Dear Reviewer,

Thank you so much for your valuable comments, and the positive assessment of our paper. Please find below a preliminary answer to your comments.

Best regards,

Paolo Peruzzo (on behalf of all authors)

The article by Peruzzo et al explores the gas exchange dynamics in a step-and-pool situation, which is a common geomorphic feature in many streams with some relieve like mountain streams. To do this they use a lab setup, with a unique monitoring set that measures water velocity and turbulence in multiple directions and at a high spatial and temporal resolution. This is something I have rarely seen so the authors can explore experimentally at a great resolution the drivers of gas exchange in waters. They can also separate bubble mediated gas exchange which is important ad often ignored.

Thank you for the nice summary of our work.

They find a high spatial in the gas exchange, both turbulence and bubble mediated, with a very small area accounting for most of the gas exchange with the atmosphere. The article is well written and figures of high quality…

Thank you for the positive assessment of our manuscript.

…but the technical level of some of the section may be hard to follow for not only the average reader of "Biogeosciences", but also for someone who works with gas exchange in rivers without a hydrologic engineering background like me. With this I provide some suggestion to clarify some terms, notations, and provide citations on those technical aspects of the work.

Thank you for your comment. We will work to improve the readability for the average readership of Biogeosciences starting from your suggestions. In particular we will revise Section 2.3 including additional references and explaining the most technical aspects related to the hydrodynamics of the step and pool by a plain language.

As said, the work is of high quality, but I do have a major issue with the take home message of the article, which can be summarised with the last sentence of the discussion: "Based on the above arguments, we propose that the use of the mass transfer rate, k, should be dismissed in cases in which the heterogeneity of the flow field controls the fraction of mass evaded into the atmosphere, as in our step-and-pool configuration." I fully agree that gas transfer velocities are tricky to measure, highly variable in space and tricky to translate from one spatial scale to another.

Thank you for the comment and the general agreement on the main point of the paper.

There are also multiple methodologies for different use cases, see the review by Hall and Ulseth 2020 (WIRES) for a great overview.

The reference is already included in the reference list.

In this case, a reach-scale measure of the gas transfer velocity using a gas tracer, where a inert gas is injected upstream and the loss is quantified downstream with reaches of 50-150 meters for example, is a perfectly valid method to quantify gas exchange in a stream with steps and pools. Spatial aggregation is indeed important, and how those reach scale measurements are translated to catchment scales remain an unanswered issue for instance. I will agree with the authors that for example, using chambers in a step and pool system is not a good method, but their claim is a bit overreaching.

The adoption of inert gas can allow us to quantify the whole gas exchange in a given portion of a watercourse, but not the internal patterns; however, the estimated k values do not necessarily depend on the mean physical quantities defining the reference reach (slope, velocity etc) because they are mostly related to the specific topography of the reach. Likely, we can upscale gas exchange in heterogeneous streams, as in those characterized by steps and pools, only by separating the gas evasion contributions deriving from characteristic geomorphic elements (steps, segments, cascades, etc), as shown in Botter et al. (2022). The estimation of the average value of k by the gas tracer in a 50-150 m of reach cannot allow one to recognize the role of these elements and, consequently, properly upscale and extrapolate the direct experimental measure. In the revised version of the paper we will be more cautious in proposing that k should be dismissed, and we will explain the main issues associated with methods that do not resolve the internal heterogeneity of reaches. We feel we have to thank the referee for his/her comment.

I will conclude this with an analogy to another physical and turbulent system. Temperature of a fluid is some kind of emergent property, which is related to the average movement of all molecules. You could use some great technology to track and quantify the movement of all particles, only to realise that the system is highly chaotic and heterogenous with a lot of eddies, indicating that is rally hard to quantify the movement of particles. Regardless, at a larger scale we have some other tools to estimate temperature of that system at larger scales that may be a simplification but work well enough. This study is a bit similar in this sense, it provides unique insight of the fine scale turbulent dynamics of water, suggests that is highly heterogeneous, but the link to the larger reach scale is a bit weak that may need some improvement or toning down in the text.

Thank you for your comment, which offers an interesting starting point for reflection. We think the analogy of the temperature can be useful to explain our point of view. Your preamble is correct but your reasoning does not directly face the question: "What is my temperature data for?". Whether the process of interest is linearly dependent on T, the use of this "average" of the molecules' movements works very well. On the contrary, non-linear processes involving the dynamics of the molecules are not captured by the averaged quantities. The gas exchanged between water and the atmosphere belongs to this second case, as the gas exchange and the mass transfer rate are non linear functions of the dissipated energy (see Botter et al., 2021). Of course, in practice, a perfect knowledge of the k patterns (or of the molecules' movements) is hardly feasible, and some average quantities are mandatory. However, this procedure determines small uncertainties only in nearly-homogenous conditions, i.e., when the k has the same order of magnitude along the whole study reach. This is not the case for streams with step-and-pools where k may vary by some order of magnitude, and in which we need to isolate the contribution of k owing to the jet at a scale not detectable by in-field instruments..

However, the final statement of the paper can be toned down, and the criticisms of the adoption of k at the reach scale in high-energy streams will be better substantiated in the revised version of the paper.

Below I detail some minor issues I found in the text:

Abstract

-L10: maybe is cleaner to put k in parenthesis?

We will edit the abstract accordingly.

Introduction

In the first and second paragraph, the authors focus very fast on mountain streams. Step and pools are common in other landscapes outside mountains, so maybe it would help rising the generality of the article to discuss this more broadly outside mountain streams.

We will revise the mentioned paragraph according to your advice. We will include in the description other settings where steps and pools are common, as described in Chin and Whol (2005) and the cited studies therein.

Chin, A., & Wohl, E. (2005). Toward a theory for step pools in stream channels. *Progress in physical geography*, 29(3), 275-296.

Methods

-L121: The symbol of a bar with a dot above and one below may be unfamiliar to many readers of Biogeosciences. Define it in parentheses.

We will use a better symbol instead of ÷.

-L121-132:This whole paragraph is highly technical but still can be understood by a broader readership. This would be more likely with the support of more references as only Zappa et al 2007 is cited here. For example it would be helpful to have a citation after “Batchelor scale” (L124), “in its energy cascade (L126).

We will add these additional references as suggested (see list below). Further we will rephrase the paragraph to improve the readability for a general audience.

Esters, L., Landwehr, S., Sutherland, G., Bell, T. G., Saltzman, E. S., Christensen, K. H., ... & Ward, B. (2016). The relationship between ocean surface turbulence and air-sea gas transfer velocity: An in-situ evaluation. *In IOP Conference Series: Earth and Environmental Science* (Vol. 35, No. 1, p. 012005). IOP Publishing.

Batchelor, G. K. (1959). Small-scale variation of convected quantities like temperature in turbulent fluid Part 1. General discussion and the case of small conductivity. *Journal of fluid mechanics*, 5(1), 113-133.

Tennekes, H., & Lumley, J. L. (1972). *A first course in turbulence*, pp 257-264. MIT press.

Katul, G., & Liu, H. (2017). Multiple mechanisms generate a universal scaling with dissipation for the air-water gas transfer velocity. *Geophysical Research Letters*, 44(4), 1892-1898.

-L134: citation supporting this? Maybe Hall and Ulseth 2020 WIRES water?

Thank you for the suggestion. We will add the quotation in the revised text.

In addition to the one mentioned, we will include:

Hall Jr, R. O., & Madinger, H. L. (2018). Use of argon to measure gas exchange in turbulent<? xmltex\break?> mountain streams. *Biogeosciences*, *15*(10), 3085-3092.

Raymond, P. A., Zappa, C. J., Butman, D., Bott, T. L., Potter, J., Mulholland, P., ... & Newbold, D. (2012). Scaling the gas transfer velocity and hydraulic geometry in streams and small rivers. *Limnology and Oceanography: Fluids and Environments*, *2*(1), 41-53.

Ulseth, A. J., Hall Jr, R. O., Boix Canadell, M., Madinger, H. L., Niayifar, A., & Battin, T. J. (2019). Distinct air–water gas exchange regimes in low-and high-energy streams. *Nature Geoscience*, *12*(4), 259-263.

Maurice, L., Rawlins, B. G., Farr, G., Bell, R., & Gooddy, D. C. (2017). The influence of flow and bed slope on gas transfer in steep streams and their implications for evasion of CO2. *Journal of Geophysical Research: Biogeosciences*, *122*(11), 2862-2875.

-L161: A brief explanation of this solubility coefficient?

The Ostwald solubility coefficient is the ratio between the volume of absorbed gas and the volume of absorbing liquid for a given condition of pressure and temperature. We will add this explanation in the revised version of the manuscript.

Results

-L177: “Provided by the code” might be better to say “provided by the model”

-L190 “Typo in “Negative values are observer” -> observed

Thanks. We will fix these minor issues.

-L205: It is unclear if it was one or two orders of magnitude. Might need to rephrase.

-L223: No need to say what you did or what you show in a figure. You can directly explain the observation and cite the figure in parenthesis.

We will rephrase the two sentences as suggested.

Discussion

-the discussion is very well written, despite the main comment that mostly concerns the discussion I have no small issues.

Thank you for the positive comment.