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Evaluation on the risk of contamination by mercury and classification of areas at foz of river Tapajós, Pará, Brazil.

Enilson da Silva Sousa[1], Ronaldo Rocha[2], Dulcideia da Conceição Palheta[3], Joaquim Carlos Barbosa Queiroz [4]

[1] Universidade Federal do Pará – UFPA - Universidade Federal do Pará – UFPA - Rua Augusto Corrêa, n. 1. CEP: 66075-110 – Guamá – Belém-Pa. - E-mail: enilson@ufpa.br ou prof.enilson@gmail.com.
[2] Laboratório Central do Estado – LACEN - Av. Augusto Montenegro, km 10 – Icoaraci – Belém-PA. - E-mail: ronaldo.lacen@gmail.com.
[3] Universidade Federal Rural da Amazônia – UFRA - Avenida Presidente Tancredo Neves 2501 - 66077-530 Belém – PA - Telefone: (91) 274 3493/(91) 274 0900 Fax: (91) 274 3814 - Site: www.ufra.edu.br - E-mail:deiapalheta@hotmail.com.
[4] Universidade Federal do Pará – UFPA - Instituto de Geociências – IG - Rua Augusto Corrêa, n. 1. CEP: 66075-110 – Guamá – Belém-PA E-mail: joaquim@ufpa.br

Abstract — *Geostatistics is a tool that, in the last years, has been emphasizing in the analysis of spatial data in several areas, especially in the environmental sciences, since it is a technique that presents advantages in relation to other methods of spatial interpolation, mainly, by the possibility of analysis of data in small quantity, informing the errors of estimates and consideration of the anisotropy. In this work, mercury maps were elaborated from spatially distributed data, using Geostatistics to infer results for the location and risk of contamination of this metal along the mouth of the Tapajós River, Santarém-PA. Mercury contamination risk or probability maps were also constructed, which enabled the classification and quantification of the areas contaminated by this metal for several levels of confidence in and around the mouth of the Tapajós River. The methodological procedures will include the use of the "R" program to quantify mercury concentrations in the study area and to map the spatial distribution of these concentrations. To determine the total mercury in the samples, they were submitted to acid digestion and determinations of Hg performed by Atomic Absorption Spectrophotometry with cold steam generation at the Central Laboratory of the State of Pará. Two campaigns were carried out: in July and December 2014, with 37 and 45 sampling points in each one. The results point to a possible contamination of the area in some points, presenting indices above the one recommended by resolution n. 357/2005 of CONAMA.*
Keywords— *Geostatistics. Mercury contamination. Rio Tapajós. Santarém.*

I. INTRODUCTION

Geostatistics deals with simple but important questions ranging from spatial interpolation and quantification of uncertainty to variables that have spatial continuity that can be measured anywhere in the area / region / zone under study and is based on traditional statistical concepts. It is based on the Theory of Regionalized Variables, initially formulated by Matheron (1960), based on practical studies developed for estimates of mineral reserves by mining engineer D. Krige and H.S. Sichel, in the calculation of reserves in the gold mines of South Africa in the 1950s (CAMARGO, 2002). Nowadays, Geostatistics is applied in many fields, from earth and atmosphere sciences, agriculture, soil science and hydrology, environmental studies and, more recently, epidemiology.

It is the 60th International Congress of Geostatistics held in April 2000 in South Africa, where the work of Matheron and Sichel was highlighted, as well as the extraordinary development and diffusion of the use of computers and programs aimed at these areas that have fostered a rapid development of Geostatistics. It was during this period that regression estimates were used for mineral reserve blocks for the first time (CAMARGO, 2002). In the 1960s, Matheron made this technique known as kriging and established the first global geostatistical model for reserve recovery estimation and many fundamental concepts in Geostatistics and valid to this day. From there, this knowledge was disseminated throughout the world following the French formation (JOURNEL, 1984; KRIGE, 2000; QUEIROZ, 2003).

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In the 1970s, there was great growth not only of the geostatistical community but also of new models. In the following decade a rapid expansion of computers and softwares occurred, especially in the areas of spatial modeling (variograms), kriging and other techniques, such as kriging and universal kriging (QUEIROZ, 2003), and thus geostatistics has made great progress. The 1990s were the period when several ideas were introduced in South Africa, such as co-kriging of virgin areas using limited data from drilling holes and regularized adjacent data from other explored areas, and practical studies of the effects and implications of bias conditionals in block estimates.

The discovery and modeling of these patterns of spatial correlations allow a better understanding of the underlying physical processes, as well as assist in spatial interpolation. In general, it is desired to estimate the concentration of high values of a given element - and to assess its connectivity in space or time - instead of the spatial mean values, and therefore, the generated uncertainties generally need to be documented in a quantitative way to assist in making of decision-making. Initially, an exploratory analysis of the data is performed. In these exploratory analyzes the most relevant characteristics of the data set of the study area are presented. This purely descriptive part is a preliminary step for the construction of spatial distribution models of the variables used that are used in kriging, which makes estimates of non-sampled values of the study área. The Hg data were treated using the geostatistical procedures, with the objective of generating a probability map of the concentrations of Hg, which exceeded the limit allowed by Conama (0.0002 ppm), and from there to construct classification maps of contaminated areas for the mean and median values. In this case, the values of 80%, 85% and 90% were established, that is, for the 20%, 15% and 10% values above those established by the regulatory body, respectively.

II. MATERIALS AND METHODS LOCATION OF SAMPLE COLLECTION POINTS

To select the sampling sites for water samples in 2014, 37 samples were used in the first campaign (July 18-28, 2014) and 45 samples in the second campaign (December 13-20, 2014). total of 82 samples, in the period from rainy to less rainy in the Amazon basin, respectively (Figure 1).

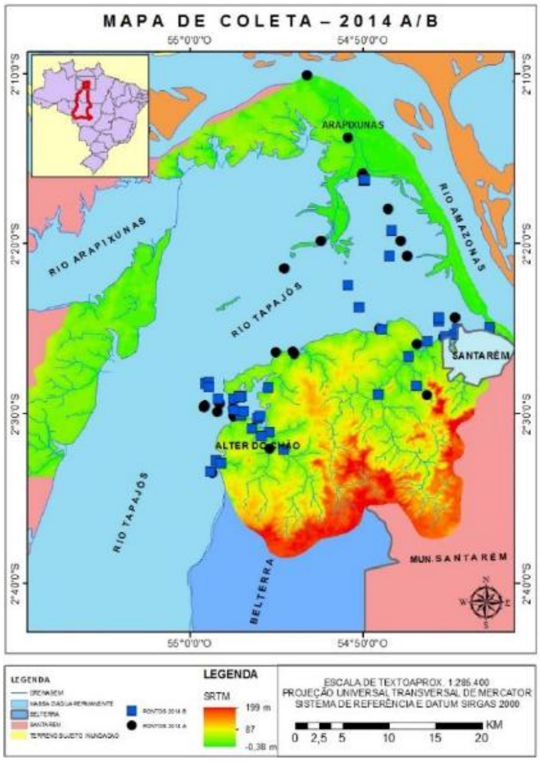

Fig.1 - Sampling area in Santarém, with the respective collection points in the first and second campaigns of 2014.

Elaboration: Author of the research (2016)

Mercury determination contents in samples

The laboratory analyzes to determine the total mercury values found in the samples were performed by Atomic Absorption Spectrophotometry, with cold vapour generation, at the Central Laboratory of the State of Pará, Belém-PA. In the field, about 5 liters of samples were collected in polypropylene flasks previouly treated in acid bath. The results had as reference the resolution n. 357/05, of CONAMA/BR. The decantation / flocculation procedure with aluminum sulphate was done by emptying the supernatant in the field and transferring the moist residues to Falcon tubes.

Geostatistical method

Initially the exploratory analysis was carried out to determine if the sample distributions satisfy the requirements (normality and stationarity) for the construction of the geostatistical models. Then, the spatial distribution is modeled by means of variograms. In the classification of contamination risk in the study area, geostatistical techniques were used to characterize the area in order to know the estimates of the magnitude, extent and location of contaminated areas in non-sampled locations, with the inclusion of associated uncertainty measures. All Hg data were treated using geostatistical

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procedures, which allowed the analysis and construction of variable maps from spatially distributed data to infer results of the location and risk of contamination along the mouth of the Tapajós River.

III. RESULTS AND DISCUSSION

Analytical results for water in 2014

The Hg values in the samples collected along the Arapixunas Hole, especially at the entrance to the Amazon River, were elevated, and may be related to the large amount of organic matter in suspension brought by this river into the small hole. At this site, it receives a significant volume of muddy river waters, forcing it into the mouth at its opposite end.

It was observed that the concentration of 5.15 (µg / L) in Boca do Arapixunas (Foz do Amazonas) would be an out lie, which would require a more detailed monitoring of Amazonas' contribution to the system. Concentrations in 5 samples of the first season of 2014, Ponta do Tauá, Foz do Tapajós (Ponta do Cururu), Foz do Tapajós (Sample 4) and Foz do Tapajós (Sample 5) show optimal detection limit (LOD - 0.011 ppb) , less than can be quantified by this experimental procedure used in this work. Therefore, it can not be stated that there are Hg concentrations in the samples in question, but they were not determined and quantified by this technique.

The average found for samples from the first campaign of 2014 was 1,146 (µg / L) average above 0.119 µg / L, found in the river Rato, por Silva (1997) and in the Acre tributaries of 0.116 µg / L, by Mascarenhas et al. al. (2004). In the samples from the second campaign of 2014, only 4 (four) samples: Boca do Igarapé do Camarão, Boca do Igarapé do Camarão (Direct Sample), Igarapé do Miritiapina (Nascente II) and Igarapé Miritiapina (Direct Sample) (LOD) above the detectable. The remaining samples, 41, could not be quantified because they were below the LOD. This fact can be justified because it is a drought period and most of the samples are within the mouth, a fact that facilitates the adsorption of mercury in this type of environmen.

Key Descriptive Statistics for the Hg Variable in 2014

In the evaluation of the mercury concentration data, Table 1 presents the descriptive statistics for the months of July and December of 2014, in which the values occur well above those established by CONAMA, mainly in July. In December, most of the values (more than 75%) are below the LOD. Due to the large number of values below the LOD, greater variability is observed in the month of December in relation to July. This is shown by the standard deviations and the mean and median measurements that are most distant from each other in the month of December.

Table 1 - Statistical summary for mercury concentrations (ppb), 2014

Statistic	Dez.	Jul.
Minimum	0,011	0,011
Quantil 25	0,011	0,710
Mediana	0,011	1,020
Mean value	0,443	1,155
Standard deviation	2,622	1,016
Quantil 75	0,011	0,710
Maaximum	17,6	5,15
Interval	17,589	5,139
Score	45,000	37,000
Conama/357**	0,2	0,2

(*) LOD < 0,011 ppb
** CONAMA RESOLUTION 357/2005
Elaboration: Author of research (2016)

Results of geostatistical analyzes

The data were submitted to geostatistical analysis to perform the mapping of mercury Concentrations. According to Guimarães (2004), the choice of the variogram model to be used is one of the most important aspects of Geostatistics (Figure 2).

All calculations depend on the adjusted variogram model, and therefore, if the adjusted model is not appropriate, all of the following calculations will contain errors that may affect the inferences.

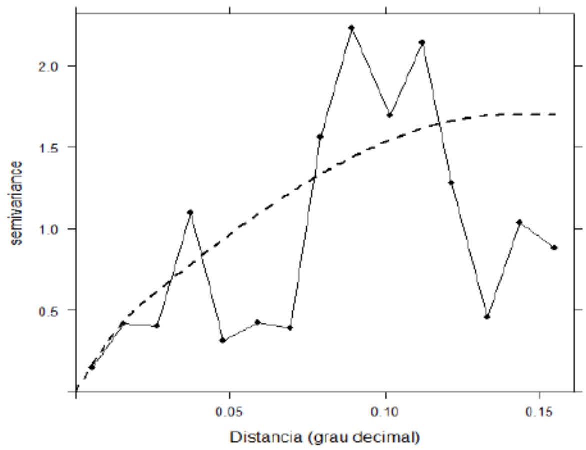

Fig.2 – Experimental semivariogram (continuous line) and fitted model (dashed line) for Mercury.

Elaboration: Author of the research "R" (2016)

With the results of the semivariographic model, kriging could be used to calculate the estimates of mercury concentration values in non-sampled locations in the field. With the mesh of interpolated points, we have a greater detail of the area under study, and a better visualization of the behavior of the variable in the region, due to the increase in the number of observations (DEUTSCH; JOURNEL, 1997).

From the data interpolated by kriging the map of the distribution of mercury concentration in the study area (Figure 3) was constructed, where two zones of concentration Hg can be observed along the mouth of the Tapajós. The Lago Preto region, in the community of Itaparí, on the right bank of the river, and the internal mouth of the Arapixunas hole (region within the mouth: Arapixunas mouth), these are the places that are most likely to be contaminated by Hg.

It was not possible to construct the concentration map in the second campaign because most of the samples were below the detection limit of the method. In this campaign, only four points could be quantified.

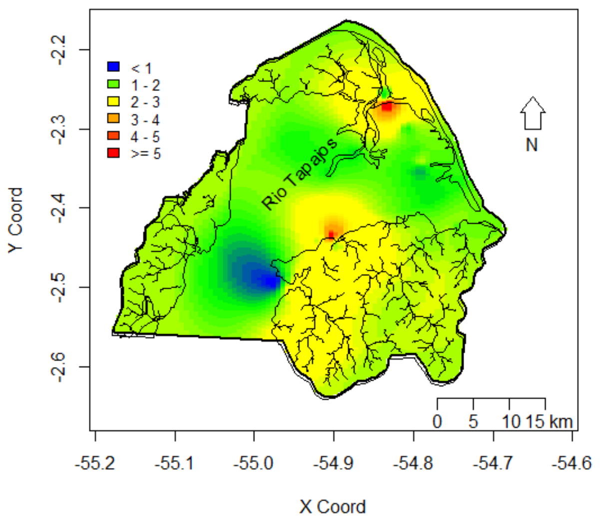

Fig.3 - Map of the concentration of Hg for samples 2014 A, with hydrography
Elaboration: Author of the research (2016)

IV. MAPS OF CHALLENGES OR CONTAMINATION RISKS

Local uncertainty models for risk of contamination

A non-sampled location can be classified based on the estimate of its true value or, alternatively, on the probability that the point is below (or above) a given cut-off value. Estimates involve errors. Errors are subject to costs. If a point is incorrectly classified as high when in fact it is low, there is a cost of super classification (or super estimation). On the contrary, if the point is incorrectly classified as low when in fact it is high, there is a cost of subclassing (or understating).

In the case of environmental contamination, overestimation of the extent of contamination will increase the cost of remediation, but the underestimation of contamination may result in premature loss of life or high cost of health. In such a situation, the cost of underestimation may be much higher than the cost of overestimation. The unit of cost of super estimation and underestimation depends on each situation.

The uncertainty assessment can be represented by probability maps of the concentrations of the study variable. In this case, the probabilities of occurrence above a certain cut-off point, generally defined by legislation, are represented. Figure 4 shows the probability map for mercury, highlighting two regions with probability above 80% of the concentrations presenting values above 0.0002 ppm (limit established by CONAMA). These sites were identified in the regions of Arapixunas and Itaparí, left and right bank of Tapajós, respectively.

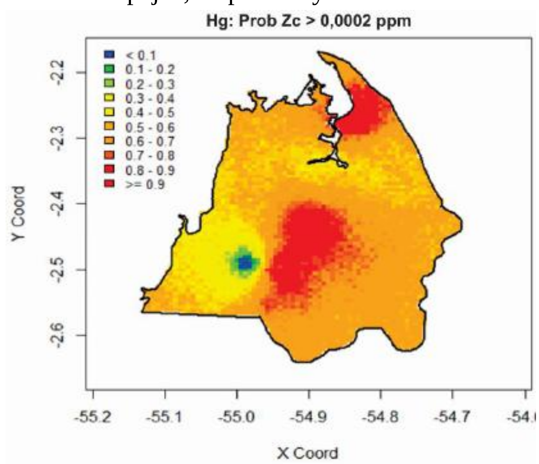

Fig.4 - Probability maps of Hg concentrations exceed the maximum permissible limit of 0.0002 ppm
Elaboration: Author of the research (2016)
Classification maps with probability of contamination above 80%, 85% and 90%

Based on the probability map it was possible to construct classification maps of the contaminated areas for the mean, median or quantil values. In this case, maps with quantiles, 80%, 85% and 90%, that is, for the 20%, 15% and 10% values above those established by CONAMA, respectively, were elaborated. The research area corresponds to 115,950 km2, from the results generated and the size of the pixels of the vector produced, it was possible to reach the sizes of the areas classified as contaminated or uncontaminated, with each pixel having an area of 100x105 m.

For the classification map that used the 80% quantil (Figure 5), the contaminated area (in red on the map) corresponds to 30.06% of the total area, which corresponds to 34.06 km2, and 69.93% of probability that the area is not contaminated, that is, 81.89 km2, of the research area.

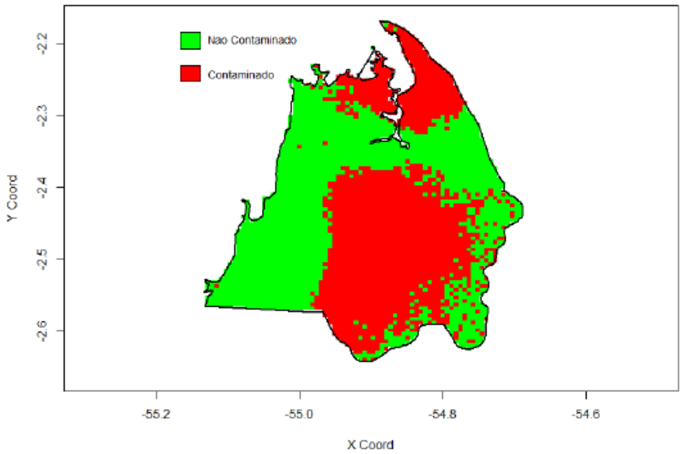

Fig.5 - Classification Map with probability above 80% of Hg contamination exceeds the allowed maximum limit of 0.0002 ppm

Elaboration: Author of the research (2016)

For the classification of locations based on the probability that concentrations above the maximum permissible limits were greater than 85% of contamination (Figure 6), the following values were found: of 7.81% probability of the area being contaminated, or 9, 05 km2, and 92.18%, probability of the area not being contaminated or 106.89 km2.

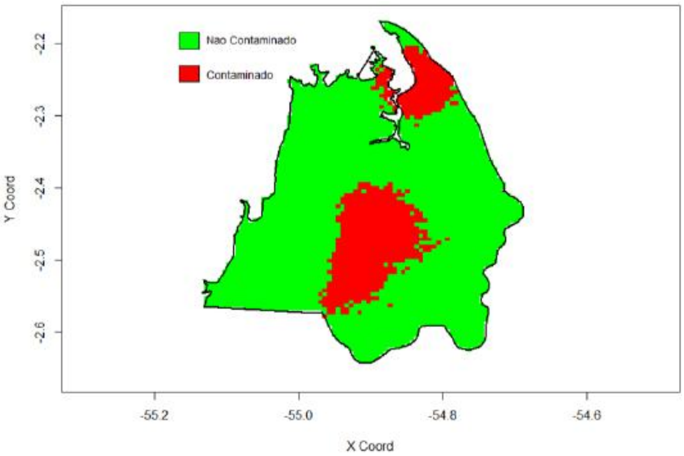

Fig.6 - Classification Map with probability above 85% of Hg contamination exceeds the allowed maximum limit of 0.0002 ppm

Elaboration: Author of the research (2016)

For classification of locations based on the probability that concentrations above the maximum permissible limits are greater than 90% of contamination (Figure 7). These values are 7.81% probability of the area being contaminated or 3.30 km2, and 92.18%, probability of the area not being contaminated or 106.89 km2 of the studied area.

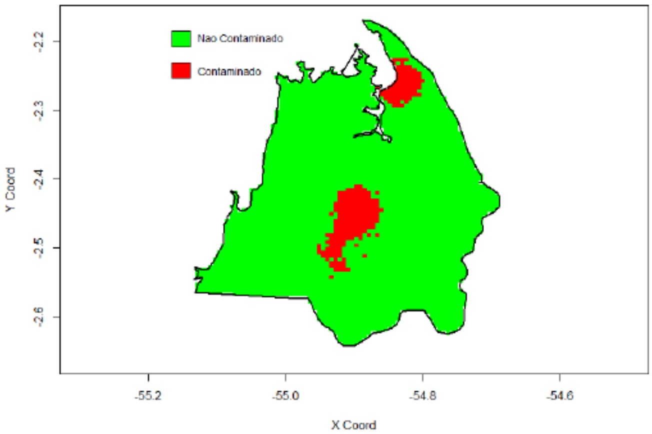

Fig.7 - Classification Map with probability above 95% Hg contamination exceeds the allowed maximum limit of 0.0002 ppm

Elaboration: Author of the research (2016)

V. DISCUSSION OF RESULTS

Among the spatial analyzes performed in the Hg data, considering the months of July and December, it was observed that the distribution of Hg values does not occur in the same proportion in the two periods. The samples that presented higher values of mercury occurred predominantly in July and suffered a retraction in December, especially along the mouth, being able to be associated to the adsorption of this metal. The highest values in the Tapajós river flood period are related to the possible enrichment of Hg concentrations in the suspended solids in this region, due to regional soils and the increase of erosion caused by the expansion of agriculture, according to Gomes (2007).

The pattern of the spatial distribution of the values presented in the probability maps suggests that in the period of July the processes of deposition and mobilization of the Hg in the sediment predominate, since the greater adsorption of the mercury is associated to the increase of the contents of compounds (oxides and hydroxides) of iron, aluminum and manganese and organic matter in the sediments. In them, the ionic content of sodium and potassium, which is quite high, contributes significantly to the increase of the local ionic strength, acting in a way to promote the mobilization of mercury. In the sediments, mercury is associated with organic matter and little influence has the oxide-hydroxides present, evidenced in the triangular diagram (Hg / Fe / Al) (HYPOLITO, 2005 and LONGO, 2012).

It is at this time that the largest contribution of material transported from the highest areas to the flat ones occurs, especially of particulate matter, functioning as an important transport vehicle of Hg and is attributed to this erosion process through the superficial drainage of rainwater after washing and leaching of the soils of the region, carrying trace elements to the talvegue (GOMES, 2007).

With the arrival of December, the Hg is adsorbed to the sediments of the mouth and they are transported to the hydrological system of the mouth of the Tapajós, until more distant area (PALHETA, 2011). The transport of this mercury occurs more efficiently in areas close to the canal, since they present a great flow and as the Tapajós is a river of drowned mouth (SOUSA, 2009), the narrowing of the channel near the urban area of Santarém, undertakes to the great river strength and speed, interconnecting it to the Amazon. It was observed in field work that in this area there is a very large flow of water that drains the mouth into the Amazon River, thus facilitating the transport of sediments from the bottom of the mouth of the Tapajós, with mercury adsorbed to the Amazon (LACERDA, 2009).

The spatial distribution of samplings by ordinary kriging evidenced, points to areas of higher concentration of Hg, related to regions of higher sediment deposition, presence of organic matter in large concentrations, as in the mouth of Arapixunas and Itapari community. Such direct observations on probability maps show a direct correlation of the presence of organic matter and higher concentration of Hg. Although it is not conclusive to establish a direct relationship between these values, it is possible to make a correction between the formation of preference zones of sediment deposition and accumulation of mercury (JAQUET, 1989).

The presence of the Arapixunas hole, which acts as a connecting channel between the Amazonas and Tapajós rivers, is another factor to consider, because in July, the Amazon basin facilitates the deposition of sediments by the Amazon within the mouth of the Tapajós and the reverse direction around the month of December, hence the association with the high values found in Boca do Arapixunas. A similar phenomenon is pointed out by Maurice-Bourgoin et al. (1999), when studying the Beni River, claiming that Hg adsorbed the sedimentary particles is enriched by the regional soils in oxide and Fe hydroxide during the rainy season. Thus, it would be transported to the lower area and, by analogy, this same phenomenon may be occurring at the mouth of the Tapajós.

Another factor to be taken into account, as pointed out by the specialized literature, is the presence of the Alter-do-Chão Formation in the surroundings of the mouth, which would have an important contribution to this correlation, since it is a region of higher average altitude than those of the surrounding of the mouth and has a great concentration of Fe, that would make important connections with the Hg, associated with the organic matter deposited by the Amazon, which would justify the high values found in the locality (Brauba, 2010; Wasserman, 2001).

Deforestation, monoculture and subsistence agriculture, responsible for the erosive process along the upstream mouth, releases the mercury contained in the soil and drags the mercury adsorbed to the organic and inorganic particles into the aquatic system, thus another variable to be taken into account to justify the high quantified value of Hg at some points in the sampled area (LACERDA, 2009).

Taking into account that the Tapajós river basin, over the decades, has been greatly impacted by agropastoral and mineral projects, and large projects, such as the construction of hydroelectric plants in progress or projected, may be another factor responsible for this contamination and observation of values higher in future samples, when due to the large hydroelectric projects upstream of the basin are in progress or implanted.

A atividade mineradora artesanal, ao longo das últimas décadas, é outra variável a ser considerada, pois, os garimpos artesanais ao longo das décadas de 1980 e meados de 1990, contribuíram para remoção do solo do médio e alto curso do rio, assim como a deposição direta de Hg, com o processo de amálgama da queima do ouro, sem a menor tentativa de conter o vapor ou o metal em estado líquido, mesmo considerando que o vapor entraria no sistema global de mercúrio (SANTOS et al. 1995; 2000).

In the month of December, when the second campaign occurred, most of the values (more than 75%) are below the LOD. Due to the large number of values below the LOD, greater variability is observed in the month of December in relation to July. This is shown by the standard deviations and the mean and median measurements that are most distant from each other in the month of December.

These results are noteworthy because it is a period that is not favorable to large concentrations of organic matter, and the adsorption of Hg may be occurring with greater intensity (not quantified by the method), which may have contributed to these results.

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VI. CONCLUSION

The use of kriging as an interpolation method was of fundamental importance for the calculation of the estimates of the concentration values of the mercury in location not sampled during the field steps. In this way, it was possible to obtain a greater detail of the study area and better visualization of the behavior of the variable searched throughout the region, which was visualized in the distribution map of the concentrations.

The use of this tool allowed the quantitative evaluation of the concentrations of mercury in the mouth of the Tapajós river. With the mapping of the concentrations, one can have an idea of the locations, their extensions and where the highest concentration levels of mercury are concentrated. With the use of probabilistic or risk maps it was possible to quantify, in percentage or probabilistic terms, the degree of danger that may be being submitted to the population that is located in the areas of greatest risk.

Classification maps of contaminated areas may serve as a basis for remediation, prevention or safety plans by the agencies concerned. This practice is used in various parts of the world, especially when dealing with contamination of anthropic origin, such as industrial waste, garimpos, factories, etc., where classification maps can be used for the negotiation of alternative remediation plans with the responsible actors by the problem.

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