, Brazil	(LIMA *et al*., 2007)
4	Spectrophotometric determination of arsenic in the city of Santana-AP using the modified silver diethyldithiocarbamate (SDDC) method	(PEREIRA *et al*., 2009)
5	Evaluating and classifying contaminated areas based on loss functions using annealing simulations	(QUEIROZ *et al*., 2009)
6	Arsenic in thehairoftheindividuals in Santana-AP-Brazil: significanceofresidencelocation	(PEREIRA*et al*., 2010)
7	Socioenvironmental damages resulting from improper handling of manganese waste and the implications for the life and health of residents of Vila do Elesbão	(FACUNDES, 2011)
8	Report on the monitoring of surface and sub surface waters	(ANGLO AMERICAN, 2012)

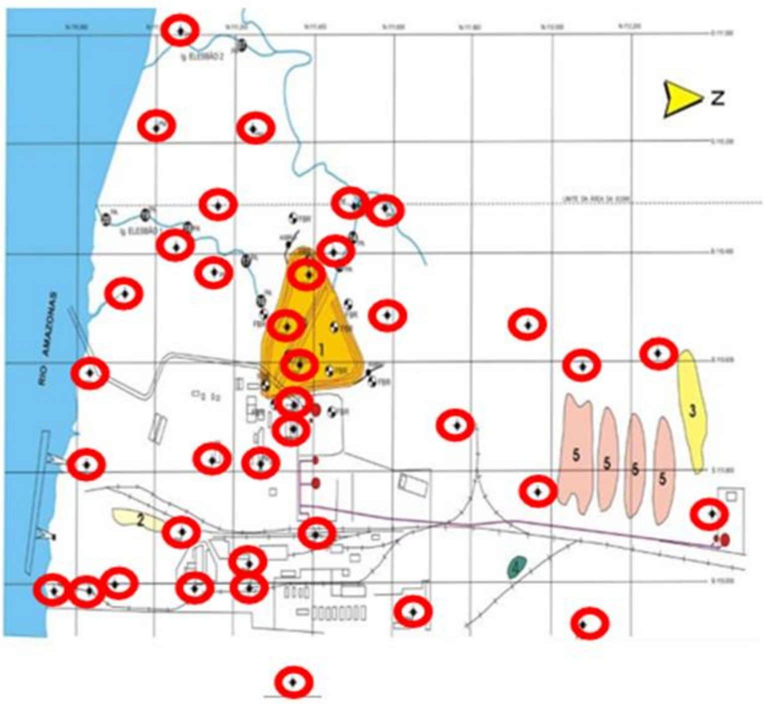
9	Report on the monitoring of surface and sub surface waters	(ANGLO AMERICAN, 2013)
10	Biomarcadores bioquímicos em duas espécies aquáticas amazônicas na avaliação da qualidade de ambientes com histórico de contaminação por arsênio	(DERGAN, 2015)

3 Results and Discussion

3.1 Concentration of Arsenic in water in Santana (AP)

The study conducted by Santos *et al*., (2003) entitled "Exposure to mercury and arsenic in Amazonian states: a synthesis of Evandro Chagas/ FUNASA studies", analyzed 33 monitoring wells located in the port and industrial area of ICOMI (Figure. 03), these collection points were the same as those used by the company's water quality monitoring program, in which the concentrations of As varied from <0.5 to 1970 μg/L, with a mean of 131.4 μg/L.

Figure 03. Map of the collection points and the location of the old industrial and port facilities of ICOMI in Santana



Source: Ampla Engineering (1997, *apud* FACUNDES, 2011)

Figure 3 identifies in the red circles the ICOMI monitoring wells, in the orange color (1) tailings deposit developed by ICOMI, which identified the anomalous values for As concentration

In the mentioned study the authors report that anomalous values, restricted only in the area of waste deposition (1976 μg/L, 1320 μg/L, 760 μg/L) were identified. However, when the anomalous values are not considered, the average concentration does not exceed 8.43 mg/L, according to the authors in question the value established by the legislation for drinking water is 10 mg/L. However, this value does not correspond to that described in Administrative Order No. 1469 of 2000 (referenced in the study) (BRASIL, 2000) and CONAMA No. 003 (2005), both establish the maximum limit of 0.01mg/L or 10 $\mu g.L^{-1}$. Therefore, the mean values identified (131.4 μg/L and 8.43 mg/L) are above the reference value. So, we assume that the concentrations were written incorrectly. This aggravating factor is worrying, since such a study has been referred to in legal decisions (TJ-AP, 2005).

In the monitoring report of surface and groundwater developed by technical representatives of Anglo American (2012; 2013) in the Elesbão community, high levels of As, with values of 9.7 μg/L were found. This result is within the scope of the legislation. However, the collection points were not the same as those of the study conducted by Santos *et al*., (2003), so we cannot infer if the concentration of As in deposition of the tailings decreased. Queiroz *et al*., (2009) examined 42 water samples from the municipality of Santana, 26 from residential wells, 10 from ICOMI monitoring wells, and 6 from surface water and streams in Elesbão. High concentrations of As were found in this study, with values ranging from 0.19 to 5.45 mg/L. The authors attribute such results to the exploration and commercialization of manganese. Dergan (2015) discovered similar results in water samples closer to the source of Santana contamination, the study in question analyzed 20 samples, 10 in the dry season and 10 in the rainy season, the concentration of total As ranged from 1.2 to 1.5 g As/L, such values are above the maximum value allowed for fresh waters with fishing activity (0.14 g As/L) (CONAMA, 2005).

Lima *et al*., (2007) developed a study on the evaluation of total arsenic of trace elements in drinking water in the community of Elesbão, Santana municipality, in three periods: May 2003 with 50 samples, presenting the average concentration of As 5.93 μg/L; November of 2003 with 52 samples, with an average concentration of As 1.95 μg/L and March of 2004 with 52 samples, with an average concentration of As 2.22 μg/L. Thus, the mean levels of As found in this study are within the values considered normal (BRASIL, 2007). In this way, the mentioned study concludes that it is not possible to identify route of exposure from the ingestion of the water.

This study recognizes that the industrial and port area of ICOMI has negative impacts, making the risk of exposure to As imminent, but that company attribute the results of the concentration of As, to regional and environmental factors, such as the possibility of dilution of metals in the Amazon River. However, this study does not question the possibility of adsorption in the sediments, thus, the possible contamination of benthic trophic levels, which may occur the bioaccumulation process or even biomagnification, since these factors were not evaluated.

3.2 Concentration of Arsenic in aquatic species

increases the concentration of a chemical species in living organisms as it accumulates at the highest trophic level. Thus, it is always necessary to evaluate different levels in the food chain.

In the above study, Santos *et al*., (2003) collected fish samples (n=262) from species consumed in the community of Elesbão, and samples of shrimp from different localities of the community. Fish species were divided into two groups: group 1 (carnivorous) and group 2 (non carnivorous), group 1 content ranged from 12.1 to 156 mg/kg, and group 2 of 10.1 and 348.4 mg/kg. The shrimp samples presented levels ranging from 51.0 to 127.5 mg/kg. Even with the predominant presence of organic (less toxicity) in fish and shrimp, these results represent the concentration of As total, so we can not infer the difference in concentration of organic and inorganic forms, the latter with reference values in fish and shrimp of 1.0 mg/kg (ANVISA, 2013; MAPA, 2013).

About 90% of total As found in marine aquatic organisms are in the form of organic As and only 10% in inorganic form. In this way, considering that the speciation pattern of As in marine and freshwater fish and shrimps does not present variation (RAHMAN; HASEGAWA; LIM, 2012) we can infer that the inorganic fractions of the results of Santos et al., the As content ranged from 1.21 to 15.6 mg/kg in carnivorous, and in the non-carnivorous group it varied from 1.01 to 34.84 mg/kg. The As levels in the shrimp samples ranged from 5.1 to 12.75 mg/kg. From the inorganic As fractions we can see that the results are above the established values. Thus, further studies are needed to assess the environmental condition of the area in question.

Pei *et al.*, (2019) describe that the high concentration of As in fish is a result of the process of bioaccumulation, that is, when water passes through the gills, the dissolved substances may have a greater affinity to the organism in question, substances tend to accumulate in the tissues. However, when bioaccumulation occurs indirectly, the phenomenon too will be called biomagnification, defined as the accumulation of a xenobiotic or its derivatives in the different trophic levels, that is, bioaccumulate in the fish, and in the human being through the feeding, occurring the process of biomagnification.

In the literature it is shown that most As-containing compounds, whether organic or inorganic, pentavalent or trivalent, end up interfering with cellular homeostasis, inhibiting the action of enzymes, as well as blocking cellular respiration (PATACA; BORTOLETO; BUENO, 2005). Santos *et al*., (2003), did not differ As organic and inorganic forms.

3.3 Arsenic concentration in blood, human hair and soil

Pereira *et al*., (2010) evaluated the concentration of As in the hair of individuals from the city of Santana, dividing into two areas: urban (23 individuals) (Central, Nova Brasília and Vila Amazonas) and peripheral (98 individuals) (Vila do Elesbão and Vila Daniel). The analysis was carried out using an atomic absorption spectrophotometer, with samples from 121 donors. These groups presented specific socioeconomic characteristics and different habits. After analyzing the results, 52.17% of the individuals in the urban area and 88.87% in the peripheral region had concentrations of As superior at 1.00 $mg.kg^{-1}$, a level considered as a threshold (PEREIRA *et al*., 2009). Although hair is metabolically dead in the epidermis, roots are highly influenced by the health status of living beings, therefore, their analysis is essential in monitoring occupational and environmental exposure to toxic elements (MANDAL; OGRA; SUZUKI, 2003).

In the Elesbão community, the average number of As in the blood of 1,927 people surveyed reached 5.95 mg/L. In the hair samples, the average of 1,986 individuals was 0.56 $mg.kg^{-1}$, and there was no significant difference values found (SANTOS *et al*., 2003).

After evaluating the aforementioned data from Santos *et al*., (2003) and Pereira *et al*., (2010) we can observe a divergence of results, both evaluated the concentration of As in the hair of residents who lived for more than 10 years in Santana, especially residents of the Elesbão neighborhood (a region close to the ICOMI contamination area). Thus, the need for further studies to assess the real concentration of As in the hair samples of the population of the Elesbão neighborhood is evident.

Pereira *et al*., (2009) determined by spectrophotometric analysis the concentration of As in the city of Santana (AP) using the modified silver diethyldithiocarbamate (SDDC) method, 19 samples were evaluated. The results showed that 94.74% (mean of As, 682.96 $mg.kg^{-1}$) of the samples had concentrations (50 $mg.kg^{-1}$) of As above the limit published by Casarini *et al*., (2001) for residential soils evidencing soil contamination by As.

3.4 Socio-environmental aspects of exposure to Arsenic

Facundes (2011) evaluated the socio-environmental damages resulting from the inadequate management of manganese waste and the implications for the life and health of the residents of Vila do Elesbão, who interviewed 33 residents and ex-residents, of whom seven were with visible signs of health problems that resemble some of the adverse effects caused by As-contamination, the most common signs are: severe itching of the body, prominent spots on the skin, back, face, hand and foot injuries.

Such signs, according to those interviewed in the aforementioned study, came from the involuntary exposure to As released with the manganese tailings at Vila do Elesbão by ICOMI. In addition, informants reported headaches, breathing problems, leg pains, stomach problems, kidney and liver problems, eye irritation, body wounds, urinary tract infection, among others. There is the possibility of latency of some days or even 30 years, so that As can produce cutaneous manifestations (GONTIJO; BITTENCOURT, 2005).

In this context, how to explain the low levels of concentration of As in the water consumed by the community and the high levels identified in fish, shrimp, soil and hair besides signs and symptoms of a possible contamination. Bioaccumulation and biomagnification of As in environments are not well known (HAYASE *et al*., 2014). However, Barwick and Maher (2003) evaluating the biotransference of As in marine grasses in Australia have identified evidence of biomagnification. Benthic species are more likely to bioaccumulate, especially in muscle tissue (SLEJKOVEC *et al*., 2014).

The question of the concentration of As of this study area, is an environmental problem still unresolved, since the wastes of As with an average concentration of 1,877.7 μg/g, remain stored in Santana and therefore being leached to the surroundings of the industrial and port area of ICOMI (MONTEIRO, 2003; LIMA *et al*., 2007).

4 Conclusions

The exposure risk to high concentrations of As is evident, which can cause several deleterious effects to the ecosystem, so the present study identified that the values of As concentration in the water for consumption by the Elesbão community were within the established parameters, but in fish and shrimp the total As level was not differentiated (organic/inorganic). In this way, the high level of As in the hair of individuals from Elesbão community is justified, since most residents survive from fishing. and the phenomenon of biomagnification may be occurring between trophic levels, expressed through signs and diseases that are manifesting in some residents over time.

Due to the fact that the review aimed at scientific information on arsenic contamination in the city of Santana-AP, the number of studies evaluated was limited to eight. Thus, further aquatic ecotoxicological studies, clinical and biochemical evaluation of the local population are needed, as well as new studies of As concentration in water and soil. Both All works will aim to clarify and better understand the factors that harm the health of the exposed population.

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