

Diurnal and Seasonal Variation of CO₂ and CH₄ Fluxes in Tomago Wetland

David Safari¹, Grant. C. Edwards¹, Faustina Gyabaah²✉

¹Macquarie University, Faculty of Science and Engineering, Department of Environmental Sciences

²Bolgatanga Polytechnic, School of Engineering, Civil Engineering Department

Abstract: The measurement of greenhouse gas (GHG) fluxes in estuaries is crucial in expressing the impacts of these GHGs on global warming, and hence climate change. In this study, we investigated the effect of various environmental and micrometeorological factors on diurnal and seasonal variations of methane (CH₄) and carbon dioxide (CO₂) in a tidal inundated saltmarsh. Measurements of GHG fluxes were taken by using eddy covariance technique from August 2015 to July 2016 in Tomago wetland, Newcastle, NSW, Australia. In this paper, a positive flux is defined as the one directing into the atmosphere. The highest average diurnal emissions were 2.54 µg m⁻² s⁻¹ CH₄ during the day and 0.45 mg m⁻² s⁻¹ CO₂ at night. Monthly average fluxes peaked in February (0.365 µg m⁻² s⁻¹ CH₄ and 0.137 mg m⁻² s⁻¹ CO₂). There was a significant negative relationship between CO₂ flux and water level ($p < 0.001$), tidal height ($p = 0.02$) and positive relationship with water temperature ($p = 0.002$). CH₄ flux showed positive correlation with water level and negative correlation with EC although not statistically significant. Although tidal flooding did not demonstrate clearly carbon sequestration before and after tidal reinstatement, freshwater events (rainfall) were seen to influence the wetland carbon balance.

Keywords: GHG Flux, Water Level, Tidal Inundation, Salinization, Rainfall

Introduction

While wetlands function as sinks in the carbon cycling process through storing carbon in the vegetation and soil, they can also function as atmospheric sources of CO₂ and CH₄ (Mitsch et al., 2013). Through the process of photosynthesis, carbon dioxide is absorbed by green plants to form organic carbon, as well as autotrophic bacteria (Olsson et al., 2015). The absorbed carbon dioxide eventually gets released back into the atmosphere through decomposition of organic matter under aerobic and anaerobic processes. Tidal marshes are considered to be among the most efficient carbon sinks, with capacity to accumulate and store carbon at rates which are 55-times faster on millennial timescales, compared to tropical rainforests (McLeod et al., 2011). In Australia and other parts of the world, the capacity to store carbon by tidal marshes has been significantly affected by human activities such as land reclamation (Bu et al., 2015), chemical and physical disturbances (Macreadie, Hughes, & Kimbro, 2013), as well as eutrophication (Deegan et al., 2012).

Studies have revealed that estuarine wetlands, to a greater extent have high carbon sequestration capacity per unit area than other wetland ecosystems especially in undisturbed conditions (Bridgman, Megonigal, Keller, Bliss, & Trettin, 2006). Conversion of wetlands into agricultural zones, such as pasture lands, and urban development can reduce carbon storage capacity of the wetland. In many parts

of the world including the US, studies indicate that wetland reclamation into pasture lands and crop farms can substantially reduce carbon storage within the first decade of land use change (Armentano & Menges, 1986). Similar studies that were conducted in Australia also demonstrate that about 50% of the wetlands that were converted into other land use practices, such as pasture land (Streever, 1997), have been affected due to modifications by tidal flows (Howe, Rodriguez, & Saco, 2009).

Tidal flow can be managed through construction of flood mitigation drains with modified flood gates (Smart Gates). On the other hand, tidal restriction can cause acidification of coastal wetlands, which is a significant environmental, economic and social problem in many parts of Australia (Indraratna, Blunden, & Nethery, 1999), including the Hunter estuary of New South Wales. Acidification resulted into the formation of acid sulphate soils (ASS) due to pyrite as a dominant iron sulphide formed during the last major interglacial in coastal NSW (Indraratna, Golab, Glamore, & Blunden, 2005), and this is one of the major problems that affected Tomago wetland, and thus, lowering its ecosystem productivity. Tidal flow restoration allows flushing water to decrease acidity, increase dissolved oxygen, remove exotic freshwater weeds, enhance runoff during wet periods and create healthy fish breeding grounds (Dick & Osunkoya, 2000; Glamore, 2003; Indraratna, Glamore, & Tularam, 2002). Studies on coastal saltmarshes in Australia have concentrated mainly on

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Faustina Gyabaah (Correspondence)

assessing carbon storage and accretion, geochemistry of acid sulphate soils and GHGs production. This research project sought to investigate diurnal and monthly patterns of CO₂ and CH₄ fluxes, and the influence of environmental factors before and after tidal flow reinstatement. Knowledge about diurnal and seasonal fluctuations of GHG fluxes gives an insight on interventions to reduce GHG emissions from saltmarshes. Therefore, the findings demonstrate specific recommendations to reduce the effects of climate change and global warming by limiting emissions of GHGs from the coastal saltmarsh. The study outcomes are essential in understanding the carbon cycling functions of coastal wetlands, and the role they play in GHG emissions and climate change as a whole. While as the results from this research project will give a strong argument for protection and restoration of degraded saltmarsh ecosystems in NSW and other coastal areas in Australia, the research findings are relevant to enrichment of the scientific understanding of the

processes that can control GHG emissions due to tidal inundation.

1.0 Materials and methods

1.1 Study site

Measurements were carried out in Tomago wetlands in Hunter estuary, NSW, Australia (32°51'52"S, 151°42'15"E), and fluxes were recorded for 1 year. In its natural state, Tomago wetland was an estuarine wetland mainly covered by saltmarsh and mangroves. There was construction of levee and a drainage system between 1913 and 1928, and this led to dominance of grasses for grazing animals, in addition to weeds which were initially less adapted to flooded aquatic environment. Further expansion of the drainage system was done between 1968 and 1980 with the construction of flood gates by the NSW works department to; promote agriculture in the wetland, direct water from smaller floods downstream and provide a flood detention basin in a bid to protect New Castle from heavy floods.

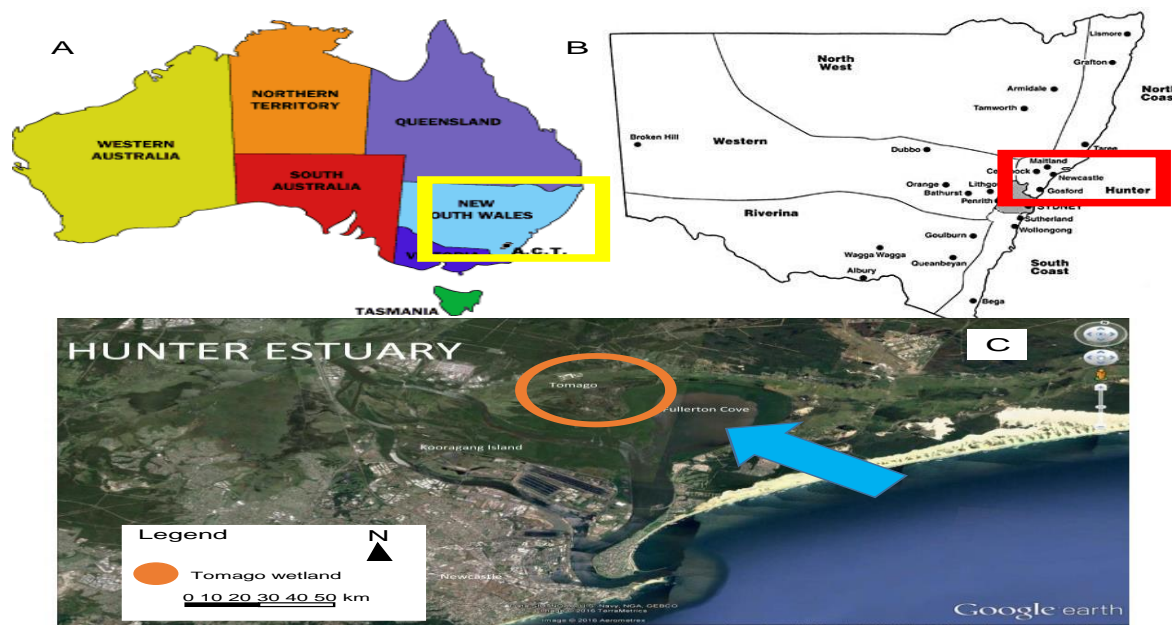


Figure 1 Shows the study area. A-Map of Australia showing New South Wales, in which the study area is located. B-Map of New South Wales showing neighbouring urban centres to the study site. C-Google map of the Hunter Estuary in which the study site is located.

1.2 Gas flux measurement

The eddy covariance (EC) technique was used to measure gas fluxes. The system comprised of: the LI-7700 CH₄ open-path gas analyser (LI-COR Biosciences, Lincoln, NE, USA), a Gill WindMaster 3D sonic anemometer (Gill Instruments Limited Lymington Hampshire, UK) and the LI-COR 7500 CO₂/H₂O open-path gas analyser (LI-COR Biosciences). The data were then logged through the

LI-COR 7550 and smart flux system to produce high resolution (i.e., tenth of a second data for all parameters) and half hour fluxes. Data was collected at 3.5 m above the ground, and was recorded after every 30 minutes. Instruments were installed in August 2015 before flooding, and data was collected up to July 2016.

The eddy covariance (EC) method was used to provide direct measurement of the turbulent flux of

CH₄ and CO₂. The Tomago site offered an ideal setting for the application of the EC method (Foken, 2008; Pattey et al., 2006). The eddy turbulent flux was computed based on the following assumptions among others; terrain was horizontal and uniform, steady environmental conditions (Pattey et al., 2006), and detection of small changes at very high frequency.

Mathematically, in turbulent flow, vertical flux can be expressed as,

$$F = \bar{\rho} \bar{w} \bar{s}$$

Where **F** is the flux equal to a mean product of air density (*p*), vertical wind speed (*w*) and the dry mole fraction (*s*) of the gas of interest in the air.

1.3 Measurement of environmental parameters

Soil water temperature and radiation parameters were recorded onto a CR3000 data logger (CR3000; Campbell Scientific). Electrical conductivity (μS cm⁻¹) data were logged by the HOBO instrument that was placed below the water table. This type of data logger was selected to be used because of the larger dynamic range that could be encountered in the experiment. The EC logger was placed in a perforated pipe to allow entry of water so that the logger sensor was submerged in water within the pipe. It was then held in suspense within the pipe by using a string tied at the upper end of the pipe. The pipe was placed at a depth of 90 cm in the soil under water at the inundated area. Data was downloaded monthly by using the installed HOBO software.

Water level was measured using the HOBO U20L-004 research-grade water level data logger which has a 0.1 % level of accuracy in measurement. The logger was fixed flat on the ground surface and then any water depth above it was recorded as a positive water depth value and the negative value was returned whenever there was no water over the logger. All the values of water depths were recorded in meters.

Rainfall data (mm) from August 2015 to May 2016 was obtained from Williamtown station, Bureau of Meteorology (BOM), NSW, and it was measured using a tipping bucket rain gauge.

Data about tidal times and heights for Newcastle-NSW were downloaded from the Bureau of meteorology website. The corresponding flux data was selected basing on the time for the occurrence of the tide.

1.4 Data capture, processing and QA/QC

The eddy covariance flux data at a frequency of 10 Hz was processed from the LI-COR smart flux

system and EddyPro[®] software (available through LI_COR Biosciences). Foken level 2 QA/QC criteria were applied to all the Eddy Covariance data. CH₄ and CO₂ flux values associated with spikes resulting from signal loss or instrument malfunctions were removed. In addition, high fluctuations in turbulence, especially at night due to low winds, resulted in poor data quality and such data was also filtered out.

The installed LI-7700 has two diagnostic outputs which were used to filter half-hourly values where the instrument was performing poorly. It has the received signal strength indicator (RSSI), which indicates the cleanliness of the mirrors, and a coded value that represents one or more pieces of diagnostic information. Low RSSI values do not always lead to outliers or spikes, but need to be combined with diagnostic code values to properly filter instances of the instrument malfunction from the data. There are also occasions when the mirrors are clean but the diagnostic value records malfunctions (Dengel, Levy, Grace, Jones, & Skiba, 2011). For the case of this study, data with signal strength above 20 for CH₄ was considered while the signal strength for CO₂ sensor was set at 80.

Sensible heat flux data was also used to perform quality assurance and quality control data for outliers. This was achieved by considering fluxes that were in the range of -50 – 500 W m⁻². All the values that were outside this range were deleted before computing diurnal sensible heat fluxes. A filter of three times the standard deviation was applied to remove spikes that were not attributable to non-optimal instrument operation. The daily averages of sensible heat flux (W m⁻²) were used to calculate the day-night transition to determine day and night time hours for CH₄ and CO₂ fluxes. If the sensible heat flux value was greater than 10 W m⁻², then the point was considered to be day while values less than 10 W m⁻² were labelled night.

1.5 Statistical analysis

The averages, standard deviations, and standard of errors for daily and monthly fluxes CH₄ and CO₂ were determined using excel. Diurnal variations of CH₄ and CO₂ fluxes were calculated using averages values of fluxes for every 30 minutes interval for each month. Results of diurnal and monthly variation of CH₄ and CO₂ flux were presented on line and bar graphs... The relationships between environmental factors (soil-water temperature, rainfall, water level, tidal height and electrical conductivity) and GHG fluxes were explored using line and bar graphs, and linear regression with associated correlations at 95% confidence interval.

2.0 Results

2.1 Diurnal variation of CH₄ and CO₂ fluxes with water level

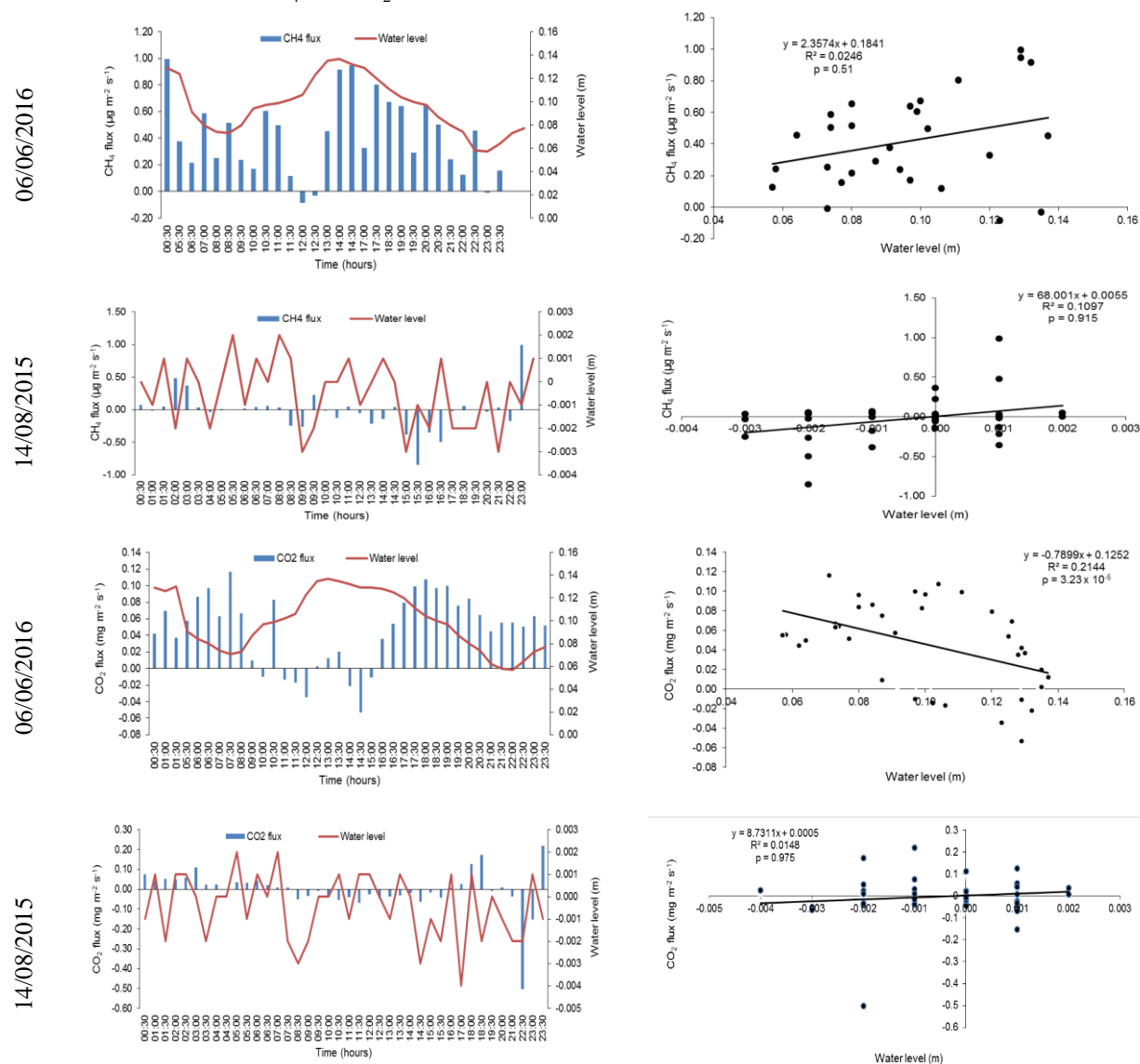


Figure 2 Diurnal variation of CO₂ and CH₄ fluxes with water level during tidal inundation (6th June 2016) and with exposed soil surface stage (14th August 2015)

The variation of CH₄ flux on 06th June (during tidal inundation) demonstrated a distinct pattern with low emissions during the night time and high emissions in the day time, and uptake of CH₄ around mid-day. CH₄ flux showed positive correlation with water level on a diurnal scale although the relationship was not statistically significant ($p = 0.51$). On the other hand,

CO₂ fluxes indicated a significant negative correlation with water level ($p < 0.001$).

On 14th August, there were fluctuations in diurnal variations, with no distinct trends for both CH₄ and CO₂ fluxes, and water level. However, CH₄ and CO₂ fluxes indicated a positive relationship with water level which was not statistically significant.

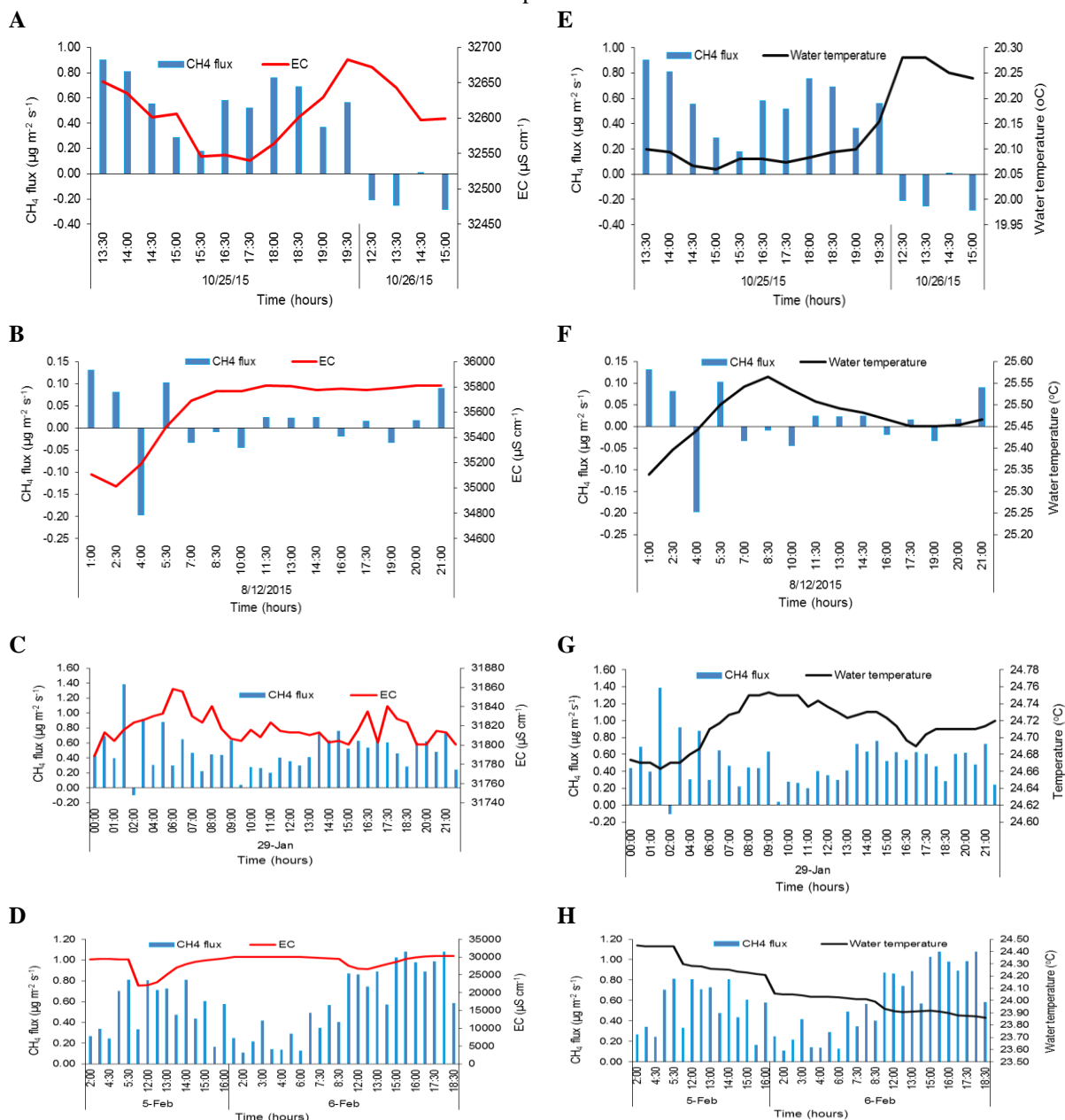
2.2 Diurnal variations of CH₄ with EC and water temperature

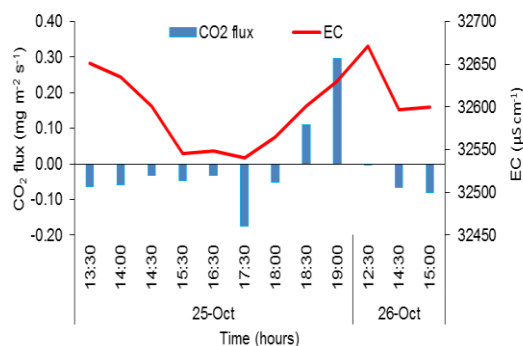
Figure 3 Diurnal variation of CH₄ fluxes with electrical conductivity (EC) and water temperature before tidal inundation-October 2015 (A&E), and after tidal inundation-December 2015 (B &F), Jan 2016 (C&G) and Feb 2016 (D&H)

Generally, CH₄ fluxes showed a diurnal variation which was less correlated with EC and water temperature for the dates indicated in figure 3 above. There were low negative correlations between CH₄ fluxes and EC (i.e. $r = -0.17, -0.09, -0.01$ and -0.20 for October, December, January and February respectively). However, the relationships were not statistically significant. Water temperature was also

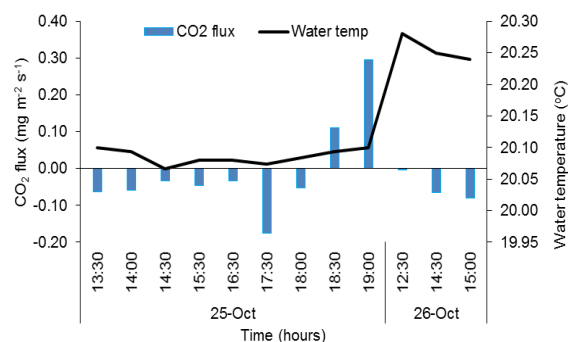
negatively correlated with CH₄ fluxes ($r = -0.80, r = 0.31, r = -0.35$ and $r = -0.32$ for October, December, January and February respectively) but all these relationships were not statistically significant. Water temperature was positively correlated with EC except during February ($r = -0.29$), and the highest positive correlation ($r = +0.93$) was identified in January 2016.

2.3 Diurnal variations of CO₂ with EC and water temperature

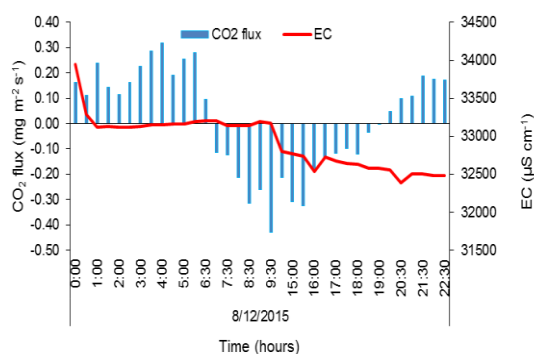
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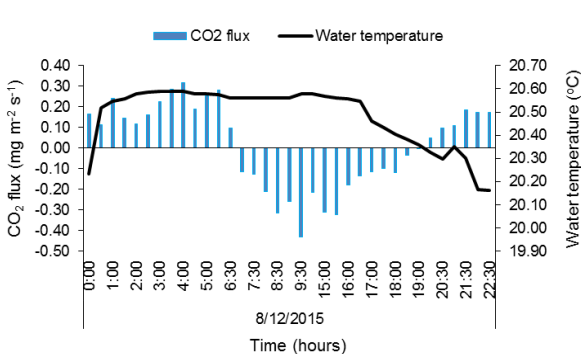
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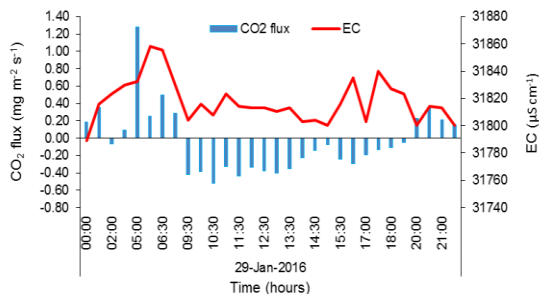
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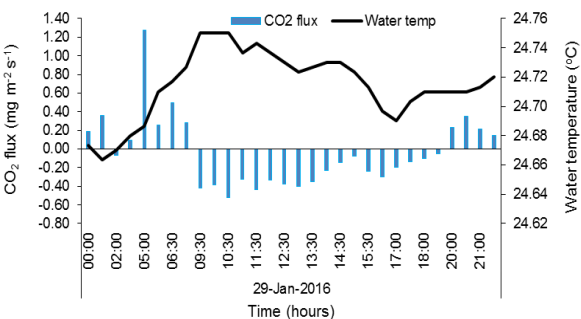
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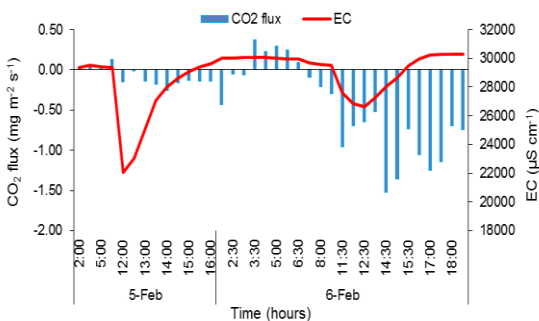
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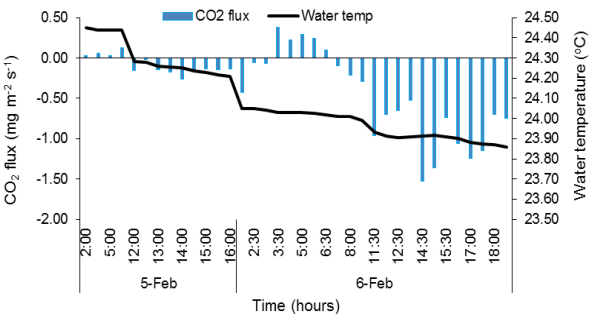


Figure 4 Diurnal variation of EC and water temperature with CO₂ fluxes before tidal inundation-October 2015 (S&T), and after tidal inundation-December 2015 (U&V), January 2016 (W&X) and February 2016 (Y&Z)

The diurnal variation of CO₂ flux with EC indicated low positive correlation before and after tidal flooding and all these relationships were not statistically significant at 95% level of confidence except for January ($r = 0.36$, $p = 0.25$ for October, $r = 0.15$, $p = 0.38$ for December, $r = 0.32$, $p = 0.03$ for January and $r = 0.009$, $p = 0.74$ for February). Water temperature appeared to be negatively correlated with

CO₂ ($r = -0.077$ for October, $r = -0.214$ for December, $r = -0.55$, $p = 0.002$ for January) except in February ($r = 0.62$, $p < 0.001$) which indicated a high positive correlation. The relationship between CO₂ flux and water temperature was statistically significant for diurnal variations in January and February 2016, and this occurred after tidal flooding.

2.4 Diurnal variation of CH₄ and CO₂ fluxes with tidal water height

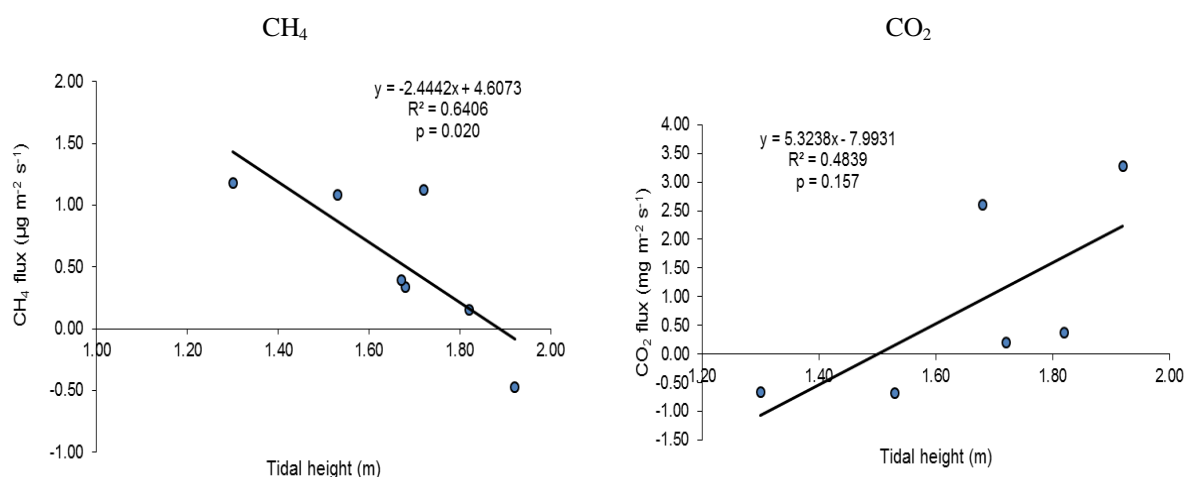


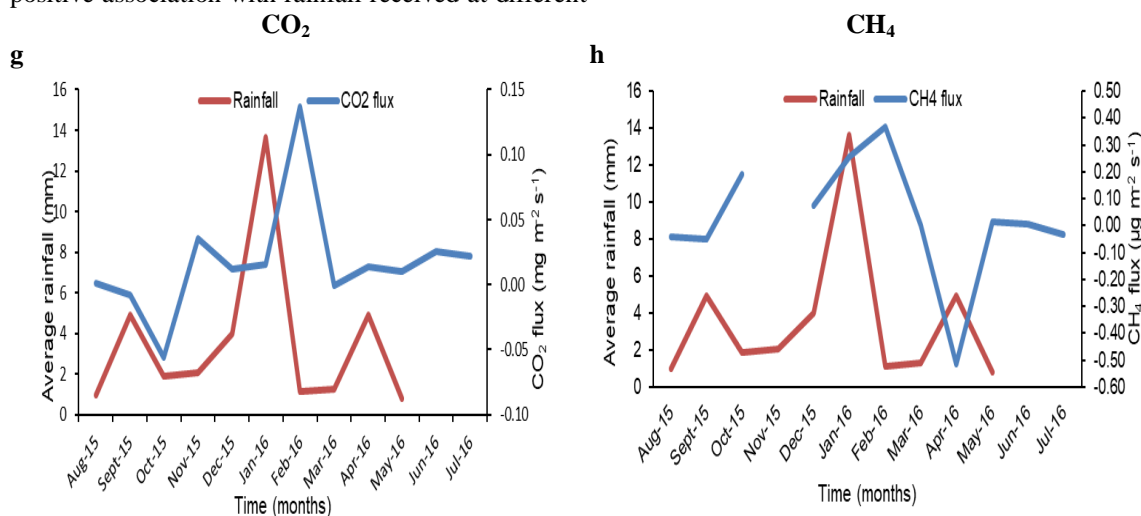
Figure 5 Linear relationship between CH₄ and CO₂ fluxes at the water-air interface and tidal water height in February 2016

Analysis of CH₄ flux data for four days (11th, 12th, 13th, and 15th February 2016) from the tidal marsh, indicated a significant negative correlation with tidal height ($r = -0.80$, $p = 0.020$) while CO₂ flux during the same period indicated a positive correlation with tidal height ($r = 0.69$, $p = 0.157$).

2.5 Seasonal variation of CH₄ and CO₂ fluxes with environmental factors

Generally, CO₂ and CH₄ fluxes showed a weak positive association with rainfall received at different

periods of the months. Increase in rainfall resulted in an increase in CO₂ and CH₄ fluxes except in September 2016 during which an increase in CH₄ fluxes was associated with a decrease in rainfall. Rainfall was irregular except for September, December, January and April which received rainfall almost throughout the months.



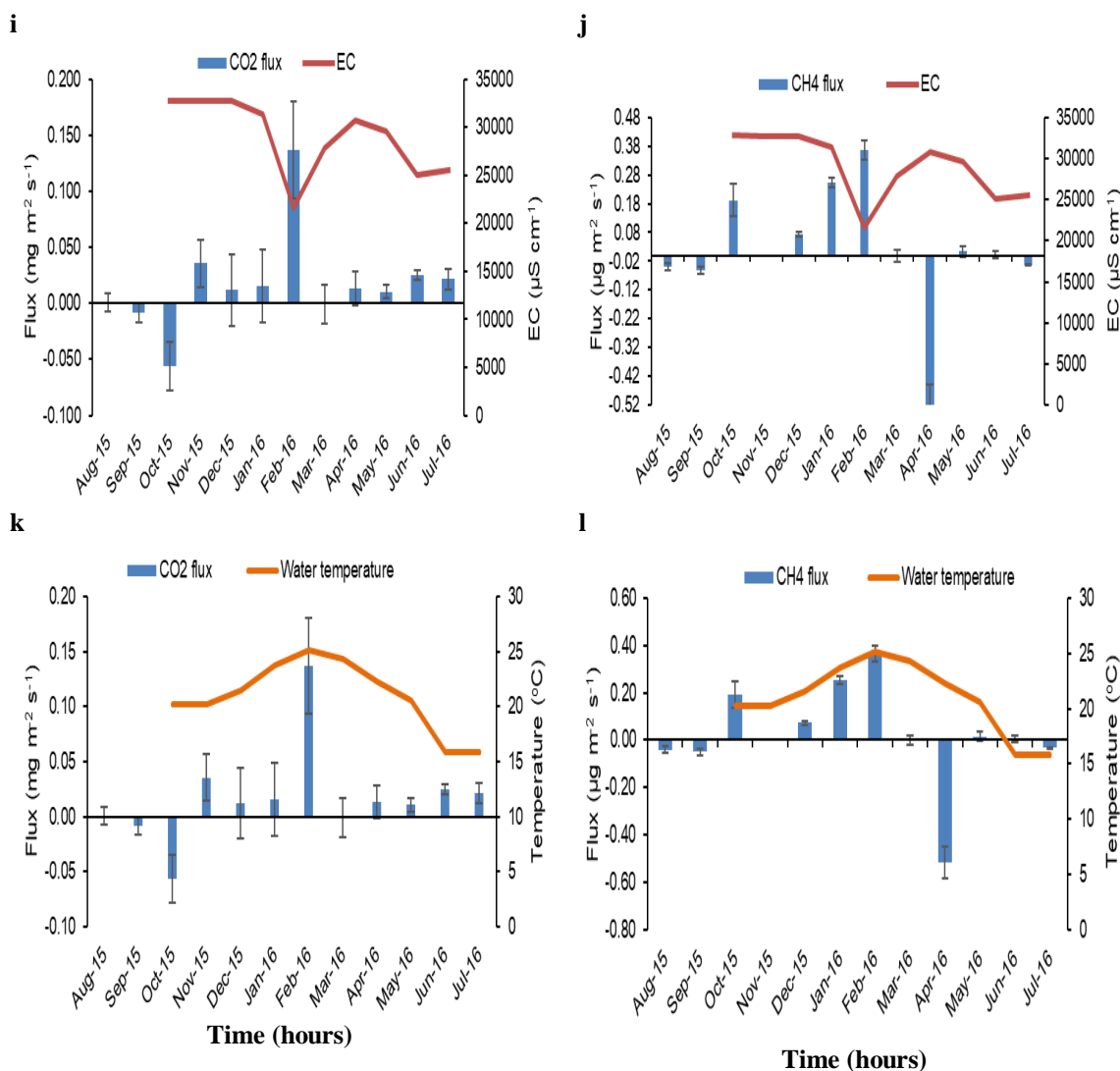


Figure 6 Seasonal variation of rainfall, electrical conductivity (EC), and water temperature with CO₂ and CH₄ fluxes in a period of one year (from August 2015 to July 2016). Water temperature and EC measurements started in October 2015, and also there was no rainfall data for June and July due to technical challenges

High CO₂ uptake occurred from August to October, with the highest consumption rate of $-0.06 \text{ mg m}^{-2} \text{ s}^{-1}$ in October. During this time a small rainfall event occurred, and before tidal reinstatement for the saltmarsh. Emissions of CO₂ increased from $0.016 \text{ mg m}^{-2} \text{ s}^{-1}$ in January to a maximum of $0.137 \text{ mg m}^{-2} \text{ s}^{-1}$ in February, and this increase in average monthly CO₂ flux was noted after receiving heavy rainfall in January (monthly average of 13.62 mm). Generally, CH₄ fluxes increased with increase in the monthly rainfall although the relationship was not consistent throughout the study period. Results indicate that the highest monthly average CH₄ emissions ($0.37 \text{ μg m}^{-2} \text{ s}^{-1}$) occurred in February after receiving heavy rainfall in January.

The highest CH₄ emissions ($0.37 \text{ μg m}^{-2} \text{ s}^{-1}$) and CO₂ emissions ($0.14 \text{ mg m}^{-2} \text{ s}^{-1}$) were obtained at the same

time in February when soil water indicated the highest average water temperature (25.15°C).

3.0 Discussion

3.1 Diurnal variations of CH₄ and CO₂ fluxes with environmental factors

A decrease in CO₂ flux with an increase in the water level on a diurnal scale could have been due to lowered rate of CO₂ production by aerobic respiration and enhances the rate of CH₄ production by anaerobic methanogenesis. High water levels might have resulted into saturation of the soils with water, thus reducing the ability of oxygen to diffuse from the atmosphere into the soils. On the other hand, low water levels enable oxygen to diffuse from the atmosphere into the soil leading to aerobic CO₂ production but limiting CH₄ production. Figure 2 shows that while CO₂ flux decreased with an increase

in water level, CH₄ flux increased as water level increased on 06th June 2016. These relationships clearly demonstrated the water-saturated soil conditions favourable for CH₄-producing microbial communities after tidal flow inundation and rainfall during the summer season. The diurnal variations of CH₄ and CO₂ on 14th August 2016 showed a very low positive correlation with water level because the tidal flow re-instatement had not been effected. The diurnal change in water level during this time was insignificant and therefore, had less or no influence on CH₄ and CO₂ fluxes.

The negative correlation between the tidal height and methane fluxes may imply that low methane emissions could have occurred during tidal submergence. When the saltmarsh soils are flooded with inflowing water, it's likely that gas diffusion from soil air spaces may be interfered with, and also the inflowing water may dilute CH₄ gas dissolved in the pore water and hence reducing the amount of CH₄ emitted from the saltmarsh (Yamamoto et al., 2009). The decrease in the amount of CH₄ may also be attributed to certain proportions of CH₄ diffusing into the tidal water may get oxidised in the water column before it reaches the water-air interface.

The negative diurnal relationship between electrical conductivity and CH₄ flux from Figure 2 probably indicated inhibition of methanogenesis due to presence of certain ionic species such as sulphates in the soil sediments. In presence of sulphate ions, the sulphate reducing bacteria outcompete methanogens for the appropriate substrates available such as acetate and hydrogen (D'Angelo & Reddy, 1999). The negative diurnal correlations between CH₄ flux and temperature was not consistent with most research findings, and this demonstrated that temperature was not a driving factor for CH₄ fluxes in Tomago saltmarsh.

The positive correlation between diurnal CO₂ flux and electrical conductivity was found to be contradictory to some research studies in which EC (linked to salinity) was negatively correlated with CO₂ flux due to high CO₂ absorbing capacity of alkaline wetland soils (Xie, Li, Zhai, Li, & Lan, 2009). The increase in EC in Tomago can be linked to the ionisation of acid salts while the increase in CO₂ flux was caused by high decomposition rates of organic matter in addition to high respiratory capacity of the ecosystem. Tomago is known to be one of the wetlands in the hunter estuary which was faced with the problem of formation of acid sulphate soils (Rogers, Saintilan, & Copeland, 2014; Saintilan, 2013) before tidal reinstatement. EC was not the controlling factor for CO₂ flux during January, 2016. The negative relationship between CO₂ flux and diurnal water temperature indicated that the later was

dependent upon solar radiation. Water temperature increased during the day when solar radiation is available to stimulate uptake of CO₂ by plants. During tidal inundation period, the marsh showed uptake of CO₂ in the day and at certain time intervals during the night, and this is explained by the solubility driven diffusion of CO₂ into the inflowing tidal water.

3.2 Seasonal variation of environmental factors and, CH₄ and CO₂ fluxes

Rainfall dilutes wetland water solutes causing a reduction in electrical conductivity, EC. EC is linked to salinity, which refers to the total soluble salts in water. Results indicated that the highest average rainfall received in January led to a dramatic decrease in EC (Figure 6). Rain water dissolves salts, and infiltrates and carries salts from the subsurface layers to deeper soil layers and thus reduces their concentrations in surface waters (Nachshon, Ireson, van der Kamp, Davies, & Wheeler, 2014).

Changes in precipitation patterns influenced the amount of soil water, and also the level of water above the ground level. The rain pulse that occurred during January resulted in increased flooding which in turn influenced CH₄ emissions in February due to favourable conditions for anaerobic microbial communities to perform their biochemical activities. Oxidation of CH₄ decreases when soil water content is high, and this could be attributed to the low solubility and diffusion of oxygen and CH₄ in the soil water as well (Del Grosso et al., 2000). At high moisture content levels, methanogenesis is highly favoured due to low diffusion and solubility rates of oxygen. In areas with water saturated soils, net oxidation has been reported due to the presence of anaerobic CH₄ oxidation (Khalil & Baggs, 2005), and this indicates the importance of flooding water in wetland soils in relation to CH₄ production and oxidation.

The formation of CH₄ depends on the quantity and composition of the methanogenic population present in a wetland ecosystem. High water salinity inhibits methanogenesis probably due to competition from sulphate reducing bacteria (Le Mer & Roger, 2001). Because of the low levels of salinity during February; there was high emission of CH₄ (0.37 $\mu\text{g m}^{-2} \text{s}^{-1}$). The lower the EC, the lower will be the salt ion concentration in water, and hence low salinity levels. The results therefore concur with previous work by Chambers et al., (2011) who stated that high salt concentration reduces microbial populations in wetlands, by lowering microbial respiration, and hence the rate of methanogenesis. The high emissions of CO₂ during the same period (January – February) can be explained by the ability of microbial populations to decompose organic matter, and the availability of organic matter in the sediments, in

addition to CO₂ that resulted from respiration of plants and animals. Although the reduction in EC was followed by a slight increase in CO₂ production rate, EC might have had more of an indirect influence on CO₂ flux since the reduction in EC was as a result of dilution due to rainfall.

High temperatures in summer are favourable for physiological processes in soil microbial processes, and plant respiration. This effect of temperature is supported with the research by Pulliam (1993) and Wang et al., (2016), from which they found that high temperatures promote the oxidative breakdown of organic matter and plant tissue (root and shoot) respiration. In a related study, it was found that wetlands act as sources of CO₂ if climate, hydrologic conditions and soil temperatures enhance oxidation of organic matter and root respiration (Altor & Mitsch, 2008).

4.0 Conclusions

CH₄ and CO₂ fluxes showed distinct diurnal variation patterns, with low CH₄ emissions during the night and high CH₄ emissions in the daytime, and the diurnal variation patterns demonstrated a relationship with water level and tidal heights. Almost all the diurnal patterns of CO₂ flux indicated CO₂ uptake during the day and CO₂ emissions at night with the exception of January, February and May when heavy rainfall occurred. CH₄ fluxes were linked to changes in EC and water level although these relationships were not statistically significant, while CO₂ flux was found to be driven by rainfall and water level. There was no definitive impact on carbon sequestration by the measurements that were carried out before and after tidal flow reinstatement although results demonstrate that freshwater events were seen to influence the carbon balance in the tidal marsh.

CH₄ uptake during the day and some hours in the night for the soil exposed stage (August, September, 2016), suggests a biogeochemical activity consuming CH₄ after its production. This research suggests that there might have been a possibility of aerobic microbial oxidation of CH₄, causing its uptake during August and September 2015, basing on the fact that during this period, there was no flooding in the wetland, and the soil was virtually exposed to air. The effect of reducing EC of the water in the marsh by rainfall between December and February was not a normal seasonal event. A lot of rainfall (225 mm) on 6th January was received and this contributed significantly towards flooding of the tidal marsh, leading to dilution of the tidal flooding water. Consequently, EC of the water reduced tremendously from 31321.09 µS cm⁻¹ in January to 21586.36 µS cm⁻¹ in February. Results indicated that after this rainfall event, EC started to increase from June through December 2016 as the inflow of tidal water continued. The effect of rainfall was noted to have

changed the behaviour of the tidal marsh system in relation to other environmental factors. For instance, there was no relationship between water temperature and EC not until after the December to February rainfall events that water temperature showed a positive relative relationship with electrical conductivity.

There was a very weak relationship between CH₄ flux and CO₂ flux ($R^2 = 0.101$, $p = 0.33$) which was not statistically significant. It is therefore likely that CH₄ flux might have had negligible influence on CO₂ flux during the study period. High seasonal CO₂ fluxes that occurred (max 0.14 mg m⁻² s⁻¹ in February) were due to respiration and decomposition of organic matter. Decomposition of organic matter occurs during aerobic and anaerobic processes although the former are more efficient and mainly produce CO₂ while the latter is much slower and can produce CH₄ in addition to CO₂ (Olsson et al., 2015). The negative relationship between diurnal water temperature and CO₂ fluxes was linked to the influence of solar radiation in stimulating photosynthesis.

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