Environmental Research 150 (2016) 438–445

Contents lists available at ScienceDirect

Environmental Research

journal homepage: www.elsevier.com/locate/envres

Waterscape determinants of net mercury methylation in a tropical wetland

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ARTICLE INFO

Article history:
Received 19 February 2016
Received in revised form
30 May 2016
Accepted 17 June 2016

Keywords:
Periphyton
Radiotracers
MeHg
Landscape ecology

ABSTRACT

The periphyton associated with freshwater macrophyte roots is the main site of Hg methylation in different wetland environments in the world. The aim of this study was to test the use of connectivity metrics of water bodies, in the context of patches, in a tropical waterscape wetland (Guapore River, Amazonia, Brazil) as a predictor of potential net methylmercury (MeHg) production by periphyton communities. We sampled 15 lakes with different patterns of lateral connectivity with the main river channel, performing net mercury methylation potential tests in incubations with local water and *Eichhornia crassipes* root-periphyton samples, using $^{203}HgCl_2$ as a tracer. Physico-chemical variables, landscape data (morphological characteristics, land use, and lateral connection type of water bodies) using GIS resources and field data were analyzed with Generalized Additive Models (GAM). The net $Me^{203}Hg$ production (as % of total added ^{203}Hg) was expressive (6.2–25.6%) showing that periphyton is an important matrix in MeHg production. The model that best explained the variation in the net $Me^{203}Hg$ production (76%) was built by the variables: connection type, total phosphorus and dissolved organic carbon (DOC) in water (AICc=48.324, p=0.001). Connection type factor was the best factor to model fit (r^2=0.32; p=0.008) and temporarily connected lakes had higher rates of net mercury methylation. Both DOC and total phosphorus showed positive significant covariation with the net methylation rates (r^2=0.26; p=0.008 and r^2=0.21; p=0.012 respectively). Our study suggests a strong relationship between rates of net MeHg production in this tropical area and the type of water body and its hydrological connectivity within the waterscape.

1. Introduction

The conversion of mercury (Hg) to methylmercury (MeHg), one of its organic forms and a potent neurotoxin, is one of the key processes to access the potential contamination impact of the metal (Clarkson, 2002). The inorganic mercury (Hg^0 and Hg^{2+}) released into the aquatic environment is converted into MeHg by biotic and abiotic methylation processes (Weber, 1993; Barkay and Wagner-Dobler, 2005; Avramescu et al., 2011). The MeHg formed in this process can be transferred the aquatic food chain, bioaccumulate, biomagnify and reaches final consumers, including humans (Watras et al., 1998; Díez, 2009; Molina et al., 2010).

In tropical and some temperate environments, the root zone of floating macrophytes and its associated periphyton sustains very high net methylation rates, producing more MeHg than the sediments and water column (Cleckner et al., 1999; Correia et al., 2012; Lázaro et al., 2013). The classic definition of periphyton, and that we use in this work, is a community of microorganisms (microalgae, fungi, macroinvertebrates, and prokaryotes) joined and with organic and inorganic particles by an exopolysaccharide matrix (produced by microbial metabolism), attached to dead and living substrates in the aquatic environment (Costerton et al., 1995).

The periphyton supports a diverse bacterial community, including sulfate-reducing bacteria (SRB) (Achá et al., 2005) and methanogens (Hamelin et al., 2011). These microorganisms are considered as the organisms responsible for a significant fraction of MeHg production in aquatic ecosystems (Mauro et al., 2002; Achá et al., 2011, Achá et al., 2012; Si et al., 2015).

The aquatic habitats of tropical floodplain are mostly governed

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http://dx.doi.org/10.1016/j.envres.2016.06.028
0013-9351/

by the flood pulse, which strongly influences the hydrological connectivity (Junk et al., 2011). The latter defines networks of matter and energy flow through the patches of the wetland landscape/waterscape. Patches are discrete spatial units in landscape scale, surrounded by a matrix (pasture, forest, land, crops) and may be connected to corridors: elongated environments that connect patches (Forman and Godron, 1986). In the waterscape context, the rivers and streams forms the corridors and the lakes and flooded areas can act as patches. These patches and corridors form networks in the waterscape/landscape, mediated by the hydrological (or biological) connectivity between them (Cantwell and Forman, 1993). Spatial characteristics have been associated with methylmercury production in peatlands (Mitchell et al., 2008) and boreal wetlands soils (Tjerngren et al., 2012a, 2012b). Other important aspects of the Hg cycle have been related to spatial distribution (Liu et al., 2009), flood hydrology (Bradley et al., 2010) and anthropogenic landscape change (Golden and Knightes, 2011; Myers et al., 2014; Wang et al., 2014).

Despite such research efforts, little is known about the distribution of Hg methylation potentials in aquatic habitats of tropical floodplains and their surroundings. In this sense, this work aims to assess the importance of limnological variables, and to test the use of connectivity metrics of water bodies, in the context of patches, and of land use indicators as predictors of potential net MeHg production by periphyton communities in a tropical wetland waterscape (Guaporé River, Mato Grosso, Brazil).

2. Methods

2.1. Study area

The Upper Guaporé River Basin is located in the northwestern part of the Mato Grosso state, Brazil. The Guaporé River is part of the Amazon Basin, being the major tributary of the Madeira River and a water/biological corridor between Amazonia and Pantanal (Ruffino, 2004). Along the course of the Guaporé River, there are several oxbow lakes that present different types of lateral connection to its main channel. The sampling campaign included 15 oxbow lakes along the Guaporé River and was done during the drought season to better document the contrasting lateral connectivity patterns. (Fig. 1). Each lake was sampled in four repetitions for all data, totaling 60 samples.. The lateral connections were classified as DC, directly connected (lake directly connected with the river during all the hydroperiod), indirectly connected (IC, connected during all the hydroperiod but mediated by a floodplain channel) and temporarily connected (TC, connected to the river during flood periods only) The delimitation of the lateral connection dynamics of each lake was based on the field monitoring of a complete hydrological cycle of the area.

2.2. Mercury methylation assays

The Hg methylation assays and analytical procedures followed Guimarães et al. (1995). Unwashed, freshly sampled *E. crassipes* roots with their associated periphyton (equivalent to 0.5 g dry weight) were suspended in 30 mL of local water, filteredin 48 μm porosity filters), sampled on the macrophyte bedsand incubated (24 h) "*in situ*" within the macrophyte beds in 50 mL screw cap glass tubes with 23,000 to 25,000 DPM. (disintegrations per minute) of ^{203}Hg added (1.2 ± 0.2 ng L^{-1}). as $^{203}HgCl_2$ The samples were pre-incubated with ^{203}Hg for 1 h to allow equilibration with dissolved ligands. Control samples as above were killed with 1 mL 4 N HCl.

All incubations were stopped by adding HCl (4 mol L^{-1}, 1 mL). Methylmercury was extracted by addition of 4 mL of NaBr (3 mol. L^{-1} in 11% H_2SO_4) and 1 mL of (0.5 mol L^{-1}) $CuSO_4$, with 15 min shaking and centrifugation. The supernatant was removed, and $Me^{203}Hg$ was extracted by shaking in separation funnels for 15 min with 30 mL of scintillation cocktail, consisting of POP (2,5-diphenyloxazole) and POPOP [1,4-bis-2-(5-phenyloxazolyl)-benzene] salts dissolved in toluene. The organic phase was removed and sodium thiosulphate added to remove any trace of water. $Me^{203}Hg$ was quantified by beta spectrometry in a Perkin Elmer

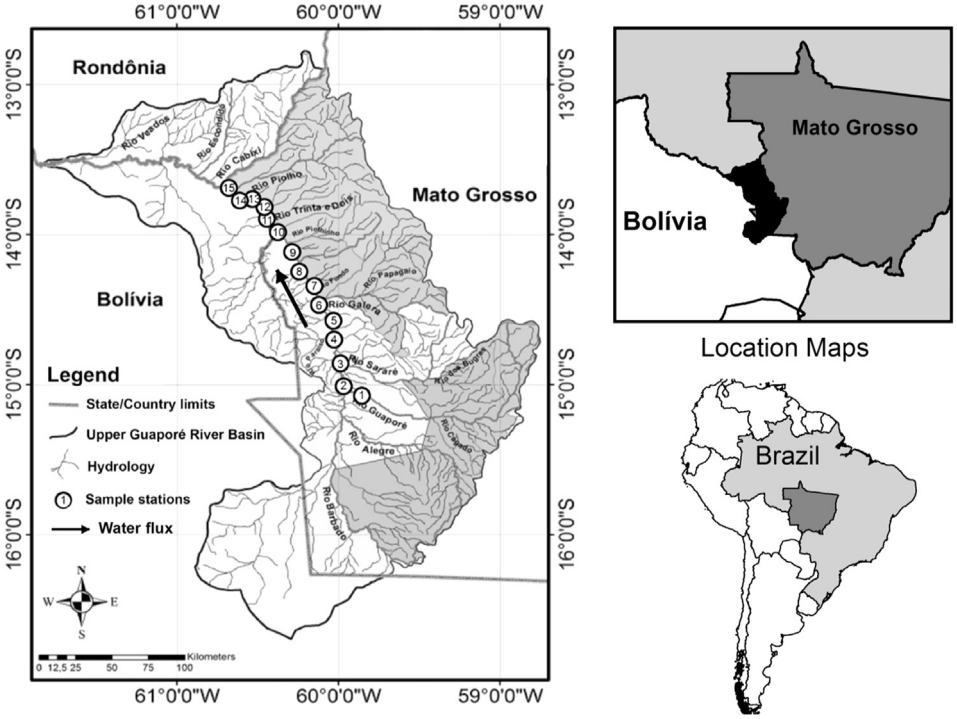

Fig. 1. Location map of the lakes sampled in the Guapore River floodplain, Amazonia, Brazil.