General comments

This manuscript presents results from a sampling campaign in the riparian zone shallow groundwater and draws conclusions on the redox conditions influencing DOM exports into the stream of a small agricultural catchment. This work builds on preceding works in the same catchment and extents the previous findings and hypotheses. The manuscript is a good match for HESS and should be of interest for scientists working on catchment water quality.

The manuscript is written in a concise way. Figures are mostly good and references up to date. My specific comments given below add up to a quite long list but are not substantial. My most critical point is the temporal averaging of lysimeter data that nees a better justification. However, from my point of view some work is needed to bring this manuscript into a final acceptable form.

REPLY: We thank the reviewer for their positive evaluation of our work.

Specific comments

Abstract

The abstract uses DOC while title and manuscript text uses DOM. Homogenize that?

REPLY: The abstract was modified as followed:

" […] we combined monitoring of nitrates, iron, soluble phosphorus and dissolved organic matter concentration (as dissolved organic carbon; DOC) […] "

L7: Check this first sentence. Not clear what seasonal variations are meant here. Seasonal variations of environmental conditions that control DOC exports or controls of the seasonal variations of the DOC export itself?

REPLY: We meant the seasonal variations of environmental conditions regulating DOC export. The sentence will be modify accordingly:

"To better constraint the seasonal variations of the environmental conditions regulating dissolved organic matter (DOM) export in lowland catchments, we […]"

L8: Why "nitrates" not "nitrate"?

REPLY: "Nitrate" will be used in the revised manuscript instead of "nitrates".

L13: "visit" is maybe not the best choice here. I hope you also sampled them.

REPLY: Indeed, we "collected" samples.

L14ff: Increase of DOC concentrations and release into the soil water seems to be the same thing. Our do you mean release into surface water?

REPLY: We meant an increase in soil waters:

"We observed a large increase in DOC concentrations in soil waters of the riparian areas […]"

L15: I have mixed feelings about "notably due to". Is that your interpretation of the data or a proof? Maybe another choice can make that level of certainty in the underlying processes more clear.

REPLY: It is based on the data that show a strong link between iron biodissolution and DOC release in soils. We changed "notably due to" by "linked to".

Introduction

L36-39: Consider to state the hydroclimatic conditions under that these statements were made. Also consider the work of Winterdahl in this context (10.1002/2013gb004770).

REPLY: Thank you for the additional reference. We suggest modifying the text as follow:

"Numerous research carried out in temperate and boreal catchments have shown that headwater catchments are the main entry point of DOM into fluvial networks (Ågren et al., 2007; Creed et al., 2015) and identified riparian areas as the dominant sources of DOM at the catchment scale owing to their location at the terrestrial-aquatic interface (Sanderman et al., 2009; Lambert et al., 2014; Laudon et al., 2012; Winterdahl et al., 2014)."

L41-42: Is this statement underpinned with papers later in the text? Maybe it make sense to state that time-scale question after the next section? Here it seems quite strong without underpinning.

REPLY: This statement is supported by several papers, indeed cited in the following paragraph. Please note that the introduction will be modidy following comments from B. Selle:

"Numerous research carried out in temperate and boreal catchments have shown that headwater catchments are the main entry point of DOM into fluvial networks (Ågren et al., 2007; Creed et al., 2015) and identified riparian areas as the dominant sources of DOM at the catchment scale owing to their location at the terrestrial-aquatic interface (Sanderman et al., 2009; Lambert et al., 2014; Laudon et al., 2012; Winterdahl et al., 2014). The flushing of shallow organic-rich soil layers during storm events typically represents the majority of annual DOC loads (Inamdar et al., 2006), and the DOC versus discharge relationships during storm events show that DOC export is transport-limited at the event scale (Buffam et al., 2001; Zarnetske et al., 2018). Although geomorphological and climatic conditions regulate DOC loads in stream waters (Winterdahl et al., 2014; Laudon et al., 2012), DOC export at the annual scale is commonly conceptualized as a two-steps process in which DOM is produced and stored in the catchment during the hot and dry period, and then exported toward surface waters during the wet and cold period (Boyer et al., 1996). This two-steps process model often described in temperate catchments (Deirmendjian et al., 2018; Strohmenger et al., 2020; Wen et al., 2020; Ruckhaus et al., 2023) is also supported by numerous studies carried out in tropical (Bouillon et al., 2014), boreal (Tiwari et al., 2022), Mediterranean (Butturini and Sabater, 2000) or Arctic fluvial networks (Neff et al., 2006). However the processes regulating the size of the pool of riparian DOM remain unclear (Tank et al., 2018 and references below)."

L43-56: Again some mentioning of climatic settings may help. I noted that often there is the assumption that DOC export works basically similar from boreal to Mediterranean/ subtropic conditions. Partly justified but (not exhaustively) stating where some of the findings have been made would be great. See also Laudon (10.1029/2012gl053033).

REPLY: Researches on DOC export have shown that stream DOC export is very similar across different geomorphological and climatic settings. Please find below a proposition of modification:

"Although geomorphological and climatic conditions regulate DOC loads in stream waters (Winterdahl et al., 2014; Laudon et al., 2012), DOC export at the annual scale is commonly conceptualized as a two-steps process in which DOM is produced and stored in the catchment during the hot and dry period, and then exported toward surface waters during the wet and cold period (Boyer et al., 1996). This two-steps process model often described in temperate catchments (Deirmendjian et al., 2018; Strohmenger et al., 2020; Wen et al., 2020; Ruckhaus et al., 2023) is however also supported by numerous studies carried out in tropical (Bouillon et al., 2014), boreal (Tiwari et al., 2022), Mediterranean (Butturini and Sabater, 2000) or Arctic fluvial networks (Neff et al., 2006)."

L67-72: I have problems following the line of argumentation in this sentence. Why is the impact of Fe reduction (on DOC export? Not stated here) limited but then rather favoring conditions are mentioned.

REPLY : We reformulated this sentence for clarity:

"However, the triggering of Fe reduction (and the consequences in terms of DOC release in soils and export toward stream waters) could be limited in agricultural catchments owing to inputs of nitrate (an oxidizing specie) from upslope and/or groundwaters that may prevent the reductive biodissolution of iron (Mcmahon and Chapelle, 2008; Christensen et al., 2000)."

L75-77: This is true but it would be fair to cite some studies that make this attempt of bringing together soil and stream water (Knorr 2013, Dupas et al. 2015 – though P-centered, or even Seibert et al. 2009 and Ledesma et al. 2015).

REPLY: We will add several papers in addition to those suggested by the reviewer: Knorr, 2013; Dupas et al., 2015; Ledesma et al., 2015; Seibert et al., 2009; Sanderman et al., 2009; Lambert et al., 2013.

L82-91: Maybe mention if and how stream water quality was monitored as well.

REPLY: Indeed. The text will be modify as follow:

"To this end, zero-tension lysimeters were installed in the riparian area of a well-instrumented agricultural catchment whose stream waters are continuously monitored for water quality, including DOC at high frequency (Fovet et al., 2018).".

Material and methods

L102: Some more details on the riparian soils would be helpful to later on make more clear, why a depths of 15 cm was chosen.

REPLY: Values of soil organic carbon content have been added in the revised manuscript:

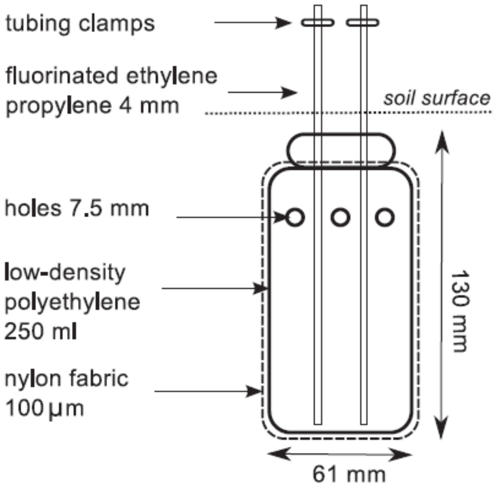
"Soil organic carbon content presents lateral (riparian versus upland soils) and vertical (surface versus deep soils) gradients, with highest values about 5.3 – 5.6 % reached in the uppermost soil horizons (0-20 cm depth) of the riparian area while soil organic content fall under 1% below 20 cm depth (Lambert et al., 2011)."

L107: Figure reference is misleading here since there is no land use in this figure.

REPLY: Correct. Figure will be modify.

L122: Some details on the zero tension lysimeters would be helpful.

REPLY: A figure showing how lysimeters are build will be added in supplements, based on Dupas et al. (2015):



L128: Not sure if “degradation” is the right word.

REPLY: This will be replaced by “damages”.

L129: “consecutive dates of data” sounds a bit strange.

REPLY: “of data” will be removed.

L132: Figure 1 reference does not fit here.

REPLY: Indeed, we meant Figure 2.

L137 and L146: Remove the XXX.

REPLY: Done.

L157: On which bases was the decision made to dilute the sample.

REPLY: Samples were diluted following the Ohno et al. (2002) recommendation (absorbance at 254 nm < 0.3) to reduce inner filter effects and iron quenching.

L177: How were missing data handled?

REPLY: Missing data were very few (less than 10 sampling) and were not included in the PCA.

L181: Does normalization not include an averaging to a mean of zero and a standard deviation of 1? Is the averaging justified? Temporal variability may be higher than spatial variability and maybe each single sample would be better in the PCA? This needs more justification somewhere in the paper.

REPLY: Similar comments were raised by C. Williams. Please find below our answer:

The aim of the PCA-clustering approach was to discriminate and group lysimeters based on the occurrence or absence of iron biodissolution in soil waters in order to investigate the temporal pattern of each cluster that would help to identify patterns compared to individual time series. For this reason, data we aggregated for each lysimeters. Otherwise, a given lysimeter would switch clusters and the temporal figure per cluster would make no sense.

Although we agree that the data aggregation per lysimeter erases the temporal dimension, this is necessary for the clustering and the temporal aspect is described in the next step of the analysis. Please note that including all the dates lead to similar result compared to the “temporally-normalized PCA” used in the manuscript, although, obviously, more ‘noisy’ (Figure 1).

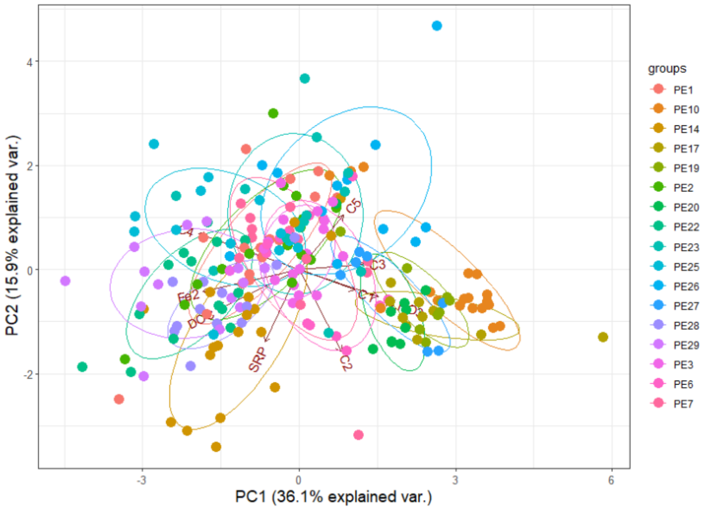


Figure 1 - PCA using all dates

The revised manuscript gives more details to justify the approach (section 2.5):

“A principal component analysis (PCA) coupled to a clustering analysis was used to discriminate and group lysimeters based on the occurrence or absence of iron biodissolution in soil waters in order to investigate the temporal pattern of each cluster that would help to identify patterns compared to individual time series. For this reason, data (DOC, NO3-, SRP and Fe(II) concentrations and the relative contribution of PARAFAC components) were aggregated for each lysimeters and normalized.”

Results

L189: In a stricter sense this opener connects your catchment to a general behavior in the region that would actually better fit the discussion and, moreover, needs referencing. I suggest to just state three phases. You may mention the general regional behavior in that type of catchment in the introduction or method section also. Moreover, the sharp mid-months boundaries of the phases need a criterion to be mentioned here. Is this visually decided?

REPLY: The different phases are based on the water table fluctuation along the hillslope and were defined by Lambert et al., 2013. Following this comment we proposed to modify the text as below:

“The hydrological regime of the study site is characterized by the succession of three distinct periods determined by water table fluctuations along the hillslope, corresponding to different hydrological regimes for the riparian soils (Fig. 2; Lambert et al., 2013): (i) a period of progressive rewetting of riparian soils after the dry season and of low groundwater flow and low stream discharge (01/09/2022 – 18/12/2022, mean and cumulated precipitation = 5.1±5.3 mm d-1 and 338.5 mm, respectively); (ii) a period of prolonged waterlogging of riparian soils induced by the rise of water table in the upland domain, corresponding to high values of hillslope groundwater flow and stream discharge (18/12/2022 – 9/05/2023, mean and cumulated precipitation = 6.6±7.9 mm d-1 and 577 mm, respectively); and (iii) a period of drainage and progressive drying of the riparian soils induced by the drawdown first in the upland domain then in the bottomland domain and corresponding to the decrease of both the hillslope groundwater flow and stream discharge (09/05/2023 – 01/07/2023, mean and cumulated precipitation = 2.1±4.5 mm d-1 and 23 mm, respectively).”

Fig. 2: Consider to indicate the phases in the three time series.

REPLY: Indeed, the figures will be modified.

L197: You mention frequent intense rainfall with 2 mm/d but wrote about moderate precip of 5 mm/d above. Are the numbers correct?

REPLY: The 2 mm per day referred to the short period inside the second period during which no significant rainfall events occurred. In the revised manuscript we removed this sentence for clarity, only mentioning the precipitation values for each period.

L201: Why not giving precipitation values for this third period?

REPLY: we add the values.

L209ff: The chapter on fluorescence properties does not fit here in my opinion. It feels more naturally for me to first state water quality in terms of concentrations and then jump to the DOM properties in detail. But you may have a good reasoning at hand.

REPLY: We found more logical to first begin with the PARAFAC model as the temporal and spatial variations in DOM composition are linked to the dynamics of iron. We thus found sequence PARAFAC > Seasonal Variations > PCA & cluster more relevant than Seasonal Variations > PARAFAC > PCA & cluster.

Fig. 3: Be precise in the captions. What temperature is this? Mention that C-F are soil water concentrations? Homogenize captions and axis – e.g. nitrates – N-NO3? The latter also applies to the other figures.

REPLY: The 'soil' mention will be added on axes, and we will homogenize captions and axis by keeping, for instance, N-NO3 rather than nitrate.

Fig. 4: Does it make sense to give nitrate in B on a log axis?

REPLY: Some values were equal to 0, as measurements were below detection limit. A log axis would therefore not be adequate.

L231: I suggest to also give a Pearson's correlation coefficient to underline the claim of a linear relationship. However, nitrate-iron relationship seems to be far away from linear and rank correlation would be maybe a better fit.

REPLY: Correlation coefficients will be added in the figures and/or in the text. Given the relationship, we will use either Pearson or Spearman correlation coefficients.

L234: "Some" lysimeter not following that cannot be seen in the data. You probably did the correlation across all data. Maybe state lysimeter-individual correlation coefficient (mean, range) to point out that some do not follow the general behavior?

REPLY: Details will be added.

L243: This title should make clear that this is about soil water quality.

REPLY: This will be modify.

L244: Would be good to write clearly what variance is explained.

REPLY: We modified the text as below:

"Overall, the two first components of the PCA explained 69.4 % of the total variance of the dataset"

"discriminating lysimeters depending on the degree of Fe(II) biodissolution" does not sound well written and seems to be more a discussion than a result. I would leave that statement more to the lines 251-253 at the end of the section.

REPLY: The sentence was modified as below for clarity, but we still mentioned that lysimeters were discriminated based on occurrence or absence of iron biodissolution. Indeed, we think that this is a result from the PCA.

"[…] discriminated lysimeters depending on the occurrence or absence of Fe(II) biodissolution in soil waters of the riparian area"

L254: In this chapter it may be elaborated on if averaging the lysimeters in time was justified? Does one lysimeter seem to switch from high-nitrate:low-Fe to low-nitrate:high-Fe over time? This is hard to be seen in Fig. 6 as here all lysimeters of one sampling data and one cluster are averaged.

REPLY: Please see our previous answer on the PCA approach we used.

Fig. 6: Same as fig. 2 – indicating the different wetness phases would be very helpful here.

REPLY: This will be done.

L258 and 270: I don`t understand the idea of a flushing dynamic. Does that mean the solutes are flushed out and replaced by a different water with a different quality? Moreover, I find this to be more an interpretation of that data and thus more a discussion part.

REPLY: We agree that this paragraph is unclear and that we used terms more relevant for the discussion. Following this comment, we modified the text as below:

"In the cluster 1, DOC, N-NO3 and SRP decreased from 39.8±13.3 to 23.4±8.4 mg L-1, from 2.6±3.6 to 1.2±1.8 mg L-1, and from 0.18±0.18 to 0.08±0.15 mg L-1, respectively, during the rewetting phase of the catchment while Fe(II) was no measured at significant levels. During the high flow period, however, Fe(II) increased gradually from 3.7±3.2 to 26.5±7.8 mg L-1, and both DOC and SRP followed a similar trend with concentrations raising from 27.3±9.5 to 54.9±25.0 mg L-1 and from 0.07±0.13 to 0.18±0.11 mg L-1, respectively. During this period and until the end of the hydrological cycle, N-NO3 were very low, falling from 0.54±0.66 mg L-1 at the beginning of the high flow period to values below 0.15 mg L-1 the rest of the survey. The start of the third hydrological period corresponding to the drawdown of the water table and the consecutive aeration of riparian soils was marked by the rapid drop of Fe(II) at 8.1±7.4 mg L-1, DOC at 17.5±10.9 mg L-1, and SRP at 0.02±0.02 mg L-1."

Please note that we applied the same kind of modification for the following paragraph where the term flushing was also used:

"Similarly to cluster 1, soil solutions from the cluster 2 exhibited a decline in DOC and SRP concentrations during the rewetting phase of the catchment but these trends continued during the high flow period, with minimal values reached in the middle of February. Thus, DOC dropped from 34.5±7.1 to 9.4±3.1 mg L-1 and SRP from 0.19±0.08 to 0.02±0.01 mg L-1 during this period, before showing an increasing trend to reach concentrations about 21.0±6.1 mg L-1 for DOC and 0.16±0.13 mg L-1 for SRP at the end of the high flow period. DOC remained elevated (24.1±3.1 mg L-1) at the start of the dry period, but SRP dropped close to depletion. To the inverse, N-NO3 first increased from 0.57±0.81 in November to maximum values of 6.5±5.9 in the middle of March, and then exhibited decreasing concentrations until a complete depletion at the beginning of the third hydrological period. Contrary to cluster 1, Fe(II) was not measured at significant concentrations in cluster 2 (i.e. below 0.5 mg L-1) except in March, during which Fe(II) increased from 1.2±1.9 to 4.1±0.2 mg L-1."

Discussion

L290: appearance, abundance maybe better than apparition.

REPLY: We replaced by the term 'release' that fit more the purpose.

L303: What about a temperature effect here? Or can rainwater just not infiltrate deep enough?

REPLY: Although it is true that temperature may affect oxygen solubility and exchanges with atmosphere, we did not found any relationship between Fe(II) and soil temperatures. The gradual increase in Fe(II) also suggests that rainfall events did not impacted iron biodissolution.

L306f: I don't understand the role of the hydraulic gradient here. Translating to soil water/ shallow groundwater travel time? What I read is more about depth to groundwater than hydraulic gradient.

REPLY: The hydraulic gradient determines the flow of groundwater coming from upland domain to the riparian soils, impacting therefore the hydrological functioning of valley bottoms (Lambert et al., 2013). Although we are not certain about the exact mechanism involved, we attributed the slight decrease in Fe(II) in February/March to the slight decrease in water table in the upland domain. The paragraph was modified to take into account this comment and the previous one:

"A fundamental condition for the establishment of reductive conditions is the prolonged waterlogging of riparian soils. As shown earlier for this and other lowland catchments on impervious bedrock, the increase of the hydraulic gradient induced by the rise of groundwater in the upland domain during the high flow period maintains a strong hydrologic connection between upland and riparian domains (Pacific et al., 2010; Molenat et al., 2008). Under these conditions, riparian soils remain waterlogged owing to a high and continuous hillslope groundwater flow, leading to the gradual establishment of reductive conditions and the subsequent triggering of Fe-biodissolution as long as inputs of oxidizing species remained limited and/or counterbalanced by higher rate of consumption through microbial activity (Lotfi-Kalahroodi et al., 2021; Lambert et al., 2013). This pattern was well illustrated by records from lysimeters grouped in the first cluster (Fig. 6). After a quick depletion of an initial stock of nitrate accumulated during the previous summer, reductive conditions were rapidly established at the beginning of the high flow period and increasing Fe(II) concentrations in soil solutions evidenced the triggering of the reductive Fe biodissolution in riparian soils. The gradual increase in Fe(II) during all the high flow period despite variations in temperature or rainfall patterns (with some intense precipitation events > 20 mm d-1) pointed to limited impact, except during a period of low precipitation during which both Fe(II) and DOC exhibited a slight decrease in February/March. Although the exact reason remains to be determined, we attributed this small drop to the drawdown of the water table in upland groundwater flow owing to a prolonged absence of precipitations (see PK3 fluctuations, Fig. 2) that may have favoured the penetration of oxygen within soil waters (as no changes in N-NO3 occurred)."

L307: If DOC and SRP are similarly affected by oxygen and iron presence, why are they weighted differently in the two clusters?

REPLY: Likely because SRP concentrations were lower compared to DOC.

L316: Double word biodissolution here.

REPLY: Indeed.

L318f: I am not sure if I understand what is meant by "net depletion pattern".

REPLY: Both DOC and SRP concentrations decreased in soil waters, that we interpreted as the progressive flushing of a finite DOC/SRP pool.

L327 (and 313): Spatial patterns are not shown but may be helpful and interesting? So maybe map the clusters back to the catchment figure? Could be in the SI or an additional panel in Fig. 5.

REPLY: Lysimeters were aligned along three lines parallel to the stream channel. These lines, about 10-20 m from each other, were located at different distance from the stream with the aim to capture the heterogeneity of water flow paths and nitrates concentrations coming from the upland domain. Despite our sampling design, the distance between each lysimeter is not a variable that we could integrate in our analysis. We would need to set distance to an independent point (the nearest field? the river?) but we don't think this is would lead to an interesting pattern as no spatial pattern was visible: two neighbouring lysimeters could be more different than lysimeters on the opposite side of the

Following this comment, more details were added in the Material and methods (section 2.2):

"Lysimeters were aligned along three lines parallel to the stream channel. These lines, about 10-20 m from each other, were located at different distance from the stream with the aim to capture the heterogeneity of water flow paths and nitrates concentrations coming from the upland domain. Lysimeters were all located in the hydromorphic soils unit (Figure 1)."

And in Results (section 3.3):

"Despite the fact that lysimeters were installed along three lines that were more or less closed to the stream channel, no spatial pattern was identified. Thus, two neighbouring lysimeters could be more different than lysimeters on the opposite side of the transect."

L337: Water circulation is maybe not the most precise word here. Does soil water really circulate?

REPLY: We modified the sentence:

"A first explanation can be related to the heterogeneity in water flowpaths in soils."

L353-357: This is an interesting part but would, at this process-scale, better fit the end of chapter 4.1? But I am not fully sure either. I noted that this is an initial statement on something explained in more detail below (L373ff). Maybe make more clear that details are given in the following text?

REPLY: We think it is a good opening for the 4.2 section. Indeed, the idea behind these sentences (that winter should be considered as an active phase of DOC export) are developed in the next paragraphs where we try to link soil and stream dynamics.

L366-367: I know what you mean here but generally a “supply-limited pool” is associated with a dilution behavior not a flushing behavior. So, check your choice of words here and maybe better describe what concentration dynamics you see at this point in time in the stream.

REPLY: We replace by “DOM pool limited in size” which is more relevant.

Conclusion

L401: The word “but” indicates some contradiction which I do not clearly see here. Fe-reduction can only establish when nitrates are not present, right?

REPLY: Indeed, we should reconsider this sentence. We proposed the following changes:

“In agreement with previous studies (e.g. Selle et al., 2019; Knorr, 2013), the establishment of Fe-reducing conditions in riparian areas was identified as a major mechanism for the release of large amount of DOM in soil waters. In agricultural catchments, however, we found that this process can be buffered by nitrate, leading to a strong heterogeneity on the degree of iron biodissolution and its consequences on soil DOC dynamics. Our study also evidenced that another production mechanisms unrelated to Fe dynamics contributed to release DOM in riparian soils, pointing the need to investigate deeper stream DOC export at the soil/stream interface.”

L409: Any reference for the “wetter winter” statement?

REPLY: Yes: Strohmenger et al. (2020).

L404ff: This last section in the conclusions seem to make a new story on the relation of the small-scale redox soil processes to the long-term trends. While I appreciate this part I think this may be already part of the introduction and motivation.

REPLY: In fact this was part of the motivation, but indeed this was very briefly mentioned in the manuscript lines 82-85. The last paragraph of the introduction will be modify to better appreciate the goal of our study:

“To this end, zero-tension lysimeters were installed in the riparian area of a well-instrumented agricultural catchment, the so-called Kervidy-Naizin catchment, whose stream waters are continuously monitored for water quality, including DOC at high frequency (Fovet et al., 2018). This catchment is located in Brittany (France), a region where stream DOC concentrations exhibited contrasting trends (increasing, decreasing or no trend) over the 2007-2020 period despite similar geomorphological and climatic conditions (Supplementary Fig. S1). The Kervidy-Naizin site for instance shows a weak but significant increase in stream DOC concentrations over the last decades (Strohmenger et al., 2020). In this context, another goal of this study was to explore the hyoothesis that long-term decline in nitrate inputs from agricultural practices (Abbott et al., 2018) may have impacted long-term trends in DOC through a potential impact on iron dynamics in riparian soils.”

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