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Interactive comment on "Methane in the Danube Delta: The importance of spatial patterns and diel cycles for atmospheric emission estimates" *by* Anna Canning et al.

Anonymous Referee #1

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General comments

Overall, the study is an important contribution into methane dynamics in highly heterogenous system of river delta. It also emphasizes important (and often neglected) aspect of diel variability of CH4 dynamics. I see this study as an ambitious project as it aims to examine spatial and temporal variability of CH4 in various water bodies including river, side channels and lakes. It is important that authors distinguish between different aquatic systems and focus on more refined delta variability (river, channels, lakes) as these systems may experience distinct and highly variable CH4 dynamics. Also, this study not only aims to elucidate high spatial variability in the above subsys-

tems of delta, but also adds additional layer of high temporal variability (diel) of CH4. Such study design allows to obtain more comprehensive picture of CH4 fluxes from such delta systems. In general, from reading the abstract and introduction I had an understanding of the study design and the rationale behind the investigations.

However, I see few substantial points which should be addressed before the manuscript can be published. These points are addressed in specific comments. Briefly, my major concerns are:

1. Gas transfer model used to calculate fluxes. As river delta is highly heterogenous system I believe that Cole and Caraco model is not the best choice to derive fluxes from measured concentrations. In the specific comments I propose the alternative gas transfer model, which could be more suitable in this system.

2. Although diel study is very important aspect here, extrapolation and recommendations based on just two full diel studies is certainly not enough to suggest how to design sampling campaign (time of samplings to reduce bias).

3. CO2 variability is certainly important and worth to study, however this manuscript focuses mostly on CH4. There is no information, which would introduce the reader the CO2 dynamics and in general CO2 appears in the text only fragmentary. I would suggest that spatial and temporal CO2 variability has a potential for separate manuscript.

4. There are numerous repetitions, especially regarding results. Values of concentrations and fluxes appear in tables and they are also described in the text, which is redundant.

5. Language is occasionally missing fluency and precision. Some sentences are too composite and reader gets lost, so their structure could be simplified to assure nicer reading of this interesting study.

Specific Comments

Abstract Overall abstract is understandable; however it is missing flow in reading occasionally. Below I include several specific comments. However, if authors decide to implement other of my suggested comments (results and discussion) then the abstract would require modifications as well.

Line 7: please add how many days in total? "Over 3 seasons" is misleading and suggests that the concentrations were measured continuously for 3 seasons which is not the case.

Line 14-15: Did authors measured temperature profile to conclude there was a stratification? If not and this is an assumption (may be correct), it is not a result of the study

Line 15 – 16: Please correct this sentence, it doesn't have correct English structure.

Introduction:

I suggest to shorten introduction slightly. Especially selected parts (please see comments below) should be removed as they provide details which rather distract the reader from the main path of the manuscript instead of leading to the main issue. Instead I would suggest that authors elaborate slightly more on diel variability as this is main topic of the paper.

Line 29: It is not clear:, please specify "Potential driver" of what ?

Line 37 : substitute "although" with "yet"

Line 39-40: repetition "covering" and "covered", please change

Line 39: "give a range", just "range" is enough

Line 43-44: Do authors aim to assess the role of rivers and channels in methane emissions, not the role of methane?

Line 46-50: In my opinion this part is redundant as the methanogenesis or methanogenesis related processes are not a topic of this paper. According to me it deviates the

reader from the main (interesting) story.

Figure 1. This scheme is nice and illustrative but rather redundant in the context of this manuscript. This is rather text book knowledge and this scheme does not illustrate neither findings of the paper or concept of the study. In my opinion this scheme is not necessary.

Line 63: Please provide reference for this statement

Line 64 - 65: Although I see how browning or warming could affect seasonal variability of CH4, I cant see how they would impact diel variability?

Line 67-68: Real strength of this study: high spatial and temporal variability in different systems

Line 73: Please specify what is "extremely high resolution" 10 times a day, every hour, every minute. . .?

Methods:

Set up: As a reader not specifically familiar with this type of set up, I would appreciate more details. As far as I know this type of setup uses membrane-based equilibrators? I think this information should be in the method description so the reader obtains information about principle of the method. Also, equlibrators may underestimate concentrations (especially CH4), it would be highly appreciated if authors acknowledge drawbacks of this specific method. I do not try undermine this method and I am aware that every single method has its pros and cons, but for the reader it would be beneficial to know (very briefly) to know its drawback (and benefits too).

Study area is described sufficiently, however I am missing specific information how many km2 were covered regarding each system (lakes, channels, river)?

Line 109: Wind speeds may be locally different from what was occurring at the delta therefore its important how far from measurement sites Gorgova weather station was located?

Line 110: I encourage authors to use reference Wannikhof 2014 instead of Wannikhof 1992

Line 113: I think its not correct to disregard stream velocities, especially without information about their values? What are the stream velocities (range) in this system?

Without this information it is difficult to assess if Cole and Caraco is the most suitable gas transfer model used. Please also see my comments below.

Line 114: Since authors didn't measure fluxes directly but derived them from the concentrations, it is very important to assure that authors apply gas transfer model which is the most suitable for measured sites and conditions. Cole and Caraco was developed for lakes mostly and doesn't include impact of variable hydrological settings in delta system, mainly flow velocity, which has an impact on fluxes too. Therefore, I would suggest to calculate fluxes using gas transfer model, including both variables (wind and flow velocity), which have an impact on k in such dynamic system. As there is no "ideal" model, I would still suggest to rather use the way k was derived in "Borges et al 2004" (as I see in reference list authors are already familiar with this publication) than Cole and Caraco gas transfer model.

Line 122: application of different model will have an impact on n value too

Results and discussion: As I already wrote in the General comments above, there are numerous places where results (values) are repeated. This is redundant. If value is already presented in the table, there is no need to repeat it in the text. It allows to keep the manuscript more concise. Also, regarding reporting values, I suggest to use mikromol L-1 instead of nmol L-1 all throughout the text.

Line 147: Please rephrase the sentence, oxygen by itself cant be undersaturated, lake water can be.

Line 152: "some variability ultimately due to temperature and wind": Currently I cant

see a support for this claim in the presented data. Where do the data show that flux variability was due to the temperature and wind?

Line 154: What do authors mean by "outgassing flux density"? Do you mean "mean flux"?

Line 154-156: If such extrapolation exercise is done, then it should rather be described in the method section. Thus, I suggest to move it to method section and in this section present only obtained results.

Also, I think it is important that authors measured and presented separate values for different types of water bodies in the delta. Its highly appreciated since such information is less common and shows that even it is one system, CH4 variability in water bodies located in close proximity to each other can be high. That is why I do not see the reason to pool all the data together and just derive one mean for the whole delta. To include flux variability within such complex system I suggest to (at least) calculate mean flux for each type of water body (channel, river, lake), calculate how much of area is occupied by each type of waters/wetlands and then extrapolate and derive total flux per year.

Line 162- 168: Good point, it is appreciated that authors acknowledge and discuss importance of ebullition in this system

Line 169 – 170: I am not familiar with specific set up of Maier et al 2020, but I do not agree that the method used by authors in the manuscript and chamber method cannot be compared at all. Sure, such high variability is missed by chambers, but at least fluxes at the same depths and same systems obtained by both studies could be compared.

Line 173: I agree that its not possible to assess impact of environmental drivers (such as wind) if it wasn't measured in situ. However, if authors suggest that distribution patterns were rather driven by biological or physical factors it should be supported by the data. And I'm not sure if I see such evidence. Plus, which biological and physical factors do authors mean? It would be important to be more specific.

Line 181: Flooding is an important aspect in a frame of seasonality. As I imagine such system experiences reoccurring flooding, which may have big impact on CH4 fluxes (see for example Gatland et al 2014, JGR). To obtain more comprehensive picture on CH4 seasonality in delta system, it would be important to acknowledge flooding phenomena in a bit more detail.

Line 185-187: Please rewrite this sentence, I'm not sure what authors meant here.

Line 189-190: The sentence is structured like authors actually measured decomposition of OM and subsequent CH4 liberation. If authors didn't do such measurements, I would suggest to change it to:"....of biodegradation of OM could have triggered...which could be responsible for...."

Line 192: Please change to "may explain" or "could explain"

Line 193-194: Not only lakes are sources of labile organic matter, also rivers and channels too. Please provide reference

Line 195 - 196: "Channels.......in O2": If authors make such statement, reference would be appreciated or these are results of this study? It is not clear.

Line 201: Why such conductivity points into groundwater influence?

Line 199 - 203 : Authors raise here the potential impact of groundwater on 'hot spot' conductivity. Also, the impact of groundwater on surface water CH4 concentrations may be very important too (especially in wetlands). So, if authors raise here importance of groundwater, then its potential impact on CH4 in this system should be acknowledged as well. Of course, one cant measure everything and I wouldn't expect the actual results but one or two sentences about potential impact of 'hot spot' groundwater on surface water CH4 in this system.

Line 202: Please clarify. What do authors mean by "water dropping further into the 'hot spot' ?

Line 206: In general the word "significant" is usually used in a statistical context, which is not the case here. I would suggest using other word

Line 211-222: I agree that this may be the case that more production happens in the delta, not in the river, and that such hot spots may impact the system's concentrations. Thus, this paragraph is important, however I have difficulty to follow the way it is currently presented, I guess because many values are included in the text. I would suggest maybe to draw simple conceptual figure, which could illustrate possible impact of channels and hot spot on a system. With colors representing concentrations? It would be easier to grab this interesting concept. If authors do not wish to illustrate this idea and stick to written text, I would suggest to rewrite it so its easier to follow.

Line 216- 229 Many values described in this paragraph are already in the table 1, so there is no need to write and describe all the values here again. It is unnecessary repetition and can be removed.

Situation of CH4 in lakes

I suggest changing the paragraph header to "CH4 concentrations and fluxes in lakes" or "CH4 dynamics in lakes" Many values described in this paragraph are already in the table 1, so there is no need to write and describe all the values here again. The discussion of obtained results is enough.

Line 254: What do authors mean by "averaging" the lakes? Is it average of fluxes, of total fluxes, from all lakes? Its unclear from the text.

Line 260: What authors mean by "intermediate" sampling ?

Diel CH4 cycling

Line 269 – 270: This sentence is not clear, requires clarification what authors intended to say here?

Line 266: Please indicate the letters in Fig. 4 instead of “left”

Line 273-275: Do authors mean here convective mixing? If yes, its good to mention this term here

Line 284- 310 As I wrote in the general comments, CO2 deserves its own story. Including it suddenly into the manuscript distracts the reader from the main story, which as my understanding is, covers almost solely CH4. Also, so far the whole text above included only CH4 patterns. Additionally, this section has a title which points to CH4 daily patterns only. I suggest to remove CO2 dynamics from the manuscript.

Figure. 4: Nice and informative figures.

Line 326 – 327: I think authors mean here “day light data”, not “day night data” ? Please clarify.

Line 334-354: It is a valid and very good point that authors’ aim is to emphasize importance of diel variability and bias which occurs due to spot sampling. This may be one of the reasons for bias in current CH4 estimates or upscales. However, I would avoid giving recommendations how to minimize bias during lake CH4 sampling based on 2 full diel cycles. I see this manuscript as important work to bring attention into neglected issue of CH4 diel cycle in delta system, but to draw firm conclusions and give sampling recommendations, more full diel surveys would be necessary. Also, it is important to acknowledge that these two diel cycles captured by this study could be a snapshot as well. Thus, if being measured another day (or season), the opposite situation could have occurred with higher late daytime fluxes compared to the nigh time (for example, see Sieczko et al 2020 and references therein).

Table 2. I propose to visualize the table in a form of figure. It will be easier for the reader to see more clear the impact of day light (DL) vs. full diel cycle (FD) sampling on CH4 flux and concentration.

Line 349-350: The cited studies actually acknowledge and emphasize existence of CH4 diel cycle. They do not undermine it as this sentence suggests. The sentence or the references requires modification.

Line 379: Did authors measure organic carbon and showed that it was derived from macrophytes?

Technical Comments Line 170: please change “location” to “locations” Line 177: Please change “decease” to “decrease”

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