Mapping of Nitrate Contamination (NO₃⁻) in an Urban Area on the Brazil/Bolivia Border

Carlos Alberto Paraguassú-Chaves¹, Charlles da Silva Barata², Eliomar Pereira da Silva Filho³, Fabrício Moraes de Almeida⁴, Lenita Rodrigues Moreira Dantas⁵, Fábio Robson Casara Cavalcante⁶, Ana Maria Morais da Fonseca Cavalcante⁷, Izan Fabrício Neves Calderaro⁸, Leonardo Severo da Luz Neto⁹, Delson Fernando Barcellos Xavier ¹⁰ and Marco Antônio Domingues Teixeira

¹PhD in Health Sciences - University of Brasília - UnB, Brazil. PhD in Science - University of Havana (Cuba); Post-Doctor in Health Sciences - UnB and Degli Studi D'Aquila University - IT. Professor at the Federal University of Rondônia - Brazil. E-mail: carlos.paraguassu@gmail.com

²Master in Geography - Federal University of Rondônia. Researcher at the Higher Institute of Health Sciences and Environment of the Amazon, Brazil.

³PhD in Geosciences and Environment at Universidade Estadual Paulista - UNESP. Professor at the Federal University of Rondônia - Brazil.

⁴PhD in Physics (UFC), with post-doctorate in Scientific Regional Development (DCR/CNPq). Researcher of the Doctoral and Master Program in Regional Development and Environment (PGDRA/UNIR). Leader of line 2 - Technological and Systemic Development, and Researcher of GEITEC — Federal University of Rondonia, Brazil. E-mail: dr.fabriciomoraes001@gmail.com

⁵Bachelor and Specialist in Geography. Bachelor in Law. Researcher at the Higher Institute of Health Sciences and Environment of the Amazon - AICSA. E-mail: lenita_dantas@gmail.com

⁶PhD and researcher linked to the graduate program in administration and the academic Department of Social and Environmental Sciences (DACS) - Federal University of Rondônia, Brazil. E-mail: frcasara@hotmail.com

⁷Mestre em Agronomia pela Universidade Federal de Pernambuco (UFPE). Researcher of the Institute of Health Sciences and the Amazon environment - AICSA.

⁸Master in Regional Development and Environment (PGDRA/UFRO). TI Analyst of Federal University of Rondônia Foundation, Porto Velho, Rondônia (Brazil). Member of GEITEC/UNIR/ CNPq. Brazil. E-mail: izancalderaro@gmail.com

⁹Master in Education - Autonomous University of Barcelona, Spain. Master in Psychology - University of São Paulo. Master in Religious Studies - Faculty of Theological Education Logos of São Paulo. Bachelor of Nusring. Professional Physical Education. Bachelor in Theology. Professor at the Federal University of Rondonia, Brazil - Department of Collective Health, Researcher at the GEITEC and GEISC of the Federal University of Rondonia, Brazil. Email: lluz@unir.br

¹⁰Doctor of Law - University of the State of Rio de Janeiro, UERJ, Brazil. Professor of the law department of the Federal University of Rondônia Foundation.

¹¹PhD in Sustainable Development of the Humid Tropics - Federal University of Pará, Brazil. Professor at the Federal University of Rondônia - Brazil.

Abstract: The objective of this research was to map and evaluate nitrate contamination in an urban area on the Brazil / Bolivia border. The evaluation of the groundwater table occurred in the water of cacimbas wells or Amazonian wells with an average depth of 10 meters in the city of Nova Mamoré, in the State of Rondônia, located in the southern region of Western Amazonia bordering the Republic of Bolivia. METHODS: Eighty (80) water samples were collected in the city’s urban center, defined and evaluated for nitrate (NO₃⁻) in a proportion above or below the values defined by Ministry of Health Ordinance No. 2.914/2011, which governs standards of water standardization and potability in Brazil. From the data obtained, maps with georeferenced information were made to locate the areas with the highest and lowest contamination potential. The use of geostatistics assisted in the accomplishment of the spatialization of the samples and in the analysis of the spatial patterns allowing a higher quality of the generated data. Results: Sixty-two and a half percent
(62.5%) of the samples presented nitrate levels above 10 mg/L, making it unfit for human consumption.

Conclusions: This study indicated that the source of the contamination originates from the result of anthropogenic activities, evidenced by the inefficiency of the sanitary sewage system, one of the basic sanitation axes in which, specifically, it addresses the collection, treatment and adequate disposal of sanitary sewage and other hygiene practices. It is necessary, the urgent insertion of public policies in the sector of basic sanitation in Nova Mamoré and in the Amazonian cities.

Keywords— Mapping. Wells. Contamination. Nitrate (NO3-). Indicative Kriging.

I. INTRODUCTION

Brazil is considered a country where water resources are abundant. Approximately 12% of the planet’s fresh water is concentrated in the Brazilian territory, more specifically in the northern region of Brazil, where about 7% of its population lives. It is the region with the lowest demographic density in the country, 0.4 inhab / km², which would make it a region with excellent quality of life indicators for its inhabitants in terms of water resources; low supply in the provision of basic sanitation services, places it in the ranking of the worst Brazilian region in this indicator.

As mentioned by several authors in urban areas and in rural areas, water quality is questionable [1]; [2]; [3]; [4]. The consumption of water in the northern region of Brazil occurs without major concerns and this fact shows both the lack of efficacy in the treatment of water by the public authorities and the lack of knowledge of the population that consumes it without further questioning.

The concern with the quality of the water must be observed from the moment it is destined, be it for the productive system, watering of animals or directly for the human consumption. The prevailing uses are established in regulations, where different standards and parameters were established according to the activity to be performed, but it is in the direct use associated to human consumption that the present study evaluated the consequences of the low water quality, considering for analysis the small urban nuclei of the North region of Brazil, specifically in the State of Rondônia, where the capture of water in most of its cities is carried out through cacimbas wells, that is, with the use of groundwater.

The scientific awakening with the quality of water for human consumption has become a growing interest in the subjects related to the possible negative consequences on the life of the human being. Queiroz [5] states that abundant and quality water is essential to public health, preventing diseases such as diarrhea and intestinal infections.

Paraguassu-Chaves et al [6] points out several health problems associated with non-standard levels of nitrate and nitrite, which can promote diseases such as childhood cyanosis and possibly different types of cancer.

According to Ordinance no. 2914/2011 of the Brazilian Ministry of Health [7] and the World Health Organization, which provides for the procedures for controlling and monitoring water quality for human consumption and its drinking water standard, the concentration greater than 10 mg/L of Nitrogen (N) in the form of Nitrate (NO3-), is unfit for human consumption.

Considering the question of the population health and socioenvironmental risk involved, the present work mapped and evaluated the concentration of contamination in the free aquifer of the city of Nova Mamoré, in the State of Rondônia, located in the southern region of Western Amazonia.

According to Tomaz [8], lack of water is one of the serious global problems that can affect the survival of humans. Disorganized use, wastefulness and growth in demand are contributing factors to intensifying the scarcity of potable water on the planet.

Brazil has one of the largest hydrographic basins in the world. However, the severe shortage of drinking water in several regions has been caused by the imbalance between the demographic, industrial and agricultural distribution in relation to water availability.

The underground water potential in the Amazon region has an important contribution to human supply according to Campos [9]. Its use is made in an unplanned way and without adequate knowledge of its potentialities and qualities.

Studies on contaminating agents in groundwater have been occurring gradually in the North, [10]; [11]. The city of Nova Mamoré in the state of Rondônia, located south of the Amazon on the border with Bolivia, is an example of an isolated area with contamination problems in its groundwater.

For Alaburda & Nishihara [12], virtually all human activities pose some risk of environmental pollution and often become sources of groundwater contamination. Among the substances that may constitute a risk to human health are the nitrogen compounds in their different states of oxidation: ammoniacal and albuminous nitrogen, nitrite and nitrate.

The use of groundwater in Brazil has increased as a result of demographic concentration and economic expansion due to its qualitative and quantitative advantages over surface waters. The logic of this demand has directly influenced its quality and, consequently, the health of the populations, since these sources are degraded, according to studies done by Hirata and Cagnon [13].
According to IBGE [14] in Brazil, the underground aquifer supplies approximately 20% of the total households and of these, 68.78% are located in the rural area, covering 12% of the entire population of the country.

In the State of Rondônia, groundwater represents an important resource in human supply. According to the Water and Sewage Company of Rondônia - CAERD [15], of the total water distributed by the company, about 35% originates from the underground spring.

According to Campos [9], groundwater, because it is a low-cost alternative, is accessible to all, especially the low-income population, both in daily supplementation and in the total replacement of water provided by the public service.

However, Melo Junior et al [16] point out that in Rondônia, another aspect that is characterized by the inefficiency of basic sanitation services is the lack of sanitary sewage in the urban area that perpetuates to the present day.

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For Lima [17], water supply, be it public or private, may be compromised by the lack of sanitary sewage in urban areas, where different substances, whether natural or anthropogenic, are present.

According to Finotti et al. [18], Brazilian groundwater reserves are already seriously compromised. Through studies carried out in all parts of Brazil, we can see the significant degree of contamination found in groundwater, whether urban, industrial or agricultural.

Nitrate is found naturally in groundwater but its presence at high concentrations is a result of human activities, mainly due to the use of in situ sanitation systems the nitrogenous substances of the organic waste are oxidized by chemical and biological reactions and the result is presence of nitrate in the soil and consequently to groundwater [19].

Freezer and Cherry [19] state that nitrate is extremely soluble in water and can move easily, contaminating the aquifer at long distance due to its persistence and mobility. The nitrate present in the soil or directly in the water has very easy to contaminate the groundwater. According to Brazil's regulatory standards for the public or individual water supply system, water containing concentrations greater than 10 mg/L of nitrogen (N) as nitrate (NO3-) is classified as unfit for consumption (Ministry of Health of Brazil, Ordinance 2.914/2011).

In the case of the city of Nova Mamoré, the basic sanitation, specifically, water supply and sanitary sewage collection, of the municipality is administered by CAERD. The number of households with access to the general water network is still very low, only 16.88% of the households, while 83.67% are supplied by wells or spring on the property.

This fact, in itself, demonstrated the need for studies in relation to physical and chemical data, in order to evaluate water quality throughout the urban perimeter of the municipality of Nova Mamoré, aiming at the diagnosis of parameters of water contamination.
In this aspect, the present study had as general objective to map the plume of contamination of groundwater (free aquifer), potentially impacted by nitrate (NO₃⁻), in the municipal headquarters of Nova Mamoré - Rondônia, under the prism of geostatistics using the Indicative Kriging. And as specific objectives, (a) Register cacimbas wels in the study area to carry out the chemical analysis of nitrate (NO₃⁻); (b) Identify the areas of isoprobability of nitrate contamination (NO₃⁻) with cut-off level <3 mg/L; (c) Identify areas of isoprobability of nitrate contamination (NO₃⁻) with cut-off content> 10 mg/L; and (d) map nitrate (NO₃⁻) isoconcentration areas in the Free Aquifer of the study area.

Through a simple method, geostatistics and kriging, it was possible to determine the anomalous areas and the plume of contamination of the water table in the city of Nova Mamoré and demonstrated through maps with georeferenced information, the areas of greatest and least contamination potential in order to have a spatial model of the problem in question.

II. METHODOLOGY

2.1. Methodological procedures

Some hypotheses were formulated to explain the origin of nitrate in groundwater in the city of Nova Mamoré. 80 shallow wells were delimited, the water table was delimited for the study and water samples were collected in the urban area of the city, in the wells cacimbas, defined and evaluated for the existence of Nitrate (NO₃⁻) in a ratio above or below the values defined by Ministry of Health Ordinance No. 2.914/2011, which govern standards of water standardization and potability in Brazil.

The results were spatialized in the form of maps with the use of Geostatistics through the software "ArcGIS for Desktop Advanced 10.2" [20], so that, later, they can serve as a basis for the development of public policies in the prevention of water contamination underground of the city of New Mamoré in Rondônia, Western Amazon, in the border with Bolivia.

2.2. Field and Laboratory Procedures

Two methods were used to register the wells. The first one according to the National Guide of Collection and Preservation of Samples: water, sediment, aquatic communities and liquid effluents of Water of the Company of Technology of Environmental Sanitation - CETESB [21] and National Agency of Waters - ANA. The second method was Stratified Random, according to Yamamoto and Landim [23] so that a certain number of observations, that is, of samples, can estimate the behavior of the set of all the potential observations of the population, it is necessary that these subsets are collected in such a way that each observation has the same chance of being chosen.

2.3. Collection and Preservation of Water Samples

For the field collection of the samples, it was made available by the Water and Sewage Company of Rondônia - CAERD, a technician of its staff. The owners or real estate agents, in the sites of the selected wells, were registered and signed the Term of Free and Informed Consent - TCLE.

For data collection and water samples, they were carried out according to the methodology proposed in the National Guide for Collection and Preservation of Samples: water, sediment, water communities and liquid effluents from the Company of Environmental Sanitation Technology [21] and Agency National Agence of Water [22], adopted by the CAERD Water Laboratory as follows:

a) The waters were collected at the exit of the well, using the local electric pumps. The water drawn in the initial three minutes is discarded to eliminate stagnant water.

Then, the water samples were conditioned in 500 ml plastic bottles, sterilized and adequately identified by Sampling Well (PA), numbered according to the collection sequence, in ascending order, and stored at approximately 4°C in a polystyrene box with ice to maintain the original characteristics of the waters.

b) The data of the wells, as well as the location of the collection, date and time, depth, rainfall in the last 24 hours, hygienic conditions of the soil were collected and prepared according to the instructions contained in CETESB Guide, wells, cesspools, animals and sewage nearby. At each point of water collection, the UTM coordinates were georeferenced using the Global Positioning System (GPS).

c) the bottles with water samples were transported to the Laboratory of the Federal University of Rondônia, where Nitrate levels were determined.

2.4. Ionic constituents

The water samples were previously filtered in cellulose acetate filters of 0.22 μm porosity and 13 mm in diameter (Sartorius Biolab Products) and analyzed by ion chromatography with conductivity detector (Ion Chromatograph with ion conductivity detector, METROHM - 882 Compact IC plus). The calibration of the equipment was performed by a calibration curve with specific standards, the concentrations of the ions present being calculated by comparison with external standards.

The analytical columns used were: Metrosep A Supp. 5 - 150/4.0 and Metrosep C 4 -150 / 4.0 (METROHM), with the fixed volume of injection of 100 μL and flow always maintained in 0.7 mL min -1. The anion and cation standards were introduced separately with 5 ml disposable hypodermic syringe into the ion chromatography injection system.

Determination of Nitrate content with the use of equipment made available by the Federal University of
Rondônia, through the Ion Chromatography with Conductivity Detector technique.

2.5. Spatial Distribution of Sampling

Through the random sampling, stratified in the urban area of Nova Mamoré, we selected 80 cacimba wells that are used continuously in the local supply.

Of the thirteen existing neighborhoods in the district of Nova Mamoré, four neighborhoods were excluded from the survey: the neighborhood called the Green Area, which still has no occupation. In the Nova Redenção neighborhood, five wells were selected according to the methodology adopted, but due to the absence of cacimba wells or Amazonian wells, because they were conditioned to the presence of massive rock a few meters deep (Complexo Nova Mamoré) was one of the excluded, the neighborhood Ambrósio, located in the industrial sector, was not allowed the registration of wells and neighborhood hortifrutigranjeiro did not have wells.

The spatial distribution of the samples was found in nine neighborhoods: Centro, Chacarê, Cidade Nova, João Francisco Clímaco, Nossa Senhora de Fátima, Novo Horizonte, Planalto, Santa Luzia and São José, according to Figure 1.

2.6. Contamination Mapping

One of the most efficient methods to characterize the contamination plume in the free aquifer of the Nova Mamoré Urban area, adopted in this work was the geostatistical method of kriging, allowed the identification of two anomalous areas of nitrate (NO3-) occurrences.

2.7. The Geostatistical Method of Kriging

According to Isaak & Srivastava [24] Kriging is termed as spatial estimator of regionalized variables, from adjacent values while considered independent in varigraphic analysis. Through it, one can obtain: a) The prediction of the point value of a regionalized variable and in a specific place within the geometric space, it is an exact interpolation procedure that takes into account all the observed values. b) The average calculation of a regionalized variable for a larger volume than the geometric support and the estimation of the main trend (drift), similar to the trend surface.

Krigagem provides, in general, non-tendentious estimates with minimal variation, bringing together several types of estimation methods, such as: simple, ordinary, universal, indicative, disjunctive and cokriging [25].

2.8. Ordinary Kriging

Ordinary Kriging is a linear estimation technique for a regionalized variable that satisfies the intrinsic hypothesis, whose objective is to minimize the bias estimation error, that is, where the mean residual error is equal to zero [26].

According to Landim [27], the most usual forms of linear kriging are simple, ordinary, universal and intrinsic. Nonlinear Krigages use some nonlinear transformation of the original data and are: lognormal, multigaussian, indicative, probabilistic, and disjunctive.
III. RESULTS AND DISCUSSION
3.1. Histogram and Box Plot Analysis of Nitrate Content Data in the Study Area
The histogram and the mean concentration graph of Nitrate contents were elaborated using the R Program for Statistical Computation.

Figure 2 shows a heterogeneous distribution of the contamination plume, with 37.5% of the samples having <10 mg/L Nitrate and 62.5% of the samples presenting >10 mg/L of Nitrate (NO₃⁻).

The results presented in the histogram show a heterogeneous distribution of Nitrate values and, according to the box-plot relative to figure 3, they are positively asymmetric, considering the approximation of the median value to the quartiles Q1.
Fig. 3: Average NO₃⁻ (mg/L) nitrate concentration in the study area.

3.1.1. Nitrate Isocontents

By observing the Map of Isocontents of nitrate (NO₃⁻) in figure 4, it can be seen that the highest levels were presented on the lateritic residual plateau, which covers a large part of the urban area of Nova Mamoré. These values are associated to the periods of occupation of the urban space considered older, since the central area of the urban nucleus, denominated "center", was the first one to be occupied.

The oldest population density in the municipality is associated with Nitrate levels ranging from 21.51 mg/L to 45.77 mg/L, with a gradual decrease in the most recent occupation areas (Table 1). Extreme values of 42.0 and 45.8 mg/L are observed in the urban area of the oldest human occupation, as opposed to minimum values of 0 and 2 mg/L, located outside the central area of the city and associated with the sector with low population density (Table 1).
Still based on the existing districts in New Mamore: New city; Chacareiro; Novo Horizonte and Ambrósio, we found levels of Nitrate (NO3-) below 9.70 mg/L and, therefore, lower than the maximum limit of 10 mg/L determined by Ministry of Health Ordinance No. 2914/2011. As a result, the other districts, in a total of eight, had higher levels than indicated by the aforementioned Ordinance.

3.2. Nitrate Content Occurrence Isoprobability > 10 mg/L

The isovalues, determined in the study area with indexes above 10 mg/L, are shown in figure 5.

Fig.5: Nitrate concentration map (NO3-) with cut level > 10mg/L
When analyzing the mentioned figure, referring to the isoprobabilade of occurrence values above 10 mg/L, it was verified that the area with the highest occurrence of nitrate levels and with probability of occurrence in a percentage between 70 and 100% is that of the nucleus urban center of Nova Mamoré, cut by Federal Highway BR 425, this act and acted as facilitating agent of the occupation and urban transformation of the city, the consequences of this urban concentration motivated by federal highway BR 425, resulted in the contamination of groundwater of the place studied. The study by Alaburda and Nishiara [12] corroborates, with this study when he mentions that the anthropogenic action contributed to the degradation of the waters associated with low indicators of infrastructure in networks that collect sanitary sewage.

Studies carried out by Campos [9], in the municipality of Mirante da Serra, State of Rondônia, showed high values of Nitrate (NO₃⁻), above 10 mg/L in the most densely populated areas. Low probability values of contamination, in general, range from 0 to 53% in areas farther from the central axis with urban influence. It is also observed the appearance of a transition band with probability of up to 53% and greater than 72% of occurrence, above 10 mg/L, which, in this case, is associated with the presence of drainage of the Ambrósio channel and Olaria stream.

Results evidencing the nitrate content, generally exceeding 10 mg/L, which limits the potability for human consumption, were detected in shallow tubular wells and cacimba type, in the municipality of Urânia-SP, [28]; [29]; [30], a situation similar to that found in the municipality of Nova Mamoré in the cacimba wells mapped.

3.3. Nitrate Content Occurrence Isoprobability <3 Mg/L

The probability of having nitrate contents below 3 mg/L nitrate (NO₃⁻) in the study area is 70 to 100%, as shown in figure 6, corresponding to the red band of the legend. In spite of its predominant position of lower third of valley slope, the lowest levels of nitrate are in this range. In contrast to the lateritic plateau area, in this figure n 18 indicated by the blue color where the probability of finding contents below 3 mg/L is lower than 12%, this low percentage confirms the theories of Campos [9] and Hirata [28], where the anthropic action resulting from urban activities have a strong impact on the quality of groundwater, and in this space the groundwater is already in a high degree of impaction.

The areas with low levels of nitrate are little occupied by the urbanization of the city of Nova Mamoré, where the population concentrated on the axis of federal highway BR 425, shown in figure 6, the high levels of contamination.

At various points, the open ombrophilous forest, which covers approximately 66% of the city of Nova Mamoré, is present in these areas, where the contamination was less than 3 mg/L of nitrate.
Figure 6: Nitrate concentration map (NO₃⁻) with cut-off level <3mg/L nitrate

Table 1a - Location, Coordinates, Nitrate Content and Depth of PAs

| NEIGHBORHOOD | UTM East  | UTM North  | Well | Content Nitrate (NO₃⁻) | Depth (m) |
|--------------|-----------|------------|------|------------------------|-----------|
| Centro       | 244893,205 | 8848763,191 | 21   | 23,072                 | 7         |
| Centro       | 244781,464 | 8848640,577 | 41   | 34,125                 | 8         |
| Centro       | 244700,527 | 8848621,249 | 42   | 42,084                 | 13        |
| Centro       | 244538,652 | 8848324,077 | 51   | 12,772                 | 10        |
Table 1b (continuation) - Location, Coordinates, Nitrate Content and Depth of PAs

| NEIGHBORHOOD        | UTM East  | UTM North | Well | Content Nitrate (NO₃⁻) | Depth (m) |
|---------------------|-----------|-----------|------|------------------------|-----------|
| Novo Horizonte      | 243958.804 | 8848637.255 | 48   | 4,432                  | 6         |
| Novo Horizonte      | 243892.968 | 8848791.277 | 45   | 0                      | 9         |
| Novo Horizonte      | 243833.775 | 8848965.835 | 44   | 14,625                 | 7         |
| Novo Horizonte      | 243997.461 | 8848298.406 | 59   | 0.399                  | 12        |
| Novo Horizonte      | 244087.458 | 8848082.775 | 58   | 4,986                  | 3         |
IV. CONCLUSIONS AND RECOMMENDATIONS

Eighty cacimba wells were registered in the urban area of the municipality of Nova Mamoré, State of Rondônia, and these were spatialized through the stratified random sampling method, with water collection and chemical analysis of the Nitrate (NO₃⁻) parameter, where high levels of contamination in the most urbanized area. Sixty-two and a half percent (62.5%) of the samples presented nitrate levels above 10 mg/L, the maximum limit allowed for human consumption according to Ministry of Health Ordinance No. 2.914/2011, which governs standardization norms and potability of water in Brazil. With the application of the geostatistics method, through the non-linear indicator Kriging, through the study of its spatial distribution and variability, two areas of anomalous levels of Nitrate (NO₃⁻) were identified: one up to 3 mg/L and another above 10 mg/L. The two identified areas demonstrated that the urban occupation evidenced in the BR-425 highway axis, as well as the high number of wells excavated, corroborate...
with the theories of the authors mentioned in this study where the lack of basic sanitation, specifically, allied to the high population concentration in this place contributed to the concentration of the contamination in the water table, making underground water unfit for some uses, especially human consumption.

The other area with lower levels of nitrate, less than 3 mg/L, is still little occupied by urbanization, characterized in this study by native vegetation cover and low number of excavated wells.

This study indicated that the source of the contamination originates from the result of anthropogenic activities, evidenced by the inefficiency of the sanitary sewage system, one of the basic sanitation axes in which, specifically, it addresses the collection, treatment and proper disposal of sanitary sewage and other hygiene.

Even with a high amount of Nitrate (NO$_3^-$), in the largest number of analyzed samples, we must consider that there are still areas with low levels of Nitrate (NO$_3^-$) and those that have not undergone anthropogenic processes.

It is necessary, therefore, the insertion of public policies in the sector of basic sanitation of the municipality.

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