

SPECIAL ISSUE-LETTER

Carbon dioxide and methane emissions of Swedish low-order streams—a national estimate and lessons learnt from more than a decade of observations

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Scientific Significance Statement

Streams have been identified as disproportional emitters of CO₂ to the atmosphere across all inland waters. Despite their suggested importance, reliable large-scale stream C emission data are often lacking which makes current estimates uncertain. Here, we show that Swedish low-order streams emit much higher amounts of C to the atmosphere than previously reported, corresponding to 21% of the estimated terrestrial C sequestration. We also show that local scale spatiotemporal variability in stream gas concentrations often exceeds variability across regions, and that stream surface area matters. Without such fundamental information, large-scale stream C emission estimates will always be associated with a large degree of uncertainty.

Abstract

Low-order streams are suggested to dominate the atmospheric CO₂ source of all inland waters. Yet, many large-scale stream estimates suffer from methods not designed for gas emission determination and rarely include other greenhouse gases such as CH₄. Here, we present a compilation of directly measured CO₂ and CH₄ concentration data from Swedish low-order streams (> 1600 observations across > 500 streams) covering large climatological and land-use gradients. These data were combined with an empirically derived gas transfer model and the characteristics of a ca. 400,000 km stream network covering the entire country. The total

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Author Contribution Statement: MBW brought the idea and compiled the concentration data. MBW and TG designed the study, analyzed the data and conducted the modelling component. AC, JA, DB, KB, JK, HJ, EL, SL, SN, SB, CT and GAW provided data, ideas and catchment/region specific information. MBW wrote the manuscript with great support from all co-authors.

Data Availability Statement: Data are available in the Uppsala University data repository at <http://urn.kb.se/resolve?urn=urn:nbn:se:uu:diva-332472>.

Additional Supporting Information may be found in the online version of this article.

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This article is part of the Special Issue: Carbon cycling in inland waters

Edited by: Emily Stanley and Paul del Giorgio

stream CO₂ and CH₄ emission corresponded to 2.7 Tg C yr⁻¹ (95% confidence interval: 2.0–3.7) of which the CH₄ accounted for 0.7% (0.02 Tg C yr⁻¹). The study highlights the importance of low-order streams, as well as the critical need to better represent variability in emissions and stream areal extent to constrain future stream C emission estimates.

Although streams and rivers comprise a small part of the water surface area (~ 20%) (Downing et al. 2012; Raymond et al. 2013), they have been shown to dominate the carbon dioxide emissions of all inland waters (85%). Despite a clear bias in geographical coverage of investigated streams toward North-America and Europe, low-order streams (stream order [SO] 1–4) are suggested to be disproportional contributors, emitting more than 70% of the total stream and river CO₂ emissions (Raymond et al. 2013). Despite the suggested importance, reliable large scale emission data for these small streams are often lacking, making current estimates uncertain (Marx et al. 2017). Spatial scaling of stream emission requires in principle three components; the gas concentration gradient between water and atmosphere, the gas transfer velocity (k) describing the physical efficiency for gas exchange across the air–water interface, and the areal extent of stream surfaces. Most existing large-scale stream and river emission studies have, in the absence of direct measurements, derived their CO₂ concentration estimates from indirect methodologies using standard water chemistry measurements often provided by various monitoring programs (Humborg et al. 2010; Butman and Raymond 2011; Raymond et al. 2013; Lauerwald et al. 2015). By using alkalinity, pH, and water temperature together with known chemical equilibrium reactions, the CO₂ concentration could be calculated. Although such indirect methods might be suitable for certain systems, they are known to generate questionable CO₂ in systems with low (or no) alkalinity, low pH, and high organic carbon concentrations, if it is possible to estimate the CO₂ at all (Hunt et al. 2011; Wallin et al. 2014; Abril et al. 2015). Such conditions are typically found in streams draining landscapes with low weathering rates and with organic-rich soils which characterize much of the boreal and subarctic regions. In addition, low-pH streams without alkalinity have higher CO₂ concentrations than streams containing alkalinity (Wallin et al. 2014). Although such streams without alkalinity may not be that common at a global scale, 25% of all Swedish streams with less than a 5 km² catchment area are estimated to have a pH under 5.6 (Bishop et al. 2008). These streams are systematically excluded from current CO₂ data sets, simply because CO₂ cannot be estimated in a reliable way. Furthermore, there are other greenhouse gases (GHG) such as methane (CH₄) that might be important for the total emissions especially when considering the different global warming potentials (GWP). Hence, representative large-scale estimates of stream GHG emissions should be based on direct measurements of stream GHG concentrations.

Estimates of stream gas transfer velocities (k) are in most recent large scale studies (Butman and Raymond 2011; Raymond et al. 2013; Lauerwald et al. 2015) based on an empirical model with k being dependent on the stream channel slope and water velocity (Eq. 5 in Raymond et al. 2012). Although this is the most comprehensive study to date that parameterize gas transfer velocities in streams, the model output has rarely been validated against independent measurements. For example, the most used model found in Raymond et al. (2012) (Eq. 5) has an intercept that returns a k of ~ 2 m d⁻¹ at zero stream channel slope and/or water velocity. This is at odds with several catchment-scale studies that have reported stream $k < 2$ m d⁻¹ in slow-flowing stream sections with low elevation differences (Wallin et al. 2011; Campeau et al. 2014; Natchimuthu et al. 2017).

The quantification of a representative surface area is a critical and challenging task for any area-based C emission estimate, whether it is made for terrestrial or aquatic systems. This is especially true for low-order streams that are often very dynamic in their lateral and longitudinal extent, morphology and channel geometry along the network. This makes scaling of local observations to catchment or regional estimates highly challenging. Although recent efforts have been made to quantify the areal extent of stream and river network at catchment (Wallin et al. 2013), regional (Ran et al. 2015) and global scales (Downing et al. 2012; Raymond et al. 2013), there are still large uncertainties partly due to stream and river surface identification being restricted by the resolution of current remote sensing techniques (Benstead and Leigh 2012). In addition, high spatiotemporal dynamics with areal contraction and expansion of stream networks according to the hydrological conditions complicate the determination of stream area (Stanley et al. 1997; Godsey and Kirchner 2014; Ågren et al. 2015).

The aim of this study was to estimate stream CO₂ and CH₄ emissions of all low-order streams (SO 1–4) in Sweden. The data basis was a compilation of studies using direct concentration measurements conducted during the last 15 yr. More specifically, we used a combination of available directly measured stream CO₂ and CH₄ concentration data (both published and unpublished), a spatiotemporally distributed gas transfer model based on measurements from one of the included studies, together with the characteristics of a high-resolution national stream network inventory. This effort aimed to present a national stream CO₂ and CH₄ emission inventory that is as accurate as is currently possible, but maybe even more importantly highlighting knowledge gaps

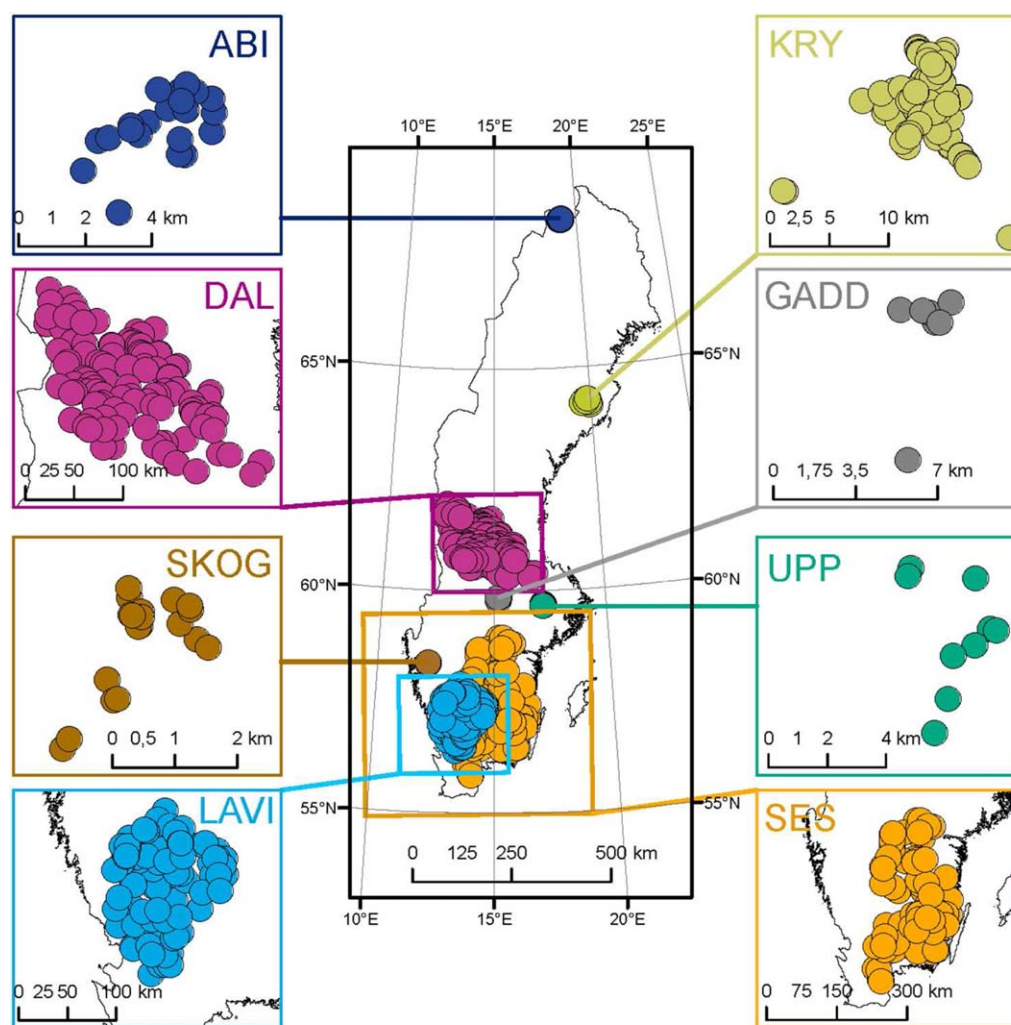


Fig. 1. Catchments/regions where stream CO_2 and CH_4 concentrations have been directly measured. Each sampled stream site is given for each of the catchments/regions.

that need to be filled to constrain stream emission studies in general.

Methods

CO_2 and CH_4 concentration data sets as well as brief site descriptions

The stream CO_2 and CH_4 concentration data set is based on a compilation of published and unpublished data collected during 2005–2016. There were two criteria for data inclusion, (1) the data must be based on direct concentration measurements of CO_2 and CH_4 , and (2) streams must be of low-order, ranging from 1st to 4th Strahler SO. The first criterion excludes data where CO_2 has been indirectly determined by calculation from alkalinity, pH, and temperature. The data set represents a mixture of sampling methods including different headspace (CO_2 and CH_4) and sensor (CO_2) techniques. The sensor- and headspace-derived CO_2

concentrations have shown close correspondence (Bastviken et al. 2015). For further sampling and analysis details concerning data used, see the specific data sources (Supporting Information Table S10). All unpublished data were based on sampling and analytical methods described in (Wallin et al. 2010, 2014) and (Åberg and Wallin 2014). Concentrations of CO_2 and CH_4 are reported in mg C L^{-1} and $\mu\text{g C L}^{-1}$, respectively, with associated interquartile range (IQR). For comparing purpose, median partial pressures of CO_2 and CH_4 for the entire data set are reported assuming a stream water temperature of 8°C. The concentration data represents a combination of regional and catchment surveys within Sweden (see Fig. 1). Regional studies are characterized by many stream sites (typically ca. 100 per region) sampled on one or a few occasions (DAL, LAVI, SES, KRY) (Wallin et al. 2014; unpublished). Catchment studies include fewer sampled streams (typically <20) but with more observations over time (ABI, KRY, GADD, UPP, SKOG) (Audet et al. 2017;

unpublished, Wallin et al. 2010; 2013; unpublished, Kocic et al. 2015, Kocic 2017, Lundin et al. 2013, Natchimuthu et al. 2017). As the dataset of CO₂ and CH₄ concentrations was very heterogeneous in terms of spatiotemporal coverage and number of observations, with different sampling designs and purposes of the original studies, it was hard to fully evaluate concentration differences among the different catchments/regions. The catchments/regions spanned a large geographical and hydro-meteorological range covering a large part of Sweden including subarctic (ABI), boreal (KRY, DAL, GADD), and hemiboreal (UPP, SKOG, LAVI, SES) regions (Fig. 1, Supporting Information Table S10). The main land-use classes in each stream specific catchment were dominated by mires and shrubs in ABI, forest and mires in KRY, DAL, GADD, SKOG, LAVI, and SES, and agriculture in UPP. Meteorological data (precipitation and annual mean air temperature) for each catchment/region represents the 30 yr mean values (1961–1990) according to the Swedish Meteorological and Hydrological Institute (Raab and Vedin 1995).

Stream network determination

The stream and river network with associated catchment boundaries were collected from a virtual stream network of Sweden (VIVAN), based on a digital elevation model (50 × 50 m resolution) (Nisell et al. 2007). From this network, a total of ca. 425,000 km of stream and river length (excluding lakes and reservoirs) were identified (Supporting Information Table S11). The total network was further divided into ca. 472,000 individual stream segments without any stream junctions. This resulted in an average stream segment length of ca. 900 m. Stream characteristics (catchment area, main land-use, stream channel slope) were derived for each individual stream segment. Land-use distribution within each catchment was based on the digital versions of the topographic map (1 : 50,000) and the road map (1 : 100,000) (Swedish Land Survey) separated into three land-use classes; Agricultural (including agricultural land, other open and populated areas), Forest and Mire (including clear cut areas) and Alpine. Out of the total stream and river network, stream segments of 1st to 4th order streams ($n = 443,763$) were used in this study. The stream surface area for each stream segment of 1st to 4th order streams was assumed to be constant over the year and was calculated by multiplying stream section length with SO specific width estimates derived from data published by Downing et al. (2012) (Supporting Information Table S13).

Modeling of C emission and statistical analysis

Stream CO₂ and CH₄ emission was calculated for each of the 443,763 individual stream segments by using the diffusive flux equation (Liss and Slater 1974):

$$E_g = \Delta[g] \times k_g \times A \quad (1)$$

Where E_g is the emission of the specific gas (CO₂ or CH₄) for the individual stream segment (mg m⁻² d⁻¹); $\Delta[g]$ (mg C

L⁻¹) is the difference between the in-stream CO₂ or CH₄ concentration and the concentration that would exist if the stream was in equilibrium with the atmosphere (assuming an atmospheric $p\text{CO}_2$ and $p\text{CH}_4$ of 400 μatm and 1.9 μatm , respectively); k_g is the gas transfer velocity (m d⁻¹) and A is the stream surface area of the specific stream segment. Mean stream CO₂ emission was calculated using SO specific median concentrations of Δ_{CO_2} based on all concentration observations. For emission of CH₄, the median Δ_{CH_4} concentration for all observations included in the study was used for each of the stream segments (*see* Supporting Information for motivation). Mean k was modeled for each individual stream segment based on data presented by Natchimuthu et al. (2017) (*see* more details in Supporting Information). Mean k values are reported in m d⁻¹ with associated standard deviation (SD). Total annual median CO₂ and CH₄ emission rates with associated 95% confidence interval (CI) were determined using an extensive Monte Carlo experiment. A detailed description of the emission calculations and uncertainty estimation is presented in the Supporting Information. Differences in gas concentrations and gas transfer velocities between the different catchments/regions, SOs, and land-use categories were tested on each pair using the nonparametric Wilcoxon's test (for gas concentrations) and Tukey-Kramer's test (for gas transfer velocities). Spatial differences between catchments/regions, SO or land-use classes were considered significant if $p < 0.05$. The data were logarithmically transformed when needed to achieve normal distribution. JMP 12.0.1 (SAS Institute, Cary, North Carolina) was used for all statistical calculations except for the Monte-Carlo experiment which was run in MATLAB (R2017a).

Results

CO₂ and CH₄ concentration patterns

The median CO₂ concentration for all low-order stream observations (1962 individual observations across 596 different stream sites) was 1.7 (± 1.7) mg C L⁻¹ or equal to a $p\text{CO}_2$ of 2468 (± 2509) μatm (Fig. 2). The CO₂ concentration for individual samples ranged from 0.2 mg C L⁻¹ to 65.8 mg C L⁻¹. The median CO₂ concentrations for the individual regions/catchments ranged from 1.1 (± 0.9) mg C L⁻¹ in GADD to 9.0 (± 9.1) mg C L⁻¹ in UPP. The median CO₂ concentration in UPP was more than three times higher than the second highest regional/catchment specific median CO₂ observed in LAVI (2.4 [± 1.5] mg C L⁻¹) ($p < 0.0001$). The median CO₂ concentrations were different across the four SOs and decreased with increasing SO (Fig. 3A) (median concentrations per SO; SO1 = 2.0 mg C L⁻¹, SO2 = 1.4 mg C L⁻¹, SO3 = 1.2 mg C L⁻¹, SO4 = 0.8 mg C L⁻¹). The median CH₄ concentration for all stream observations (1696 individual observations across 525 different stream sites) was 6.7 (± 13.5) $\mu\text{g C L}^{-1}$ or equal to a $p\text{CH}_4$ of 272 (± 550) μatm (Fig. 2). The CH₄ concentration for individual samples ranged

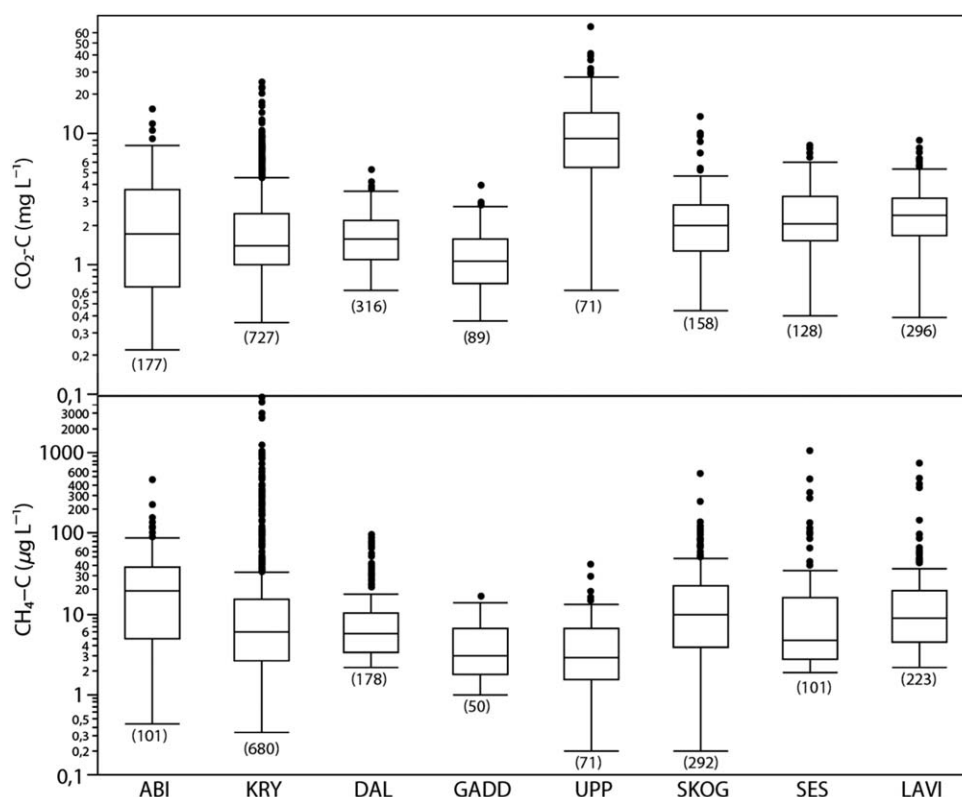


Fig. 2. Distribution of stream CO_2 and CH_4 concentrations measured in the different catchments/regions. Numbers of observations are given within brackets. Catchments/regions are organized by their north-south geographical location with the furthest northern located catchment, ABI, to the left.

from $0.2 \mu\text{g C L}^{-1}$ to $4829.4 \mu\text{g C L}^{-1}$. The median CH_4 concentrations for the individual regions/catchments ranged from $2.9 (\pm 5.0) \mu\text{g C L}^{-1}$ in UPP to $19.1 (\pm 33.6) \mu\text{g C L}^{-1}$ in ABI. The median CH_4 concentration in ABI was almost two times higher than the second highest regional/catchment specific median CH_4 observed in SKOG ($10.0 [\pm 17.9] \mu\text{g C L}^{-1}$) ($p = 0.0002$). CH_4 concentrations did not display a similar trend in decreasing concentration with increasing SO as observed for CO_2 , instead the median CH_4 concentration was highest in SO3 ($\text{SO1} = 7.3 \mu\text{g C L}^{-1}$, $\text{SO2} = 5.7 \mu\text{g C L}^{-1}$, $\text{SO3} = 8.9 \mu\text{g C L}^{-1}$, $\text{SO4} = 3.6 \mu\text{g C L}^{-1}$) (Fig. 3B).

The distribution of stream CO_2 and CH_4 concentrations across all regions/catchments was relatively similar (Fig. 2), and the variability (expressed as the IQR) within one catchment/region was generally higher than the variability between the different catchments/regions. The exception was the agriculturally dominated catchment UPP, which showed both the highest median CO_2 concentration and SD ($9.0 [\pm 9.1] \text{mg C L}^{-1}$) relative to the remaining catchments/regions. When excluding UPP, the catchment/region specific variability in CO_2 concentration, expressed by the IQR ($0.8\text{--}3.0 \text{mg C L}^{-1}$), was higher for all catchments/regions than the variability among the region specific median concentrations ($\text{IQR} = 0.8 \text{mg C L}^{-1}$). A similar comparison for CH_4 concentrations showed that the variability across all catchment/

region specific median CH_4 concentrations ($\text{IQR} = 6.3 \mu\text{g C L}^{-1}$) was lower than the variability within most of the catchments/regions. The two exceptions were the GADD and UPP catchments ($\text{IQR} = 4.9 \mu\text{g C L}^{-1}$ and $\text{IQR} = 5.1 \mu\text{g C L}^{-1}$, respectively), while the range of IQR for remaining catchments/regions was from $6.8 \mu\text{g C L}^{-1}$ to $33.6 \mu\text{g C L}^{-1}$.

Despite the heterogeneity in these data, monthly median stream CH_4 concentration displayed a significant seasonal variability as a function of sampling month, best described through an intra-annual cubic relationship with highest CH_4 concentrations observed in August–September (Supporting Information Figs. S5, S6). In contrast, no seasonal pattern was observed for CO_2 concentration. The observed seasonal pattern was not related to monthly air temperature patterns, either for CO_2 or CH_4 concentrations (Supporting Information Fig. S7). The stream CO_2 and CH_4 concentrations were significantly related to each other both when treating the regions/catchments individually (data not shown) and when combining all observations (Supporting Information Fig. S8). Again, the exception was the agriculture dominated UPP catchment, where the stream CO_2 and CH_4 concentrations were unrelated. In addition, the UPP catchment displayed both the highest median CO_2 concentration and the lowest median CH_4 concentrations across all regions/catchments (Fig. 2, Supporting Information Table S10).

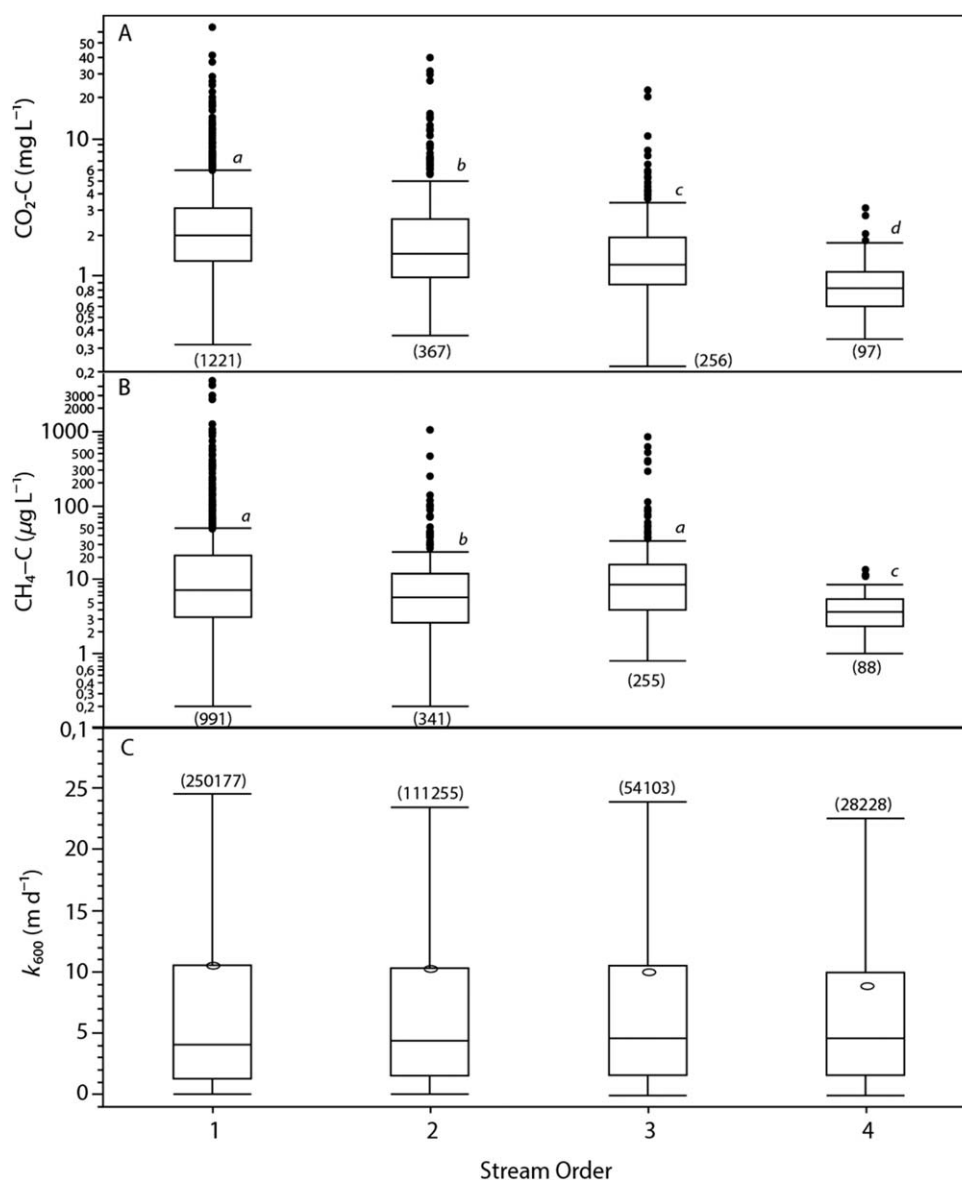


Fig. 3. Stream CO₂ (A) and CH₄ (B) concentrations for all observations included in the study and separated by SO. Numbers of observations are given within brackets and with italic letters describing statistical differences. (C) Distribution of modeled gas transfer velocities (k_{600}) for the stream network of Sweden. Number of stream segments are given within brackets. Median values, IQRs, and 10th and 90th percentiles are indicated by box-plots. Mean values are indicated by circles.

Distribution of gas transfer velocities

The overall mean and median (\pm SD) k_{600} across all 1st to 4th order streams was 9.5 and 4.2 (\pm 19.5) m d⁻¹, respectively. The mean k_{600} decreased slightly with increasing SO, from 9.9 m d⁻¹ in 1st order streams to 8.8 m d⁻¹ in 4th order streams (Fig. 3C, Supporting Information Table S13) ($p < 0.0001$). The mean stream k_{600} were different among the land-use classes ($p < 0.0001$), with 33.5 (\pm 48.3) m d⁻¹, 3.9 (\pm 7.5) m d⁻¹, and 8.6 (\pm 14.5) m d⁻¹ for alpine, agricultural, and forest/mire regions, respectively. Land-use specific k_{600} decreased with increasing SO in alpine regions (from 37.4 m d⁻¹ to 15.6 m d⁻¹). In contrast, k_{600} increased with increasing SO in

agricultural regions (from 3.5 m d⁻¹ to 5.1 m d⁻¹). For forest and mire covered regions, k_{600} did not differ statistically between all stream sizes ($p = 0.28$).

Stream network distribution

The majority (74%) of the total stream and river length of Sweden drain forested regions, with agricultural and alpine regions representing 19% and 7% of the total stream length, respectively. About 400,000 km, or 95% of the total stream and river length, were represented by low-order streams (\leq SO4), draining catchments with a median catchment area of < 51 km² (Supporting Information Table S11). For these

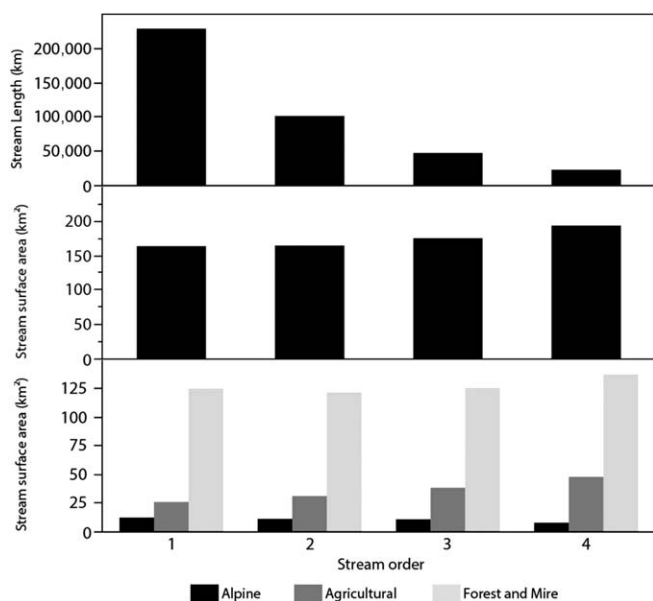


Fig. 4. Stream length and stream surface area of the Swedish stream network (Strahler SO1–4) in total and separated by land-use class.

1st to 4th order streams, the total stream length roughly decreased by 50% for each step in increased SO (from SO1 = ca. 230,000 km to SO4 = 23,000 km), but the estimated total surface areas were similar across the four SOs ranging from 164 km² to 194 km² (Fig. 4, Supporting Information Table S13). A majority (73%) of the total low-order stream surface area (697 km²) was located in areas draining forest and mire, while agricultural and alpine areas represent 21% and 6%, respectively.

Stream CO₂ and CH₄ emissions

The total C (CO₂ + CH₄) emission from low-order streams of Sweden was estimated at 2.7 Tg C yr⁻¹ (95% CI: 2.0–3.7) with CH₄ being responsible for 0.7% of the C share (Fig. 5). The majority of the total emissions derived from 1st and 2nd order streams (70%) or from streams draining areas dominated by forest and mires (67%). Alpine areas were estimated to emit 26% of the total stream C emissions although being responsible for just 7% of the total stream surface area. The CH₄ emissions contributed to 17% of the total stream GHG emissions when considering a 28 times higher GWP for CH₄ (excluding climate-carbon feedbacks) over a 100-yr horizon (IPCC 2013).

Discussion

Here, we show that the total C emissions from Swedish low-order streams alone (2.7 Tg C yr⁻¹) equals the sum of a previously reported national CO₂ emission estimate for all inland waters (total 2.6 Tg C yr⁻¹; lakes 1.8 Tg C yr⁻¹; streams 0.8 Tg C yr⁻¹) (Humborg et al. 2010) together with a national estimate of total lake CH₄ emission (0.1 Tg C yr⁻¹) (Bastviken et al. 2004). The much larger importance of low-

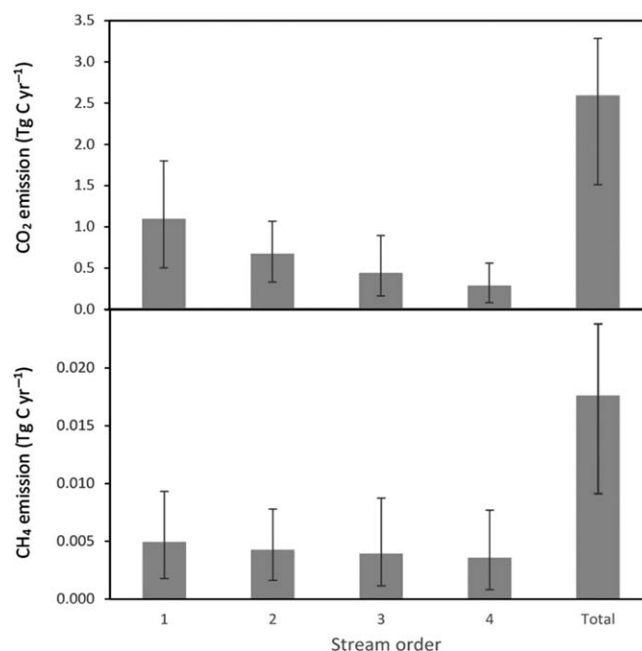


Fig. 5. Annual stream emissions of CO₂ and CH₄ for the stream network of Sweden separated by Strahler SO1–4 and in total. Error bars represent the 95% CI.

order streams as presented here was mainly an effect of the ca. 2.5 times more stream surface area estimated in the present study. The stream C emissions correspond to 21% of the total net atmospheric C uptake by land-use, land-use change and forestry (12.8 Tg C yr⁻¹ for 2014) reported by Sweden to the UNFCCC and the Kyoto protocol (SEPA 2016). Hence, neglecting stream C emissions in landscape C balances will significantly overestimate the terrestrial C sequestration. The national stream C emission estimate we present here is consistent with the findings of catchment studies on low-order boreal streams which have shown that lateral fluxes of organic and inorganic C are significant fractions (7–28%) of the terrestrial C uptake (Huotari et al. 2013; Wallin et al. 2013; Öquist et al. 2014). Corresponding estimates for the conterminous U.S. and Alaska showed that total stream and river CO₂ emissions averaged between 8% and 27% of the terrestrial Net Ecosystem Productivity (Butman et al. 2016; Stackpoole et al. 2017).

The stream CO₂ and CH₄ concentrations in Swedish streams were highly variable in both space and time. Although the included catchments/regions covered a large geographical and hydro-meteorological range, much of the entire observed concentration range found across Sweden was covered in any of the individual catchments/regions (Fig. 2). The exception was for CO₂ concentrations in the UPP catchment, which displayed more than three times higher median CO₂ concentration (9.0 mg C L⁻¹) than any of the other catchments/regions. This was the only catchment/region with significant agricultural land-use (around

50%), while the agricultural influence was less than 5% in the other catchments/regions. Stream CO₂ concentration data from agricultural systems is generally underrepresented in the literature, but Sand-Jensen and Staehr (2012) reported high CO₂ concentrations in Danish agricultural streams, with a median of 2.5 mg C L⁻¹ and individual streams with up to 35 mg C L⁻¹. Studies from Germany and Belgium have also found that streams draining agriculture-dominated areas had higher CO₂ concentration than streams draining forested areas (Bodmer et al. 2016; Borges et al. 2018). The source of the high CO₂ concentrations in the UPP streams is unclear, but potentially arises from weathering of ancient glacier deposits containing carbonates that are commonly found in the soils of the area. Further investigations using isotopic characterization (both radiogenic and stable) (e.g., Campeau et al. 2017a,b) of the dissolved CO₂ may allow better constraint on the source of the high CO₂ concentrations in UPP.

For CH₄, it was striking to see the high variability in concentrations across Swedish streams, but also how similar these data are with the extensive global stream and river CH₄ concentration data compiled by Stanley et al. (2016). The median CH₄ concentration of all the stream observations of this study was 6.7 μg C L⁻¹, while the median headwater CH₄ concentrations of the compiled literature are ~ 4 μg C L⁻¹. More strikingly, the total concentration range found for streams and rivers globally (0–4632 μg C L⁻¹) was close to the range found in this study (0.2–4829 μg C L⁻¹, Fig. 2). Local hotspot sources and sinks, terrestrial or in-stream, often result in very variable CH₄ concentrations within stream networks, but also along individual streams (Crawford et al. 2017). In addition to very variable concentration and emission patterns, CH₄ also has additional transport pathways across the water-atmosphere interface through ebullition and through the stem of plants. The role of nondiffusive CH₄ emission processes is generally understudied in aquatic systems, and this is especially true for streams with just a few ebullition examples found in the literature. Crawford et al. (2014) found high ebullition of CH₄ for low gradient wetland streams in the U.S., with ebullition being close to equaling the diffusive CH₄ emission. Similarly, Baulch et al. (2011) found ebullition to contribute 20–67% of the total CH₄ emission from Canadian agricultural streams. As there are no measurements on CH₄ ebullition from Swedish streams, our estimate for CH₄ represents diffusive emission only. Hence, additional efforts concerning stream CH₄ emission are clearly needed.

Due to the heterogeneity of stream CO₂ and CH₄ concentration data, the observed differences between the catchments/regions should be taken with caution. A previous study has shown that the median CO₂ and CH₄ concentrations observed in the DAL and LAVI regions, where identical, spatially representative regional sampling designs were used, were statistically different (Wallin et al. 2014). Still we generally conclude that due to the large variability in gas

concentrations within each of the catchments/regions and with overlaps in concentration ranges for a majority of the observations, any degree of oversaturation for both CO₂ and CH₄ can roughly be found anywhere in Sweden. Instead local conditions related to the presence of, for example, peat soils (Wallin et al. 2010; Crawford et al. 2017), stream channel morphology (Wallin et al. 2011; Natchimuthu et al. 2017), extent and character of stream bed sediments (Crawford et al. 2017), and hydrological conditions (Wallin et al. 2010; Dinsmore et al. 2013) are more important for controlling spatiotemporal concentrations dynamics of CO₂ and CH₄ than geographical location. In order to understand this small-scale variability in sources and sinks of C gas concentrations, there is a need for spatially distributed studies specifically designed to link stream gas observations to terrestrial (catchment or riparian) or in-stream sources, and to physical features of the stream channel. As the stream gas concentrations often vary over the scale of meters (especially for CH₄), a key for this work is sufficiently high resolution of potential predictor variables so that they can explain these local variations.

We generally believe that the majority of CO₂ in these small streams is of terrestrial origin, either through degradation of organic matter or by root respiration, being exported to the draining stream (Hotchkiss et al. 2015; Winterdahl et al. 2016). We propose that in-stream processes are relatively minor contributors to the observed oversaturation of CO₂ as short water residence times, typically from minutes to a few days (Wallin et al. 2013), allow for limited biological organic matter degradation (Catalán et al. 2016), pH and nutrient concentrations often are low, and since many of the streams draining forested areas are limited in sun-light exposure which restricts the photochemical oxidation of organic matter to CO₂. This interpretation is supported by strong terrestrial-aquatic linkage and low in-stream contribution to stream CO₂ reported by detailed studies in the forested KRY catchment (Öquist et al. 2009; Leith et al. 2015). However, as the stream network of Sweden is heterogeneous spanning from fast flowing, nutrient limited, alpine or forested streams to lowland, slow flowing, nutrient rich, agricultural streams, the relative importance of in-stream vs. terrestrial CO₂ sources is likely very variable across the landscape. Our data only includes low-order streams (SO ≤ 4) and efforts to study changes in CO₂ sources along Swedish stream networks of this size are limited (Wallin et al. 2010). Hotchkiss et al. (2015) suggested for U.S. streams and rivers that terrestrial CO₂ sources dominate in low-order streams but that the relative contribution of in-stream sources increase with stream size.

With the exception of the UPP catchment, CO₂ and CH₄ concentrations were weakly but still significantly related, both when treating all data together and for the different catchments/regions individually (Supporting Information Fig. S8). Although the weak relationship should be handled

with caution, the correlation between the two gases could be viewed in three ways, (1) the two gases derive from a common biogeochemical source, (2) the two gases derive from different sources but have a common source control, or (3) their stream-atmosphere emission has a common physical control. Of course, it could also be a combination of these regulators and that their relative importance might vary across landscapes and over time for a specific point. A common biogeochemical source alternative could be anaerobic metabolism as it contributes not only to the oversaturation of CH₄ commonly observed in streams but also to CO₂ (Richey et al. 1988; Stanley et al. 2016). Also, although no analysis on the influence of land use on the CO₂ and CH₄ concentrations were made within this compilation of data, several of the original studies have found correlations between both CO₂ and CH₄ concentration with the peatland coverage in the individual catchments (Wallin et al. 2010, 2014), further supporting occurrence of anaerobic CO₂ sources for these small streams. On the other hand, the ABI catchment, where many of the sampled streams are influenced by peat-rich soils, showed the highest median CH₄ concentration (Fig. 2), the highest CH₄ : CO₂ ratio (0.01) and a relatively weak (but still significant) relationship between CH₄ and CO₂ concentrations ($r^2 = 0.11$, $p = 0.001$). The subarctic ABI catchment is the only catchment/region affected by permafrost, which might explain why the coupling of CO₂ and CH₄ is different, compared to other catchments/regions. However, several of the 2nd to 3rd order stream sites in the ABI catchment are located downstream of small ponds/lakes likely influencing the CH₄ : CO₂ ratio as well. Interesting to note is the decoupling of CO₂ and CH₄ observed in the UPP catchment. The high CO₂ concentrations in UPP may be due to a geogenic C source related to the presence of calcareous soils in the area, and could thereby also explain the decoupling with CH₄. To meet the challenge of spatiotemporally very variable gas concentrations, and to understand the different sources and sinks, direct measurement techniques of aquatic gas concentration need to be incorporated into stream monitoring programs. Furthermore, the high spatiotemporal variability in stream gas concentrations requires systematic approaches that ideally (1) can capture concentration dynamics continuously, i.e., sensor techniques, and (2) that are cheap enough to also enable high spatial coverage. There are several expensive sensor solutions available meeting the first requirement, but only a few existing solutions (and to our knowledge so far just for CO₂) that meet both, e.g., (Johnson et al. 2010; Bastviken et al. 2015). Further technological development is clearly needed, with cheap and robust sensors that also include other GHGs (e.g., CH₄).

To apply a representative stream k is critical in any kind of emission scaling exercise, independent whether it is at catchment or global scales. Here, we used a spatiotemporally resolved model based on data published by Natchimuthu

et al. (2017) (see Supporting Information). This model generated lower k values for streams with low slope or low velocity than Eq. 5 in Raymond et al. (2012). In contrast, our model predicted higher k values in steeper streams and at higher velocities (see Supporting Information Fig. S10 for further comparison of the model outputs). The overall mean stream k modeled in our study was slightly higher than what Butman and Raymond (2011) derived in a similar scaling exercise for the conterminous U.S. An overestimation in C emission rates has been suggested when applying existing gas transfer models on steep streams (Crawford et al. 2015). In our case, unrealistically high segment specific k would cause disproportional influence of a few stream segments on the total emission estimate. To overcome this problem, we applied an upper limit on k_{600} of 600 m d⁻¹. A similar use of an upper limit of k was used by Butman et al. (2016) in a scaling study for the conterminous U.S., but where they did not allow k to exceed 30 m d⁻¹. We base our higher limit on the range in k values measured in the included studies (Wallin et al. 2011; Kokic et al. 2015; Natchimuthu et al. 2017), but also due to measurements of k values higher than 30 m d⁻¹ in the Colorado River, U.S.A. (Hall et al. 2012) and in UK peatland streams (Billett and Harvey 2013). Clearly more research is needed on k in streams, and particularly on the degree to which current gas transfer models correctly reflect stream gas exchange in steep terrain.

Here, we used a “virtual” stream and river network of Sweden, based on a combination of available map information and a national digital elevation model. The stream network represents perennial conditions excluding ephemeral streams. Hence, the emission estimates are conservative from an areal extension perspective. Still the total stream surface area estimate for 1st to 4th order streams, 697 km², is almost 2.5 times larger than the surface area (288 km²) suggested by Humborg et al. (2010) in an earlier inland water emission estimate of Sweden. Although Humborg et al. (2010) did not explicitly present measures of stream lengths, the large discrepancy in estimated stream area between these two studies must be an effect of the differences in determined stream length, as the SO-specific widths used in our study were generally narrower than the widths used in the Humborg et al. (2010) study. Here, we used width data derived from the global dataset presented by Downing et al. (2012) as we did not have representative national width estimates covering all four SOs. The width estimate for 1st order streams (0.7 m) used here was slightly lower than the median stream width (0.9 m) manually measured at the catchment outlets for 228 1st order streams in the KRY, DAL, and LAVI regions (Supporting Information Fig. S9). Applying the Strahler SO concept on remotely identified stream networks is associated with inherent uncertainty, simply because it is hard to judge where exactly streams begin. Hence, the given SO is an arbitrary measure that is highly dependent on the resolution and quality of the remote sensing product as well as the

hydrological conditions. We still find the concept useful for separating streams of different sizes, and interpreted the relatively good agreement in 1st order stream width between our observations and the global data as a motive for also using the global width data as a basis for estimating width in streams with higher SO. According to the total stream length (Nisell et al. 2007) and land surface area (SCB 2012), the drainage density of Sweden at average stream flow is ~ 1.0 km of stream per km² of land surface area. As shown by the VIVAN data, a clear majority of this terrestrial-aquatic interface is located in low-order stream systems which correspond to $\sim 0.2\%$ of the Swedish land surface area. However, such small streams are dynamic in their extent. Ågren et al. (2015) showed for the KRY catchment that going from base flow to high flow conditions increased the total stream length by a factor of 4.5. Similarly, Godsey and Kirchner (2014) showed that the stream length in low-order California streams could vary by factors of 2.6–7.5 related to discharge conditions. Correct understanding of the dynamic extent of streams is critical as the terrestrial source areas change over time, but also due to the variability in the emission contributing surface area. Hence, improving the temporal resolution of the stream network extent is of utmost importance to constraining stream C emission estimates independent of scale.

From being more or less neglected in large-scale estimates of inland water C emissions, streams and rivers are currently suggested to be responsible for the dominant share of global aquatic CO₂ emissions (Raymond et al. 2013). Before this could be seen as a full paradigm shift within the inland water C community, a stronger basis in appropriate data is needed where suitable sampling and scaling methodologies are used. Here, we show this importance, but also the challenge, in estimating C emissions from low-order streams at large scales. Clearly, an improved understanding of the spatiotemporal controls on stream gas concentrations and the physical efficiency (k) for gas exchange in streams is critical. From a more fundamental point of view, we suggest that quantification of the extent of streams and the dynamics of that extent might be an even more important area for improvement. Without this fundamental information, large-scale stream C emission estimates will always be associated with major uncertainties.

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Acknowledgments

MBW was supported by the Knut and Alice Wallenberg foundation (Grant KAW 2013.0091 to Lars J. Tranvik). DB and SN were supported by The Swedish research councils FORMAS (214-2009-872) and VR (2012-00048, 201604829), SKB, and ERC (725546). JA was supported by the Carlsberg Foundation and FORMAS (2015-1559). We further thank SITES Water, funded by the Swedish Research Council (VR), for supporting several of the field stations used for the stream studies. In addition, we thank the financial support for each of the original studies that together have enabled the large CO₂ and CH₄ concentration data set. The sampling in the DAL, LAVI, and SES were financed by the Swedish Environmental Protection Agency, the Swedish Agency for Marine and Water Management, and the Swedish Energy Agency. Many thanks to all samplers of the different catchments including personnel from the county administrations sampling all headwaters in the DAL, LAVI, and SES regions. Reinert Huseby-Karlsen is acknowledged for sharing ADCP data from the KRY catchment. Johan Temnerud and Ishi Buffam are acknowledged for initiating the early work on stream water GHG in Sweden.

Submitted 03 July 2017

Revised 30 October 2017

Accepted 18 December 2017