Estimating the contribution of monsoon-related biogenic production to methane emissions from South Asia using CARIBIC observations

Angela K. Baker,[1] Tanja J. Schuck,[1] Carl A. M. Brenninkmeijer,[1] Armin Rauthe-Schöch,[1] Franz Slemr,[1] Peter F. J. van Velthoven,[2] and Jos Lelieveld[1,3]

Received 20 March 2012; revised 26 April 2012; accepted 3 May 2012; published 31 May 2012.

[1] During the summer monsoon the upper troposphere over South Asia is characterized by the monsoon anticyclone centered above the Tibetan Plateau. Surface air that has been rapidly transported upwards through deep convection becomes trapped within the strong anticyclonic circulation. Observations of trace gases within this anticyclone by the CARIBIC flying observatory revealed large enhancements in the greenhouse gas methane (CH_4), which increased over the course of the monsoon. Meteorological analysis indicated that these air masses originated primarily in India, for which relatively little is known about CH_4 emissions. Using correlations between concentrations of CH_4 and carbon monoxide (CO) we estimated total emissions of 30.8 Tg CH_4 during the 2008 monsoon season (June–September), 19.7 Tg of which were identified as additional, monsoon-related biogenic methane using the relationship of CH_4 to ethane (C_2H_6). After accounting for the ~3.9 Tg attributed to rice agriculture in the current inventories, ~15.8 Tg of additional CH_4 remain. Underestimated rice emissions provide a partial explanation, with the remainder most likely attributable to microbial production in waterlogged areas such as landfills, polluted waterways and wetlands.
Citation: Baker, A. K., T. J. Schuck, C. A. M. Brenninkmeijer, A. Rauthe-Schöch, F. Slemr, P. F. J. van Velthoven, and J. Lelieveld (2012), Estimating the contribution of monsoon-related biogenic production to methane emissions from South Asia using CARIBIC observations, *Geophys. Res. Lett.*, *39*, L10813, doi:10.1029/2012GL051756.

1. Introduction

[2] Methane (CH_4), second only to carbon dioxide (CO_2) in its contribution to anthropogenic climate forcing, is a potent greenhouse gas whose atmospheric burden has strongly increased during the last 250 years [*Dlugokencky*, 2003; *Forster et al.*, 2007]. Its short lifetime compared to CO_2 means that a reduction in CH_4 emissions can produce a rapid response in abating climate change, making CH_4 sources ideal targets for mitigation [*Montzka et al.*, 2011]. However, constraining the atmospheric budget of CH_4 has proven difficult, particularly the relative contributions of its various sources, as reflected in the debate over the cause of fluctuations in its growth rate [*Aydin et al.*, 2011; *Heimann*, 2011; *Kai et al.*, 2011]. For South Asia, including India, the second most populous nation in the world, observational data for calculating CH_4 emissions are scarce.

[3] The dominant meteorological feature in South Asia is the summer monsoon, which begins in late May/early June and ends in mid/late September. During the monsoon there exists persistent large-scale deep convection over the Bay of Bengal and the Indian subcontinent, which rapidly transports low-level air to higher altitudes [*Park et al.*, 2009; *Randel et al.*, 2010]. This ultimately results in well-mixed air masses representative of South Asian emissions, primarily from India, being present in the upper tropospheric monsoon anticyclone, which is centered over the Tibetan Plateau [*Randel et al.*, 2010].

[4] Over the course of the 2008 monsoon season the CARIBIC (Civil Aircraft for the Regular Investigation of the atmosphere Based on an Instrument Container) observatory conducted monthly measurement flights from Frankfurt, Germany to Chennai, India and probed the UT anticyclone over South Asia [*Baker et al.*, 2011; *Patra et al.*, 2011; *Schuck et al.*, 2010]. A prominent feature we observed in monsoon outflow was a large enhancement in CH_4 abundances reaching from southern India to the Middle East, which waxed and waned with the monsoon [*Schuck et al.*, 2010]. While this distinct upper tropospheric CH_4 "plume" has also been observed using satellites [*Xiong et al.*, 2009], CARIBIC provides the first detailed in situ observations, flying through this synoptic scale feature of upper tropospheric air burdened with surface emissions. Using tracer-tracer correlations, namely those of CH_4 to carbon monoxide (CO) and ethane (C_2H_6), we estimate total CH_4 emissions from India during the monsoon period and identify and estimate the fraction of CH_4 emitted by biogenic sources, i.e., not arising from fossil fuels, biofuels, or biomass burning.

2. Methods

[5] CARIBIC (www.caribic-atmospheric.com) is a long-term atmospheric measurement program wherein a large (1.6 ton) instrument container is deployed monthly aboard a Lufthansa Airbus A340-600 for a sequence of four long-distance passenger flights between Frankfurt, Germany and various destinations across the globe [*Brenninkmeijer et al.*, 2007]. Between April and December 2008 the container was deployed to Chennai in southern India. At aircraft cruise

[1]Atmospheric Chemistry Division, Max Planck Institute for Chemistry, Mainz, Germany.
[2]Royal Netherlands Meteorological Institute, De Bilt, Netherlands.
[3]Cyprus Institute, Energy, Environment and Water Research Center, Nicosia, Cyprus.

Corresponding author: A. K. Baker, Atmospheric Chemistry Division, Max Planck Institute for Chemistry, Hahn-Meitner-Weg 1, D-55128 Mainz, Germany. (angela.baker@mpic.de)

0094-8276/12/2012GL051756

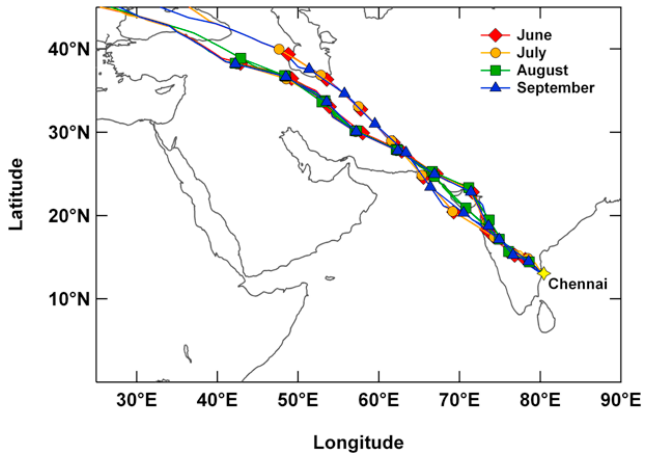

Figure 1. CARIBIC flight tracks between 10°N and 40°N from June to September 2008 for flights where whole air samples were collected. Markers indicate air sample collection points.

altitudes between 10 and 12 km, the CARIBIC observatory intersected the monsoon anticyclone along the flight path between ~10°N and ~40°N, with the extent depending on the stage of the monsoon (Figure 1).

[6] The CARIBIC scientific payload consists of 15 measurement systems, is fully automated, and carries out in-situ trace gas and aerosol measurements as well as remote sensing by DOAS and collection of aerosol and whole air samples [*Brenninkmeijer et al.*, 2007]. During flight whole air samples are collected in two sampling units, each of which contains 14 glass sampling flasks of 2.7 L volume pressurized to ~4.5 bar during collection. Samples were collected at pre-determined, evenly spaced intervals of ~35 min (~480 km) with filling times between 0.5 to 1.5 min (~7 to ~22 km). When all four monthly flights were between Frankfurt and Chennai, sample collection only took place during the first two flights in order to achieve higher spatial and temporal resolution. After the flights whole air samples are analyzed for greenhouse gases and non-methane hydrocarbons. For this analysis we selected only those samples that were collected within the monsoon anticyclone as indicated by meteorological analyses and backward trajectory calculations based on ECMWF re-analyzed data determined along the flight track for each individual sample [*Scheele et al.*, 1996; P. van Velthoven, Meteorological analysis of CARIBIC by KNMI, 2011, available at http://www.knmi.nl/samenw/campaignsupport/CARIBIC/#LH]. Of the 69 samples collected between 10°N and 40°N during the monsoon season, 58 were collected within the monsoon anticyclone. For each sample greenhouse gases and non-methane hydrocarbons were measured post-flight using separate gas chromatography (GC) systems coupling GC separation with flame ionization detection or electron capture detection (GC-FID, GC-ECD) [*Baker et al.*, 2010; *Schuck et al.*, 2009]. Mixing ratios of CO were measured in flight using a vacuum ultraviolet fluorescence based instrument with a time resolution of 1 s, and fast data is integrated over the air sample collection times to obtain the corresponding average CO mixing ratios [*Scharffe et al.*, 2012].

3. Results and Discussion

[7] During flights to Chennai observed latitudinal trace gas profiles (mixing ratios measured along the flight track) of CH_4, CO, and C_2H_6 from June to August revealed a distinct and persistent monsoon pattern, with clear Gaussian-shaped enhancements between 10°N and 40°N, peaking near 25°N (Figure 2). These profiles persisted through September, becoming weaker and narrower with the southward recession and weakening of the monsoon. Over the course of the monsoon season, mean and maximum mixing ratios of CO and ethane remained nearly constant, with slightly higher levels in June than in later months. Conversely, monthly mean and maximum mixing ratios of CH_4 significantly increased from June until August, and remained about the same in September. Increasing CH_4 in the absence of concomitant increases in CO and ethane points to independent sources of these gases. The large increases in CH_4 are

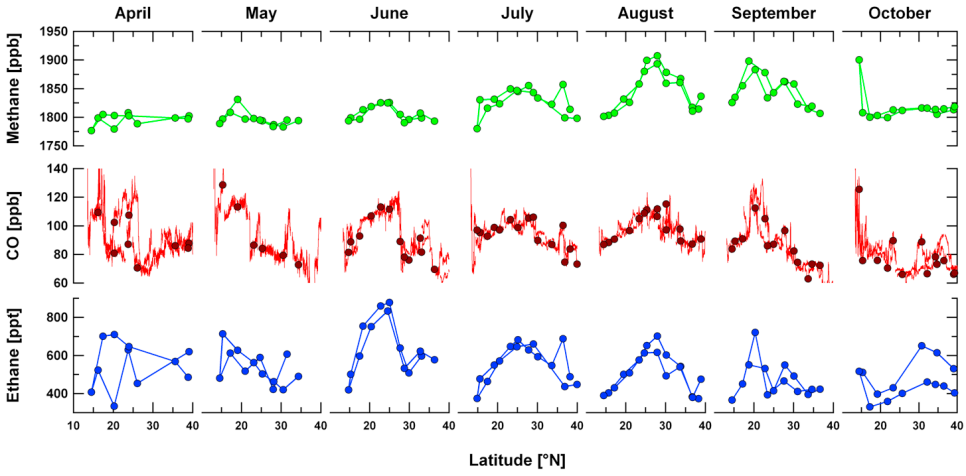

Figure 2. Latitudinal profiles of methane, CO and ethane mixing ratios in the UT between 10°N and 40°N from April to October 2008, measured during CARIBIC flights between Frankfurt (Germany) and Chennai (India). Methane and ethane data points represent measurements from individual whole air samples; in the plot of CO (second from top), lines represent 10s data from in situ measurements and circles represent integrals corresponding to air sample collection times.

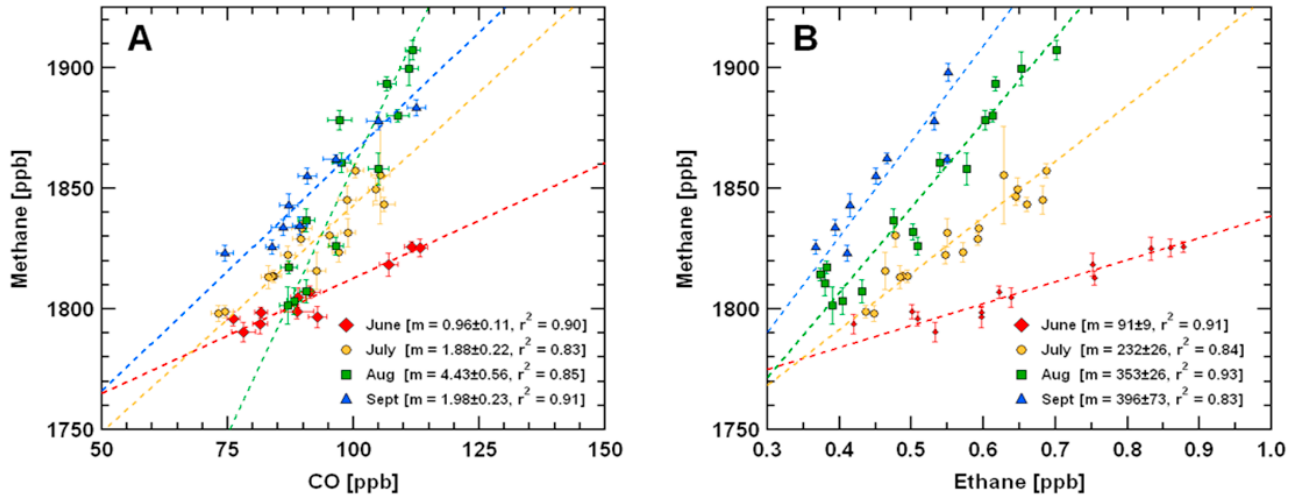

Figure 3. Correlation plots of methane with (a) CO and (b) ethane in monsoon outflow south of 40°N from June to September 2008. Error bars represent one standard deviation. Slopes (m) are for the orthogonal distance regressions and their errors represent one standard deviation.

readily apparent in the CH_4/CO and CH_4/C_2H_6 enhancement ratios ($\Delta CH_4/\Delta CO$ and $\Delta CH_4/\Delta C_2H_6$, respectively), which are determined from the slopes of the respective linear correlation plots and which represent the species' relative enhancements above background levels (Figure 3).

[8] Enhancement ratios between two compounds are commonly interpreted as emission ratios and can be used to estimate emissions of one compound when those of the other are known [*Andreae et al.*, 2001; *Mauzerall et al.*, 1998]. This can be complicated by photochemical losses during transport and by mixing with air masses having different tracer ratios (i.e., background air). Very short (<2 day) transport times in the boundary layer/lower troposphere coupled with much longer lifetimes (>2 months) upon reaching the UT means that photochemical losses have only a small effect on these ratios. Based on the conditions for the CARIBIC samples we estimate that these losses result in reductions of ~5–10% for both ratios, attributable to larger losses of CO and C_2H_6 relative to methane. As these losses are not exact we do not include them in subsequent calculations, but note that they are generally smaller than uncertainties arising from errors in the measurements themselves. The effect on concentrations from dilution by mixing with background air is also difficult to calculate, however, previous examination of monsoon non-methane hydrocarbon signatures from CARIBIC samples provided strong evidence that once the air parcels reached the upper tropospheric anticyclone the mixing in of outside (background) air was minimal [*Baker et al.*, 2011]. This is in agreement with other findings of chemical isolation within the monsoon anticyclone [*Lawrence and Lelieveld*, 2010; *Park et al.*, 2008, 2009; *Randel et al.*, 2010]. Furthermore, during each of the four months enhancement ratios relative to CO and ethane were the result of robust correlations, showing that although observed over distances of more than 3000 km these air parcels must have been well-mixed, implying representativeness for a large region.

[9] To estimate monthly CH_4 emissions, we chose CO as a reference compound, as its sources are widespread and relatively well-studied, making it an ideal tracer for surface emissions. Additionally, although not all sources of CO are significant sources of CH_4, they tend to be co-located with CH_4 sources, resulting in strong correlations between the two in well-mixed air parcels, as observed within monsoon outflow (Figure 3a). Methane emission estimates (E_{CH4}) can then be calculated from CO emissions (E_{CO}) using $E_{CH4} = E_{CO} \bullet (\Delta CH_4/\Delta CO)$.

[10] Indian emissions of CO were derived by combining emissions data for India given in EDGAR v.4.1 (Emission Database for Global Atmospheric Research [*European Commission Joint Research Centre* (*JRC*) *and Netherlands Environmental Assessment Agency* (*PBL*), 2009]) and GFED v.3.2 (Global Fire Emissions Database [*van der Werf et al.*, 2010]). To calculate monthly CO emissions for June through September we have taken the monthly emissions from open biomass burning reported in gridded GFED data and added these to the emissions from other sources reported in monthly gridded EDGAR data. Both datasets were integrated over the land area of India. The most recent emissions data available in EDGAR are from 2005 and 2010, and a linear interpolation was used to determine 2008 values. Monthly CO emissions were 5.82 Tg, 5.81 Tg, 5.82 Tg and 5.85 Tg in June, July, August and September, respectively. Although uncertainties in CO emission estimates are not provided in the EDGAR database, a general uncertainty of 50% has been suggested [*Olivier et al.*, 2001]. Since there is no explicit value available for India we do not include this in our uncertainty calculations, opting to provide the more readily quantifiable errors attributable to our own analyses, but note that these large uncertainties in the CO estimates should be kept in mind throughout the subsequent discussion, as higher (lower) CO emissions would result in higher (lower) CH_4 emission estimates from our data.

[11] From these CO emissions we infer monthly CH_4 emissions between 3.3 ± 0.4 and 14.7 ± 1.9 Tg, with a four month total of 30.8 ± 2.2 Tg CH_4 (Table 1 and Figure 4); uncertainties are derived from the 1-σ uncertainty of the slope of the orthogonal distance regression (Figure 3). Surprisingly, available studies estimate total annual CH_4 emissions from India to be not much higher, ~29–41 Tg CH_4 yr^{-1} [*JRC and PBL*, 2009; *Patra et al.*, 2009; *Streets et al.*, 2003]. Therefore the 30.8 ± 2.2 Tg CH_4 estimated

Table 1. Monthly Emission Ratios of Methane to Ethane and CO, Percent Excess (Monsoon-Related) Methane, and Monthly Total and Monsoon-Related Methane Emissions[a]

Month	$\Delta CH_4/\Delta CO$[b] (g/g)	Monthly CH_4[c] (Tg)	Excess CH_4 Using CO[d] (Tg)	$\Delta CH_4/\Delta C_2H_6$[b] (g/g)	Percent Excess CH_4[c,d]	Excess CH_4 Using Ethane and Monthly Fluxes[d] (Tg)
June	0.57 ± 0.06	3.3 ± 0.4	0	49 ± 5	0	0
July	1.07 ± 0.12	6.2 ± 0.7	2.9 ± 0.8	124 ± 14	60 ± 20	3.8 ± 0.9
August	2.52 ± 0.32	14.7 ± 1.9	11.4 ± 1.9	188 ± 14	74 ± 11	10.5 ± 1.8
September	1.13 ± 0.13	6.6 ± 0.8	3.3 ± 0.8	211 ± 39	77 ± 24	5.1 ± 1.4
Total		30.8 ± 2.2	17.6 ± 2.2			19.7 ± 2.4

[a]Uncertainties represent one standard deviation.
[b]derived from slopes of the orthogonal distance regressions (Figure 3).
[c]Estimated using CO emissions derived from EDGAR v4.0 and GFED.
[d]Using June emissions as a baseline (see text).

from the CARIBIC CH_4-CO correlations for the four month monsoon period indicates a large amount of additional CH_4 clearly not accounted for in the current inventories. If emissions in June are assumed to represent a baseline for emissions at the beginning of the monsoon, we can interpret the difference between emissions in June and in subsequent months to be roughly equivalent to the additional emissions of CH_4 related to the monsoon. We then find that 2.4 ± 0.7 to 9.6 ± 1.7 Tg per month of additional CH_4 are emitted in July, August and September, or 15.2 ± 2.0 Tg total (Table 1). We note here that June emissions should already contain a monsoon-related component, meaning our calculations most likely do not capture all monsoon-related methane emissions, and our results provide a conservative estimate.

[12] Given the widespread flooding during the monsoon, the near absence of open biomass burning, and the coincidence with the (rice) growing season, increased production of CH_4 by biogenic sources is the obvious explanation for the extra CH_4, although this information cannot be distilled from its relationship with CO. To further elucidate the nature of the sources we turn to C_2H_6, which has shown itself to be a useful tracer for identifying CH_4 sources [*Aydin et al.*, 2011; *Simpson et al.*, 2006]. The most significant sources of C_2H_6, namely fossil fuel use and the burning of biomass/biofuels, also emit CH_4; in fact, these two sources emit the bulk of non-biogenic CH_4. Conversely, biogenic CH_4 sources do not emit C_2H_6 [*Redeker et al.*, 2003], and it stands to reason that variations in CH_4 relative to C_2H_6 are the result of changes in biogenic emissions. Thus, although ethane emissions are not as well known as those of CO, making it less useful for quantifying absolute CH_4 emissions, C_2H_6 can be used to differentiate between biogenic and non-biogenic CH_4 sources.

[13] Enhancement ratios clearly show that the amount of CH_4 increases relative to C_2H_6 until August (Figure 3b). In fact, from June to September the ratio increases fourfold, from 91 ± 9 to 396 ± 73 ppb CH_4/ppb C_2H_6. To quantify the relative increase in emissions the respective emission ratios for July, August and September are related to those for June, which, as with the emission estimates from CO, is used as a baseline. The changing emission ratios correspond to enhancements of 75 ± 15, 139 ± 15 and 162 ± 39 g of additional CH_4 per g C_2H_6 emitted in July, August and September, respectively. This translates to between 60 and 77% of monthly CH_4 emissions arising from additional, biogenic sources during the monsoon. If these percentages are then multiplied by the monthly total CH_4 emissions derived using CO, additional monthly emissions can be calculated. Estimates of between 3.8 ± 0.9 and 10.5 ± 1.8 Tg additional biogenic CH_4 each month are comparable to estimates made from CO, and total additional CH_4 is estimated to be 19.7 ± 2.4 Tg (Table 1 and Figure 4).

[14] As the monsoon season is coincident with the primary growing season for rice, a fraction of this excess CH_4 must arise from rice cultivation. Emissions of CH_4 from rice in India are currently estimated to be ~3.9 Tg CH_4 yr^{-1}, with the bulk emitted between June and September [*JRC and PBL*, 2009; *Garg et al.*, 2011; *Panigrahy et al.*, 2010]. Taking into account these rice emissions leaves 15.8 ± 2.2 Tg CH_4 from our estimate unaccounted for. Underestimated rice paddy emissions undoubtedly contribute to this large discrepancy, although it seems unlikely that they account for all of the additional CH_4, making it highly likely that other sources are at play. For example, satellite observations of India show that waterlogged areas increase nearly threefold from the beginning to the end of the monsoon resulting in increased wetland CH_4 emissions [*Agarwal and Garg*, 2009]. Furthermore, there is evidence that unmanaged waste sites and landfills as well as wetlands and waterways polluted by urban waste and sewage are considerable sources of CH_4, and that emissions peak during the monsoon [*Purkait and Chakrabarty*, 2011; *Purvaja and Ramesh*,

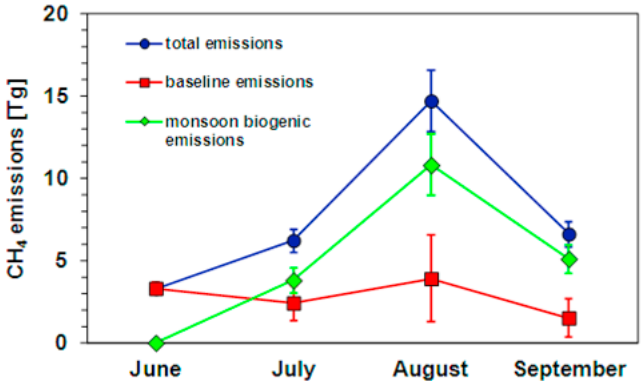

Figure 4. Monthly emissions of methane from India during the monsoon season. Monsoon biogenic emissions (green diamonds) represent those calculated from fractions of excess methane relative to ethane. Baseline emissions (red squares) represent the difference between total and monsoon biogenic emissions. Error bars represent one standard deviation.

2001]. Since few other non-rice agricultural CH_4 sources seem likely to be influenced by the monsoon, waste-related emissions are prime candidates for some of the extra CH_4 measured. Current inventories estimate landfills, wastewater treatment and wetlands to contribute only ~2.5 Tg CH_4 annually, much less than indicated by our estimates [*JRC and PBL*, 2009; *Garg et al.*, 2011].

4. Conclusions

[15] Measurements by the CARIBIC observatory of trace species in the outflow of the South Asian summer monsoon revealed large enhancements in the greenhouse gas methane, originating primarily in India. Using correlations between CH_4 concentrations and those of CO and C_2H_6 measured in monsoon outflow we estimate total CH_4 emissions of 30.8 ± 2.2 Tg between June and September 2008, 19.7 ± 2.4 Tg of which are attributable to biogenic sources. Recent greenhouse gas inventories for India account for only a fraction of these emissions. According to these emission inventories ~3.9 Tg of the excess CH_4 can be explained by rice agriculture, leaving 15.8 ± 2.2 Tg of additional CH_4. While underestimates in rice emissions provide a partial explanation, a large part is likely attributable to biogenic emissions arising from microbial production in waterlogged landfills, sewers, and wetlands, which must be several times larger than current estimates. Clearly, bottom-up estimates of biogenic CH_4 emissions from a wide range of ill-definable sources based on spot measurements and extrapolations are difficult, if not impossible, to make. However, the large source of biogenic CH_4 we infer is of concern, especially as organic waste production, which is difficult to assess and manage, is expected to increase with rapid economic and population growth. Even larger questions arise when considering the potential influence of climate change-related changes in monsoon meteorology on these methane emissions, and any subsequent climate feedbacks. On a positive note, successful mitigation strategies could aim to tap this large biogenic source for local energy generation (e.g., capping of landfills, biogas production), thereby doubling the benefit through reducing CH_4 related climate forcing in the short term, pending fundamental changes in the fossil fuel-based mode of the early Anthropocene.

[16] **Acknowledgments.** The authors would like to thank Lufthansa Airlines and Fraport for their support of the CARIBIC project, as well as all members of the CARIBIC team, both in Mainz and at partner institutions. We cordially thank Paul Crutzen for his continued support of CARIBIC. We also thank Andrea Pozzer for assistance with the EDGAR CO data. We acknowledge the support of the European Commission through the GEOmon (Global Earth Observation and Monitoring) Integrated Project under the 6th Framework Program (contract FP6-2005-Global-4-036677).

[17] The Editor thanks Prabir K. Patra anonymous reviewers for their assistance in evaluating this paper.

References

Agarwal, R., and J. K. Garg (2009), Methane emission modelling from wetlands and waterlogged areas using MODIS data, *Curr. Sci.*, *96*(1), 36–40.

Andreae, M. O., et al. (2001), Transport of biomass burning smoke to the upper troposphere by deep convection in the equatorial region, *Geophys. Res. Lett.*, *28*(6), 951–954, doi:10.1029/2000GL012391.

Aydin, M., K. R. Verhulst, E. S. Saltzman, M. O. Battle, S. A. Montzka, D. R. Blake, Q. Tang, and M. J. Prather (2011), Recent decreases in fossil-fuel emissions of ethane and methane derived from firn air, *Nature*, *476*(7359), 198–201, doi:10.1038/nature10352.

Baker, A. K., F. Slemr, and C. A. M. Brenninkmeijer (2010), Analysis of non-methane hydrocarbons in air samples collected aboard the CARIBIC passenger aircraft, *Atmos. Meas. Tech.*, *3*(1), 311–321, doi:10.5194/amt-3-311-2010.

Baker, A. K., T. J. Schuck, F. Slemr, P. van Velthoven, A. Zahn, and C. A. M. Brenninkmeijer (2011), Characterization of non-methane hydrocarbons in Asian summer monsoon outflow observed by the CARIBIC aircraft, *Atmos. Chem. Phys.*, *11*(2), 503–518, doi:10.5194/acp-11-503-2011.

Brenninkmeijer, C. A. M., et al. (2007), Civil Aircraft for the regular investigation of the atmosphere based on an instrumented container: The new CARIBIC system, *Atmos. Chem. Phys.*, *7*(18), 4953–4976, doi:10.5194/acp-7-4953-2007.

Dlugokencky, E. J. (2003), Atmospheric methane levels off: Temporary pause or a new steady-state?, *Geophys. Res. Lett.*, *30*(19), 1992, doi:10.1029/2003GL018126.

European Commission Joint Research Centre (JRC) and Netherlands Environmental Assessment Agency (PBL) (2009), Emission Database for Global Atmospheric Research (EDGAR), release version 4.0, http://edgar.jrc.ec.europa.eu, Ispra, Italy.

Forster, P., et al. (2007), Changes in atmospheric constituents and in radiative forcing, in *Climate Change 2007: Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, edited by S. Solomon et al., pp. 131–234, Cambridge Univ. Press, Cambridge, U. K.

Garg, A., B. Kankal, and P. R. Shukla (2011), Methane emissions in India: Sub-regional and sectoral trends, *Atmos. Environ.*, *45*(28), 4922–4929, doi:10.1016/j.atmosenv.2011.06.004.

Heimann, M. (2011), Atmospheric science: Enigma of the recent methane budget, *Nature*, *476*(7359), 157–158, doi:10.1038/476157a.

Kai, F. M., S. C. Tyler, J. T. Randerson, and D. R. Blake (2011), Reduced methane growth rate explained by decreased Northern Hemisphere microbial sources, *Nature*, *476*(7359), 194–197, doi:10.1038/nature10259.

Lawrence, M. G., and J. Lelieveld (2010), Atmospheric pollutant outflow from southern Asia: A review, *Atmos. Chem. Phys.*, *10*(22), 11,017–11,096, doi:10.5194/acp-10-11017-2010.

Mauzerall, D. L., J. A. Logan, D. J. Jacob, B. E. Anderson, D. R. Blake, J. D. Bradshaw, B. Heikes, G. W. Sachse, H. Singh, and B. Talbot (1998), Photochemistry in biomass burning plumes and implications for tropospheric ozone over the tropical South Atlantic, *J. Geophys. Res.*, *103*(D15), 19,281–19,282, doi:10.1029/98JD01786.

Montzka, S. A., E. J. Dlugokencky, and J. H. Butler (2011), Non-CO_2 greenhouse gases and climate change, *Nature*, *476*(7358), 43–50, doi:10.1038/nature10322.

Olivier, J. G. J., J. J. M. Berdowski, J. A. H. W. Peters, J. Bakker, A. J. H. Visschedijk, and J.-P. J. Bloos (2001), Applications of EDGAR emission database for global atmospheric research, *Rep. RIVM 773301 001/NOP 410200 051*, Natl. Inst. for Public Health and the Environ., Bilthoven, Netherlands.

Panigrahy, S., K. R. Manjunath, R. P. Singh, A. Chhabra, and J. S. Parihar (2010), Spatio-temporal pattern of green house gases over India and upscaling of methane emission from agriculture using space technology, *Bull. Natl. Nat. Resour. Manage. Syst.*, *35*, 50–56.

Park, M., W. J. Randel, L. K. Emmons, P. F. Bernath, K. A. Walker, and C. D. Boone (2008), Chemical isolation in the Asian monsoon anticyclone observed in Atmospheric Chemistry Experiment (ACE-FTS) data, *Atmos. Chem. Phys.*, *8*(3), 757–764, doi:10.5194/acp-8-757-2008.

Park, M., W. J. Randel, L. K. Emmons, and N. J. Livesey (2009), Transport pathways of carbon monoxide in the Asian summer monsoon diagnosed from Model of Ozone and Related Tracers (MOZART), *J. Geophys. Res.*, *114*, D08303, doi:10.1029/2008JD010621.

Patra, P. K., et al. (2009), Growth rate, seasonal, synoptic, diurnal variations and budget of methane in the lower atmosphere, *J. Meteorol. Soc. Jpn.*, *87*(4), 635–663, doi:10.2151/jmsj.87.635.

Patra, P. K., Y. Niwa, T. J. Schuck, C. A. M. Brenninkmeijer, T. Machida, H. Matsueda, and Y. Sawa (2011), Carbon balance of South Asia constrained by passenger aircraft CO_2 measurements, *Atmos. Chem. Phys.*, *11*(9), 4163–4175, doi:10.5194/acp-11-4163-2011.

Purkait, N. N., and D. K. Chakrabarty (2011), Pattern of methane emission from a garbage dump, *Curr. Sci.*, *101*(4), 528–531.

Purvaja, R., and R. Ramesh (2001), Natural and anthropogenic methane emission from coastal wetlands of south India, *Environ. Manage. N. Y.*, *27*(4), 547–557, doi:10.1007/s002670010169.

Randel, W. J., M. Park, L. Emmons, D. Kinnison, P. Bernath, K. A. Walker, C. Boone, and H. Pumphrey (2010), Asian monsoon transport of pollution to the stratosphere, *Science*, *328*(5978), 611–613, doi:10.1126/science.1182274.

Redeker, K. R., S. Meinardi, D. Blake, and R. Sass (2003), Gaseous emissions from flooded rice paddy agriculture, *J. Geophys. Res.*, *108*(D13), 4386, doi:10.1029/2002JD002814.

Scharffe, D., F. Slemr, C. A. M. Brenninkmeijer, and A. Zahn (2012), Carbon monoxide measurements onboard the CARIBIC passenger

aircraft using UV resonance fluorescence, *Atmos. Meas. Tech. Discuss.*, *5*(2), 2681–2702, doi:10.5194/amtd-5-2681-2012.

Scheele, M. P., P. C. Siegmund, and P. F. J. Van Velthoven (1996), Sensitivity of trajectories to data resolution and its dependence on the starting point: In or outside a tropopause fold, *Meteorol. Appl.*, *3*(3), 267–273, doi:10.1002/met.5060030308.

Schuck, T. J., C. A. M. Brenninkmeijer, F. Slemr, I. Xueref-Remy, and A. Zahn (2009), Greenhouse gas analysis of air samples collected onboard the CARIBIC passenger aircraft, *Atmos. Meas. Tech.*, *2*(2), 449–464, doi:10.5194/amt-2-449-2009.

Schuck, T. J., C. A. M. Brenninkmeijer, A. K. Baker, F. Slemr, P. F. J. von Velthoven, and A. Zahn (2010), Greenhouse gas relationships in the Indian summer monsoon plume measured by the CARIBIC passenger aircraft, *Atmos. Chem. Phys.*, *10*(8), 3965–3984, doi:10.5194/acp-10-3965-2010.

Simpson, I. J., F. S. Rowland, S. Meinardi, and D. R. Blake (2006), Influence of biomass burning during recent fluctuations in the slow growth of global tropospheric methane, *Geophys. Res. Lett.*, *33*, L22808, doi:10.1029/2006GL027330.

Streets, D. G., et al. (2003), An inventory of gaseous and primary aerosol emissions in Asia in the year 2000, *J. Geophys. Res.*, *108*(D21), 8809, doi:10.1029/2002JD003093.

van der Werf, G. R., J. T. Randerson, L. Giglio, G. J. Collatz, M. Mu, P. S. Kasibhatla, D. C. Morton, R. S. DeFries, Y. Jin, and T. T. van Leeuwen (2010), Global fire emissions and the contribution of deforestation, savanna, forest, agricultural, and peat fires (1997–2009), *Atmos. Chem. Phys.*, *10*(23), 11,707–11,735, doi:10.5194/acp-10-11707-2010.

Xiong, X., S. Houweling, J. Wei, E. Maddy, F. Sun, and C. Barnet (2009), Methane plume over south Asia during the monsoon season: Satellite observation and model simulation, *Atmos. Chem. Phys.*, *9*(3), 783–794, doi:10.5194/acp-9-783-2009.