Gross ecosystem photosynthesis causes a diurnal pattern in methane emission from rice

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[1] Understanding the relative contribution of environmental and substrate controls on rice paddy methanogenesis is critical for developing mechanistic models of landscape-scale methane (CH_4) flux. A diurnal pattern in observed rice paddy CH_4 flux has been attributed to fluctuations in soil temperature physically driving diffusive CH_4 transport from the soil to atmosphere. Here we make direct landscape-scale measurements of carbon dioxide and CH_4 fluxes and show that gross ecosystem photosynthesis (GEP) is the dominant cause of the diurnal pattern in CH_4 flux, even after accounting for the effects of soil temperature. The time series of GEP and CH_4 flux show strong spectral coherency throughout the rice growing season at the diurnal timescale, where the peak in GEP leads that of CH_4 flux by 1.3 ± 0.08 hours. By applying the method of conditional Granger causality in the spectral domain, we demonstrated that the diurnal pattern in CH_4 flux is primarily caused by GEP. **Citation:** Hatala, J. A., M. Detto, and D. D. Baldocchi (2012), Gross ecosystem photosynthesis causes a diurnal pattern in methane emission from rice, *Geophys. Res. Lett.*, *39*, L06409, doi:10.1029/2012GL051303.

1. Introduction

[2] Rice is the dominant staple food crop for over 5 billion people worldwide [*Hossain and Narcisco*, 2005] and contributes 11% of annual global methane (CH_4) emissions [*Smith et al.*, 2007]. Constraining carbon turnover times in rice paddy agroecosystems is particularly important for improving mechanistic predictions of CH_4 flux magnitude and timing. A diurnal pattern in CH_4 emissions from rice paddies has previously been attributed to daily fluctuations in temperature physically driving diffusive CH_4 transport [*Denier van der Gon and van Breemen*, 1993; *Hosono and Nouchi*, 1997; *Schütz et al.*, 1989]. Methanogens in rice paddy soils only produce CH_4 in reduced soil conditions [*Conrad*, 2007], and as a result many models treat CH_4 flux as a function of temperature and redox potential [*Li*, 2000].

[3] However, ecosystem CH_4 flux not only depends on physiochemical environmental conditions, but is also highly regulated by the ecological function of rice plants. Rice plants provide the dominant transport mechanism for CH_4 flux from soil to atmosphere by diffusive emission through their porous aerenchyma tissue [*Cicerone and Shetter*, 1981; *Holzapfel-Pschorn et al.*, 1986; *Nouchi et al.*, 1990] and they are the primary source of carbon substrates for metha-

nogenic metabolism on a range of timescales [*Cicerone et al.*, 1992; *Holzapfel-Pschorn et al.*, 1986; *Huang et al.*, 1997; *Minoda et al.*, 1996; *Sass et al.*, 2002]. Net ecosystem production is highly correlated with CH_4 flux across a range of climatically diverse wetlands, indicating that substrate supply is an important control on the magnitude of CH_4 flux [*Whiting and Chanton*, 1993]. Stable isotope labeling studies in controlled laboratory settings have revealed a strong transient link between rice photosynthesis and CH_4 flux, with time lags between plant carbon dioxide (CO_2) assimilation and CH_4 emission from 2 hours to 3 days [*Dannenberg and Conrad*, 1999; *Minoda and Kimura*, 1994; *Minoda et al.*, 1996].

[4] While there is a clear link between plant productivity and CH_4 flux at the plant and plot scale, the mechanisms controlling short-term CH_4 flux in the field remain unclear based on a history of CH_4 fluxes measured with soil chambers. Diurnal coupling between photosynthesis and heterotrophic microbial respiration is an emergent property of many ecosystems [*Kuzyakov et al.*, 2000; *Vargas et al.*, 2010], although most chamber-based studies that directly measure CH_4 flux are not accompanied by simultaneous measurements of photosynthesis. In this analysis, we tested the hypothesis that daily carbon substrate supply by rice photosynthesis, not soil temperature, causes the diurnal pattern in rice paddy CH_4 flux by using the eddy covariance technique to measure fluxes of CO_2, CH_4, and evaporation [*Baldocchi et al.*, 1988] over the course of a rice growing season.

2. Methods

[5] We examined the relative roles of gross ecosystem photosynthesis (GEP) and soil temperature in modulating the temporal spectrum of CH_4 flux with high frequency micrometeorological data collected continuously over the course of a growing season. We measured landscape-scale fluxes of CO_2, H_2O, CH_4, and energy at a rice paddy located on Twitchell Island, CA, USA (latitude: 38.1087°N, longitude: 121.6530°W; elevation: 4 m below sea level) from the emergence of rice seedlings on 15 June 2011 to harvest on 15 October 2011. The water table at the field was maintained at 5 cm above the soil surface for the duration of the growing season. Winds during the study period were strong in magnitude and stable in direction where the 90% flux area footprint fell entirely within the bounds of the rice paddy. We measured soil temperature at 2 cm depth below the soil surface with three replicate copper-constantan thermocouples at a rate of 0.2 Hz, recorded as half-hourly averages. At a height of 3.05 m and a rate of 10 Hz, we measured 3-dimensional wind velocities (u, v, w) with a sonic anemometer (Gill WindMaster Pro; Gill Instruments Ltd,

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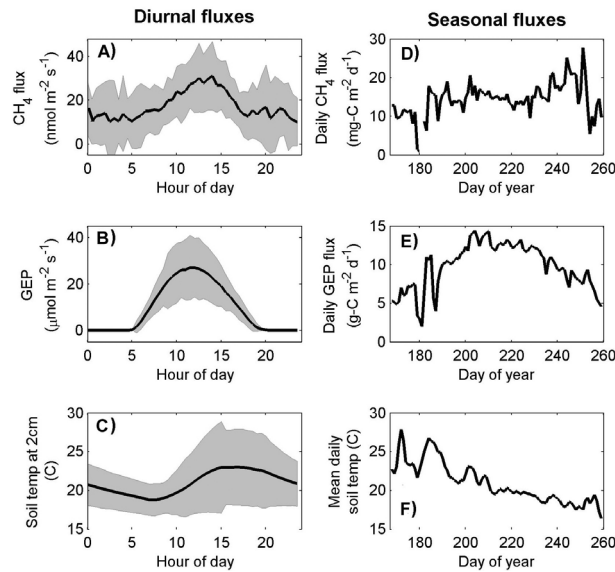[2]Now at Smithsonian Tropical Research Institute, Balboa, Panama.

Figure 1. (a) CH_4 flux, (b) gross ecosystem productivity (GEP), and (c) soil temperature at 2 cm depth all demonstrate a diurnal pattern for the duration of the rice growing season, where the solid line represents the growing season mean, and the shaded area represents the growing season standard deviation for each half-hour interval. The diurnal peak in GEP leads that of CH_4 flux, soil temperature, and evaporation, indicating that GEP is a more likely driver of the daily peak in CH_4 flux than soil temperature. The trend across the growing season for (d) CH_4 flux, (e) GEP, and (f) soil temperature at 2 cm depth. GEP shows strong seasonality with a mid-summer peak, while CH_4 flux increases throughout the growing season and soil temperature decreases over the course of the season. Low values of GEP at day of year 178–180 are caused by the application of a general herbicide to the field to control the growth of weeds with a concomitant decrease in CH_4 flux, indicating a strong link between these two variables.

Lymington, Hampshire, England), CO_2 and H_2O density with an open-path infrared gas analyzer (LI-7500; LI-COR Biogeosciences, Lincoln NE, USA), and CH_4 density with a closed-path tunable diode laser CH_4 analyzer (FMA, Los Gatos Research, CA, USA). This sampling rate allowed for a 5 Hz cut-off for the co-spectra between the scalars (CO_2, CH_4, H_2O) and turbulence, which was adequate for eddy covariance measurements at this site [*Detto et al.*, 2011]. Additional micrometeorological instrumentation (air temperature, humidity, barometric pressure, incoming and net radiation, precipitation, and water table depth) is detailed by *Hatala et al.* [2012].

[6] Using standard eddy covariance processing techniques, we analyzed fluxes of CO_2, H_2O, CH_4, and heat after applying corrections with in-house software [*Detto et al.*, 2010] explained in detail by *Hatala et al.* [2012]. Briefly, the procedure removed artificial data spikes (greater than six standard deviations from the mean) from the 10 Hz data and filtered bad readings that resulted from very infrequent fog events. For each 30-minute block of 10 Hz values, we applied a coordinate rotation to align the mean vertical and lateral wind velocities to zero and removed effects of air density fluctuations by the Webb-Pearman-Leuning

correction [*Detto and Katul*, 2007; *Webb et al.*, 1980]. We applied co-spectral corrections to CO_2, H_2O, and CH_4 fluxes to account for sensor separation, and additional co-spectral corrections to CH_4 fluxes to correct for tube attenuation, residence time in the analyzer cell, and small changes in analyzer flow rate [*Detto et al.*, 2011]. We filtered 30-minute flux values with anomalously high and low friction velocity (u* > 1.2 m/s and |uw| < 0.02 m/s) to constrain our analysis to periods where the air near the sensors was well-mixed. Of all possible 30-minute flux values during the growing season period in this analysis, 9% of CO_2, H_2O, and CH_4 fluxes were eliminated due to low friction velocity and an additional 10% of half-hourly CH_4 fluxes were not available due to brief FMA sensor malfunction.

[7] We gap-filled CO_2 fluxes using an artificial neural network approach standardized within the international Fluxnet project with meteorological variables driving the fitting [*Papale et al.*, 2006]. To partition CO_2 fluxes into gross ecosystem photosynthesis (GEP) and ecosystem respiration (R_{eco}), we extrapolated nighttime CO_2 flux as R_{eco} using a short-term (2 week) 2 cm-depth soil temperature response and subtracted daytime R_{eco} from net CO_2 flux to obtain GEP [*Reichstein et al.*, 2005]. To avoid spurious correlations between CH_4 flux and GEP, we did not gap-fill CH_4 flux data with the artificial neural network technique, and instead replaced missing values with the median for the entire growing season. There were no gaps in the soil temperature time series for this measurement period. For spectral analysis, we standardized the CH_4 flux, GEP, and soil temperature time series to have zero mean and unit variance.

2.1. Spectral Analysis Methods

[8] We used the continuous wavelet transform (CWT) with the Morlet mother wavelet to examine correlation between the spectra of GEP, CH_4 flux, and soil temperature [*Torrence and Compo*, 1998]. Compared with Fourier analysis, wavelet spectral analysis is a more powerful tool for analyzing geophysical time series with nonstationarity, including trace-gas flux data measured by eddy covariance [*Katul et al.*, 2001; *Vargas et al.*, 2010]. The wavelet coherence spectrum is interpreted as the local correlation between two variables in frequency-time space where high coherence indicates phase-locked behavior between the two time series [*Grinsted et al.*, 2004]. We tested the statistical significance of wavelet power against the null hypothesis of a red noise first order autoregressive process with lag-1 autocorrelation [*Grinsted et al.*, 2004]. For time periods with significant wavelet coherence, we used the phase angle to calculate the time lag between the correlated oscillations of the two series.

[9] We used the method of Granger causality to determine whether the patterns of wavelet coherence between the time series of GEP and temperature and CH_4 flux represented causal relationships. Granger causality is a method whereby a time series of one variable is determined to cause a second time series if it can successfully be used to predict the response of the second lagged time series [*Granger*, 1988], and here we applied the principles of conditional Granger causality to the nonparametric spectral domain [*Chen et al.*, 2006; *Dhamala et al.*, 2008] (auxiliary material).[1] We tested

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[1]Auxiliary materials are available in the HTML. doi:10.1029/2012GL051303.

A) GEP-CH$_4$ Flux
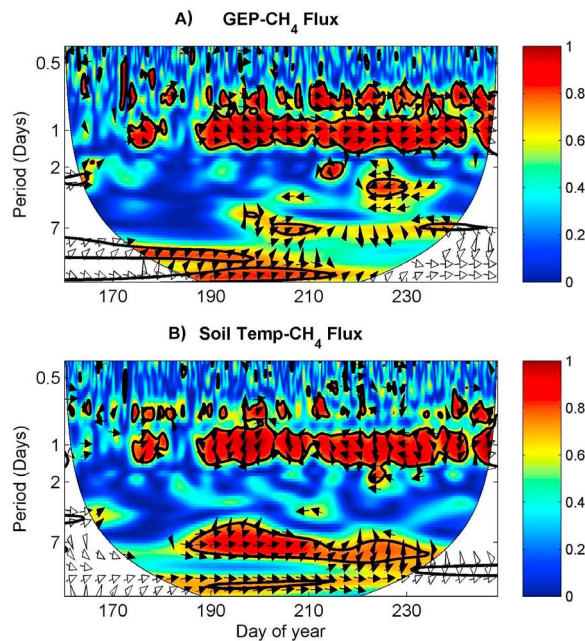
B) Soil Temp-CH$_4$ Flux

Figure 2. (a) The wavelet coherence between GEP and CH$_4$ flux for the rice paddy growing season shows high in-phase coherence between the two time series at the daily timescale for the duration of the growing season, and lower periodic coherence at the 18 hour, weekly, and bi-weekly timescales. (b) Soil temperature and CH$_4$ flux have the highest coherency at the daily and weekly timescale, whereas soil temperature and GEP are coherent at the daily timescale. Significant coherency (at the 5% level with 1000 Monte Carlo simulations of AR-1 autocorrelation) is indicated by the bold black lines. The direction of arrows indicate the phase angle between the two time series, where an arrow with an inclination of zero pointed to the right indicates zero lag (the series are perfectly correlated). The cone of influence represents the limit where wavelet power dropped to e^{-2} of the edge values.

the significance of nonparametric Granger causality against the null hypothesis that no direct interaction exists by the iterative amplitude adjusted Fourier transform, which preserves the power spectrum and distribution of the original time series but eliminates the correlation structure [*Molini et al.*, 2010]. We tested the hypothesis that the periodic signal of GEP caused a periodic response in CH$_4$ flux after we conditioned the response for the effect of soil temperature on CH$_4$ flux. Support for this hypothesis indicates that GEP modulates CH$_4$ flux at the time lags indicated by the spectral analysis.

3. Results and Discussion

[10] GEP, CH$_4$ flux, and soil temperature all demonstrated a strong diurnal pattern for the duration of the growing season, where the daily peak in GEP leads that of CH$_4$ flux and soil temperature (Figures 1a–1c). The mean growing season peak in GEP occurred in late morning at 11:00 hours, the mean peak in soil temperature occurred in late afternoon at 16:30 hours, and the mean peak in CH$_4$ flux occurred in mid-afternoon at 14:30 hours. If either GEP or soil temperature are driving the diurnal pattern in CH$_4$ flux, we would predict that the diurnal peak of GEP or soil temperature would temporally lead the peak in CH$_4$ flux. However, this is not the case, as GEP leads CH$_4$ flux, but the peak in soil temperature lags both of these variables. All variables also demonstrate coherent seasonal trends, where GEP peaks mid-season, CH$_4$ flux increases and soil temperature at 2 cm depth decreases throughout the season (Figures 1d–1f). The contrary seasonal pattern of decreasing soil temperature but increasing CH$_4$ flux provides further evidence that soil temperature might not be driving CH$_4$ flux. The application of a general herbicide for weed control in the field at day of year 179 caused a sharp decline in GEP (Figure 1e) with a concomitant decrease in CH$_4$ flux (Figure 1d), also providing qualitative mechanistic insight regarding the strong control of GEP on CH$_4$ emission.

[11] We examined the wavelet coherence spectra [*Grinsted et al.*, 2004; *Torrence and Compo*, 1998] to determine the dominant timescales and strength of
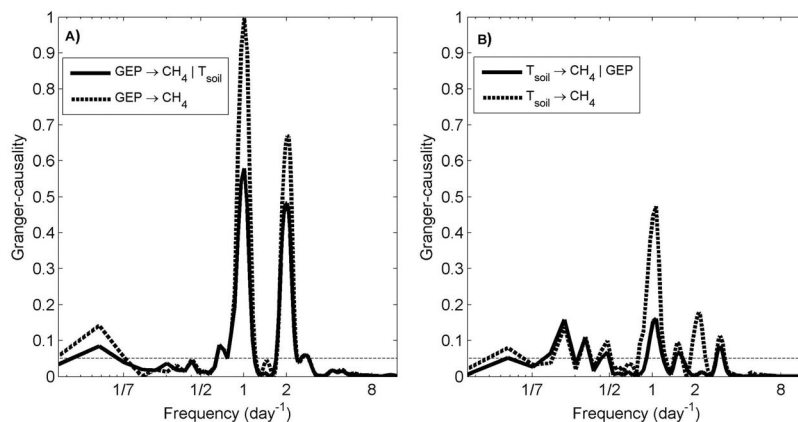

Figure 3. The Granger-causality spectra are plotted for the causal relationships between (a) GEP and (b) soil temperature and CH$_4$ flux. When conditioned on soil temperature, GEP still has a strong causal relationship with CH$_4$ flux at the daily timescale. Conversely, when soil temperature is conditioned on GEP the Granger-causality at the daily timescale becomes insignificant, although temperature is still a significant driver of CH$_4$ flux for frequencies smaller than 0.5 day^{-1}.

coupling between CH$_4$ flux and GEP and soil temperature. GEP and CH$_4$ flux are strongly coherent throughout the growing season at the daily timescale, where the mean lag time with 95% confidence interval is 1.3 ± 0.08 hours (0.350 ± 0.021 radians) (Figure 2a). The soil temperature and CH$_4$ flux time series are also significantly coherent at the daily period (Figure 2b), however soil temperature lags CH$_4$ flux by 5.5 ± 0.1 hours (-1.47 ± 0.030 radians). The soil temperature-CH$_4$ flux wavelet coherence is also significantly coherent at the weekly time period (Figure 2b), and the mean time lag here is 14 ± 1.8 hours (0.52 ± 0.079 radians). Although both GEP and soil temperature are correlated with CH$_4$ flux at multiple timescales, strong coherencies occur at the daily timescale that are maintained for the duration of the growing season. Soil temperature is also highly correlated with CH$_4$ flux at the weekly period, which might indicate a relationship where soil temperature drives kinetic rate changes in CH$_4$ flux on longer timescales.

[12] From the spectral Granger causality analysis, we found strong support for the hypothesis that GEP modulates CH$_4$ flux at the daily timescale. The GEP-CH$_4$ flux Granger causality spectrum (Figure 3a) shows strong power at the daily timescale as well as at the harmonic 12-hour timescale after accounting for the effects of soil temperature on CH$_4$ flux. The spectrum of GEP-induced CH$_4$ flux demonstrates that carbon cycling between plants and methanogens occurs rapidly with a coherent temporal pattern that is maintained for the duration of the growing season. We also tested the alternative hypothesis that soil temperature causes a diurnal pattern in CH$_4$ flux, and conditioned this relationship on GEP. Soil temperature modulates CH$_4$ flux over a longer five-day timescale and demonstrates a much weaker daily signal than that of GEP-induced CH$_4$ flux (Figure 3b). Since a few observations have suggested that stomatal conductance might drive CH$_4$ flux [*Chanton et al.*, 1997], we tested but did not find strong support for this other alternate mechanism (auxiliary material).

4. Conclusions

[13] Understanding the temporal lags of carbon turnover from plants to methanogens is essential for scaling methanogenesis to ecosystem-level CH$_4$ flux. Our analysis concludes that in rice, CH$_4$ flux rapidly response to GEP in a coherent pattern for the duration of the growing season. Although this analysis is conducted in a spatially homogeneous rice paddy, it may yield insight into the high-frequency mechanisms that contribute to variability in CH$_4$ flux measurements at spatially heterogeneous sites. For example, if photosynthetic rates vary across the landscape and high-frequency CH$_4$ flux is driven by GEP, accurately measuring and modeling photosynthesis might help explain at least some of the heterogeneity in CH$_4$ flux. The diurnal pattern of GEP-regulated CH$_4$ fluxes also has direct implications for the daily and seasonal extrapolation of studies that measure only daytime CH$_4$ flux, due to an inability to account for diurnal variation in CH$_4$ flux due to changes in GEP. Furthermore, the strong connection between GEP and CH$_4$ flux found in this study highlights a possible trade-off in using flooded ecosystems for carbon capture and sequestration, a subject of research that warrants further study at other sites. This analysis re-examines assumptions about the importance of biotic and abiotic factors in

regulating landscape-scale CH$_4$ flux on the timescale of hours to days, and concludes that gross ecosystem photosynthesis is the primary cause of the diurnal pattern in rice paddy CH$_4$ flux.

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