Author comments on RC1

Interactive comment on "Spatio-temporal variations of lateral and atmospheric carbon fluxes from the Danube Delta" *by* Marie-Sophie Maier et al.
Anonymous Referee #1

Dear Anonymous Referee #1,

Thank you very much for your constructive comments regarding our MS, we highly appreciate the time and effort you invested. In the following, you find our replies (black, indented) to your *individual comments* (blue, italic).

The MS by Maier et al. presents some extensive and original data of C concentrations and CO2 and CH4 fluxes in the Danube delta. This is a well-designed study. The methods are appropriate, the results are well presented and the interpretations are sound. I recommend the publication of this paper. However, I found few weak points that can easily be improve before publication, based on a more detailed analysis of the data and reading of the literature: In general, the text is insisting more on spatial variations, rather than temporal variations. Most of the calculated flux numbers are annual averages for the two years of the study. It would be interesting to interpret more precisely these data in relation with seasonal flooding of the wetland (how do flooded areas change seasonally?), the spring/summer primary production in the wetland, eventually the winter C recycling in the wetland: potentially changing the CO_2, O_2 and CH_4 concentrations and air-water fluxes particularly in the channels. The results of BOD appear in the discussion but not in the result section. A special paragraph in the result section to describe the discrepancies and concordance between in stream respiration and CO_2 outgassing would be useful. It should be made very clear in the discussion that wetland C metabolic and burial fluxes shown in the last figure are from the literature, not necessarily valid for same study period, and that they do not consider seasonal variations.

It was our intention in this paper to focus on the functional differences between lakes and channels. A detailed analysis of the seasonal variability in relation to the flooded area would require a detailed remote-sensing study and this was beyond the scope of this paper. Also, we did not have the capacity to perform direct primary productivity measurements of the reed stands, which would have required a rather complex study design with aquatic plus terrestrial observations. We welcome, however, the other suggestions:

- In the discussion section, we will now include literature data that report the seasonal cycle of reed growth and decomposition. See detailed text below (L600).
- In the revised version, we will also add a short paragraph to the results section documenting the relations between stream respiration and outgassing. Fig. 6 will also be shifted to this section.

"**3.2.3 CO_2 production vs. CO_2 flux**

We find respiration rates ranging between 0.8–390 mM m^{-2} d^{-1} for rivers, while in the channels and lakes they ranged from 2.3–560 mM m^{-2} d^{-1} and 1.0–350 mM m^{-2} d^{-1}, respectively (Fig. 6 and Fig. S5-S7). Median respiration rate is highest in rivers (54 mM m^{-2} d^{-1}), followed by lakes (48 mM m^{-2} d^{-1}) and channels (45 mM m^{-2} d^{-1}). Many stations showed a pronounced seasonality with highest respiration rates occurring mostly between July to October. Respiration rates, i.e. CO_2 production rates generally exceed CO_2 fluxes in river and lake stations throughout the year (Fig. 6), which implies that local instream CO_2 production sustained the observed fluxes. At the channel stations we frequently observed fluxes exceeding the local production, even if we account for potential underestimation of the CO_2 production, which implies the presence of other CO_2 sources. This was most striking at station 10, the CO_2 hotspot, where CO_2 outgassing exceeded local respiration on average by a factor of 40. At the other channel stations (also see Fig S6), there seems to be a seasonally occurring pattern: CO_2 fluxes exceed local production in the first half of the year, while for the remainder of the year they fall below. While this pattern is very distinct in 2016, it is less pronounced in the drier year 2017, which suggests that the additional CO_2 source is linked to hydrology."

- Finally, we will point out more clearly in Fig. 8, that the rates of burial and wetland metabolism were taken from the literature with different inherent timescales. For this purpose, we will expand the figure caption accordingly:

  "Italic values refer to estimates based on literature data from different study periods (carbon burial and net primary production do not explicitly consider seasonality): *carbon burial in lakes, based on average sedimentation rate measured in 7 lakes in the Danube Delta with an organic carbon content range of 3 – 30 % (Begy et al., 2018), ** sink capacity of *phragmites australis* upscaled to the area covered by scripo-phragmitetum plant community (Zhou et al., 2009), *** upscaled net primary productivity of scripo-phragmitetum plant community (Sarbu, 2006)."

Line by line comments

Abstract: I miss some information about seasonal variations please provide standard deviations on flux numbers L21 & 22 L25, explicit what form of C is exported from the delta: is it OC or DIC?

L21 & L22: As our data is not normally distributed, we are reporting median concentrations for the CO_2 and CH_4 fluxes to the atmosphere from the different compartments. Providing standard deviations along with the median values would be inconsistent. In the revised version we will provide standard deviations for the overall annual fluxes of GHG including the ranges we obtain from calculations with the 25 and 75 percentile: "65 Gg C yr^{-1} (30–120 Gg C yr^{-1}, range calculated using 25–75 percentile of observed fluxes)"

L25: The number refers to the total export of carbon, i.e. the sum of OC and DIC. We add the standard deviations calculated using gaussian error propagation and define the kind of carbon exported: "In terms of lateral export, we estimate the net total export (DIC+DOC+POC) from the Danube Delta to the Black Sea to about 160 ± 280 GgC yr^{-1}, which only marginally increases the carbon load from the upstream river catchment (8490 ± 240 GgC yr^{-1}) by about 2 %. "

L43: the fact "carbon inputs from terrestrial ecosystems degas as CO_2 and CH_4 along the way to the ocean" is known for a long time, and not only from "recent estimates". In the introduction, it is important to cite pioneer papers and not refer all the time to very recent work that only confirmed the previous study, and do not provide any new information about the mentioned statement.

Thanks for the suggestion. While there are different valid approaches to cite recent or more classical literature, we will add a classical reference that already documents the CO_2 supersaturation in freshwater ecosystems: Stumm and Morgan (1981) and Cole et al. (2007)

L59: The statement "riparian wetlands in the Amazon basin have been identified as significant sources for the outgassing of terrestrial carbon in the form of CO2" Cite also Abril et al. 2014 here.

We will add this reference.

L62: "While wetlands are estimated to contribute 1.1 PgC yr-1 (Aufdenkampe et al., 2011) to the global carbon emissions, Amazonian wetland emission alone could contribute another 0.2 PgC yr-1 (Abril et al., 2014). Specifically, riparian systems in the lowlands could provide significant lateral carbon inputs (Sawakuchi et al., 2017)." These references to the literature are partially inappropriate. There is a confusion here between CO_2 outgassing from waters and CO_2 emissions from wetland ecosystem. Abril et al. proposed that central amazon wetland + river channel could be at equilibrium (zero flux), the flooded forest and marophytes being a sink and the open waters a source. Also, no need to be so precise on Amazon numbers in an introduction of a MS on the Danube delta.

Thank you for these critical remarks. In the revised version, we will restrict the discussion to estimates of the global contribution of wetlands to aquatic emissions and add a note of caution regarding the confounding factors:

"Global wetlands were estimated to contribute 1.1 PgC yr^{-1} (Aufdenkampe et al., 2011) to the carbon emissions in the land-ocean aquatic continuum. The uncertainty of these estimates is large, due to the difficulty to delineate global wetland areas (Tootchi et al., 2019) and the complex interaction between potential emissions and carbon uptake by vegetation and soils (Hastie et al., 2019)"

L68, mention that flooding has been recently described an important transport mechanism of terrestrial C to aquatic system, additional to drainage and surface runoff.

We will change this passage to the following wording and cite Abril and Borges (2019) here.

"Therefore, these deltas experience seasonal flooding, instead of (semi)-diurnal flooding determined by tidal action. Flooding can, in addition to groundwater drainage and surface runoff, transport substantial amounts of terrestrial carbon to aquatic systems (Abril & Borges, 2019). We thus anticipate seasonal variability in CO_2 and CH_4 emissions and in lateral carbon transport from the Danube Delta to the ocean. "

L162: "As tests showed that there was no significant difference between the lab- and field-based methods, we pooled the data in our analysis." I suggest you provide the result of these tests as a figure as supplementary material

We will add the following paragraph to the supplementary material, section 1:

"In October 2017, we conducted a comparison of CH_4 measurement procedures using the GC and the Los Gatos using field samples from the Danube Delta. We calculated average values for the lab-based GC procedure (n=2) and the field-based LG procedure (n=3). Considering the standard deviation of the samples, only 2 samples deviate from the 1:1 line, however they are still within the 10% measurement uncertainty of our GC system. Based on the results of this comparison, we deemed it appropriate to pool data acquired using the two different methods."

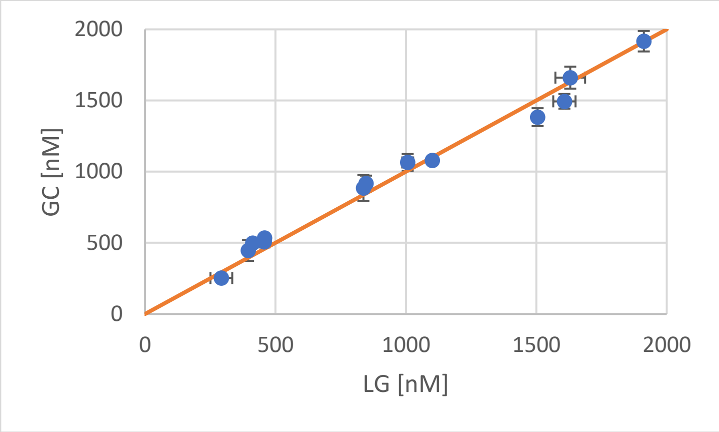

Figure 1 Average CH_4 concentration measured with lab-based GC method (n=2) versus field-based LG method (n=3). Error bars show the standard deviation, the orange line symbolizes the 1:1 line.

L184: "In the high-resolution LGR time series, the influence of gas bubbles could easily be identified." The method is described graphically in Grasset et al. Freshwater Biology. doi:10.1111/fwb.12780.

Thank you for the reference. Grasset et al. (2016), however, calculated the total flux as the diffusive flux determined from a linear regression plus the partial pressure increase during an ebullition event divided by the total observation time, i.e. $F_{tot} = F_{dif} + \frac{\Delta p_{ebu}}{t}$. In our study, we calculated the total flux as the difference between the initial and the final observed CH_4 partial pressure, i.e. $F_{tot} = \frac{p_{final} - p_{initial}}{t}$, a method that was for example also used by Beaulieu et al. (2016).

A visual representation of our approach is provided in the supplementary material and we will add a reference to Beaulieu et al. (2016).

L242 In section "2.6 Import by Danube River and Export to Black Sea", please provide here or maybe in the results section, more precise verbal information (equation is ok) on how you calculate C lateral fluxes before and after the wetland in the delta and ***how you deal with the problem that these two fluxes might be too close to each other to allow a precise calculation of the net lateral export from the wetland in the delta as a small differences between two large numbers that contain some uncertainty****. What are* ***the representativity of stations and data****, with respect to observed spatial and temporal differences in the C forms and discharge data between sampling points.*

"We calculated the lateral transfer of carbon between the Danube Delta and its River by subtracting the load exported to the Black Sea via the three main branches from the load imported to the Delta from the catchment:

$$F_{lateral} = F_{\mathrm{St1}} - (F_{\mathrm{St3}} + F_{\mathrm{St4}} + F_{\mathrm{St5}})$$

Station 1 is located in the Tulcea branch close to the apex of the delta and represents the water signature from the catchment, while stations 3, 4 and 5 are located in the 3 main branches close to the Black Sea. Stations 4 and 5 are located shortly upstream of the settlements of Sulina and St.George to avoid measuring the effect of these two settlements. Station 3 is located in a small side arm of the Chilia branch marking the border between Romania and Ukraine, which during comparison measurements showed the same water composition as the main branch.

The resulting lateral flux in our case is comparably small and we used gaussian error propagation to estimate its range. The basis were the measurement uncertainties in concentrations (0.5% DIC, 4% DOC, 10% POC) and discharge (3%, assumed), which were used to calculate the loads".

L290: figure 3 and next ones would be easier to read if simultaneous discharge could be shown

We agree and will add the discharge (daily average discharge close to the apex at Isaccea, also shown in Fig 2) as additional panel above right figure panel of Fig 4 (see below).

L350 and k values. Lake are more exposed to wind indeed, however rivers and channels are exposed to current which may also contribute to k_{600}

We are of the opinion, that the display of k_{600} is most meaningful with respect to the three different categories and therefore add this plot next to the seasonal plot of the discharge in Fig 4. See below for the upper section of the updated Fig. 4.

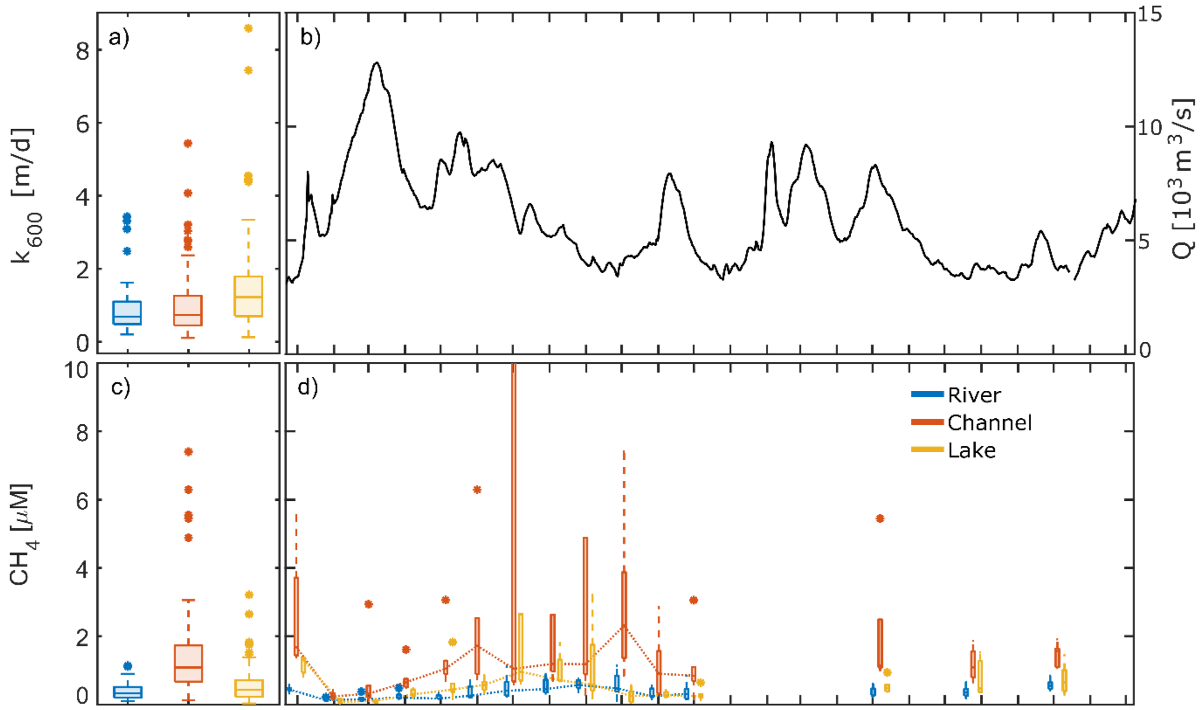

Figure 2 Figure 4 k_{600} (a), daily average discharge close to the apex (b) and measured concentrations of dissolved gases in the different waterscapes, i.e. river, channel and lake (a, c-h). Left panels (a, c, e, g): pooled data from 2-years. Right panel (d, f, h): seasonal dynamics with dotted lines connecting median values. X-axis ticks indicate day 15 of the respective month. c) & d) CH4 in 2016: four channel values (ranging from 22.2 to 58.0 µM) and one lake station (12.5 µM) exceeding 10 µM were cutoff. e) & f) dotted black line represents equilibrium concentration of CO2 at 15°C (18.2 µM). Boxplots indicate 25 and 75 percentiles, as well as median, whiskers indicate maximum and minimum, with data > 1.5*IQR is shown as outliers

L380 you are repeating what has been said in Mat and met about CH_4

Thank you, we will delete the respective sentence here.

L394. The calculation of lateral flux is indeed poorly constrained and it would be interesting to see the data that support the statements "POC import from the catchment exceeds the export to the Black Sea in February and March, while DOC import exceeds export only during August (data not shown)."

We will add this data in the form of a Figure to the appendix and rephrase the paragraph as following:

"The water export from the delta, however, is poorly constrained. The balance between precipitation minus evaporation is negative, poorly quantified and quite variable. We therefore rely on the flux balance of the three branches to estimate carbon export from the delta. The resulting export to the Black Sea via the Danube's main branches amounts to 8650 ± 147 GgC yr^{-1} and is less than 2 % higher than the inflow load reaching the apex of delta. It mainly relates to increased DOC levels reaching the main branches from the delta, especially during the spring flood in March and April. The relatively small fraction of water that passes through the delta changes the relative fraction of DOC and POC only marginally to 7 % and 4 %, respectively, while the largest fraction in the water reaching the Black Sea remains DIC (89 %, Fig. 8). DIC import and export is fairly comparable throughout the year, while POC export to the Black Sea strongly exceeded the imports from the catchment in April. DOC exports are highest in the first half of the year (see Fig Sxy)."

Supplementary Information:

"3.x Seasonality of carbon import and export

Import and Export loads varied seasonally, which was to a large extent driven by variations in discharge (Fig. Sxy).

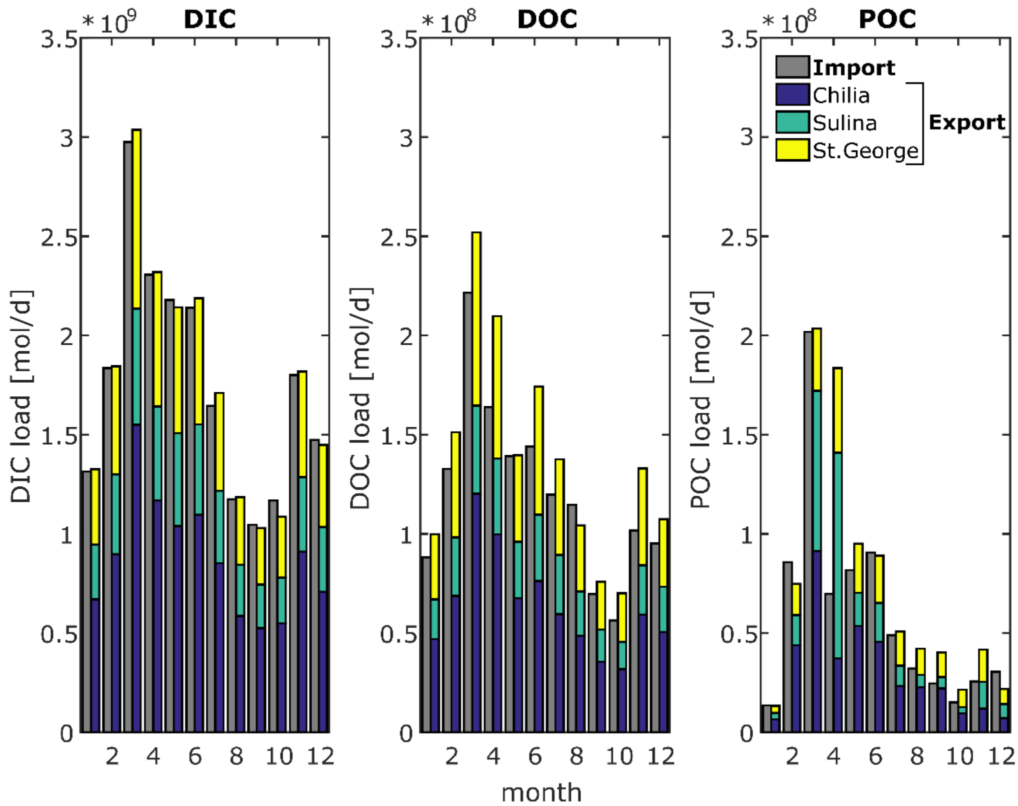

Figure Sxy: Carbon import to the delta and export to the Black Sea (sum of Chilia, Sulina and St. George branch) in the forms of DIC, DOC and POC loads. Please observe the different order of magnitude for DIC."

L400-410, please simplify the story about statistics.

The simplified version will read as follows:

"The non-parametric Kruskal-Wallis test does not require normal distribution of the data, but it requires equal variance of the data groups investigated for difference in median (Hedderich & Sachs, 2016). Our observations in the seasonal plots (Fig. 3 & 4) support the results of the test: in most cases, the boxplots do not overlap, indicating that the three groups are significantly different. For example, DOC is significantly higher in the delta lakes and channels due to the strong primary productivity of these systems. O_2 is significantly lower in the channels than in the other two categories due to lateral inflow of oxygen-depleted waters from the wetland (Zuijdgeest et al., 2016; Zurbrügg et al., 2012). The large difference between the waterscapes with respect to CO_2 and CH_4 fluxes supports our approach to treat the waterscapes independently when upscaling the flux measurements to the total water surface of the delta."

L442: what about summer stratification in lakes?

Unfortunately, we did not measure depth profiles and I couldn't find anything on summer stratification in Danube Delta lakes. The shallow character of most investigated lakes (maximum depth of 3.5 m in Lake Rosu) led us to speculate that the lakes may be fully mixed or maybe stratified during individual days in summer. Since we don't have any prove for either condition, we did not speculate about it at this point.

L445: the study of CH_4 emissions from various plant types by Grasset et al 2016 might be helpful here

Thank you for the reference. We plan to adapt this paragraph as follows:

"In the lakes, longer residence times of 10–30 days allow primary production and local decomposition of organic matter to become important factors driving carbon cycling. We observed abundant macrophytes like *Ceratophyllum demersum* and *Elodea canadensis* growing in spring and early summer, which, depending on lake depth, even reached the lake water surface. A change in abundance of submerged vegetation to vegetation with floating leaves might be linked to changes in the CO_2 and CH_4 fluxes (Grasset et al., 2016). Around July, algal blooms coincided with a significant reduction in macrophyte abundance. This pattern seems to be reoccurring due to the eutrophic state of the delta lakes (Coops et al., 2008; Coops et al., 1999; Tudorancea & Tudorancea, 2006). During our observations, both macrophytes and algal blooms caused a drawdown of CO_2 and supersaturation in O_2 (Fig. 4d & 4f). The algal blooms also partly explain the peak in measured POC from July to November, which extended to most of the delta's channels (Fig. 3d). The degradation of the macrophyte biomass coincided with locally elevated CH_4 concentrations from July to October (Fig. 4b).

L584 drylands are not defined as the contrary of wetlands. The difference here is between floodable land and non-floodable land

Yes, thank you. We will correct this:

"Abril and Borges (2019) recently suggested that the active pipe concept of carbon transport in the aquatic continuum indeed needs to be extended to consider floodable and non-floodable land as separate carbon sources."

L594 "Assuming a carbon content of 0.42 gC gBiomass-1 determined by Greenway and Woolley (1999) for Phragmites australis, between 1000 and 1210 GgC yr^{-1} are bound in the form of macrophyte biomass in the reed area of the Danube Delta". Not clear the meaning of "bound" here. A great portion of the macrophyte biomass is supposed to be recycled. "The wetlands thus hold 12 to 17 times the total input of organic C to the delta from the catchment" do you mean the total ANNUAL input?

The idea was to compare input of organic carbon from the catchment with the primary production from the vegetation. The annual primary production of the reed is between 1000-1210 GgC yr^{-1}, while we estimate the total annual input of organic carbon from the catchment to 79 GgC yr^{-1} (i.e. 10% of the organic carbon load transported by the river). You are right, the term "bound" is ambiguous in this context, since we are referring to the reed biomass and not the carbon stored in the sediment. Please see comment L600

L595 "Nevertheless, wetlands are considered to be net C sinks and Zhou et al. (2009) estimate the sink capacity of a Phragmites australis dominated wetland to -62 gC m^{-2} yr^{-1} considering CO_2 and CH_4 release from the wetland itself." You must be more precise here in the vocabulary used. CO_2 sink might be different from C sink. Please specify that C sink is OC burial in sediments and not the atmospheric CO_2 sink. Same in the following sentences.

Thank you, we will correct the imprecise wording, see comment L600.

L600 “Assuming that carbon emitted from the channels originated only from the wetland source, this would suggest that up to 20 % of the potential wetland sink might be exported laterally, eventually finding its way to the atmosphere.” Please explain more clearly

Since the comments L594, L595 and L600 all concern the same paragraph and are somehow linked to each other, we present the planned changes as follows:

“The Danube Delta is dominated by the plant association Scirpo-Phragmitetum, which covers nearly 89% of the total marsh area (1600 km^2). Its net primary productivity ranges between 1500–1800 g m^{-2} yr^{-1} (Sarbu, 2006), which is slightly higher than the average net primary productivity of intertidal salt marshes and mangroves (1275 gC m^{-2} yr^{-1}) (Cai, 2011). Assuming a carbon content of 0.42 gC $gBiomass^{-1}$ determined by Greenway and Woolley (1999) for *Phragmites australis*, primary production in the reed amounts to 1000–1210 GgC yr^{-1} (Fig. 8), which is about 8 times less than the carbon load transported by the river. A large fraction of the net carbon assimilation by the *phragmites* stands is decomposed and released back to the atmosphere. In a Danish wetland, more than 50 % of the carbon was respired and released back to the atmosphere, with 48 % being released as CO_2 and 4 % as CH_4 (Brix et al., 2001). In the Danube Delta, the 50 % accretion rate would correspond to about 500 gC m^{-2} yr^{-1}. However, net primary production and carbon accretion change seasonally with environmental factors such as temperature and irradiation. Accordingly, net CO_2 assimilation in the Danish study was limited to the warm season from Arpil to September, whereas CO_2 and CH_4 emission occurred during the whole year but with maxima of 0.2 mol Cm^{-2} d^{-1} during July-August. Qualitatively, we observed the same seasonality in CO_2 oversaturation in the Channels that drain water from the *Phragmites* stands (Fig. 4d).

For a *phragmites australis* dominated wetland in China, at a latitude comparable to the Danube Delta, Zhou et al. (2009) estimated the annual net uptake of CO_2 to 62 gC m^{-2} yr^{-1}. Scaled to the area of the Danube Delta, this would result in 99 GgC yr^{-1} remaining in the delta, which is in the same order of magnitude as the total annual input of organic C from the catchment (79 GgC yr^{-1}). Similar to the Danish study, also Zhou et al. (2009) do not account for potential lateral transport of carbon to adjacent water bodies. Our results show that channels in the Danube Delta are receiving carbon from the wetland, with peaks in CO_2 and CH_4 concentrations that match the maxima in the gross ecosystem production in China. Comparing the estimated carbon fluxes from the channels with the yearly carbon accumulation estimates of the wetland suggests that up to 20% of the latter could be released to the atmosphere via lateral transport, assuming the carbon flux from the channel were exclusively sustained by the wetland. With a lag phase of about 3 months, the Danube Delta reed beds release peak concentrations of DOC and POC during October to November, when the biomass in the reed stands start degrading (Figure 3 d,e).”

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