Nitrous oxide (N\textsubscript{2}O) and methane (CH\textsubscript{4}) in rivers and estuaries of northwestern Borneo

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Abstract. Nitrous oxide (N\textsubscript{2}O) and methane (CH\textsubscript{4}) are atmospheric trace gases which play important roles in the climate and atmospheric chemistry of the Earth. However, little is known about their emissions from rivers and estuaries, which seem to contribute significantly to the atmospheric budget of both gases. To this end concentrations of N\textsubscript{2}O and CH\textsubscript{4} were measured in the Rajang, Maludam, Sebuyau and Simunjan rivers draining peatland in northwestern (NW) Borneo during two campaigns in March and September 2017. The Rajang River was additionally sampled in August 2016 and the Samunsam and Sematan rivers were additionally sampled in March 2017. The Maludam, Sebuyau, and Simunjan rivers are typical “blackwater” rivers with very low pH (3.7–7.8), very high dissolved organic carbon (DOC) concentrations (235–4387 mmol L\textsuperscript{-1}) and very low O\textsubscript{2} concentrations (31–246 µmol L\textsuperscript{-1}, i.e. 13 %–116 % O\textsubscript{2} saturation). The spatial and temporal variability of N\textsubscript{2}O and CH\textsubscript{4} concentrations (saturations) in the six rivers or estuaries was large and ranged from 2.0 nmol L\textsuperscript{-1} (28 %) to 41.4 nmol L\textsuperscript{-1} (570 %) and from 2.5 nmol L\textsuperscript{-1} (106 %) to 1372 nmol L\textsuperscript{-1} (57 459 %), respectively. We found no overall trends of N\textsubscript{2}O with O\textsubscript{2} or NO\textsubscript{3}\textsuperscript{-}, NO\textsubscript{2}\textsuperscript{-} or NH\textsubscript{4}\textsuperscript{+}, and there were no trends of CH\textsubscript{4} with O\textsubscript{2} or dissolved nutrients or DOC. N\textsubscript{2}O concentrations showed a positive linear correlation with rainfall. We conclude, therefore, that rainfall is the main factor determining the riverine N\textsubscript{2}O concentrations since N\textsubscript{2}O production or consumption in the blackwater rivers themselves seems to be low because of the low pH. CH\textsubscript{4} concentrations were highest at salinity = 0 and most probably result from methanogenesis as part of the decomposition of organic matter under anoxic conditions. CH\textsubscript{4} in the concentrations in the blackwater rivers showed an inverse relationship with rainfall. We suggest that CH\textsubscript{4} oxidation in combination with an enhanced river flow after the rainfall events might be responsible for the decrease in the CH\textsubscript{4} concentrations. The rivers and estuaries studied here were an overall net source of N\textsubscript{2}O and CH\textsubscript{4} to the atmosphere. The total annual N\textsubscript{2}O and CH\textsubscript{4} emissions were 1.09 Gg N\textsubscript{2}O yr\textsuperscript{-1} (0.7 Gg N yr\textsuperscript{-1}) and 23.8 Gg CH\textsubscript{4} yr\textsuperscript{-1}, respectively. This represents about 0.3 %–0.7 % of the global annual riverine and estuarine N\textsubscript{2}O emissions and about 0.1 %–1 % of the global riverine and estuarine CH\textsubscript{4} emissions. Therefore, we conclude that rivers and estuaries in NW Borneo – despite the fact their water area covers only 0.05 % of the global river/estuarine area – contribute significantly to global riverine and estuarine emissions of N\textsubscript{2}O and CH\textsubscript{4}.

1 Introduction

Nitrous oxide (N\textsubscript{2}O) and methane (CH\textsubscript{4}) are atmospheric trace gases which influence the climate and atmospheric chemistry of the Earth (IPCC, 2013; WMO, 2014). They act as greenhouse gases in the troposphere and are indirectly involved in stratospheric ozone depletion. Emission estimates indicate that rivers and estuaries contribute significantly to the atmospheric budget of both N\textsubscript{2}O and CH\textsubscript{4}. N\textsubscript{2}O emission estimates for rivers and estuaries range from 0.05
to 3.3 Tg N\textsubscript{2}O yr\textsuperscript{-1} and from 0.09 to 5.7 Tg N\textsubscript{2}O yr\textsuperscript{-1}, respectively (see overview in Maavara et al., 2019). Thus, the combined riverine and estuarine emissions may contribute up to 32% of the global natural and anthropogenic emissions of N\textsubscript{2}O (28.1 Tg N\textsubscript{2}O yr\textsuperscript{-1}; IPCC, 2013). CH\textsubscript{4} emission estimates for rivers and estuaries are in the range of 1.5–26.8 Tg CH\textsubscript{4} yr\textsuperscript{-1} (Bastviken et al., 2011; Stanley et al., 2016) and 0.8–6.6 Tg CH\textsubscript{4} yr\textsuperscript{-1} (see overview in Borges and Abril, 2011), respectively. The combined emissions from rivers and estuaries can contribute up to 6% of the global natural and anthropogenic atmospheric emissions of CH\textsubscript{4} (556 Tg CH\textsubscript{4} yr\textsuperscript{-1}; IPCC, 2013). As indicated by the wide range of the estimates cited above, the emission estimates of both gases are associated with a high degree of uncertainty, which is mainly caused by an inadequate coverage of the temporal and spatial distributions of N\textsubscript{2}O and CH\textsubscript{4} in rivers and estuaries and the inherent errors of the model approaches to estimate their exchange across the water–atmosphere interface (see, e.g., Alin et al., 2011; Borges and Abril, 2011).

N\textsubscript{2}O is produced by microbial processes such as nitrification (i.e. oxidation of ammonia, NH\textsubscript{3}, to nitrite, NO\textsubscript{2}\textsuperscript{-}) in estuarine waters (see, e.g., Barnes and Upstill-Goddard, 2011) and heterotrophic denitrification (i.e. reduction of nitrate, NO\textsubscript{3}\textsuperscript{-}, to dinitrogen, N\textsubscript{2}) in river sediments (Beaulieu et al., 2011). The yields of N\textsubscript{2}O from these processes are enhanced under low-oxygen (i.e. suboxic) conditions (see, e.g., Brase et al., 2017; Zhang et al., 2010), whereas N\textsubscript{2}O can be reduced to N\textsubscript{2} under anoxic conditions via sedimentary denitrification in rivers (see, e.g., Upstill-Goddard et al., 2017). Apart from ambient oxygen (O\textsubscript{2}) concentrations, riverine and estuarine N\textsubscript{2}O production is also dependent on the concentrations of dissolved inorganic nitrogen (DIN; = NH\textsubscript{4}\textsuperscript{+} + NO\textsubscript{2}\textsuperscript{-} + NO\textsubscript{3}\textsuperscript{-}) and organic carbon (Quick et al., 2019). There seems to be a general trend towards high estuarine/riverine N\textsubscript{2}O concentrations when DIN concentrations are high as well (Barnes and Upstill-Goddard, 2011; Quick et al., 2019; Zhang et al., 2010). However, this trend masks the fact that in many cases the spatial and temporal variability of riverine and estuarine N\textsubscript{2}O is often not related to DIN (see, e.g., Borges et al., 2015; Brase et al., 2017; Müller et al., 2016a; Quick et al., 2019).

CH\textsubscript{4} is produced during microbial respiration of organic matter by anaerobic methanogenesis in riverine and estuarine sediments (see, e.g., Borges and Abril, 2011; Romeijn et al., 2019; Stanley et al., 2016). A significant fraction of the CH\textsubscript{4} produced in sediments can be oxidized to carbon dioxide (CO\textsubscript{2}) via anaerobic CH\textsubscript{4} oxidation in sulfate-reducing zones of estuarine sediments (see, e.g., Maltby et al., 2018) and aerobic CH\textsubscript{4} oxidation in riverine sediments (see, e.g., Shelley et al., 2017). When released to the overlying riverine or estuarine water, CH\textsubscript{4} can be oxidized by aerobic CH\textsubscript{4} oxidation before reaching the atmosphere (see, e.g., Borges and Abril, 2011; Sawakuchi et al., 2016; Steinle et al., 2017).

In general, the temporal and spatial distributions of N\textsubscript{2}O and CH\textsubscript{4} in rivers and estuaries are driven by the complex interplay of microbial production and consumption pathways (see above) as well as physical processes such as input via shallow groundwater, river discharge, tidal pumping, release to the atmosphere and export to coastal waters (Barnes and Upstill-Goddard, 2011; Borges and Abril, 2011; Quick et al., 2019; Stanley et al., 2016).

Peatlands, which are found in the tropics and at high latitudes, constitute one of the largest reservoirs of organic-bound carbon worldwide (Minasny et al., 2019; Page et al., 2011; Treat et al., 2019; Yu et al., 2010). Rivers and streams draining peatlands have exceptionally high concentrations of dissolved organic carbon (DOC) and low pH and, thus, belong to the “blackwater” river type, which is also found in southeast (SE) Asia (see, e.g., Alkhatib et al., 2007; Martin et al., 2018; Moore et al., 2016).

Despite the fact that a number of studies about N\textsubscript{2}O and CH\textsubscript{4} emissions from peatlands in SE Asia have been published (see, e.g., Couwenberg et al., 2010; Hatano et al., 2016; Jauhiainen et al., 2012), only a few studies about their emissions from peatland-draining rivers in SE Asia have been published so far (Jauhiainen and Silvennoinen, 2012; Mù et al., 2016a). Therefore, our knowledge about the biogeochemistry and emissions of N\textsubscript{2}O and CH\textsubscript{4} from peatland-draining rivers is still rudimentary at best.

Here we present measurements of dissolved N\textsubscript{2}O and CH\textsubscript{4} in six rivers and estuaries in northwestern (NW) Borneo during August 2016, March 2017 and September 2017. The objectives of our study were (i) to measure the distributions of dissolved N\textsubscript{2}O and CH\textsubscript{4}, (ii) to identify the major factors influencing their distributions, and (iii) to estimate the N\textsubscript{2}O and CH\textsubscript{4} emissions to the atmosphere.

2 Study site description

Discrete samples of surface water were taken at several stations along the salinity gradients of the Rajang, Maludam, Sebuyau and Simunjan rivers in NW Borneo during two campaigns in March and September 2017 (Fig. 1, Table 1). The Rajang River was additionally sampled in August 2016, and the Samunsam and Sematan rivers were additionally sampled in March 2017. The environmental settings of the river basins are summarized in Table 2. Based on the areas affected by oil palm plantations and logging in combination with our own observations during several sampling campaigns, we classified the Rajang and Simunjan River basins as “disturbed” and the Maludam, Sebuyau, Sematan and Samunsam River basins as “undisturbed” (Table 2).

3 Methods

3.1 Measurements of N\textsubscript{2}O and CH\textsubscript{4}

Water was collected from 1 m depth by using a Niskin sampler. Subsamples for N\textsubscript{2}O and CH\textsubscript{4} were taken as duplicates or triplicates in 20 or 37 mL glass vials. The vials were
first rinsed with sample water, then filled to the maximum (without air bubbles), and finally sealed on the spot using a crimper. The samples were kept on ice for a maximum of 3 h. When returned to the field station, 50 µL of saturated aqueous mercuric chloride (HgCl₂) solution was immediately added to stop any biological activity, and samples were stored at 4 °C until shipment. The samples were shipped to GEOMAR Helmholtz Centre for Ocean Research Kiel, Germany, for further analysis within a few weeks after sampling. For the determination of the N₂O and CH₄ concentrations, we applied the static-headspace equilibration method followed by gas chromatographic separation and detection with an elec-

Figure 1. Map of the study area with locations of the sampling stations. Sampling stations from August 2016 are displayed in red circles, from March 2017 in blue triangles, and from September 2017 in green diamonds. Major cities are highlighted in bold plus symbols. Inset is adapted from Staub et al. (2000).

Table 1. Overview of sampling and sampled ranges of salinity, pH as well as O₂ concentration and saturation (in percent, given in parentheses) and concentrations of dissolved inorganic nitrogen (DIN = NO₃⁻ + NO₂⁻ + NH₄⁺), silicate (SiO₂) and dissolved organic carbon (DOC). All concentrations are given in µmol L⁻¹. NA stands for not available and “Stat.” stands for sampling station. DOC data were taken from Martin et al. (2018).

| River    | Date       | No. of Stat. | Range of Salinity (µmol L⁻¹) | pH (°C) | O₂ (µmol L⁻¹) | DIN (µmol L⁻¹) | SiO₂ (µmol L⁻¹) | DOC (µmol L⁻¹) |
|----------|------------|--------------|------------------------------|--------|--------------|---------------|----------------|----------------|
| Rajang   | 20–27 Aug 2016 | 30           | 0–32                         | 6.5–8.1| 85–153       | 6.7–29        | 4.0–179        | NA             |
|          | 4–7 Mar 2017  | 14           | 0–30                         | 6.0–8.2| 142–237      | 8.1–18        | 16–158         | 96–201         |
|          | 5–14 Sep 2017 | 8            | 0–18                         | 6.9–8.2| 164–227      | 6.7–14        | 12–98          | NA             |
| Maludam  | 9 Mar 2017    | 9            | 0–20                         | 3.7–7.6| 34–213       | 3.9–10        | 5.8–32         | 266–4387       |
|          | 14–15 Sep 2017| 9            | 0–15                         | 4.1–6.7| 43–155       | 2.1–3.0       | 0.1–8.0        | 3072–3245      |
| Sebuyau  | 11 Mar 2017   | 11           | 0–24                         | 4.3–7.8| 43–246       | 2.9–13        | 33–78          | 206–1968       |
|          | 15 Sep 2017   | 5            | 0–10                         | 7.2–7.7| 65–179       | 1.1–13        | 0.9–44         | 235–2052       |
| Simunjan | 12 Mar 2017   | 6            | 0–0.4                        | 4.7–6.3| 31–81        | 2.2–16        | 73–114         | 2016–3039      |
|          | 17 Sep 2017   | 6            | 0–4.6                        | 4.7–6.7| 95–131       | 2.0–13        | 1.4–2.6        | 925–1960       |
| Sematan  | 9 Mar 2017    | 5            | 0–28                         | 6.8–8.3| 184–208      | 5.9–10        | 6.3–141        | 100–240        |
| Samunsam | 11 Mar 2017   | 5            | 0–27                         | 6.3–8.2| 174–208      | 3.9–6.6       | 9.7–98         | 87–1188        |
Table 2. Summary of the environmental settings of the river basins. Based on the area percentage of oil palm, logging, and our own surveys and observations, we classified the river basins as undisturbed (U) and disturbed (D). All areas are given in kilometres.

| River       | Total basin | Peatland$^a$ | Oil palm plantations$^b$ | Logging$^c$ | River water surface$^d$ | Remarks                                                                 | Classification |
|-------------|-------------|--------------|--------------------------|-------------|-------------------------|--------------------------------------------------------------------------|----------------|
| Rajang      | 50 000$^e$  | 3844         | 4514                     | 29 379      | 455$^e$                 | The longest river in Malaysia. The major town is Sibu (163 000 population). Smaller townships are Kapit, Kanowit and Sarieki. There is a large number of villages and longhouses (traditional buildings inhabited by local communities) located along the river and its tributaries. Two hydroelectric power plants were built at two tributaries in the upper Rajang Basin. The river mouth is surrounded by peat lands, and most of these peat lands have been converted to commercial oil palm plantations. | D              |
| Maludam     | 197         | 172          | 16                       | 0           | 0.36                    | The upstream part of the river is surrounded by the Maludam National Park. The Maludam Peninsula is bordered by the Lupar and Saribas rivers and is the biggest undisturbed peat forest in Malaysia. The National Park had been subjected to selective logging before it was gazetted as a totally protected area in 2000. Well-preserved peat land. There are oil palm cultivations near the few villages. | U              |
| Sebuyau     | 538         | 288          | 24                       | 0           | 2.11                    | Major town is Sebuyau (14 000 population), surrounded by a few villages. Other agricultural activities were observed. | U              |
| Simunjan    | 788         | 346          | 240                      | 0           | 4.73                    | Major town is Simunjan (22 000 population), a few villages. Two streams combine to form the main Simunjan River. One of the streams passes an oil palm mill which discharges into the river. | D              |
| Sematan     | 287         | 0            | 0                        | 0           | 1.47                    | Major town is Sematan (7600 population); small villages. We observed agricultural activities by the local people. | U              |
| Samunsam    | 163         | 0            | 0                        | 0           | 0.85                    | Well-preserved tropical forest. Some peat in the upper catchment area. | U              |

$^a$ Estimate is based on “Wetlands International”. “Malaysia peat lands”. Accessed through Global Forest Watch on 22 November 2018 (https://www.globalforestwatch.org).
$^b$ Estimate is based on “Oil palm concessions”. Accessed through Global Forest Watch on 22 November 2018 (https://www.globalforestwatch.org).
$^c$ Estimate is based on “Managed forest concessions”. Accessed through Global Forest Watch on 22 November 2018 (https://www.globalforestwatch.org).
$^d$ Estimate is based on “Managed forest concessions”. Accessed through Global Forest Watch on 22 November 2018 (https://www.globalforestwatch.org).
$^e$ Area estimates are based on the length and width of the primary course and main tributaries of the rivers. Length and width of the rivers were estimated using Google Earth (multiple readings).
$^f$ Estimate from Staub et al. (2000).

Electron capture detector (ECD; for N$_2$O) and a flame ionization detector (FID; for CH$_4$) as described in Bastian (2017) and Kallert (2017). Calibration of the ECD and FID was performed with standard gas mixtures of 348.4–1476.1 ppb N$_2$O and 1806.10–3003.79 ppb CH$_4$ in synthetic air which have been calibrated against NOAA-certified primary gas standards in the laboratory of the Max Planck Institute for Biogeochemistry in Jena, Germany.
Dissolved N$_2$O/CH$_4$ concentrations ($C_{\text{obs}}$ in nmol$L^{-1}$) were calculated with

$$C_{\text{obs}} = x' PV_{hs}/(RTV_{wp}) + x'\beta P,$$

where $x'$ is the dry mole fraction of N$_2$O or CH$_4$ in the headspace of the sample, $P$ is the ambient pressure (set to 1013.25 hPa), and $V_{hs}$ and $V_{wp}$ are the volumes of the headspace and the water phase, respectively. $R$ stands for the gas constant ($8.31451 \text{ m}^3 \text{ Pa} \text{ K}^{-1} \text{ mol}^{-1}$), $T$ is the temperature during equilibration, and $\beta$ is the solubility of N$_2$O or CH$_4$ (Weiss and Price, 1980; Wiesenburg and Guinasso Jr., 1979). The estimated mean relative errors of the measurements were $\pm 9\%$ and $\pm 13\%$ for N$_2$O and CH$_4$, respectively. These comparably high relative errors most probably resulted from the long storage time ($6$–$7$ months after sampling) for some of the samples. The higher mean measurement error of the CH$_4$ samples (compared to the N$_2$O measurements) was attributed to the fact that CH$_4$ samples are more sensitive to storage time than N$_2$O samples (Wilson et al., 2018).

### 3.2 Ancillary measurements

Water temperature, dissolved oxygen and salinity were recorded with an Aquaread® 2000. Nutrient measurements are described in detail in Sia et al. (2019). In short, all samples were collected within the upper $1$ m (surface) using pre-washed bottles via a pole sampler to reduce contamination from the surface of the boat and engine coolant waters (Zhang et al., 2015). Samples were filtered through a 0.4 µm pore-size polycarbonate membrane filter (Whatman) into pre-rinsed bottles, conserved with concentrated HgCl$_2$ solution and kept in a cool, dark room. Nutrients were determined utilizing a Skalar SANplus auto analyser with an analytical precision $< 5\%$. pH was measured using a YSI Aquaread® multiple-parameter probe (AP-2000). The measurements of DOC are described in detail in Martin et al. (2018). The performance of the DOC measurements was monitored by using deep-sea water samples with a certified DOC concentration of $42$–$45 \mu$mol L$^{-1}$ provided by the Hansell Laboratory, University of Miami. Our analyses consistently yielded slightly higher concentration for the reference water, with a long-term mean (±1 SD) of $47 \pm 2.0 \mu$mol L$^{-1}$ ($n = 51$). The DOC data are available from the Supplement in Martin et al. (2018).

### 3.3 Computations of saturations and flux densities

The saturations (Sat, %) for N$_2$O, CH$_4$ and O$_2$ were calculated as

$$\text{Sat} = 100\% \frac{C_{\text{obs}}}{C_{\text{eq}}},$$

where $C_{\text{eq}}$ is the equilibrium concentration of N$_2$O/CH$_4$ or O$_2$ calculated according to Weiss and Price (1980), Wiesenburg and Guinasso Jr. (1979), or Weiss (1970), respectively, with the in situ temperature and salinity as well as the mean dry mole fractions of N$_2$O/CH$_4$ at the time of the sampling. Mean monthly N$_2$O/CH$_4$ dry mole fractions of $329/1841 \times 10^{-9}$ (ppb), $331/1880$ and $330/1852$ ppb for August 2016, March 2017 and September 2017, respectively, were measured at the atmospheric monitoring station Bukit Kototabang, located on the west coast of Sumatra (Indonesia). This station is operated by the NOAA/ESRL Global Monitoring Division program and data are available from http://www.esrl.noaa.gov/gmd (last access: 4 November 2019). A saturation $< 100\%$ indicates a concentration lower than the theoretical equilibrium concentration (i.e. undersaturation), and a saturation $> 100\%$ indicates supersaturation.

Flux densities ($F$, nmol m$^{-2}$ s$^{-1}$) were calculated as

$$F = k_w(C_{\text{obs}} - C_{\text{eq}}),$$

$$k_w = k_{600}(Sc/600)^{-0.5}.$$

$k_w$ is the gas transfer velocity and $Sc$ is the Schmidt number, which was calculated with the equations for the kinematic viscosity of water (Siedler and Peters, 1986) and the diffusion of N$_2$O or CH$_4$ in water (Jähne et al., 1987; Rhee et al., 2009). $k_{600}$ was determined in a study for the Lupar and Saribas rivers which are located in close vicinity to the Maludam River (Müller et al., 2016a, b). Both rivers have similar environmental and morphological settings in comparison to the rivers studied here. Therefore, we assume that the $k_{600}$ values measured by Müller et al. (2016a) are representative of the rivers in NW Borneo studied here. Mean $k_{600}$ ranges from $13.2 \pm 11$ to $23.9 \pm 14.8$ cm h$^{-1}$. On the basis of the data in Müller et al. (2016a), we computed a mean $k_{600}$ of $19.2$ cm h$^{-1}$ ($5.33 \times 10^{-5}$ m s$^{-1}$), which we used to estimate the flux densities of N$_2$O and CH$_4$. This $k_{600}$ is in good agreement with the mean $k_{600}$ for rivers $< 100$ m wide ($22.4 \pm 14.3$ cm h$^{-1}$) and estuaries/rivers $> 100$ m wide ($10.3 \pm 7.7$ cm h$^{-1}$) listed in Alin et al. (2011), which range from $6.0$ to $35.3$ and $4.8$ to $30.6$ cm h$^{-1}$, respectively. $k_w$ in rivers depends on the turbulence at the river is water–atmosphere interface, which in turn is mainly affected by water current velocity, water depth and riverbed roughness and to a lesser extent by the wind speed (Alin et al., 2011; Borges and Abril, 2011). Since the $k_{600}$ reported by Müller et al. (2016a) was determined only during the wet season (March 2014), our mean $k_{600}$ is biased because it does not account for a lower $k_{600}$, which is to be expected during the dry season (resulting from a lower water current velocity; Alin et al., 2011). This results in an overestimation of the flux densities.

### 3.4 Rainfall data

In order to account for the regional variability of the rainfall in NW Borneo, we used rainfall data with a 3 h resolution recorded at the weather stations in Kuching, Bandar Sri Aman and Sibu (all in NW Borneo). The rainfall
data were provided by World Weather Online (Dubai, UAE, and Manchester, UK) and are available via https://www.worldweatheronline.com/ (last access: 4 November 2019). Representative weather stations were chosen for each river basin studied here and allocated as follows. The rainfall data for the Simunjan, Sematan and Samunsam River basins are represented by the data from Kuching; the Maludam–Sebuyau and the Rajang River basins are represented by the data from the Bandar Sri Aman and Sibu weather stations, respectively. We also included the N\textsubscript{2}O and CH\textsubscript{4} concentration data from two measurement campaigns to the Lupar and Saribas rivers in June 2013 and March 2014 (Müller et al., 2016a). The Lupar and Saribas data were associated with the rainfall data from the weather station in Bandar Sri Aman. Accumulated rainfall amount was computed by summing up the 3 h rainfall data for the periods of 1–4 weeks prior to the sampling dates.

4 Results and discussion

All rivers showed low concentrations of DIN in the range of 1.1 to 29 µmol L\textsuperscript{−1} (Table 1). NO\textsubscript{3}\textsuperscript{−} concentrations ranged from below the detection limit of 0.14 up to 19 µmol L\textsuperscript{−1}, and NH\textsubscript{4}\textsuperscript{+} concentrations were in the range of 0.3 to 17 µmol L\textsuperscript{−1}. The Maludam, Sebuyau and Simunjan rivers can be classified as blackwater rivers with low pH (3.7–4.8), high DOC concentrations (1960–4387 µmol L\textsuperscript{−1}) and low O\textsubscript{2} concentrations (31–95 µmol L\textsuperscript{−1}; 13 %–39 % saturation) at salinity = 0 (Table 1). Comparable settings have been reported from other tropical blackwater rivers in SE Asia as well (Alkhatib et al., 2007; Baum et al., 2007; Moore et al., 2011; Rixen et al., 2008; Wit et al., 2015).

4.1 Nitrous oxide

The measured ranges of N\textsubscript{2}O concentrations and saturations are listed in Table 3 and the distributions of N\textsubscript{2}O saturations along the salinity gradients are shown in Fig. 2. N\textsubscript{2}O concentrations (saturations) were highly variable and ranged from 2.0 nmol L\textsuperscript{−1} (28 %) in the Rajang River (at salinity = 0 in August 2016) to 41.4 nmol L\textsuperscript{−1} (570 %) in the Simunjan River (at salinity = 0 in March 2017). N\textsubscript{2}O concentrations in the Rajang, Maludam and Sebuyau rivers were generally higher in September compared to March 2017 (Fig. 2a–c). A decreasing linear trend of the N\textsubscript{2}O saturations with salinity was only observed for the Rajang River in March 2017 (Fig. 2a) indicating a conservative mixing and no N\textsubscript{2}O sources or sinks along the salinity gradient. Our results are in general agreement with the N\textsubscript{2}O measurements in the Lupar and Saribas rivers (which are located in close vicinity of the Maludam River) in June 2013 and March 2014: Müller et al. (2016a) measured N\textsubscript{2}O concentrations (saturations) from 6.6 to 117 nmol L\textsuperscript{−1} (102 % to 1679 %) in the Lupar and Saribas rivers. Salinity and N\textsubscript{2}O concentrations in the Lupar and Saribas rivers were negatively correlated in June 2013 but were not correlated in March 2014 (Müller et al., 2016a).

In contrast with our study, no N\textsubscript{2}O undersaturations have been observed by Müller et al. (2016a). Our results are at the lower end of N\textsubscript{2}O concentrations reported from rivers around the globe, which can range from extreme undersaturation (down to about 3 %, i.e. almost devoid of N\textsubscript{2}O) as measured in a tropical river in Africa (Borges et al., 2015) to extreme supersaturation (up to 12 500 %) as measured in an agriculture-dominated river in Europe (Borges et al., 2018).

Maximum N\textsubscript{2}O saturations measured in March 2017 were in the range of 106 % to 142 % for the rivers classified as undisturbed (Maludam, Sebuyau, Sematan and Samunsam), whereas the maximum saturation for the rivers classified as disturbed (Rajang and Simunjan) was in the range of 329 % to 570 % (Tables 2 and 3) indicating higher emissions from the disturbed rivers. The maximum N\textsubscript{2}O saturations in September 2017 ranged from 329 % to 390 %, and no differences were observed between undisturbed and disturbed rivers (Table 3).

We found no overall trends of N\textsubscript{2}O with O\textsubscript{2} or NO\textsubscript{3}\textsuperscript{−}, NO\textsubscript{2}\textsuperscript{−}, NH\textsubscript{4}\textsuperscript{+} and DIN. Therefore, it is difficult to decipher the major consumption or production processes of N\textsubscript{2}O or to locate the influence of (local) anthropogenic input of nitrogen compounds on riverine N\textsubscript{2}O cycling. This is in line with results from studies of other tropical rivers (Borges et al., 2015; Müller et al., 2016a). There are, however, occasional observations of N\textsubscript{2}O correlations with O\textsubscript{2} or nutrients in tropical rivers which were attributed to river types such as swamp and savannah rivers (Upstill-Goddard et al., 2017). Figure 3 shows the N\textsubscript{2}O concentrations along the pH gradients. Obviously there are no trends except for an enhancement of the N\textsubscript{2}O concentrations in September 2017. N\textsubscript{2}O production via nitrification depends on the prevailing pH because nitrifiers prefer to take up ammonia (NH\textsubscript{3}). The concentration of dissolved NH\textsubscript{3} drops significantly at pH < 8–9 (Bange, 2008) because of its easy protonation to ammonium (NH\textsubscript{4}\textsuperscript{+}). A low pH of about 5–6 can reduce nitrification (NH\textsubscript{4}\textsuperscript{+} oxidation) significantly as was recently shown for the Tay Ninh River in Vietnam (Le et al., 2019). Moreover, the optimum for a net N\textsubscript{2}O production by nitrification, nitrifier denitrification and denitrification lies between a pH of 7 and 7.5 (Blum et al., 2018). Therefore, a net N\textsubscript{2}O production may be low in the blackwater rivers studied here because of their low pH (see Table 1). The observed N\textsubscript{2}O supersaturations, therefore, might have been mainly the result of external inputs of N\textsubscript{2}O-enriched waters or groundwater. The observed N\textsubscript{2}O undersaturations were most probably resulting from heterotrophic denitrification which could have taken place in organic matter-enriched anoxic river sediments or in anoxic environments of the surrounding soils. However, the main factor for riverine N\textsubscript{2}O under- or supersaturation might be rainfall because rainfall events determine the height of the water table in the surrounding soils, which, in turn, determines the
amount of suboxic–anoxic conditions favourable for N\textsubscript{2}O production or consumption (Jauhiainen et al., 2016). See also discussion in Sect. 4.3.

### 4.2 Methane

The measured ranges of CH\textsubscript{4} concentrations and saturations are listed in Table 3, and the distributions of CH\textsubscript{4} saturations along the salinity gradients are shown in Fig. 4. CH\textsubscript{4} concentrations (saturations) were highly variable and ranged from 2.5 nmol L\textsuperscript{-1} (106 %) in the Simunjan River (at salinity = 0 in September 2017) to 1372 nmol L\textsuperscript{-1} (57 459 %) in the Simunjan River (at salinity = 0 in March 2017).

(Delete note that we also measured a CH\textsubscript{4} concentration of 14 999 nmol L\textsuperscript{-1} (624 070 %) at one station in the Simunjan River at salinity = 0 in March 2017, which, however,)

Table 3. Overview of N\textsubscript{2}O and CH\textsubscript{4} concentrations, saturations and flux densities in rivers and estuaries of NW Borneo.

| River    | Date   | N\textsubscript{2}O | CH\textsubscript{4} |
|----------|--------|----------------------|----------------------|
|          |        | Concentration nmol L\textsuperscript{-1} | Saturation % | Flux density nmol m\textsuperscript{-2} s\textsuperscript{-1} | Concentration nmol L\textsuperscript{-1} | Saturation % | Flux density nmol m\textsuperscript{-2} s\textsuperscript{-1} |
| Rajang   | Aug 2016 | 2.0 to 14.1 | 28 to 215 | −0.33 to 0.48 | 13.2 to 233 | 719 to 9988 | 0.77 to 15 |
|          | Mar 2017 | 5.9 to 24.0 | 100 to 329 | 0 to 1.08 | 11.1 to 1008 | 455 to 40 598 | 0.34 to 62 |
|          | Sep 2017 | 18.6 to 24.6 | 277 to 390 | 0.76 to 1.22 | 7.4 to 150 | 350 to 6019 | 0.35 to 9.05 |
| Maludam  | Mar 2017 | 4.5 to 6.7 | 62 to 106 | −0.20 to 0.03 | 312 to 829 | 12 603 to 32 988 | 19 to 50 |
|          | Sep 2017 | 10.8 to 20.7 | 150 to 331 | 0.23 to 1.00 | 3.3 to 18 | 163 to 717 | 0.09 to 0.93 |
| Sebuyau  | Mar 2017 | 3.5 to 7.7 | 55 to 118 | −0.18 to 0.08 | 8.4 to 1228 | 396 to 50 774 | 0.41 to 78 |
|          | Sep 2017 | 12.8 to 23.0 | 176 to 335 | 0.36 to 1.08 | 6.4 to 29 | 299 to 1285 | 0.28 to 1.79 |
| Simunjan | Mar 2017 | 2.5 to 41.4 | 35 to 570 | −0.31 to 2.20 | 39 to 1372 (14 999)\footnote{This extreme value was not included in further computations.} | 1642 to 57 459 (624 070)\footnote{This extreme value was not included in further computations.} | 2.37 to 88 |
|          | Sep 2017 | 5.1 to 26.5 | 73 to 365 | −0.13 to 1.24 | 2.5 to 21 | 106 to 878 | 0.01 to 1.18 |
| Sematan  | Mar 2017 | 4.3 to 8.2 | 71 to 109 | −0.11 to 0.04 | 8.6 to 12 | 433 to 47 055 | 0.43 to 72 |
| Samunsam | Mar 2017 | 4.0 to 9.5 | 67 to 142 | −0.13 to 0.19 | 16.5 to 978 | 830 to 43 807 | 0.95 to 63 |

* This extreme value was not included in further computations.
Figure 3. Concentrations of N$_2$O (a) and CH$_4$ (b) from rivers or estuaries along the pH gradients. The open red squares depict data from August 2016, the filled red squares depict data from March 2017 and the filled blue triangles depict data from September 2017. The vertical bars in (a) and (b) roughly indicate salinity = 0. Concentrations to the left of the vertical bar are at salinity = 0 and concentrations to the right of the vertical bars are at salinity > 0. The horizontal bar in (a) indicates the equilibrium concentration of N$_2$O. Please note that in August 2016, only the Rajang River was sampled.

was not included in Fig. 4 and which was excluded in the emission estimates for statistical reasons.) CH$_4$ saturations in the Rajang, Maludam, Sebuyau and Simunjan rivers were higher in March 2017 compared to September 2017. Maximum CH$_4$ concentrations were measured at salinity = 0, and there was a general decrease in CH$_4$ concentrations with increasing salinity. Exceptions from this trend occurred at individual stations in the Maludam, Sebuyau and Samunsam rivers which point to local sources of CH$_4$ (Fig. 3). The range of CH$_4$ concentrations (saturations) from our study is larger compared to the concentration range measured in the Lu-par and Saribas rivers (3.7–113.9 nmol L$^{-1}$; 168 %–5058 %) (Müller et al., 2016a). Borges et al. (2015) reported a maximum CH$_4$ concentration (saturation) of 62 966 nmol L$^{-1}$ (approx. 954 000 %) in their study of tropical rivers in Africa, which is much higher than the maximum concentration measured in our study. We found no differences in the CH$_4$ saturations between the rivers classified as undisturbed and those classified as disturbed in both March and September 2017.

We found no overall trends of CH$_4$ with O$_2$ or dissolved nutrients or DOC along the salinity gradients. There are, however, occasional observations in tropical rivers of CH$_4$ relationships with O$_2$, which were attributed to different river types such as swamp and savannah rivers (Upstill-Goddard et al., 2017). High CH$_4$ concentrations, which were often associated with high DOC and low O$_2$ concentrations at salinity = 0 and pH < 7 (see Fig. 3b), might have been produced by methanogenesis in anoxic riverine sediments rich in organic material or in anoxic parts of the surrounding soils drained by the rivers. The decrease in CH$_4$ with increasing salinity can be attributed to the gas exchange across the river water–atmosphere interface in combination with CH$_4$ oxidation (Borges and Abril, 2011; Sawakuchi et al., 2016).

4.3 N$_2$O/CH$_4$ concentrations and rainfall

Mean N$_2$O concentrations showed linear correlations with accumulated rainfall during different periods from 1–4 weeks before the dates of sampling (Fig. 5, Table 6). Enhanced N$_2$O emissions from (peat) soils are usually associated with rainfall when the water table approaches the soil surface (Couwenberg et al., 2010; Jauhiainen et al., 2016). A high water table, in turn, allows decomposition of previously deposited fresh organic material (Jauhiainen et al., 2016) and, thus, will result in favourable conditions for microbial N$_2$O production mainly via denitrification in a suboxic–anoxic soil environment (Espenberg et al., 2018; Pihlatie et al., 2004). N$_2$O production via nitrification may be less important at a high water table (Pihlatie et al., 2004; Regina et al., 1996). Therefore, the positive linear relationship of the riverine N$_2$O concentrations with rainfall might result from
enhanced N₂O production in the adjacent soils drained by the rivers. A decreasing trend of N₂O concentrations, which would be expected to be caused by enhanced river discharge after the rain events – which in turn can lead to dilution of the concentrations and enhanced fluxes across the river–atmosphere interface (Alin et al., 2011) – is obviously out-competed by an enhanced input of N₂O.

In contrast with N₂O, the response of riverine or estuarine CH₄ concentrations to increasing rainfall does not result in increasing CH₄ concentrations (Fig. 5). When considering the periods of 1 or 1.5 weeks of accumulated rainfall there seems to be a pronounced decrease in CH₄ concentrations with increasing rainfall (Fig. 5c and Table 6). This trend is no longer significant when considering the periods of 2–4 weeks of accumulated rainfall (Table 6). A closer inspection of the data reveals that the response to increasing rainfall seems to be different for individual rivers or estuaries. There is a clear negative relationship with rainfall for the Maludam, Sebuyau and Simunjan rivers, whereas no obvious trends were observed for the other rivers (Fig. 5c and d). Under the assumption that rainfall is a predictor for river discharge/high water we can argue that our results are in agreement with the often observed inverse relationship between CH₄ concentrations and river discharge (Anthony et al., 2012; Bouillon et al., 2014; Dinsmore et al., 2013; Hope et al., 2001). This relationship can be explained by an interplay of various processes such as (i) a decrease in CH₄ concentrations caused by a higher water flow (i.e. dilution under the assumption that the net CH₄ production does not change significantly), (ii) higher flux across the river–atmosphere interface during periods of higher discharge (caused by an enlarged river surface area and/or a more turbulent water flow) (Alin et al., 2011) and (iii) the enhancement of CH₄ oxidation during high waters: Sawakuchi et al. (2016) showed that CH₄ oxidation in blackwater rivers of the Amazon Basin was maximal during the high-water season.

4.4 Emission estimates

The N₂O flux densities from the six rivers studied here are comparable to the N₂O flux densities from other aqueous and soil systems reported from Borneo and other sites in SE Asia; see Table 4. The corresponding CH₄ flux densities are higher than the CH₄ flux densities reported for the Lupar and Saribas rivers but much lower than the flux densities from drainage canals in Central Kalimantan and Sumatra (Jauhiainen and Silvennoinen, 2012) (Table 4). Our CH₄ flux densities are, however, comparable to recently published CH₄ eddy covariance measurements (Tang et al., 2018) in the Maludam National Park, which is drained by the Maludam River, and measurements of the CH₄ release from peat
soils when the water table is high and CH$_4$ from rice paddies (Couwenberg et al., 2010); see Table 4. The mean annual N$_2$O and CH$_4$ emissions for the individual rivers were calculated by multiplying the mean flux density, $F$, for each river (Table 4) with the river surface area given in Table 2. The results are listed in Table 5. The resulting total annual N$_2$O emissions for the rivers in NW Borneo — including the emissions from the Lupar and Saribas rivers (Müller et al., 2016a) — are 1.09 Gg N$_2$O yr$^{-1}$ (0.7 Gg N yr$^{-1}$). This represents about 0.3–0.7 % of the global annual riverine and estuarine N$_2$O emissions of 166–322 Gg N$_2$O (106–205 Gg N yr$^{-1}$) recently estimated by Maavara et al. (2019). The total annual CH$_4$ emissions from rivers in NW Borneo are 23.8 Gg CH$_4$ yr$^{-1}$. This represents about 0.1 %–1 % of the global riverine and estuarine CH$_4$ emissions of 2300–33 400 Gg CH$_4$ yr$^{-1}$ (the emission range is based on the minimum and maximum estimates given in Bange et al., 1994; Bastviken et al., 2011; Borges and Abril, 2011; and Stanley et al., 2016). However, we caution that our estimates are associated with a high degree of uncertainty because (i) our data are biased by the fact that for some rivers it was not possible to cover the entire salinity gradient, (ii) seasonal and interannual variabilities of the N$_2$O and CH$_4$ concentrations are not adequately represented in our data set, (iii) the wind-speed-driven gas exchange in estuaries is not adequately represented, and (iv) the mean $k_{600}$ used here is most probably too high (see Sect. 3.3), resulting in an overestimation of the emissions.

5 Summary and conclusions

N$_2$O and CH$_4$ were measured in the Rajang, Maludam, Seb yaw and Simuntan rivers and estuaries in NW Borneo during two campaigns in March and September 2017. The Rajang River was additionally sampled in August 2016, and the Samunsam and Sematan rivers were additionally sampled in March 2017. The spatial and temporal variability of N$_2$O and CH$_4$ concentrations was large. N$_2$O concentrations (saturations) ranged from 2.0 nmol L$^{-1}$ (28 %) in the Rajang River (at salinity = 0 in August 2016) to 41.4 nmol L$^{-1}$ (570 %) in the Simunjan River (at salinity = 0 in March 2017). CH$_4$ concentrations (saturations) were in the range of 2.0 nmol L$^{-1}$ (28 %) to 1372 nmol L$^{-1}$ (57 459 %) in the Simunjan River (at salinity = 0 in March 2017). N$_2$O concentrations showed a positive linear correlation with rainfall. We conclude, therefore, that rainfall, which determines the N$_2$O production or consumption in the surrounding soils, is the main factor determining the riverine N$_2$O concentrations. N$_2$O production in the blackwater rivers themselves seems to be low because of the low pH. CH$_4$ concentrations were highest at salinity = 0 and most probably result from methanogenesis as part of the decomposition of organic matter under anoxic conditions. CH$_4$ concentrations in the blackwater rivers showed an inverse relationship with rainfall. We suggest that enhanced CH$_4$ oxidation in combination with a higher flux across the river–atmosphere interface during
Table 4. Overview of N\textsubscript{2}O and CH\textsubscript{4} flux densities from aqueous and soils ecosystems in SE Asia.

| Site                        | Location                        | N\textsubscript{2}O flux density, nmol m\textsuperscript{-2} s\textsuperscript{-1} | CH\textsubscript{4} flux density, nmol m\textsuperscript{-2} s\textsuperscript{-1} | Measurement or Reference                  |
|-----------------------------|---------------------------------|---------------------------------------------------------------------------------|---------------------------------------------------------------------------------|------------------------------------------|
|                             |                                 | Range    Mean\*                      | Range    Mean\*                      |                                                                                   |
| **Aqueous systems**         |                                 |          |                                   |                                                                                   |
| Rajang River/Estuary        | Sarawak, NW Borneo              | −0.33 to 1.22 0.53                 | 0.34 to 62 5.52                     | Aug 2016; Mar, Sep 2017                                                           | This study                              |
| Maludam River/Estuary       | Sarawak, NW Borneo              | −0.20 to 1.00 0.32                 | 0.09 to 50 15.9                     | Mar, Sep 2017                                                                     |                                        |
| Sebuyau River/Estuary       | Sarawak, NW Borneo              | −0.18 to 1.08 0.39                 | 0.28 to 78 15.4                     | Mar, Sep 2017                                                                     |                                        |
| Simunjan River/Estuary      | Sarawak, NW Borneo              | −0.31 to 2.20 0.50                 | 0.01 to 88 18.7                     | Mar, Sep 2017                                                                     |                                        |
| Sematan River/Estuary       | Sarawak, NW Borneo              | −0.11 to 0.04 −0.05                | 0.43 to 72 21.1                     | Mar 2017                                                                           |                                        |
| Samunsam River/Estuary      | Sarawak, NW Borneo              | −0.13 to 0.19 0.05                 | 0.95 to 63 21.7                     | Mar 2017                                                                           |                                        |
| Lupar River/Estuary         | Sarawak, NW Borneo              | 0.04 to 0.04 0.04                  | 0.59 to 0.84 0.72                   | Jun 2013; Mar 2014                                                                | Müller et al. (2016a)                   |
| Saribas River/Estuary       | Sarawak, NW Borneo              | 0.04 to 0.08 0.06                  | 0.45 to 1.01 0.73                   | Jun 2013; Mar 2014                                                                |                                        |
| Saribas River tributary     | Sarawak, NW Borneo              | 0.37 to 0.39 0.38                  | 0.81 to 4.84 2.83                   | Jun 2013; Mar 2014                                                                |                                        |
| Drainage canal, Kalimantan, settled | Central Kalimantan, S Borneo | −0.02 to 0.03 0 | 0 to 943 119 | Sep 2007; Apr 2008 | Jauhiainen and Silvennoinen (2012) |
| Drainage canal, Kampar, settled | Riau, eastern central Sumatra | 0.03 to 5.80 0.73                  | 0 to 3672 776 | Sep 2007; Apr 2008 |                                        |
| Drainage canal, Kampar, disturbed | Riau, eastern central Sumatra | 0.02 to 0.84 0.20                  | 2.17 to 281 64.4 | Sep 2007; Apr 2008 |                                        |
| **Soil systems**            |                                 |                                    |                                    |                                                                                   |
| Forest                      | Sarawak, NW Borneo              | −0.03 to 0.20 0.08                 | −0.10 to 0.19 0.04                  | Aug 2002–Jul 2003                                                                | Melling et al. (2005, 2007)            |
| Sago plantation             | Sarawak, NW Borneo              | 0.01 to 1.75 0.88                  | −0.17 to 2.36 1.10                  | Aug 2002–Jul 2003                                                                |                                        |
| Oil palm plantation         | Sarawak, NW Borneo              | 0.01 to 0.58 0.29                  | −0.76 to 0.11 −0.33                 | Aug 2002–Jul 2003                                                                |                                        |
| Undrained forest            | Central Kalimantan, S Borneo    | −0.09 to 1.16 0.02                 | NA NA                               |                                                                                   |
| Drained forest              | Central Kalimantan, S Borneo    | −0.42 to 22.9 1.11                 | NA NA                               |                                                                                   |
| Drained recovering forest   | Central Kalimantan, S Borneo    | −0.06 to 0.45 0.02                 | NA NA                               |                                                                                   |
| Drained burned peat         | Central Kalimantan, S Borneo    | −0.70 to 0.88 0.11                 | NA NA                               |                                                                                   |
| Agricultural peat in Kalampangan | Central Kalimantan, S Borneo | −0.95 to 0.89 0.12                 | NA NA                               |                                                                                   |
| Agricultural peat in Marang | Central Kalimantan, S Borneo    | −0.86 to 0.59 0.07                 | NA NA                               |                                                                                   |
| Canopy soil of oil palm     | Jambi, eastern central Sumatra  | NA 0.001                           | NA 0.0004                           | Feb 2013–May 2014                                                                | Allen et al. (2018)                    |
| Drained burned land         | Central Kalimantan, S Borneo    | NA 0.001                           | NA 21.1                             | Jul 2011                                                                           | Ishikura et al. (2018)                |
| Drained forest              | Central Kalimantan; S Borneo    | NA 0.08                            | NA 0.23                             | Jul 2011                                                                           |                                        |
| Undrained forest            | Central Kalimantan; S Borneo    | NA 0.15                            | NA 17.6                             | Jul 2011                                                                           |                                        |
| Drained agricultural land   | Various locations in SE Asia    | 0.81 to 29.3 10.3                  | 0.05 to 6.74 3.39                   | Various dates                                                                     | Cauwenberg et al. (2010)              |
| (fertilized)                |                                 |                                    |                                    |                                                                                   |
| Drained, open vegetation    | Various locations in SE Asia    | −0.12 to 0.45 0.08                 | NA NA                               | Various dates                                                                     |                                        |
| (abandoned, not fertilized) |                                 |                                    |                                    |                                                                                   |
| Forested (drained and       | Various locations in SE Asia    | −0.06 to 1.51 0.39                 | −0.73 to 11.6 5.45                  | Various dates                                                                     |                                        |
| undrained peat swamp,       |                                 |                                    |                                    |                                                                                   |
| agro-forestry)              |                                 |                                    |                                    |                                                                                   |
| Rice paddies                | Various locations in SE Asia    | −0.04 to 0.23 0.07                 | 7.17 to 98.1 52.7                   | Various dates                                                                     |                                        |
| Peat soil                   | Various locations in SE Asia    | NA NA                              | 0 to 52.1                           | Various dates                                                                     |                                        |
| Maludam Natl. Park          | Sarawak, NW Borneo              | NA NA                              | 23.1 Nov–Dec 2013                   | Tang et al. (2018)                                                               |                                        |

* Values in italics indicate a mean flux density computed from the range given in the table (when no mean flux density was given in the reference). NA stands for not available/not measured.
periods of higher river flow (after rainfall events) is responsible for the reduction in the CH$_4$ concentrations along the salinity gradient. The rivers and estuaries studied here were an overall net source of N$_2$O and CH$_4$ to the atmosphere. The total annual N$_2$O and CH$_4$ emissions were 1.09 Gg N yr$^{-1}$ (0.7 Gg N yr$^{-1}$) and 23.8 Gg CH$_4$ yr$^{-1}$, respectively. This represents about 0.3%–0.7% of the global annual riverine and estuarine N$_2$O emissions and about 0.1%–1% of the global riverine and estuarine CH$_4$ emissions. Rivers and estuaries in NW Borneo contribute only 0.05% (= 7.9 × 10$^7$ km$^2$ including the surface areas of the Lupar and Saribas rivers; Müller et al., 2016a) to the global water surface area of rivers and estuaries (= 1.7 × 10$^9$ km$^2$; Maavara et al., 2019). Therefore we conclude that rivers and estuaries in NW Borneo contribute significantly to the global riverine and estuarine emissions of both N$_2$O and CH$_4$.

The environment of Borneo (and SE Asia) is affected by rapid changes due to (i) anthropogenic activities such as conversion of peatland into oil palm plantations (see, e.g., Austin et al., 2018; McAlpine et al., 2018; Schoneveld et al., 2019) and (ii) climatic changes (see, e.g., Sa’adi et al., 2017a, b; Tang, 2019) which, in turn, could significantly affect N$_2$O and CH$_4$ emissions from soils (see, e.g., Jauhiainen et al., 2016; Oktarita et al., 2017). But little is known about how these changes will affect N$_2$O and CH$_4$ emissions from aquatic systems such as rivers and estuaries in the future. The obvious relationship of N$_2$O and CH$_4$ concentrations and rainfall could be used to predict future concentrations and its associated emissions to the atmosphere. However, the trends of rainfall and river discharge in Borneo show a high local variability and no general common trend (Sa’adi et al., 2017a; Tang, 2019). Therefore, predictions of future trends of N$_2$O and CH$_4$ emissions will be associated with a high degree of uncertainty. In order to improve our knowledge of predicted future changes in N$_2$O and CH$_4$ riverine or estuarine emissions, we suggest establishing regular measurements in the rivers and along the salinity gradients. This will help decipher the temporal and spatial variability of N$_2$O and CH$_4$ emissions from tropical rivers and estuaries. Moreover, studies of the relevant production or consumption pathways (and their main driving factors) for both gases are required. A suitable framework for this could be the recently published concept of the global N$_2$O Ocean Observation Network (N2O-ON) (Bange et al., 2019).

Data availability. All N$_2$O/CH$_4$ data presented here are archived in and available from the MEMENTO (the MarineE Methane and NiTrous Oxide) database: https://memento.geomar.de (last access: 4 November 2019).

Author contributions. MM, CHS, AM and HWB designed the study. CHS performed the sample preparation during the campaigns. DB and JK performed the N$_2$O/CH$_4$ measurements with support from AK. HWB prepared the paper with contributions from all co-authors.

Competing interests. The authors declare that they have no conflict of interest.

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