

Dear Editor,

Thank you for your efforts and those of the Reviewers in evaluating and handling our manuscript bg-2019-486, “Carbon dioxide dynamics in an agricultural headwater stream driven by hydrology and primary production.”. We have changed the manuscript in response to the reviewer’s comments, and a detailed breakdown of our responses and revisions to the manuscript follows. In the revised manuscript we feel that we have dealt fully with the points raised and as a result the manuscript has been significantly improved.

Best regards / Marcus Wallin (corresponding author)

## **Reviewer (R#1) comments and author responses to ms bg-2019-486**

### **Reviewer comments are given in normal style and with author responses in *italic***

Headwater streams are known hotspots for CO<sub>2</sub> emissions, although studies of headwater streams draining agricultural catchments, and specially studies that includes a temporal dimension, are sparse. In this study, a headwater stream draining an agricultural catchment was continuously monitored during for approximately one year, and the responses in CO<sub>2</sub> concentrations to hydrological variations were studied.

#### General comments

This study provides important insights of CO<sub>2</sub> and discharge dynamics in a headwater stream draining a catchment impacted by agriculture. We need more studies like this in order to better understand the exchange of greenhouse gases between inland waters and the atmosphere. Overall, I think the manuscript is very good. The study is well designed and presented in a well-structured way. I have only a few, although important, remarks that I think would improve the manuscript.

*Response: We thank reviewer #1 for their overall positive evaluation of our manuscript and appreciate that it is found “very good” and “well designed and presented in a well-structured way”. We believe that the revised manuscript has been significantly improved following the comments given by reviewer #1.*

Firstly, this paper would benefit from the authors emphasizing the relevance of their study better. For instance, this study points out potential effects of stream intermittency for streams draining agricultural catchments. This finding is highly important with respect to climate change. Despite this, the authors do no mention this neither in the abstract nor in the conclusions of the paper.

*Response: We agree that this is an important finding that we do not well enough lift up as one of the main messages. We do not have enough years of measurements for saying how common the intermittency of this specific stream is. The spring and summer of 2018 was unusually dry, but this kind of conditions are expected to occur more frequently in the future. We have in the revised version included this finding in the abstract (ln 22-25) and also more explicitly in the conclusions of the study (ln339-343).*

Secondly, the manuscript would benefit from a more extensive discussion, for example how this stream compares to other agricultural influenced streams, if the type or insensitivity of the agriculture matters, land use change etc.

*Response: We have in the revised version tried to improve the discussion on the spatial representativeness of our findings. However, this is not easy as there are not many relevant studies existing, while the current study contributes to fill this knowledge gap. We have further included references concerning DOC-discharge responses in agricultural areas. Ln285-291*

Lastly, the readability of the manuscript could be greatly improved by simple sentence adjustments, such as shortening sentences and inserting more commas. Also, the figures could be designed in a more intuitive way.

*Response: We have in the revised version tried to improve the readability of the text where appropriate, and we agree that some of the figures needed a quality lift up, although no suggestions on how were given by the reviewer. We have improved the quality of the figures (mostly improved font sizes) and believe that they are now clear and informative for the reader.*

#### Specific comments

##### Abstract

L15: It is unclear what "one year of open-water season" means. It would be helpful to add the dates and/or number of monitored days.

*Response: We agree that this was a bit unclear, in the revised version we have removed "open-water season" and also added "(in total 339 days excluding periods of ice and snow cover)" at the end of the sentence. Ln16*

L22: I recommend the authors to add a sentence about the effects of indeterminacy of streams draining agricultural catchments here, since this is an important finding of the paper.

*Response: We agree that this was missing. In the revised version we have added two sentences on this topic in the abstract. Ln22-25*

##### Introduction

L41-42: This sentence is unclear. What do you mean with positive and negative responses? Please clarify.

*Response: We mean that variations in stream CO<sub>2</sub> concentration have been found to be both positively and negatively related to variations in stream discharge, i.e. either that CO<sub>2</sub> concentrations increase when discharge does, or that CO<sub>2</sub> concentrations decrease when discharge increase (dilution). This is now clarified in ln47-48*

L44: "...dominant CO<sub>2</sub> source areas of catchment soils"? Please rephrase this sentence.

*Response: This is now clarified. Ln50-51*

L45: Please specify what kind of other catchments.

*Response: There is no consensus (partly due to few existing studies) in which catchments CO2 are mainly controlled by hydrology or biology so it is hard to specify them more than catchments where the hydrological influence is low or non-existing. Hence, we keep the original formulation.*

L48: New paragraph needed.

*Response: Now added*

L50: Please specify what “relevant” time-scales are.

*Response: “(<hourly resolution)” is now added in ln58.*

L69: Please specify what you mean with high-resolution. Also, as mentioned before, it is unclear what "one year of open-water season" actually means.

*Response: Both “(hourly)” and “(in total 339 days excluding periods of ice and snow cover)” are now added to this sentence. Ln77-78.*

## Methods

L76: Please rephrase this sentence. Is it unclear if you mean the annual mean temperature or the January and July temperatures. This is especially important since you do not mention the precipitation in January or July - perhaps this could be added.

*Response: This is now clarified in ln85-86.*

L82: Stream pH ranging between 7.4 and 8.4. Also, this sentence would be much more readable if you would add a comma. In general, I would recommend using commas more frequently.

*Response: This is now clarified in ln93.*

L83-84: How much lower? Please provide a reference percentage.

*Response: It is hard to give exact percentages as the variables included in “nutrients” are many. We have instead added, as a general approximation, that the studied stream is within the 25<sup>th</sup> percentile of the monitored agricultural streams in Sweden when it comes to DOC and nutrient levels. Ln94*

L86-87: This sentence could be moved to the beginning of the paragraph.

*Response: We agree and in the revised version have moved it to the start of the paragraph in ln84-85.*

L90: influences

*Response: Correct, now changed in ln100.*

L91: Table S1; Figure S2

*Response: Correct, now changed in ln101.*

L93: Would it be possible to add here the percentage that were snow/ice-free (and included in your study) as well as the percentage when the stream was falling dry?

*Response: We have again added the total number of measurement days in ln103-104. Concerning the number of days of the dry periods, this is more of a result and is already given in the text.*

L100: This is quite confusing for the reader, especially since you have not mentioned before that the stream is falling dry during some periods of the year. In general, I would recommend you to highlight the stream intermittency better, including adding some sentences in the introduction about this.

*Response: Again, the dry periods are here seen as result rather than description the methods. Still we needed to explain that analysis of the CO<sub>2</sub> data was only made when runoff was generated. It would therefore not be logical to introduce the stream intermittency already in the introduction as this was not included in the aims of the study. However, as it became evident during the study that stream intermittency occurred and also was highly influential for the CO<sub>2</sub> dynamics of the studied stream, this is something that needs to be discussed in a more extended way. See response to the first general comment.*

L108-109: Please clarify. What is the temporal resolution of your data?

*Response: As for many sensor-based systems averaging high-frequency data reduces the noise of the measurements and makes them more reliable. The given averaging time needs to account for relevant time-scales for the processes you want to study but also consider practical limitations as power consumption, data storage etc. In this case we measured at a 1 min interval and stored average values based on these 1 min measurements every 30 min (in 2017) or 60 min (in 2018). This is now clarified in ln118-119.*

L112-114: Please rephrase. Also, how many replicates?

*Response: We have clarified that one sample was taken at each sampling occasion in ln122-123.*

L117: Please clarify. When was the phosphoric acid added?

*Response: The phosphoric acid was pre-injected in the vial before the sample was injected. This is already stated and so no change has been made.*

L121: Did you run any standards?

*Response: Yes, certified standards were analysed. This is needed as the DIC-values are given in relation to the PDB standard. This is clarified in ln132.*

L129: Add reference to Figure S2 here.

*Response: Correct, that is now added in ln 141.*

L140: It would probably be easier to follow if you move this paragraph to the beginning of the methods section.

*Response: It is not clear how that this would clarify the text and make the methods more logical to follow. We prefer to keep this section where it is.*

L145: Another example of a sentence where the overall readability could be greatly improved if more commas are added.

*Response: We agree, and a comma has been added-*

## Results

L155: This sentence is confusing. Precipitation is usually in mm/year however the period is for a bit more than a year. I assume that the "total precipitation" represent the precipitation for the whole period. Thus, it would be easier to read if the sentence would first state the mean air temperature (XXX) and then the total precipitation (XXX).

*Response: We write that "The mean air temperature and total precipitation for the entire period (Sep 26, 2017-Dec 12, 2018)". We believe this is already clear and have not made any changes.*

L205: It would be good to also add the corresponding pCO<sub>2</sub> here for reference.

*Response: We have chosen to present the CO<sub>2</sub> data as a concentration in the unit of mg C/L as this normalizes for solubility and makes it directly comparable with for example DOC/TOC concentrations if total aquatic C export would be of interest. We give corresponding pCO<sub>2</sub> values in ln ?? as an example for how they compare. But we don't think it is reasonable to give pCO<sub>2</sub> values to all given CO<sub>2</sub> concentrations in the manuscript while no addition has been made.*

L212: Same as above, the corresponding pCO<sub>2</sub> values would be helpful as reference values.

*Response: Same as above*

## Discussion

L231: "highly dynamic pattern in streamwater CO<sub>2</sub> concentration".

*Response: Yes, we have added "concentration" for clarity in ln245.*

L250: Please add references.

*Response: Two suitable references for this statement are added in ln264.*

L256: could

*Response: We agree, is now changed in ln272.*

L258-260: Please rephrase.

*Response: We have removed one piece of this sentence that might have been unclear. Ln274*

L266-270: Great paragraph. Would it be possible to develop more on this?

*Response: We have extended this paragraph to further develop the discussion about similarities/dissimilarities in carbon dynamics observed for agricultural streams. Ln285-291.*

L271-272: Please rephrase.

*Response: The sentence is now rephrased in order to clarify in ln 294-295.*

L309-311: Another great paragraph. This could also be further developed and better highlighted.

*Response: We thank the reviewer for this positive comment. We have in the revised version developed this section further in ln333-334, and also highlighted this finding in the abstract (ln 22-25) and the conclusions (ln339-343).*

## Tables

Table 1: Throughout the manuscript, you write either "land-use" or "land use". In the table it is obviously a spelling mistake; however, please be consistent with the terminology throughout the whole manuscript.

*Response: Thank you for noting, we have now used a consistent spelling "land use" throughout the ms.*

Table 2: Would be good to add the name of the catchment and not only the abbreviation.

*Response: The name is now fully spelled out.*

## Figures

Figures: I recommend the authors to redo all figures. They are not intuitively designed or appealing for the reader.

*Response: Although the comment is very un-specific concerning what to improve, we agree that some of the figures needed polishing, especially concerning font sizes etc. We have updated many of the figures in order to make them easier to read.*

Figure 7: Add regression line?

*Response: Here we have used Spearman's Rank (which assumes a monotonic, non-linear, relationship), not regression, so fitting a line would not be appropriate. The given statistics in the figure refer to the Spearman rank test.*

Figure 10: In the text it is written that d13C-DIC was NOT a function of Q?

*Response: Yes, we write "Although there was a tendency towards more negative  $\delta^{13}\text{C-DIC}$  values at higher discharge, no significant relationship was found (Figure 10)". Although not significant from a statistical point of view, we still think it is useful information provided by the figure. One can imagine that the relationship might have been significant if the number of observations would have been more.*

## **Reviewer (R#2) comments and author responses to ms bg-2019-486**

### **Reviewer comments are given in normal style and with author responses in *italic***

Inland waters and specifically headwaters emit significant amounts of CO<sub>2</sub> to the atmosphere; however, studies focusing in agricultural streams and including continuously measured in-situ CO<sub>2</sub> from are rather rare. In this MS, the authors continuously monitored CO<sub>2</sub> with cost-effective Co<sub>2</sub> sensors during one year and explored the spatio-temporal variations of CO<sub>2</sub> throughout the year as a function of hydrology and metabolism.

#### General comments

The MS bg-2019-486 provides an interesting study about CO<sub>2</sub> dynamics in one stream draining a catchment largely dominated by agriculture. An important finding is that stream intermittency can cause rapid pulses of CO<sub>2</sub> even in catchment with no pronounced dry and wet seasons. I think this is an important matter to better understand carbon emissions from streams at the global scale, in the context of climate change (change in hydrology). In line with this result, it could be useful to add somewhere in the discussion the spatial representativeness at the global scale of the stream studied here. In addition, it could be nice to add discussion/comparison of this agricultural stream with other agricultural streams worldwide, because the hydrology should be very different. To increase there adability, I suggest to better define some terms used in this study, particularly, open-water season, and the different periods, and also define better the time-intervals of these seasons throughout the text. Indeed, to my opinion, those terms are specific to boreal systems, and sometimes it is difficult to follow for a reader who is novice with boreal landscapes. A second important finding is the strong biologic control (aquatic primary production) of the CO<sub>2</sub> dynamics during baseflow that should decrease CO<sub>2</sub> emissions during this period. Indeed, during base flow it is common to observed higher CO<sub>2</sub> concentration in streams because deeper levels of groundwater are involved. Perhaps the authors could further developed this. Overall, I found the dataset very interesting; it is rare to have such continuous measurements for CO<sub>2</sub> in streams. In addition, I found the paper well written. Perhaps the quality of some figures could be improved. Overall, I support publication of this manuscript and below are some more detailed comments.

*Response: We thank reviewer #2 for their overall positive evaluation of our manuscript and are happy that publication in Biogeosciences is recommended after a revision. We believe that we in the revised manuscript have better discussed the spatial representativeness of our findings and also elaborated more on the primary production part. We further improved the quality of the figures (also in line with comments from R#1).*

#### Specific comments

##### Abstract

L. 15-16: It would be nice for the reader adding the size of the catchment, the date of open-water season, and the time-step of CO<sub>2</sub> measurements.

*Response: We agree and have in the revised version of the abstract added catchment size and total number of days of measurements. We have further replaced “continuous” with “hourly” to clarify the temporal resolution of the measurements.*

##### Introduction

L.31-33: The authors can check this reference that suit with their study (Deirmendjian et al, 2019. Importance of the vegetation-groundwater-stream continuum to understand transformation of biogenic carbon in aquatic systems – a case study based on a pine maize comparison in a lowland sandy watershed), where the concentration of CO<sub>2</sub> in agricultural and forested streams (and in groundwater) in a temperate catchment was compared. They found no differences between both streams because degassing in agricultural streams was prevented.

*Response: We agree that this reference is very suitable and have added two sentences using information from it in ln40-44.*

L. 43-45: Please clarify this sentence. You mean that different level of soils are exported in function of the change in hydrology?

*Response: Yes, we mean that dependent on hydrological conditions different source areas in the catchment soils are hydrologically connected and contribute differently to the stream CO<sub>2</sub>. The variability in source areas are both vertically and laterally distributed in the soils and are hence activated differently dependent on groundwater position and dominating pathways. This pattern is further dependent on the catchment characteristics and land use. We have in the revised version tried to clarify the lateral and vertical consideration of source areas in ln49-51.*

L-40-55: To my opinion, there is a lightly lack of spatial references in this paragraph. Indeed, I guess that agricultural streams in tropical or boreal areas are very different in terms of hydrology and carbon dynamics. Could you mention spatial references?

*Response: We agree that the spatial coverage among the given references might look limited. We base this section solely on studies that have used high-frequency CO<sub>2</sub> sensor data. This is now clarified in ln46. Also, the two references originally given (Dinsmore et al. 2013 and Crawford et al. 2017) both include data from multiple sites including boreal, temperate, alpine and subtropical areas. They further represent a large variety of forest, wetland and mountainous coverage. Hence, we believe that we already have a relatively good spatial coverage, but to further support the tropical side we have added the very suitable Johnson et al. 2007 paper and adjusted the text according to this in ln46-49..*

L. 69: High-resolution: what is the time-step of measurements?

*Response: We have in the revised version added “(hourly)” after “high-resolution”(ln77). Although we used 30 min resolution in 2017 and 60 min resolution in 2018 (in order to save power) “hourly” is likely the best option here. Further details on the different temporal resolutions are given in the method section.*

## Methods

L.78: What kind of cropland it is? This is important for the d13C-DIC

*Response: The land is mainly used for cereal production and pasture. This clarification is now added in ln88.*

L. 83: Lower end: how much lower?



*Response: It is hard to give exact percentages as the variables including in “nutrients” are so many. We have instead added, as a general approximation, that the studied stream is within the 25<sup>th</sup> percentile of the monitored agricultural streams in Sweden when it comes to DOC and nutrient levels. Ln93-95.*

L. 85: Growing season: what is the time interval?

*Response: The length of the growing season is on average ca 210 days starting in mid-April and ending in early November. This information is now included in ln86-87.*

L. 97: what was the concentration of gas standards?

*Response: Four standards were used (400, 1000 and 5000 ppm as well as 2%). This is now added in ln107.*

L.100: discharge rates lower than 0 L/s: so you mean when the stream was dry or when the stream was frozen? Or both? It is a bit confusing.

*Response: This mean that CO2 data was just analyzed if runoff was generated over the V-notch dam i.e. excluding standing water or completely dry conditions. The instrument was never measuring during ice or snow conditions. This is now clarified in 103-104.*

L.101: Figure S1

*Response: Yes, this figure reference is now given in ln111.*

L109: You wrote one measurements each minute but then a temporal resolution of 30. It is a bit confusing what is the meaning of temporal resolution here?

*Response: As for many sensor-based systems averaging high-frequency data reduce the noise of the measurements and make them more reliable. The given averaging time needs to account for relevant time-scales for the processes you want to study but also consider practical limitations as power consumption, data storage etc. In this case we measured at a 1 min interval and stored average values based on these 1 min measurements every 30 min (in 2017) or 60 min (in 2018). This is now clarified in ln118-119.*

L.120: What is the volume of the injections?

*Response: The volume of the injections was 100  $\mu$ L i.e.  $7 \times 100 \mu$ L per sample. This info is now added in ln130.*

L.129: Please specify that these streams were not located in your catchment and add the reference to the figure S2

*Response: This is now clarified in ln141.*

L. 145: Please define better your four periods. What are the time intervals?

*Response: We have now added number of days per period in ln158-162. We have also added a new table (Table S2) to the supplementary information that gives the full period description.*

## Results

L.157: Please refer to figure 3

*Response: Figure 3 is now referred to in ln172.*

L.168-172: Please add corresponding pCO<sub>2</sub> for reference, as you did

*Response: We have chosen to present the CO<sub>2</sub> data as a concentration in the unit of mg C/L as this normalizes for solubility and makes it directly comparable with for example DOC/TOC concentrations if total aquatic C export would be of interest. We give corresponding pCO<sub>2</sub> values as an example for how they compare. But we don't think it is reasonable to give pCO<sub>2</sub> values to all given CO<sub>2</sub> concentrations in the manuscript while no addition has been made.*

L.166. To my opinion, I suggest to do that for the remainder of the text because pCO<sub>2</sub> in ppmv is more “understandable” than CO<sub>2</sub> in mg/L.

*Response: See comment above. We think this is also very much a matter of personal taste and as stated above we see clear advantages of presenting the CO<sub>2</sub> data as concentrations rather than a volume fraction i.e. ppmv.*

Discussion:

L.225: I would not rush on conclusion about zero/limited tree cover along agricultural streams, at the global scale. I am agree considering your figure S2 that this is the case in your catchment. However, in temperate climate it is very common to observe riparian forest along agricultural streams.

*Response: We agree that the statement was maybe too strong and have revised it in ln239. We still believe that canopy cover is important and that agricultural streams to a larger extent than for example forest streams are exposed to direct sun-light, even at the global scale.*

Figures

Figure 1: In the left part, I suggest to add a map of Europe rather than just Sweden. Please add a scale in the left part too.

*Response: We have updated the figure for the revised version.*

Figure 2: I suggest to separate the different periods (autumn, snowmelt, spring, dry period) with dotted lines, as you did in the next figure.

*Response: Good idea, this is now added*

Figure 4: It is not very intuitive what the time interval is for A, B, C and D.

*Response: The figure is now updated with appropriate font sizes*

Figure 5: Same remark

*Response: The figure is now updated with appropriate font sizes*

Figure 7: Perhaps add regression line with slope

*Response: Here we have used Spearman's Rank (which assumes a monotonic, non-linear, relationship), not regression, so fitting a line would not be appropriate.*

# Carbon dioxide dynamics in an agricultural headwater stream driven by hydrology and primary production

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**Abstract.** Headwater streams are known to be hotspots for carbon dioxide (CO<sub>2</sub>) emissions to the atmosphere and are hence important components in landscape carbon balances. However, surprisingly little is known about stream CO<sub>2</sub> dynamics and emissions in agricultural settings, a ~~land-use~~ land use type that globally cover ca 40% of the continental area. Here we present e  
15 continuously-hourly measured in-situ stream CO<sub>2</sub> concentration data from a 11.3 km<sup>2</sup> temperate agricultural headwater stream  
catchment covering more than one year of open-water season (in total 339 days excluding periods of ice and snow cover). The  
stream CO<sub>2</sub> concentrations during the entire study period were generally high (median 3.44 mg C L<sup>-1</sup>, corresponding to partial  
pressures (*p*CO<sub>2</sub>) of 4778 μatm) but were also highly variable (IQR = 3.26 mg C L<sup>-1</sup>). The CO<sub>2</sub> concentration dynamics covered  
a variety of different time-scales from seasonal to hourly, and with an interplay of hydrological and biological controls. The  
20 hydrological control was strong (although with both positive as well as negative influences dependent on season) and CO<sub>2</sub>  
concentrations changed rapidly in response to rainfall and snowmelt events. However, during growing-season baseflow and  
receding flow conditions, aquatic primary production seemed to control the stream CO<sub>2</sub> dynamics resulting in elevated diel  
patterns. During the dry summer period, rapid rewetting following precipitation events generated high CO<sub>2</sub> pulses exceeding  
the overall median level of stream CO<sub>2</sub> (up to 3 times higher) observed during the whole study period. This finding highlights  
25 the importance of stream intermittency and its effect on stream CO<sub>2</sub> dynamics. Given the observed high levels of CO<sub>2</sub> and its  
temporally variable nature, agricultural streams clearly need more attention in order to understand and incorporate these  
considerable dynamics in large scale extrapolations.

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## 1. Introduction

Fluvial systems (streams and rivers) are estimated to dominate the inland water CO<sub>2</sub> source globally, surpassing CO<sub>2</sub> emissions by lakes and reservoirs by a factor of six (Raymond et al. 2013). However, this estimate relies on a number of assumptions and the scarcity of empirical data makes it uncertain. One of the critical gaps in the global upscaling is the lack of direct measurements from agriculture dominated areas (Osborne et al. 2010). Globally, agricultural land covers about 40% of the total continental area (Ramankutty et al., 2008) but there are few studies specifically focusing on the magnitude and dynamics of CO<sub>2</sub> emissions from agricultural streams. The few studies that do exist ~~have shown~~ ~~conclude~~ that agricultural stream CO<sub>2</sub> concentrations in such streams are generally high and up to ~~five~~ 5 times greater than those in streams draining forested areas which are more extensively studied (Borges et al. 2018; Bodmer et al. 2016; Wallin et al. 2018). For example, Bodmer et al. (2016) measured partial pressure of CO<sub>2</sub> (pCO<sub>2</sub>) in German and Polish streams and examined differences between forested and agricultural catchments. They found that pCO<sub>2</sub> was generally 2-3 ~~higher~~ times higher in agricultural streams compared to streams draining forested areas. Similarly, Borges et al. (2018) found high CO<sub>2</sub> concentrations in streams and rivers dominated by agriculture in the river system Meuse, Belgium. They linked the higher pCO<sub>2</sub> in agricultural streams ~~(up to 5 times higher than in forested areas)~~ to elevated levels of dissolved organic carbon (DOC), particulate organic carbon (POC) and inorganic nitrogen. ~~On the other hand, Deirmendjian et al. (2019) showed that there was no difference in pCO<sub>2</sub> between forest and cropland streams in south-west France despite higher pCO<sub>2</sub> in forest groundwater compared to cropland groundwater. They explained the similar stream pCO<sub>2</sub> by lessmore efficient gas exchange in the forest streams compared to the low-gradient cropland streams.~~

There are numerous factors influencing CO<sub>2</sub> patterns in stream systems and ~~often~~ site-specific controls often dominate. Hence, large scale generalizations are difficult to make (Crawford et al. 2017). ~~Based on high-frequency data, Although the hydrology of agricultural streams will differ across climate zones (and this in turn will have an effect on C dynamics), climate itself has not directly been observed to act as a control on fluvial CO<sub>2</sub> concentrations (Raymond et al. 2013).~~ CO<sub>2</sub> concentrations in streams draining nutrient-poor forest and peatlands, as well as tropical forests, streams are ~~are often found~~ often related to variations in stream discharge but with site-specific response patterns, with CO<sub>2</sub> found either positively or negatively related

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55 ~~to stream discharge both positive and negative~~ (Crawford et al. 2017; Dinsmore et al. 2013; Johnson et al. 2007). These response patterns have often been connected to the catchment characteristics and changes in hydrological pathways, which in turn control the dominant  $\text{CO}_2$ -source areas (~~both from a vertical and lateral point of view~~) of  $\text{CO}_2$  in the catchment soils (Campeau et al. 2018; Leith et al. 2015; Dinsmore and Billett 2008). In contrast, other catchments lack a strong hydrological control and instead display clear diel cycles in ~~the~~ stream  $\text{CO}_2$  concentration indicating a metabolic control (Crawford et al. 2017). Here the interplay of photosynthesis and respiration (in-stream or terrestrial) could result in large day to night time differences in stream  $\text{CO}_2$ .

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60 These recent findings concerning dynamics and controls on stream  $\text{CO}_2$  concentrations have been possible due to the development of cost-effective  $\text{CO}_2$  sensors (e.g. Johnson et al. 2010; Bastviken et al. 2015) which have enabled continuous data collection covering relevant time-scales (~~<hourly resolution~~). However, very little information about stream  $\text{CO}_2$  dynamics exists from agricultural areas, a ~~land-use/land use~~ type that is heavily managed by humans from multiple aspects including hydrological drainage, nutrient additions, soil cultivation etc. As a consequence,  $\text{CO}_2$  patterns in agricultural streams could potentially be very different than in other ~~land-use/land use~~ types with amplified diel  $\text{CO}_2$  dynamics due to high metabolism and/or quicker response to hydrological events due to effective drainage systems.

70 In addition to the concentration gradient between the stream water and the above air, gas exchange is also highly dependent on the physical conditions at the air-water interface. For stream systems, the gas transfer velocity (often the variable given to describe the efficiency of the air-water gas exchange) is related to a combination of hydrological and morphological conditions of the stream channel, often including slope, velocity and ~~water~~ depth (Raymond et al. 2012; Wallin et al. 2011). All these variables are proxies for describing the ~~water~~-turbulence of the stream ~~water~~, which controls the gas exchange but that is rarely directly measured (Kokic et al. 2018). Agricultural areas are often located in flat landscapes resulting in drainage systems that are low-gradient and slow-flowing (Rhoads et al. 2003; Hughes et al. 2010), conditions that prevent effective air-water gas exchange (Hall & Ulseth, 2019). However, whether the elevated  $p\text{CO}_2$  observed in agricultural streams is an effect of ~~land-~~

use and use specific hydro-morphological stream conditions preventing efficient gas exchange or an effect of high internal (aquatic) or external (terrestrial) CO<sub>2</sub> production is currently unknown.

80 Although recent studies have ~~shown the potential importance of identified~~ agricultural streams as high pCO<sub>2</sub> systems, there are still large knowledge gaps to be filled in order to improve our understanding concerning the influence of these waterbodies in landscape C cycling. Here we present high-resolution (hourly) CO<sub>2</sub> concentration measurements in a Swedish agricultural headwater stream during more than a year ~~of open water season (in total 339 days excluding periods of ice and snow cover)~~. The study aimed to 1) quantify CO<sub>2</sub> concentration levels in an agricultural stream and explore its temporal dynamics, 2) identify  
85 the main drivers causing temporal variability in stream CO<sub>2</sub> concentration and how they might vary with season.

## 2. Methods

### 2.1. Study area

The study was conducted within the 11.3 km<sup>2</sup> Sundbromark (SBM) catchment (59°55'N, 17°32'E), located 5 km NW of the city of Uppsala, Sweden (Figure 1B). The catchment is a part of the hydro-meteorological observatory Marsta that was  
90 established in the late 1940s (Halldin et al. 1999). The 30 year (1960-1991) mean annual temperature for the area is 5.3°C (mean January and July temperatures are -4.5 and 16.0°C, respectively) and with a mean annual precipitation of 535 mm ~~The 30 year (1960-1991) mean annual, January and July temperatures for the area are 5.3°C, -4.5 and 16.0 and with a mean annual precipitation of 535 mm (SMHI).~~ The length of the growing season is on average ca 210 days from early April to the end of  
October (SMHI). The catchment is dominated by agricultural land (86%) mainly used for cereal production and pasture, and  
95 with minor influence of forest (8%) and urban areas (6%). The area is flat with only 28 m elevation difference from 41 m.a.s.l. at the highest point to 13 m.a.s.l. at the catchment outlet (Table 1). The bedrock consists of gneissic granites and the soils are dominated by post glacial clay at lower elevations and with some influence of glacial clay and silt at higher elevations. Although the bedrock does not contain any known carbonates, the soils are alkaline due to glacial carbonate containing deposits resulting in a stream pH ranging between 7.4- and 8.4 (Table 2), and with high electrical conductivity (EC, ranging 791-1908  
100 μS cm<sup>-1</sup>) (Osterman 2018). The nutrient and DOC levels of the stream water (Table 1) are at the lower end (within the 25<sup>th</sup> percentile) of monitored agricultural catchments in Sweden (Linefur et al. 2018; Kyllmar et al. 2014). The oxygen conditions

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are mainly undersaturated (median D.O. = 53%) during the growing season. The arable fields are to a large extent artificially drained with extensive tile drainage pipe systems connected to the stream network. ~~The catchment is a part of the hydro-meteorological observatory Marsta that was established in the late 1940s (Halldin et al. 1999).~~

105 To explore how representative the SBM catchment is for streams draining agricultural areas in the region, a snapshot sampling survey was performed across 10 streams (denoted region UPP 2 in the study by Audet et al. (2019)) of various sizes (catchment area 8.5-740 km<sup>2</sup>) and agricultural influences (30-86%) distributed within a radius of 10 km from the city center of Uppsala (Table S4 Figure 1A, Table S1).

## 2.2. Field sampling and analysis

110 The measurements were conducted ~~during the open water season from September 26, 2017 to December 12, 2018 (in total 339 days of measurements excluding periods of ice, and snow cover and drought).~~ Stream CO<sub>2</sub> concentration was monitored using an EosGP sensor (Eosense, Dartmouth, Canada). The sensor was covered by copper tape in order to avoid biofouling. Sensor accuracy is <1% of the calibrated range (0-2% CO<sub>2</sub>) + 1% of the reading corresponding to a maximum error of ca 0.3 mg C L<sup>-1</sup> based on the maximum CO<sub>2</sub> measured in the current study. The CO<sub>2</sub> sensor was calibrated against known gas standards (400, 1000, 5000 and 20,000 ppm) before and after deployment. No significant drift (exceeding the above given uncertainty) in the instrument was observed during the period. Volume fraction outputs from the sensor were corrected for variations in temperature and pressure (atmospheric and water depth) using the method described in Johnson et al. (2010) and expressed in the unit of mg C L<sup>-1</sup>. ~~Only CO<sub>2</sub> data measured at discharge rates > 0 L s<sup>-1</sup> (i.e. excluding standing water or completely dry conditions) were used in the analysis of the stream CO<sub>2</sub> data (Figure S1).~~

120 Water level, water temperature and EC were measured together with CO<sub>2</sub> concentration at a V-notch weir (Figure S1). Water level was measured using a pressure transducer (1400, MJK Automation, Sweden) mounted in a stilling well representing the stream water level at the V-notch weir. Discharge was calculated from a stage-discharge rating curve based on a series of manual measurements and according to a rating curve presented in Holmqvist (1998). Water temperature and EC were 125 monitored using a thermocouple (Type T) and a CS547A-L conductivity sensor (Campbell, UK), respectively. The sensors

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(except for the pressure transducer) were deployed under the water surface attached to a wooden rod in the center of the stream just upstream of the weir. All sensors were connected to a CR1000X data logger (Campbell, UK) measuring at a 1 min interval and storing average values at a temporal resolution of 30 (in 2017) or 60 (in 2018) minutes, which stored average data (measurements every 1 minute) at a temporal resolution of 30 (in 2017) or 60 (in 2018) minutes.

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Stable isotopic analysis of the dissolved inorganic carbon (DIC) ( $\delta^{13}\text{C-DIC}$ ) was performed on six occasions during the falling limb of the snowmelt discharge peak in 2018 in order to explore the temporal variability in DIC source. At each sampling occasion a 5 samples for analysis of  $\delta^{13}\text{C-DIC}$  were taken in a 60 ml-mL glass vial completely filled with stream water and closed airtight with a rubber septum below the water surface. In order to preserve the sample, 1 ml-mL of highly concentrated  $\text{ZnCl}_2$  solution was injected in each sample (with subsequent release of 1 ml of sample in order to keep atmospheric pressure) directly after sample collection. Samples were kept cold and dark until analysis. Prior to analysis, 2 ml-mL of sample was injected into 12 ml-mL septum-sealed pre-combusted glass vials (Labco Limited) pre-filled with  $\text{HeN}_2$  gas, and pre-injected with 0.1 ml-mL of concentrated phosphoric acid in order to convert all DIC species to  $\text{CO}_2(\text{g})$  (Campeau et al. 2017a). The samples were analyzed using an isotope ratio mass spectrometer (DeltaV Plus, Thermo Fisher Scientific, Bremen, Germany) Gasbench II (Thermo Fisher Scientific, Bremen, Germany) measuring the  $\text{CO}_2$  in the headspace. Each sample was analyzed seven times (sample volume: 100  $\mu\text{L}$  per sample) and the first two injections for each sample were discarded to avoid memory effects, and the mean was taken of the other five to give the final result. The  $\delta^{13}\text{C-DIC}$  values are given in terms of deviation from the known carbonate standards Pee-Dee-Belemnite (PDB) in per mille where R is the isotopic ratio of  $^{13}\text{C}/^{12}\text{C}$ :

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$$\delta^{13}\text{C-DIC} (\text{‰}) = (R_{\text{sample}}/R_{\text{standard}} - 1) \times 1000$$

Precipitation, air temperature and incoming shortwave (global) radiation data (Figure 2) were obtained from the Marsta meteorological observatory located within the catchment ca 2.5 km from the stream sampling station (Halldin et al. 1999). In the absence of direct measurements of photosynthetically active radiation (PAR) shortwave incoming radiation was used as a proxy for available photosynthetic light.

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A spatial sampling campaign for CO<sub>2</sub> concentration, pH, EC and water temperature was conducted on June 21, 2018 across ten agricultural streams (including the SBM stream) located in different catchments around the city of Uppsala (Figure 1A). The sampling was performed between 10.00 and 14.00 during the day. Samples for CO<sub>2</sub> analysis were collected using the headspace method (Hope et al., 2004; Kokic et al. 2015). Briefly, 30 mL bubble-free water were collected in 60 mL polypropylene syringes and equilibrated with a known volume of ambient air by shaking vigorously for 1 min. The equilibrated headspace (15-20 mL) was recovered and analysed on an Ultraportable Greenhouse Gas Analyzer (UGGA) (Los Gatos Research, USA) equipped with a soda lime filter and manual injection port. In situ CO<sub>2</sub> concentration was calculated from the UGGA-determined ppm values using Henry's law considering stream temperature (Weiss 1974), atmospheric pressure, the added ambient air, as well as the water-air volume ratio in the syringe. pH, EC and water temperature were measured in-situ in the streams with handheld instruments, for pH with a pH110 pH-meter (VWR, USA), and for EC and temperature with a HI 99300 (Hanna Instr., USA).

### 2.3. Delineation of the stream network and catchment characteristics

Catchment area and characteristics were calculated in QGIS 3.8 based on a high resolution (2×2 m) digital elevation model (DEM) derived from LIDAR data (GSD Elevation data, grid 2+, Swedish Land Survey). Land use distribution within the catchment was derived from the CORINE Land Cover 2018 product (European Environment Agency), and soil and bedrock characteristics were based on digital versions of the Quaternary deposits (1:25,000 – 1:100,000) and bedrock (1:50,000 – 1:250,000) maps (Swedish Geological Survey).

### 2.4. Data analysis

Out of the total data set (339 days) from the SBM catchment, only data measured at discharge rates > 0 L s<sup>-1</sup> (i.e excluding standing water or completely dry conditions) were used in the analysis of the stream CO<sub>2</sub> data (Figure S1). For further evaluation of the control on stream CO<sub>2</sub> concentration, ~~The continuous~~ the data set-data from the SBM catchment was divided into four periods (Autumn (49 days), Snowmelt (17 days), Spring (91 days) and Dry period (138 days)) according to distinct phases in the hydrograph ~~in order to further analyze the control on stream CO<sub>2</sub> concentration~~ (Figure 3, Table S2). The stream CO<sub>2</sub> dynamics observed among the different periods were examined visually and any hydrological controls on the CO<sub>2</sub> were

175 identified by the presence and direction of CO<sub>2</sub>-discharge hysteresis loops (Evans and Davies, 1998). Similar hysteresis  
analysis was used to investigate diel patterns in the CO<sub>2</sub> concentration data. Spearman's rank correlation coefficient was used  
to test for monotonic relationships between the diel amplitude in stream CO<sub>2</sub> concentration and potential drivers. Correlations  
were considered significant if  $p < 0.05$ . The software JMP 14.2.0 (SAS Institute Inc., Cary, NC, USA) was used for all statistical  
calculations.

### 180 3. Results

The mean air temperature and total precipitation for the entire period (September 26, 2017-December 12, 2018) were 6.8 °C  
and 704 mm, respectively. The summer and autumn of 2018 were dry with generally low precipitation, the exception was on  
July 29 with 82 mm rain within 24 hours (Figure 2). Mean and median stream discharge for the open-waterstudy period were  
30.6 and 0.9 L s<sup>-1</sup>, respectively, and with a total range from 0 to 668 L s<sup>-1</sup> (corresponding to a range from 0 to 5.0 mm day<sup>-1</sup>)  
185 (Figure 3). However, due to high water table exceeding the range of the pressure transducer the absolute peak discharge  
occurring during April 5 to April 7 was missed in the measurements. The large skewness between mean and median discharge  
was an effect of the large number of days without waterflow over the weir during the summer and autumn 2018, 128 days  
(38%) out of the open-waterstudy period. According to frequency analysis, 67% of the days had a mean daily discharge <5 L  
s<sup>-1</sup>. Despite the few days with discharge >100 L s<sup>-1</sup> (7% of the entire period), those days accounted for 69% of the accumulated  
190 discharge. The majority (84%) of these high discharge days occurred during the snowmelt in April.

#### 3.1. General CO<sub>2</sub> patterns

The stream CO<sub>2</sub> concentrations during the entire study period (median and mean 3.44 mg C L<sup>-1</sup> and 3.94 mg C L<sup>-1</sup>, respectively,  
corresponding to a  $p\text{CO}_2$  of 4778 μatm and 5324 μatm) were highly variable (IQR = 3.26 mg C L<sup>-1</sup>) (Figure 3) and displayed  
a bimodal distribution with frequency peaks at ~2.7 mg C L<sup>-1</sup> and ~6.1 mg C L<sup>-1</sup> (Figure S3S2). The lower peak was associated  
195 with the snowmelt and spring period, whereas the higher peak was attributed to the autumn period 2017 and to rain events  
during the dry period of summer/autumn 2018. In addition to the bimodal shape a very distinct peak in frequently measured  
concentrations was observed at ~1.6 mg C L<sup>-1</sup>. This peak was attributed to the minimum concentrations values for the diel  
cycles observed during the spring period.

### 3.2. Controls on stream CO<sub>2</sub> concentration

200 The autumn period started dry with low discharge ( $<3 \text{ L s}^{-1}$ ) for the initial month of measurements. The CO<sub>2</sub> concentrations were at the same time highly dynamic but unrelated to variations in discharge. The CO<sub>2</sub> concentration reached the maximum for the autumn (10.89 mg C L<sup>-1</sup>, which was also the maximum for the entire study period) during late October followed by a decline in CO<sub>2</sub> to ca 2 mg C L<sup>-1</sup> in early November. During November and December four main rain events were identified which all displayed an increasing stream CO<sub>2</sub> concentration with increasing discharge. In three of these events a positive clockwise hysteresis loop was observed (Figure 4) where the CO<sub>2</sub> concentration reached its maximum before the discharge did. At the last event during the autumn 2017, the relationship between CO<sub>2</sub> concentration and discharge was close to linear, but still positive. During the snowmelt period the hydrograph was characterized by a diel cycle with melting during day-time resulting in daily discharge peaks which were suppressed during night-time freezing. In contrast to the autumn events the daily discharge peaks were negatively related to the stream CO<sub>2</sub> concentration, and with an anti-clockwise hysteresis loop where the minimum

210 CO<sub>2</sub> concentration was reached before the highest discharge of the event (Figure 5). After the snowmelt discharge peak the spring and early summer periods (late April to early July) were dry with limited precipitation and with a steady decline in runoff (Figure 3). During this period the CO<sub>2</sub> concentration displayed a pronounced diel cycle with daily maximum and minimum CO<sub>2</sub> concentrations reached during early mornings (06:00) and late afternoons (18:00), respectively (Figure 6). The medium amplitude of the diel CO<sub>2</sub> cycle for this period was 2.03 mg C L<sup>-1</sup>, corresponding to  $p\text{CO}_2 = 2974 \text{ } \mu\text{atm}$  (IQR = 1.23

215 mg C L<sup>-1</sup>, corresponding to  $p\text{CO}_2 = 2212 \text{ } \mu\text{atm}$ ), and with the size of the diel CO<sub>2</sub> concentration amplitude being related to both the daily mean water temperature and the shortwave radiation (Figure 7). The diel pattern displayed a clear negative anti-clockwise CO<sub>2</sub>-streamwater temperature hysteresis loop, where the median CO<sub>2</sub> concentration could differ up to 75% between day and night-time although being measured at the same stream water temperature (Figure 8).

220 From early July the stream dried out and hence no runoff over the V-notch weir was generated. During this period the CO<sub>2</sub> sensor was mostly recording an atmospheric signal. However, for five rain events during the summer and early autumn runoff was generated which allowed stream CO<sub>2</sub> determination for shorter periods (Figure 9). During these runoff events ( $< 2$  days long) high CO<sub>2</sub> concentration pulses were recorded (up to 11 mg C L<sup>-1</sup>). At all events CO<sub>2</sub> was recorded for a longer period

than the discharge as the small dam above the v-notch weir was still water-filled for some time after runoff over the weir  
225 ceased. Also, common for all events was that the stream CO<sub>2</sub> concentration continued to increase although the discharge peak  
had passed. During July 29 a heavy rain storm occurred with 82 mm precipitation ~~during-in~~ 24 hours. Although more than  
15% of the long-term annual mean precipitation fell during one day, low discharge was generated (maximum discharge 6.1 L  
s<sup>-1</sup>) due to high evapotranspiration and dry soils (Figures 3 and 9). However, the rainstorm event resulted in close to the highest  
stream CO<sub>2</sub> concentration (10.81 mg C L<sup>-1</sup>) being observed during the studied period. As soon as the stream was more  
230 permanently refilled in early December and with discharge generated over the weir, the stream CO<sub>2</sub> concentration was back to  
similarly high levels (typically 5-8 mg C L<sup>-1</sup>) as observed in the autumn of 2017.

### 3.3. Sources of DIC

The δ<sup>13</sup>C-DIC data collected during the falling limb of the spring discharge peak (discharge range 130-9.6 L s<sup>-1</sup>) were ranging  
from -13.8 to -12.2‰. This narrow range suggests a relatively constant source of inorganic C during the spring period.  
235 Although there was a tendency towards more negative δ<sup>13</sup>C-DIC values at higher discharge, no significant relationship was  
found (Figure 10). δ<sup>13</sup>C-DIC was also unrelated to the stream CO<sub>2</sub> concentration (data not shown).

### 3.4. Spatial representativeness

The ten streams manually sampled around Uppsala displayed a wide range in CO<sub>2</sub> concentrations (1.8-4.6 mg C L<sup>-1</sup>) on the  
day of sampling (June-21 2018), and with the SBM stream (site 3 in [Table S1](#)) being close to the overall median (SBM, 2.7  
240 mg C L<sup>-1</sup>; overall median, 3.0 mg C L<sup>-1</sup>) (Table S1). Furthermore, the CO<sub>2</sub> concentration manually sampled at SBM was close  
to the sensor recorded CO<sub>2</sub> (2.59 mg C L<sup>-1</sup>) at the hour of sampling. The SBM stream was also close to the spatial median  
DOC concentration but slightly elevated in NO<sub>3</sub> and PO<sub>4</sub>. The CO<sub>2</sub> concentration was on a spatial scale related to pH but  
unrelated to catchment area or ~~land-use~~[land use](#) distribution within the catchment. Furthermore, the CO<sub>2</sub> concentration was on  
a spatial scale unrelated to ~~open-water~~[mean values stream concentrations](#) of DOC, PO<sub>4</sub> and NO<sub>3</sub>, although these variables  
245 were sampled during a different period than the CO<sub>2</sub>.

#### 4. Discussion

In order to produce large scale estimates of the exchange of GHGs between inland surface waters and the atmosphere, a basic requirement is to know the aqueous concentrations of the gases of interest and how they might vary over time. Headwater streams have been identified as “hotspots” for CO<sub>2</sub> emissions (Raymond et al. 2013; Wallin et al. 2018), but there is limited data capturing the temporal resolution, specifically from streams draining agricultural regions, making large scale generalizations uncertain. Due to effective drainage, high nutrient conditions and often high sun-light exposure (due to ~~zero~~ limited tree cover), agricultural streams could potentially be very different in their CO<sub>2</sub> dynamics compared with streams draining other environments. Here we continuously measured stream CO<sub>2</sub> concentration in a headwater catchment dominated by agricultural ~~land use~~ land use (86%) covering more than one year of the snow-free period. In line with findings from similar studies from other environments (arctic tundra, boreal forest, temperate peatlands, alpine) (e.g. Rocher-Ros et al. 2019; Riml et al. 2019; Crawford et al. 2017; Peter et al. 2014; Dinsmore et al. 2013) we found a mixture of controls on stream CO<sub>2</sub> operating at different time-scales generating a highly dynamic stream CO<sub>2</sub> concentration pattern. These time-scales covers seasonal patterns to diel cycles, or even shorter scales associated ~~to~~ with discharge events. Both the magnitude of CO<sub>2</sub> concentrations, and their associated temporal dynamics were found to be high in the current agricultural stream when compared with the literature. The mean CO<sub>2</sub> concentration (3.94 mg C L<sup>-1</sup> corresponding to a pCO<sub>2</sub> of 5324 µatm) is at the high end when compared with other high-frequency CO<sub>2</sub> data sets covering low-order (<3<sup>rd</sup> stream order) catchments draining multiple environments, including arctic tundra, boreal forest, hemi-boreal forest, temperate forest, temperate peatlands and alpine areas (typically ranging from ca 0.2 to 6 mg C L<sup>-1</sup>) (Crawford et al. 2017; Natchimuthu et al. 2017; Peter et al. 2014; Dinsmore et al. 2013). Still, CO<sub>2</sub> concentrations in SBM do not seem to be exceptionally high compared to snapshot-based data from other agricultural streams.

The spatial variability seen in this study, although only based on snapshot samples, and previous studies indicate that CO<sub>2</sub> concentrations in agricultural streams are comparably high (Borges et al. 2018; Bodmer et al., 2016; Sand-Jensen & Staehr, 2012). In addition, the observed temporal dynamics presented here are, to our knowledge, among the most pronounced in the literature, although the number of high-frequency stream CO<sub>2</sub> data sets are limited. For example, the rapid decrease in stream

CO<sub>2</sub> during the autumn of 2017, the strong diel cycle (diel amplitude up to almost  $<5.0$  mg C L<sup>-1</sup>) during the spring/early summer period, or the rapid and high CO<sub>2</sub> pulses (up to  $<11.0$  mg C L<sup>-1</sup>) occurring in accordance to rain events during the dry late summer/autumn period. These high CO<sub>2</sub> dynamics clearly illustrate the need for continuous high frequency CO<sub>2</sub> concentration measurements in streams in general, and in agricultural streams more specifically. Without such high-frequency data, representative estimates of agricultural stream CO<sub>2</sub> will be associated with high uncertainty. Although based on measurements from a single stream, these findings in turn indicate that current large-scale stream CO<sub>2</sub> emission estimates (e.g. [Raymond et al. 2013](#); [Humborg et al. 2010](#)), which are largely based on snapshot concentration data with low (or no) resolution in time, might be specifically uncertain for agricultural regions.

According to our continuous data the highly dynamic pattern in stream CO<sub>2</sub> concentration is driven by a complex interplay of hydrology and biology. The high autumn concentrations observed both in 2017 and 2018 are likely an effect of high respiration of organic matter in the stream channel and/or in the adjacent soil water (Figure [3e3D](#)). This is supported by efficient aquatic microbial DOC degradation ( $<800$   $\mu$ g C L<sup>-1</sup> d<sup>-1</sup>) observed during the autumn period across the ten streams (agricultural ~~land-use~~land use, 30-86%) included in the spatial sampling campaign (Peacock et al. unpublished [20192020](#)). This ~~shc~~ould be compared with organic C degradation rates determined in boreal forest and mire streams displaying typically lower rates ( $<300$   $\mu$ g C L<sup>-1</sup> d<sup>-1</sup>, Berggren et al. 2009). The positive CO<sub>2</sub>-discharge relationships indicated that event flow pathways, ~~whether those are more surficial or different spatially~~, were in contact with soils with higher concentrations of CO<sub>2</sub> compared to flow pathways during base flow (Evans & Davies, 1998; Seibert et al., 2009). Also, the clock-wise shape of the hysteresis loop suggests that there is a buildup of CO<sub>2</sub> in the catchment that is flushed out during rain events (Figure 4). The CO<sub>2</sub> pool seems to be limited as the CO<sub>2</sub> concentration drops before the maximum discharge peak occurs, or that vertical patterns in the CO<sub>2</sub> soil profile control the stream CO<sub>2</sub> dependent on dominating flow paths (Evans and Davies, 1998; Öquist et al. 2009). This could explain that the stream CO<sub>2</sub> increase did not reach any source limitation at rain events of lower magnitude (Figure [4d4D](#)). Similar positive CO<sub>2</sub> concentration-discharge patterns have been observed across different low-order streams (e.g. Crawford et al. 2017; Dinsmore et al. 2013), but the absolute patterns are often concluded to be highly site-specific and even event-specific. Here we suggest, by exploring the hysteresis loops, that such positive relationships are influenced by the size of the

available catchment CO<sub>2</sub> pool or the hydrological connectivity to it. In a highly drained low-elevation agricultural landscape where much of the stream runoff is generated through drainage pipes (Castellano et al. 2019), the extent and spatial distribution of these terrestrial source areas and connections between ground- and surface water are central for the CO<sub>2</sub> patterns observed in the stream. ~~Similar results~~ Strong hydrological control hasve been found for DOC in agricultural streams in USA and France, ~~where high discharge events flush allochthonous DOC, via subsurface drainage pipes, into streams (Morel et al. 2009; Royer & David, 2005). In contrast to the seasonally variable CO<sub>2</sub>-discharge response patterns observed in the current study, Morel et al. (2009) suggested ~~that~~ Furthermore, fluvial CO<sub>2</sub> concentrations have previously been found to positively correlate with DOC concentrations (Campeau & del Giorgio, 2014). We therefore suggest that, in agricultural catchments that also feature wetland soils, CO<sub>2</sub> delivery into the stream DOC is would be non-limited (Morel et al. 2009), and would continue to rise until the maximum discharge peak is reached. Whether this discrepancy in source limitation between CO<sub>2</sub> and DOC (although based on different studies and environments) indicate differences in the source areas of the different carbon components require further investigation. ~~Whether this discrepancy in source limitation between CO<sub>2</sub> and DOC (although based on different studies and environments) indicate differences in the source areas require further investigation.~~~~

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In contrast to the patterns observed during the autumn, during the snow melt period the stream CO<sub>2</sub> was diluted ~~at~~ when discharge increases following a diel pattern (Figure 5). The melting and freezing between day and night-time suggests that melt-water from the surface snowpack during day time to a larger extent reached the stream without picking up an elevated CO<sub>2</sub> signal. Similar dilution patterns in conjunction with snowmelt have been observed in catchments of various ~~land use~~ land use but specifically in peatland catchments with limited forest cover (e.g. Wallin et al. 2013). The similarity between this agricultural catchment and open peatlands could potentially be the effect of an efficient melting of the snowpack. Both non-forested peatlands and agricultural fields are open areas subject to direct sunlight, and wind and rain exposure, while the soil under the snow remains frozen. As a result, a large share of the melt-water will never infiltrate the soil but instead reach the surface drainage system as overland flow (Laudon et al. 2007). This is further accompanied by the low hydraulic conductivity of clay soils, which are dominating the catchment of the current study. Although we did not capture the 2-3 days of peak spring flood (due to a water level out of the range of the pressure transducer) it was evident that the stream CO<sub>2</sub> concentration was

diluted from ca 6.0 mg C L<sup>-1</sup> to ca 2.0 mg C L<sup>-1</sup> during these days, something that is further supported by the similar drop in EC during the peak spring flood from ca 900 to ca 150  $\mu$ S cm<sup>-1</sup>. However, as soon as the ~~absolute~~-discharge peak passed, the stream CO<sub>2</sub> concentration recovered rapidly to the pre-peak levels suggesting a shift to hydrological pathways that mobilize a high CO<sub>2</sub> pool, again supported by the concurrent increase in EC. April and May 2018 were characterized by warm and clear  
325 weather with an average 4.2°C higher air temperature and 255 more sun hours than the 30-year mean (1961-1990, SMHI). Altogether, this stimulates a kick-start of the aquatic primary production upon snowmelt, which likely explains the steady decline in CO<sub>2</sub> that occurred during late April/early May. During the spring and early summer, a strong diel pattern in CO<sub>2</sub> concentration further developed, likely driven by aquatic primary production consuming CO<sub>2</sub> during day-time. Such diel CO<sub>2</sub> patterns are commonly observed in stream CO<sub>2</sub> time series at base-flow or during receding flow conditions (e.g. Riml et al.  
330 2019; Peter et al. 2014) and are especially pronounced in amplitude in nutrient-rich streams or in streams without canopy shading (Alberts et al. 2017; Crawford et al. 2017; Rocher-Ros et al. 2019). Initial evaluation of the  $\delta^{13}\text{C}$ -DIC data collected during the spring period suggests a relatively steady mixture of geogenic and biogenic DIC although somehow related to variations in discharge (Figure 11-10). However, given the suppressed stream CO<sub>2</sub> during the spring period, together with the strong diel cycle caused by aquatic primary production, fractionation of a strict biogenic DIC pool (with a  $\delta^{13}\text{C}$ -DIC from -28  
335 to -20‰) could theoretically push the  $\delta^{13}\text{C}$ -DIC towards the less negative values observed in the current study (from -13.8 to -12.2‰) (Campeau et al. 2017b). Combined studies on aquatic metabolism, C dynamics and stable isotopic composition would further be recommended to disentangle the dynamic CO<sub>2</sub> source patterns in this type of agricultural system.

The spring and early summer of 2018 were generally dry leading to the stream channel drying out during long periods. The  
340 rapid rewetting periods (< 2 days) that occurred following larger precipitation events resulted in high CO<sub>2</sub> pulses (3-11 mg C L<sup>-1</sup>) generally exceeding the overall median level of stream CO<sub>2</sub> (3.44 mg C L<sup>-1</sup>) observed during the study period. The intermittent nature of streams, with distinct drying and rewetting episodes, is known to generate high CO<sub>2</sub> concentration pulses and subsequent emissions (Marcé et al. 2019). Such rapid pulses are generally suggested to be a result of intense respiration in the stream bed sediments upon rewetting, or due to a rapid mobilization of terrestrial C, both organic (DOC) and inorganic  
345 (CO<sub>2</sub>) in connection to precipitation events. However, the findings of high CO<sub>2</sub> pulses upon rewetting have mostly been done



in areas that display pronounced dry and wet seasons e.g. Mediterranean areas or Australia (e.g. Gomez-Gener et al. 2015; Looman et al. 2017). Here we show that such stream intermittency can also cause high and rapid CO<sub>2</sub> pulses in a Swedish agricultural setting, highlighting the need for expanding the geographical coverage of studies that investigate stream intermittency in relation to GHG dynamics and emissions. [Areas that display stream intermittency will likely also increase in the future given the predicted changes in temperature and precipitation patterns.](#) An obvious tool in this work is the use of continuous sensor-based measurements which allow capturing the episodic and unpredictable nature of these phenomena.

## 5. Conclusions

It is evident from the current study that the stream CO<sub>2</sub> dynamics in an agricultural headwater catchment are highly variable across a variety of different time-scales and with an interplay of hydrological and biological controls. The hydrological control was strong (although with both positive as well as negative influences dependent on season) and rapid in response to rainfall and snowmelt events. However, during growing-season baseflow and receding flow conditions, the aquatic primary production seems to control the stream CO<sub>2</sub> dynamics, which in turn sets the basis for atmospheric emissions. [During the dry summer period, rapid rewetting following precipitation events generated high CO<sub>2</sub> pulses exceeding the overall median level of stream CO<sub>2</sub> \(up to 3 times higher\). This finding thus highlights the importance of stream intermittency in agricultural areas and its effect on stream CO<sub>2</sub> dynamics.](#) Given the observed high levels of CO<sub>2</sub> and its temporally variable nature, agricultural streams clearly need more attention in order to understand and incorporate these considerable dynamics in large scale extrapolations.

## 6. Data availability

Data is available from the Uppsala University data repository, LINK WILL BE ADDED

## 7. Author contribution

MBW and MW brought the idea and designed the study. MBW funded and instrumented the catchment and analysed the data. MW conducted the GIS analysis. JA, MP and ES provided ideas and data. MBW wrote the manuscript with great support from all co-authors.

## 8. Competing interests

The authors declare that they have no conflict of interest

## 370 9. Acknowledgements

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## 375 References

- Alberts, J.M., Beaulieu, J.J. and Buffam, I., 2017, Watershed land use and seasonal variation constrain the influence of riparian canopy cover on stream ecosystem metabolism. *Ecosystems* 20(3), 553-567.
- Audet, J., Bastviken, D., Bundschuh, M., Buffam, I., Feckler, A., Klemedtsson, L., Laudon, H., Löfgren, S., Natchimuthu, S., Öquist, M., Peacock, M. and Wallin, M.B. 2019, Forest streams are important sources for nitrous oxide emissions. *Global Change Biology*, [26\(2\)](#) doi:10.1111/gcb.14812
- 380 Berggren M., Laudon H., Jansson M., 2009, Hydrological control of organic carbon support for bacterial growth in boreal headwater streams. *Microbial Ecology*, 57, 170-178. doi:10.1007/s00248-008-9423-6
- Bodmer, P., Heinz, M., Pusch, M., Singer, G. and Premke, K., 2016, Carbon dynamics and their link to dissolved organic matter quality across contrasting stream ecosystems. *Science of The Total Environment*, 553, 574–586.
- 385 Borges, A.V., Darchambeau, F., Lambert, T., Bouillon, S., Morana, C., Brouyère, S., Hakoun, V., Jurado, A., Tseng, H.-C., Descy, J.-P. and Roland, F.A.E., 2018, Effects of agricultural land use on fluvial carbon dioxide, methane and nitrous oxide concentrations in a large European river, the Meuse (Belgium). *Science of The Total Environment*, 610–611, 342–355.
- Campeau, A., Bishop, K., Nilsson, M. B., Klemedtsson, L., Laudon, H., Leith, F. I., Öquist, M. G., Wallin, M. B., 2018, Stable carbon isotopes reveal soil-stream DIC linkages in contrasting headwater catchments, *Journal of Geophysical Research – Biogeosciences*, 123 (1), 149-167, doi:10.1002/2017JG004083
- 390 Campeau, A., Bishop K., Billett, M. F., Garnett, M. H., Laudon, H., Leach, J. A., Nilsson, M. B., Öquist, M. G., Wallin, M. B., 2017a, Aquatic export of young dissolved and gaseous carbon from a pristine boreal fen: implications for peat carbon stock stability, *Global Change Biology*, 23 (12), 5523-5536, doi:10.1111/gcb.13815
- 395 Campeau, A., Wallin, M. B., Giesler, R., Löfgren, S., Mörth, C.-M., Schiff, S. L., Venkiteswaran, J. J., Bishop, K., 2017b, Multiple sources and sinks of dissolved inorganic carbon across Swedish streams, refocusing the lens of stable C isotopes. *Scientific Reports*, 7, 9158, doi:10.1038/s41598-017-09049-9
- Castellano, M.J., Archontoulis, S.V., Helmers, M.J., Poffenbarger, H.J. and Six, J., 2019, Sustainable intensification of agricultural drainage. *Nature Sustainability*, 2(10), 914-921.
- 400 Crawford, J. T., Stanley, E. H., Dornblaser, M. M., & Striegl, R. G., 2017, CO<sub>2</sub> time series patterns in contrasting headwater streams of North America. *Aquatic Sciences*, 79(3), 473-486. doi:10.1007/s00027-016-0511-2
- [Deirmendjian, L., Anschutz, P., Morel, C., Mollier, A., Augusto, L., Loustau, D., Cotovicz, L.C., Buquet, D., Lajaunie, K., Chaillou, G., Voltz, B., Charbonnier, C., Poirier, D. and Abril, G., 2019, Importance of the vegetation-groundwater-stream continuum to understand transformation of biogenic carbon in aquatic systems – A case study based on a pine-maize comparison in a lowland sandy watershed \(Landes de Gascogne, SW France\). \*Science of the Total Environment\* 661, 613-629. doi:10.1016/j.scitotenv.2019.01.152](#)
- 405 [doi:10.1016/j.scitotenv.2019.01.152](#)
- Dinsmore, K. J., M. B. Wallin, M. S. Johnson, M. F. Billett, K. Bishop, J. Pumpanen & A. Ojala, 2013. Contrasting CO<sub>2</sub> concentration discharge dynamics in headwater streams: A multi-catchment comparison. *Journal of Geophysical Research: Biogeosciences*, 118, 445-461.
- 410 Dinsmore, K. J. & M. F. Billett, 2008, Continuous measurement and modeling of CO<sub>2</sub> losses from a peatland stream during stormflow events. *Water Resources Research*, 44, 11.
- Evans, C., Davies, T.D., 1998, Causes of Concentration/Discharge Hysteresis and its Potential as a Tool for Analysis of Episode Hydrochemistry. *Water Resources Research* 34, 129–137.

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- Gómez-Gener, L., Obrador, B., von Schiller, D., Marcé, R., Casas-Ruiz, J.P., Proia, L., Acuña, V., Catalán, N., Muñoz, I., Koschorreck, M., 2015. Hot spots for carbon emissions from Mediterranean fluvial networks during summer drought. *Biogeochemistry* 125, 409–426.
- 415 Hall Jr, R.O. and Ulseth, A.J., 2019, Gas exchange in streams and rivers. *Wiley Interdisciplinary Reviews: Water*, p.e1391
- Halldin, S., Bergström, H., Gustafsson, D., Dahlgren, L., Hjelm, P., Lundin, L.C., Mellander, P.E., Nord, T., Jansson, P.E., Seibert, J., Stähli, M., Szilágyi Kishné, A. and Smedman, A.S. (1999) Continuous long-term measurements of soil–plant–atmosphere variables at an agricultural site. *Agricultural and Forest Meteorology*, 98–99, 75–102.
- 420 Holmqvist, M., 1998, Avrinningsdynamik i fem små områden. Vattenbalans, recession, magasininkoefficient och dynamiskt vattenmagasin. MSc thesis, Uppsala University, 54 pp
- Hope, D., S. M. Palmer, M. F. Billett, and J. J. Dawson, 2004, Variations in dissolved CO<sub>2</sub> and CH<sub>4</sub> in a first-order stream and catchment: an investigation of soil–stream linkages, *Hydrol. processes*, 18, 3255–3275.
- Hughes, R.M., Herlihy, A.T. and Kaufmann, P.R., 2010, An Evaluation of Qualitative Indexes of Physical Habitat Applied to Agricultural Streams in Ten US States 1. *JAWRA Journal of the American Water Resources Association*, 46(4), 792–806.
- 425 Johnson, M. S., M. F. Billett, K. J. Dinsmore, M. Wallin, K. E. Dyson, and R. S. Jassal, 2010, Direct and continuous measurement of dissolved carbon dioxide in freshwater aquatic systems - methods and applications, *Ecohydrology*, 3, 68–78, doi:10.1002/eco.95.
- 430 Johnson, M. S., Weiler, M., Couto, E. G., Riha, S. J., & Lehmann, J., 2007, Storm pulses of dissolved CO<sub>2</sub> in a forested headwater Amazonian stream explored using hydrograph separation. *Water Resources Research*, 43(11), w11201.
- Kocic J, Wallin MB, Chmiel HE, Denfeld BA, Sobek S., 2015, Carbon dioxide evasion from headwater systems strongly contributes to the total export of carbon from a small boreal lake catchment. *J Geophys Res-Biogeo*. 120:13–28. doi:10.1002/2014JG002706
- 435 Kocic J, Sahlée E, Sobek S, Vachon D, Wallin MB, 2018, High spatial variability of gas transfer velocity in streams revealed by turbulence measurements, *Inland Waters*, 8:4, 461–473, doi:10.1080/20442041.2018.1500228
- Kyllmar, K., Forsberg, L. S., Andersson, S., & Martensson, K., 2014, Small agricultural monitoring catchments in Sweden representing environmental impact. *Agriculture Ecosystems & Environment*, 198, 25– 35. doi:10.1016/j.agee.2014.05.016
- Laudon, H., V. Sjöblom, I. Buffam, J. Seibert, and M. Mörth, 2007, The role of catchment scale and landscape characteristics for runoff generation of boreal streams, *J. Hydrol.*, 344(3–4), 198–209,
- 440 Leith, F. I., Dinsmore, K. J., Wallin, M. B., Billett, M. F., Heal, K. V., Laudon, H., Öquist, M. G., Bishop, K., 2015, Carbon dioxide transport across the hillslope–riparian–stream continuum in a boreal headwater catchment, *Biogeosciences*, 12, 1881–1892, doi:10.5194/bg-12-1-2015
- Linefur, H., Norberg, L., Kyllmar, K., Andersson, S. och Blomberg, M., 2018, Växtnäringsförluster i små jordbruksdominerade avrinningsområden 2016/2017. Uppsala: Sveriges lantbruksuniversitet. (Ekohydrologi, 155).
- 445 Looman, A., Maher, D.T., Pendall, E., Bass, A., Santos, I.R., 2017. The carbon dioxide evasion cycle of an intermittent first-order stream: contrasting water–air and soil–air exchange. *Biogeochemistry*, 132, 87–102.
- Marcé, R., Obrador, B., Gómez-Gener, L., Catalán, N., Koschorreck, M., Arce, M.I., Singer, G. and von Schiller, D., 2019, Emissions from dry inland waters are a blind spot in the global carbon cycle. *Earth-Science Reviews* 188, 240–248.
- 450 Morel, B., Durand, P., Jaffrezic, A., Gruau, G., & Molenat, J., 2009, Sources of dissolved organic carbon during stormflow in a headwater agricultural catchment. *Hydrological Processes*, 23(20), 2888–2901.
- Natchimuthu, S., Wallin, M. B., Klemedtsson, L., Bastviken, D., 2017, Spatio-temporal patterns of stream methane and carbon dioxide emissions in a hemiboreal catchment in Southwest Sweden, *Scientific Reports*, 7, 39729, doi:10.1038/srep39729
- Osborne, B., Saunders, M., Walmsley, D., Jones, M., Smith, P., 2010, Key questions and uncertainties associated with the assessment of the cropland greenhouse gas balance. *Agriculture, Ecosystems & Environment* 139, 293–301, doi:10.1016/j.agee.2010.05.009
- 455 Osterman, M., 2018, Carbon dioxide in agricultural streams - magnitude and patterns of an understudied atmospheric carbon source, MSc thesis, Uppsala University, 58 pp
- Öquist, M. G., M. Wallin, J. Seibert, K. Bishop, and H. Laudon, 2009, Dissolved inorganic carbon export across the soil/stream interface and its fate in a boreal headwater stream, *Environmental Science & Technology*, 43(19), 7364–7369, doi:10.1021/es900416h.
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- Peter, H., Singer, G. A., Preiler, C., Chiffard, P., Steniczka, G., & Battin, T. J., 2014, Scales and drivers of temporal pCO<sub>2</sub> dynamics in an Alpine stream. *Journal of Geophysical Research: Biogeosciences*, 119(6), 1078-1091. doi:10.1002/2013JG002552
- 465 Ramankutty, N., Evan, A.T., Monfreda, C. and Foley, J.A., 2008, Farming the planet: 1. Geographic distribution of global agricultural lands in the year 2000. *Global Biogeochemical Cycles*, 22(1).
- Raymond PA, et al. 2013, Global carbon dioxide emissions from inland waters. *Nature*, 503 (7476), 355-359.
- Raymond, P. A., C. J. Zappa, D. Butman, T. L. Bott, J. Potter, P. Mulholland, A. E. Laursen, W. H. McDowell & D. Newbold 2012, Scaling the gas transfer velocity and hydraulic geometry in streams and small rivers. *Limnology and Oceanography - Fluids and Environments*, 2, 41-53.
- 470 Rhoads, B.L., Schwartz, J.S. and Porter, S., 2003, Stream geomorphology, bank vegetation, and three-dimensional habitat hydraulics for fish in midwestern agricultural streams. *Water Resources Research*, 39(8).
- Riml, J., Campeau, A., Bishop, K., Wallin, M. B., 2019, Spectral decomposition of high-frequency CO<sub>2</sub> concentrations reveals soil-stream linkages, *Journal of Geophysical Research - Biogeosciences*, doi:10.1029/2018JG004981
- Rocher-Ros, G., Sponseller, R.A., Bergström, A.-K., Myrstener, M. and Giesler, R., 2019, Stream metabolism controls diel patterns and evasion of CO<sub>2</sub> in Arctic streams. *Global Change Biology*, doi:10.1111/gcb.14895
- 475 Royer, T. V., & David, M. B., 2005, Export of dissolved organic carbon from agricultural streams in Illinois, USA. *Aquatic Sciences*, 67(4), 465-471.
- Sand-Jensen, K., Staehr, P.A., 2012, CO<sub>2</sub> dynamics along Danish lowland streams: water-air gradients, piston velocities and evasion rates. *Biogeochemistry* 111, 615-628. doi:10.1007/s10533-011-9696-6
- 480 Seibert, J., Grabs, T., Köhler, S., Laudon, H., Winterdahl, M., Bishop, K., 2009, Linking soil- and stream-water chemistry based on a Riparian Flow-Concentration Integration Model. *Hydrology and Earth System Sciences* 13, 2287-2297. https://doi.org/10.5194/hess-13-2287-2009
- Wallin, M. B., Campeau, A., Audet, J., Bastviken, D., Bishop, K., Kokic, J., Laudon, H., Lundin, E., Löfgren, S., Natchimuthu, S., Sobek, S., Teutschbein, C., Weyhenmeyer, G., Grabs, T., 2018, Carbon dioxide and methane emissions of Swedish low-order streams – a national estimate and lessons learnt from more than a decade of observations, *Limnology and Oceanography - Letters*, 3 (3), 156-167, doi:10.1002/lol2.10061
- 485 Wallin, M. B., Grabs, T., Buffam, I., Laudon, H., Ågren, A., Öquist, M. G., Bishop, K., 2013, Evasion of CO<sub>2</sub> from streams – The dominant component of the carbon export through the aquatic conduit in a boreal catchment, *Global Change Biology*, 19(3), 785-797, doi:10.1111/gcb.12083.
- 490 Wallin, M. B., Öquist, M. G., Buffam, I., Billett, M. F., Nisell, J., Bishop, K. H., 2011, Spatiotemporal variability in the gas transfer coefficient (KCO<sub>2</sub>) of boreal streams; implications for large scale estimates of CO<sub>2</sub> evasion, *Global Biogeochemical Cycles*, 25, GB3025, doi:10.1029/2010GB003975

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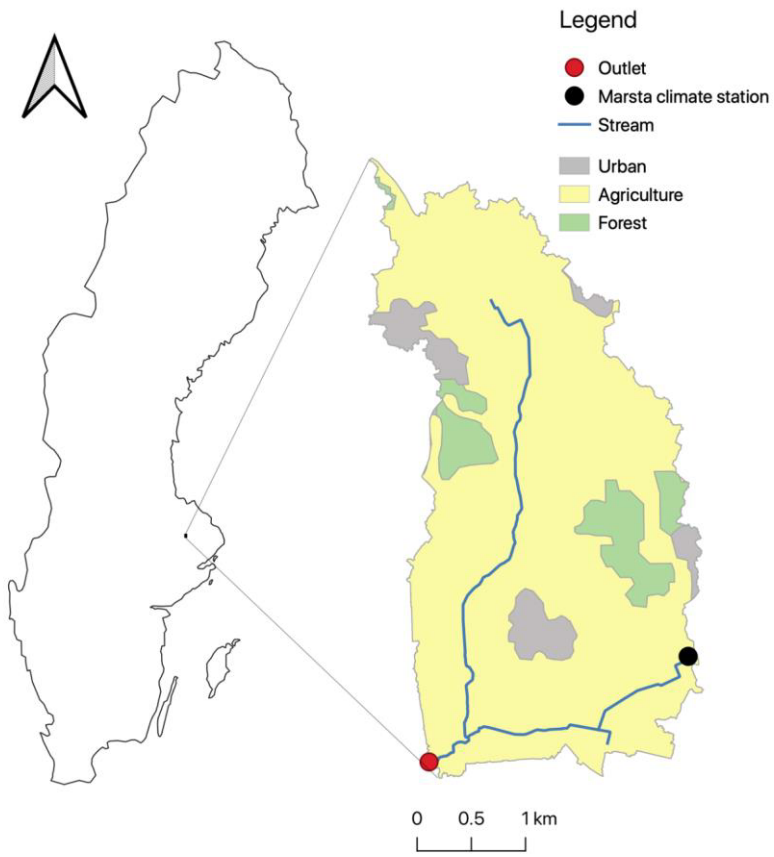
495 **Table 1. Catchment characteristics of the Sundbromark (SBM) catchment**

Catchment area (km <sup>2</sup> )	11.3
Elevation range (masl)	13-41
<i>Land use distribution (%)</i>	
Agricultural land	86
Forest	8
Urban	6
<i>Main Soil type distribution (%)</i>	
Post glacial clay	48
Glacial silt	22
Glacial clay	14
Sandy till	12
<i>Main bedrock distribution (%)</i>	
Granodiorite granite	89
Tonalite granodiorite	6
Dacite rhyolite	3
Granite	2

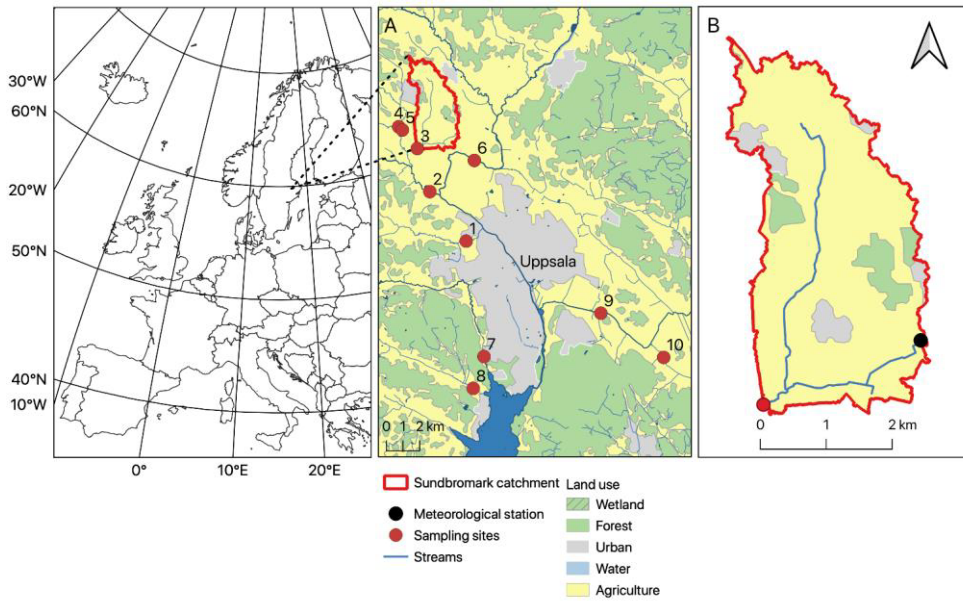
**Table 2. Water chemistry at the outlet of the SBM catchment collected during June-November 2017 (n = 8) (Osterman 2018).**

	Median	Mean	Min-Max
pH	7.7	7.8	7.4-8.4
EC ( $\mu\text{S cm}^{-1}$ )	1082	1273	791-1908
NH <sub>4</sub> -N ( $\text{mg L}^{-1}$ )	0.10	0.08	0.01-0.1
NO <sub>3</sub> -N ( $\text{mg L}^{-1}$ )	0.7	1.9	0.09-6.5
PO <sub>4</sub> -P ( $\text{mg L}^{-1}$ )	0.07	0.09	0.01-0.2
DOC ( $\text{mg L}^{-1}$ )	10.0	9.6	4.2-13.1
D.O. (%)	53	62	31-119









**Figure 1. Location of the study with A) sampled sites of the spatial survey, and B) The Sundbromark (SBM) catchment. Catchment delineation and Land use distributions within the SBM catchment are given according to (GSD elevation data, grid 2+ (©Swedish Land Survey) and; CORINE Land Cover 2018, (European Environment)). The stream-based measurements were conducted at the catchment outlet (red dot) whereas the meteorological data derived from the Marsta Observatory (black dot).**

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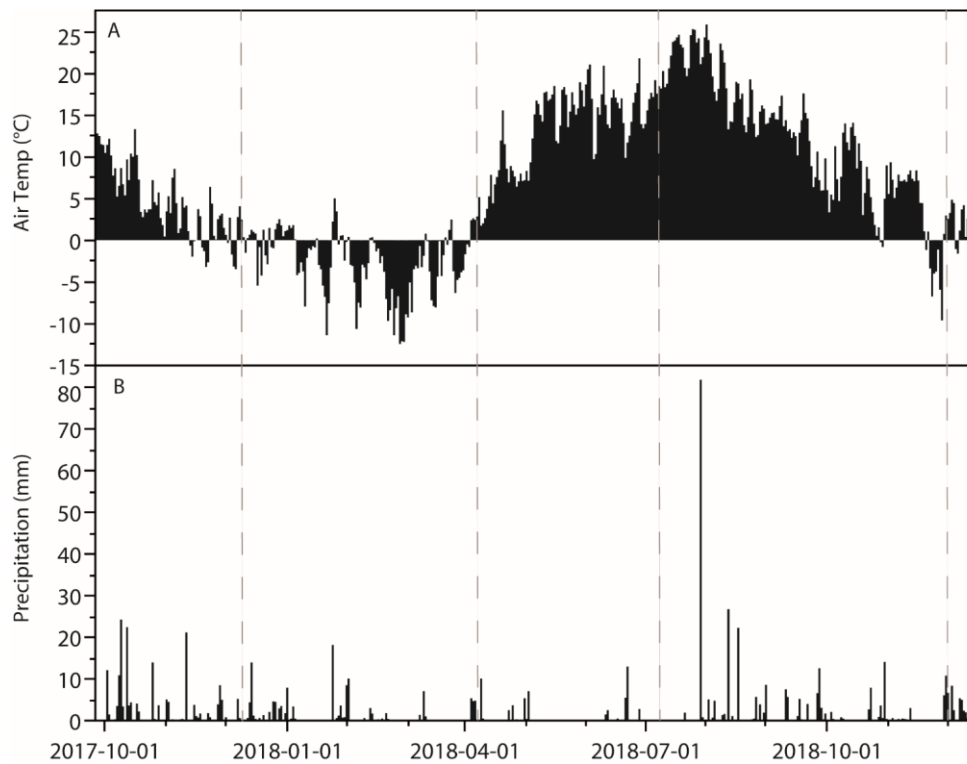


Figure 2. (A) Daily mean air temperature, and (B) daily precipitation during the study period (Sep 26, 2017-Dec 12, 2018) at the Marsta Observatory. Due to malfunctioning sensor the precipitation data for July 29 2018 is collected from the nearby (3 km) SMHI station, Ärna. The dotted lines refer to the hydrological periods displayed in figure 3.

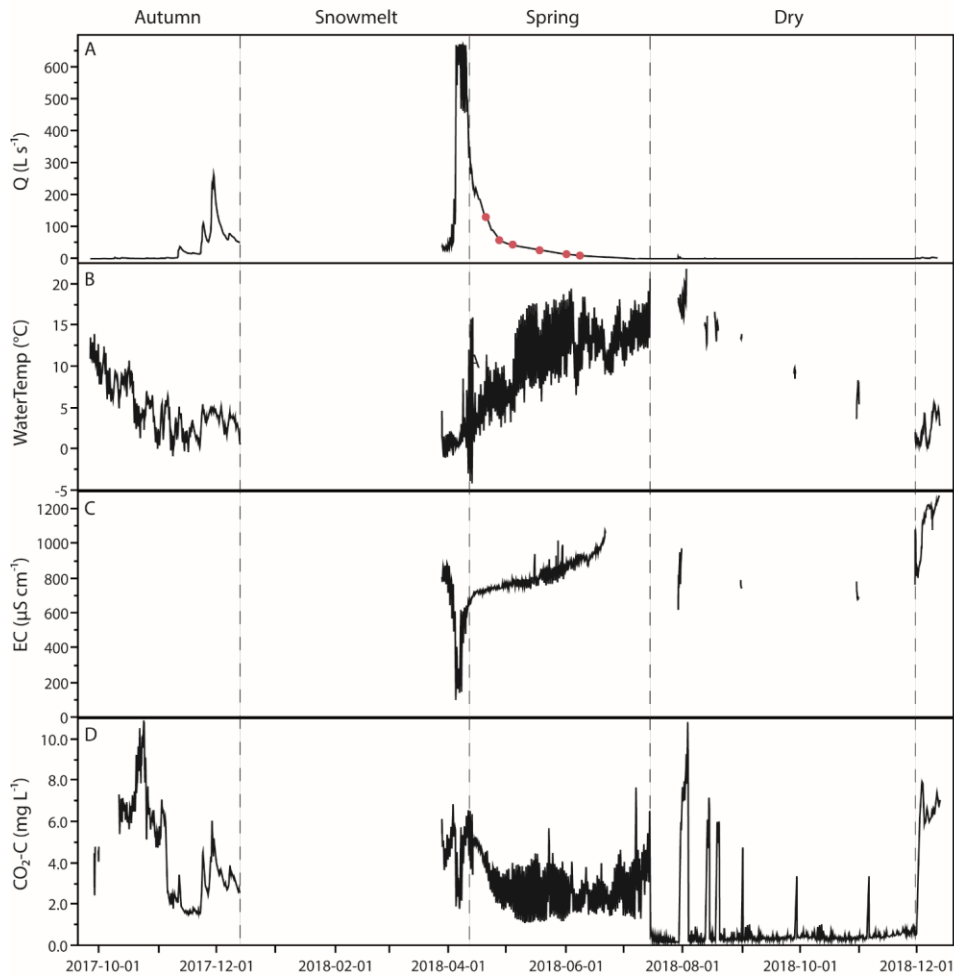
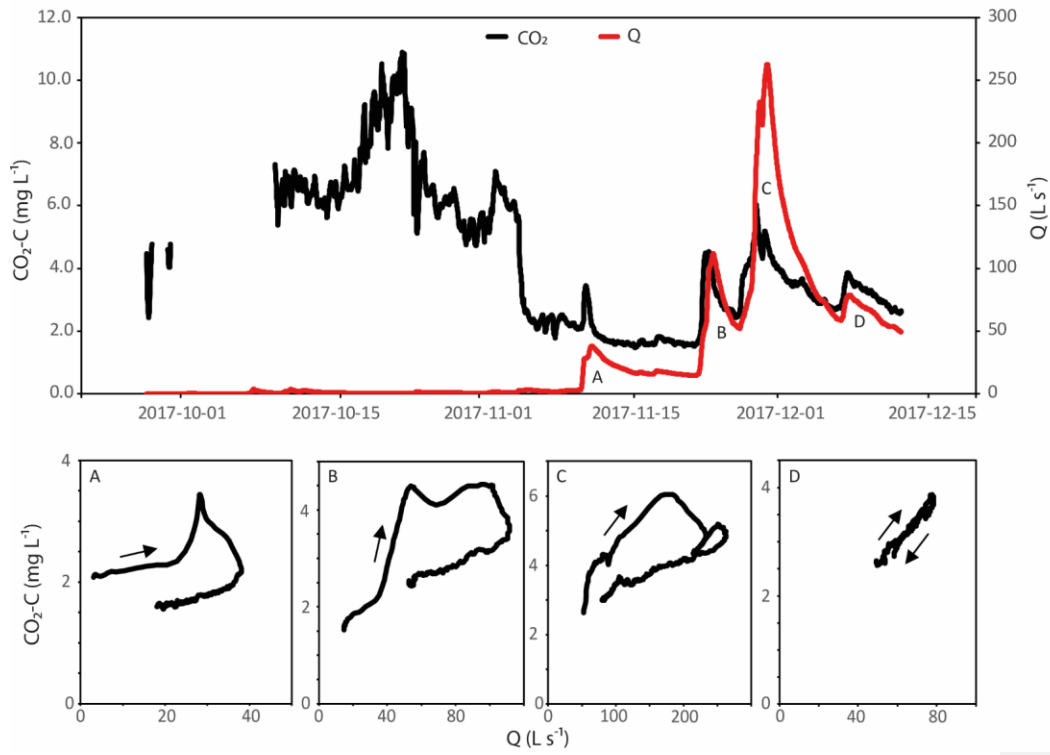
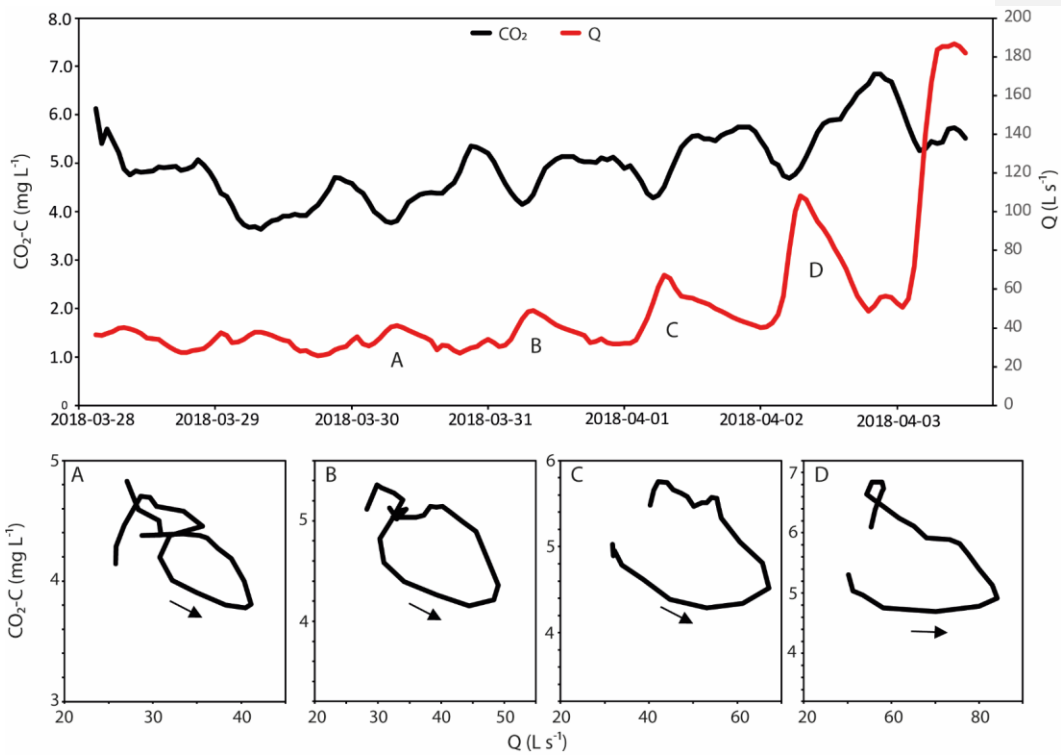


Figure 3. Time series of A) stream discharge ( $Q$ ) with sampling days for  $\delta^{13}\text{C-DIC}$  highlighted by red dots, B) stream water temperature, C) electrical conductivity (EC), and D)  $\text{CO}_2$  concentration for the study period September 26, 2017-December 12, 2018, with break for the ice- and snow-covered period December-March. The  $\text{CO}_2$  data include periods when the sensor was above the water surface during dry periods in summer/autumn of 2018.



**Figure 4.** Stream CO<sub>2</sub> concentration (black) and discharge (red) for the autumn 2017 period with CO<sub>2</sub>-Q hysteresis plots for four rain events.

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**Figure 5.** Stream CO<sub>2</sub> concentration (black) and discharge (red) for the snowmelt period 2018 with CO<sub>2</sub>-Q hysteresis plots for four discharge events.

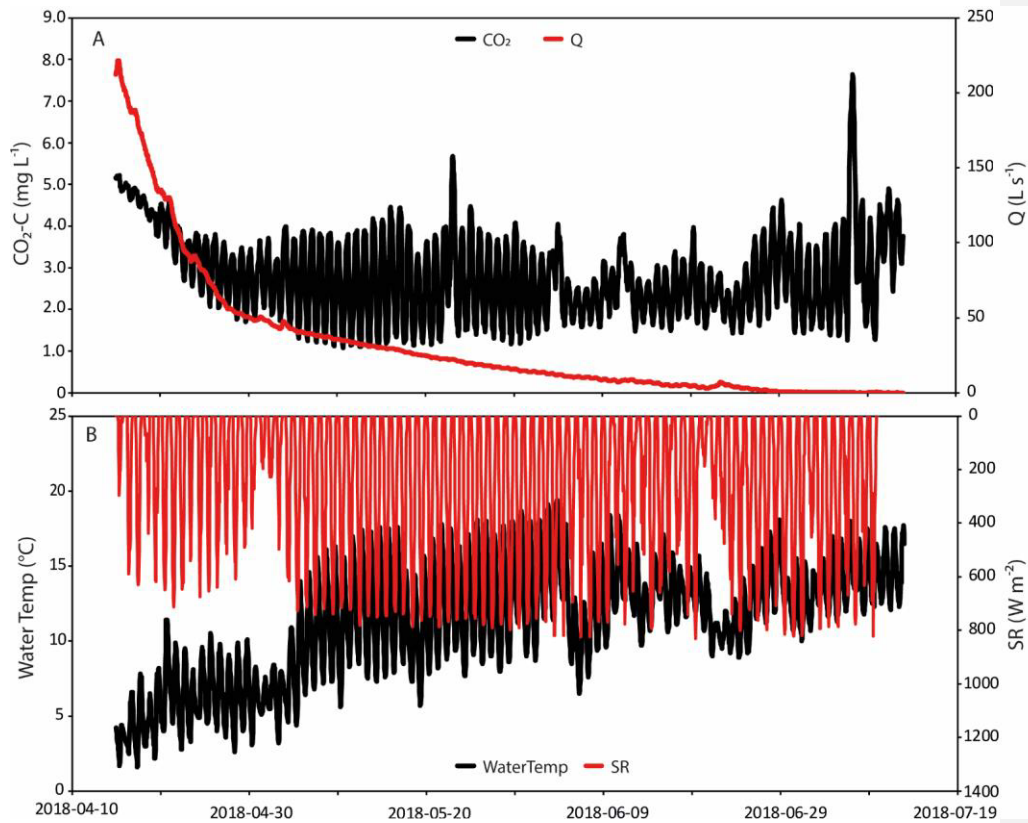
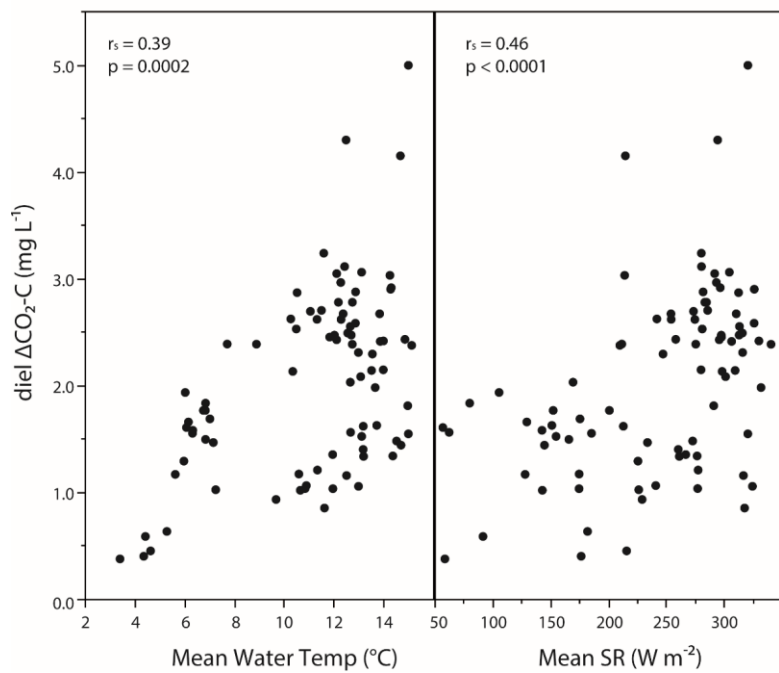


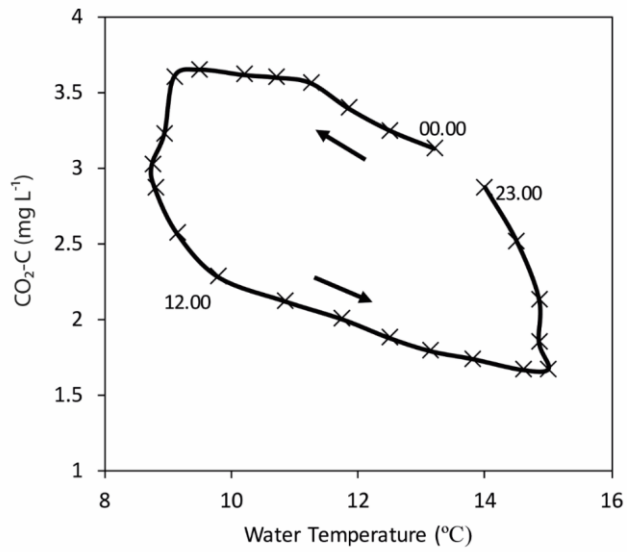
Figure 6. Time series of (A) Stream CO<sub>2</sub> concentration (black) and discharge (red), and (B) water temperature (black) and shortwave incoming radiation (SR, red) covering the period April-July 2018. Note the reverse axis for shortwave incoming radiation.

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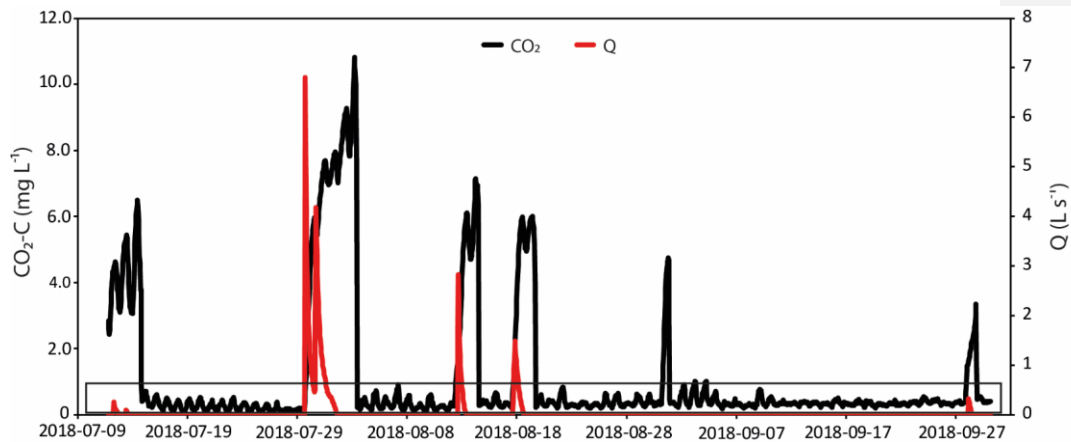
**Figure 7. Diel amplitude in stream CO<sub>2</sub> concentration in relation to A) daily mean stream water temperature, and B) daily mean shortwave radiation (SR), covering the period April-July 2018. Statistics are given according to Spearman's rank correlation.**



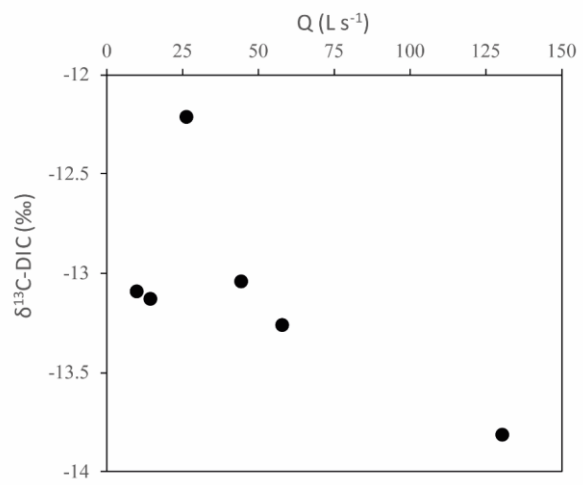
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**Figure 8.** CO<sub>2</sub>-Water temperature hysteresis loop based on the median daily values presented in figure 7 covering the period April-July 2018.





545 **Figure 9.** Stream CO<sub>2</sub> concentration (black) and discharge (red) for the dry period (July-September 2018). Periods when the CO<sub>2</sub> sensor was above the water table capturing an atmospheric signal (i.e. with concentrations <0.5 mg C L<sup>-1</sup>) are highlighted by the lower box.



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**Figure 10.**  $\delta^{13}\text{C-DIC}$  as a function of stream discharge. The six sampling occasions covered the falling limb of the snowmelt peak April-June 2018.