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CARBON DIOXIDE AND METHANE FLUX MEASUREMENTS AT A LARGE UNSANITARY DUMPING SITE IN THE AMAZON REGION

MEDIDAS DE FLUXO DE DIÓXIDO DE CARBONO E METANO EM UM DEPÓSITO DE RESÍDUOS INSALUBRE NA AMAZÔNIA

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ABSTRACT

Dumps are important anthropogenic sources of greenhouse gas emissions into the atmosphere, mostly CH_4. However, few studies on the subject have been carried out in the Amazon region. Several factors affect the production and emission of dumps gases. The objective of this study was to quantify the spatial variation of CO_2 and CH_4 production in an Amazonian dump and seek the relationship between the relative importance of some environmental factors and the gas fluxes. This study was carried out in an open-air dump in the metropolitan region of Belém, where approximately 11.0 million Mg of waste was deposited within 25 years, of which 6.4 million Mg were organic. The CH_4 and CO_2 emission rates from the surface of the dump were determined using the closed dynamic flux chamber technique. The study was conducted in three cells of different ages, sampled in two times between the rainy and the dry season in Amazon. The Aurá dump has an area of 30 ha and emits a total of 51.49 Mg CO_2 ha^{-1} $month^{-1}$ and 3.16 Mg CH_4 ha^{-1} $month^{-1}$ to the atmosphere. This results in an expressive production of 1,359,961.04 Mg CO_2-e y^{-1}, being that 58.54% is due to CH_4 flux. The spatial variability in CO_2 and CH_4 fluxes is very large, especially for CH_4, forming hotspots of high concentrations. Perhaps for this reason, the flow has not been correlated with micrometeorological variations.

Keywords: pollution; flux chamber; open dumping; Amazon.

RESUMO

Lixões são importantes fontes antropogênicas de emissão de gases de efeito estufa na atmosfera, principalmente CH_4. No entanto, poucos estudos sobre o assunto foram realizados na região amazônica. Diversos fatores afetam a produção e emissão de gás de aterro. O objetivo deste estudo foi quantificar a variação espacial da produção de CO_2 e CH_4 em um lixão da Amazônia e tentar associar a importância relativa de alguns fatores ambientais e os fluxos de gás. Este estudo foi realizado em um lixão ao ar livre na região metropolitana de Belém, onde aproximadamente 11,0 milhões de Mg de resíduos foram depositados em 25 anos, dos quais 6,4 milhões de Mg eram orgânicos. As taxas de emissão de CH_4 e CO_2 da superfície do aterro foram determinadas usando a técnica de câmara de fluxo dinâmico fechado. O estudo foi realizado em três células de diferentes idades, amostradas em dois momentos entre a estação chuvosa e seca da Amazônia. O lixão Aura tem uma área de 30 ha e emite um total de 51,49 Mg CO_2 ha^{-1} $mês^{-1}$ e 3,16 Mg CH_4 ha^{-1} $mês^{-1}$ para a atmosfera. Isso resulta em uma produção expressiva de 1.359.961,04 Mg CO_2-e ano^{-1}, sendo 58,54% devido ao fluxo de CH_4. A variabilidade espacial no fluxo de CO_2 e CH_4 é muito grande, especialmente para CH_4, formando pontos ativos ("*hotspots*") de altas concentrações, e talvez por isso, o fluxo não tenha sido correlacionado com variações micrometeorológicas.

Palavras-chave: poluição; câmaras de fluxo; lixão a céu aberto; Amazônia.

INTRODUCTION

Atmospheric methane (CH_4) concentrations have increased to over 1,800 ppb in 2016 (IPCC, 2013; REAY *et al*., 2018), of which 70% result from anthropogenic activities such as rice cultivation, domestic ruminants, biomass burning, leakage of natural gas, coal mining, landfills, and the remainder from natural wetlands (MATTHEWS; THEMELIS, 2007). Aerobic soil consumes and oxidizes atmospheric CH_4 (6% of total sink), while anaerobic soils can be a significant source of CH_4 (BIAN *et al*., 2018a; DALAL *et al*., 2008). The sink/source ratios and controls on the production and emission of CH_4 in the Amazon basin come mainly from studies on individual wetlands, lakes, and floodplains (POTTER *et al*., 2014). No studies have addressed how much Amazonian dumps produce and how long these deactivated dumps have contributed to global warming with carbon dioxide (CO_2) and CH_4 emissions.

Landfills are a significant global source of anthropogenic atmosphere CH_4 (BARLAZ *et al*., 2010) and a non-negligible source of CO_2 (AGAMUTHU, 2013). Global CH_4 emissions are responsible for approximately 40% of the global warming in the last 150 years (HANSEN *et al*., 2013), given that its global warming potential (GWP, molar basis, 100-year period) is about 21 to 27 times greater than that of CO_2 (AGAMUTHU, 2013; LELIEVELD *et al*., 1998). This is due to the high ultraviolet absorption coefficient and long residence time in the atmosphere (IPCC, 2013; LELIEVELD *et al*., 1998).

Currently, landfills contribute with about 22% of the total anthropogenic emissions of CH_4, which are expected to increase globally from 58 Mt to 365 Mt by 2030, assuming no further implementation of control measures (BAJAR *et al*., 2017). Current estimates from the Intergovernmental Panel on Climate Change (IPCC) for the annual CH_4 emissions from landfills range from 67 to 90 Mt CH_4 y^{-1}, which is equivalent to a CO_2 emission (CO_2-e) of 500 to 800 Mt CO_2-e (IPCC, 2013). Brazil has a considerable unexplored potential for landfill biogas production (LIMA *et al*., 2018), which is lost due to the lack of technology in the construction of landfills (AHOUGHALANDARI; CABRAL, 2017b; BARROS *et al*., 2018).

The characterization of landfill emissions is a complicated task, mainly because emissions are the result of a complex matrix of biological, physical and engineering factors (SPOKAS *et al*., 2003). These factors depend on parameters such as organic content, age and distribution of residues (GEORGAKI *et al*., 2008), climate (CHANTON *et al*., 2011), soil porosity, water content, nutrient availability, pH, texture, cracks and fissures (BOGNER *et al*., 2008; GEBERT *et al*., 2011). These factors are numerous and variable. Therefore, CH_4 emissions may exhibit prominent spatial and temporal variations (ABICHOU *et al*., 2011; GONZALEZ-VALENCIA *et al*., 2016; SPOKAS *et al*., 2003).

Landfill gases consist mainly of CH_4 (50–70% v/v) and CO_2 (30–50% v/v), nitrogen, hydrogen sulfide and non-methane hydrocarbons (SCHEUTZ *et al*., 2009). In Brazil, there are three main destinations for solid urban waste: landfills, controlled landfills and open-air dumps (LIMA *et al*., 2018). The biological process is commonly applied, for being a simple and economical approach and is often the only technique used in most municipalities (COSTA *et al*., 2019). Open-air dumps are the least recommended way to dispose solid waste as they have no cover layers, no leachate collection or treatment systems, and the gas produced is not used as an energy source (ABRELPE, 2016).

The objective of this study was to quantify both CO_2 and CH_4 production in an open-air dump (Aurá dump) that is located in the Amazon region and has emitted a total 9.4 to 9.8 Tg of CO_2 equivalent (IMBIRIBA *et al*., 2018) after it was closed, and to evaluate the relative importance of some environmental factors to gas surface fluxes, in both time and space. The main hypothesis is that there is a high production of greenhouse gases and that the substrate humidity and temperature would influence CO_2 and CH_4 fluxes, even assuming a high spatial variability.

MATERIALS AND METHODS

Study area

The Aurá open-air waste dump (1°25'19.04"S and 48°23'18.68"W) has an area of 30 ha (Figure 1) and began activities in 1987, receiving waste from the metropolitan area of Belém, which comprises the municipalities of Belém, Ananindeua and Marituba (estimated population of over two million people) (MATOS *et al*., 2011). The initial project included an incineration, and a recycling and composting plant. However, neither were implemented (SIQUEIRA *et al*., 2016). Therefore, all the solid wastes were deposited and distributed sequentially in layers, and compacted with track loaders, forming an open-dump with no environmental control and protection techniques. As such, this dump disrespects the technical specification of the Brazilian Association of Technical Standards (ABNT, 2010). Until now, leachate material infiltrates the soil or reaches the water resources through runoff, while all the gas produced escapes to the atmosphere.

The Aurá open-air dump received approximately 1,200 Mg of waste per day from 1989 to 2014, of which 58% was organic (SANTO, 2014). This is equivalent to approximately 11.0 million Mg of waste deposited in twenty-five years, of which 6.4 million Mg was organic. The deposition of domestic waste was forbidden on 2015, being allowed only the deposition of civil construction and urban cleaning waste. No soil

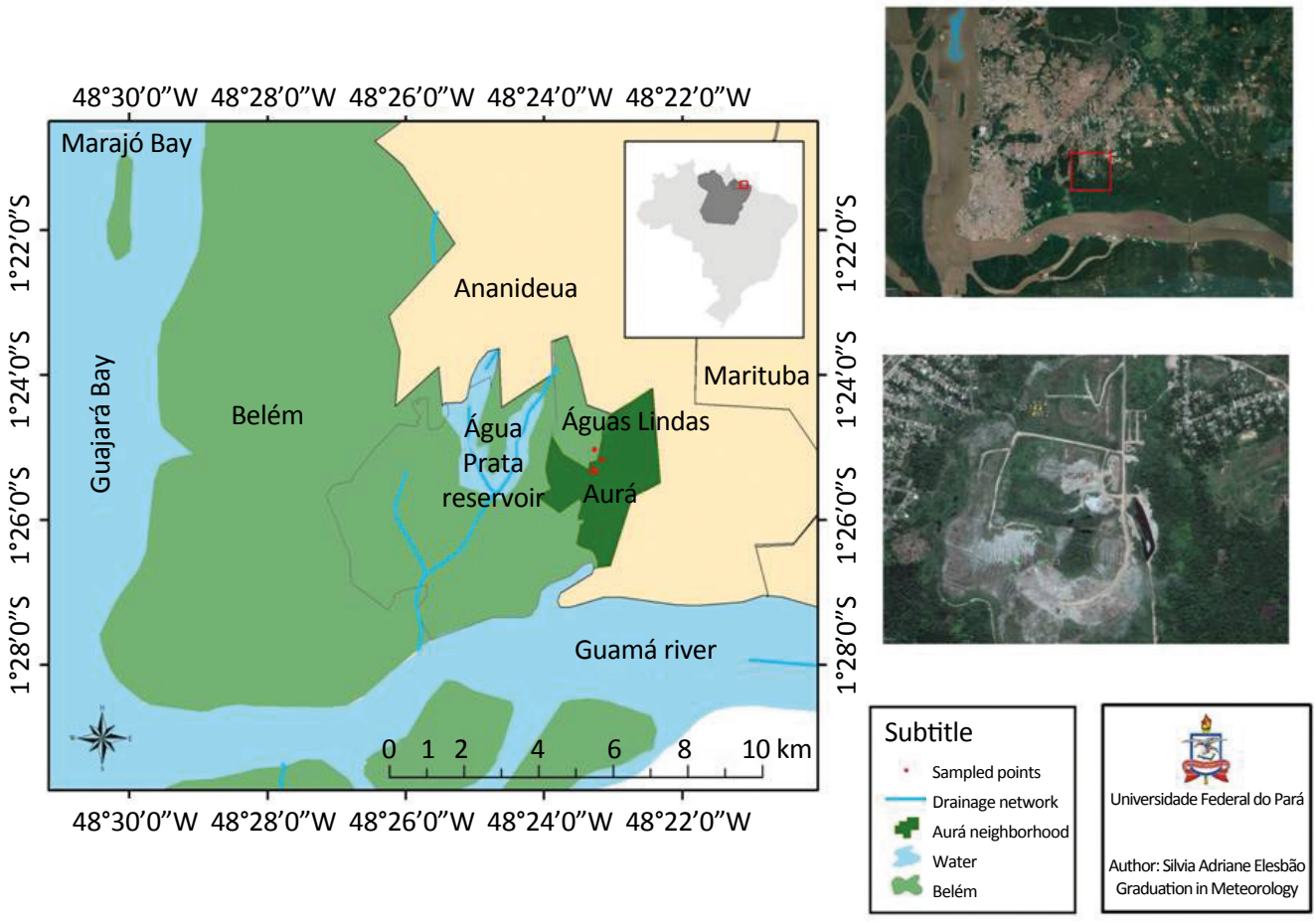


Figure 1 – Location of the Aurá open-air dump with the identification of the studied sites.

layer was placed over the waste, layer which could act as a reactive biological barrier, reducing CH_4 emission into the atmosphere (BÖRJESSON; SVENSSON, 1997). Thus, the soil in this study will be called substrate from here on.

The Köppen climate classification of the study area is *Afi*, with an annual average air temperature of 26.7°C, relative humidity of 84%, precipitation of 3,001 mm, and 2,338 hours of sunshine (BASTOS *et al*., 2002). There are two well-defined rainy periods, one is rainier (December to June), called here wet season, and the other is less rainy (July to November), called dry season.

In 2007, a biogas burning project, predicted to last 10 years, was established by Conestoga-Rovers and Associates (CRA). The landfill gas was captured using a technology that consists on a network of ducts and wells connected to a central ventilation system by vacuum induction. A total of 2,608,401.0 Mg CO_2-e (tons of carbon dioxide equivalent) was burnt from April 2007 to June 2016, according to a CRA report (CRA, 2006) available on the United Nations website on Certified Emissions Reductions (CER), and 139,092.0 Mg CO_2-e in the last CERs measurement (01/01/2016 to 06/30/2016). The most efficient systems are able to capture 75% of the biogas generated in a landfill (HASNAIN *et al*., 2012). However, in most cases, the efficiency ranges from 40 to 60% (BARLAZ *et al*., 2004). The measurements of CO_2 and CH_4 fluxes showed in this study were obtained in 2017, after the pipes used for conduction and flaring of the gases were removed.

Three different sites were selected to measure CH_4 and CO_2 fluxes. The municipal urban waste was deposited in the first site (S1, Figure 1) for a five-year period and street cleaning and commercial waste are currently deposited. At this site, there is no vegetation cover and the gas fluxes were measured on May 11th and June 8th, 2017. The second site (S2, Figure 1) is still receiving municipal waste, however in smaller amounts than when it was officially active. S2 is approximately 12 years old and has no vegetation cover, and the gas flux measurement was performed on June 29th, 2017. The third site (S3) was located in an older area (approximately 13 years old), where municipal waste was deposited until 2016. S3 is currently covered with undergrowth and was sampled on November 9th, 2017. The measurements at S1 and S2 were made in the middle of the site, forming a circle (10 m radius), where eight flow chambers (samples or point) were randomly distributed. A rectangular area of 16 × 22 m was delimited within S3. The chambers were placed every 2 m in each direction of the area, yielding of 88 samples.

Carbon dioxide and methane flux measurements

Emission rates of CO_2 and CH_4 from the surface of the open-air dump were determined using the closed dynamic flux chamber technique, which measures variation of gas concentration inside the chambers (NORMAN *et al*., 1997). The Ultra-Portable Greenhouse Gas Analyzer (Los Gatos Research, Mountain View, CA, USA) model 915-0011 was used for simultaneous measurements of CO_2, CH_4, and H_2O (MAHESH *et al*., 2015). Two devices were used for simultaneous measurements.

The chambers consisted of polyvinyl chloride (PVC) rings (diameter of 0.20 m and height of 0.12 m) and were inserted 0.05 m deep into the substrate at each sample location (within the sites). The rings that didn't pierce the substrate were placed on the surface of the dump and externally sealed with clay soil. Any vegetation found inside the chamber was previously removed. All rings were then closed with a PVC cap, forming a 4-liter chamber. An air circulation was established between the Ultra-Portable LGR analyzer and the flux chamber through polytetrafluoroethylene (PTFE) tubes using a vacuum circulation pump at a rate of 0.50 L min^{-1}.

CO_2 and CH_4 concentrations (ppmv) were recorded at 1 s intervals over a 3–4 min period. Fluxes were calculated from the rate of increase in concentration using the steepest linear portion of the accumulation curve as a function of time elapsed after the chamber was closed, adjusting to chamber volume and covered area, as proposed by Abichou *et al*. (2006). For a significantly non-zero flow, r^2 would have to be less than 0.3 (SUNDQVIST *et al*., 2014).

Environmental variables

Wind velocity (m s^{-1}), relative humidity (%), air temperature (°C) and barometric pressure were measured with an AK821 thermo-hydro anemometer at each flux measurement interval. Substrate humidity (%) was analyzed with a Soil Water Measurement System (Hydrosense; Campbell Scientific Inc.), and the substrate temperature (°C) was measured with a digital soil thermometer when the flow chambers were closed. The monthly rainfall and climatology data (1961–1990) were made available by the National Institute of Meteorology (INMET), which has an automatic weather station at a site relatively near the Aurá dump.

Geospatial analysis

Geostatistical analysis tools were used to evaluate the spatial variation of the carbon dioxide and methane fluxes and to detect spatial dependence. This analysis was performed at S3 with 88 sampling units distributed in a grid design (OPROMOLLA *et al*., 2006). The semi variance function is one of the tools in geostatistics most used to determine spatial dependence of a variable, generating a variogram (MELLO *et al*., 2005; OPROMOLLA *et al*., 2006). The variogram shows the spatial variability among the samples and the dependence level among the sites. A variogram $\gamma(h)$ describes the variance of the quadratic difference of a spatial variation between pairs of samples at distance h. Variograms were constructed, assuming isotropic spatial variation (i.e., independent of direction). In the absence of spatial dependence, i.e., in cases of large sample-to-sample variation at short distances, the variogram will show a nugget effect (OPROMOLLA *et al*., 2006).

Statistical analysis

Data normality was analyzed through the Shapiro-Wilk test, and the data were log-transformed when the residues did not present a normal distribution. The experiments were performed with at least eight chambers for each hour analyzed. In addition, 88 chambers were used in the geospatial (S3) analysis, as described above. The analysis of variance was used to assess the significance of the variation. When the differences were significant, the Tukey test was used to evaluate which samples differed from each other. Pearson's and Spearman's correlation were used to analyze the correlation between fluxes and environmental variables. Pearson's correlation evaluates the linear relationship between two continuous variables, while Spearman's measures the monotonic relationship between two continuous or ordinal variables, which tend to change together, but not necessarily at a constant rate. All analyzes were performed using the software InfoStat.

RESULTS AND DISCUSSION

Precipitation

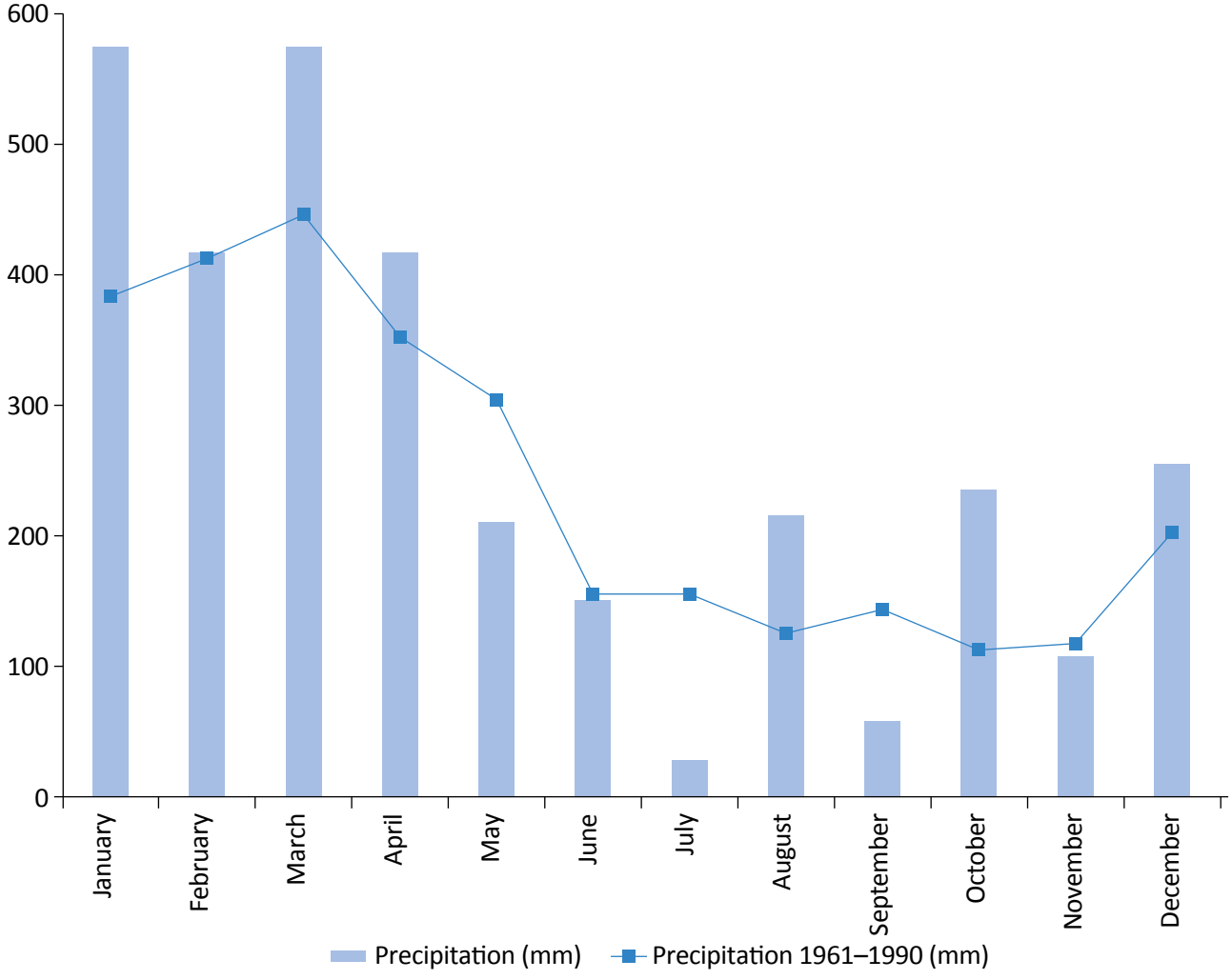
Precipitation in 2017 was 328.2 mm higher than the climatological average (1961–1990). The precipitation recorded for the months of May, June, July, September, and November was below the climatological average (Figure 2), and the remaining months exhibited above average precipitation records. Precipitation values when samples were collected were below the climatological average, and the highest variation occurred in May 2017, when the precipitation was 94.9 mm below the climatological average. The precipitation in the months of June and November 2017 was, respectively, 3.9 and 8.0 mm below the average.

An increased response time between a precipitation event and a change in the dump humidity may occur, given the direct relationship between landfill depth and response time (SCHEUTZ *et al*., 2017). These time intervals between humidity waves do influence the production of gas fluxes (RISK *et al*., 2008). In other words, when humidity decreases, the oxidative regions increase CH_4 production, with a consequent flux increase (TIAN *et al*., 2016; YANG; SILVER, 2016). In contrast, an increased CO_2 production is expected as humidity increases, with a consequent increased flow (DAVIDSON *et al*., 2000). The study was conducted at the end of the rainy season, and the beginning and the end of the dry season.

Carbon dioxide and methane fluxes at two sites that were simultaneously analyzed

CO_2 and CH_4 fluxes were measured at S1 on May 11th, 2017, where two areas were simultaneously analyzed (Figure 3). The distance between sites was approximately 30 m. The fluxes had a non-normal distribution ($p < 0.05$). Therefore, the data was log-transformed to carry out the statistical analysis, thus reaching statistical normality ($p > 0.05$) for the two gases investigated. The average CO_2 flux at S1 and S2 was 133.04 ± 51.47 g m^{-2} d^{-1} and 370.80 ± 184.84 g m^{-2} d^{-1} (mean ± standard error, n = 8), respectively. The mean CH_4 flow at the same sites was 40.00 ± 22.59 g m^{-2} d^{-1} and 77.32 ± 54.36 g m^{-2} d^{-1}, respectively. No significant difference ($p > 0.05$) was found between the two sites studied for either of the gases analyzed. Air temperature varied significantly ($p < 0.05$) in the first chamber, ranging initially from 33.97 to 36.26°C on the last measurement. The mean temperature was 35.19 ± 0.26°C, and the relative humidity and wind velocity were 89.13 ± 0.63% and 1.10 ± 0.43 m s^{-1}, respectively.

Both flux measurements showed large variability, with CO_2 fluxes ranging from 61.69 to 1,655.43 g m^{-2} d^{-1} (coefficient variation — CV = 140.99%), and CH_4 fluxes ranging from 2.77 to 455.95 g m^{-2} d^{-1} (CV = 67.04%). These results confirm that the surface emissions in



*Data provided by INMET.

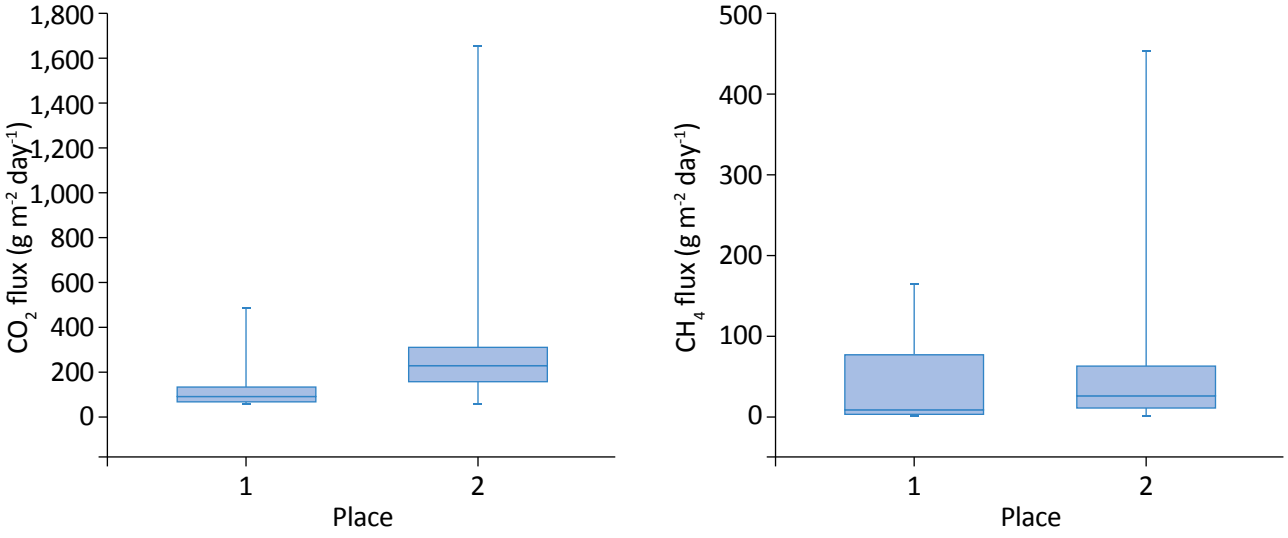
Figure 2 – Cumulative monthly precipitation for 2017 and climatological mean (1961–1990) at the metropolitan region of Belém*.

dumps are not uniform, with paths of lower resistance, creating hotspots (AHOUGHALANDARI; CABRAL, 2017b; ALLEN *et al*., 2019; GONZALEZ-VALENCIA *et al*., 2016; RACHOR *et al*., 2013). It is possible that emission areas have a higher air-filled porosity or improved pore connectivity compared to the larger dump area (BIAN *et al*., 2018b; RACHOR *et al*., 2013), resulting in preferred pathways for gases. The high CO_2 emissions in the Aurá open-air dump may be consequence of the ecosystem respiration, and aerobic decomposition of organic matter was as well as of the indirect CO_2 emissions generated by CH_4 oxidation (BIAN *et al*., 2018a; FJELSTED *et al*., 2019). The hotspots of CH_4 was the same for CO_2, meaning that the methanotrophic bacteria are possibly consuming CH_4 and producing CO_2 when CH_4 is transported from deeper layers to the surface (ROSLEV; KING, 1996). This can be confirmed because both CO_2 ($p = 0.0548$) and CH_4 ($p = 0.0402$) fluxes are negatively correlated with temperature (Figure 4).

Carbon dioxide and methane fluxes at different periods of the day

CO_2 and CH_4 fluxes were measured at the same site (S2, Figure 1), at different hours of the day, on June 8th, 2017. Samples were conducted at the end of the rainy season and the beginning of the dry season (Figure 2). The air temperature was significantly different for all measurement hours ($p < 0.01$), ranging from 37.55 ± 0.32 to 42.55 ± 0.07°C (Figure 5). The CO_2 fluxes measured at the site were 198.22 ± 20.17, 188.93 ± 25.94, 216.53 ± 48.14 and 222.40 ± 31.73 g m^{-2} d^{-1} for the hours of 10 a.m., 11 a.m., 12 and 12:30 p.m., respectively (Figure 5). CH_4 fluxes were 2.65 ± 1.46, 4.91 ± 1.92, 4.47 ± 3.34 and 2.99 ± 1.78 g m^{-2} d^{-1}, respectively, for the aforementioned hours. CO_2 and CH_4 fluxes didn't vary significantly ($p > 0.05$) among measurements.

Temperature was not correlated with either CO_2 or CH_4 fluxes ($p > 0.05$), despite the significant variation ($p < 0.05$) in air temperature observed among measurement hours (ABUSHAMMALA *et al*., 2013) (Table 1). Atmospheric pressure was 1013.0283 ± 0.0004 mb, and did not vary significantly ($p > 0.05$). Wind speed ranged from 1.51 ± 0.42 to 1.90 ± 0.46 m s^{-1}, and no significant difference was found ($p > 0.05$) among measurements (Table 1). The parameters analyzed were extremely homogeneous during the hours studied, except



* Each box represents eight chambers, and bars show the standard error of the mean. Vertical lines represent the distribution of the chamber values, and horizontal lines inside the gray box indicate the mean value. The box height indicates the standard deviation of the mean.

Figure 3 – The flow of carbon dioxide and methane measured simultaneously on both locations at S1 within the Aurá open-air dump, on May 11th, 2017*.

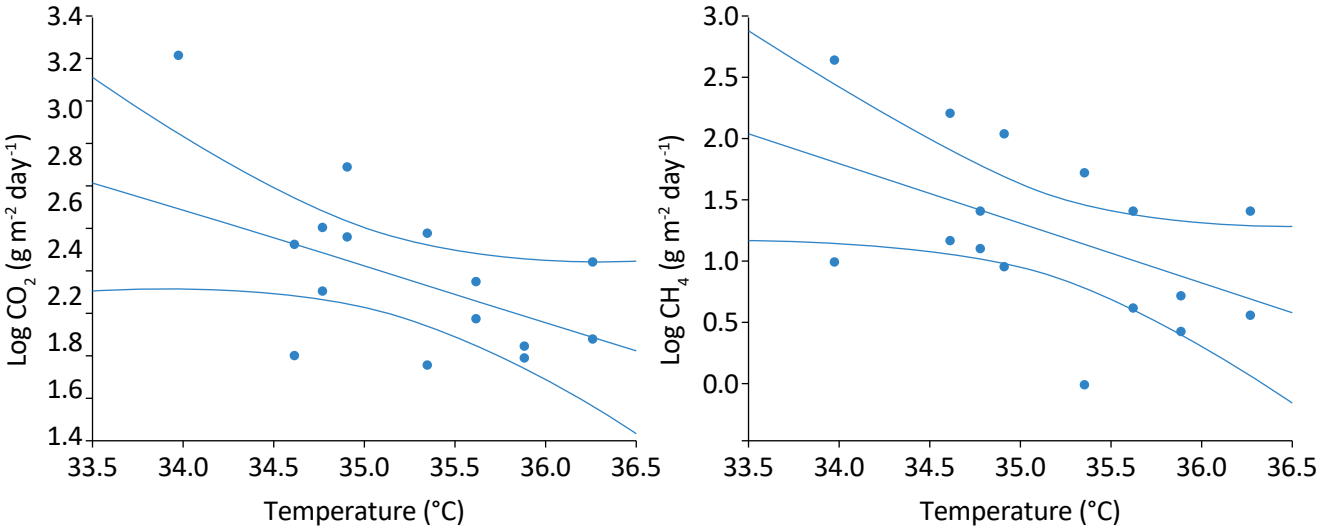
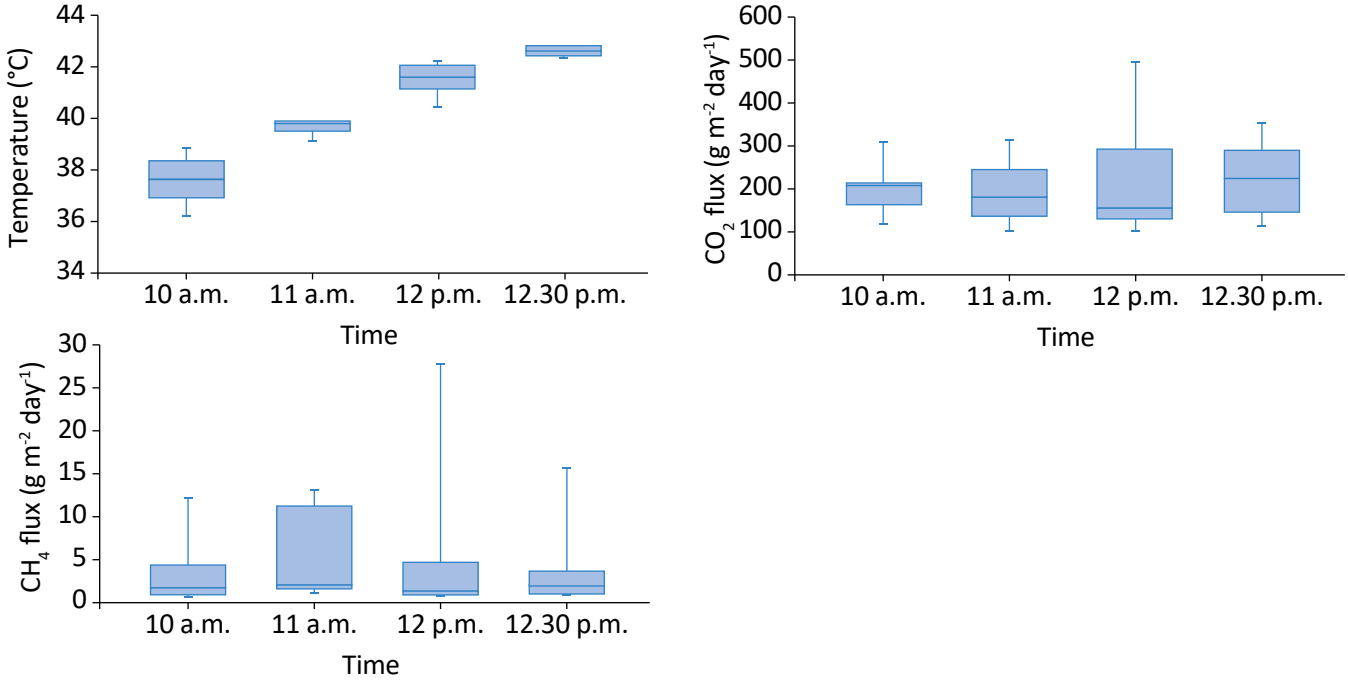


Figure 4 – Regression analysis between the logarithm of CO_2 and CH_4 fluxes (g m^{-2} d^{-1}) and the temperature (°C) in cell 1 (S1) in the Aurá dump.



* Each box represents the mean of eight chambers and the bars, the standard error of the mean.

Figure 5 – Variation of temperature (°C) and fluxes of carbon dioxide and methane measured at 10 a.m., 11 a.m., 12 p.m. and 12:30 p.m., on June 8th, 2017, at S2*.

for temperature. However, no variable was significantly correlated ($R^2 < 0.1218$) with either CO_2 or CH_4 fluxes. The maximum CH_4 oxidation activity was recorded at 15 to 20% moisture contents (ABICHOU *et al*., 2015; VISVANATHAN *et al*., 1999). In our study, 81.25% of the analyzed points had moisture values below 10%. Thus, CH_4 fluxes were expected to be larger than CO_2 fluxes (HANSON; HANSON, 1996; MEI *et al*., 2015), suggesting that CH_4 oxidation in depth is occurring (BIAN *et al*., 2018a; FJELSTED *et al*., 2019).

Spatial variation in substrate permeability, air porosity, methane concentration in substrate gas and humidity content affect CH_4 emission rates (SPOKAS *et al*., 2003). Some advective mechanisms may be locally important for gas fluxes (SCHEUTZ *et al*., 2009). Inductive mechanisms of advective gas movement in the substrate may be: variations in atmospheric pressure (AGHDAM *et al*., 2019; FJELSTED *et al*., 2019; XU *et al*., 2014), temperature (CHRISTOPHERSEN *et al*., 2001; FENG *et al*., 2017; PARK; SHIN, 2001; UYANIK *et al*., 2012), wind velocity in the substrate surface (AGHDAM *et al*., 2019; XIN *et al*., 2016), substrate humidity and water percolation (HANSON; HANSON, 1996; BOGNER *et al*., 2008), and differences in substrate density (BIAN *et al*., 2018b; RACHOR *et al*., 2011). However, the results presented here show no variation in the fluxes, and no correlation between the variables analyzed and the gas emissions, despite the significant variation in temperature (Table 1).

Carbon dioxide and methane fluxes in three locations and at three different hours

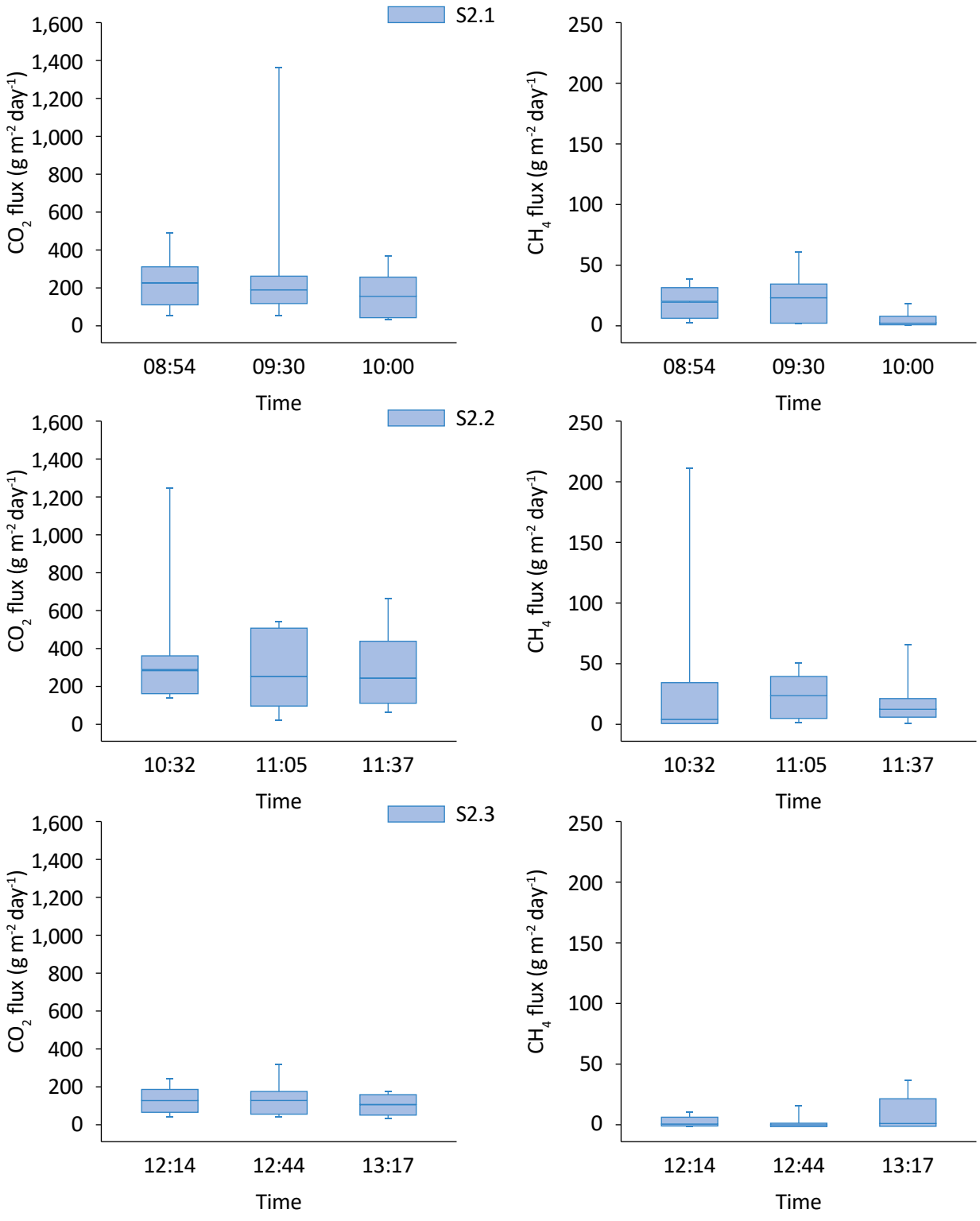
CO_2 and CH_4 fluxes were measured on three different locations at S2 on June 29th, 2017 (Figure 1), with three sequential measurements on each location (Figure 6). Mean CO_2 fluxes were 222.43 ± 52.47, 299.52 ± 155.32 and 153.56 ± 47.82 g m^{-2} d^{-1} at S2.1; 346.88 ± 133.06, 265.69 ± 76.99 and 280.39 ± 75.21 g m^{-2} d^{-1} at S2.2; and 126.73 ± 25.78, 124.78 ± 33.65 and 105.28 ± 23.08 g m^{-2} d^{-1} at S2.3. Residues of CO_2 fluxes did not reach a normal variation, were log-transformed, and did not exhibit a significant variation ($n = 8$, $p > 0.05$) among the sampled locations. A significant difference was recorded only between locations S2.2 and S2.3 (Tukey test, $n = 24$; $p < 0.05$) in the CO_2 flux (Figure 6).

In the same experiment, mean CH_4 fluxes were 18.91 ± 5.03, 22.58 ± 7.57 and 4.48 ± 2.24g m^{-2} d^{-1} at S2.1; 33.43 ± 26.00, 23.56 ± 6.88 and 17.76 ± 7.32g m^{-2} d^{-1} at S2.2; and 2.78 ± 1.63, 2.17 ± 1.99 and 9.46 ± 6.08 g m^{-2} d^{-1} at S2.3 (Figure 6). The residues did not have a normal distribution, and hence were log-transformed. No significant variation ($n = 8$; $p > 0.05$) was found within either studied location. Comparison among locations showed that only S2.3 differed significantly (Tukey's test, $n = 24$; $p < 0.01$) from the other two analyzed locations (Table 2). Air temperature varied significantly (Tukey's test, $n = 8$, $p < 0.05$) throughout the analyzed hours (Table 2), where the temperature during the gas emission measurement at S2.3 was statistically higher than at S2.2 (Tukey's test, $n = 24$, $p < 0.05$), which was greater than S2.1 (Tukey's test, $n = 24$, $p < 0.05$). Thus, air temperature ranged from 33.53 ± 0.39 to 41.57 ± 0.06°C, with a difference of 8.04°C. Wind speed did not vary significantly among hours and locations analyzed, ranging between 1.10 ± 0.20 and 2.43 ± 0.29 m s^{-1}. Relative atmosphere humidity (Table 2) varied significantly within each hour analyzed (Tukey's test, $n = 8$, $p < 0.05$). It was significantly higher at location S2.1 than at S2.2 (Tukey's test, $n = 24$, $p < 0.05$), which did not differ significantly from S2.3 (Table 2). Substrate

Table 1 – Variation of CO_2 and CH_4 fluxes (g m^{-2} d^{-1}), air temperature (°C), barometric pressure (mb), wind speed (m s^{-1}), relative humidity (%) and substrate humidity (%) analyzed on June 8th, 2017*.

Hour (hr)	CO_2 flux (g m^{-2} d^{-1})	CH_4 flux (g m^{-2} d^{-1})	Air temperature (°C)	Pressure (mbar)	Wind velocity (m s^{-1})	Relative humidity (%)	Substrate humidity (%)
10 a.m.	199.41 ± 20.29^a	2.67 ± 1.47^a	37.55 ± 0.32^d	1013.0274 ± 0.0002	1.90 ± 0.46^a	51.48 ± 1.70^a	11.63 ± 3.91^a
11 a.m.	190.04 ± 26.08^a	4.94 ± 1.93^a	39.60 ± 0.10^c	1013.0288 ± 0.0001	1.51 ± 0.42^a	37.58 ± 1.72^b	9.00 ± 3.71^a
12 p.m.	217.77 ± 48.41^a	4.50 ± 3.36^a	41.49 ± 0.22^b	1013.0301 ± 0.0001	1.55 ± 0.27^a	37.93 ± 1.43^b	4.38 ± 1.78^a
12.30 p.m.	223.65 ± 31.91^a	3.01 ± 1.79^a	42.55 ± 0.07^a	1013.0309 ± 0.0001	1.76 ± 0.37^a	41.73 ± 0.32^b	4.25 ± 1.08^a

*Numbers represent the mean ± standard error, and the different letters represent the significance in the difference among the means by Tukey's test ($n = 8$, $p < 0.05$).



*Each box represents the mean of eight chambers and the bars are the standard error of the mean.

Figure 6 – CO_2 and CH_4 fluxes (g m^{-2} d^{-1}) at the three hours, and three different locations in the Aurá dump (sampled on June 29th, 2017)*.

humidity (%) did not vary significantly within each hour analyzed and between sites (Table 2). CO_2 and CH_4 fluxes were not correlated with the environmental parameters analyzed. However, the figures show that an air temperature increase to over 40°C causes a significant decrease ($p < 0.05$) in CO_2 and CH_4 fluxes (Table 2).

The advective movement of gases through the substrate can be induced by variations on atmospheric pressure (AGHDAM *et al.*, 2019; FJELSTED *et al.*, 2019; XU *et al.*, 2014), air temperature (ABICHOU *et al.*, 2015; BOWDEN *et al.*, 1998; XU *et al.*, 2014), surface wind speed (AGHDAM *et al.*, 2019; XIN *et al.*, 2016), displacement of the water that infiltrates the substrate (ABICHOU *et al.*, 2009; BAJAR *et al.*, 2017; HANSON; HANSON, 1996; ROSLEV; KING, 1996; WHALEN *et al.*, 1990), and differences in dump compaction (GEBERT *et al.*, 2011; KAUSHAL; SHARMA, 2016; RÖWER *et al.*, 2011). CH_4 is less dense than the atmospheric air, and therefore tends to rise, while CO_2 and almost all of the vapors produced by volatile organic liquids are denser than air, tending to sink when released into the gaseous portion of the substrate (SEINFELD; PANDIS, 2006). These results found on the Aurá open-air dump confirm what has been stated throughout this paper, that both CO_2 and CH_4 fluxes do not depend on the external factors of the site, nor on substrate humidity. The main flow of the landfill gas seems to be driven by gas concentration and free pathways (porosity) to reach the surface. Large amounts of plastic material placed in the dump can produce hotspots (MØNSTER *et al.*, 2015; RACHOR *et al.*, 2013; SCHEUTZ *et al.*, 2017), operating as small "chimneys" for CO_2 and CH_4 fluxes (Figure 6).

Specialization of the carbon dioxide and methane fluxes

The geospatial analysis performed on November 9th, 2017 (at the end of the dry season), for CO_2 and CH_4 fluxes, showed a non-uniform distribution of gases emission into the atmosphere (Figure 7). The CO_2 flux ranged from 20.54 to 413.73 g m^{-2} d^{-1}, and CH_4, from -0.11 to 25.32 g m^{-2} d^{-1}. Large hotspots were found on the surface of the dump at different points for CO_2 flow and only at one point for CH_4 flow (Figure 7). On the

Table 2 – Variation of CO_2 and CH_4 fluxes (g m^{-2} d^{-1}), at different locations in S2, at different sampling hours (hour) on June 29th, 2017, compared to air temperature (°C), wind speed (m s^{-1}), relative air humidity (%) and substrate humidity (%), in the Aurá dump*.

Location	Hour	CO_2 flow (g m^{-2} d^{-1})	CH_4 flow (g m^{-2} d^{-1})	Air temperature (°C)	Wind speed (m s^{-1})	Relative humidity (%)	Substrate humidity (%)
S2.1	1	222.43 ± 52.47^{a}	18.91 ± 5.03^{a}	33.53 ± 0.39^{c}	2.43 ± 0.29^{a}	60.80 ± 2.13^{a}	4.50 ± 1.09^{a}
	2	299.52 ± 155.32^{a}	22.58 ± 7.57^{a}	35.58 ± 0.15^{b}	1.38 ± 0.24^{b}	39.65 ± 1.50^{b}	1.88 ± 0.40^{b}
	3	153.56 ± 47.82^{a}	4.48 ± 2.24^{a}	36.59 ± 0.22^{a}	1.16 ± 0.17^{b}	40.55 ± 1.71^{b}	2.00 ± 0.27^{b}
Mean		225.17 ± 55.80^{AB}	15.32 ± 3.40^{A}	35.23 ± 0.31^{C}	1.65 ± 0.18^{A}	47.00 ± 2.27^{A}	2.79 ± 0.45^{A}
S2.2	1	346.88 ± 133.06^{a}	33.43 ± 26.00^{a}	38.39 ± 0.17^{c}	1.41 ± 0.42^{a}	51.65 ± 1.49^{a}	3.75 ± 0.98^{a}
	2	265.69 ± 76.99^{a}	23.56 ± 6.88^{a}	39.56 ± 0.16^{b}	1.10 ± 0.20^{a}	34.50 ± 1.70^{b}	1.63 ± 0.38^{a}
	3	280.39 ± 75.21^{a}	17.76 ± 7.32^{a}	40.19 ± 0.15^{a}	1.58 ± 0.25^{a}	29.79 ± 1.10^{b}	1.50 ± 0.19^{a}
Mean		297.65 ± 55.01^{A}	24.92 ± 8.98^{A}	39.38 ± 0.18^{B}	1.36 ± 0.17^{A}	38.65 ± 2.12^{B}	2.29 ± 0.40^{A}
S2.3	1	126.73 ± 25.78^{a}	2.78 ± 1.63^{a}	40.35 ± 0.16^{c}	1.73 ± 0.21^{a}	45.94 ± 0.70^{a}	2.75 ± 0.77^{a}
	2	124.78 ± 33.65^{a}	2.17 ± 1.98^{a}	41.57 ± 0.06^{a}	1.33 ± 0.35^{a}	35.70 ± 1.43^{c}	1.50 ± 0.27^{a}
	3	105.28 ± 21.59^{a}	9.46 ± 6.08^{a}	40.69 ± 0.18^{b}	1.29 ± 0.36^{a}	41.44 ± 1.44^{b}	1.38 ± 0.18^{a}
Mean		119.52 ± 15.72^{B}	5.01 ± 2.14^{B}	40.48 ± 0.43^{A}	1.45 ± 0.18^{A}	41.03 ± 1.13^{AB}	1.88 ± 0.30^{A}
Total		215.45 ± 27.98	15.23 ± 3.42				

*Numbers represent the mean ± standard error, and the different letters represent the statistical difference ($p < 0.05$) between the averages by the Tukey test, where lowercase letters compare the hours within each site (n = 8), and capital letters between the sites (n = 24).

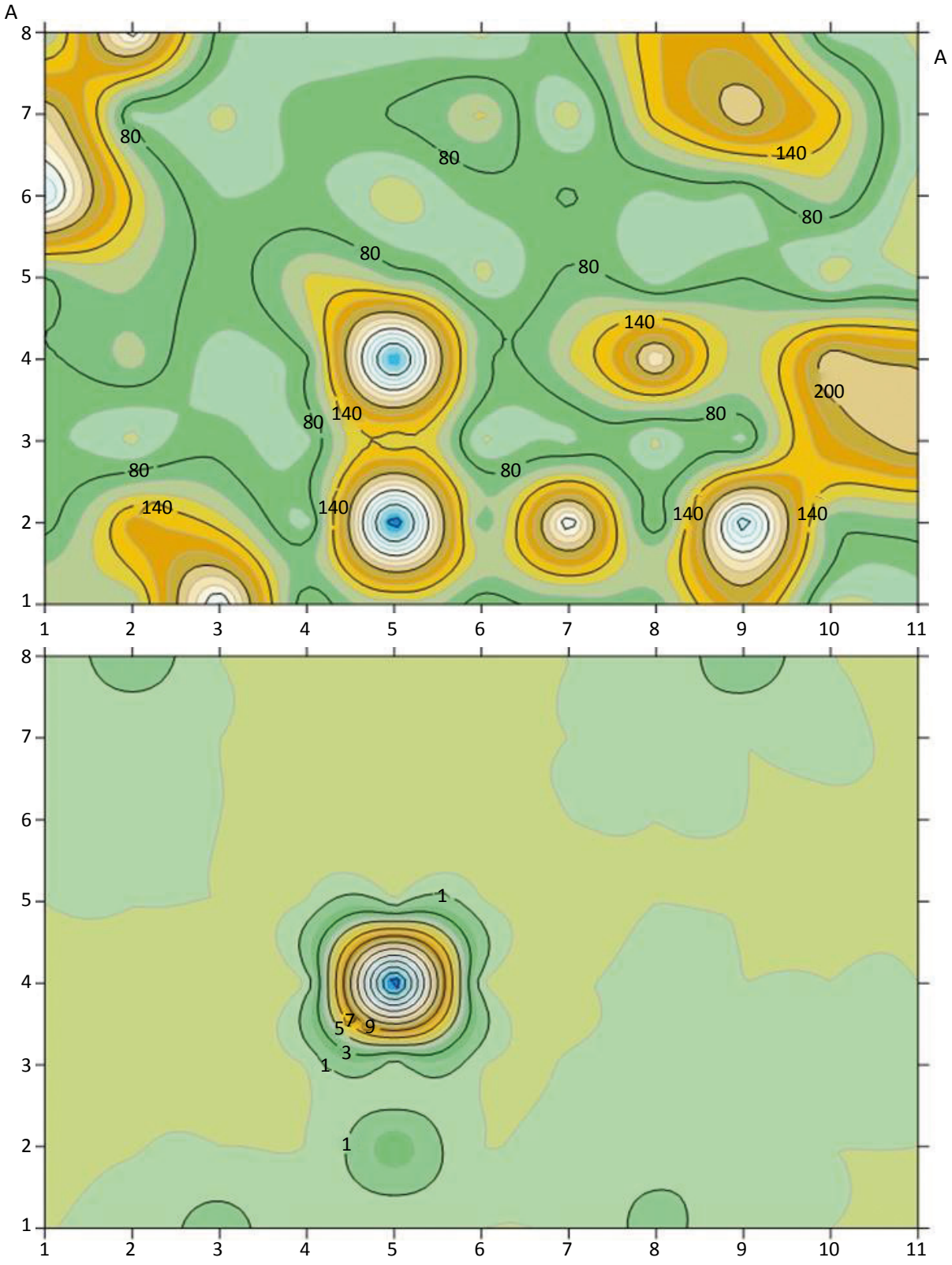


Figure 7 – Geospatial variation (with flow chambers allocated every two meters) for CO_2 and CH_4 fluxes (g m^{-2} d^{-1}), studied on November 9th, 2017 in the Aurá open-air dump.

other hand, in this older area, which has not been receiving residues from many years, several places had a zero CH_4 flow, and some exhibited a sink of atmospheric CH_4 (SAUNOIS *et al*., 2016; STERN *et al*., 2007). The presence of preferential emission points was probably due to changes in gas concentrations within the dump (ABICHOU *et al*., 2006; BAJAR *et al*., 2017; KRAUSE, 2018). These changes are due to the uneven spreading of residues, lack of substrate layers or no use of plastic waterproofing between residue layers, or any gas collection strategy. These intense CO_2 fluxes may be consequence of the oxidation rate of CH_4 by the methanotrophs located in the substrate under the chambers (BIAN *et al*., 2018a; CHRISTOPHERSEN *et al*., 2001). This methane oxidation may be intensifying due to the less rainy period of the region (Figure 2), with November being the last month of the dry season.

These results confirm the enormous spatial variability of gas fluxes in the Aurá dump, which shows an uneven residue distribution (AHOUGHALANDARI; CABRAL, 2017b; RÖWER *et al*., 2011). The variogram is an essential tool in a geospatial analysis, determining the amount of spatial dependence (autocorrelation) in the spatial data underlying the variations (SPOKAS *et al*., 2003). It is calculated from sampling sites in a uniform geospatial distribution and at least 100 sites are required for a good variogram accuracy using a stationary random function (SPOKAS *et al*., 2003). The variogram data presented in this study used 88 sampling sites, measured with two devices simultaneously on opposite sides of the geospatial design.

Results from the semi variance analysis (variogram) revealed that CO_2 fluxes at 4 to 10 m from the samples are independent and that, before and after this distance, the samples are dependent on the sampling site (Figure 8). However, the distance explains very little of the variation in CO_2 flux ($R^2 = 0.04$, $p = 0.668$). The semi variance of CH_4 showed that the fluxes are dependent on the sampling site and that there are possible spots with a higher flux between 2 and 6 m, and the distance among the sites reasonably explains the CH_4 flux variation ($R^2 = 0.69$; $p = 0.022$). However, the results for the gas emissions indicated that the non-spatial variability was high in comparison with the spatial variability. Most studies show intense spatial variability (ABUSHAMMALA *et al*., 2016; AHOUGHALANDARI; CABRAL, 2017b; CHANTON *et al*., 2011; DI TRAPANI *et al*., 2013). However, if the variables had been studied in greater detail, the heterogeneity defined as non-spatial variability may have exhibited a spatial structure. However, substrate temperature does not appear to vary spatially (Figure 8), and substrate humidity was not measurable due to a device malfunction.

CO_2 and CH_4 fluxes spatialization did not depend on the substrate temperature, but only on the enormous spatial variability as seen above (ABUSHAMMALA *et al*., 2013). The dump was constructed in a disorderly manner, without waterproofing and without covering the layers with substrate, isolating the concentration of organic material among plastics and other materials of difficult degradation (KARANJEKAR *et al*., 2015; SPOKAS *et al*., 2006). At the same time, this disordered arrangement can produce paths that facilitate gas flow, creating a hotspot (RACHOR *et al*., 2013; TAYLOR *et al*., 2018). Due to the hotspots and the methanotrophic activity, the use of CH_4 to generate energy for a long time in open dumps in Brazil is unfeasible (AHOUGHALANDARI; CABRAL, 2017a; COSTA *et al*., 2019). On the other hand, the presence of hotspots implies in a limited recovery effort to produce significant recovery results (GONZALEZ-VALENCIA *et al*., 2016).

The fluxes during the analyzed months were on average 171.62 ± 13.46 and 10.54 ± 2.70 g m^{-2} d^{-1} for CO_2 and CH_4, respectively. Thus, total monthly emissions from the Aurá dump to the atmosphere were 51.49 ± 4.04 Mg CO_2 ha^{-1} $month^{-1}$ and 3.16 ± 0.81 Mg CH_4 ha^{-1} $month^{-1}$. World landfill production ranges from 518.28 ± 448.28 and 184.11 ± 112.70 g m^{-2} d^{-1} of CO_2 and CH_4, respectively (GOLLAPALLI; KOTA, 2018). As a result, the CO_2 flow in Aurá's dump remains similar to the measured flux in other active landfills, with a rapid decrease in CH_4 flux. Also, exploiting Aurá dump for energy production may be economically unfeasible due to the large CH_4 flux spatial variation and the low generation.

Since the Aurá dump area is 30 ha, the total gas emitted to the atmosphere is 1,544.61 and 94.84 Mg CH_4 $month^{-1}$. Thus, when converting CH_4 to CO_2 equivalent (CO_2-e), we consider the global warming potential of CH_4 in 100 years to be 23 times greater than of CO_2 (IPCC, 2013), which results in a production of 1,359.96 Gg CO_2-e y^{-1}. That is, even after being closed for the domestic waste deposit and burned 2,608.40 Gg CO_2-e (between 2007 and 2016), Aurá dump is still a significant contributor to the intensification of the greenhouse effect.

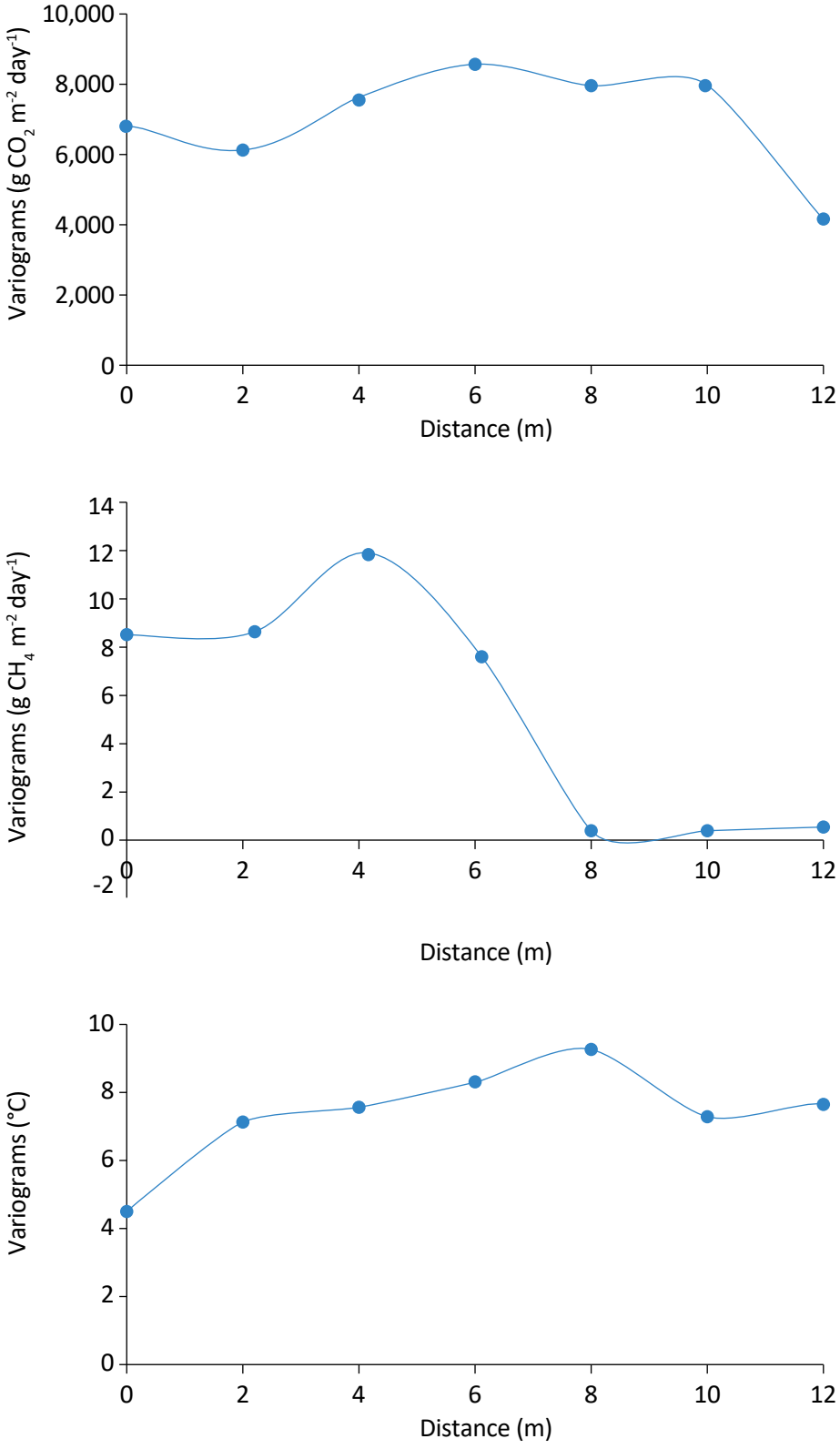


Figure 8 – Variogram of the (A) CO_2 flux, (B) CH_4 flux, and (C) temperature: variance γ(h) per distance h (m).

CONCLUSION

Based on the results, we can conclude that:

- The gas emission did not show a significant difference between the end of the rainy period and the end of the dry period;
- The spatial variability in the flux of CO_2 and CH_4, especially, is very large, forming hotspots of high concentrations;
- Aurá's dump generates approximately 172.0 and 11.0 g m^{-2} d^{-1} for CO_2 and CH_4, respectively;
- The fluxes were not clearly correlated with any micrometeorological variable studied, i.e., only the gas concentration and the free paths to the surface flow motivate the release to the atmosphere;
- The oxidation of CH_4 is apparently the main source of high CO_2 production on the surface, which is due to the low relative humidity of the open-air dump surface;
- Aurá open-air dump was active for 28 years and has been closed for three. In addition, a significant amount of CO_2-e was taken from the open-air dump by the CRA Company. Still it continues to release 1,359.96 Gg CO_2-e y^{-1} into the atmosphere;
- This result can be used with the IPCC waste model to accurately estimate the total CH_4 emissions from the open-air dump in Amazon, which can be used to assess how much the CO_2-eq emissions from the Amazonian dump contributes to the global warming.

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