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Seasonal variability of tropical wetland CH$_4$ emissions: the role of the methanogen-available carbon pool

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Abstract. We develop a dynamic methanogen-available carbon model (DMCM) to quantify the role of the methanogen-available carbon pool in determining the spatial and temporal variability of tropical wetland CH$_4$ emissions over seasonal timescales. We fit DMCM parameters to satellite observations of CH$_4$ columns from SCIAMACHY CH$_4$ and equivalent water height (EWH) from GRACE. Over the Amazon River basin we found substantial seasonal variability of this carbon pool (coefficient of variation = 28 ± 22 %) and a rapid decay constant ($\phi = 0.017$ day$^{-1}$), in agreement with available laboratory measurements, suggesting that plant litter is likely the prominent methanogen carbon source over this region. Using the DMCM we derived global CH$_4$ emissions for 2003–2009, and determined the resulting seasonal variability of atmospheric CH$_4$ on a global scale using the GEOS-Chem atmospheric chemistry and transport model. First, we estimated that tropical emissions amounted to 111.1 Tg CH$_4$ yr$^{-1}$, of which 24 % was emitted from Amazon wetlands. We estimated that annual tropical wetland emissions increased by 3.4 Tg CH$_4$ yr$^{-1}$ between 2003 and 2009. Second, we found that the model was able to reproduce the observed seasonal lag of CH$_4$ concentrations peaking 1–3 months before peak EWH values. We also found that our estimates of CH$_4$ emissions substantially improved the comparison between the model and observed CH$_4$ surface concentrations ($r = 0.9$). We anticipate that these new insights from the DMCM represent a fundamental step in parameterising tropical wetland CH$_4$ emissions and quantifying the seasonal variability and future trends of tropical CH$_4$ emissions.

1 Introduction

Wetlands are the single largest source of methane (CH$_4$) into the atmosphere and account for 20–40 % of the global CH$_4$ source (Denman et al., 2007; Ito and Inatomi, 2012), of which tropical wetlands account for 50–60 % of this global wetland CH$_4$ source (e.g. Cao et al., 1996; Bloom et al., 2010). Tropical wetland biogeochemistry is poorly understood compared to boreal peatlands (Mitsch et al., 2010), resulting in large inter-model discrepancies of the magnitude and distribution of tropical wetland CH$_4$ emission estimates (Riley et al., 2011). Tropical climate variability (e.g. resulting in widespread droughts, Lewis et al., 2011) can lead to large year to year variations in tropical wetland CH$_4$ emissions and subsequently the global CH$_4$ budget (Hodson et al., 2011). Moreover, Bousquet et al. (2011) found substantial disagreements between tropical wetland CH$_4$ emissions from process-based and atmospheric inversion estimates. An improved quantitative understanding of the magnitude, distribution, and variation of tropical wetland CH$_4$ emissions is therefore essential to further understanding of the global CH$_4$ cycle. Here, we parameterise tropical wetland CH$_4$ emissions, and hence introduce a predictive capability that can be used to determine future emissions and to help quantify global CH$_4$ climate feedbacks.

In wetlands and rice paddies, methanogenesis (the biogenic production of CH$_4$) occurs as the final step of anoxic organic matter decomposition (Neue et al., 1997). Factors influencing methanogenesis rates include substrate availability, soil pH, temperature, water table position and redox potential (Whalen, 2005). Wetland vegetation type and aquatic herbivore activity can also affect the transport of CH$_4$ between the soil and atmosphere (Joabsson et al., 1999; Dingemans et al.,...
2822

A. A. Bloom et al.: Seasonal variability of tropical wetland CH₄ emissions

2011). On a global scale, seasonal variations in wetland CH₄ fluxes are mostly determined by temporal changes in wetland water volume and soil temperature (Walter et al., 2001; Gedney et al., 2004), and from seasonal changes in wetland extent and wetland water table depth (Ringeval et al., 2010; Bloom et al., 2010). Recent work that used SCIAMACHY lower tropospheric CH₄ column concentrations and Gravity Recovery And Climate Experiment (GRACE) equivalent water height (EWH) retrievals showed that the seasonality of wetland CH₄ emissions can be largely explained by seasonal changes in surface temperature and water volume (Bloom et al., 2010). The Amazon and Congo River basins were the only major exceptions in this study, where CH₄ concentrations peaked several weeks before EWH, highlighting our incomplete understanding of the processes controlling tropical wetland CH₄ emissions over seasonal timescales.

In this paper we focus on the seasonal lag between CH₄ emissions and flooding over the Amazon River basin area (Oki and Sud, 1998). We use SCIAMACHY CH₄ retrievals and GRACE EWH (both described in Sect. 2.2) to determine the seasonal lag between wetland CH₄ emissions and wetland water volume. Figure 1 shows that seasonal flooding of the Amazon basin occurs typically 1–3 months after the peak CH₄ concentrations, and to a lesser extent the lag persists throughout tropical wetland areas. Although typical time-lags between EWH and CH₄ in the Congo River basin are comparable (0–2 months), in this paper we choose to focus on the Amazon basin as it covers a larger area extent, and larger time-lags between EWH and CH₄ are found over this river basin. In Sect. 2, we test the hypothesis that this lag is related to the depletion of methanogen-available carbon during the onset of the tropical wet season by explicitly accounting for this carbon pool in a parameterised model of tropical wetland CH₄ emissions (Bloom et al., 2010). We optimise model parameters by fitting them to SCIAMACHY CH₄ column and GRACE EWH measurements, and use the resulting model to estimate global wetland emission estimates. In Sect. 3 we (1) compare our results to previous estimates of wetland CH₄ emissions and to decomposition rates of methanogen-available carbon in anaerobic environments; (2) provide an overview of additional factors which potentially influence the seasonal variability of CH₄ emissions in tropical wetlands; and (3) use our estimated emission rates to drive the GEOS-Chem atmospheric chemistry model as an approach to test the consistency between our emission estimates and observed variations of atmospheric CH₄ concentration. We conclude the paper in Sect. 4.

2 Process-based model and application

Here, we introduce a methanogen-available carbon pool (Cₙ) that typically originates from labile plant litter, recalcitrant organic matter decomposition and root exudates (e.g. Wania et al., 2010). Typically, soil carbon pool decay constants are more than an order of magnitude lower than those of leaf litter (Sich et al., 2003; Wania et al., 2010). Therefore, if Cₙ originates mostly from the slow-decomposing recalcitrant carbon pool, then variations in Cₙ over seasonal timescales are likely to be small. Conversely, if Cₙ is drawn from leaf litter, then large variations in Cₙ abundance may arise as a result of rapid litter decomposition in the tropics. Miyajima et al. (1997) measured CH₄ accumulation of anaerobic decomposition of incubated tropical withered tree leaves over a 200 day period. These observations showed a rapid decrease in decomposition rates over the incubation period. Bianchini Jr. et al. (2010) found similar results for anaerobic decomposition from dried and ground Oxycaryum cubense at 20°C: following a 20-day lag (where no emissions were observed) CH₄ produced from organic carbon decomposition peaked after a 50-day period, and then rapidly decreased. On a tropical river-basin scale, flooded areas expand at the onset of the wet season and engulf newly available plant litter; as a result, CH₄ emissions from plant litter may peak before the height of the water table. The occurrence of anaerobic CH₄ emissions from litter decomposition within sub-seasonal timescales raises the question as to whether Cₙ significantly varies in time.

2.1 Model description

We base our model on previous work (Bloom et al., 2010) that describes the temporal variability of wetland emissions FCH₄ₜ (mg CH₄ m⁻² day⁻¹) as a function of EWH and surface temperature:

\[ F_{CH₄}^{t} = k(T_{w}^{t} + D_{a})Q_{10}(T_{s})^{\frac{T_{s}}{10}}, \]

where at time t (days), Γₗ is the EWH, T_s is the surface temperature (K), D_a is the equivalent depth of the wetland soil (m), Q_{10}(T_s) is the temperature dependence function implemented by Gedney et al. (2004), and k is a scaling constant (mg CH₄ m⁻² day⁻¹) accounting for all temporally constant factors (e.g. Gedney et al., 2004).

Equation (1) assumes an inexhaustible source of methanogen-available carbon. Here, we account for the potential seasonal changes in Cₙ by substituting k with \(\phi_{0}C_{\nu}^{t}\), where \(\phi_{0}\) (day⁻¹) is the water and carbon independent decay constant of wetland methanogenesis, and Cₙ is the value of Cₙ (mg CH₄ m⁻²) at time t:

\[ F_{CH₄}^{t} = \phi_{0}C_{\nu}^{t}(T_{w}^{t} + D_{a})Q_{10}(T_{s})^{\frac{T_{s}}{10}}. \]

To determine temporal changes in Cₙ, we define Cₙ+1 in terms of Cₙ:

\[ C_{\nu}^{t+1} = C_{\nu}^{t} + \Delta t - F_{CH₄}^{t} \Delta t, \]

where \(\Delta t\) is the time interval, \(F_{CH₄}^{t}\) is the carbon loss due to emitted CH₄ (Eq. 2), and \(\Delta t\) is the net influx of carbon...
available for methanogenesis from plant litter, root exudates, and breakdown of complex polymers from the recalcitrant carbon pool. We assume $N_\mu$ is temporally constant, and we assume wetland carbon stocks are in quasi-equilibrium on annual timescales, hence $\overline{N}_\mu = \frac{F_\mu}{c_\mu}$. Note that when $\phi_0$ is small, the equilibrium $C_\mu \gg N_\mu \Delta t$. In this case, $C_\mu^{t+1} \approx C_\mu^t$ and Eq. (2) converges to Eq. (1) (Bloom et al., 2010), which assumes $\phi_0 C_\mu$ is constant over seasonal timescales. In order to compare derived decay constants with observed and model values (e.g. Miyajima et al., 1997; Wania et al., 2010), we determine the annual mean decay constant of wetlands areas as $\overline{\phi} = \frac{F_\mu}{c_\mu} \phi_\mu$ (day$^{-1}$). Equations (2) and (3) constitute the dynamic methanogen-available carbon model (DMCM).

2.2 Data

For the sake of brevity, we only include a brief description of the datasets for our analysis and refer the reader to dedicated papers. Solar backscatter data from the Scanning Imaging Absorption Spectrometer for Atmospheric Cartography (SCIAMACHY) instrument onboard Envisat was used to retrieve the mean column concentrations of CH$_4$ in the atmosphere (Frankenberg et al., 2005). The spatial resolution of CH$_4$ retrievals is 30 km × 60 km, and the Envisat orbital geometry ensures global coverage at 6-day intervals. CH$_4$ retrievals were only achievable in daytime cloud-free conditions. The Gravity Recovery and Climate Experiment (GRACE) is a twin satellite system from which the Earth’s gravity field was retrieved at 10-day intervals. Tides, atmospheric pressure and wind are included in the applied corrections on GRACE gravity retrievals; the remaining temporal variation in GRACE gravity is dominated by terrestrial water variability (Tapley et al., 2004). We incorporated SCIAMACHY CH$_4$ concentrations, GRACE EWH and NCEP/NCAR daily $1.9^\circ \times 1.88^\circ$ temperature re-analyses (Kalnay et al., 1996) into a process-based model following Bloom et al. (2010). We used the 2003–2008 SCIAMACHY column CH$_4$ retrievals (Frankenberg et al., 2008), and the CNES GRACE EWH $1^\circ \times 1^\circ$ 10-day resolution product (Lemoine et al., 2007). We aggregated all three datasets to a daily $3^\circ \times 3^\circ$ horizontal grid (see Bloom et al., 2010).

2.3 Global parameter optimisation

We implemented the DMCM (as shown in Eqs. (2) and (3)) on a global $3^\circ \times 3^\circ$ grid for the period 2003–2009. We drove the DMCM using the aggregated daily values of $T_s^t$ and $\Gamma_w^t$. We spun up the DMCM using 2003 $T_s^t$ and $\Gamma_w^t$ values until it reached an annual equilibrium ($\overline{N}_\mu = \frac{F_\mu}{c_\mu}$). In contrast to Bloom et al. (2010), we supplemented the $Q_{10}(T_s)$ function with a gradual linear cut-off for temperatures for $0^\circ\mathrm{C} < T_s^t < -10^\circ\mathrm{C}$, and when $T_s^t < -10^\circ\mathrm{C}$, $F_\mu$ = 0 as a first order approximation to wintertime CH$_4$ emission inhibition in boreal wetlands. As the $Q_{10}$ function never reached zero, this supplementary constraint effectively suppressed wintertime CH$_4$ emissions, which is broadly consistent with our current understanding of CH$_4$ emissions in boreal wetlands. We applied the DMCM globally in order to determine (i) the magnitude of $\phi_\mu$ and $c_\mu$ in the tropics within each $3^\circ \times 3^\circ$ gridcell, (ii) the potential of $c_\mu$ temporal variability on extratropical wetland environments, and (iii) CH$_4$ emissions from wetlands and rice paddies at a global scale. We determined
the global distribution and seasonal variability of wetland CH$_4$ emissions by optimising parameters $\phi_0$ and $D_0$ at each gridcell by minimising the following cost function ($J$):

$$J = \sum_{i=1}^{n} (\kappa \times \Delta F_{\text{CH}_4}^i - \Delta S_{\text{CH}_4}^i)^2,$$

where $\Delta S_{\text{CH}_4}^i$ denotes the SCIAMACHY CH$_4$ variability, $\Delta F_{\text{CH}_4}^i$ is derived from Eqs. (2) and (3), and the conversion factor $\kappa$ (ppm kg$^{-1}$ CH$_4$ m$^{-2}$ day$^{-1}$) relates CH$_4$ emissions to the equivalent column concentration in the lower troposphere (see Bloom et al., 2010). We removed the interannual trend (represented as a second order polynomial) from $\Delta S_{\text{CH}_4}^i$ in order to minimize the influence of global atmospheric CH$_4$ trends. We then implemented the global $Q_{10}(T_s)$ optimisation approach of Bloom et al. (2010). Like other top-down parameter optimisation methods of global wetland CH$_4$ emissions (Gedney et al., 2004; Bloom et al., 2010), our method was unable to distinguish between the seasonality of CH$_4$ emissions from wetlands and rice paddies due to the concurring fluxes over seasonal timescales. However we anticipated that varying fertilisation and irrigation practices also influence the seasonality in rice paddy CH$_4$ emissions (Conen et al., 2010). We hence distinguished the sources spatially (Bloom et al., 2010), for which we had more confidence in the distribution of rice paddies. Finally, we used the IPCC global wetland and rice paddy CH$_4$ emissions median of 227.5 Tg CH$_4$ yr$^{-1}$ (Denman et al., 2007) as a base value for 2003 emissions.

We propagated the following uncertainties through our global wetland and rice paddy CH$_4$ emissions estimation (Bloom et al., 2010): (i) SCIAMACHY CH$_4$ observation errors; (ii) the uncertainty of the linear fit between $F_{\text{CH}_4}$ and $S_{\text{CH}_4}$; (iii) the uncertainty $\sigma_\kappa = \pm 16$ % associated with $\kappa$; and (iv) a global wetland and rice paddy uncertainty of $\pm 58$ Tg CH$_4$ yr$^{-1}$ (Denman et al., 2007).

We propagated SCIAMACHY CH$_4$ VMR errors to a $3^\circ \times 3^\circ$ resolution; compared to a global mean of $\pm 18.0$ ppb, we found mean CH$_4$ error values of $\pm 19.2$ ppb (5th–95th percentile = 6.3–37.8 ppb) over the Amazon River basin. Temporal CH$_4$ VMR error variability was dominated by the number of cloud-free CH$_4$ VMR observations within each $3^\circ \times 3^\circ$ gridcell for each daily timestep. We found little seasonal variability in the three-monthly mean propagated CH$_4$ errors (18.28–20.36 ppb). As the correlation of CH$_4$ errors within each gridcell was unknown, we chose not to weight the cost function (Eq. 4) using propagated SCIAMACHY CH$_4$ errors.

We incorporated the uncertainty of $\kappa$, $\sigma_\kappa = \pm 16$ % in our estimated CH$_4$ emissions, where $\sigma_\kappa$ is the estimated uncertainty between surface CH$_4$ emission amplitude and CH$_4$ column VMR amplitude; this value was derived from an atmospheric chemistry transport model (GEOS-Chem) using prior emissions and SCIAMACHY averaging kernels (Bloom et al., 2010). Finally, we implemented a global wetland and rice paddy uncertainty equivalent to the variance of IPCC wetland emission estimates ($\pm 58$ Tg CH$_4$ yr$^{-1}$).

### 3 Results and discussion

Over the Amazon River basin, we find wetland CH$_4$ fluxes coincide with small values of $C_\mu$, resulting in a highly variable $C_\mu$ over seasonal timescales. Assuming an annual mean inundated fraction of 3.3 % (Prigent et al., 2007), the median CH$_4$ flux over a flooded area is 1.06 Mg C ha$^{-1}$ yr$^{-1}$ (387 mg CH$_4$ m$^{-2}$ day$^{-1}$). The median Amazon basin $C_\mu = 0.16$ Mg C ha$^{-1}$ yr$^{-1}$ with a range of 0.02–7.89 Mg C ha$^{-1}$ yr$^{-1}$ (5th–95th percentile). The large spatial variability of $C_\mu$ is consistent with the complexity of methanogenesis rates in wetlands (Neue et al., 1997; Whalen, 2005). Large temporal changes of $C_\mu$ are observed in the Amazon River basin, where the mean $C_\mu$ coefficient of variation ($C_v$) is $28 \pm 22$ % over the period 2003–2009. When we allow $C_\mu$ to vary in extra-tropical regions, we find a median of $C_v < 0.1$ %, and as a result the relatively small $C_\mu$ variability does not influence the seasonality of CH$_4$ emissions outside the tropics. For rice paddy areas in southeast Asia we find a median of $C_v = 4.8$ %. We acknowledge that due to the varying rice cultivation practices around the world (Conen et al., 2010), the effects of rice paddy irrigation and the timing of fertilisation on $C_\mu$ cannot be captured by the DMCM approach.

To determine whether our derived values for $C_\mu$ and $\phi$ are relevant to tropical ecosystems, we compared them against laboratory measurements of anaerobic decomposition of withered leaves from a wetland region in Narathiwat, Thailand (Miyajima et al., 1997). We simulated CH$_4$ production from $C_\mu$ at each model gridcell for a 200-day period without fresh carbon input ($N_p=0$), and we used inundated fraction observations (Prigent et al., 2007) to determine the flux magnitude over flooded areas only. Figure 2 shows the cumulative CH$_4$ production over a 200-day period for (i) simulated decomposition from derived $\phi$ and $C_\mu$ values over the Amazon, (ii) simulated decomposition from derived $\phi$ and $C_\mu$ values over boreal wetlands, and (iii) upscaled withered leaf mineralisation rates by Miyajima et al. (1997) using a median of 17.5 Mg C ha$^{-1}$ yr$^{-1}$ fine and coarse woody debris (Malhi et al., 2009). For boreal and tropical $C_\mu$ decomposition, the median cumulative CH$_4$ emissions, 68 % confidence interval, and mean decay constants ($\langle \phi \rangle$) are shown. For the withered leaf mineralisation rates, we show the mean fitted decay constant ($\langle \phi \rangle$) and the range and median cumulative CH$_4$ emissions.

The top-down parameter estimation of $\phi$ and $C_\mu$ suggest plant litter $C_\mu$ is a fundamental component of tropical CH$_4$ emission seasonality. Our top-down estimation of anaerobic decomposition rates for tropical wetland CH$_4$ emissions compare favourably with laboratory measurements of anaerobically produced CH$_4$; while the magnitude of tropical
A. A. Bloom et al.: Seasonal variability of tropical wetland CH\textsubscript{4} emissions

Table 1. Model and observed decay constants for organic matter decomposition in anaerobic environments.

<table>
<thead>
<tr>
<th>Study</th>
<th>Decay Constant (yr\textsuperscript{-1})</th>
<th>Study</th>
</tr>
</thead>
<tbody>
<tr>
<td>This Study: Top-down wetland CH\textsubscript{4} emission parameter optimisation</td>
<td>2.6—9.6\textsuperscript{a} (median=5.9)</td>
<td>Withered Leaves (35 °C)</td>
</tr>
<tr>
<td>Miyajima et al. (1997): Decay constant from anaerobic tropical leaf CH\textsubscript{4} mineralisation</td>
<td>4.0</td>
<td></td>
</tr>
<tr>
<td>Longhi et al. (2008): Measured decomposition rates in Paluda di Ostiglia, Italy</td>
<td>1.0—5.5</td>
<td></td>
</tr>
<tr>
<td>Wania et al. (2010): Bottom-up CH\textsubscript{4} Emissions from Northern Peatlands</td>
<td>0.001—0.03</td>
<td></td>
</tr>
<tr>
<td>Root Exudates (10 °C)</td>
<td>0.35</td>
<td></td>
</tr>
<tr>
<td>Root Exudates (10 °C)</td>
<td>13</td>
<td></td>
</tr>
</tbody>
</table>

\textsuperscript{a} 68\% confidence interval

\textsuperscript{b} Mass-loss decomposition rates

Fig. 2. Cumulative CH\textsubscript{4} emissions over a 200-day period from model C\mu mineralisation and incubated withered leaves. Blue: median and range of values from Miyajima et al. (1997). Red (green): median and 68\% confidence interval range of CH\textsubscript{4} emissions from the Amazon River basin (boreal wetland) from C\mu and \phi values when N\mu = 0. A total litter stock of 17.5 Mg C ha\textsuperscript{-1} (Malhi et al., 2009) was used to upscale the Miyajima et al. (1997) CH\textsubscript{4} mineralisation rates.

C\mu decomposition is more than a factor of two smaller than laboratory measurements (Miyajima et al., 1997), the mean decay constant \bar{\phi}_{\text{Amazon}} = 0.017 day\textsuperscript{-1} compares well to \bar{\phi}_{\text{leaf}} = 0.011 day\textsuperscript{-1} for withered leaf decomposition. The larger laboratory measurements (Miyajima et al., 1997) are partially explained by an incubation temperature of 35 °C (cf. a mean surface temperature in the Amazon basin of 23 °C), and the lack of observations for coarse woody debris decomposition. As a result of relatively high \phi values, measured leaf decomposition and model CH\textsubscript{4} emissions both show a significant reduction of CH\textsubscript{4} emission rates throughout the 200-day period. In contrast, the boreal decay constant (\bar{\phi}_{\text{Boreal}} = 0.0003 day\textsuperscript{-1}) indicates relatively constant CH\textsubscript{4} emission rates throughout the 200-day period.

Fig. 3. Daily wetland CH\textsubscript{4} emissions for 2003–2009 (blue) and GRACE equivalent water height (green) over the central branch of the Amazon River (0°–6° S, 40°–80° W).

Figure 3 shows the total CH\textsubscript{4} flux over the central branch of the Amazon River (0°–6° S, 40°–80° W). The temporal changes in C\mu result in a significantly different timing for CH\textsubscript{4} emissions over the tropics in comparison to the Bloom et al. (2010) water volume and temperature dependence
approach. While in the dry season the minimum CH$_4$ fluxes coincide with the lowest GRACE EWH, peak CH$_4$ fluxes occur during the rising water phase. The DMCM optimisation predicts that the accumulation of carbon in the dry season results in higher $C_{\mu}$ values at the onset of the wet season. This carbon pool is then rapidly depleted during the wet season. As a result, CH$_4$ emission rates begin to decrease before the peak water phase in the wet season.

In order to determine the importance of temperature variability in estimating $\Phi$ and the $C_{\mu}$ coefficient of variation, $c_{\nu}$, we performed a temperature-driven sensitivity analysis on the DMCM. Using a range of $Q_{10}(T_\nu) = 1.2 - 4$ to repeat the global parameter optimisation (Sect. 2.3), we derived a corresponding ranges of $\bar{\Phi} = 0.018 - 0.012$ and $c_{\nu} = 20.2 - 31.3\%$, respectively, for the Amazon River basin. Hence, larger $C_{\mu}$ seasonality is associated with higher $Q_{10}(T_\nu)$. We note that the seasonal variability of $C_{\mu}$ and the relatively high turnover rates are a prominent feature in the Amazon River basin across a range of prescribed $Q_{10}(T_\nu)$ values.

Other hypotheses that could explain the lag between CH$_4$ and EWH include the temporal variability of (a) macrophyte biomass, (b) water column oxidation, (c) redox potential, and (d) soil pH. The presence of aquatic macrophytes plays an important role in the production of methane in wetland soils, as macrophytes produce carbon available for methanogenesis, facilitate the transport of CH$_4$ to the atmosphere, transport $O_2$ to the subsurface (e.g. Laanbroek, 2010), and can inhibit light and re-aeration in aquatic environments (Pierobon et al., 2010). Hence, an increase in macrophyte biomass during the rising water phase (e.g. Silva et al., 2009) could result in seasonal changes in CH$_4$ emissions in tropical wetland environments due to enhanced plant-mediated transport and an increase in labile carbon in aquatic environments. Conversely, seasonal macrophyte growth may result in increased methanotrophy due to increased $O_2$ transport to the subsurface. Seasonal variability in redox potential in wetland environments is controlled by microbial activity, and hence is indirectly controlled by temperature, nutrient availability, water table level and root biomass, amongst other factors (Seybold et al., 2002; Thompson et al., 2009; Schmidt et al., 2011). However, few long-term measurements of redox potential have been performed over seasonal timescales, and further research is required to determine the large-scale redox variability in wetland environments. CH$_4$ oxidation within the water column (e.g. Schubert et al., 2010) has also been proposed as a mechanism explaining reduced CH$_4$ emissions during the peak of the wet season (Mitsch et al., 2010), although this would result in a second CH$_4$ peak at the end of the wet season. The absence of this peak in our analysis suggests this process plays only a minor role in the seasonality of tropical wetland CH$_4$ emissions. Soil pH has been found to temporally vary over seasonal timescales and, in particular, has been found to increase with decreasing redox potential (e.g. Singh, 2001; Seybold et al., 2002), although pH responses to redox potential and water table can vary widely (e.g. Singh et al., 2000; Thompson et al., 2009). Although seasonal variation in wetland pH as a significant control on CH$_4$ emissions is a viable hypothesis, to our knowledge there are currently no repeat measurements of pH in response to flooding in tropical wetlands. Other mechanisms that could temporally affect CH$_4$ emissions include the subsurface sulfur and iron cycles (Laanbroek, 2010).

We also expect uncertainties in the seasonal variability of tropical wetland CH$_4$ fluxes to arise from (i) the use of GRACE EWH as a proxy for wetland water volume and (ii) the first order approximation of a temporally constant $N_{\mu}$. GRACE monthly EWH change uncertainties of 1.0–2.1 cm were reported by Wahr et al. (2004); over the Amazon River basin, EWH variability is between 0.30–0.42 m for 2003–2009. However, GRACE EWH is only a proxy for wetland water volume. Papa et al. (2008) found a strong concurrence in the seasonal cycle of GRACE EWH, inundated fraction, and modelled water storage. We also find strong seasonal covariance over the main branch of the Amazon River (Fig. 1). Although independently GRACE and inundated fraction (Prigent et al., 2007) provide proxies for wetland water volume, a better understanding of basin-scale hydrology could ultimately be achieved via a synthesis of all available hydrological parameters (e.g. Azarderakhsh et al., 2011).

We chose a constant value of $\bar{N}_{\mu} = \frac{P_{CH_4}}{F_{CH_4}}$ as a first order approximation of methanogen-available carbon input in wetlands. However, the likely factors influencing the temporal variability of $N_{\mu}$ are the seasonal variability of root exudates and leaf litter. While root exudates are strongly dependent on

\[ \text{CH}_4 \text{ flux} = \frac{P_{CH_4}}{F_{CH_4}} \]
reduction in boreal wetland emissions (3.2 %), primarily due to the introduction of a gradual cut-off in methanogenesis rates under 0 °C (Sect. 2.3). During 2003–2008, the global change in CH₄ wetland emissions amounted to an increase of 7.7 Tg CH₄ yr⁻¹, mostly as a result of boreal wetlands (3.1 Tg CH₄ yr⁻¹) and tropical wetlands (3.4 Tg CH₄ yr⁻¹), while there was also a significant increase of 1.1 Tg CH₄ yr⁻¹ from mid-latitude wetlands. The increase in Southern Hemisphere extra-tropical wetland emissions (0.13 Tg CH₄ yr⁻¹) did not significantly contribute to the CH₄ wetland emissions growth during 2003–2008. Boreal wetland emissions increased by 1.6 Tg CH₄ yr⁻¹ in between 2006–2007 and decreased by 0.1 Tg CH₄ yr⁻¹ in 2007–2008. Tropical wetland emissions increased by 1.4 Tg CH₄ yr⁻¹ in 2006–2007 and 1.2 Tg CH₄ yr⁻¹ in 2007–2008. Other work shows a larger interannual variability, a similar year-to-year trend for boreal wetlands, and a decrease in the atmospheric CH₄ inversion estimates of tropical wetland emissions for 2007–2008 (Bousquet et al., 2011).

Finally, we used our wetland and rice CH₄ emission estimates to drive the GEOS-Chem global 3-D atmospheric chemistry and transport model (described and evaluated by Fraser et al., 2011), allowing us to test consistency between our emissions to surface measurements of CH₄ concentrations. We sampled the model at the time and geographical location of the surface CH₄ measurements from the GasLab, AGAGE and ESRL networks (Francey et al., 1996; Prinn et al., 2000; Cunnold et al., 2002; Dlugokencky et al., 2009). Figure 5 shows model and observed CH₄ concentration anomalies (i.e. minus the mean trend) for the Northern and Southern Hemispheres. We chose to remove the interannual trend from all CH₄ concentrations to compare the seasonality of model and surface measurements of CH₄. We show that the DMCM approach better describes the observed seasonality in both hemispheres \( r_{\text{NH}} = 0.9, r_{\text{SH}} = 0.9 \), and the amplitude of the Southern Hemisphere seasonality is largely improved in comparison to the GEOS-Chem runs using Fung et al. (1991) and Bloom et al. (2010) CH₄ emissions.

**Table 2. Estimates of total annual Amazon River basin wetland CH₄ emissions (Tg CH₄ yr⁻¹).**

<table>
<thead>
<tr>
<th>Study</th>
<th>Amazon Wetland CH₄ Emissions (Tg CH₄ yr⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Melack et al. (2004)</td>
<td>22</td>
</tr>
<tr>
<td>Fung et al. (1991)</td>
<td>5.3</td>
</tr>
<tr>
<td>Riley et al. (2011)</td>
<td>58.9*</td>
</tr>
<tr>
<td>Bloom et al. (2010)</td>
<td>20.0</td>
</tr>
<tr>
<td><strong>This study</strong></td>
<td><strong>26.2 ± 9.8</strong></td>
</tr>
</tbody>
</table>

* High tropical fluxes by Riley et al. (2011) are a result of anomalously high predicted net primary productivity in the Community Land Model (CLM version 4).

**Fig. 5.** Northern Hemisphere (NH, top) and Southern Hemisphere (SH, bottom) mean observed and model methane anomalies from surface concentration measurements, 2003–2008. Surface concentration measurements (black) are from the GasLab, AGAGE and ESRL networks (Francey et al., 1996; Prinn et al., 2000; Cunnold et al., 2002; Dlugokencky et al., 2009). The GEOS-Chem global 3-D chemistry transport model (Fraser et al., 2011) is driven by wetland CH₄ emission estimates from Fung et al. (1991) (blue), Bloom et al. (2010) (red), and our new top-down approach (green).

NPP, in-situ and modelled estimates of leaf litter seasonality in the Amazon River basin have been found to vary widely (Chave et al., 2010; Caldararu et al., 2012). Ultimately, a temporally variable and complete representation of NPP is required in order to further understand the temporal variability of CH₄ in wetlands.

By globally integrating the DMCM method, we estimated tropical wetlands emit 111.1 Tg CH₄ yr⁻¹, where Amazon wetlands account for 26.2 Tg CH₄ yr⁻¹ (24 %). Table 2 shows our estimates are within the range of other independent Amazon wetland emission CH₄ estimates. Figure 4 shows the zonal profile of our top-down approach with the associated uncertainty estimates. We capture three main features of global wetland and rice paddy emissions – i.e. peaks over the tropics, subtropics and lower mid-latitudes (mainly due to rice), and boreal latitudes – in agreement with previous studies (Bloom et al., 2010; Fung et al., 1991; Riley et al., 2011). In comparison to our previous work (Bloom et al., 2010), we find a slight
4 Concluding remarks

Understanding the temporal controls of temperature, water volume and carbon content of wetlands is crucial in determining the global and regional seasonal cycle of wetland CH4 emissions. We showed that incorporating a temporally variable methanogen-available carbon pool, Cμ, in our top-down approach results in a significant improvement in describing the temporal behaviour of tropical and global CH4 emissions.

By implementing our dynamic methanogen-available carbon model (DMCM) on a global scale, we determined the effects of a seasonally variable Cμ on the seasonality of wetland CH4 emissions in the Amazon River basin. We found a median decay constant of $\overline{\Phi_{\text{Amazon}}}=0.017$ day$^{-1}$ over the Amazon River basin. Seasonal changes in Cμ in the tropics largely explained the seasonal lag between SCIAMACHY observed CH4 concentrations and GRACE equivalent water height. The relatively high seasonal variability in Cμ (mean $c_v=28 \%$) over the Amazon River basin resulted in peak CH4 emissions occurring mostly 1–3 months prior to the peak water height period; in contrast, the median boreal Cμ variability was $c_v<0.1 \%$. We showed a substantial improvement in simulating the seasonality of surface concentrations when using the GEOS-Chem atmospheric chemistry transport model with our wetland and rice CH4 emission estimates ($r=0.9$). These improvements in the magnitude and temporal dynamics of tropical CH4 emissions will ultimately help constrain global inverse modelling efforts.

We anticipate that this work will lead to further and more detailed parameterisation of tropical wetland CH4 emissions, and we expect our tropical wetland CH4 emission parameterisation will reduce the uncertainty in forecasting future changes in wetland CH4 emissions.

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References


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