

## #Reviewer 1

This manuscript presents a year-long dataset of GHG concentrations and fluxes from 5 headwater catchments in Germany from streams, agricultural ditches, and WWTP outflows. It identifies controls on GHG dynamics using mixed-effects models and structural equation models. In addition, it upscales flux rates to calculate annual emissions in terms of global warming potential. The main finding, that anthropogenically impacted streams have higher and more variable GHG concentrations and fluxes, was well supported. Overall, the manuscript presents results that will be an important contribution to our understanding of GHG emissions from inland waters and I find the analysis and results novel and worthy of publication.

I have a few suggestions, although they mostly minor and easy to address.

Abstract:

-I don't think that the analysis backs up the statements about separating in situ production of GHGs and direct inputs of GHGs (e.g., In 27-28, 30-31). These statements should be removed or rephrased.

**Response:** Thank you for your critical comment and suggestion. We removed the initial sentences and rephrased them to clarify our meaning.

"Our findings also suggested that nutrient, labile-carbon, and dissolved GHG inputs from the agricultural and settlement areas may have supported these hotspots and hot-moments of fluvial GHG emissions."

-I think the authors should more clearly state that anthropogenically impacted streams have not only higher, but also \*more variable\* GHG emissions than natural streams in the abstract. (i.e., give some sort of variability stats)

**Response:** Thank you for the suggestion. We have added a sentence to the abstract to represent this finding better.

"Streams in agricultural-dominated catchments or with wastewater inflows had up to 10 times higher CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O emissions, which were also more temporally variable (CV > 55%) than forested streams."

-I would consider mentioning some of the other main findings in the abstract (if possible within word count limits): 1) the break down of the expected stream-order patterns in impacted sites and 2) the finding that CO<sub>2</sub> is the dominant contributor in terms of global warming potential

**Response:** Thank you for the suggestion. We have added several sentences to reflect both findings in the abstract.

"Overall, the annual emission from anthropogenic-influenced streams in CO<sub>2</sub>-equivalents was up to 20 times higher (~71 kg CO<sub>2</sub> m<sup>-2</sup> yr<sup>-1</sup>) than from natural streams (~3 kg CO<sub>2</sub> m<sup>-2</sup> yr<sup>-1</sup>), with CO<sub>2</sub> fluxes accounting for up to 81 % of the annual emissions, while N<sub>2</sub>O and CH<sub>4</sub> accounted for up to 18 and 7 %, respectively. The positive influence of anthropogenic activities on fluvial GHG emissions also resulted in a breakdown of the expected declining trends of fluvial GHG emissions with stream size. Therefore, future studies should focus on anthropogenically perturbed streams, as their GHG emissions

are much more variable in space and time and can potentially introduce the largest uncertainties to fluvial GHG estimates"

Methods:

-I don't see temperature/seasonality or NH<sub>4</sub> in the SEM results, even though these parameters are listed as input variables. Were they found insignificant and dropped? Please clarify.

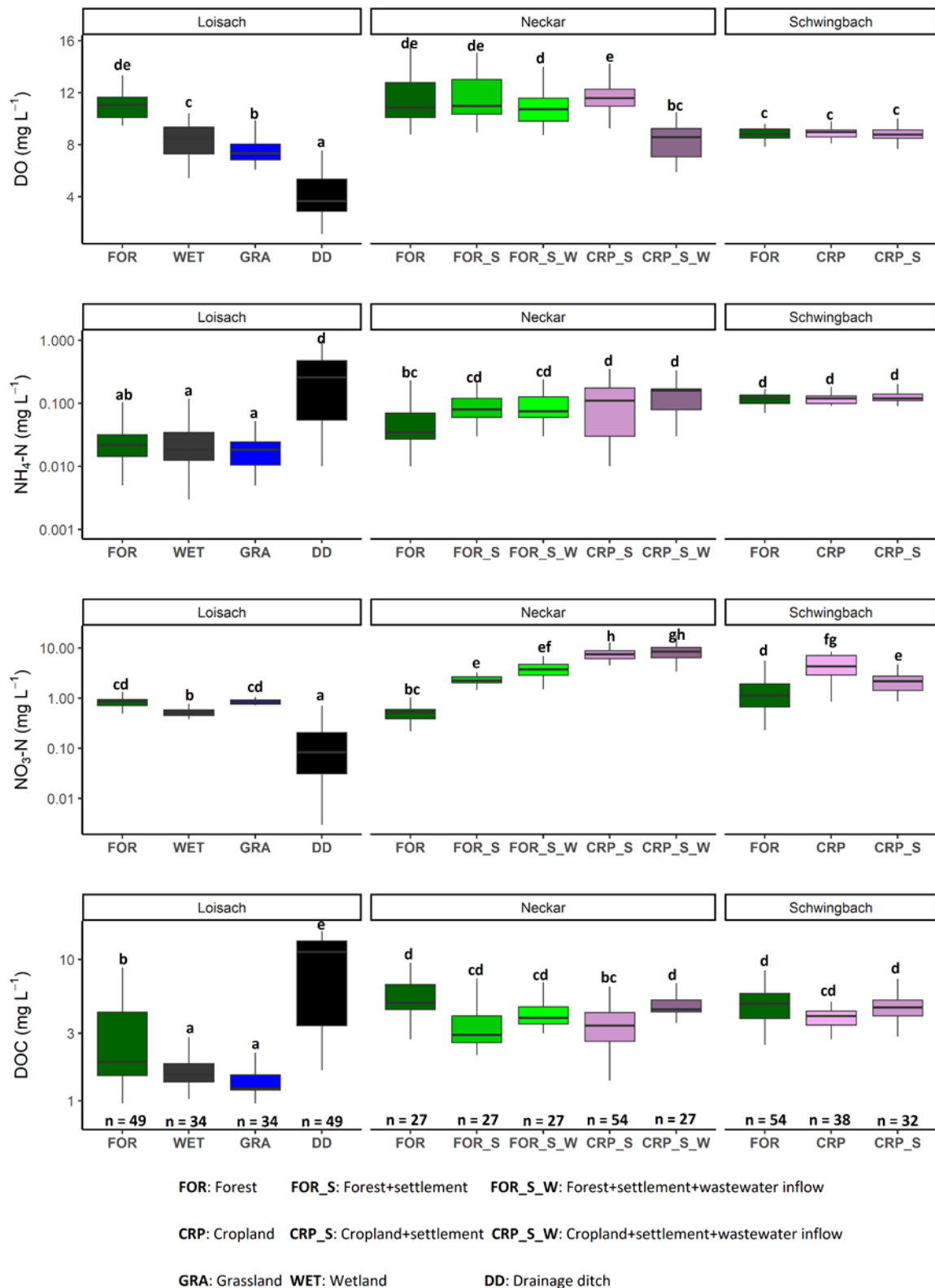
**Response:** Thank you for your question. Temperature and NH<sub>4</sub> were removed from the SEMs as they were insignificant. We have now clarified this in the results section.

"In contrast to all other variables, water temperature and NH<sub>4</sub>-N mg L<sup>-1</sup> did not contribute significantly (p-value>0.05) to the variance explained by the best-fit SEMs and were removed from the final path analyses (Table B4)."

Results:

F2. Consider using colors to represent major land-use classifications and shades to differentiate the sub-classifications. For example, crop, crop + settlement, and crop + settlement + WW inflow could be given different shades of the same color. Also, yellow is somewhat difficult to see on all these plots.

**Response:** Thank you for the suggestion. We have edited all the colors in our plots to reflect this suggestion.



I don't have the background to fully assess how the SEM analysis was applied but it seems to make sense and F5 is great.

**Response:** Thank you for the compliment. We also found it practical in explaining how multivariate drivers interact to drive the intra-annual trends in GHG concentrations.

I find the conclusion that typical stream order patterns break down in anthropogenically impacted streams interesting (L573-584). However, I don't see the data presented in the results section. Please include it here (and perhaps add to the abstract as well).

**Response:** Thank you for the observation. We have added this information in the results section and also in the abstract.

## Abstract

"The positive influence of anthropogenic activities on fluvial GHG emissions also resulted in a breakdown of the expected declining trends of fluvial GHG emissions with stream size."

## Results

" In addition to land use effects, we also examined spatial variability in the GHG concentrations and fluxes linked to stream order differences. We found tendencies of higher CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O concentrations and fluxes with increasing stream orders in the Schwingbach and Neckar catchments dominated by croplands and settlement areas. In contrast to the Neckar and Schwingbach catchments, GHG concentrations and fluxes in the more natural Loisach catchment decreased with stream order (Fig. A4). Comparing across catchments, higher stream orders (5&6) in the human-influenced Neckar catchment had higher or comparable GHG concentrations and fluxes than lower stream orders (1–3) in the Schwingbach and Loisach catchments(Fig. A4). "

Discussion:

Consider discussing the result that CO<sub>2</sub> was the main contributor when emissions of all three gases are converted to CO<sub>2</sub> equivalences (F6). I found this result to be interesting and perhaps it deserves more attention in the manuscript.

**Response:** Thank you for the observation. We have added a sentence at the beginning of the discussion to indicate this finding and further expanded on the fact that an increase in upstream human activities increased the contributions of the CH<sub>4</sub> and N<sub>2</sub>O relative to CO<sub>2</sub>.

"In agreement with previous studies, CO<sub>2</sub> accounted for most (>81 %) of the annual fluvial fluxes in CO<sub>2</sub> equivalents (e.g., Marescaux et al., 2018; Mwanake et al., 2022; Li et al., 2021). However, the presence of upstream agricultural and settlement areas seemed to alter these trends by reducing the contribution of CO<sub>2</sub> and increasing N<sub>2</sub>O and CH<sub>4</sub> contributions. The effects of the above anthropogenic activities on aquatic GHG dynamics were twofold. Drainage ditches were landscape hotspots for CH<sub>4</sub> emissions, while increasing upstream agricultural and settlement areas resulted in fluvial N<sub>2</sub>O hotspots."

Conclusion:

L620 - It seems like CH<sub>4</sub> is also higher in the anthropogenically impacted sites?

**Response:** Thank you for the observation. Yes. Methane fluxes were also higher in human-influenced streams. We have edited the conclusion to reflect this view.

"Streams and ditches in agricultural and settlement areas were characterized by significantly higher GHG fluxes with more significant intra-annual variabilities than forests and wetlands. A combination of wastewater inflows and agricultural land use resulted in the highest fluvial CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O fluxes, particularly during high discharge periods with substantial external dissolved GHGs."

Minor:

L40 – "contributors to global greenhouse gas budgets" or "contributors of greenhouse gases"

**Response:** Thank you for the observation. Rivers are contributors to global greenhouse gas budgets. We have edited the statement to make it clearer.

"Streams and rivers cover only a small fraction of the earth's land surface (0.4%; Allen et al., 2018), yet they are significant contributors to global greenhouse (CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O) budgets, emitting approximately 7.6 (6.1–9.1) Pg-CO<sub>2</sub> equivalent into the atmosphere per year. (Li et al., 2021)."

L41 – Li et al., 2021 citation only refers to headwater streams

**Response:** Thank you for the question. The study also included the most recent cumulative GHG emissions from headwaters and large rivers, and that's why we used it to get an idea of the contributions of fluvial GHG fluxes to global budgets.

L52 – "in situ N<sub>2</sub>O production"

**Response:** Thank you for the observation. We have made the change in the text to make the sentence clearer.

"Elevated hydrological inputs of dissolved GHGs, nutrients, and labile carbon to streams from fertilized croplands have been shown to increase their N<sub>2</sub>O (e.g., Beaulieu et al., 2009), CO<sub>2</sub> (e.g., Bodmer et al., 2016; Borges et al., 2018), and CH<sub>4</sub> fluxes (e.g., Mwanake et al., 2022), by favoring *instream* GHG production processes and also ensuring steady supplies in periods of low in-situ biogeochemical production."

L70 – "where"

**Response:** Redone.

"Similar findings were also found in urban-impacted rivers in China, where their GHG emissions were up to 14 times higher than those in other land uses (Zhang et al., 2021)."

L260 – "May 31"

"Fixed effects in the models consisted of land use classes in each catchment (Table 1) and seasons: summer June 1–August 31, autumn September 1–November 30, winter December 1–February 28, and spring March 1–May 31."

L369 – consider using uatm for N<sub>2</sub>O, instead of natm, as it is more commonly used

**Response:** Thank you for the suggestion. We agree that uatm data units are commonly used. However, we used nano (uatm/1000) to represent better the low N<sub>2</sub>O concentrations measured in the forests and drainage ditches.

L375-376 - It looks like negative fluxes are actually presented in F3.

**Response:** Thank you for the observation. We indeed had negative fluxes. However, they were only limited throughout the year, hence the use of the word "mostly" to represent the majority of the fluxes, which were positive and dominated the annual emissions