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## JAGUARIBE VALLEY: A THREATENED OASIS IN THE BRAZILIAN SEMI-ARID REGION – SYSTEMATIC REVIEW ON ENVIRONMENTAL CONTAMINATION AND POTENTIAL HAZARD TO THE BASIN AND ITS USERS

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### Abstract

The Jaguaribe River is the largest watercourse in Ceará, one of the semi-arid states of northeastern Brazil, in which the effects of seasonal droughts are most significant. To manage the water scarcity, the Government has tried to perpetuate hydrographic basins, including the Jaguaribe River Hydrographic Basin. Since the 1960s, the elaboration of policies and the construction of reservoirs has encouraged the beginning of irrigated agriculture in the state, bringing large multinational companies and the need for intense agribusiness modernization. Over the past 15 years, scientific papers have revealed the pesticides accumulation, heavy metals, and polycyclic aromatic hydrocarbons in the waters, sediments, and living organisms in the Jaguaribe Hydrographic Basin environment. Besides, the increase in the prevalence of acute poisoning and cancer concerns about severe harmful effects of the traditional agricultural activity in the population of municipalities within the basin. The objective of the

current work was to quantify and qualify the toxicological studies in the Jaguaribe River Basin, expose the affected species, and discuss their importance in a "One Health" context, alerting to neglectation issues involving this vital source in the northeast of Brazil.

**Keywords:** Ceará; environmental contaminants; ecotoxicology

**VALE DO JAGUARIBE: UM OÁSIS EM PERIGO NO SEMIÁRIDO BRASILEIRO – REVISÃO SISTEMÁTICA SOBRE CONTAMINAÇÃO AMBIENTAL E POTENCIAIS DANOS À BACIA E SEUS USUÁRIOS**

### Resumo

O Rio Jaguaribe é o maior curso d'água do Ceará, um dos estados do semiárido no qual os efeitos da seca são mais expressivos. Como solução para a escassez de água, o Governo tem atuado tentando perenizar suas bacias hidrográficas, dentre elas, a Bacia Hidrográfica do Rio Jaguaribe. A partir dos anos 1960, a construção de reservatórios e as políticas de incentivo ao desenvolvimento possibilitaram o início da agricultura irrigada no estado, trazendo grandes empresas multinacionais e a necessidade de intensa modernização para o incremento da produtividade agrícola. Nos últimos 15 anos, a preocupação com os efeitos da intensa atividade agrícola tradicional sobre o ambiente da Bacia Hidrográfica do Jaguaribe vem se mostrando em forma de artigos publicados sobre o acúmulo de agrotóxicos, metais pesados e hidrocarbonetos policíclicos aromáticos nas águas, sedimentos e organismos vivos do rio, além do aumento na prevalência de intoxicações agudas e casos de câncer na população de municípios da Bacia. O objetivo desta revisão foi quantificar e qualificar os estudos toxicológicos realizados na bacia do Rio Jaguaribe, expondo as espécies afetadas e discutindo sua importância num contexto de saúde animal, ambiental e humana, alertando para prováveis negligências com um importante manancial no nordeste brasileiro.

**Palavras-chave:** Ceará; contaminantes ambientais; ecotoxicologia.

## VALLE DE JAGUARIBE: UN OASIS EN PELIGRO EN EL SEMIÁRIDO BRASILEÑO - REVISIÓN SISTEMÁTICA DE CONTAMINACIÓN AMBIENTAL Y POTENCIAL DE DAÑOS A ESTA CUENCA Y A SUS USUÁRIOS.

### Resumen

El río Jaguaribe es el curso de agua más grande de Ceará, uno de los estados de Brasil donde los efectos de la sequía son más fuertes. Como solución a la escasez de agua, el Gobierno ha estado tratando de perpetuar sus cuencas hidrográficas, incluso la Cuenca Hidrográfica del río Jaguaribe. Desde la década de 1960, la construcción de embalses y las políticas de fomento del desarrollo han permitido el inicio de la agricultura de regadío en el estado, trayendo grandes empresas multinacionales y la necesidad de una intensa modernización para incrementar la productividad agrícola. En los últimos 15 años, la preocupación por los efectos de la intensa actividad agrícola tradicional en el medio ambiente de la Cuenca Hidrográfica del Jaguaribe se ha manifestado a través de artículos publicados sobre la acumulación de plaguicidas, metales pesados e hidrocarburos aromáticos policíclicos en las aguas, sedimentos y organismos vivos del río. También ha ocurrido un aumento de la prevalencia de intoxicaciones agudas y casos de cáncer en la población de los municipios de la Cuenca. El objetivo del trabajo actual fue cuantificar y calificar los estudios toxicológicos en la Cuenca del Río Jaguaribe, exponer las especies afectadas y discutir su importancia en un contexto de "Una Salud", alertando sobre temas de negligencia que involucran esta fuente vital en el noreste de Brasil.

**Palabras-clave:** Ceará, contaminantes ambientales, ecotoxicología.

### 1. INTRODUCTION

The world demand for water has grown by 1% per year since 1980, predominantly attributed to population growth and changes in production and consumption profiles. By the year 2050, a 20 to 30% increase in demand is expected, putting billions of people under pressure due to water restrictions (UNESCO, 2019). According to the non-governmental organizations WaterAid (2019) and FAO (2019), Brazil and some South American countries are considered at low risk of drinking water scarcity crisis because they concentrate the largest source globally with more than 28% of renewable sources.

In Brazil, however, there are many conflicts between containing the largest source of drinking water and managing it. Although abundant, water is not evenly distributed among different country regions due to geography, climate, and indiscriminate use. In the early 1990s, new regulations were progressively installed in Brazilian states, culminating in the Water Law (Law No. 9,433/1997), which, based on the Dublin Protocol (1992), decentralized water governance and established the National Water Resources Policy (JOHNSON & KEMPER, 2005; LEGACY BRAZIL, 2010).

Brazilian Semi-Arid occupies the entire center of the northeast and north of the Southeast region, presenting intense insolation, relatively high temperatures, irregular and scarce rains. The rainy season comprises three to four months a year, which is

considered insufficient to supply the local springs. Social-economic indicators are the worst in the country, still linked to outdated agrarian structure, income concentration, and poor land distribution (SILVA et al., 2010). In this context, Ceará is one of the semi-arid states in which the effects of drought are more expressive. Among the measures to prevent drought issues, the Government has been trying to perpetuate its river basins, including the Jaguaribe River basin (BRASIL, 2010).

The Jaguaribe River is located between 4°30' and 7°45' south latitude and 37°30' and 41°00' west longitude. It is born in Tauá, southwest of the state, and flows northeast for 610 km into the Atlantic Ocean, on the east coast of Ceará, between Aracati and Fortim. Its basin occupies almost 76.000 km<sup>2</sup>, representing more than 50% of the total area of Ceará. It is divided into five sub-basins (Upper, Middle, and Lower Jaguaribe; The Salgado River, and the Banabuiú River) and serves 80 municipalities. Caatinga is the biome of the Jaguaribe River. Caatinga reliefs are composed of river plains, hinterland depressions, and residual massifs, with plant formations such as steppes savanna prairies and seasonal tropical forests. The climate is primarily semi-arid, with an average annual temperature of 27°C and a rainfall index of around 740mm per year. Due to its vast extent, Caatinga comprises several areas of climatic and floristic transition, wetter environments, and seasonal herbaceous strata (GATTO, 1999; CEARÁ, 2009). The Jaguaribe River estuary region is a fluvial marine plain, predominantly with mangrove forests, cliffs, and dunes, in a subhumid tropical climate, classified as part of the "northern coast of Brazil" (AB'SABER, 2001).

Over many decades, the water governance in the semi-arid region has been a challenger. As original weather conditions worsen and population density grows, the area is becoming more vulnerable to drought effects. Permanent interventions to mitigate the economic, social, and environmental impacts of droughts include constructing "dams", artificial reservoirs that have changed the landscape, supported life and the productive activities of the semi-arid (BRASIL, 2017). The perpetuation of the Jaguaribe River was due to more than four thousand dams, including Orós, Banabuiú, and Castanhão, which hold about 10 billion m<sup>3</sup> of water, 80% of the entire volume of the basin (JOHNSON & KEMPER, 2004).

Since the 1960s, the construction of reservoirs and incentive policies has enabled the development of irrigated agriculture. Irrigated perimeters increased agricultural productivity and attracted large multinational companies (PEREIRA & CUELLAR, 2015). The "Green Revolution" pressed governments to create specific legislation for agricultural financing to acquire equipment and supplies, including "pesticides" (FREITAS & BOMBARDI, 2019).

Pesticides ('agrotóxicos' in Portuguese) are chemical compounds used to control harmful organisms in agriculture, increasing productivity. They include insecticides, fungicides, herbicides, rodenticides, molluscicides, nematocides, among others. In theory, pesticides should only be lethal to target species; nonetheless, indiscriminate use reaches other species, as humans, vertebrate animals, and the adjacent environment. It justifies the need for pesticides selling strict control (AKTAR et al., 2009). Pesticides, fertilizers, and other supplies may also contain heavy metals, which cause environmental damages as well. Endocrine disruption and its metabolic consequences, neurological

alterations, reproductive disorders, and carcinogenesis are some pesticide effects on human and animal health (GUPTA, 2007c).

Over the past fifteen years, researchers have pointed out contamination in agribusiness areas associated with Jaguaribe's irrigated perimeters and anthropic impact on its river and marine bed. Herbicides, insecticides, and heavy metals have already been detected in surface, groundwaters and sediments, in the organism of fish, and in some invertebrates. Other studies include the epidemiology of human poisoning.

The objective of the current work was to quantify and qualify the toxicological studies in the Jaguaribe River Basin, expose the affected species, and discuss their importance in a "One Health" context, alerting to neglectation issues involving this vital source in the northeast of Brazil.

## 2. METHODOLOGY

We carried out a bibliographic survey in Google Scholar, using the descriptors "Jaguaribe", "contamination", "pesticides", "metals", in September 2019, with an update in January 2020. We organized the results in spreadsheets and classified them according to the type, year of publication, primary and secondary theme. We screened the publications related to the subject, including all those that contained the toxicological information of interest about the Jaguaribe River Basin region. We excluded citations, publications not directly associated with the theme, dissertations or theses that generated articles published in indexed journals (that contained repeated information). We performed descriptive statistics and few meta-analyses of published data, including Chi-square test, correlation, and linear regression, using the software R v.3.6.1.

## 3. RESULTS

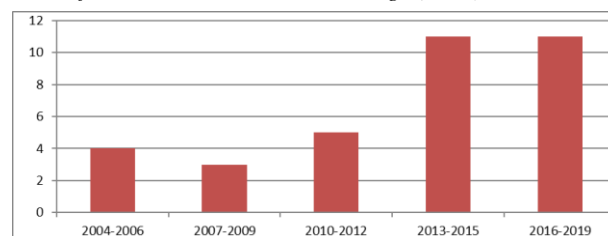
The search identified 183 nonduplicated results, including scientific articles, dissertations or theses, and citations. We recognized 34 titles of those as eligible by content. They were published between 2003 and 2019, including 21 research articles, six master's dissertations, two review articles, one scientific event proceeding, one research project, one book in full, one book chapter, and one technical report. We classified the primary themes as "pesticides", "metals", "PAHs" (polycyclic aromatic hydrocarbons), and "descriptive" (referring to geographical, political, social, or economic description papers). We referred to secondary themes as the type of sample studied or method, including water, sediment, human health, bioindicators, and epidemiological risk analysis. Other secondary topics included reports and dossiers on political and economic management (Table 1).

Table 1 - Selected publications, classified and distributed by themes. Source: Braga (2019).

Theme	Primary				TOTAL	%
	PESTICIDES	METALS	PAHS	DESCRIPTIVE		
Water	3	8	-	-	11	32,3
Sediments	2	1	1	-	4	11,7
Human Health	7	-	-	-	7	20,5
Bioindicators	-	4	-	-	4	11,7
Risk Analysis	3	-	-	-	3	8,8
Other	-	-	-	5	5	14,7
<b>TOTAL</b>	<b>15</b>	<b>13</b>	<b>1</b>	<b>5</b>	<b>34</b>	<b>100,0</b>
<b>%</b>	<b>44,1</b>	<b>38,2</b>	<b>2,9</b>	<b>14,7</b>	<b>100,0</b>	

Graphic 1 shows the absolute frequency of publications on the subject during the evaluated temporal series. In 2013 we highlighted the highest number of publications (n=7), especially studies on the pesticides and metals accumulation in water.

Graphic 1 - Absolute frequency of publications on selected themes, from 2004 to 2019. Source: Braga (2019).



### 3.1. Descriptive publications

Johnsson and Kemper (2004) coordinated a World Bank research project, which yielded a dossier about the Jaguaribe River Basin, its users, managers, public policies, and sustainable use strategies. They reviewed the primary documents and interviews with users. The most concerning issues were water scarcity and recurrent droughts in the semi-arid region; urban growth and supply of other areas (throughout the construction of riverbed diversions); water quality and environmental contamination; periodic flooding; irregular precipitation, and inadequate operation/maintenance of infrastructure. After analyzing the complex information network, they concluded that there were issues in the parity of utilization and the transparency of decision-making. They judged that the 1980s and early 1990s were more favorable to change the outdated management model because the post-dictatorship years had a significant predisposition to re-democratization and decentralization. The authors concluded that, even at a slow pace, the process was in progress, and it would need more integration to achieve better results.

From 2007 to 2010, the Ceará State Legislative Assembly coordinated "The Waters Pact", 80 states' institutions meeting to discuss a consensus to guarantee high-quality water sources for future generations. It generated six e-book reports discussing the status of regional water resources and a strategy for implementing public policies (CEARÁ, 2010). As "the result of great efforts",

the "Regional Notebook of the Middle Jaguaribe Sub-Basin" (CEARÁ, 2009) stood out among the publications, which gathered the essential information to guide the planning of necessary actions in this basin, in a summarized way. The Notebook contains geography and geopolitics; quality, demand and water balance; socioeconomic aspects, and management actions. Maps and graphs illustrate the publication, and tables show the applications of the Waters Pact resolutions. The algorithms demonstrate: an issue to be resolved; what to do/how/who/when; partnerships; formal instruments, and the final action of the supervisory Committee.

Oliveira and Uchôa (2013) compiled and published the actions carried out by the Upper Jaguaribe Sub-Basin Management Committee. From 2006 to 2010, the Committee held 16 ordinary (quarterly) and seven extraordinary meetings. The meetings topics were decentralizing sub-basin water resources use, conflicts between users, water use appraisal, water and natural sources conservation campaigns. "Effects of chemicals" were also present within the educational campaigns. The authors also emphasized equal participation in this Committee. Most members (60%) were civil society, including users from the neighborhood of sub-basin, and 40% were technical representatives of municipal and state public institutions.

Pantaleña and Maia (2014) described the history of occupation of the Jaguaribe Basin region. They related the potential impacts to the different economic cycles occurring since the 17th century, such as soil erosion and riverbanks, decreased flow, waste of surface and deep resources, pollution, and increased salinity. Mineral exploitation, tourism, wind energy production and aquaculture are recent processes, but they have also contributed to deforestation, erosion, and spring pollution.

### 3.2. Evaluation of pesticides environmental contamination

Even before laboratory chemical analyses are made in the Jaguaribe Valley, some authors performed a risk analysis for pesticide contamination using mathematical models. Milhome et al. (2009) gained information about the Jaguaribe-Apodi irrigated perimeter in Lower Jaguaribe, identifying more than 200 small producers and 20 companies dedicated to irrigated fruit farms of cotton, custard apples, bananas, beans, guava, soursop, papaya, mango, corn, soybeans, and grapes. More than 30 types of pesticides used to be employed, especially insecticides (organophosphates, followed by pyrethroids and carbamates). According to the United States Environmental Protection Agency (EPA) risk analysis criteria, 36 to 60% of the evaluated chemical compounds were considered potential contaminants of surface and or groundwater in Upper and Middle Jaguaribe regions, based on their physical-chemical characteristics (water solubility, organic matter adsorption coefficient, liquid gas solubility, half-life in water and soil). The authors created a list of priority compounds for environmental monitoring (MILHOME et al., 2009; GAMA et al., 2013; PINHEIRO et al., 2016).

Several authors evaluated pesticides in surface and groundwater samples in the Jaguaribe-Apodi irrigated perimeter, Quixeré and Limoeiro do Norte municipalities in the Lower Jaguaribe region. Avelino et al. (2013) detected 14.2% of contaminated samples, with the highest frequencies of Imidacloprid and Clotianidine insecticides and Atrazine herbicide.

Milhome et al. (2015) found 100% contamination on the surface and 62% in groundwater samples at global concentrations of 1.1-17.3 µg/L and n.d.-8.9 µg/L, respectively. Propiconazole and Difenconazole fungicides revealed the most frequent and highest concentrations in surface waters, followed by Chlorpyrifos organophosphate insecticide and Atrazine. Sousa et al. (2016) evaluated water samples from 10 reservoirs in Ceará state. They detected Atrazine in 60% out of the samples, in levels 5-6 times higher than the maximum residue limits for pesticides established by the Federal Health Department, including 7.0-8.0 µg/L of Atrazine in samples from Jaguaribe Valley reservoirs. Oliveira et al. (2016) investigated the fate of organochlorinated insecticides applied in the irrigated perimeters of the Jaguaribe Basin. They observed concerning levels of Heptachlor and Lindano in the river and estuarine sediments samples, which were considered particularly alarming in the estuarine domain due to ecological risks to the local microbiota.

We observed no articles on pesticide concentrations in bioindicator tissues (vertebrates or invertebrates) in the Jaguaribe Valley.

### 3.3. Pesticide effects on human health

In 2009, Rigotto et al., from the Community Health Department of the Federal University of Ceará (DSC-UFC), coordinated an epidemiological survey about pesticide exposure with the Lower Jaguaribe population. In the 2000s, Limoeiro do Norte municipality stood out due to an increase in the acute human poisonings recorded by the State Health Department. A large fruit farming company was installed in Limoeiro do Norte, exploiting large-scale pineapple crops for internal and external markets. It used mechanized agriculture, a low-cost workforce, and pesticide applications (RIGOTTO & MATOS, 2009). Alexandre (2009) interviewed 75 farmers in the region, applying for a socioeconomic status questionnaire, professional and family aspects, lifestyle, and pesticide awareness information. The farmers were also referred to clinical and laboratory tests. All the interviewees acknowledged being exposed to pesticides during their routine. Out of them, 94% admitted direct contact more than 8 hours a day; 60% reported have already felt sick at work, and 53% correlated such episodes to pesticide exposure (ALEXANDRE, 2009). Clinical findings pointed out 63% of neurological symptoms (headache, mental confusion, tremors, memory lapse) and 45% eye irritation, which, according to our meta-analysis, were significantly higher than expected ( $\chi^2 = 13,782$ ,  $p=0.001017$ ). Laboratory tests showed that 48% had altered liver enzymes. Although the authors collected workers' clinical data, they executed no correlation tests between clinical findings and pesticide exposure. Based on Alexandre's (2009) descriptive tables, we performed a correlation test and a linear regression to investigate high TGO/TGP ratios over the months of exposure. We observed a temporal linear increasing trend; however, our model was not enough to explain the relationship since several other factors may alter liver enzymes ( $R^2=14.62\%$ ,  $p=0.0872$ ). In a case-control study, Rigotto et al. (2013) detected significantly increased neoplasms prevalence and fetal mortality in the case group (Russas, Limoeiro do Norte, and Quixeré), with a 1.76 higher rate of hospitalizations and 1.38 higher neoplasms mortality than the control group. Diogenes (2017) observed that

the Social Security rural customers in Ceará had five times more risk of receiving early health retirement for neoplasms than the urban customers. Especially Icó, Limoeiro do Norte, and Russas municipalities showed the highest frequencies of retirement, comprising 57% of male workers. The most prevalent neoplasms were skin, followed by hematological ones. Barbosa et al. (2019) evaluated a possible association between childhood cancer and irrigated perimeters population using the State Cancer Registries. The authors observed an increase of 16% in the mortality rates temporal trend due to neoplasms in the Lower Jaguaribe. It was considered the highest mortality rate among the studied non-metropolitan regions.

Freitas and Bombardi (2017) reviewed the bibliography about contamination and poisoning in the Ceará irrigated perimeters, specifically in the Lower Jaguaribe. They elaborated epidemiological maps based on the Diseases Notification System (SINAN) and the Droughts National Department (DNOCS) databases. The results showed that acute pesticide poisoning could reach 26 cases/100,000 inhabitants in the perimeters of Morada Nova, Jaguaruana, Tabuleiro de Russas, and Jaguaribe-Apodi. This rate might be 10-30 times higher than those from urban areas, reaching up to 75 cases/100,000 inhabitants in other state irrigated perimeters. In addition to prohibiting aerial spraying in cultivated areas, State Draft Law No. 18/2015 (approved in December/2018) proposed new standards and limits for the acquisition, use, storage, and transportation of pesticides. Its approval was intended to replace the former Law No. 1478/2009, known as "The Zé Maria Tomé Law." That law referenced an activist who denounced land grabbing and pesticide abuse by agribusiness companies in the Jaguaribe-Apodi perimeter; nonetheless, it was repealed by the Limoeiro do Norte city council in 2010 after the activist has been murdered (FREITAS & BOMBARDI, 2017). Not only do large companies make use of pesticides in their production. In the Russas municipality, 37% of the land reform settlement farmers confirmed using one or more chemical compounds in their small temporary polyculture crops. Out of them, more than 20% reported having already suffered acute poisoning by organophosphate compounds. They mentioned dizziness, headache, malaise, weakness, skin itching, and inappetence. In spite of that, none sought health services, awaiting spontaneous improvement of the symptoms (CASTRO, 2008).

### 3.4. Heavy metal detection in the Jaguaribe River Basin

Lacerda et al. (2004) estimated the annual emissions of the most impactful heavy metals to the environment: Zinc (Zn), Copper (Cu), Lead (Pb), and Cadmium (Cd). They concluded that the anthropic emissions of Cu are the most concerning, assuming that 95% come from agricultural activities, comprising more than 9 tons/year dumped in the Jaguaribe River Basin.

Nobre et al. (2018) investigated metal contamination in surface water and sediments from an alluvial aquifer in the Itaiçaba municipality in Lower Jaguaribe. They found Aluminium (Al) (0.7-1.8 mg/L) and Pb (11-68 µg/L) in 10% samples, higher than the maximum limits of the Brazilian National Environmental Council (CONAMA). The sources of contamination were probably from shrimp farming and poor basic sanitation (by the time, most inhabitants still used the septic tanks).

The Institute of Marine Sciences (Labomar-UFC) research group investigated heavy metal contamination in the final riverbed and estuary zones of the Jaguaribe River, including the impacts of erosion, effluent discharge, and shrimp farming. Lopes (2006) observed Cu up to 8mg/g and Zn up to 68.5mg/g in shrimp ponds' water and sediments, but no significant difference over the old or recent ponds' time of use. Excessive heavy metal concentration in ponds could affect the shrimps' metabolism, making them potential sources of food contamination. Soares (2011) detected dissolved (~5.4ng/L), particulate (~0.6ng/L), and total (~11.9ng/L) Mercury (Hg) levels in the Jaguaribe River estuary fluvio-marine gradient. These findings suggested that Hg was coming from the continent, while a large amount was being retained in the estuary zone during the tidal range. According to CONAMA maximum limits, Hg observed levels were not enough to consider a high impact over the Jaguaribe River estuary. Notwithstanding, their bioavailable fraction was sufficient to affect the trophic chain, causing chronic effects for local aquatic biota. Lacerda et al. (2011; 2013) expected a more significant particulate Hg (1.8-12.6 mg/s) load from the river to the estuary during the rainy season. Along with contaminated aquaculture food, fertilizers and lime would increase total Hg levels in the shrimp ponds, reaching 230mg Hg Total/hectare/cycle. Despite concerns, Lacerda et al. (2017) estimated that only 0.15% of the annual Hg load in the Jaguaribe River estuary would correspond to shrimp farming residues.

Da-Silva-Dias et al. (2013) compared particulate Cu, Zn, Iron (Fe), and Al levels in the Jaguaribe River estuary during rainy and dry seasons. They found out that Cu, Fe, and Al loads were original from erosion sources, therefore lithogenic, and positively correlated with rainfall. In contrast, Zn showed higher levels in the dry season, positively correlated with anthropogenic sources.

### 3.5. Bioindicators toxicity evaluation

Once in 2000, Jaguaribe River estuary fishermen reported increased mortality in the macrofauna, Valentim Neto (2004) was concerned about the swamp ghost crab (*Ucidis cordatus*). Aware of the toxic residues dumped in the estuary, the author performed a bioassay, exposing a group of live specimens to the shrimp farming effluents for 30 days. After this, he evaluated water quality and sample histology. His findings detected no significant morphologic damages, so the experiment failed to evaluate an impact over that sample. He also failed to suggest causation about the prior reported mortality increases, as long as it could have been just an isolated event, quickly dissipated for the tides and rainfall.

Peres (2012) investigated how invertebrate bioindicators influenced metal accumulation in the sediments of three important estuary rivers in Ceará. In the Jaguaribe River estuary, he observed that the colonization for *Mytella* spp. bivalve mollusks supported Fe, Al, and Manganese (Mn) higher accumulation in sediments up to 15 cm depth. Conversely, the colonization for *Uca* spp. reduced Fe and Al accumulation in sediments up to 10 cm depth, probably due to the crabs' bioturbation (behavior of oxidizing sediments while feeding). On the other hand, bivalves' and crabs' colonization influenced Cu, Zn, and Pb accumulation equally. The author estimated the origins of metals by quantifying not colonized areas and using

correlation matrices, concluding that Fe, Cu, Zn, and Pb had the same origin (highly correlated to each other), different from Al and Mn.

Costa e Lacerda (2014) examined Hg levels in consumed fish meat along the several small villages from Lower Jaguaribe. They detected 0.1 – 107.5 ng/g Hg in muscle samples of 13 fish species, still considered below the maximum residue limits established by the National Health Surveillance Agency (ANVISA). The highest Hg levels were detected in the carnivore fish muscles, which demonstrated trophic biomagnification phenom. After interviewing the consumers, the authors estimated the local Hg human exposure for fish consumption. They concluded that the observed fish meat Hg levels offered no human poisoning risk at that time (even for the increased local fish consumption). The same authors compared Hg levels in fish muscle of the native wildlife to exotic cultivated species from the Castanhão Pond in the Middle Jaguaribe. The levels in the native wildlife were found significantly higher than in the cultivated ones but still lower than the ANVISA maximum residual limits. Moura et al. (2018) prospectively evaluated Hg accumulation in invertebrates and fishes with different diets, finding it higher in carnivores crustaceans but not significantly different between the diets. Again, Hg levels were found to be lower than the maximum official limits; however, the authors believed that the hydrochemical and hydrodynamical aspects of the present ecosystem could have contributed to the Hg bioavailability, what along with the increased regional fish consumption, consequently, could cause chronic damages in fauna and human population.

### 3.6. Aromatic hydrocarbons detection

Andrade et al. (2019) evaluated 18 polycyclic aromatic hydrocarbons (PAHs) along the Lower Jaguaribe. They detected PAHs at 0.6 – 3752.0 ng/g, unevenly distributed among studied areas, showing higher concentrations at the estuary. Due to these findings, the Jaguaribe River estuary was considered as moderate to highly contaminated, according to EPA (1000 – 5000 ng/g). The authors found predominantly high molecular weight compounds (four to more aromatic rings), including pyrene and fluoranthene. The benzopyrene, another high weight compound known as carcinogenic in animal models, was detected in lower levels in the estuary. Domestic biomass and brickyard coal combustions contributed to the riverbed, while possibly boat oil spills mainly contributed to the estuary PAH accumulation.

Despite the considerable efforts to measure environmental impacts and water quality in the Jaguaribe River Basin, no studies have yet dedicated to examining the effects of contamination on its terrestrial vertebrates during this temporal series.

## 4. DISCUSSION

The environmental contamination by pesticides and PAHs (organic chemicals) and heavy metals (inorganic elements) cause a complex of adverse effects. Therefore, it is necessary to approach it in an interdisciplinary way within the "One Health" concept (human-animal-environmental health). Agriculture residues are dumped in soil and waters, reaching microbiota and macrofauna. Residues may travel inside the trophic chain organisms, affecting plants, invertebrates, vertebrate animals,

until they affect humans throughout the feeding and topical absorption. Given the above, it is not possible to approach only one of the three vertices of health.

Chemical compounds called pesticides began to be produced in the 1950s in India, expanded to all continents in the following decades, contributing to increasing productivity in agriculture. Among their benefits were reduced losses, disease vector control, and increased food quality. Nevertheless, even the most modern and specific pesticides can be dangerously toxic to human and animal health, contaminating food products, air, vegetation, soil, surface and groundwaters. Be dangerous to the ecosystem means affecting the trophic chain's basal components, as plants and invertebrates accumulate and produce harmful effects to the whole natural system (AKTAR et al., 2009).

Pesticides detected in the highest levels in the Jaguaribe River Valley may affect humans and vertebrate animals, damaging liver, reproductive and nervous systems primarily. Triazole fungicides may cause hepatic acute toxicity and carcinogenesis. Besides, Ilyushina et al. (2019) observed bone marrow inhibition of erythropoiesis in mice exposed to triazoles fungicides.

As endocrine disruptors, Atrazine and Lindane may cause male feminization (by aromatase expression) and testis toxicity. Chlorpyrifos, among organophosphates, interferes in nerve conductivity, acetylcholinesterase activity, and central nervous system oxidative stress, resulting in neurobehavioral changes. Imidacloprid and Clotianidine can also alter cholinergic reactions but are considered highly selective and safe (GUPTA, 2007c). Besides those previously listed, other health effects were recently discovered on humans and animals. Lindane and its isomers ( $\alpha$ ,  $\beta$ ,  $\delta$ -hexachlorobenzene) were detected in the serum of 96% of patients with Parkinson's disease, being  $\beta$ -hexachlorobenzene the most significantly correlated compound (RICHARDSON et al., 2009). Navaratne et al. (2014) observed the acute death of exposed tadpoles to Chlorpyrifos at 0.1 ppm. Other authors reported that Atrazine causes myocardium degeneration in quails and failed transgenerational development in rats (LIN et al., 2016; MCBIRNEY et al., 2017). Sun et al. (2016) noticed imidacloprid to induce lipogenesis and insulin resistance in mice, while other researchers checked sublethal chronic damage and ecological dysfunctioning in pollinated bees, among other invertebrate communities (VAN DIJK et al., 2013; DIVELY et al., 2015; SUN et al., 2016; MILES et al., 2017).

Occupational exposure to pesticides is widely associated with increased risk for hematological neoplasms (non-Hodgkin lymphoma, leukemias, and multiple myeloma). In addition, serum levels of non-persistent pesticides have significantly correlated to anomalous counts of lymphoid and myeloid cells in rural workers in southern Brazil (MERHI et al., 2007; PICCOLI et al., 2019). Even domestic insecticide or herbicide exposure, used in lower levels and frequency, may increase the risk by 1.23-1.47 times for children's neoplasms, including neuroblastomas, nephroblastomas, and leukemias (CHEN et al., 2015).

Heavy metal contamination in the Jaguaribe riverbed and estuary is concerning. In general, heavy metal produces sulfur (S), nitrogen (N), and oxygen (O) complexes, inactivates enzymatic systems, changes protein structures, and affects cell metabolism, leading to cell death. The central and peripheral nervous system, gastrointestinal, cardiovascular, hematopoietic, and renal are the most affected organic systems (SHARMA et al., 2014).

Cu was one of the most concerning detected metals. Toxic doses vary among different animal species. Acute poisoning, liver and kidney failure (generally lethal), frequently manifests up to 48 hours (THOMPSON, 2007a). Bovine and ovine may have chronic poisoning whenever they feed in fields contaminated with Bordeaux mixtures (fungicide CuSO<sub>4</sub> solution), copper-based feed supplements, poultry, or pig manure (O'DELL, 1997). Cu and Zn high levels are present in shrimp farming in the Jaguaribe River estuary. Once metals accumulate in tissues and deplete through the exoskeleton, those shrimps could be contaminated food sources. Human Zn poisoning is rare, probably associated with supplement accidental overdose; very serious, anyhow, promoting oxidative stress in different tissues, especially in the nervous system (PLUM et al., 2010). Although its levels were considered lower than the maximum legal limits, Cd must be monitored, for it caused carcinogenesis (LEE & WHITE, 1980), osteoporosis and compact bone vascularization failure (DURANOVA et al., 2014), dystrophic enamel (ŚWIETLICKA et al., 2019) and prostate inflammation (SANTANA et al., 2016) even in lower but chronic exposure in rats. In humans, occupational exposure may cause nephropathy, osteoporosis with osteomalacia (HOOSE, 2007).

As stated in Food and Drug American Agency (FDA), Hg levels in fish meat for consumption must not be over 0.1 ppm (GUPTA, 2007b), i.e., 1.0mg/kg for carnivore and 0.5 mg/kg for non-carnivore fishes established by ANVISA (BRASIL, 2013). Amorim et al. (2000) found a positive correlation between minimal levels (50µg/g) of methylmercury contamination (Hg inorganic metabolite which accumulates in fish meat) and lymphocytic cytogenetic damage in the Amazon riverside population. Even in considered low levels, methylmercury and phenylmercury chronic exposure may cause kidney, stomach, and bowel damages; arterial tension and heartbeat changing; failed development and abortion; and spermatogenic disturbance in laboratory animals. In addition to the prior listed alterations, livestock animals may manifest evident neurological signs (GUPTA, 2007b; BRASIL, 2013; GENCHI et al., 2017).

Al and Pb were detected in high levels in a groundwater well in the Lower Jaguaribe. Both metals are related to general oxidative stress and hypochromic anemia, but the organic damages depend on which tissue they accumulate. Pb causes blood-brain barrier disruption, neurotoxicity, and neurotransmitters alteration in humans and animals; in contrast, Al may cause neurodegenerative diseases (GUPTA, 2007a).

Among the PAHs, Pyrene and fluoranthene were detected in high levels in the Lower Jaguaribe. Aggravated by UV light, PAHs induce acute metabolic toxicity in wild birds and aquatic organisms. Chronic toxicity may lead to carcinogenesis, immunity diseases, reproductive and developmental problems (ABDEL-SHAFY & MANSOUR, 2016). In humans, acute exposure to PAHs high levels may cause eye irritation, queasiness, vomiting, diarrhea, mental confusion (UNWIN et al., 2006). Chronic exposure results in cataracts, liver and kidney disease, asthma, respiratory and digestive carcinogenesis (OLSSON et al., 2010; DIGGS et al., 2011). In lower levels than the prior listed, benzopyrene was the first described carcinogenic PAH. If inhaled, benzopyrene caused lung cancer; if drunk, it caused stomach cancer; if topical, it caused skin cancer in laboratory animals (ABDEL-SHAFY & MANSOUR, 2016).

## 5. FINAL CONSIDERATIONS

Throughout this bibliographic review, we realized the significance of the Jaguaribe River not only to Ceará state but to Brazil, whether for domestic or industrial supply, whether for irrigation, transportation, or ecosystem conservation. In summary, critical to regional progress. We discussed the detected contaminants in the light of human, animal, and environmental health. Although most studies conclude that "the contamination levels are still acceptable (compared to the official maximum limits and other rivers)", pesticides, metals, and hydrocarbons can cause long-term irreversible damage, even in low concentrations or doses.

Monitoring is essential, controlling is unavoidable, and penalties must be legitimately applied. Environmental education and population awareness must be continuous. Abuse of chemical compounds must be fought, not only by the authorities as by everyone.

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