Science of the Total Environment 650 (2019) 394–407

Contents lists available at ScienceDirect

Science of the Total Environment

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

Conjunctive use of *in situ* gas sampling and chromatography with geospatial analysis to estimate greenhouse gas emissions of a large Amazonian hydroelectric reservoir

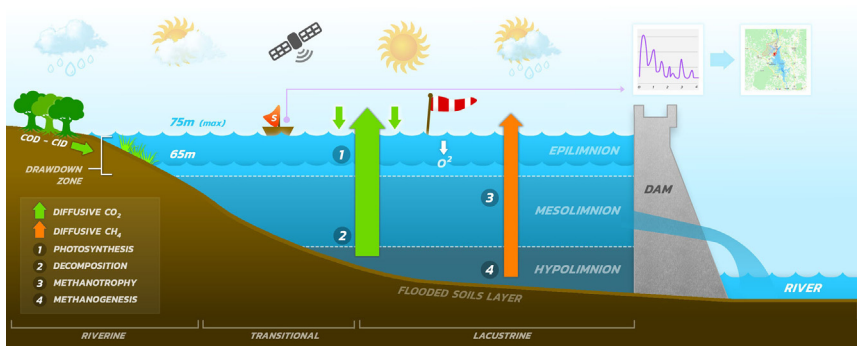
Isabel L. de Sousa Brandão [a,*], Chris M. Mannaerts [a], Isaque W. de Sousa Brandão [b], Joaquim Carlos Barbosa Queiroz [c], Wouter Verhoef [a], Augusto C. Fonseca Saraiva [d], Heronides A. Dantas Filho [b]

[a] Department of Water Resources, Faculty of Geo-Information Science and Earth Observation, University of Twente, Enschede, NL 7500, AE, the Netherlands
[b] Faculty of Chemistry, Federal University of Pará, Belém 66075-110, Brazil
[c] Geoscience Institute, Federal University of Pará, Belém 66075-110, Brazil
[d] Analytical Center Laboratory, Belém 66815-140, Brazil

HIGHLIGHTS

- We assessed CO_2 and CH_4 emissions for different seasonal and operating conditions of a large Amazonian impoundment.
- Emissions of the reservoir system were derived from in-situ sampling, laboratory chromatography and geospatial techniques
- Remote sensing data were used to extract the reservoir area and spatial interpolation for deriving the total GHG emissions
- The total diffusive emissions over a year, in units of tons of C, added up to 6.82 $\times 10^3$ for CH_4 and 1.19 $\times 10^6$ for CO_2
- We suggest to monitor emissions for longer periods to capture its temporal and spatial variabilities

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 18 May 2018
Received in revised form 27 August 2018
Accepted 28 August 2018
Available online 29 August 2018

Editor: Yolanda Picó

Keywords:
GHG fluxes
Tucuruí
Amazon

ABSTRACT

Hydroelectric power reservoirs are considered potential contributors to the greenhouse effect in the atmosphere through the emittance of methane and carbon dioxide. We combined *in situ* sampling and gas chromatography with geostatistical and remote sensing approaches to estimate greenhouse gas (GHG) emissions of a large hydropower reservoir. We used remote sensing data to estimate the water surface and geospatial interpolation to calculate total emissions as a function of reservoir surface area. The CH_4 and CO_2 gas concentrations were linearly correlated to sampling time, confirming the adequacy of the *in situ* sampling method to measure GHG diffusive fluxes from reservoir water surfaces. The combination of high purity (99.99%) ISO-norm gas standards with a gas chromatograph, enabled us to achieve low measurement detection limits of 0.16 and 0.60 μmol mol^{-1}, respectively, for CH_4 (using a flame ionization or FID detector) and CO_2 (using a thermal conductivity or TCD detector). Our results show that CO_2 emissions are significantly (an order of 5.10^2–10^3) higher than those of CH_4 in both the spatial and temporal domain for this reservoir. The total diffusive GHG emissions over a year (June 2011 to May 2012) of the Tucuruí hydropower reservoir being in operation, in units of tons of carbon, added up to 6.82

* Corresponding author.
E-mail addresses: i.l.desousabrandao@utwente.nl (I.L. de Sousa Brandão), c.m.m.mannaerts@utwente.nl (C.M. Mannaerts), isaque@ufpa.br (I.W. de Sousa Brandão), joaquim@ufpa.br (J.C.B. Queiroz), w.verhoef@utwente.nl (W. Verhoef), herondantas@ufpa.br (H.A. Dantas Filho).

https://doi.org/10.1016/j.scitotenv.2018.08.403
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Diffusive emissions
Spatial interpolation

$\times 10^3$ for CH_4 and 1.19×10^6 for CO_2. We show that *in situ* GHG sampling using small floating gas chambers and high precision gas chromatography can be combined with geospatial interpolation techniques and remote sensing data to obtain estimates of diffusive GHG emissions from large water bodies with fluctuating water surfaces such as hydropower reservoirs. We recommend that more measurements and observations on these emissions are pursued in order to support and better quantify the ongoing discussions on estimates and mitigation of GHG emissions from reservoirs in the Amazon region and elsewhere in the world.

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1. Introduction

Emissions of methane (CH_4) and carbon dioxide (CO_2) gases from hydroelectric reservoirs into the atmosphere form a growing concern among researchers around the world due to their potential to contribute to global warming (Huttunen et al., 2002; Kemenes et al., 2007; Soumis et al., 2004; St. Louis et al., 2000). Greenhouse gases (GHGs) and their greenhouse effect play an important role in Earth's climate as they are responsible for maintaining the temperature gradient in the atmosphere (Samimi and Zarinabadi, 2012). Carbon dioxide is distinguishable from other gases due to its ability to absorb and re-emit infrared energy, making it an effective heat-trapping greenhouse gas (Lacis et al., 2010; Somerville, 1998) and according to the European Environmental Agency (EEA, 2010) CO_2 emissions account for the largest share of GHGs, contributing 80–85% of emissions. Investigating methane emissions is also important, as reservoirs may create favourable conditions for the production of CH_4, which has a global warming potential of 34 times that of CO_2, considering the 100 time horizon years adopted by the Kyoto Protocol (Myhre et al., 2013).

Hydroelectric reservoirs have long been considered to be a source of "clean" GHGs-free energy (Barros et al., 2011). However, recently there is increased awareness that these reservoirs could be contributing to the increase of GHGs in the atmosphere (Deemer et al., 2016). Concerns have been raised around the world (IHA, 2010), also creating awareness in the Amazon region, where several studies and monitoring campaigns have taken place (Devol et al., 1990; Kemenes et al., 2007; Lima, 2005; Richey et al., 2002; Rosa et al., 2016; Rosa et al., 2004; Santos et al., 2008; Santos et al., 2006).

Studies undertaken in the Amazon region have revealed diverging results regarding GHGs emitted by amazonian reservoirs, as the variables that might be contributing to the total emission by reservoirs are partly unknown. Rosa et al. (2002), for instance, who conducted earlier studies on estimates of GHGs emissions using a model of plane geometry to calculate emissions in unit of area, understimated the total area of the Tucuruí Hydropower Reservoir (THR) by 15%, because they did not consider the variability occurring in the reservoir area during the year. An example of overestimation cited in the literature is the study conducted by Kemenes et al. (2007) in the Balbina reservoir, which found methane emissions exceeding those estimated by Rosa et al. (2002) by >20%. This was due to Kemenes et al. (2007) miscalculating the methane flux downstream of the Balbina reservoir. According to Santos et al. (2008), they should have based calculations on the mean variation of this flux in the water column. Instead, they used the maximum depth, where methane emissions were higher, overstimating results.

Deshmukh (2013) also conducted a study on estimates of GHGs in a subtropical reservoir, revealing that the diffusive emission from the water surface has been the main source of release of carbon dioxide (CO_2) into the atmosphere, namely 95% of total monthly emissions. A recent study, including a three-dimensional approach, conducted by Curtarelli et al. (2016) in the Tucuruí hydropower reservoir underestimated rather than overestimated the total emission by this reservoir. Despite the great achievements by these authors, the major drawback in their studies is not including the river downstream area or reference sites in their estimates, both necessary to allow emissions from a reservoir to be accurately estimated (IHA, 2010).

With the uncertainties surrounding these researchers' results, concerns arise regarding the models used to estimate their emissions (Parekh and Mccully, 2004; Santos et al., 2008) and whether hydropower reservoirs really provide "clean" GHGs-free energy. It is important to improve the available information regarding the GHG status of existing and new reservoirs and to provide tools to support decision making on mitigating measures that may be necessary, as stated in the guidelines for GHG measurement for freshwater reservoirs (IHA, 2010).

Despite progress made in GHG emission monitoring in amazonian reservoirs, more research is needed to provide a better understanding of the spatial and temporal distribution of GHG fluxes. Seasonal changes in climate, hydropower operations and carbon load may all affect spatial and temporal variations in GHG fluxes (IHA, 2010; Marcelino et al., 2015).

In this study we use an approach different from previous studies, which is based on a combination of *in situ* sampling with laboratory chemical analysis, geostatistics and remote sensing to model the spatial and temporal variations in CO_2 and CH_4 in the THR. Geostatistics, as a predictive tool, provides accurate and reliable estimations of phenomena at unsampled locations (Rivoirard, 2005).

The first step when applying geostatistic models is to determine the statistical relationship between sample points. In order to generate a surface model, some type of interpolation method must be used to estimate data values for locations where no samples were taken (Babish, 2000; Vieira and Paz Gonzalez, 2003). A fundamental tool in geospatial analysis is the variogram/semivariogram, which determines the amount of spatial dependency in the dataset (Spokas et al., 2003). The variogram/semivariogram is calculated based on the sampled sites. It is usually recommended that at least 100 data points are used to develop an accurate variogram. However, in many circumstances, like in GHG sampling and analysis, it is difficult to obtain a high enough data density to fully characterize the surface emissions of GHGs. This is due to practical constraints such as the sampling time needed, and the displacement time on large reservoirs, such as Tucuruí (Spokas et al., 2003). The optimal GHG sampling accuracy recommended by the International Hydropower Association (IHA) requires between 20 and 30 sample sites, including areas such as the main upstream tributary, the main reservoir body and the downstream area. This recommendation was adopted in this research.

Remote sensing can be used to monitor surface water extent and its variation using visible and infrared next to Synthetic Aperture Radar (SAR) observations (Alsdorf et al., 2007; Prigent et al., 2007). Estimations of surface water extent using visible and infrared satellite measurements can be attained using open access data at medium-high (*e.g.* 30 m) spatial resolution, but possibilities of detecting water surfaces under clouds or dense vegetation are limited (Papa et al., 2010). Interannual variability in methane surface emissions combined with water surface extent analysis has been studied by Bousquet et al. (2006), Papa et al. (2010), and Ringeval et al. (2010). The purpose of using remotely sensed water surface extent is to determine and monitor variations in the reservoir area in relation to the hydrological operation phases. It is already known that the surface area of this reservoir varies over the course of a year from 1500 km^2 to 2500 km^2 (Curtarelli et al., 2016).

The objective of this study was to evaluate a methodology, which may be used as input to the ongoing discussion regarding estimates of greenhouse gas emissions by hydroelectric power reservoirs in the Amazon region, as well as to provide information on the quantitative spatial

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