Biogeosciences Discuss.,
doi:10.5194/bg-2016-4-AC2, 2016

Interactive comment on "Nitrous oxide and methane in two tropical estuaries in a peat-dominated region of North-western Borneo" *by* D. Müller et al.

D. Müller et al.

dmueller@iup.physik.uni-bremen.de

Received and published: 23 March 2016

We thank Anonymous Referee 1 for his helpful comments and suggestions. Our detailed answers can be found below.

This paper describes CH4 and N2O distributions in two tropical estuaries. Current literature for estuarine CH4 and N2O concentrations is still limited, and this type of study is significant for our scientific understanding and relevant to BG. The paper is well written and reads easily. However, there are several issues that need to be addressed prior to publication.

1) The dataset of CH4 and N2O concentrations does not cover the region of low

salinity (0-5), where high CH4 and N2O might be expected. Furthermore, no sample was taken at the river-end member. Hence it is hard to take a full picture of trace gas variation in the whole estuary system.

The correlation of N_2O with salinity during the dry season indicates that the freshwater end-member might indeed exhibit the highest N_2O during the dry season. We calculated the expected river end-member using the correlation of N_2O with salinity during the dry season and found concentrations of 9.1 nM, (Lupar, r = 0.5), 9.3 nM (Saribas, r=0.8) and 15 nM (Saribas tributary, r=0.9). For CH_4, this is a bit harder to do, as we did not observe a correlation between methane and salinity. While we agree that the lack of data in the low salinity region deserves a more thorough discussion, which we will include in the revised manuscript in sections 4.2 and 4.3, our main conclusions can be maintained despite the lack of data for the upper estuaries:

1) Eutrophication did not lead to enhanced N_2O. - Data from the river end-member would not provide additional information, as eutrophication was not observed in the river end-member.

2) DIN was a poor predictor of N_2O. - This was mainly inferred from Figure 2b), a river end-member data point is not likely to change this overall observation.

3) Postulation of additional N_2O and CH_4 sources during the wet season. - This conclusion is based mainly on the observation of high N_2O and CH_4 values at salinities 10-20. Of course, it would be extremely interesting to see if a similar observation could be made in the freshwater region.

2) Page 5, line 13: Does the DO sensor calibrated with Winkler titration method?

The FDO 925 sensor was calibrated by the manufacturer (WTW, Germany). According to the manufacturer, user calibration is not required for the specified lifetime of the sensor. Nevertheless, a routine function check was performed in water vapor saturated air, using the check and calibration vessel (FDO (R) check) that was provided with the sensor. This information will be added in the revised manuscript.

3) Page 6, line 25: I think the authors should give a little more detail about the k600 they used. For example, were they measured in situ or calculated by widely used wind-speed related relationships in the literature?
The k600 that we used were derived from CO_2 fluxes measured with a floating chamber in 2014. The measurements are described in detail in Müller et al., 2015a. From simultaneous measurements of the CO_2 flux and the water and air pCO_2, we derived kCO_2 and, ultimately, k600 for these estuaries. In the revised manuscript, we will change the following sentence:

For k, we used k600 values that were reported for the Lupar and Saribas estuaries in Müller et al. (2015a).

to

For k, we used k600 values that were derived for the Lupar and Saribas estuaries using the floating chamber method (Müller et al., 2015a). Floating chamber measurements were conducted at several locations along the estuaries during the wet season campaign and averaged over the spatial extent of the individual estuaries. We argued in Müller et al. (2015a) that the k600 values determined in this way are more appropriate than commonly used wind speed parameterizations, which neglect the influence of tidal currents and the water flow velocity.

4) Table 3 showed that dissolved CH4 in the Lupar estuary was low and undersaturated (2nM and 88%). What's possible reasons for this?
This seems to be an artifact. The reasons why we think so are the following: 1.) There are only two datapoints directly following each other which indicate undersaturation. 2.) With the exception of CO, all gas concentrations that we could retrieve from the respective spectra indicate undersaturation or atmospheric equilibrium values (N_2O, CH_4 and CO_2). This indicates that atmospheric air was in the measurement cell. 3.) No measurements were taken during the 20 minutes preceding the undersaturation values. A background measurement (empty cell) had been performed before that, and

the cell was filled with atmospheric air afterwards before the measurements of equilibrator air continued. If we reconsider these facts, it seems likely that atmospheric air was still in the cell. We would therefore exclude the two data points from the analysis. The numbers will be corrected in the revised manuscript.

5) Table 4 didn't show k600 for dry season, and the authors should explain what k600 was used for the calculation for dry season. k600 for Saribas tributary and Lupar estuary are almost twice of that for Saribas. Do the authors have any idea of the reason?
We used the same k600 for the dry and wet season. In the revised manuscript, we will add a more detailed description of the k600 values used in the methods section (see comment 3).
We discussed the variability of the k600 values in Müller et al., 2015a and after adding more detailed information in the methods section, the reader will be referred to that publication. There, we reasoned that the strong currents in the Lupar estuary are responsible for the relatively higher k600. The higher gas exchange velocity in the Saribas tributary if compared to the Saribas is consistent with the notion that the gas exchange velocity decreases with increasing stream order (Raymond et al., 2012).

6) Figure 1: Scales should be added and the South China Sea should be located on the map.
The Figure will be revised accordingly.

7) Figure 2: N2O vs salinity, it was shown that there are great N2O peaks during wet seasons between the areas with salinity of 12-15, suggesting a significant N2O source. The authors should discuss this in the text.
On page 10, lines 23-27 of the discussion paper, we tried to combine several lines of evidence as to where this N_2O comes from. Our argument is as follows. 1) There are sources in the estuary (i.e., salinities 10-20) in the wet season which we did not observe in the dry season. 2) Another feature of N_2O that we observed in the wet but not in the dry season was the co-variation of N_2O with CH_4. 3) In the wet season,

both N_2O and CH_4 varied with the tides, with higher concentrations during low tide. We inferred from 1) that there are additional sources of N_2O in the wet season, from 2) that they are sources of both N_2O and CH_4 and from 3) that either tidal creeks or the estuarine sediments constitute this additional source. We will modify the sections 4.2 and 4.3 in the revised manuscript in order to provide a more specific and detailed discussion.

References

Müller et al. (2015a): Fate of peat-derived carbon and associated CO2 and CO emissions from two Southeast Asian estuaries. Biogeosciences Discussions 12:8299-8340, doi: 10.5194/bgd-12-8299-2015

Raymond et al. (2012): Scaling the gas transfer velocity and hydraulic geometry in streams and small rivers. Limnology and Oceanography: Fluids and Environments 2:41-53. doi: 10.1215/21573689-1597669

Interactive comment on Biogeosciences Discuss., doi:10.5194/bg-2016-4, 2016.

Biogeosciences Discuss.,
doi:10.5194/bg-2016-4-AC1, 2016

Interactive comment on “Nitrous oxide and methane in two tropical estuaries in a peat-dominated region of North-western Borneo” *by* D. Müller et al.

D. Müller et al.

dmueller@iup.physik.uni-bremen.de

Received and published: 23 March 2016

We thank Anonymous Referee 2 for the positive evaluation of our manuscript. Our replies to the detailed comments can be found below.

This paper presents new information on seasonal differences in N2O and CH4 fluxes in mangrove environments. It furthers our understanding of the role of natural factors such as salinity, DO and DOC on greenhouse gas production in a generally sparsely-sampled region. There are a few minor issues to address:

Spelling/Grammar: This paper is very well written, there were only a few issues that I could find.

Printer-friendly version

Discussion paper

1. Page 8, line 10: "N2O was correlated with DOC (Fig. 2a), whereas this correlation was strong..." I think this should be "N2O was correlated with DOC (Fig. 2a), *and* this correlation was strong..." or just "N2O was correlated with DOC (Fig. 2a); this correlation was strong..."
This will be corrected in the revised manuscript (suggestion 2).

2. Page 9, like 17: "...the Lupar and Saribas rivers are no blackwater rivers..." Is this meant to say "...are *not* blackwater rivers..." It's technically correct either way, but the first phrasing sounds more colloquial.
We will change this to the more formal phrasing in the revised manuscript.

3. Page 10, line 15: "Either, a source of N2O exists on the continental shelf..." This comma is unnecessary.
The comma will removed from the sentence in the revised manuscript.

Other Comments:
1. This paper refers to k-value calculations derived from floating chamber experiments (covered in an earlier paper, Müller et al., 2015). It might be a good idea to make it clear in the methods section that these k values are from floating chambers, to distinguish this k calculation from the more common technique of estimating k-values from equations well-known in the literature.
We agree, we will replace the following sentence:
For k, we used k600 values that were reported for the Lupar and Saribas estuaries in Müller et al. (2015a).

with

For k, we used k600 values that were derived for the Lupar and Saribas estuaries using the floating chamber method (Müller et al., 2015a). Floating chamber measurements were conducted at several locations along the estuaries during the wet season campaign and averaged over the spatial extent of the individual estuaries. We argued in Müller et al. (2015a) that the k600 values determined in this way are more appropri-

ate than commonly used wind speed parameterizations, which neglect the influence of tidal currents and the water flow velocity.

2. On a related point, gas transfer velocities can be temporally and spatially heterogeneous. Were the floating chamber measurements made near the field site? Were they made upstream, downstream, or along the length of the estuaries? A brief mention of the location or timing of the floating chamber measurements might give a better idea of the precision of this approach to k-value calculation.
We will add this information in the methods section as indicated in the reply to your comment above.

3. The authors thoroughly document the source of the atmospheric mixing ratios of N2O and CH4 (which are needed in order to calculate water-to-air fluxes). However, from what I understand, the N2O flux totals are likely going to be much more sensitive to the choice of k-value. Is there some reason why the k-value would be particularly sensitive to the atmospheric mixing value (say if it were 325.25 ppb rather than 325.15 ppb)? Considering that the local air mixing ratio could be slightly greater or less than the Mauna Loa value, it might be good to mention that this isn't a large source of error in the calculation.
The k600 values were derived from CO_2 fluxes measured with a floating chamber and simultaneous measurements of the pCO_2 in water and in the air. That is, when we determined the k600 in the first place, we used atmospheric concentrations measured at the site and not mixing ratios determined elsewhere. Therefore, for the k600 values themselves, the error for $pCO2^{air}$ is not significantly larger than the instrument's uncertainty. Nevertheless, as you mentioned in comment 2 as well, the k600 can be temporally and spatially variable. Therefore, with respect to the calculated N_2O and CH_4 fluxes, k600 still causes the largest uncertainty. We did a simple error propagation calculation on the average values, assuming an error of 60% for k, 0.5% for K_0, 2% for $pCH_4{}^{water}$ and 0.2% for pN_2O^{water} (disequilibrium error, Johnson 1999) and 1% for pCH_4 and pN_2O in air. As a result, the uncertainty in k accounts for 96% of the flux un-

Printer-friendly version

Discussion paper

certainty. Therefore, we think our approach of reporting the flux with the % uncertainty of k600 is justified. We will change the caption of Table 4 to

k600 values and median N_2O and CH_4 areal and total fluxes from the Lupar, Saribas and the Saribas tributary. The uncertainties relate to the maximum variability of the k600 value, as the k600 uncertainty propagation was responsible for approximately 96% of the flux uncertainty. Values are given for the spatial extent of the rivers that was covered in 2013.

Interactive comment on Biogeosciences Discuss., doi:10.5194/bg-2016-4, 2016.

List of relevant changes in the manuscript

Pages and line numbers refer to the marked-up version of the manuscript.

- Methods: Details about the oxygen sensor calibration (p 5, line 14-15) and the k600 values used (p.6 , lines 27-31) were added.

- Results: Since the apparent undersaturation was an artifact, the two datapoints were excluded from the analysis. As a consequence, a few numbers were corrected in Tables 2,3 and 4 and in the text (p. 8, l. 3-4 & 22, p.9, l. 17-18).

- Discussion: A few discussion points were extended or made more explicit. This includes the additional N2O sources in the wet season (p. 10, l.25-26, p.11, l. 1-10), the lack of data for the upper estuary (p.10, l.30- p.11, l. 1 and 22-23) and the explanation for the low CH4 values (p. 11,l. 25-26).

- References: A few references were added to support the discussion (Abril et al. 2007, Barnes and Owens 1998, de Angelis and Scranton 1993, de Wilde and de Bie 2000, Robinson et al. 1998). One study was updated (Müller et al., 2015a: previously a discussion paper, now finally published and referred to as Müller et al. 2016).

- Tables: The caption of Table 4 was changed, numbers in the Tables were corrected as indicated above.

- Figures: The map (Fig 1) was modified according to Referee 1's suggestions; all other Figures were slightly modified (two datapoints were removed as indicated above).

- Grammar, phrasing and spelling: Changes were made in p. 2 l. 16, p. 7 l. 19, p. 88, l. 11 & 15, p. 9. l. 29, p 10, l. 25-26, p 27 l. 5

Nitrous oxide and methane in two tropical estuaries in a peat-dominated region of North-western Borneo

D. Müller[1], H. W. Bange[2], T. Warneke[1], T. Rixen[3,4], M. Müller[5], A. Mujahid[6], and J. Notholt[1,7]

[1] Institute of Environmental Physics, University of Bremen, Otto-Hahn-Allee 1, 28359 Bremen, Germany
[2] GEOMAR Helmholtz Centre for Ocean Research Kiel, Düsternbrooker Weg 20, 24105 Kiel, Germany
[3] Leibniz Center for Tropical Marine Ecology, Fahrenheitstr. 6, 28359 Bremen, Germany
[4] Institute of Geology, University of Hamburg, Bundesstr. 55, 20146 Hamburg, Germany
[5] Swinburne University of Technology, Faculty of Engineering, Computing and Science, Jalan Simpang Tiga, 93350 Kuching, Sarawak, Malaysia
[6] Department of Aquatic Science, Faculty of Resource Science & Technology, University Malaysia Sarawak, 94300 Kota Samarahan, Sarawak Malaysia
[7] MARUM Center for Marine Environmental Sciences at the University of Bremen, Leobener Str., 28359 Bremen, Germany

Correspondence to: D. Müller (dmueller@iup.physik.uni-bremen.de)

Abstract. Estuaries are sources of nitrous oxide (N_2O) and methane (CH_4) to the atmosphere. However, our present knowledge of N_2O and CH_4 emissions from estuaries in the tropics is very limited because data is scarce. In this study, we present first measurements of dissolved N_2O and CH_4 from two estuaries in a peat-dominated region of north-western Borneo. Two campaigns (during the dry season in June 2013 and during the wet season in March 2014) were conducted in the estuaries of the rivers Lupar and Saribas. Median N_2O concentrations ranged between 7.2 and 12.3 nmol L^{-1} and were higher in the marine end-member (13.0 ± 7.0 nmol L^{-1}). CH_4 concentrations were low in the coastal ocean (3.6 ± 0.2 nmol L^{-1}) and higher in the estuaries (medians between 12.2 and 64.0 nmol L^{-1}). The respiration of abundant organic matter and presumably anthropogenic input caused a slight eutrophication, which did not lead to hypoxia or enhanced N_2O concentrations, however. Generally, N_2O concentrations were not related to dissolved inorganic nitrogen concentrations. Thus, the use of an emission factor for the calculation of N_2O emissions from the inorganic nitrogen load leads to an overestimation of the flux from the Lupar and Saribas estuaries. N_2O was negatively correlated with salinity during the dry season, which suggests a riverine source. In contrast, N_2O concentrations during the wet season were not correlated with salinity but locally enhanced within the estuaries, implying that there were additional estuarine sources during the wet (i.e. monsoon) season. Estuarine CH_4 distributions were not driven by freshwater input but rather by tidal variations. Both N_2O and CH_4 concentrations were more variable during the wet season. We infer that the wet season dominates the variability of the N_2O and CH_4 concentrations and subsequent emissions from tropical estuaries. Thus, we speculate that any changes in the Southeast Asian monsoon system will lead to changes in the N_2O and CH_4 emissions from these systems. We also suggest that the ongoing cultivation of peat soil in Borneo is likely to increase N_2O emissions from these estuaries, while the effect on CH_4 remains uncertain.

1. Introduction

Nitrous oxide (N_2O) and methane (CH_4) are greenhouse gases whose global warming potentials exceed that of carbon dioxide (CO_2) by far (a factor of 265 for N_2O and 28 for CH_4 on a 100 year time horizon, Myhre et al., 2013). Thus, an assessment of the natural and anthropogenic sources and sinks as well as the formation pathways of N_2O and CH_4 is essential to understand present Earth's climate variability and to predict its future development. The world's oceans, including its coastal zones, are sources of N_2O and CH_4 to the atmosphere and play a major role in the global budget of atmospheric N_2O, but only a minor role in the global budget of atmospheric CH_4 (Ciais et al., 2013). Rivers and estuaries are considered hotspots for the production and emission of both N_2O and CH_4 (Bange, 2006; Bastviken et al., 2011; Borges et al., 2015; Murray et al., 2015; Seitzinger and Kroeze, 1998).

In aquatic systems, N_2O is mainly formed as a byproduct during nitrification ($NH_4^+ \rightarrow NO_3^-$) and to minor extent as an intermediate during denitrification ($NO_3^- \rightarrow N_2O \rightarrow N_2$) (Freing et al., 2010). In both processes, the yield of N_2O strongly depends on the concentration of ambient oxygen (O_2) (e.g. Bange, 2008). Both nitrification and denitrification are microbial processes and occur in the water column and in sediments (e.g. Bange, 2008). CH_4 is formed during the decomposition of organic material by microbial methanogenesis (e.g. Reeburgh, 2007; Valentine, 2011; Ferry, 2010). Since CH_4 formation requires strictly anaerobic conditions, it is produced in anoxic environments as found in sediments, in the interior of suspended particles or in zooplankton guts (see e.g. Reeburgh, 2007; Valentine, 2011). Additionally, CH_4 is oxidized under aerobic and anaerobic conditions in the water column and in the sediments, respectively (e.g., Valentine, 2011). On the continental shelf so-called geological CH_4 can be released directly to the water column through mud volcanoes, via groundwater input or seeping at pockmark structures (e.g., Valentine, 2011). Alternative aerobic CH_4 formation pathways in the nutrient poor (oligotrophic) surface ocean have been suggested (see e.g. Karl et al., 2008; Damm et al., 2010; Zindler et al., 2013), but they are negligible in nutrient rich (eutrophic) coastal systems.

In this study, we present first measurements of dissolved N_2O and CH_4 from the estuaries of the rivers Lupar and Saribas, which are located in north-western Borneo (Sarawak, Malaysia, Southeast Asia). These measurements are complemented by dissolved N_2O and CH_4 measurements from a cruise on the adjacent coastal shelf of the South China Sea. The Lupar and Saribas rivers drain catchments which are covered by tropical peatlands, parts of which have been converted to oil palm plantations or other crops. This study aimed at investigating the effect of the carbon-rich peatlands on the N_2O and CH_4 estuarine distributions and emissions and the potential impact of estuarine eutrophication. Two intensive sampling campaigns took place during the dry and wet seasons in June 2013 and March 2014, respectively, and provide a first conception of the seasonality in these systems.

2. Materials and Methods

2.1 Study Area

Our study was conducted in Sarawak, the largest Malaysian state, which is located in the north of the island of Borneo (see Fig. 1a). About 12 % of the area of Sarawak is covered by peatlands (Chai, 2005), approximately 41 % of which have been converted to oil palm plantations (SarVision, 2011). Sarawak has a tropical climate with a mean annual air temperature of 26.1°C in the capital Kuching and average (1961-1990) annual rainfall of 4101 mm yr^{-1} (DWD, 2007). Between November and February, Sarawak experiences enhanced rainfall due to the Northeast monsoon, while June and July are the driest months (DWD, 2007).

Two macrotidal estuaries which enclose Malaysia's largest peat dome, the Maludam peninsula, are in the focus of this study. The catchment areas of the Lupar and Saribas rivers are 6558 km^2 and 1943 km^2, respectively (Lehner et al., 2006), with approximate discharges of 490 $m^3\,s^{-1}$ and 160 $m^3\,s^{-1}$ (Müller et al., 2016~~5a~~). The Lupar catchment comprises mainly the division of Sri Aman, which has a population of approximately 95,000. The Saribas basin belongs largely to the Betong division, with a population of approximately 110,000 (SPU, 2012). Major settlements along the Lupar River and estuary are Sri Aman, Lingga and Sebuyau (Fig. 1b). Important settlements along the Saribas River and estuary are Betong, Pusa and Beladin. Oil palms are being cultivated in both catchments (Fig. 1b).

In the Lupar and Saribas estuaries, sampling took place during two campaigns in June 2013 and March 2014. Our sampling strategy is described in detail in Müller et al. (2016~~5a~~). In that study, we showed that precipitation during June 2013 and March 2014 did not deviate much from the historical average, so that we considered the measurements in June 2013 representative of the dry season and those in March 2014 representative of the wet season. Therefore, we refer to the two campaigns as "MLD dry" (June 2013) and "MLD wet" (March 2014). Note that the peak of the monsoon season is between December and February, so the monsoon recedes in March, which is why our statements about seasonality are conservative.

We complement this dataset with measurements that were performed during the cruise SO218 – SHIVA with the German research vessel (R/V) *Sonne* (15-29 November 2011) (Quack and Krüger, 2013). The cruise started in Singapore and went along the Sarawakian coastline to end in Manila, Philippines (see Fig. 1a). For this study, we selected those samples that were taken in the coastal ocean off north-western Sarawak (latitude <2.5, 110.5<longitude<111.5) in order to complement our dataset from the Lupar and Saribas estuaries.

2.2 Water chemistry

In the Lupar and Saribas inner and outer estuaries, we sampled 20 stations during the dry season and 23 during the wet season. Water samples were taken from approximately 1 m below the water surface. Samples were taken for dissolved organic carbon (DOC), dissolved inorganic (nitrogenous) nutrients (DIN = NO_3^-, NO_2^-, and NH_4^+), salinity, water temperature and O_2:

1. Samples for DOC were filtered (pore size 0.45 µm) and acidified with 21 % phosphoric acid until the pH had dropped below 2. DOC concentrations were determined by high temperature combustion and subsequent measurement of resultant CO_2 with a non-dispersive infrared detector (NDIR) (EPA method 415.1). Details are given in Müller et al. (2016~~5a~~).
2. Inorganic nutrient samples were filtered through a Whatman glass microfibre filter (pore size 0.7 µm), preserved with a mercuric chloride ($HgCl_2$) solution and frozen until analysis. Concentrations of NO_3^-, NO_2^- and NH_4^+ were determined spectrophotometrically (Hansen and Koroleff, 1999) at a wavelength of 540 nm with a Continuous Flow Analyzer (Alliance, Austria).
3. Salinity and temperature were measured with a CastAway CTD at each station during the MLD cruises. Additionally, we measured conductivity using a TetraCon 925 conductivity sensor (WTW, Germany). We converted conductivity to salinity using the equations from Bennett (1976). During SO218, salinity and temperature were measured continuously with a thermosalinograph on board.
4. Dissolved oxygen (DO) was measured using a Multi3420 with an FDO 925 oxygen sensor (WTW, Germany). A routine function check in water vapor saturated air was conducted prior to the measurements using the check and calibration vessel that was provided with the sensor.

2.3 N_2O and CH_4 measurements

Our setup during the MLD cruises is described in detail in Müller et al. (2016~~5a~~). Surface water was pumped through a Weiss equilibrator (Johnson, 1999) at a rate of approximately 20 L min^{-1}. The headspace air was analyzed using an in-situ Fourier Transform InfraRed (FTIR) trace gas analyzer (University of Wollongong, Australia). This instrument allows for the continuous and simultaneous measurements of several trace gas species, such as N_2O and CH_4 as well as CO_2 and CO (Müller et al., 2016~~5a~~) with high accuracy and precision over a wide range of concentrations (Griffith et al., 2012). Spectra were averaged over 5 min and dry air mole fractions were retrieved using the software MALT5 (Griffith, 1996). The gas dry air mole fractions were corrected for pressure, water and temperature cross-sensitivities (Hammer et al., 2013). The CO_2 and CO data obtained from these measurements have been reported by Müller et al., (2016~~5a~~). Here, we present the N_2O and CH_4 measurements. Calibration was achieved by measuring a suite of gravimetrically prepared gas mixtures (Deuste Steininger) ranging from 324 to 3976 ppb N_2O and 1.8 to 239 ppm CH_4 in synthetic air, which were calibrated against the World Meteorological Organization (WMO) reference scale at the Max-Planck-Institute for Biogeochemistry in Jena, Germany. The error associated with the FTIR retrieval is usually small. In ambient air, the total uncertainties reported by Hammer et al. (2013) are 0.084 nmol mol^{-1} for N_2O and 0.25 nmol mol^{-1} for CH_4, corresponding to approximately 0.01 % and 0.03 %. The larger source of uncertainty is a potentially remaining disequilibrium between water and headspace in the equilibrator, which can cause an error of <0.2 % for N_2O and 2 % for CH_4 (Johnson, 1999).

Water temperature in the equilibrator and in the water as well as ambient air temperature and pressure were monitored as described in Müller et al. (2016~~5a~~). N_2O and CH_4 partial pressures were calculated. Since the sample air was dried before entering the FTIR analyzer, we corrected for the removal of water vapor (Dickson et al., 2007). N_2O molar concentrations were calculated from N_2O fugacity and solubility (K_0) according to Weiss and Price (1980). CH_4 molar concentrations were calculated from CH_4 partial pressure and solubilities were derived from the equations given by Wiesenburg and Guinasso (1979).

During the R/V *Sonne* cruise SO218, surface sea water was continuously supplied from the ship's hydrographic shaft (moon pool) using a submersible pump at about 4 m water depth. N_2O and CH_4 samples were taken in triplicates, preserved with $HgCl_2$ and analyzed in the lab using headspace equilibration and gas chromatography. Details about the analytic procedures can be found in Walter et al. (2006) and Bange et al. (2010). The average of the three samples was calculated and data was discarded if the standard deviation exceeded 30 % of the average value. For N_2O, two additional data points were taken from the surface N_2O concentrations determined in depth profiles.

Atmospheric mixing ratios of N_2O were taken from the Mauna Loa (Hawaii) monitoring station of the NOAA/ESRL halocarbons in situ program (Dutton et al., 2015) which was the nearest atmospheric N_2O monitoring station in the northern hemisphere. Atmospheric N_2O in Mauna Loa averaged 325.15 ppb in November 2011, 326.26 ppb in June 2013 and 327.08 ppb in March 2014. Atmospheric CH_4 was derived from the NOAA/ESRL GMD Carbon Cycle Cooperative Global Air Sampling Network (Dlugokencky et al., 2014). The nearest atmospheric CH_4 monitoring station was Bukit Kototabang, Indonesia. Unfortunately, CH_4 data from NOAA/ ESRL were not available for March 2014, so we estimated the atmospheric CH_4 during that month from the value reported for March 2013 and an annual growth rate of 4 ppb between 2004-2013 at Bukit Kototabang. Atmospheric CH_4 averaged 1841.64 ppb in November 2011, 1798.64 ppb in June 2013 and a value of 1879.35 ppb was derived for March 2014.

2.4 Flux estimation

In order to calculate N_2O and CH_4 flux densities F (in nmol m^{-2} s^{-1}) across the water-air interface, we used the thin film model, which reads

$$F = kK_0(pGas_{water} - pGas_{air})f, \tag{1}$$

where k is the gas exchange velocity (m s^{-1}), K_o ist the solubility of N_2O and CH_4 in seawater (mol L^{-1} atm^{-1}, see Sect. 2.3), $pGas_{water}$ is the partial pressure derived from the equilibrator measurements (natm), $pGas_{air}$ is the partial pressure of the gas in air (natm) as measured at the atmospheric monitoring stations (see Sect. 2.3), and f is a conversion factor from L^{-1} to m^{-3}. For k, we used k_{600} values that were derived for the Lupar and Saribas estuaries using the floating chamber method (Müller et al., 2016). Floating chamber measurements were conducted at several locations along the estuaries during the wet season campaign and averaged over the spatial extent of the individual estuaries. We argued in Müller et al. (2016) that the k_{600} values determined in this way are more appropriate than commonly used wind speed parameterizations, which neglect the influence of tidal currents and the water flow velocity. ~~For k, we used k_{600} values that were reported for the Lupar and Saribas estuaries in Müller et al. (2015a).~~ In-situ k was calculated based on the Schmidt numbers of N_2O and CH_4, which relates the kinematic viscosity to the diffusivity of the gas in water. Kinematic viscosity was calculated according to Siedler

Formatted: Normal

Formatted: Font: 10 pt, Not Italic, Font color: Auto, English (U.S.)

Formatted: Font: Not Italic, Font color: Auto, English (U.S.)

Formatted: Font: 10 pt, Not Italic, Font color: Auto, English (U.S.)

Formatted: Font: 10 pt, Not Italic, Font color: Auto, English (U.S.)

Formatted: Font: 10 pt, Not Italic, Font color: Auto, English (U.S.)

Formatted: Font: 10 pt, Not Italic, Font color: Auto, English (U.S.)

and Peters (1986), the diffusivity of N_2O was computed using Eq. 2 in Bange et al. (2001) and the diffusivity of CH_4 was calculated with the formula given in Jähne et al. (1987). The annual areal flux density was estimated as the average of the wet and dry season values (for the spatial extent that was covered during the 2013 cruise, see Müller et al., 2016~~5a~~). The total N_2O and CH_4 fluxes (t N yr^{-1} and t C yr^{-1}) were calculated assuming an estuarine surface area of 220 km² for the Lupar and 102 km² for the Saribas estuary (Müller et al., 2016~~5a~~). For N_2O, we compare this estimate to one derived using the DIN export and the emission factor for estuaries suggested in Mosier et al., (1998) (0.0025 kg N_2O-N / kg N leaching and runoff). The DIN export was calculated from river discharge (see Sect. 2.1) and the median DIN concentration.

3. Results

3.1 Water chemistry

In the estuaries of Lupar and Saribas, salinity ranged from 0-30.6 in the dry season and from 0-31.0 in the wet season. N_2O and CH_4 concentration data are available for salinities of 4.3-26.5 (MLD dry) and 6.9-26.4 (MLD wet). For the coastal ocean off north-western Sarawak, N_2O measurements covered salinities between 31.3 and 32.7 and CH_4 measurements between 32.2 and 32.7. DO saturation in the estuaries ranged between 63.6 to 94.6 % (MLD dry) and 79.0-100.4 % (MLD wet) (Müller et al., 2016~~5a~~).

The total DIN concentrations were already published in Müller et al. (2016~~5a~~). Here, we report the concentrations of the different inorganic nitrogen species. Generally, DIN concentrations were quite low, but locally enhanced. NO_3^- ranged between 6.3 µmol L^{-1} and 36.3 µmol L^{-1} in the dry season and between 2.8 and 17.9 µmol L^{-1} in the wet season. A maximum value of 84.0 µmol L^{-1} was observed during MLD dry approximately 20 km offshore in north-western direction from Sebuyau. There, an influence from the Lupar river plume, but possibly also from the close-by Sadong R~~r~~iver was detected (salinity = 22.2).

NO_2^- concentrations ranged between 0.1 µmol L^{-1} and 0.6 µmol L^{-1} in the dry season and between <0.1 µmol L^{-1} and 2.3 µmol L^{-1} in the wet season. NH_4^+ ranged between <0.1 µmol L^{-1} and 2.6 µmol L^{-1} in the dry season, whereas a maximum value of 8.0 µmol L^{-1} was observed at one station in the Lupar estuary. In the wet season, NH_4^+ was higher, ranging between 0.2 µmol L^{-1} to 7.8 µmol L^{-1}. However, overall, DIN concentrations were higher in the dry season than in the wet season (Müller et al., ~~2015a~~2016, Table 1).

On the Lupar River, we determined average DIN concentrations of 5.1 µmol L^{-1} (2013) and 5.3 µmol L^{-1} (2014) upstream of the town of Sri Aman, which can be considered unpolluted. This value is 2-6 times lower than the DIN concentrations in the Lupar estuary. On the Saribas River, a slightly enhanced value of 18.6 µmol L^{-1} (2013) was measured at salinity 0 outside the town of Betong, so it cannot be considered unpolluted, yet it was lower than the DIN concentrations in the estuary during that season (Table 1). We estimated that together, Lupar and Saribas deliver 6086 t N as DIN to the South China Sea every year (Table 1).

3.2 N_2O

Dissolved N_2O concentrations in the water ranged from 6.9 nmol L^{-1} to 13.4 nmol L^{-1} during MLD dry in June 2013 (corresponding to saturations of 103 to 184 %) and from 6.~~3~~6 nmol L^{-1} to 116.8 nmol L^{-1} during MLD wet in March 2014 (~~93~~102 to 1679 % saturation). Most of the time, N_2O was close to atmospheric equilibrium. However, local enhancements were observed: During both the dry and the wet season, the Saribas tributary exhibited markedly higher N_2O concentrations than the Saribas main river (Fig. 3a,b, Table 2). During the wet season, N_2O concentrations were more variable (higher standard deviation (SD)) and higher maximum concentrations were observed (Table 2). Although the mean N_2O concentrations in the Saribas estuary and Saribas tributary were higher in the wet season as well, no difference was found with regards to the medians (Table 2).

Salinity and N_2O concentrations were correlated in the dry season (r = -0.62), but uncorrelated in the wet season (Fig. 2c). By the town of Sebuyau at the Lupar river mouth, where the Sebuyau R~~r~~iver flows into the South China Sea, N_2O was enhanced with concentrations of up to 116.8 nmol L^{-1} in the wet season (not shown in Fig. 2~~,3~~-4). The data from SO218 revealed enhanced N_2O concentrations offshore during November 2011 (see Fig. 2c), with a median of 13.0 ± 7.0 nmol L^{-1} (218 ± 119 % saturation, see Table 2).

N_2O was correlated with DOC (Fig. 2a);~~, whereas~~ this correlation was strong in the dry season (r=0.87) and weak during the wet season (r=0.38). N_2O concentrations were not correlated with DIN (Fig. 2b), NO_3^-, NH_4^+ (not shown) or DO (Fig. 2d). During the dry season, we found no link between N_2O concentrations and tidal variations (Fig. 4a). In the wet season, however, N_2O exhibited slightly higher concentrations during low tide and its spatiotemporal variation resembled that of CH_4 (Fig. 4b).

3.3 CH_4

Dissolved CH_4 concentrations ranged from 5.2 nmol L^{-1} to 59 nmol L^{-1} during MLD dry in 2013 (228 to 2782 % saturation) and from ~~2.0~~3.7 nmol L^{-1} to 135 nmol L^{-1} during MLD wet in 2014 (~~88~~168 to 6003 % saturation) in the Lupar and Saribas estuaries and was spatially variable. The highest value was measured by the town of Sebuyau during MLD wet, whereas the highest median concentrations were detected in the Saribas estuary during the dry season and in the Saribas tributary during the wet season (Fig. 3c,d; Table 3). In general, no seasonal pattern could be identified. Mean and median CH_4 concentrations in the Lupar estuary were comparable in the dry and wet season. In the Saribas estuary, CH_4 concentrations were higher during the dry season but on the Saribas tributary, they were higher during the wet season (Table 3). However, higher maximum concentrations were observed in the wet season in both estuaries and the tributary (Table 3).

CH_4 concentrations were not correlated with salinity (Fig. 5c). A relatively low marine end-member concentration was determined during the SO218 cruise: With a median of 3.6 ± 0.2 nmol L^{-1} (176 ± 9 % saturation), CH_4 concentrations in the coastal ocean were only slightly enhanced in comparison to the atmospheric equilibrium concentration (see Table 3).

CH_4 was not correlated with DOC (Fig. 5a), DO (Fig. 5d), DIN or suspended particulate matter (not shown), but increased with increasing pCO_2 (Fig. 5b). This relationship was stronger in the wet season (r=0.55) than in the dry season (r=0.15). One striking feature was that CH_4 showed a strong response to the tides (Fig. 4). This is visible for most of the data, even though we changed location during the measurements. Tidal and spatial variations are overlapping in Fig. 4, but the tidal variation seems to dominate. This is confirmed by one stationary measurement that we conducted over night at one station on the Saribas tributary in 2014 (Fig. 4b): CH_4 peaked 4-fold during low tide if compared to the concentration level at high tide.

3.4 N_2O and CH_4 flux densities

Median N_2O and CH_4 flux densities are listed in Table 4. Both the highest N_2O flux density and the highest CH_4 flux density were computed for the Saribas tributary. During both seasons, N_2O flux densities from the Saribas tributary were up to one order of magnitude higher than from the Lupar and the Saribas estuaries. Annual average N_2O flux densities were low for both Lupar and Saribas, amounting to 1.3 ± 0.3 mmol m^{-2} yr^{-1} and 1.9 ± 1.6 mmol m^{-2} yr^{-1}, respectively. The N_2O flux density from the Saribas tributary was one order of magnitude higher (12.0 ± 7.5 mmol m^{-2} yr^{-1}).

The CH_4 flux density observed on the Saribas tributary during the wet season was approximately five times higher than the flux densities from Lupar or Saribas during any season. As a result, the annual average flux from the Saribas tributary (89.2 ± 55.2 mmol m^{-2} yr^{-1}) was approximately four times as high as those computed for the Lupar and Saribas estuaries, which were comparable (22.~~4~~5 ± 5.4 mmol m^{-2} yr^{-1} and 23.0 ± 19.1 mmol m^{-2} yr^{-1}, respectively).

In total, 4 ± 1 t N_2O-N yr^{-1} and ~~59~~60 ± 17 t CH_4-C yr^{-1} were emitted from the Lupar estuary and 3 ± 2 t N_2O-N yr^{-1} and 28 ± 25 t CH_4-C yr^{-1} from the Saribas (Table 4). Using the emission factor of Mosier et al., (1998), we obtained 11 t N_2O-N yr^{-1} for the Lupar and 4 t N_2O-N yr^{-1} for the Saribas estuary.

4. Discussion

4.1 Eutrophication in the Lupar and Saribas estuaries

Blackwater rivers and their estuaries usually have very low nutrient concentrations (<1 µmol L^{-1} NO_3^-, Kselik and Liong 2004; <5 µmol L^{-1} NO_3^-, Alkhatib et al., 2007). Although the Lupar and Saribas rivers are not blackwater rivers, they have several blackwater tributaries (Kselik and Liong 2004; Müller et al., ~~2015a~~2016), and 30.5 % and 35.5 % of their catchments are covered by peat (Müller et al., ~~2015a~~2016). Therefore, rather low nutrient concentrations were expected. However, several villages and smaller towns are found along the shore of both estuaries, and there is cultivation of sago and oil palm in the catchments (Fig. 1b).

Estuarine DIN concentrations were higher than in the unpolluted freshwater end-member of the Lupar R~~r~~iver, indicating that the estuary was indeed slightly eutrophic during the time of our measurements. This eutrophication can be attributed both to the release of DIN during respiration of organic matter, which was shown to be pronounced in the Lupar and Saribas estuaries (Müller et al., ~~2015a~~2016), especially in the dry season (see discussion below), and to anthropogenic input. These processes were also identified as important sources of inorganic nutrients in the Siak River, a eutrophic blackwater river in central Sumatra, Indonesia (Baum and Rixen, 2014). The DIN concentrations in the Lupar and Saribas estuaries were similar to those reported by Baum and Rixen (2014) for the Siak River.

Eutrophication can lead to enhanced estuarine primary production and consequently to hypoxia. This was not observed. Although organic matter was respired in the estuaries (Müller et al., ~~2015a~~2016), oxygen depletion was relatively moderate in the surface water due to a quick replenishment from the overlying air, as suggested by the high gas exchange velocity (Table 4). Since we did not measure oxygen profiles, we can only speculate about the oxygen levels in the bottom water. The high turbulence in the water likely prevented stratification and promoted the ventilation of the water column and, therefore, may have prevented the development of anoxic bottom waters.

4.2 N_2O

Dissolved N_2O concentrations were mostly close to atmospheric equilibrium concentrations (i.e. 100% saturation), which is in line with other comparable studies in the tropics and subtropics (Richey et al., 1988; Zhang et al., 2010; Rao and Sarma 2013; Borges et al., 2015). High N_2O concentrations were reported for eutrophic and hypoxic coastal waters in the western Indian continental shelf (up to 533 nmol L^{-1}, Naqvi et al., 2000) and for the Peruvian upwelling (up to 986 nmol L^{-1}, Arevalo-Martinez et al., 2015). Similarly, N_2O concentrations in the subtropical Brisbane estuary were higher than in the Lupar and Saribas (median concentrations between 7.2 and 12.3 nmol L^{-1}), ranging between 9.1 and 45 nmol L^{-1} (Musenze et al., 2014).

The low N_2O concentrations in the Lupar and Saribas estuaries are not surprising. Even though these estuaries were slightly eutrophic, the DIN concentrations were still below the average for tropical non-blackwater rivers (Baum and Rixen, 2014). For estuaries with low DIN concentrations, low N_2O concentrations are expected (Zhang et al., 2010). Interestingly, enhanced N_2O was measured offshore during cruise SO218. Either~~,~~ a source of N_2O exists on the continental shelf, or the enhanced N_2O concentrations are due to interannual variability: The SO218 cruise took place in 2011, while the measurements in the Lupar and Saribas estuaries were performed in 2013 and 2014. Ultimately, the SO218 measurements were performed at the onset of the monsoon season, so possibly, seasonal variability plays a role~~ as well~~. This is considered most likely, as it is in line with the tendency towards higher and more variable N_2O concentrations during MLD wet if compared to MLD dry in the Saribas tributary.

Stronger oxygen depletion and higher DIN concentrations suggest higher respiration rates during the dry season (Müller et al., ~~2015a~~2016). In accordance with this, N_2O was well correlated with DOC and salinity during the dry season, suggesting that it originated mainly from respiratory activity in ~~freshwater~~the upper estuary. In this study, samples were not collected for salinities<4, so we cannot make this claim with certainty. However, previous studies suggest that the highest N_2O concentrations are usually encountered at low salinities in the vicinity of the turbidity maximum (Robinson et al., 1998, Barnes and Owens, 1999, Zhang et al., 2010). On the other hand, N_2O maximum concentrations along estuaries may also occur at intermediate salinities (de Wilde and de Bie, 2000). This seems to have been the case during our wet season campaign, where we observed the highest N_2O concentrations at salinities between 10-20 (Fig. 2c). N_2O concentrations were not correlated with salinity during the wet season ~~In contrast, during the wet season, a correlation of N_2O with salinity was not observed~~ and the correlation with DOC was weak, suggesting that additional sources of N_2O existed in the estuary and obscured these relationships. The nature of this additional estuarine source is indicated by the covariation of ~~In the wet season,~~ N_2O ~~showed the same spatial variations as~~and CH_4.~~,~~ ~~which were~~The variability of both gases was linked to the tidal cycle, which was not the case for N_2O during MLD dry (see Fig. 4). This implies that N_2O and CH_4 had the same source during the wet season and that this source was located within the estuary at intermediate salinities. In line with the tidal variability, we propose production of both N_2O and CH_4~~, e.g., production~~ in estuarine sediments or in tidal creeks as a common source of both gases~~.~~

The most striking feature of the spatial distribution of N_2O was its strong variability, with enhanced concentrations in the Saribas tributary during both seasons, indicating a local source. The most obvious candidate for a point source is anthropogenic, i.e. sewage. However, NO_3^-, NO_2^-, and NH_4^+ were not enhanced in the same way as N_2O, suggesting that DIN concentrations are a poor predictor for estuarine N_2O concentrations (see discussion below).

4.3 CH_4

Similar to N_2O, dissolved CH_4 concentrations were relatively moderate. For example, while CH_4 concentrations in the partially hypoxic Pearl River estuary ranged between 23 and 2984 nmol L^{-1} (Chen et al., 2008), median CH_4 concentrations in our study area varied between 4-64 nmol L^{-1}. Similarly, the CH_4 concentration range reported by Musenze et al. (2014) for the Brisbane estuary, Australia, was substantially higher (31-578 nmol L^{-1}) than in this study, although both the DOC concentration range and the DO saturation range were similar. This is surprising, as the peat-draining tributaries are extremely oxygen-depleted and contain large amounts of organic matter (Müller et al., 2015~~b~~). These conditions are usually suitable for CH_4 production. Although we do not have data on the river end-member CH_4 concentration, we can infer from ~~However,~~ the lack of correlation between CH_4 and salinity ~~implies~~ that freshwater input was not the main source of CH_4 in the estuaries. Since CH_4 is a poorly soluble gas, a large fraction might be released to the atmosphere before reaching the estuary and the coastal ocean. Another possible explanation is methane oxidation, which was shown to remove large fractions of CH_4 in some estuaries (deAngelis and Scranton 1993, Abril et al. 2007). ~~In addition, salt intrusion may inhibit CH_4 production in the estuaries.~~ Although CH_4 concentrations tended to be higher during the wet season, a clear seasonal pattern of CH_4 concentrations was not apparent, which is in line with observations at other tropical (Barnes et al., 2006; Teodoru et al., 2014) and subtropical (Musenze et al., 2014) sites.

The generally positive relationship between CH_4 and pCO_2 has been observed in other tropical aquatic systems (Teodoru et al., 2014, Borges et al., 2015) and is indicative of organic matter decomposition as a source of both gases. The strong response of CH_4 to tidal variations indicates that CH_4 is produced in the sediments and released when the hydrostatic pressure drops during falling tide. The tidal variability of CH_4 (and N_2O in the wet season) can also be interpreted as indication of the contribution of intertidal sediments and tidal creeks to the CH_4 concentrations in these estuaries. The importance of tidal creeks (Middelburg et al., 2002) and tidal pumping (Barnes et al., 2006; Borges and Abril, 2011) for CH_4 concentrations in estuaries is widely recognized.

4.4 N_2O and CH_4 flux densities and emissions

With the exception of the Saribas tributary (F_{annual} = 12.0 ± 7.5 mmol m^{-2} yr^{-1}), N_2O flux densities were quite low (1.3-1.9 mmol m^{-2} yr^{-1}) and at the lower end of the range reported for Indian estuaries (-0.4-5.2 mmol m^{-2} yr^{-1}, Rao and Sarma, 2013). CH_4 flux densities (22.~~4~~5-89.2 mmol m^{-2} yr^{-1}) were within the ranges reported for other tropical sites: Koné et al. (2010), for example, determined flux densities of 28.5-123.4 mmol m^{-2} yr^{-1} for stratified lagoons of Ivory Coast; Shalini et al. (2006) report flux densities of 19.7-102.2 mmol m^{-2} yr^{-1} for Pulicat lake, India; and Biswas et al. (2007) measured CH_4 flux densities between 0.7 and 49 mmol m^{-2} yr^{-1} in the estuaries of the Sundarban mangrove ecosystem. However, we caution that the comparison suffers from the different approaches to determining the gas exchange velocity *k*. Koné et al. (2010), Shalini et al. (2006), Biswas et al. (2007) and Rao and Sarma (2013) used empirical equations relating *k* to wind speed, while we used estimates based on floating chamber measurements. We refer to our discussion in Müller et al. (~~2015a~~2016), where we showed that our estimates derived by floating chamber measurements yielded higher values than if we had used empirical relationships with wind speed. We argued that floating chamber measurements offered a better representation of the actual flux than gas exchange models that rely on empirical relationships with wind speed, which were initially derived for the open ocean (Wanninkhof, 1992) and do not consider current-induced turbulence as a driver of gas exchange. Musenze et al. (2014) used empirical models for both the wind-related and the current-related gas exchange velocity and added them up to derive flux estimates for the Brisbane estuary. Not surprisingly, the fluxes they report were much higher than in the aforementioned studies, with 0.8-28.0 mmol N_2O m^{-2} yr^{-1} and 7.5 to 636 mmol CH_4 m^{-2} yr^{-1}. Compared with these estimates, the N_2O and CH_4 fluxes from Lupar and Saribas are quite low, which is consistent with the lower N_2O and CH_4 concentrations as discussed above.

On the basis of our floating chamber measurements (see Müller et al, ~~2015a~~2016) we computed an overall annual flux of 7 t N_2O-N for the Lupar and Saribas estuaries. This is only half of the estimate (15 t N_2O-N yr^{-1}) computed using the Mosier et al. (1998) emission factor. The reason is obviously the missing relationship between DIN concentrations and N_2O emissions in the studied system. Although across estuaries, increasing DIN is generally associated with increasing N_2O (Murray et al., 2015), the relationship is not linear: Zhang et al. (2010) found a polynomial relationship between DIN and N_2O across estuaries in different geographic locations. The emission factor from Mosier et al. (1998), in contrast, is based on constant N_2O/DIN ratio. Therefore, it is not surprising that for low DIN environments, as our study site, the use of emission factors leads to a considerable bias. In line with the data presented here, Borges et al. (2015) did not find a relationship between N_2O and DIN from measurements in several tropical and sub-tropical African rivers and estuaries. This could indicate that this relationship is not applicable in tropical systems and demonstrates the need of further studies in low latitudes to improve estimates of global N_2O emissions from estuaries.

4.5 Implications for future land use change

Nutrient loads to estuaries are predicted to increase in the future in most of Southeast Asia (Seitzinger and Kroeze, 1998). Our results imply that eutrophication in peat-dominated estuaries does not automatically lead to enhanced N_2O emissions. However, the localized elevation of N_2O concentrations, as observed on the Saribas tributary, suggests that estuarine N_2O concentrations might be impacted by local anthropogenic sources. Indeed, it has been shown that N_2O emissions from peat soils depend on land use and that cultivated sites generally exhibit higher N_2O fluxes to the atmosphere (Hadi et al., 2000). It is likely that this behavior is mirrored in the aquatic systems as well.

As CH_4 emissions from peatlands depend largely on the water table (Couwenberg et al., 2010), CH_4 fluxes from peatlands are enhanced under sago (Melling et al., 2005) and rice (Couwenberg et al., 2010) and reduced under oil palm (Melling et al., 2005). The cultivation of sago in the catchment of the Saribas tributary might therefore partially explain the observed high CH_4 fluxes from the Saribas tributary. The future development of CH_4 dynamics in estuaries in this region in dependence of land use change is hard to predict, as there are potentially counteracting effects of the conversion of peatlands to oil palm and other industrial crops, which require different agricultural practices (flooding versus drainage).

5. Conclusions

Overall, we found that the two tropical estuaries of the rivers Lupar and Saribas in a peat-dominated region in Malaysia were small to moderate sources of N_2O and CH_4 to the atmosphere. DIN concentrations were slightly enhanced compared to the unpolluted riverine end-member. This eutrophication did not lead to hypoxia or to enhanced N_2O concentrations. DIN was generally a poor predictor of N_2O, which provides further evidence that the use of emission factors for the calculation of N_2O fluxes from tropical estuaries is inappropriate. Although predictions about the future development of the Southeast Asian monsoon are highly uncertain and locally variable, chances are that rainfall will moderately increase in this region (Christensen et al., 2013). Our results suggest that this may increase the N_2O and probably also the CH_4 emissions from estuaries, as the concentrations of both gases were more variable during the wet season, with higher maximum concentrations of both gases and additional sources of N_2O in the estuaries. For our study area, additional sampling at the peak of the monsoon season would be desirable in order to consolidate these statements. Yet, our results provide a first conception of the seasonality in these systems and underline the fact that time series measurements (seasonal sampling) are vital for the understanding of N_2O and CH_4 fluxes from aquatic systems in monsoonal regions.

Acknowledgments

We would like to thank the Sarawak Biodiversity Center for permission to conduct research in Sarawak waters (Permit No. SBC-RA-0097-MM and export permit SBC-EP-0040-MM). We thank Hella van Asperen (University of Bremen, Germany), Nastassia Denis, Felicity Kuek, Joanne Yeo, Hong Chang Lim, Edwin Sia (all Swinburne University, Malaysia) and all scientists and students from Swinburne University and the University of Malaysia Sarawak who were involved in the MLD cruises and their preparation. Lukas Chin and the crew members of the SeaWonder are acknowledged for their support. We thank Franziska Wittke (GEOMAR), who performed the sampling for N_2O and CH_4 on the R/V *Sonne* cruise SO218 and Annette Kock (GEOMAR), who computed the data from SO218. The authors thank Matthias Birkicht and Dorothee Dasbach (ZMT Bremen, Germany) for their help in the lab performing the analyses of the MLD samples. We acknowledge the University of Bremen for funding the MLD cruises through the "exploratory project" in the framework of the University's Institutional Strategy and the EU FP7 project InGOS for supporting the development of the FTIR measurements. Cruise SO218 was supported by the EU FP7 project SHIVA under grant agreement no. 226224.

References

Abril, G., Commarieu, M. V., and Guérin, F.: Enhanced methane oxidation in an estuarine turbidity maximum. Limnology & Oceanography 52(1): 470-475, 2007.

Alkhatib, M, Jennerjahn, T.C., and Samiaji, J.: Biogeochemistry of the Dumai River estuary, Sumatra, Indonesia, a tropical blackwater river. Limnology and Oceanography 52(6):2410-2417, available at: http://www.jstor.org/stable/4502390. (Accessed 17 June 2015), 2007.

Arévalo-Martínez, D.L., Kock, A., Löscher, C.A., Schmitz, R.A., and Bange, H.W.: Massive nitrous oxide emissions from the tropical South Pacific Ocean. Nature Geoscience, 8:530–533. doi: 10.1038/NGEO2469, 2015.

Bange, H.W.: Nitrous oxide and methane in European coastal waters. Estuarine Coastal and Shelf Science 70:361-374, doi: 10.1016/j.ecss.2006.05.042, 2006.

Bange, H.W.: Gaseous nitrogen compounds (NO, N_2O, N_2, NH_3) in the ocean. In: Capone, D.G., Bronk, D.A., Mulholland, M.R., and Carpenter, E.J. (eds) Nitrogen in the Marine Environment, 2nd edn, Elsevier B.V., Amsterdam, chap 2, pp 51-94, doi: 10.1016/B978-0-12-372522-6.00002-5, 2008.

Bange, H.W., Andreae, M.O., Lal, S., Law, C.S., Naqvi, S.W.A., Oatra, P.K., Rixen, T., and Upstill-Goddard, R.C.: Nitrous oxide emissions from the Arabian Sea: A synthesis. Atmospheric Chemistry and Physics 1:61-71, doi: 10.5194/acp-1-61-2001, 2001.

Bange, H.W., Bergmann, K., Hansen, H.P., Kock, A., Koppe, R., Malien, F., and Ostrau, C.: Dissolved methane during hypoxic events at the Boknis Eck time series station (Eckernförde Bay, SW Baltic Sea). Biogeosciences 7:1279-1284, doi: 10.5194/bg-7-1279-2010, 2010.

Barnes, J. and Owens, N. J. P.: Denitrification and Nitrous Oxide Concentrations in the Humber Estuary, UK, and Adjacent Coastal Zones. Marine Pollution Bulletin 37(3-7): 247-260, 1998.

Barnes, J., Ramesh, R., Purvaja, R., Nirmal Rajkumar, A., Senthil Kumar, B., Krithika, K., Ravichandran, K., Uher, G., and Upstill-Goddard, R. Tidal dynamics and rainfall control N_2O and CH_4 emissions from a pristine mangrove creek. Geophysical Research Letters 33:L15,405, doi: 10.1029/2006GL026829, 2006.

Bastviken, D., Tranvik, L.J., Downing, J.A., Crill, P.M., and Enrich-Prast, A.: Freshwater methane emissions offset continental carbon sink. Science, 331: 50, doi: 10.1126/science.1196808, 2011.

Baum, A. and Rixen, T.: Dissolved inorganic nitrogen and phosphate in the human affected blackwater river Siak, central Sumatra, Indonesia. Asian Journal of Water, Environment and Pollution 11:13-24, 2014.

Bennett, A.S. Conversion of in situ measurements of conductivity to salinity. Deep-Sea Research 23:157-165, 1976.

Biswas, H., Mukhopadhyay, S.K., Sen, S., and Jana, T.K.: Spatial and temporal patterns of methane dynamics in the tropical mangrove dominated estuary, NE coast of Bay of Bengal, India. Journal of Marine Systems 68:55-64, doi: 10.1016/j.jmarsys.2006.11.001, 2007.

Borges, A.V. and Abril, G.: Carbon dioxide and methane dynamics in estuaries. In: Wolanski E, McLusky DS (eds) Treatise on Estuarine and Coastal Science, vol 5, Academic Press, Waltham, pp 119-161, 2011.

Borges, A.V., Darchambeau, F., Teodoru, C.R., Marwick, T.R., Tamooh, F., Geeraert, N., Omengo, F.O., Guérin, F., Lambert, T., Morana, C., Okuku, E., and Bouillon, S.: Globally significant greenhouse-gas emissions from African inland waters. Nature Geoscience, 8: 673-642, doi: 10.1038/NGEO2486, 2015.

Christensen, J.H., Krishna Kumar, K., Aldrian, E., An, S.I., Cavalcanti, I.F.A., de Castro, M., Dong, W., Goswami, P., Hall, A., Kanyanga, J.K., Kitoh, A., Kossin, J., Lau, N.C., Renwick, J., Stephenson, D.B., Xie, S.P., and Zhou, T.: Climate Phenomena and their Relevance for Future Regional Climate Change. In: Stocker, T.F., Qin, D., Plattner, G.K., Tignor, M., Allen, S.K., Boschung, J., Nauels, A., Xia, Y., Bex, V., and Midgley, P.M. (eds.): Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 2013.

Ciais, P., C. Sabine, G. Bala, L. Bopp, V. Brovkin, J. Canadell, A. Chhabra, R. DeFries, J. Galloway, M. Heimann, C., Jones, C. Le Quéré, R.B. Myneni, S. Piao, and P. Thornton, 2013: Carbon and Other Biogeochemical Cycles. In: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change In: Stocker, T.F., Qin, D., Plattner, G.K., Tignor, M., Allen, S.K., Boschung, J., Nauels, A., Xia, Y., Bex, V., and Midgley, P.M. (eds.): Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 2013.

CIFOR Center for International Forestry Research (2014) Industrial plantations in Borneo. Shapefiles for ArcGIS, available at: http://www.cifor.org/. Accessed 17 June 2015, 2015.

Chai, P.K.: Management plan for Maludam National Park Betong Division Sarawak. Tech. rep., Alterra/Forest Department Sarawak/Sarawak Forestry Corporation, Wageningen, 2005.

Chen, C.T.A., Wang, S.L., Lu, X.X., Zhang, S.R., Lui, H.K., Tseng, H.C., Wang, B.J., and Huang, H.I.: Hydrogeochemistry and greenhouse gases of the Pearl River, its estuary and beyond. Quaternary International 186:79-90, doi: 10.1016/j.quaint.2007.08.024, 2008.

Couwenberg, J., Dommain, R., and Joosten, H.: Greenhouse gas fluxes from tropical peatlands in south-east Asia. Global Change Biology 16, 1715-1732, doi: 10.1111/j.1365-2486.2009.02016.x, 2010.

Damm, E., Helmke, E., Thoms, S., Schauer, U., Nöthig, E., Bakker, K., and Kiene, R.P.: Methane production in aerobic oligotrophic surface water in the central Arctic Ocean, Biogeosciences, 7: 1099-1108, doi: 10.5194/bg-7-1099-2010, 2010.

DWD Deutscher Wetterdienst (2007) Climate data worldwide. as Excel file, http://www.dwd.de/bvbw/appmanager/bvbw/dwdwwwDesktop?_nfpb=true&_pageLabel=_dwdwww_spezielle_nutzer_energiewirtschaft_historisch&T26607173141161345039102gsbDocumentPath=Navigation%20FOeffentlichkeit%20FKlima__Umwelt%20FKlimadaten%20Fklimadaten__weltweit%20Fdownload__node.html%20F__nnn%20Dtrue. Accessed 3 June 2015.

de Angelis, M. A., and Scranton, M. I.: Fate of methane in the Hudson River and Estuary. Global Biogeochemical Cycles 7(3): 509-523, 1993.

de Wilde, H. P. J., and de Bie, M. J. M.: Nitrous oxide in the Schelde estuary: production by nitrification and emission to the atmosphere. Marine Chemistry 69: 203-216, 2000.

Dickson, A., Sabine, C., and Christian, G.: Guide to best practices for ocean CO_2 measurements. North Pacific Marine Science Organization (PICES) Special Publication, 191 pp., 3rd edn, available at: http://cdiac.ornl.gov/oceans/Handbook_2007.html. Accessed 6 July 2015, 2007.

Dlugokencky, E.J., Lang, P.M., Crotwell, A.M., Masarie, K.A., and Crotwell, M.J.: Atmospheric Methane Dry Air Mole Fractions from the NOAA ESRL Carbon Cycle Cooperative Global Air Sampling Network, 1983-2013, Version: 2014-06-24. ftp://aftp.cmdl.noaa.gov/data/trace_gases/ch4/flask/surface/. Accessed 10 July 2015, 2014.

Dutton, G.S., Elkins, J.W., and Hall, B.D.: Nitrous oxide data from the NOAA/ESRL halocarbons in situ program, available at: http://www.esrl.noaa.gov/gmd/dv/data/?parameter_name=Nitrous%%2BOxide. Accessed10 July 2015, 2015.

Ferry, J.G.: How to make a living by exhaling methane. Annual Review of Microbiology, 64: 453-473, doi: 10.1146/annurev.micro.112408.134051, 2010.

Freing, A., Wallace, D.W.R., and Bange, H.W.: Global oceanic production of nitrous oxide. Philosophical Transactions of the Royal Society 367:1245-1255, doi: 10.1098/rstb.2011.0360, 2012.

Griffith, D.W.T.: Synthetic calibration and quantitative analysis of gas-phase FT-IR spectra. Applied Spectroscopy 50(1):59-70, 1996.

Griffith, D.W.T., Deutscher, N.M., Caldow, C., Kettlewell, G., Riggenbach, M., and Hammer , S. A Fourier transform infrared trace gas and isotope analyser for atmospheric applications. Atmospheric Measurement Techniques 5:2481-2498, doi: 10.5194/amt-5-2481-2012, 2012.

Hadi, A., Inubushi, K., Purnomo, E., Razie, F., Yamakawa, K., and Tsuruta, H.: Effect of land-use changes on nitrous oxide (N_2O) emission from tropical peatlands. Chemosphere – Global Change Science 2, 347-358, 2000.

Hammer, S., Griffith, D.W.T., Konrad, G., Vardag, S., Caldow, C., and Levin, I.: Assessment of a multi-species in situ FTIR for precise atmospheric greenhouse gas observations. Atmospheric Measurement Techniques 6:1153-1170, doi: 10.5194/amt-6-1153-2013, 2013.

Hansen, H.P. and Koroleff, F.: Determination of nutrients. In: Grasshoff, K., Kremling, K., and Ehrhardt, M. (eds) Methods of Seawater Analysis, 3rd edn, Wiley-VCH, Weinheim, 1999.

Jähne, B., Heinz, G., and Dietrich, W.: Measurement of the diffusion coefficients of sparingly soluble gases in water. Journal of Geophysical Research 92(C10):10,767-10,776, 1987.

Johnson, J.E.: Evaluation of a seawater equilibrator for shipboard analysis of dissolved oceanic trace gases. Analytica Chimica Acta 395:119-132, 1999.

Karl, D.M., Beversdorf, L., Björkman, K.M., Church, M.J., Martinez, A., and Delong, E.F.: Aerobic production of methane in the sea, Nature Geoscience, 1: 473-478, doi: 10.1038/ngeo234, 2008.

Koné, Y.J.M., Abril, G., Delille, B., and Borges, A.V.: Seasonal variability of methane in the rivers and lagoons of Ivory Coast (West Africa). Biogeochemistry 100:21-37, doi: 10.1007/s10533-009-9402-0, 2010.

Kselik, R.A.L., and Liong, T.Y.: Hydrology of the peat swamp in the Maludam National Park Betong Division Sarawak. Tech. rep., Alterra/Forest Department Sarawak/Sarawak Forestry Corporation, Kuching, Sarawak, Malaysia, 2004.

Lehner, B., Verdin, K., and Jarvis, A.: HydroSHEDS technical documentation. World Wildlife Funds US, Washington, D. C., 1st edn, http://hydrosheds.cr.usgs.gov. Accessed 17 June 2015, 2006.

Melling, L., Hatano, R., and Goh, K.J.: Methane Fluxes from three ecosystems in tropical peatland of Sarawak, Malaysia. Soil Biology & Biogeochemistry 37, 1445-1453. doi:10.1016/j.soilbio.2005.01.001, 2005.

Middelburg, J.J., Nieuwenhuize, J., Iversen, N., Hogh, N., de Wilde, H., Helder, W., Seifert, R., and Christof, O.: Methane distribution in European tidal estuaries. Biogeochemistry 59:95-119 , 2002.

Mosier, A., Kroeze, C., Nevison, C., Oenema, O., Seitzinger, S.P., and van Cleemput, O.: Closing the global N$_2$O budget: nitrous oxide emissions through the agricultural nitrogen cycle. Nutrient Cycling in Agroecosystems 52:225-248, 1998.

Müller, D., Warneke, T., Rixen, T., Müller, M., Mujahid, A., Bange, H.W., and Notholt, J.: Fate of ~~peat-derived~~terrestrial organic carbon and associated CO2 and CO emissions from two Southeast Asian estuaries. Biogeosciences~~-~~13, 691-705, doi:10.5194/bg-13-691-2016, 2016.~~Discussions 12:8299-8340, doi: 10.5194/bgd-12-8299-2015, 2015a.~~

Müller, D., Warneke, T., Rixen, T., Müller, M., Jamahari, S., Denis, N., Mujahid, A., Notholt, J. Lateral carbon fluxes and CO$_2$ outgassing from a tropical peat-draining river. Biogeosciences, 12, 5967-5979, doi:10.5194/bg-12-5967-2015, 2015~~b~~.

Murray, R.H., Erler, D.V., and Eyre, B.D.: Nitrous oxide fluxes in estuarine environments: response to global change. Global Change Biology doi: 10.1111/gcb.12923, 2015.

Musenze, R.S., Werner, U., Grinham, A., Udy, J., and Yuan, Z.: Methane and nitrous oxide emissions from a subtropical estuary (the Brisbane River estuary, Australia). Science of the Total Environment 472:719-729, doi: 10.1016/j.scitotenv.2013.11.085, 2014.

Myhre, G., Shindell, D., Bréon, F.M., Collins, W., Fuglestvedt, J., Huang, J., Koch, D., Lamarque, J.F., Lee, D., Mendoza, B., Nakajima, T., Robock, A., Stephens, G., Takemura, T., and Zhang, H. Anthropogenic and natural radiative forcing. In: Stocker, T., Qin, D., Plattner, G.K., Tignor, M., Allen, S., Boschung, J., Nauels, A., Xia, Y., Bex, V., and Midgley, P. (eds) Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, Cambridge University Press, Cambridge, United Kingdom and New York, 2013.

Naqvi, S.W.A., Jayakumar, D.A., Narvekar, P.V., Naik, H., Sarma, V.V.S.S., D'Souza, W., Joseph, S., and George, M.D.: Increased marine production of N$_2$O due to intensifying anoxia on the Indian continental shelf. Nature 408:346-349, 2000.

Quack, B. and Krüger, K.: Stratospheric ozone: Halogen Impacts in a Varying Atmosphere, GEOMAR- Report 12, ISSN: 2193-8113, doi:10.3289/GEOMAR_REP _NS_12_2013, 2013.

Rao, G.D. and Sarma, V.V.S.S.: Contribution of N$_2$O emissions to the atmosphere from Indian monsoonal estuaries. Tellus B 65:19,660, doi: 10.3402/tellusb.v65i0.19660, 2013.

Reeburgh, W.S. Oceanic methane biogeochemistry. Chemical Reviews, 107: 486-513, 2007.

Richey, J.E., Devol, A.H., Wofsy, S.C., Victoria, R., Riberio, M.N.G.: Biogenic gases and the oxidation and reduction of carbon in Amazon River and floodplain waters. Limnology and Oceanography 33(4):551-561, 1988.

Robinson, A. D., Nedwell, D. B., Harrison, R. M., and Ogilvie, B. G.: Hypernutrified estuaries as sources of N2O emission to the atmosphere: the estuary of the River Colne, Essex, UK. Marine Ecology Progress Series 164: 59-71, 1998.

Formatted: Left, Space Before: 0 pt, Line spacing: single, Don't adjust space between Latin and Asian text, Don't adjust space between Asian text and numbers

Formatted: English (U.S.)

SarVision: Impact of oil palm plantations on peatland conversion in Sarawak 2005-2010. Tech. rep., SarVision, Wageningen, 2011.

Seitzinger, S.P. and Kroeze, C.: Global distribution of nitrous oxide production and N inputs in freshwater and coastal marine ecosystems. Global Biogeochemical Cycles 12(1):93-113, 1998.

Shalini, A., Ramesh, R., Purvaja, R., and Barnes, J.: Spatial and temporal distribution of methane in an extensive shallow estuary, south India. J Earth Syst Sci 115(4):451-460, 2006.

Siedler, G., and Peters, H.: Properties of sea water. In: Sündermann J (ed) Oceanography, Landolt-Börnstein, vol 3a, Springer Verlag, New York, pp 233-264, 1986.

SPU State Planning Unit Sarawak Facts and Figures 2012. available at: http://www.spu.sarawak.gov.my/download_facts.html. Accessed 23 June 2015, 2012.

Teodoru, C.R., Nyoni, F.C., Borges, A.V., Darchambeau, F., Nyambe, I., and Bouillon, S.: Dynamics of greenhouse gases (CO2, CH4, N2O) along the Zambezi River and major tributaries, and their importance in the riverine carbon budget. Biogeosciences 12, 2431-2453. doi: 10.5194/bg-12-2431-2015, 2014.

Valentine, D.L.: Emerging topics in marine methane biogeochemistry. Annual Review of Marine Science, 3: 147-171, doi: 10.1146/annurev-marine-120709-142734, 2011.

Walter, S., Bange, H.W., Breitenbach, U., and Wallace, D.W.R.: Nitrous oxide in the north atlantic ocean. Biogeosciences 3:607-619, doi: 10.5194/bg-3-607-2006 , 2006.

Wanninkhof, R.: Relationship between wind speed and gas exchange over the ocean. Journal of Geophysical Research 97, No. C5:7373-7382, 1992.

Weiss, R.F., Price, B.A.: Nitrous oxide solubility in water and seawater. Marine Chemistry 8:347-359, 1980.

Wiesenburg, D.A., and Guinasso Jr, N.L.: Equilibrium solubilities of methane, carbon monoxide, and hydrogen in water and seawater. Journal of Chemical and Engineering Data 24(4):356-360, 1979.

Zhang, G.L., Zhang, J., Liu, S.M., Ren, J.L., Zhao, Y.C.: Nitrous oxide in the Changjiang (Yangtze River) estuary and its adjacent marine area: Riverine input, sediment release and atmospheric fluxes. Biogeosciences 7:3505-3516, doi: 10.5194/bg-7-3505-2010, 2010.

Zindler, C., Bracher, A., Marandino, C.A., Taylor, B., Torrecilla, E., Kock, A., and Bange, H.W.: Sulphur compounds, methane and phytoplankton: Interactions along a north-south transit in the western Pacific Ocean, Biogeosciences, 10: 3297–3311, doi: 10.5194/bg-10-3297-2013, 2013.

Tables

		DIN (μmol L^{-1})					N export (t yr^{-1})
		Min	**Max**	**Mean**	**Median**	**SD**	
Lupar	**dry**	20.9	30.8	23.6	22.2	3.6	4532
	wet	11.9	21.7	18.2	19.7	3.2	
Saribas	**dry**	25.5	37.6	31.0	30.0	6.1	1554
	wet	10.0	18.0	14.0	14.0	5.6	
Saribas tributary	**dry**	20.5	30.0	24.3	22.4	5.0	n.d.
	wet	10.9	18.2	13.0	11.4	3.5	

Table 1: Estuarine DIN concentration medians for the spatial extent that was covered in 2013 and the estimated inorganic N export to the South China Sea.

		Concentrations (nmol L^{-1})					Saturations (%)				
		Min	**Max**	**Mean**	**Median**	**SD**	**Min**	**Max**	**Mean**	**Median**	**SD**
Lupar	**dry**	7.2	9.3	7.7	7.6	0.4	106	116	108	108	1
	wet	~~6.3~~6.6	13.8	7.7	7.2	1.0	~~93~~102	208	116	109	15
Saribas	**dry**	7.3	8.4	7.9	7.9	0.2	110	118	114	113	2
	wet	8.2	19.3	9.0	8.7	1.2	118	592	134	126	46
Saribas tributary	**dry**	8.1	13.4	11.8	12.3	1.5	117	184	165	169	19
	wet	9.1	24.4	13.5	11.9	4.3	130	340	189	169	59
Coastal ocean		10.1	27.3	15.8	13.0	7.0	168	462	266	218	119

Table 2: N_2O concentrations and saturations in the Lupar, Saribas and the Saribas tributary and in the coastal ocean. Values for the estuaries are given for the spatial extent of the rivers that was covered in 2013.

		Concentrations (nmol L^{-1})					Saturations (%)				
		Min	**Max**	**Mean**	**Median**	**SD**	**Min**	**Max**	**Mean**	**Median**	**SD**
Lupar	**dry**	7.3	42.3	18.7	10.6	12.2	339	1910	837	486	520
	wet	~~2.0~~3.7	61.2	24.~~3~~4	13.5	20.3	~~88~~168	2799	~~1100~~1104	~~614~~615	915
Saribas	**dry**	8.3	58.8	28.2	25.6	15.5	397	2782	1331	1188	727
	wet	9.0	68.5	15.1	12.2	9.1	401	3050	671	545	395
Saribas tributary	**dry**	5.2	53.3	20.6	12.4	16.9	228	2458	942	554	784
	wet	23.3	113.9	63.2	64.0	27.4	1040	5058	2773	2786	1198
Coastal ocean		3.3	3.7	3.5	3.6	0.2	166	188	177	176	9

Table 3: CH_4 concentrations and saturations in the Lupar, Saribas and the Saribas tributary and in the coastal ocean. Values for the estuaries are given for the spatial extent of the rivers that was covered in 2013.

		k_{600} (cm h^{-1})	**Flux density (nmol m^{-2} s^{-1})**		**Annual average flux density (mmol m^{-2} yr^{-1})**		**Flux (t N yr^{-1} and t C yr^{-1})**	
			N_2O	**CH_4**	**N_2O**	**CH_4**	**N_2O**	**CH_4**
Lupar	**dry**	n.d.	0.04 ± 0.01	0.59 ± 0.14	1.3 ± 0.3	22.~~4~~ 5 ± 5.4	4 ± 1	~~59~~ 60 ± 17
	wet	20.5 ± 4.9	0.04 ± 0.01	0.~~83~~ 84 ± 0.20				
Saribas	**dry**	n.d.	0.04 ± 0.03	1.01 ± 0.84	1.9 ± 1.6	23.0 ± 19.1	3 ± 2	28 ± 25
	wet	13.2 ± 11.0	0.08 ± 0.07	0.45 ± 0.37				
Saribas tributary	**dry**	n.d.	0.39 ± 0.24	0.81 ± 0.50	12.0 ± 7.5	89.2 ± 55.2	n.d.	n.d.
	wet	23.9 ± 14.8	0.37 ± 0.23	4.84 ± 3.00				

Table 4: k_{600} values and median N_2O and CH_4 areal and total fluxes from the Lupar, Saribas and the Saribas tributary. The uncertainties relate to the maximum variability of the k_{600} value, as the k_{600} uncertainty propagation was responsible for approximately 96% of the flux uncertainty. Values are given for the spatial extent of the rivers that was covered in 2013.

~~k_{600} values and median N_2O and CH_4 areal and total fluxes from the Lupar, Saribas and the Saribas tributary. The uncertainties relates to the maximum variability of the k_{600} value. Values are given for the spatial extent of the rivers that was covered in 2013.~~

Formatted: Font: Not Italic

Formatted: Font: Not Italic, Subscript

Formatted: Font: Not Italic

Formatted: Font: Not Italic, Subscript

Formatted: Normal

Formatted: Font: Not Italic

Formatted: Font: Not Italic, Subscript

Formatted: Font: Not Italic

Formatted: Font: Not Italic

Formatted: Font: Not Italic

Formatted: English (U.S.)

Figures

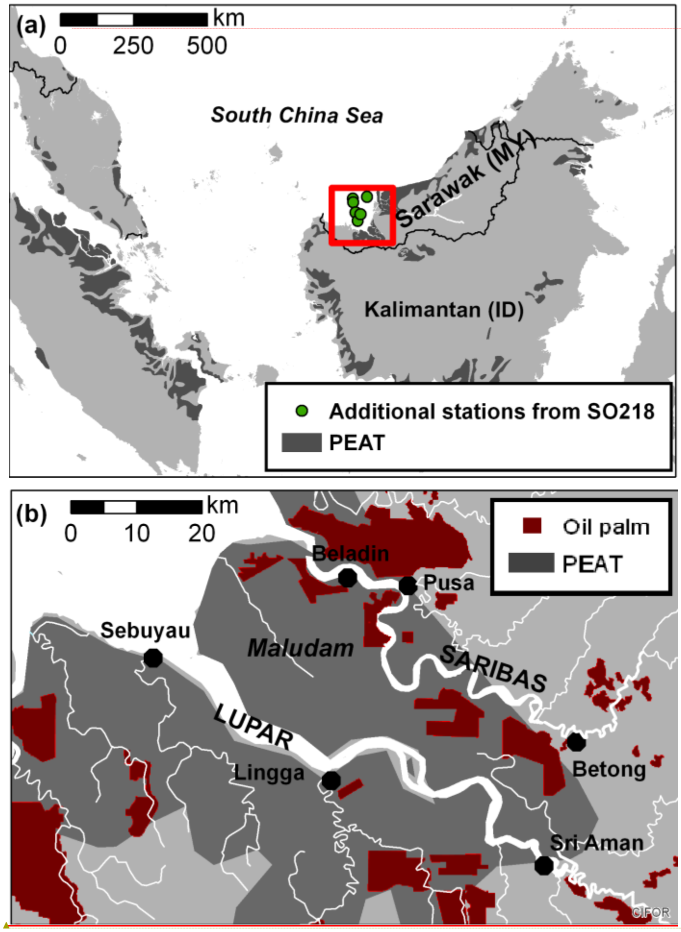

Figure 1: Map of the study area showing (a) the location of Sarawak on the island of Borneo and the cruise track of the R/V *Sonne* in November 2011 (SO218). (b) Close-up map of the Lupar and Saribas estuaries, enclosing the Maludam peninsula, showing the major settlements along the rivers and estuaries. Peat areas are indicated by the dark grey area, oil palm plantations as of CIFOR (2014) are shown in red.

Comment [d1]: Figure 1 was revised (scale bars were inserted and the South China Sea was labelled)

Formatted: Font: 10 pt, Font color: Auto

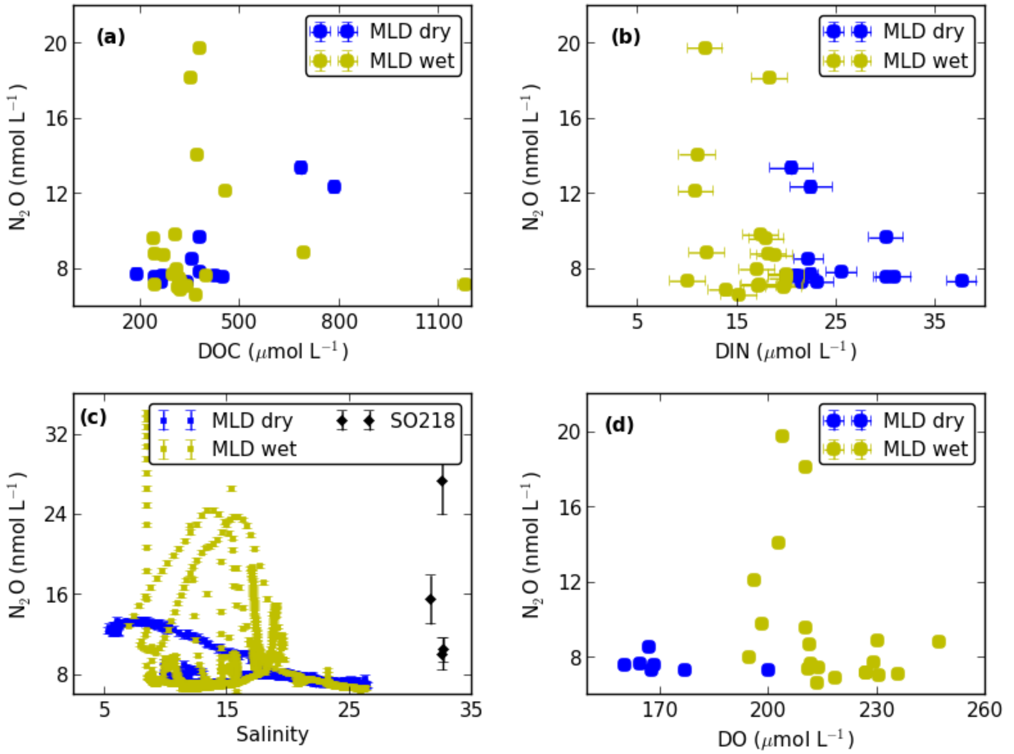

Figure 2: Relationship of nitrous oxide concentrations with (a) dissolved organic carbon (DOC), (b) dissolved inorganic nitrogen (DIN), (c) salinity and (d) dissolved oxygen (DO). MLD refers to the campaigns on the Lupar and Saribas estuaries in the dry and wet season, respectively, SO218 denotes data from the R/V *Sonne* cruise.

Formatted: Font: 10 pt, Font color: Auto

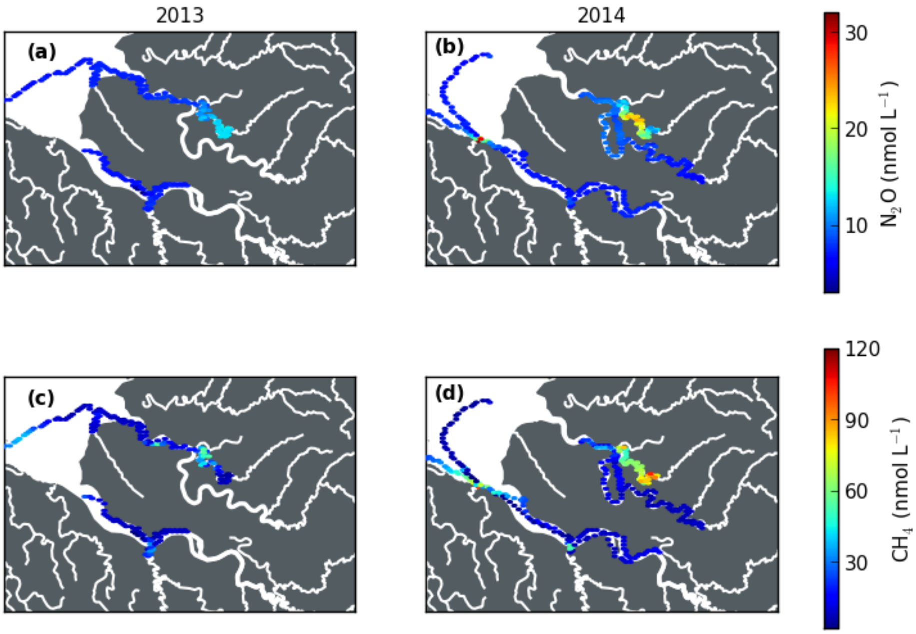

Figure 3: Dissolved N_2O (a+b) and CH_4 (c+d) concentrations measured during the 2013 (left) and 2014 (right) MLD campaigns.

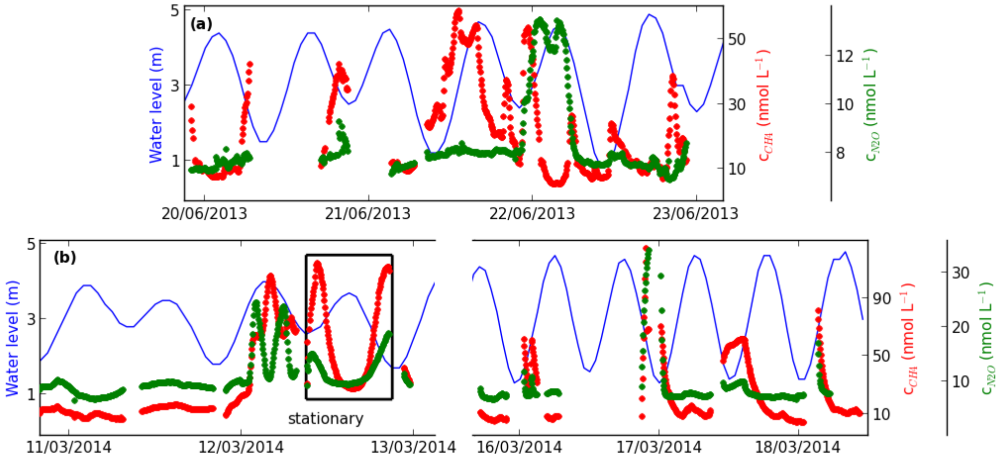

Figure 4: Time series of methane and nitrous oxide concentrations in the Lupar and Saribas estuaries measured during the dry season (2013) campaign (upper panel) and the wet season campaign (lower panel) and the water level as predicted for Pulau Lakei (+1 h for Lupar and Saribas). Spatial, temporal and tidal variations are overlapping in the Figure. One stationary measurement, as recorded on the Saribas R~~r~~iver in 2014, is denoted with the black box. Note the discontinuous time axis in the lower panel.

Formatted: Font: 10 pt, Font color: Auto

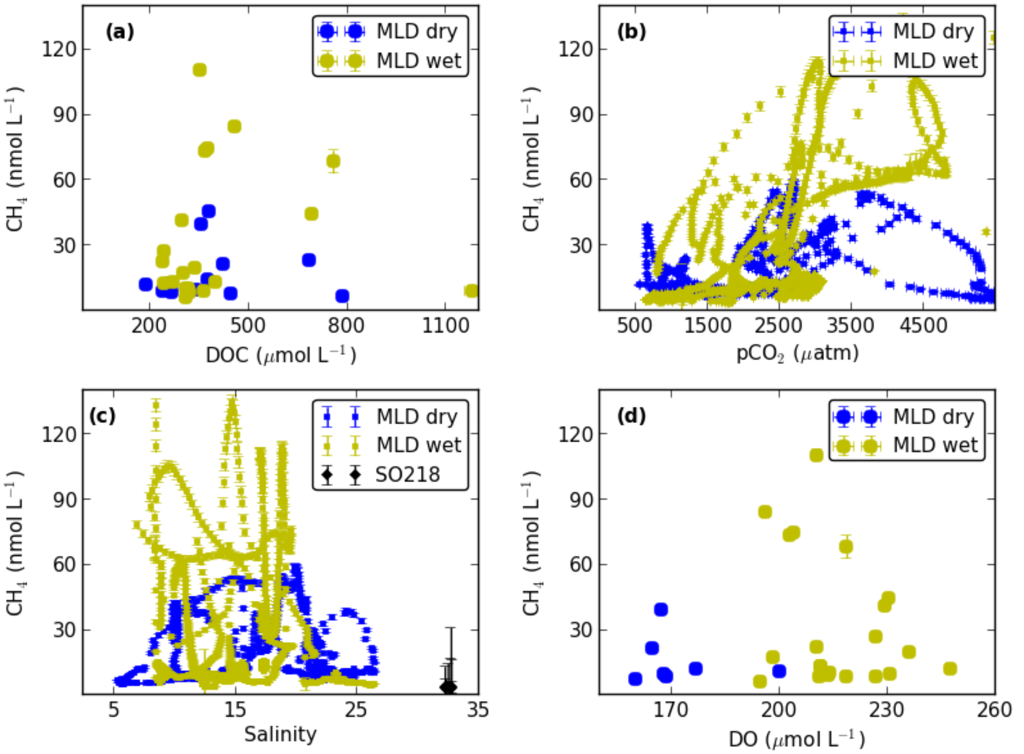

Figure 5: Relationship of methane concentrations with (a) dissolved organic carbon (DOC), (b) pCO$_2$, (c) salinity and (d) dissolved oxygen (DO). MLD refers to the campaigns on the Lupar and Saribas estuaries in the dry and wet season, respectively, SO218 denotes data from the R/V *Sonne* cruise.

Formatted: Font: 10 pt, Font color: Auto