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Large-scale projects in the amazon and human exposure to mercury: The case-study of the Tucuruí Dam

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ABSTRACT

The Tucuruí Dam is one of the largest dams ever built in the Amazon. The area is not highly influenced by gold mining as a source of mercury contamination. Still, we recently noted that one of the most consumed fishes (*Cichla* sp.) is possibly contaminated with methylmercury. Therefore, this work evaluated the mercury content in the human population living near the Tucuruí Dam. Strict exclusion/inclusion criteria were applied for the selection of participants avoiding those with altered hepatic and/or renal functions. Methylmercury and total mercury contents were analyzed in hair samples. The median level of total mercury in hair was above the safe limit (10 µg/g) recommended by the World Health Organization, with values up to 75 µg/g (about 90% as methylmercury). A large percentage of the participants (57% and 30%) showed high concentrations of total mercury (≥ 10 µg/g and ≥ 20 µg/g, respectively), with a median value of 12.0 µg/g. These are among the highest concentrations ever detected in populations living near Amazonian dams. Interestingly, the concentrations are relatively higher than those currently shown for human populations highly influenced by gold mining areas. Although additional studies are needed to confirm the possible biomagnification and bioaccumulation of mercury by the dams in the Amazon, our data already support the importance of adequate impact studies and continuous monitoring. More than 400 hydropower dams are operational or under construction in the Amazon, and an additional 334 dams are presently planned/proposed. Continuous monitoring of the populations will assist in the development of prevention strategies and government actions to face the problem of the impacts caused by the dams.

1. Introduction

Mercury exposure is a serious public health problem in the Amazon. Since 2013, Brazil, along with 128 countries, became a signatory of the Minamata Convention on Mercury (www.mercuryconvention.org), with the aim of adding international efforts to reduce and combat environmental and human exposure to this metal. The World Health Organization (WHO) endorsed this action in January 2014 (available at: http://apps.who.int/gb/ebwha/pdf_files/EB134/B134_R5-en.pdf).

Traditional gold mining is one of the main economic resources for the human population in the Amazon, using mercury to extract the gold particles found in the rivers. In the 1990s, the activity of traditional small-scale gold mining sites (named *garimpos* in Brazil) was responsible for the emission of up to 120 t of mercury per year into the environment (Veiga, 1997). Thus, in recent decades, *garimpos* and the contaminated areas downstream have been routinely monitored for human exposure to this metal (Berzas Nevado et al., 2010).

Other factors, such as river damming, may show the potential to facilitate mercury accumulation in the Amazonian environment. Both mercury methylation and bioaccumulation may be increased by submerging terrestrial areas (peatlands, upland soils, and vegetation) (Bodaly et al., 1997). Some hypotheses account for this general, if not

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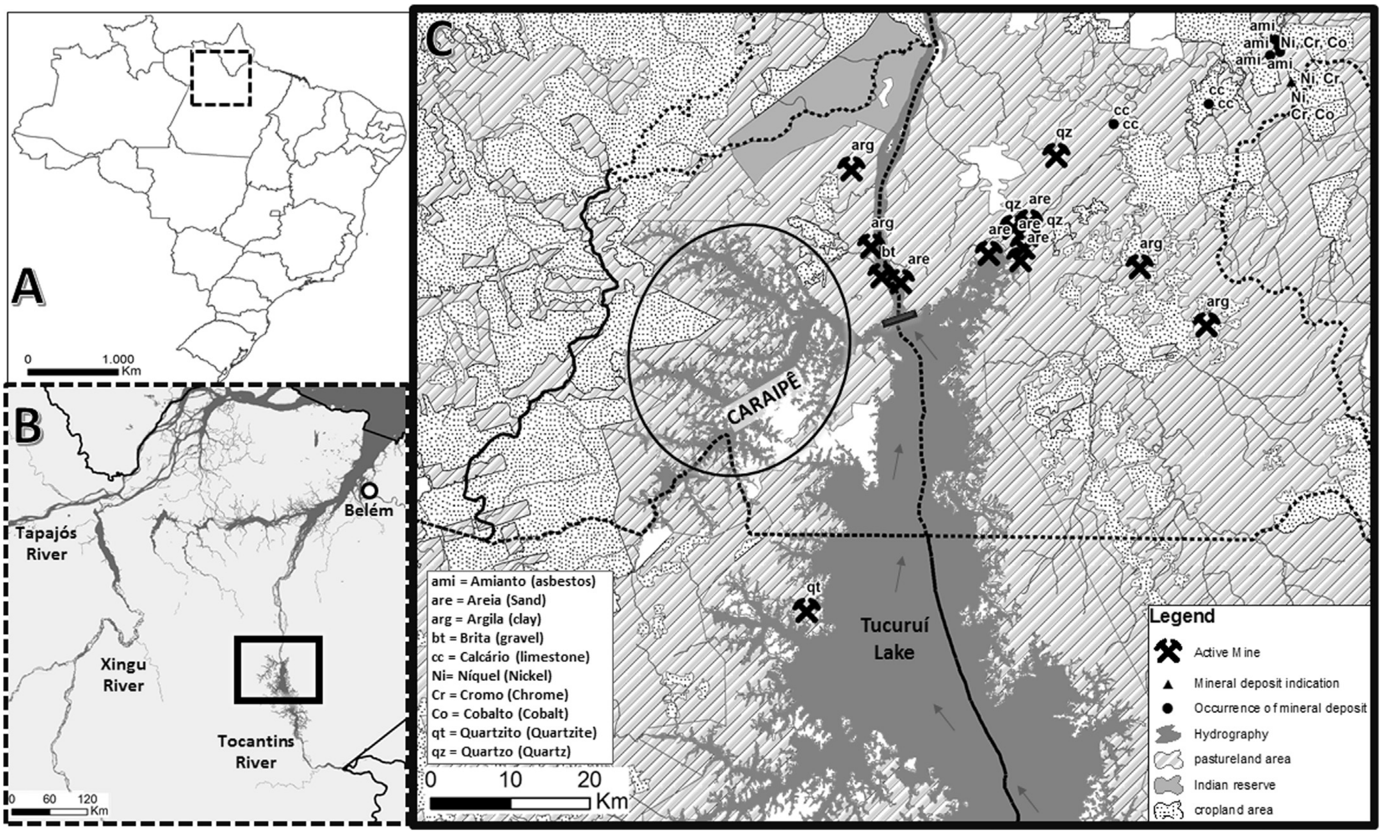

Fig. 1. Maps of Brazil (A), State of Pará (B) and Tucuruí (C). In C, the two communicated compartments (Caraipé and Lake) of the reservoir are shown in addition to the water flow, the dam (black bar), the land use and the present active mine points (note that none of these points are for gold extraction). Participants of the study were from Caraipé (area included in the circle). Maps were obtained from the Instituto Brasileiro de Geologia e Estatística (IBGE, Brazil), Departamento Nacional de Produção Mineral (DNPM, Brazil) and Ministério da Agricultura, Pecuária e Abastecimento (MAPA, Brazil).

universal, effect of impoundment: the degradation of organic carbon by microbiota and oscillations of temperature, and redox status of reservoirs, among others (Kelly et al., 1997). This occurrence has been repeatedly registered in reservoirs of different regions of the globe with no known sources (anthropogenic) of mercury (Bodaly et al., 2007; Berzas Nevado et al., 2009; Gray and Hines, 2009; Li et al., 2013; Johnson et al., 2015).

An increased number of dams are in different development or operational stages, occupying about 100,000 km^2 (or 3% of Brazil's Amazon forest) (Fearnside, 1995). Recently, the Brazilian government has approved the construction of several dams on the rivers of the Amazon rainforest, in order to provide electricity for some large cities in the country and to attend the growing industrial necessity (especially that of the Aluminum Industry) (Fearnside, 2016). The impacts of these dams will profoundly affect the environment and human health of the riverside populations of the Amazon. Yet, studies of human exposure to mercury in dam areas in the Amazon are scarce.

One of the largest dams ever built in the Amazon is the Tucuruí Dam in Tocantins River basin, East Amazon (Fig. 1 and Table 1). After its completion in 1984, this dam caused the inundation of 2430 km^2, generating the displacement of the local population, and exacerbating insect proliferation and the incidences of endemic diseases such as malaria (Fearnside, 2001). It generates electricity for a major part of the country; still, it remains controversial because of the absence of adequate studies about the environmental impact and sustainable development (Fearnside et al., 2001).

Different from other regions in the Amazon (such as Tapajós River basin), the Tucuruí area is not highly influenced by *garimpos* using mercury. About 50 years ago, mining activity was drastically reduced because it was not enough lucrative economically. Still, works in the past indicated mercury contamination in this environment (Aula et al., 1995; Porvari, 1995; Palermo et al., 2004; Kehrig et al., 2008, 2009; Rodríguez et al., 2014). Mercury was detected in water and sediments with values of 12.7 ± 8.4 ng/l and 0.012–0.037 μg/g, respectively (Aula et al., 1995; Kehrig et al., 2009). Also, floating plants such as *Eichhornia crassipes* or *Scirpus cubensis* showed levels of mercury as high as 0.075 ± 0.038 μg/g, especially concentrated in the roots of the plants (Aula et al., 1995). The highest contents of mercury were found in fish of this region, according to their positions in the food chain. Herbivorous fish such as *Anostomidae* sp. and *Prochilodus nigricans*

Table 1
Geological and limnological characteristic of Tucuruí reservoir.

Geographic localization	03°43´ – 05°15´S 49°12´ – 50°00´W
Main tributary[a]	Tocantins River
Drainage area (km^2)[a]	803,250
Inundated area (km^2)[a]	2875
Maximum depth (m)[a]	75
Total volume (km^2)[a]	45.5
Average flow (m^3/s)[a]	11,000
Water color[a]	Clear water
Trophic stage[a]	Mesotrophic
pH[a]	6.5 – 7.4
Conductivity (μS/cm)[a]	47 – 62
Vegetation[a]	Humid forest
Water Hg levels (ng/L)[b]	4.7 – 28.2
Sediments Hg levels (μg/g)[c]	0.012 – 0.037
Predatory fish Hg levels (μg/g)[d]	0.66 – 4.00

Note: Data from
[a] Espindola et al. (2000).
[b] Kehrig et al. (2009).
[c] Aula et al. (1995).
[d] Rodríguez et al. (2014).

presented lower mercury levels (0.06–0.22 μg/g) than those (0.41–2.2 μg/g) of omnivorous or piscivorous fish such as *Geophagus surinamensis* or *Cichcla* sp. (Porvari, 1995; Palermo et al., 2004; Kehrig et al., 2008, 2009; Rodríguez et al., 2014). The latter specie, *Cichcla* sp., is the most studied fish in this region and it has been showed to be consistently contaminated with high levels of mercury (as high as 2.2 ± 0.96 μg/g), being mainly methylmercury (Porvari, 1995; Kehrig et al., 2008, 2009; Rodríguez et al., 2014). Interestingly, the *Cichcla* sp. always presented levels of mercury above 0.5 μg/g (the safety limit for human consumption according to FAO-WHO, 1991).

Regarding human exposure, Leino and Lodenius (1995) revealed 20 years ago that 45 people living at the main reservoir presented median levels of total mercury (up to 51 μg/g) in hair, well above the safe limit (10 μg/g) established by WHO (1990). However, no other evaluation with human populations has been carried out to date. Recent data suggested that high methylmercury (MeHg) contents (1.7 ± 1.3 μg/g) could be currently found in Tucunaré fish (*Cichla* sp.) from Tucuruí Lake, which is one of the most consumed fishes by the population (Rodríguez et al., 2014).

Therefore, this work evaluated the mercury content in the human population living near the Tucuruí Dam.

2. Methods

2.1. Ethical aspects

All participants were informed about the study and gave written consent. This project was in accordance with the Declaration of Helsinki and it was revised and approved by the National Council for Ethics in Research (CONEP, Brazil; CAAE n° 43927115.4.0000.0018).

2.2. Participants

After the announcing of our project by radio, adult participants from Caraipé attended at a meeting for sample collection. Exclusion criteria included smokers, alcohol drinkers (more than 200 mL/day), chronic diseases, drug dependency and long-term treatment with drugs, among other factors affecting mercury distribution and elimination. Inclusion criteria involved adults (18–70 years) of both sexes, fishing for a living, consumption of ≥ 7 meals of fish/week, a minimum of 2 years living at the selected area, with ≥ 1 cm of hair length.

2.3. Hair collection and analysis of mercury species

Hair samples (1 g approximately) were collected from the occipital region of the head (1–2 cm from the scalp) with clean stainless scissors and were stored in envelopes at room temperature. Mercury speciation analysis was performed in all samples based on a previously optimized procedure (Berzas Nevado et al., 2008). Briefly, mercury species (methylmercury, MeHg, and inorganic mercury, IHg) were simultaneously extracted from the hair (0.1 g) with 10 mL of 6 N nitric acid in a closed-vessel microwave system, and they were determined by derivatization with sodium tetraethylborate and injection into a home-made GC-pyro-AFS system previously described elsewhere (Berzas Nevado et al., 2005). Total mercury concentration was calculated as the sum of both MeHg and IHg species concentrations. The method was validated by the analysis of ERM-DB001 (human hair) certified reference material from the Institute of Reference Materials and Measurement (IRMM). Both total Hg and MeHg concentrations measured for ERM-DB001 (374 ± 15 and 254 ± 23 ng/g (n = 7), respectively) were in agreement with the certified concentration for total mercury (365 ± 28 ng/g, t-value = 1.469, P = 0.192, n = 7) or proposed concentration (240 ng/g, t-value = 1.668, P = 0.146, n = 7) at 95% confidence level. To assure the quality control of the analysis, a procedural blank was prepared in each batch of samples extraction. These blanks of extraction, which were analyzed periodically between samples and standards, were used for the correction of the chromatographic signals. The quantification of mercury species was conducted by using the "standard-sample-standard" bracketing technique based on the response factors obtained for standards injected between samples. Each sample was triplicate analyzed before and after a standard and the mean value and standard deviation were provided.

2.4. Statistical analysis

Gaussian distribution and homoscedasticity of data were tested by Kolmogorov-Smirnov and Bartlett tests, respectively. Data were then analyzed with a Student´s *t*-test (parametric) or Mann–Whitney (non-parametric) test to compare groups. Also, Pearson (parametric) or Spearman (non-parametric) tests were used to study possible correlations between variables. P-value was set at < 0.05.

3. Results and discussion

The reservoir of the Tucuruí Dam is divided into two compartments communicated by a strait: Caraipé (Fig. 1) and the lake (Bonetto, 1994). The population is widespread with only a few families living in each island with a life style similar to other isolated riverine communities in the Amazon: the main occupation is fishing; fish is the main protein source of the diet; they have limited access to health care and a high prevalence of endemic diseases such as malaria; sanitary conditions are very poor (no electricity, piped water, or sewage system; water for cooking is collected from the river or hand-dug wells and trash is burned, buried, or dumped in the river).

In this work, Student *t*-test was used to analyze the parametric data of age and body mass index of the participants and Mann-Whitney test was used to verify possible differences in the non-parametric data of mercury levels between sexes. The data of participants were uniform without significant differences in age, body mass index and mercury levels between sexes (Table 2). Our work is in accordance to those that described no correlation between mercury content in hair and sex or age. This finding seems to be usual in adults exposed to high levels of mercury (Pinheiro et al., 2006; Barbosa et al., 2001; Pinheiro et al., 2008; Marrugo-Negrete et al., 2013; Hoshino et al., 2015). Differently, significant correlations between levels of mercury and age or sex are more frequently found in children (Pinheiro et al., 2007; Kim et al., 2008; Marrugo-Negrete et al., 2013; Marinho et al., 2014).

The region of Caraipé was selected because of its higher retention time of water (about 130 days), compared to that of the lake (30 days) (Bonetto, 1994), which could contribute to mercury accumulation. Interestingly, participants of this work were highly selected with rigorous exclusion/inclusion criteria (which are not always present in studies of human exposure), guaranteeing that mercury levels were not significantly influenced by altered hepatic and/or renal functions. There was no correlation of sex or age with total mercury concentrations (Mann-Whitney and Spearman tests, $P > 0.05$), revealing a homogenous pattern of exposure in our sample.

No significant difference was detected between means of total mercury levels of this study (20 ± 14 μg/g) and those (31 ± 19 μg/g) reported 20 years ago for the same region of Caraipé (Leino and Lodenius, 1995; see also Table 3). Still, the median levels of total mercury (Table 2) were apparently lower than those shown in the latter study (23 μg/g), maybe because of the different sample size (more than three times greater in our study). However, other possibilities must be not discarded (for example, the higher proportion of men in Leino and Lodenius´ work could be responsible for the higher mean because of the trend of men for higher accumulation as showed in Table 2). Still, the medians of both total mercury and MeHg presently detected were above the limit recommended by WHO (WHO, 1990). Still more worrying is the fact that a substantial number of participants (19%) showed high levels of total mercury with values 40–75 μg/g, being MeHg about 90% of the total mercury content (Table 2). The samples of all participants of

Table 2
Methylmercury (MeHg) and total mercury (Hg) contents in hair and anthropometric data of participants of this study. All samples were analyzed for both MeHg and inorganic mercury, and total mercury was the sum of these two values.

		Total	Sex		Difference between sexes	
			Male	Female		
N (%)		37 (100)	18 (48.6)	19 (51.4)	Parametric Student *t*-test	Non-Parametric Mann-Whitney test
Age (years)		43.0 ± 2.4	46.1 ± 3.7	39.6 ± 2.8	P > 0.05	–
Weight (kg)		66.8 ± 2.8	77.2 ± 3.3	57.0 ± 3.2	***	–
Height (cm)		158.5 ± 1.7	166.7 ± 1.3	150.8 ± 1.9	**	–
BMI		26.30 ± 0.85	27.6 ± 1.4	25.03 ± 0.83	P > 0.05	–
Total Hg (µg/g)	Median (Interquartile)	12.0 (7.9–23.8)	19.7 (6.2 − 47.6)	11.0 (8.0 − 18.0)	–	P > 0.05
	Min - Max	1.1 − 75.8	1.1 − 75.8	3.6 − 42.9		
MeHg (µg/g)	Median (Interquartile)	10.9 (7.0–22.2)	18.2 (5.7 – 43.0)	10.0 (7.1 − 37.7)	–	P > 0.05
	Min - Max	0.93 − 69.2	0.93 − 69.2	3.2 − 37.7		

Data are showed as mean ± SEM except for mercury levels that are presented as median and interquartile intervals. BMI = Body Mass Index. ***P < 0.001 and **P < 0.01, male *vs* female.

this study were assayed for both MeHg and IHg contents. Proportion of MeHg varied between 71% and 97%, with a mean value of 89.6 ± 2.20%. MeHg is the most toxic compound of mercury species with high capacity to biomagnify throughout the food chain, as piscivorous fishes (as Tucunaré) are a major vehicle for human exposure. In humans, the brain is the main targeted organ for MeHg. Alterations in motor performance, visual dysfunction, genotoxicity, disturbances of the immune system, alterations of blood pressure, and oxidative stress were already described in Amazonian populations that were chronically exposed to relatively low levels of MeHg (Berzas Nevado et al., 2010).

In this study, 57% of participants showed ≥ 10 µg/g of total mercury in hair with a median value above the limit (20 µg/g) (Fig. 2) proposed by Harada et al. (2001) to detect symptoms and cases of suspected and mild Minamata disease in chronically exposed Amazonian populations. In this group, four women of fertile age (18–40 years old) showed MeHg levels as high as 37.3 µg/g. The possibility of altered neurodevelopment in the fetus with long-term consequences is a serious concern in MeHg-exposed women. Prenatal exposure to MeHg causes altered neuropsychological outcomes as those showed in the Finger Tapping test (Grandjean et al., 2014).

Also, a significant group (30% of all participants) showed ≥ 20 µg/g with a median value of 42.90 µg/g (Fig. 2). Thus, we recommended an urgent biomonitoring of the total region of the reservoir to establish adequate strategies of intervention and prevention.

Surprisingly, these levels are relatively higher than those currently shown in human populations highly influenced by *garimpos* areas where large amounts of mercury were used such as in the Tapajós River basin (Berzas Nevado et al., 2010) (Table 3), although no evident anthropogenic source of mercury exists in Tucuruí. Increased levels of mercury were already shown in Amazonian rivers with a reduced influence of *garimpos* (Wasserman et al., 2003). The presence of mercury was already attributed to atmospheric deposition, as in the Balbina Dam (Kehrig et al., 1998). Amazonian soils may show high mercury content with more than 97% of the mercury accumulated at the soil surface being pre-anthropogenic (Roulet et al., 1998). Although the geological origin of natural increased levels is controversial (Nriagu et al., 1992; Porcella, 1994; Lacerda, 1995; Kehrig et al., 1998), it is recognized that the Amazonian environment has the potential to release and accumulate significant amounts of this metal in response to anthropogenic actions other than gold mining (Wasserman et al., 2003; Berzas Nevado et al., 2010).

Table 3
Total mercury (Hg) content in hair of populations from Tapajós River basin (Jacareacanga, area influenced by *garimpos*) and from Tucuruí dam (Caraipé), detected in 1995 and 2010 or 2015. Data are showed as mean ± SEM. Student´s *t*-test.

Year	Area of study	Total Hg (µg/g)	n	Differences Tapajós *vs* Tucuruí	References
1995	Tapajós basin	24.6 ± 17.8	27	P > 0.05	Akagi et al.,1995
	Tucuruí dam	31 ± 19	11		Leino and Lodenius (1995)
2010	Tapajós basin	13.7 ± 10.8	108	P < 0.0001	Grotto et al. (2010)
2015	Tucuruí dam	20 ± 19	35		This work

In the case of Tucuruí Dam, some limnologic characteristics may favor the presence of mercury in the environment (Table 1). Inundated area occupies 2875 km^2 but only 464 km^2 were deforested before the filling of the dam (Espindola et al., 2000; Fearnside, 2001). The vegetation in decomposition makes the water became acid and anoxic (Fearnside, 2001), being the ideal conditions for bacterial proliferation and mercury methylation. Different of other dams in Amazon (such as Balbina or Samuel dams), the trophic stage of the aquatic environment in Tucuruí is mesotrophic and the maximum depth is 75 m (Espindola et al., 2000). The greater depth of Tucuruí as compared to other dams in Amazon could also contribute to the present of anoxic hypolimnion with higher mercury levels.

Interestingly, environmental data seems to support this hypothesis of a higher contamination in Tucuruí as compared to other dams or impacted regions in Amazon. For example, methylmercury concentrations of *Cichla* sp. (one of the most consumed fish by Amazonian populations) from Balbina dam were significantly lower as compared to those found in Tucuruí region (0.48 ± 0.37 µg/g and 1.7 ± 1.3, respectively) (Kehrig et al., 2008; Rodríguez et al., 2014), pointing to a higher contamination of the latter environment.

Future studies analyzing environmental samples (water, soils, and plants) will give a better idea of the dynamics and the possible source of mercury in Tucuruí. Based on our data, we strongly recommend a complete ecological study of the environment.

Environmental studies about the impacts of the dams throughout the world are numerous, especially in South Africa, China and Amazon (see Winemiller et al., 2016, for an overview). However, studies evaluating human exposure to mercury are extremely scarce. For example, in 2007, 86 adults living at the region around the Inanda Dam in South Africa were evaluated for mercury content in hair (Papu-Zamxaka et al., 2010). Thirteen years after the discontinuation of the activities of a mercury processing plant located near the dam, levels of mercury in hair of inhabitants revealed that 17% of the participants were contaminated with > 7 µg/g (Papu-Zamxaka et al., 2010), demonstrating the bioaccumulation and biomagnification properties of mercury in the environment. In our work, the prevalence of exposed participants and the maximum levels of mercury detected in humans were higher than those of the latter study. Our data also revealed mercury levels above

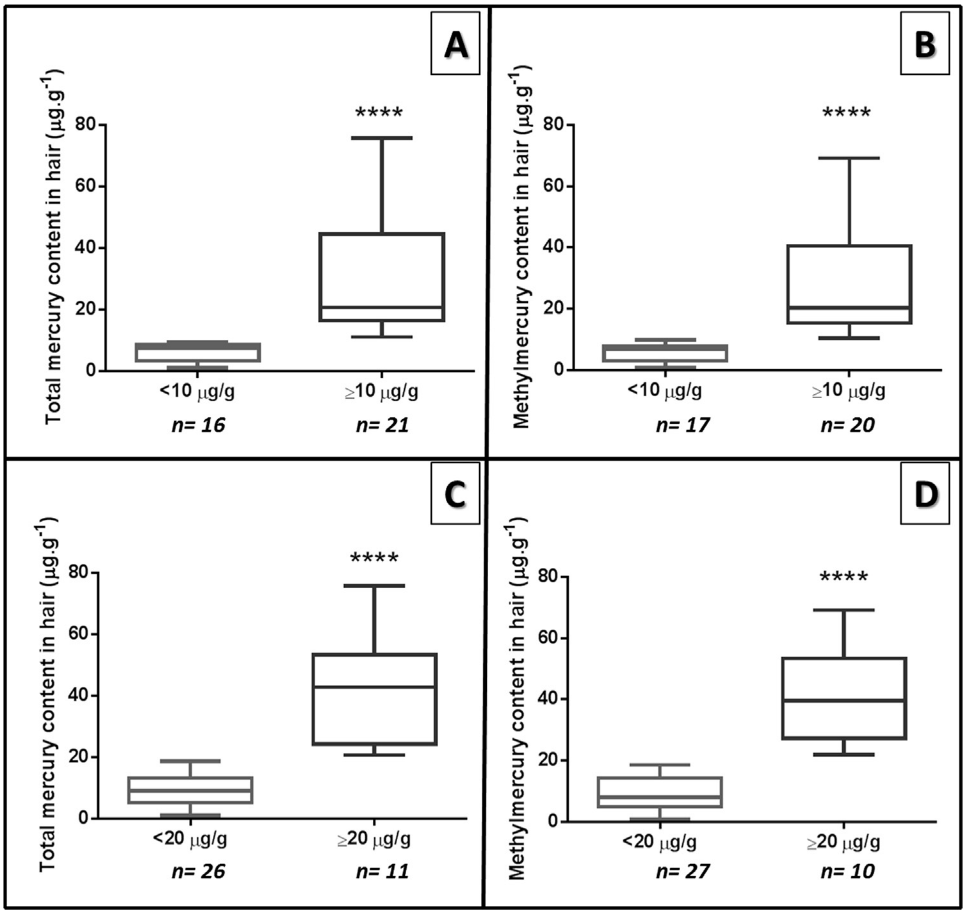

Fig. 2. Median and interquartile ranges of total mercury (A and C) or methylmercury (B and D) content in hair of participants showing levels above and below 10 µg/g (A and B) or levels above and below 20 µg/g (C and D). Mann-Whitney test. ****P < 0.0001.

the values detected in other study recently carried out in South America (not Amazonian region) with 76 individuals (24 children and 52 adults) living at the Urrá Dam in northwestern Colombia (Marrugo-Negrete et al., 2013).

Unfortunately, there are few works determining the possible human exposure of the Amazonian population surrounding large-scale projects with the potential to dramatically accumulate mercury in the environment. An interesting study on the Samuel Dam (Madeira River basin, Brazilian Amazon) found no correlation between altered neurodevelopment of young children and mercury content (Marques et al., 2011). However, in that study, no participant showed levels of mercury above the WHO limit of 10 µg/g, and median levels of total mercury (3.95 µg/g; minimum of 1.04 and maximum of 9.22) were about three times lower than those detected in our study. Inhabitants (adults and children) of another Amazonian dam, the Balbina Dam (Uatumã River basin), were also evaluated for mercury exposure (Kehrig et al., 1998), but they also showed mean levels (6.54 ± 5.45 µg/g) about three times lower than the mean levels (20 ± 14 µg/g) found in Tucuruí in our study.

To our knowledge, inhabitants of the Tucuruí Dam seem to show the highest levels of total mercury ever detected in human populations living near an Amazonian dam.

Moreover, areas downstream of the dams sometimes showed even higher levels of MeHg than those detected upstream (Kasper et al., 2014), probably because the anoxic conditions (caused by the release of hypolimnetic waters through turbines) may favor bacterial proliferation (mainly responsible for biotransformation of inorganic to organic mercury). This hypothesis is mainly supported by the different accumulation of mercury found in piscivorous fish upstream and downstream from the Amazonian dams such as Balbina, Samuel or Tucuruí dams (Palermo et al., 2004; Kasper et al., 2012, 2014). For example, in Tucuruí, piscivorous fish upstream and downstream from the dam showed 0.234 and 0.860 µg/g of mercury in muscle, respectively (Palermo et al., 2004). Previous data also demonstrated that mercury concentrations in sediments, plankton and plants (*Eichhornia crassipes* and *Scirpus cubensis*) collected downstream from the dam were approximately three times higher than those detected upstream (and especially in Caraipé) (Aula et al., 1995; Malm et al., 2004). In addition to these data, our results support the necessity to urgently analyze the exposure of populations living at regions downstream of the Tucuruí Dam. This continuous monitoring of the Tucuruí populations will assist in the development of prevention strategies and government actions to face the problem of the impacts caused by the dam.

Although additional studies are necessary to confirm the possible biomagnification and bioaccumulation of the presence of mercury by the dams in the Amazon, our data already support the necessity of adequate impact studies and continuous monitoring. More than 400 hydropower dams are operational or under construction in the Amazon, and an additional 334 dams are presently planned/proposed (Winemiller et al., 2016). Rigorous biomonitoring is especially important for the dams that are being built in areas highly influenced by *garimpos*; for example, two dams (Jatobá and São Luiz do Tapajós) are currently planned in the Tapajós River basin, downstream of the area of *garimpos* and near Jacareacanga, a region already contaminated (Table 3) and showing populations that already present symptoms of mercury intoxication (reviewed by Berzas Nevado et al. (2010)). An increase of mercury content is usually detected in the local biota after a reservoir impoundment. Inundation causes the mobilization of mercury and organic matter from submerged vegetation and soil. The forest remnants that are submerged (organic matter) suffer microbial decomposition and this fact, in addition to limnological characteristics such as acid pH and low oxygen levels (Kasper et al., 2014), favors the mercury methylation by methanogenic Archaea, allowing the bioaccumulation through food chain.

If we detect exposed populations after the impoundment of Amazonian reservoirs with reduced influence of anthropogenic mercury, the impact of these future projects in areas already contaminated, such as those presently influenced by *garimpos,* will be probably higher.

4. Conclusions

Our study found alarming levels of both total mercury and MeHg contents, above the limit recommended by the WHO, in inhabitants near the Tucuruí Dam. Based on these data, we recommend future studies: 1) a large monitoring of the population living near the reservoir and in regions downstream of the dam; 2) a complete ecological study of the environment to analyze the possible source of mercury; 3) a neurodevelopmental evaluation of young children to detect possible alterations. These future studies will permit the development of prevention strategies and government actions for Tucuruí and to learn from the example of this dam for the future large-scale projects planned or under construction in the Amazon.

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Conflict of interests

Authors declare that no conflict of interests exists. The funders had no role in study design, data collection and analysis, decision to publish, or preparation of the manuscript.

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