MERCURY AS AN ENVIRONMENTAL PROBLEM: HUMAN HEALTH RISK AND AQUATIC ECOSYSTEMS CONTAMINATION ASSESSMENT

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INTRODUCTION

Once released to an aquatic system, Hg may continue to cycle between sediments, water and biota for tens or even, hundreds of years before finally being flushed from the system, or permanently buried in sediments. The impact of mercury pollution, initially recognized as an acute and local problem, is now also understood to be global, diffuse, and chronic (Mahaffey, 2001). Environmental methylmercury (MeHg) arise largely, if not solely, from the methylation of inorganic mercury (Hg^{2+}). MeHg is listed by the International Program of Chemical Safety (ICPS) as one of the six most dangerous chemicals in the world's environment. The general population is primarily exposed to MeHg through fish consumption (WHO, 1990; Clarkson, 1994; US EPA, 2001).

In the Brazilian Amazon gold mining ("garimpos"), Hg is used to amalgamate fine gold particles from placer deposits. Hg loss from gold mining to local ecosystems was estimated to reach 1,300 t in the amazon, between 1980 and 1993 (Cid de Souza and Bidone, 1994). More than 50% of this Hg were used in "garimpos" locate in Tapajós river, in Pará State, mainly by garimpos of "Reserva garimpeira de ouro do Tapajós" as large as $28.000km^2$. It was the most important gold mining area in Northern Brazilian Amazon in the 80's when the peak in Amazon "gold rush" occured. Nowadays, the gold production is close 6 tons/year and the gold mining population are around 6.000 people in this region. Generally, the released Hg0 by "garimpos" is incorporate into the river sediments. Amazonian environmental conditions might favor methylation processes (Lacerda and Salomons, 1998).

The common dominator to managing human exposures is control of the use and release of inorganic mercury (Mahaffey, 2001). Currently there is a lack of consensus in the literature as to the importance and magnitude of several potential sources of Hg in Amazon. It is difficult to assess the major Hg source to Amazon aquatic

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ecosystems, since they integrate basin sources and direct and indirect atmospheric deposition. Atmospheric deposition can affect remote sites from sources, then affecting areas far from direct emission, which can be considered background. Major sources of Hg in the Amazon include biomass burning (Veiga et al., 1994), natural degassing (Roulet and Lucotte 1996) and gold mining (Lacerda, 1997), but we have gold mining as the principal one. Our works have been demonstrated significant differences considering Hg levels in fish from a direct influenced gold mining area (contaminated area) and a non influenced area (non contaminated area) (Castilhos et al., 2000; Castilhos et al., 2001), as well as the human health risks associated with Hg contaminated fish consumption (Bidone et al., 1997; Castilhos et al., 1998).

Socio-economic costs derived from the toxicological risks associated to this contamination should be taken into consideration, including its impact on the economic perspectives of a given region. In the case of the Amazon region, on major potential impact is on fish farming (Castilhos et al., 1998).

HUMAN HEALTH RISKS: MERCURY EXPOSURE DUE TO FISH CONSUMPTION BY AMAZON POPULATION FROM TAPAJÓS RIVER REGION

Different chemical forms of mercury have quite different metabolism and toxic effects. Thus when evaluating risks to human health, each of the various chemical forms must be considered separately. The potency to produce irreversible brain damage and teratogenic effects makes MeHg the specie of mercury of greatest public health concern (Clarkson, 1994). There are many scientific publications about this subject, so a briefly description is shown below and emphasis is done in MeHg toxicity and human health risks assessment.

Most of the mercury encountered in the atmosphere is elemental mercury vapor and it is readily absorbed through the lungs, goes directly to the Central Nervous System and may be distributed throughout the body. Occupational exposure in humans indicates that neurotoxicity is the adverse effect most likely to occur at lowest exposure level (LOEL) and usually it is reversible. Elemental mercury is

categorized as Group D, unable to be classified as to human carcinogen, according to USEPA Guideline for Carcinogen Risk Assessment (USEPA, 1986).

Inorganic mercury is generally absorbed by gastrointestinal tract. Its sentinel toxic end point is kidney damage mediated through an autoimune effect. (Schoeny, 1996). There are no data on the carcinogenic effects of inorganic mercury in humans, but it has been classified as Group C, possible human carcinogen, according to USEPA Guideline for Carcinogen Risk Assessment (USEPA, 1986), because there are some evidences from animal studies.

Methylmercury (MeHg) is rapidly and extensively absorbed through the gastrointestinal tract; once absorbed it is widely distributed in the body and across blood-brain and placental barriers. There are ample data from human and animals to consider MeHg to be a development toxicant. The most important toxic effect is on the nervous system. Neurological abnormalities have been observed in humans exposed as adults or "in utero". The developing fetus is at greater risk from MeHg exposure than are adults. In addition, children are considered to be at increased risk of MeHg exposure by virtue of their greater food consumption as a percentage of body weight (mg food/kg body weight) compare to adults exposures. Additional risk from higher mercury ingestion rates may also result from the apparent decreased ability of children's bodies to eliminate mercury. MeHg appears to be clastogenic but not to be a point mutagen; that is, mercury causes chromosome damage but not small heritage changes in DNA. EPA has classified MeHg as being of high concern for potential human germ cell mutagenicity; but the data are not sufficient to permit estimation of the amount of MeHg that would cause a measurable mutagenic effect in the human population. EPA has found MeHg to have inadequate data in humans and limited evidence in animals and has classified it as a possible human carcinogen, Group C, and has not calculated quantitative carcinogenic risk values for MeHg. Given the levels of exposure most likely to occur in the U.S. population, even among consumers of large amounts of fish, MeHg is not likely to present a carcinogenic risk (Schoeny, 1996).

It has been demonstrated that Hg usually accumulates in fish tissues as MeHg, from inorganic Hg sources (Huckabee et al., 1979). The absence of a consistent relationship between Hg concentrations in

water, sediment and various fish species illustrates the complexity and site-specific nature of mercury bioaccumulation. Thus, direct Hg determinations in the local biota appear to be crucial to adequately evaluating Hg sources, and, ultimately, the risk of the Hg exposure to human health (Peterson et al., 1996).

In a previous study, we reported on the Hg concentrations in the fish fauna and on the corresponding potential human exposure to Hg due to fish consumption, in a contaminated and non contaminated section of the Tapajós river basin. The contaminated site is located in the Tapajós river between the cities of Jacareacanga and Itaituba, into which the gold mining sites are distributed alongside the tributaries of the Tapajós river. The background site is located in a fluvial lacustrine system near Santarém, 250 Km downstream the contaminated site, which does not receive contamination from the site, but has the same basic characteristics. We sampled and analyzed 541 specimens from 22 fish species: 238 from contaminated area and 303 from noncontaminated area. They include representatives of about 85% of the species caught and commercialized in the study areas (Isaac and Ruffino, 1995). The catch and market show a relationship 1:1 between carnivorous and noncarnivorous species in this region. The same relationship is thus assumed for human consumption.

Mercury was analyzed in the fish muscle through Atomic Absorption Spectrophotometer (A-G/VARIAN MODEL) using a Vapor Generation Acessory-VGA (CVAAS). The samples were diggested in sulfuric-nitric acid solution in the presence of vanadium pentoxido 0.1%; the oxidation completed by adding potassium permanganate 6% until the fixation of the violet color. Immediately before the determination, the excess of permanganate was reduced with hydroxylamine 50% (Campos 1990). Reference standard IAEA-fish muscle tissue with a certified Hg concentration of 0.74 ± 0.13 $\mu g.g^{-1}$ were also analyzed, giving a value of 0.73 ± 0.08 $\mu g.g^{-1}$ (n=4).

The "fish enrichment factors" for Hg were calculated by equation FEF= (Hg $_{contaminated\ site}$ - Hg $_{background}$) / Hg $_{background}$, showed that the contaminated site is enriched *vis-a-vis* the background location for total fishes, noncarnivorous fish and carnivorous fish from 0.5, 0.6 to 0.8 (or plus 50%, 60% and 80%), respectively and the differences between means are statistically significant (Student's t-test; p<0.001). The results are shown in Table 1.

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Table 1. Fish Hg mean concentration (μg.Kg^{-1}) in carnivorous species and non carnivorous species from the background location and from the contaminated site and the "fish enrichment factor", FEF values for the study area; (n) = number of samples.

Food Habit	Hg mean concentration (μg.Kg^{-1})		FEF
	Background site (n)	Contaminated site (n)	
Carnivorous	228±171 (159)	420±230 (98)	0.8
Non carnivorous	39±47 (144)	62± 53 (140)	0.6
Total	138±159 (303)	210± 240 (238)	0.5

We used a screening approach to compare the potential human health hazard between areas (US EPA, 1989). Although this assessment may be simplistic, in particular for MeHg, it allows easy comparison between populations under different levels of exposure to a given pollutant. This method permits that the differences between contaminated and background area must be applied so that the toxicological - rather than simply the statistical - significance of the contamination can be ascertained. The knowledge of background (i.e., pre-impact or "natural") environmental conditions permits the establishing of physical standard reference of the environmental quality.

Risk assessments recommended by US EPA follow the paradigm established by the National Academy of Sciences. This entails a series of interconnected steps including hazard identification, dose-response assessment, exposure assessment, and risk characterization, which are briefly described in this paper. Hazard identification uses available data on biological end points related to a material to determine if that material is likely pose a hazard to human health; these data are also used to define the type of potential hazard. In the dose-response assessment, data from human and animal studies are used to estimate the amount of material that is expected to produce a given effect in humans. In this step it is generally necessary to apply mathematical models to the data to calculate a quantitative risk estimate usable for low-dose response, resulting in reference doses (RfD) and slope factors (SF) for noncarcinogenic and carcinogenic effects respectively. The exposure assessment seeks to

determine the extent to which a population is exposed to the material and uses available data relevant to population exposure. Fate and transport of the material in the environment, routes of exposure, and pharmacokinetics of material once in the body may be considered in the exposure assessment. Risk characterization is the last step of the risk assessment process. This step evaluates assessments of human health and ecological effects and delineates areas of uncertainty, limitations and assumptions made in the risk assessment.

At a screening level, a Hazard Quotient (HQ) approach, assumes that there is a level of exposure (i.e., RfD = Reference of Dose) for non-carcinogenic substances below which it is unlikely for even sensitive populations to experience adverse health effects. HQ is defined as the ratio of a single substance exposure level (*E*) to a reference of dose (E/RfD). When HQ exceeds unity, there may be concern for potential health effects. The estimated exposure level was obtained by multiplication of 95th percentil upperbound estimate of mean Hg concentration considering all fish samples (156.0 μg.kg^{-1} for background location and 240.0 μg.kg^{-1} for contaminated site) – as suggested by US EPA (1989) – by the adult human ingestion rate for riverside populations (0.2 Kg.d^{-1}) that consume more fish and therefore, the most harmful situation, and divided by 70 kg, considering the weight average human adult. The MeHg RfD value is 1 E-04 mg.Kg^{-1}.d^{-1} (IRIS, 1995) and its uncertainty factor is 10 and its confidence level is high. The resultant MeHg HQ is 4 and 7 for the background and for contaminated sites, respectively. These results suggest the need for further research on the potential health hazard from MeHg exposure in local population, even for what is considered here as background exposure. The results are shown in Table 2.

Hair concentrations of Hg are proportional to blood concentrations at the time of the formation of the hair strand. The estimates of Hg concentration in blood and in hair in contaminated and in noncontaminated sites were done by using the single-compartment model (WHO, 1990), through which the steady-state Hg concentration in blood (C) in μg.L^{-1} is related to the average daily dietary intake (d) in μg of Hg, as follows: C = 0.95 * d. The estimated hair Hg concentration (11.4 μg.g^{-1}) agree with the observed 16.6$\pm$10.5 μg.g^{-1} total Hg concentration and the observed 15.2$\pm$10.5 μg.g^{-1} MeHg concentration reported by Akagi et al. (1994) in hair samples from 48 individuals from contaminated site of the Tapajós river. The chemical Hg speciation in

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hair samples indicated that ~ 88% of the total Hg concentrations were MeHg. The total Hg in hair reported by Akagi et al. (1994) could be related to ~ 65 μg.L^{-1} in blood, using the single-compartment model. This value agree with the data reported by Cleary (1994) in blood from 12 residents of a fishing village of Jacareacanga (74.8 $\pm$ 61.0 μg.L^{-1}). The results are shown in Table 2.

Table 2. Hg concentration in fish (μg.g^{-1}); estimated intake (D; μg.kg^{-1}.d^{-1}); Reference Dose (RfD); Hazard Quotient (HQ); estimated average Hg daily intake (d; μg.d^{-1}); estimated blood Hg concentration (b; μg.L^{-1}) and estimated hair Hg concentration (h; μg.g^{-1}).

Location	Hg in fish* (μg.g^{-1})	D (μg.kg^{-1}.d^{-1})	RfD (μg.kg^{-1}.d^{-1})	HQ	D (μg.d^{-1})	B (μg.L^{-1})	H (μg.g^{-1})
Background	0.16	0.4	0.1	4	31	29.5	7.3
Contaminated site	0.24	0.7	0.1	7	48	45.6	11.4

* 95 percent upper confidence limit on the arithmetic mean

This report is of screening level, and uncertainty remains as to the health effects of eating large quantities of contaminated fish in the area studied, however our results agree with WHO recommendation: "measure to reduce methylmercury exposure via consumption of fish will need to consider the impact of these measures on the overall dietary requirements of these individuals", in view of the importance of fish consumption for the local population, particularly significant in the absence of any other abundant food resource. As a general rule, it is advisable to start the assessment with the "worst case" study; for any given environmental risk, we must assume the worst and then attempt to prove that a better situation exists (Wilson, 1991).

Also, uncertainties of the RfD statistics have been reported, suggesting an under-estimation of RfD for Hg presented in IRIS, 1995 (Smith and Farris, 1996). However, currently neurodevelopment problems in children secondary to maternal MeHg consumption during pregnancy are considered to be the most sensitive indicators of adverse effects of MeHg exposures. The oral RfD (1 E-04 mg.Kg^{-1}.d^{-1}) is associated with a maternal whole blood mercury concentration of 5.8 μg.L^{-1}. Although more protective than some other recommendation, this RfD has been found to be based on credible science by the United

States National Academy of Sciences Committee on the Toxicology of Methylmercury. Most recently additional findings describing adverse cardiovascular and immunological effects of low-dose MeHg exposure suggest that these organ systems are at least as sensitive, and possibly more sensitive, than the developing nervous system.

Table 3 shows the toxicity values derived by US EPA (2001) for elemental, inorganic and organic mercury, mainly MeHg, concerning to human exposure and critical adverse health effects. The toxicity values, presented here, are Reference Concentration (RfC) and Reference Dose (RfD) for elemental, inorganic and organic mercury, respectively. The modified factors (MF) and uncertainty factors (UF) for each toxicity value express the confidence level in them and are estimated depending on the quality of data base available.

Table 3. Toxicity values (RfD and RfC) for mercury species concerning to human exposure and critical adverse health effects.

Species	Critical effects	UF	MF	BMDL (NRC)	RfD ($\mu g.kg^{-1}.d^{-1}$)	RfC ($\mu g/m^3$)
Elemental	Neurological effects* (adults)	30	1			0.3
Inorganic	Autoimmune effects	1000	1		0.3	
Organic (MeHg)	Neurological effects (child)	10	1	58 ppb	0.1	

UF= uncertainty factor; MF= modifying factor; NRC- National Resource Council; BMDL Benchmark lower limit dose related to MeHg in cord blood; *The critical effects related to human exposure to elemental mercury: hands tremor; increases in memory disturbances; slight subjective and objective evidence of autonomic dysfunction.

Recent studies have examined populations that are exposed to lower levels of MeHg due to fish consumption, including studies of populations around the Great Lakes, the Amazon basin (Câmara and Corey, 1992; Câmara et al.,1993; OPAS/OMS, 1996), the Seychelles Island and the Faroe Island (USEPA, 1997). The last two studies are of large populations of children presumably exposed in utero. The results from the Seychelles study of fetal MeHg exposure and child development, involving a main results of 779 infant-mother pairs highlights the difficulties in interpreting epidemiologic studies of this

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type (Mayers et al. 1995; Mayers et al., 2001). Maternal total hair mercury values during pregnancy ranged from 0.5 to 26.7ppm with a median of 5.9 ppm. This value is close with those estimated for hair from the local populations in the background site.

Significant uncertainties remain, however, because of issues related to exposure, neurobehavioral endpoints, confounders and statistics, and study design. Additional data are needed on the exposure levels at which humans experience subtle, but persistent, adverse neurological effects. Data on immunologic and reproductive effects are not sufficient for evaluating of low-dose MeHg toxicity for these end-points (US EPA, 1999).

In addition, one should take in account that epidemiological control, which may represent a useful "feed-back" for the adjustment of preventive measures in the case of slight and reversible pathologies due to environmental impacts, has no preventive value in the case of irreversible damages (Zapponi, 1988). It has been expanded recognition of the range of adverse effects of MeHg on human and animal health, and the results have been a marked change in interpretation of what degree of mercury exposure is considered excessive. This parallels broad changes in approach by public and environmental health authorities: emphasis on primary prevention rather than only treatment of disease (Mahaffey, 2001).

MERCURY IN FISH: SPATIAL AND TEMPORAL ENVIRONMENTAL CONTAMINATION ASSESSMENT FOR DECISION-MAKING AND PROPOSED METHODOLOGY

Aquatic organisms accumulate MeHg from water, food and sediment. Both dissolved, inorganic Hg and MeHg accumulate in phytoplankton. However, in contrast to MeHg, inorganic Hg is not biomagnified as the trophic transfer from phytoplankton to zooplankton. At the base of the food chain MeHg typically constitutes a smaller percentage of Hg pool, but within the fish community, virtually all the Hg is MeHg. In addition, MeHg attains its highest concentration in the tissues of fish at the top of aquatic food chain, the carnivorous fish. Elimination of MeHg by fish is very slow relative to the rates of uptake and the accumulation. The direct bioaccumulation factor (BAF) or bioconcentration factor (BCF) of Hg is defined as the ratio of Hg

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concentration in fish tissue to the Hg concentration in water. The indirect bioaccumulation or biomagnification is the accumulation of a chemical in a given species according to its trophic levels in the food chain (Bruggeman, 1982).

Castilhos & Bidone (2000) have characterized the Hg biomagnification in the amazon ichthyofauna from the Tapajós River Region, suggesting a general trophic sequence as: herbivorous = or < detritivorous < omnivorous < planktivorous < carnivorous omnivorous = carnivorous ichthyophagous.

Bioaccumulation and/or biomagnification are the most direct study of mercury reaction at the environmental interface of the aquatic organisms. In environmental hazard analysis, the ecological effect of a specific substance on an organism or group of organisms in an aquatic system can be described as a function of the dose during a defined period of time. Although the bioaccumulation/biomagnification processes have not been traditionally interpreted as a pharmacological/toxicological "effect", one could suggest that as higher the internal dose (bioaccumulation), the higher is the potential aquatic risk. Moreover, several effects show a positive correlation with exposure but a causal-effect relationship is a difficult task to access, but bioaccumulation/biomagnification processes, which have to have, at least, exposure conditions for a determinant chemical agent. In this particular context, the ecological effect is defined as Hg-content in fish. In addition, the US EPA concluded that it is more appropriate, at this time, to derive a fish tissue (including shellfish) residue water quality criterion for MeHg rather than a water column-based water quality criterion. This new criterion is based directly on the dominant human exposure route to MeHg, and the resulting Fish Residue Criterion (TRC) is 0.3 mg MeHg/kg fish. This is the concentration in fish tissue that should not be exceed based on a total fish and shellfish consumption-weighted rate of 0.0175 kg fish.d-1 (US EPA, 2001).

In general, the Hg levels in fish muscles show large inter-individual variability, resulting in very high values for relative standard deviation (US EPA, 1999). Many factors have been considered as important in the bioaccumulation and/or biomagnification of Hg in fish. Among them, the Hg load-dependent factors in the aquatic environment, specially those related to Hg in sediments and environmental conditions, like bio-production (Häkanson, 1980; 1991);

as well as local biota's physiological-dependent factors, like size, length, age and metabolic rate (Phillips, 1980; WHO, 1990); and, in addition, the food-chain characteristics (Cabana et al, 1994). So, Hg levels in fish for spatial and/or temporal comparisons have been normalized by the average Hg content in 1 Kg of fish (as pike) (Johnels et al., 1967; Häkanson, 1991), by using only fish of one year old (Post et al, 1996), or a specific length (Scruton, et al., 1994), or a specific weight (Watras et al., 1998).

Castilhos et al (2001) suggested a field dose-response approach as a tool for assessing the environmental Hg contamination, through the mercury analysis in fish. Dose-response approach is usually used for analysis of data from laboratory or epidemiologic studies. However, we have proposed its use for assessment of Hg contamination in fish sampled from field. This methodology was used to assess the Hg contamination in fish from Tapajós River Region. The Tucunaré (*Cichla* spp) specie was chosen for many reasons. At the moment, there are few toxicokinetics studies from field or laboratory-controlled conditions about Hg in *Cichla* spp., but this specie may be considered good bioindicator of Hg accumulation in the Amazonian ecosystem, specially because of its time-integration capacity. According to the reproductive strategies, carnivorous icthyophagous *Cichla* spp could be classified as "in equilibrium" (Winemiller, 1989 cited in Ruffino & Isaac, 1995). The fish considered "in equilibrium" are the most sedentary and present a territorial behavior. Their density does not change strongly during the year. Spawning season is long and it is not necessarily at the beginning of the flood time. Their preferred habitat is lentic (slow moving) water. The influence of amazonian seasonality (well characterized by two hydrological periods: a low waterlevel and a high waterlevel period) on Hg accumulation in fish was studied and the results showed that carnivorous fish, including *Cichla* spp, were not affected by seasonality (Castilhos, 1999). Their fine taste and abundance in native habitat have made it an important commercial specie (Ruffino and Isaac, 1995). Also, Tucunaré was reported as most frequently consumed fish by indigenous community in Pará State (Brabo et al., 2000).

The results showed different daily uptake doses by Tucunaré (*Cichla* spp.) between non-contaminated and contaminated areas (~4.0 times), and such differences could be attributed to different Hg loading

rates between the studied areas, and also, could be a consequence of a potential pollution source, the gold mining activity.

We have also tested the applicability of the proposed methodology (DRAC) by using the literature data (Castilhos and Lima, 2001). Seven species of fishes from 8 different aquatic ecosystems, including lentic and lotic freshwater, estuarine and marine ecosystems, that were chosen from scientific publications, in which Hg levels in muscles and estimated age or measured length for individual specimens, were available.

The "DRAC" methodology was described in a previous work (Castilhos et al., 2001), and a brief description is presented as follows. The dose-response relationship has the competence to absorb inter-individual variability. The responses are of two kinds: quantal and quantitative. The quantal test are designed to estimate the concentration of a test material that affects 50% of the test organisms, the mean effective dose (ED 50% or ED50). One must choose the effect to be observed. Thus, this is a quantal rather than a graded response, since the specific effect is either present or absent. The ED50, for accumulation of Hg by fish, indicate the exposure time necessary to attain those tissue concentration levels by half of the exposed individuals. Some methods are used to calculate ED50, and, among them, there is the "probit" method (American Public Health Association, 1985; Ross and Gilman, 1985).

The D50 for accumulation of Hg by fish (accumulation dose 50 or AD50) indicates the exposure time necessary to attain those tissue concentration levels by half of the exposed individuals. This resulting time can be related to response, as follows: $t_{exposure} * C$ = constant (adapted from Dämgen and Grünhage, 1998); in which a certain response (K, constant) can be achieved from a exposure time ($t_{exposure}$) and the concentration in the aquatic environment C; such concentration will result as a potential dose or daily uptake rate (DUR), expressed in $\mu g.Kg^{-1}.d^{-1}$. From these results one could estimate the exposure time to reach either 300 $\mu g.Kg^{-1}$ or 500 $\mu g.Kg^{-1}$ and compare the contamination magnitude (or bioavailability) among different aquatic ecosystems.

The objectives were: (i) to establish and compare the dose-response relationship for Hg accumulation by different fish species, from several ecosystem and collection time (ii) to estimate and compare the daily Hg uptake rate by those different fishes; (iii) to

estimate and compare the potential exposure time necessary for Hg accumulation to reach $500\mu g.Kg^{-1}$, the limit concentration for human consumption, adopted in many countries, and, (iv) to present this approach, in addition to that expressed by U.S. EPA, for water quality criteria for methyl-mercury.

The results are presented in Table 4, which displays the popular name of the fish*,* the local and time of sampling, food habit, number of collected specimens, observed effect (OE), daily uptake rate estimate (DUR), estimate of time of exposition to reach $300\mu g.Kg^{-1}$ and $500 \ \mu g.Kg^{-1}$, respectively, and references. Comparing the OE values (mean Hg concentration in $\mu g.kg^{-1}$) in the population sampled, for piscivorous and zooplanktivorous fishes, one could suggest the following crescent order of contamination or crescent contamination magnitude/bioavailability: Sepetiba Bay (corvina) < Ilha Grande Bay (corvina) = Guanabara Bay (corvina) = lagoons-Santarém (tucunaré) < Conceição Lagoon (corvina) < Tongue River- reservoir (white crappie) < Tongue River-reservoir (walleye) = Tongue River-reservoir (pike) = Tapajós River (tucunaré) < Tongue River- reservior (sauger) < Gulf of Trieste (conger-conger) < Tucuruí River- reservoir (tucunaré). However, considering the daily uptake rate estimates and/or the time necessary for half of the specimens to reach $500\mu g.Kg^{-1}$, the order of contamination is altered to: Tongue River-reservoir (white crappie) < Conceição Lagoon (corvina) < Ilha Grande Bay < Guanabara Bay < Sepetiba Bay < Tongue River-reservoir (pike = sauguer= walleye) = lagoons-Santarém (tucunaré) < Tapajós River (tucunaré) < Gulf of Trieste (conger-conger) < Tucuruí River-reservoir (tucunaré).

Considering TRC, sauger (Tongue River), tucunaré (Tucuruí Reservoir) and conger-conger (Gulf of Trieste) showed Hg levels above the limit. However, looking at the estimated daily uptake rates, northern pike, sauger and walleye (Tongue River) and tucunaré (lagoons=Santarém) showed the same value: $0.2\mu g.kg^{-1}d^{-1}$. The estimated daily uptake rates for conger-conger (Gulf of Trieste), tucunaré from Tapajós River and tucunaré from Tucuruí Reservoir resulted in 2 times, 4 times and 8 times higher than $0.2\mu g.kg^{-1}d^{-1}$, respectively. The zooplanktivorous fish from Tongue River reservoir became, as expected, the less contaminated specie compared to carnivorous and piscivorous fish. For the Tucuruí system the possible differences in the tucunaré growth rate should be investigated, since

the literature data indicate the existence of dwarf species in reservoirs (Doyon et al., 1998).

The objective was to test the applicability of the dose-response approach as a tool for environmental assessment mercury contamination by using the literature data. Indeed, the results might not represent, actually, the magnitude of mercury contamination/availability among those sites, because there are significant differences in temporal sampling as well as in analytical procedures.

We suggest that, in addition to TRC (0.3 mg methylmercury / kg fish) for specific fish specie, one could estimate the daily uptake rate, which may express the bioavailability of mercury in a defined aquatic ecosystem, and compare the time necessary to attain the TRC value. The dose-response approach might permit an integrate comparison among different aquatic ecosystems using the same or different fish species. The DRAC is a simple and fast methodology and can be applied to any data bank including spatial and temporal contamination assessment for environmental contaminants that show bioaccumulation.

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Table 4. Popular and scientific names, food habit, locality, date and number of fish collected (N), observed effect (OE; μg.Kg^{-1}), estimated daily uptake rate (DUR; μg.Kg^{-1}.d^{-1}), estimated time of exposure to attain either 300μg.Kg^{-1} (T300, years) or 500μg.Kg^{-1} (T500, years) and references.

Popular name	Scientific name	FH*	Locality	Date	N	OE (μg.Kg^{-1})	DUR (μg.Kg^{-1}.d^{-1})	T300 (years)	T500 (years)
Northern Pike	*Esox lucius*	P	Tongue River - Reservoir (EUA)[1]	1978	56	300	0.2	4.1	6.8[5]
Sauger	*Stizostedion canadense*	P	Tongue River - Reservoir (EUA)[1]	1978	31	350	0.2	4.1	6.8[5]
Walleye	*Stizostedion vitreum*	P	Tongue River - Reservoir (EUA)[1]	1978	26	300	0.2	4.1	6.8[5]
White crapie	*Pomoxis annularis*	Z	Tongue River - Reservoir (EUA)[1]	1978	36	200	0.05	16.4	27.0[5]
Corvina	*Micropogonias furnieri*	C	Guanabara Bay (RJ, BR)[2]	1992	56	100	0.15	5.5	9.1[6]
Corvina	*Micropogonias furnieri*	C	Ilha Grande Bay (RJ, BR)[2]	1990-1991	57	100	0.13	6.3	10.5[6]
Corvina	*Micropogonias furnieri*	C	Sepetiba Bay (RJ, BR)[2]	1990-1991	60	80	0.10	8.2	13.7[6]
Corvina	*Micropogonias furnieri*	C	Conceição Lagoon (SC, BR)[2]	1990-1991	42	130	0.08	10.3	17.0[6]
Tucunaré	*Cichla spp*	P	Lagoons –Santarém (PA, BR)[3]	1992	28	100	0.2	4.1	6.8
Tucunaré	*Cichla spp*	P	Tapajós River (PA, BR)[3]	1992	41	300	0.8	1.0	2.0
Tucunaré	*Cichla spp*	P	Tucuruí River - Reservoir (PA, BR)[4]	1995	61	1000	1.65	0.5	0.8[5]
	Conger conger	P	Gulf of Trieste (Slovenia)[7]	1995-1996	25	610	0.4	2.1	3.4

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