Title: "Vertical partitioning and controlling factors of gradient-based soil carbon dioxide fluxes in two contrasted soil profiles along a loamy hillslope".

Tracking #: bg-2014-405

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Dear Editor,

This manuscript is a revised version of C8695/2015. As suggested in the decision letter, we submit a more complete and more detailed manuscript that takes into account all the comments and suggestions of the reviewers. In addition, we now highlight clear hypotheses and justification in the introduction. As requested, the manuscript also benefited from a thorough editing for language by the senior authors.

Thank you very much for the review of our manuscript. We found the comments made by both you and the referees to be very constructive and believe that the manuscript, revised in light of them, is significantly better. We list below the specific responses to the individual points raised by the referees and detail the changes made in the manuscript. For easy reference, the original comments are presented in black, and our responses in bold italic red. Below this point-to-point response, you will find the manuscript version with tracked changes.

Our results have not been published elsewhere and are not under consideration for publication elsewhere. All authors have seen and agreed to the version submitted.

Sincerely,

François Wiaux, PhD

Biogeosciences Discuss., 11, C8695–C8696, 2015

#### www.biogeosciences-discuss.net/11/C8695/2015/

Interactive comment on "Quantitative estimation and vertical partitioning of the soil carbon dioxide fluxes at the hillslope scale on a loess soil" by F. Wiaux et al.

1. Editor decision

Associate Editor Decision: Publish subject to minor revisions (Editor review) (17 Apr 2015) by Daniel Obrist

Comments to the Author:

Dear Dr. Wiaux,

I now have received detailed second reviews by two of the three previous reviewers. Both reviewers were pleased with the direction of the revisions, and both commented that the manuscript has greatly improved compared to the first submission, in particularly in regards to focus.

At the same time, both reviewers pointed out that there still are significant issues with the quality of writing, including stylistic issues, typos, as well as consistency in the use of units and order of figures. Both reviewers also commented that some sections (in particular the introduction and results section) now are extremely short and at times are lacking details, and that the manuscript lacks clear hypotheses and justification in the introduction. Similarly, the discussion sections needs improvement in order to clearly highlight the novelty and implications/impacts of the study results. Further, the manuscript needs to improve referencing to other published studies to clarify differences/similarities of results observed in this study compared to others.

In spite of the significant amount of remaining issues, I expect that these can be addressed in a relatively short time period as there are no substantial flaws with data or data analysis (so mainly editorial issues as well as improved introduction/discussion sections). I therefore decided to move ahead recommending revisions of this manuscript. I encourage the authors to address all comments of the two reviewers; if the authors can successfully address the remaining issues, I will move ahead quickly with an editorial decision accepting this manuscript.

With best regards,

Daniel Obrist

#### 2. Anonymous Referee #1

#### **Overall comments**

This is the second time I have reviewed this manuscript. I think the shortening and focusing of the manuscript has greatly improved this paper, which is now focused much more on soil CO2 profiles, vertical contributions to fluxes, and controlling variables. I still think this is a strong dataset with very sophisticated processing of results, I am particularly impressed how the authors could differentiate between contributions of various depths layers to CO2 fluxes, that is really valuable and highly interesting.

Having said that, the manuscript lacks a clear hypothesis and reasoning for the need of this study. The manuscript still has a lot of editorial issues (lines of argumentation, implications, typos, etc), and I suggest that the senior authors of this manuscript really help with editorial issues. The introduction and results sections is quite thin and very short, and should be expanded in certain areas. The results section should present quantitative relationships between variables (e.g., correlations, or other methods to clarify how much variability is explained by variables, this can be added in the text, no need to add additional figures). This would allow for better quantitative description of the controlling factors and therefore would give the discussion section more credibility (i.e., more than qualitative description of how physical processes control fluxes/emissions). Finally, I am doubtful how the authors can clearly differentiate between contributions of physical factors versus microbial factors, as they are both strongly interlinked.

I am recommending "major review" to fix these remaining issues, although I think the required changes can be implemented easily as they mainly are related to editorial issues and improving discussion/introduction, and don't reflect a structural deficit of the data or analysis performed.

#### Abstract:

Line 22. Start with the purpose and goal of this study, it needs to be clear why this study was conducted, rather than just stating "We assessed..." A clear hypothesis might help as well.

This has been considered. We have revised the introduction and have identified a clear objective (last paragraph of introduction). In summary, we highlight in our introduction that although literature suggests that soil physical controls are important, little data to illustrate it exists, especially showing the temporal evolution and the vertical profile description of  $CO_2$  fluxes and abiotic variables.

Line 30-31: I don't think that the fact that 90-95% of the soil flux originated from the surface 10 cm alone supports that soil OC at deeper depth is stabilized – there just could be very little carbon at depth, so you need to state there is plenty of OC carbon (even labile pools) present at higher depths (e.g., Figure 1) but apparently these pools aren't mineralized and don't contribute to surface flux.

There is indeed a large stock of soil OC at depth which is characterized by a large pool of labile OC. This information is given in Figure 1. We assume that these important labile OC pools are mineralized and contribute to surface flux, but the OC stocks at the footslope are so huge that important amounts of labile OC can still be observed.

Line 32-35. You should clarify the new results of this study, e.g., that CO2 fluxes at the footslope are controlled by water content and therefore diffusivity. What are the major implications and meanings of this?

There are two main implications, which are now clearly described in the discussion and at the end of the abstract:

- this study highlights the need to consider soil physical properties and their dynamics when assessing and modeling soil CO2 emissions.

- if hydrologic regimes change and that footslope soils become drier (reaching moisture conditions favorable for micro-organisms respiration and gas transfer), there is a large amount of potentially easily decomposable OC stored at depth that can suddenly decompose and be emitted to the atmosphere.

A key conclusion/discussion point of this study might be how surface flux measurements compare to gradient-based diffusion approaches, and what vertical measurement resolution and soil parameters (porosity/diffusivity/soil water content) need to be characterized to obtain reliable fluxes based on soil CO2 characterization?

Thank you for this helpful suggestion, we now briefly discuss this issue on section 2.6.

#### Introduction:

Line 50: can you expand on this study, how much did subsoil fluxes contribute.

# We have largely developed the first paragraph in this way, explaining the importance of understanding and quantifying the contribution of subsoil fluxes.

Line 54-57: Can you pls. clarify why this needs to be done in agro-ecosystems, and why you expect patterns to be different than in forests? Please expand on the results from forests, what the implications of this are, and why this needs to be repeated/studies in ago-ecosystems. E.g. how important are agro-ecosystems for CO2 fluxes worldwide? Etc. While the revised paper now is shorter and more focused, the introduction should be expanded to clarify why this study is needed, what the anticipated differences are to forest sites, and what the anticipated differences may be .

# We have largely developed the second to last paragraph in this way, explaining the importance of understanding and quantifying the contribution of crop soils compared to forest soils.

Line 63: some clear hypotheses of your expected patterns would help here.

# We added this sentence: "Based on a previous study (i.e. Wiaux et al., 2014b), we expect differences in respiration along the topographical gradient, i.e. 30% more at the downslope and 50% more at the backslope, relative to the uneroded summit position."

#### **Materials and Methods:**

Entire section: we measured....we inserted....we adjusted... I normally don't mind active language, but starting each sentence with "we" is not good style. This section (and the full manuscript) should be edited for language and flow. It seems that the writing was left to a student without appropriate internal review by all involved authors – please fix and edit.

# We apologize for this. The entire manuscript has beenrevised by the senior authors and edited for language.

Lines 79 to 86: could you clarify why these parameters were measured. It is clear, but a short statement introducing the need for these measurements would help the flow of this section.

# We have now created the section 2.2. "Soil physical and bio-chemical properties" were the goals and the protocols for measuring all soil parameters are clearly described.

Lines 120: We measured VWC at a depth of 10, 25, 35, 50, 70 and 95 cm depth – should be changed to "we measured VWC at depths of...."

#### This has been corrected.

Lines 126-128: Is it really the only goal to calibrate the soil gas diffusion model. The authors might want to expand on this stating that a goal is to compare gas diffusion-based fluxes with surface fluxes. I think the authors have a unique ability to clarify how detailed diffusion measurements and soil properties are needed in order to appropriately predict/model surface fluxes.

We agree with the reviewer that this is an interesting result. However, in order to focus our study (as requested in the previous rounds of review), we prefer not to provide an in-depth analysis of the different methods. It should also be noted that we adjusted the parameters of the gas diffusion model in order to fit the calculated  $CO_2$  fluxes to the surface flux measurements. Hence, direct diffusion measurements were not available. We measured soil water retention curve and porosity to have some indicators of the diffusion coefficients, but we later optimized these parameters in a realistic range of values. As a consequence, we prefer not focus on the importance to measure soil properties to calculate  $CO_2$  fluxes. We hope that this response satisfies the reviewer.

Lines 147-150: I don't understand these statements.

#### This has been rephrased and clarified.

Lines 190-199: ok, I guess I now understand, that the discrete surface flux measurements are used to calibrate the diffusion model, and that the measured soil CO2 gradients are used for larger temporal coverage.

Yes, this is correct, see above. In response to this comment, we have slightly reworded this section.

Still, I think this study has a unique opportunity to compare these two methods and add a good discussion about the challenges/needs when using soil CO2 gradients to predict surface fluxes (i.e., characterizing diffusivities and their temporal/spatial variability).

We agree with you but we previously made the choice to not add this comparison issue in this paper to make it shorter and more focused. However, we agree that this is a unique opportunity to explicitly show a comparison between these two methods. Without expanding too much about this in the discussion, we add a new figure (Fig. 5) which illustratesthe comparison between gradient-based calculation and observation of surface  $CO_2$  fluxes, which allowed the optimization of the calculated fluxes. However, we present it as a part of the M&M section and do not discuss it as a result.

#### <u>Results</u>

Entire section 3.1. What I am missing is how these figures link. All (most) of these variables are directly linked, e.g., soil water content affects diffusivity which in turns affects CO2 concentration profiles and fluxes. The authors need to link these figures, e.g., they could explain how much of the variability of certain factors are directly driven by others (e.g., how much of the variability in CO2 concentrations or diffusivity are directly linked to soil water content)? Maybe I am missing something here, but just showing/presenting all these variables separately without showing the connections does not make a lot of sense to me.

In Wiaux et al. (2014b), we already performed a detailed statistical analysis highlighting the role of each factor in soil respiration and how much of the variability of certain factors are directly driven by others. Hence, we prefer not to repeat this analysis with just another data set of measurements. We argue that the novelty of the present paper is related to the analysis of the temporal dynamics of each factor and its distribution along the soil profile. However, we refer to the linkages between the variables using the Wiaux et al. (2014b) study.

Lines 215: What statistical tests were done to evaluate differences? Please clarify.

While we focus here on the variability of soil temperature along time and along soil profiles, we do not have any replicates in space for each soil depth. Hence, a strong statistical analysis to compare temperature dynamics between slope positions is not possible. In addition, comparing time-series is difficult with statistics. Our comparison remains qualitative. We have identified this more clearly in the revised manuscript.

Line 218: in air? Surface soil? Please clarify.

#### This has been considered.

Lines 228 to 230: soil gas diffusivities are directly linked to water content, so the word "in contrast" doesn't seem correct. The authors should refer to the correlations (and dependence) of diffusivity to soil water content in describing the patterns (i.e., link Figures 6a and 6c).

#### This has been considered.

Line 243-244: Clearly, these differences are highly significant, but a test should be done and significance levels reported.

#### See previous comments above.

Line 246: what model? Clarify what is done here.

# The model built in this study to represent soil $CO_2$ concentration profiles (Eq. 2). This have been précised in the manuscript.

Lines 253-255. The authors should expand on the spatial/temporal patterns of CO2 fluxes, what are temporal and spatial patterns, ...

# Actually, we already expanded on the profile distribution in the section 3.3 dedicated to the vertical partitioning of $CO_2$ fluxes. Also, the temporal dynamics of $CO_2$ fluxes are discussed in the previous subsection.

...correlations to soil parameters (temperature, SWC, diffusivity, CO2 profiles), etc.

#### See previous comments above.

#### **Discussion:**

Lines 272 to 276: can you give the predictive power (e.g., r2, or percent variability explained) that is explained by temperature at both locations, rather than just describe this qualitatilvely. The correlations/%variability explained could be added to the results section as suggested above, and then could be discussed here.

The reviewer has already suggested this in his comments for the results section. We provide a detailed answer to these comments in that section.

Lines 278 - 380: you should quantify this, rather than suggest. You could add the correlation coefficients or %variability explained by each in the results section when reporting patterns in figure 6.

#### Idem

Line 283 – 288: This section needs referencing, there is a large amount of literature on this, the authors need to discuss their results with those of the literature.

# We added additional references in the manuscript (i.e. Ball, 2013; Bauer et al., 2012; Castellano et al., 2011; Davidson et al., 1998; Perrin et al., 2004; Webster et al., 2008b) and briefly discussed our results in the context of these studies.

# Note that the impact of soil gas diffusion on CO2 emission is also further discussed in the next paragraph.

Lines 293-297. The authors should put this discussion in respect to the fact that 90-95% of fluxes occurred from the top 10 cm at this location. So does the diffusion barrier prevent contribution from deeper soil levels, and would contributions be likely higher without the diffusion barrier? This discussion should be expanded.

#### This has been considered and further discussed in this section.

Line 295 to 297: This discussion is a bit thin (are key) – please discuss in detail how continuity and diffusion barriers regulate soil gas emissions, this seems a key point of this study but it is not reference or discussed well.

#### This has been considered and further discussed in this section.

Lines 298-310: this section needs improvement, and I doubt the authors clearly separate between physical processes and microbial processes since both are highly are interconnected. For example, earlier in the discussion, the authors state that a the footslope, high VWC limits the transfer of CO2 along the sol profile and/or reduces production of CO2 due to a lack of oxygen - so microbial respiration is controlling CO2 production also here, although the underlying reason is not substrate or temperature limitation but likely oxygen limitations – this seems to directly contradict the statements here (lines 307-310). This discussion should be clarified.

We do not agree that these are contradictory explanations. Oxygen limitations do not imply zero oxygen levels and no  $CO_2$  production, but a reduced potential for production. We only suggest that this reduced potential is to some extent compensated for by the very large OC stock that is found in these footslope soil profiles Hence, even if gas diffusion is limited (and that consequently oxygen supply for micro-organisms is limited),  $CO_2$  is produced. This  $CO_2$  accumulates under the diffusion barrier. This accumulated  $CO_2$  is later emitted when VWC decreases under a threshold value which allow a significant gas diffusion.

#### This has been clarified in the manuscript.

Line 312: can you clarify how? I assume you need both respiration AND carbon amount to infer about persistence?

#### Yes indeed. This has been developed in our previous publications (i.e. Wiaux et al., 2014a,b).

Line 320-322: I would disagree with this statement, why should forest soil and agro-ecosystems not be comparable. The differences should be clearly discussed, what could cause/contribute to the differences? I assume if other sites don't have such high water saturation, or not such strong diffusion barriers, then CO2 fluxes probably are related to substrate concentrations? Please clarify.

Comparing forest and crop soils is difficult because of the important part of the autotrophic respiration coming from roots in forest while this can be easily avoided in croplands. We have elaborated on this topic in the manuscript.

However, we tried to achieve a qualitative comparison of our results with the stufy of Goffin et al. (2014) in forest ecosystems. This has been detailed in the manuscript.

Line 323-324. The authors should quantify relative controls of different variables in the methods section, then they could be quantitative rather than qualitative.

See above. In Wiaux et al. (2014b), we already performed a detailed statistical analysis highlighting and quantifying the role of each factor in soil respiration and how much of the variability of certain factors are directly driven by others.

Line 355: add: and likely the transfer of O2 to deeper soil depths.

This has been considered.

#### 2. Anonymous Referee #3

#### **Overall comments:**

This review represents the second time I have seen this paper and in general I am pleased with the way the authors addressed the criticisms of the previous version. Eliminating the modeling section greatly helped focus the manuscript and for me it was easier to understand the manuscript. A few issues remained that I think need to be addressed before it can be published but these are mainly editorial. I think the methods section could be rearranged I a bit since it jumps back and forth between different methods. I also may have identified a few new issues that I did not comment on in the previous version, partly because in my previous review I focused on some 'bigger-picture' issues. My apologies for that. Below I have outlined my comments.

#### Introduction

I would suggest including Schmidt et al 2011 in either the introduction or discussion. This paper argues that low decomposition rates of organic matter in soils may be because of physical conditions (high moisture/low O2) or other means of protection rather than chemical composition. This would fit well with the objectives and results of this manuscript.

#### This has been added in the introduction.

Line 41: spell out OC the first time

#### This has been considered.

Line 41-42: whether or not climate change represents a positive feedback would depend on the current conditions and the type of change that occurs. Warming may have a positive effect in mesic, temperate environments but not in arid conditions. You may want to expand on this a little bit more and be explicit about the types of changes (temperature vs. precipitation changes) you expect to happen and how they may affect decomposition.

# This has been detailed: "Under our temperate climate, temperature increase as well as summer drought would constitute potential climatic changes (IPCC, 1990; 1992) which are supposed to increase OC turnover (e.g. Davidson and Janssens, 2006)."

Line 50-51: If the OC is highly processed you would not expect large contributions of this OC to the total decomposition flux. However, Schmidt et al argue that physical conditions may prevent decomposition of 'deep' OC even if this OC would be easily decomposable under optimal conditions.

# We have added the contrasted assumption of Schmidt et al in the mlansucript and compare these different points of view. This points to the need of in-situ measurements of both OC stocks and CO2 fluxes along deep soil profiles are needed to elucidate the issue of soil-atmosphere C exchange in the case of buried OC.

Line 55-57: Would you expect ag soils to be different from forest soils in terms of the contribution of deep vs. shallow OC to the total CO2 efflux? I am missing the development of clear hypotheses.

We agree on the need to clarify the differences between forest and agro-ecosystems. It does not differ in terms of deep OC contribution, but more on  $CO_2$  fluxes measurements. We have now explicitly developed this point at the end of the introduction.

#### **Materials and Methods**

Line 75-76: The soil classifications are based on the FAO system so I would reference that.

We have now refered the IUSS WRB classification system of soils (FAO) as requested.

Line 79-83: Was porosity mentioned in previous papers? If so, than I would also add a few lines about how OC and labile OC were measured. In other words, why describe how porosity was measured but do not describe how OC and labile OC were measured if all of these were reported in previous papers?

We have now created the section 2.2. "Soil physical and bio-chemical properties" were the goals and the protocols for measuring all soil parameters are clearly described in details.

Line 84-86: How were the SWR's determined? Either describe this in more detail or refer to another paper and see previous comment.

This has been considered. See previous comment.

Line 88: replace purpose-built with custom-built.

#### This has been considered.

Line 90-91: I am not sure I understand this sentence.

We clarified this: "The analytical precision is function of both the probe characteristic and the value of the observation. This can be calculated as the sum of 1.5% of the measurement range and 2% of the observed value."

Line 113: I would move the section from line 192 describing the LI-COR surface flux measurements to line 113. In line 123 you mention the surface flux measurements but you have not described these yet.

This has been considered. This section has been completely restructured and split intotwo parts in order to make the reading easier and the explanation more coherent.

Line 129-135: I would move this to the section after line 103 and perhaps eliminate the specific ranges at the various depths.

This seems not relevant to us to move these few lines, but we rephrased it to shorten and clarify the information about the TDR technique (which has nothing to have with the SWR curve assessment, contrary to what was suggested by the reviewer).

Line 141-146: I would also move this to the section describing the CO2 measurements.

This has been considered. This section has been completely restructured and split intotwo parts in order to make the reading easier and the explanation more coherent.

Line 147-150: Perhaps move this to after line 123.

This has been considered. This section has been completely restructured and split intotwo parts in order to make the reading easier and the explanation more coherent.

Line 162: add that 'z' is the depth

This has been considered.

Line 191: Not sure that 'punctual' is the right word here. Perhaps 'instantaneous' is more appropriate.

This has been considered.

Line 199: How were the calculated fluxes corrected? Was this done by adjusting specific parameters such as diffusivity or were the calculated fluxes simply decreased/increased by 5% or 22% to make the calculated fluxes match the measured fluxes? This is unclear.

# As already explained in the manuscript, the calibration by adjusting specific parameters such as diffusivity ensures the good precision of calculated CO2 fluxes, while the correction based on the slope of the fit ensures the accuracy of the fluxes. These two steps are necessary and complementary.

Line 201-208: This section is still a little bit unclear to me. Goffin et al. and Maier and Schack-Kirchner calculated CO2 production from each soil slice. However, the calculations used by the previous papers seems to be different from the one presented in the current manuscript. Given that this is a critical part of the paper I would recommend expanding this section and mention the differences/similarities used between the approach taken in this manuscript vs. the approaches taken in the other papers mentioned in this section.

We have expanded this section and mention the differences/similarities between this study and others: "In contrast to other studies (e.g. Pingintha et al., 2010; Turcu et al., 2005), we did not aggregate the soil diffusivity coefficient for the entire soil profile or for an entire soil layer. Contrary to Goffin et al. (2014) and Maier and Schack-Kirchner (2014), we did not calculate the CO2 production from each soil slice based on the difference of CO2 concentrations between the top and the bottom of soil horizons, but we rather assessed a continuous profile of CO2 fluxes and production.[...]"

In addition, Eq 2 does not give you the flux but rather the concentration profile so some additional steps are needed to go from Eq 2 to the actual fluxes at the various depth intervals.

Yes, indeed, but we clearly explain that Eq.2 is introduce in Eq.1 to get the  $CO_2$  flux. This Eq.1 requests the calculation of soil gas diffusivity, in addition to  $CO_2$  concentration values.

Line 207: What do the authors mean by a 'semi-seasonal' timescale? Can you be more specific?

#### We have reworded this.

#### <u>Results</u>

Line 224: I don't see in the graphs that VWC in the subsoil of the summit profiles reached 0.5 cm3/cm3 (which incidentally is higher than the total porosity). Is this a typo?

Yes this is a typo, we apologize for that. The right value is 0.39 cm3/cm3. This has been corrected.

Line 234-235: Can you expand on this a little bit more? What time periods are you talking about?

#### This has been detailed in the manuscript.

Line 228: The authors start with discussing fig 6C rather than 6A. I would change the order of the panels to make it align with the text.

## Figures have been re-ordered and renamed. Fig. 6 to Fig. 10 respectively show the spatio-temporal variation of soil temperature, moisture, CO2 fluxes, concentrations and diffusion.

Please note that different units are being used for the CO2 fluxes between the different panels. Please make sure to use the proper units consistently.

Direct observations and bulk calculated  $CO_2$  fluxes are in umol  $m^{-2} s^{-1}$ . This unit suits to comparisons between directly measured  $CO_2$  fluxes with calculated  $CO_2$  fluxes. We then kept these units in the new Fig. 5.

We used gC  $m^{-2} day^{-1}$  to smooth the temporal variations of CO<sub>2</sub> fluxes along months at the time scale of a year and at the spatial scale of a measurement station on a slope position (footprint of 5  $m^2$ ). This allows a more clear representation of the spatio-temporal dynamics at these scales. However, to illustrate the differences of CO<sub>2</sub> fluxes profiles between different dates, a daily aggregation would prevent detecting any significant differences, and we argue that it is more appropriate to express averaged daily fluxes in umol  $m^{-2} s^{-1}$ . Hence, we suggest to not homogeneize the units in the different panels of the Figure showing CO<sub>2</sub> fluxes, as the goal is not to compare data between panels but inside each panel.

This also applies to the CO2 concentrations throughout the text. Sometimes the authors use ppm, mumol/m3, or %.

We do not use umol/m3 but umol/(m2 sec), which are units of CO2 fluxes. For CO2 concentrations, we considered your remark and we now use % everywhere instead of ppm.

Line 246: Replace 'between' by 'from'.

#### Ok.

Line 257: Again, the order in which the results are discussed is different from the order in the figure. Start with discussing the summit rather than the footslope profile.

#### This has been considered.

Line 267: Is this negative uptake significant given the high standard deviations? You probably want to come back to this since this could be an artifact of the calculation method perhaps in combination with measurement uncertainty.

You're rigth, this negative uptake is not significant given the high standard deviations. We therefore not mention it anymore.

#### **Discussion**

Line 278-280: I think I understand what the authors are trying to say here but they may want to expand on this a little bit more. You are basically talking about the relative amounts of water- vs. air-filled pore space, correct?

Yes indeed, we are talking about water-filled pore spaces. This has been detailed in the manuscript.

Line 299-301: Yes but for CO2 to accumulate it has to be produced as well which is likely related to the labile OC present. The fact that it stays in the profile is due to the low diffusivity.

#### We agree that this sentence was confusing. We have rephrased it according to your comment.

Line 301-302: What is the difference between 'CO2 efflux' and 'instantaneous soil respiration'? I am not sure I understand what you are trying to say here.

The  $CO_2$  efflux is the observed  $CO_2$  flux resulting from all transfer and production mechanisms together. The soil respiration is the  $CO_2$  flux due to micro-organisms metabolic activity. Hence, if  $CO_2$  is stored into soil pore spaces and then later emitted after accumulation phases, the  $CO_2$  efflux can deviate from the instantaneous soil respiration.

#### This has been detailed in the manuscript.

Line 321-322: I would expand on this a little more. While I agree that ag soils and forest soils are very different can the authors compare/contrast the studies in terms of OC quality, diffusivity, etc.?

#### See answer to reviewer #1.

# Comparing forest and crop soils is difficult because of the important part of the autotrophic respiration coming from roots in forest while this can be easily avoided in croplands. We precised it in the manuscript.

## However, we tried to achieve a qualitative comparison of our results with the study of Goffin et al. (2014) in forest ecosystems. This has been detailed in the manuscript.

In general, I think the authors can emphasize the differences in amounts of total and labile OC between the two profiles and while the footslope profile contains more labile OC in the subsoil, there is a less of a contribution from the subsoil to the overall respiration fluxes due to physical limitations (low diffusivity and lack of O2). This information is somewhat implied in the discussion but I think the authors can bemore explicit about this.

We agree this should be more explicit. We therefore added the following sentence: "In other words, while the footslope profile contains more labile OC in the subsoil relative to the summit (Fig. 1, Wiaux et al., 2014a), there is a lower contribution from the subsoil to the overall respiration fluxes due to physical limitations (both low diffusivity and lack of  $O_2$ )."

In addition, the implications of all of this is that if hydrologic regimes change and footslope soils become drier, there is a large amount of potentially easily decomposable OC stored at depth that can suddenly decompose if moisture conditions become more favorable.

Thank you for having summarized this. We have added the following sentence at the end of the section 4.1. The main implication of these observations is that if hydrologic regimes change and that footslope soils become drier (reaching moisture conditions favorable for micro-organisms respiration), there is a large amount of potentially easily decomposable OC stored at depth that can suddenly decompose and be emitted to the atmosphere.

Line 344: This corroborates the notion by Schmidt et al suggesting that deep organic matter may be protected because of unfavorable physical conditions rather than substrate limitations.

This is right. We now cite Schmidt et al. (2011) in this paragraph to corroborate our observations.

Table 1: What does 'NI' mean?

### NI means No Information (i.e. due to a lack of replicates to allow reliable mean and S.D.). This has been identified in the figure caption.

Figure 5 and 6: I would consider splitting these up in several separate figures so rather than have 5(A) (a)(b) and 5(B)(a)(b) make two separate figures for temperature (5(a) and 5(b)) and moisture (6(a) and 6(b)). Similarly, I would split up figure 6 into three figures and make sure that the figure order matches the order in which the figures are discussed in the text (see below).

This has been considered. Figures have been re-ordered and renamed. Fig. 6 to Fig. 10 respectively show the spatio-temporal variation of soil temperature, moisture, CO2 fluxes, concentrations and diffusion.

Figure 6: Make sure the order of the panels is the same as discussed in the text.

This has been checked.

| 1                | Vertical partitioning and controlling factors of  |  |
|------------------|---|--|
| 2                | gradient-based soil carbon dioxide fluxes in two  |  |
| 3                | contrasted soil profiles along a loamy hillslope.   |  |
| 4<br>5           | Authors :Wiaux, F. <sup>†*</sup> , Vanclooster, M. <sup>†</sup> , Van Oost, K. <sup>‡**</sup>   |  |
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| 17<br>18         | <b>Key words:</b> C dynamic model; $CO_2$ flux; physical control; vertical partitioning; OC storage; Hillslope; cropland; loess soil.   |  |
| 19               | Type of paper: Regular research paper   |  |
| 20               |   |  |

#### 21 Abstract

| 22   | In this study, we aim to elucidate the role of physical conditions and gas transfer mechanism along soil  |   |
|--|---|---|
| 23   | profiles in the decomposition and storage of soil organic carbon (OC) in subsoil layers. We use a   |   |
| 24   | qualitative approach showing the temporal evolution and the vertical profile description of CO <sub>2</sub> fluxes  | Mis en forme : Indice   |
| 25   | and abiotic variables. We assessed soil CO <sub>2</sub> fluxes throughout two contrasted soil profiles (i.e. summit   |   |
| 26   | and footslope positions) along a hillslope in the central loess belt of Belgium. We measured time-  |   |
| 27   | series of soil temperature, soil moisture and CO <sub>2</sub> concentration at different depths in the soil profiles  |   |
| 28   | for two periods of 6 months. We then calculated the $CO_2$ flux at different depths using Fick's diffusion  |   |
| 29   | law and horizon specific diffusivity coefficients. The calculated fluxes allowed assessing the  |   |
| 30   | contribution of different soil layers to surface $CO_2$ fluxes. We constrained the soil gas diffusivity   |   |
| 31   | coefficients using direct observations of soil surface CO <sub>2</sub> fluxes from chamber-based measurements   |   |
| 32   | and obtained a good prediction power of soil surface $CO_2$ fluxes with a R2 of 92%.  |   |
|  |   |   |
| 33   | We observed that the temporal evolution of soil $CO_2$ emissions at the summit position is mainly   | Mis en forme : Justifié, Espace Après :<br>10 pt, Interligne : Double |
| 34   | controlled by temperature. In contrast, at the footslope, we found that long periods of $CO_2$  |   |
| 35   | accumulation in the subsoil alternates with short peaks of important CO <sub>2</sub> release. This was related to   |   |
| 36   | the high water filled pore space that limits the transfer of $CO_2$ along the soil profile at this slope  |   |
| 37   | position. Furthermore, the results show that approximately 90 to 95 % of the surface CO <sub>2</sub> fluxes   |   |
| 38   |   |   |
|  | originate from the first 10 centimeters of the soil profile at the footslope. This indicates that soil OC in  |   |
| 39   | originate from the first 10 centimeters of the soil profile at the footslope. This indicates that soil OC in this depositional context can be stabilized at depth, i.e. below 10 cm. This study highlights the need to  |   |
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| 39<br>40<br>41   | originate from the first 10 centimeters of the soil profile at the footslope. This indicates that soil OC in<br>this depositional context can be stabilized at depth, i.e. below 10 cm. This study highlights the need to<br>consider soil physical properties and their dynamics when assessing and modeling soil CO <sub>2</sub> emissions.<br>Finally, changes in the physical environment of depositional soils (e.g. longer dry periods) may affect  |   |
| 39<br>40<br>41<br>42   | originate from the first 10 centimeters of the soil profile at the footslope. This indicates that soil OC in<br>this depositional context can be stabilized at depth, i.e. below 10 cm. This study highlights the need to<br>consider soil physical properties and their dynamics when assessing and modeling soil CO <sub>2</sub> emissions.<br>Finally, changes in the physical environment of depositional soils (e.g. longer dry periods) may affect<br>the long-term stability of the large stock of easily decomposable OC that is currently stored in these                  |   |
| <ol> <li>39</li> <li>40</li> <li>41</li> <li>42</li> <li>43</li> </ol> | originate from the first 10 centimeters of the soil profile at the footslope. This indicates that soil OC in<br>this depositional context can be stabilized at depth, i.e. below 10 cm. This study highlights the need to<br>consider soil physical properties and their dynamics when assessing and modeling soil CO <sub>2</sub> emissions.<br>Finally, changes in the physical environment of depositional soils (e.g. longer dry periods) may affect<br>the long-term stability of the large stock of easily decomposable OC that is currently stored in these<br>environments. | Mis en forme : Police :11 pt, Non Gras                                |

#### 44 1. Introduction

Soils play a major role in the global C budget, as they contain 2 to 3 times more C than the 45 46 atmosphere(Eswaran et al., 1993; Lal et al., 2003). However, Courrent predictionsassessments of the 47 exchange of C between the soil and the atmosphere in response to environmental change are 48 associated with large uncertainties (Houghton et al., 2003;e.g. Peters et al., 2010). One of the sources 49 of this uncertainty is related to our poor understanding of C dynamics in the deeper layers of the soil 50 profile. Rumpel and Kögel-Knabner (2011) showed that deep soil OC is highly processed, but that 51 subsoil C fluxes from C input, stabilization and destabilization processes are still poorly constrained. 52 In addition to this, recent work has highlighted the significance of the lack of understanding of the role 53 of-buried OC in depositional setting for the C cycle on soil C emissions (e.g. Berhe et al 2007; Van 54 Oost et al., 2012; Wang et al., 2014; Wiaux et al., 2014). More specifically, deeply-buried OC that is 55 stored indownslope colluviumsal soils at the bottom of eroding hillslopes(e.g. Stallard et al., 1998)cannot be assumed to be inert to further loss as it can suddenly decompose as a result of 56 57 continued degradation or disturbances such as of global warming, desiccation of saturated soils, land 58 use change, and re-excavation by gullying, etc(e.g. Van Oost et al., 2012). Somes studies suggested an 59 "erosion-induced C source" alon hillslope ranging from 0.37 petagram C per year (Jacinthe and Lal 60 2001) to 0.8-1.2 petagram C per year (Lal 2003). Somes studies suggested an "erosion induced C source" alon hillslope ranging from 0.37 petagram C per year (Jacinthe and Lal 2001) to 0.8 1.2 61 petagram C per year (Lal 2003).Based on many recent studies, Rumpel and Kögel-Knabner (2011) 62 highlighted that deep soil OC is highly processed, and showed the need for quantitative information 63 64 about C fluxes coming from deep soil horizons. Hence, understanding controls on soil surface CO2 flux are usefulto This indicates shows that more quantitative information on the contribution of deep C 65 66 to soil-atmosphere C exchange as well an increased understanding of the controlling factors is needed 67 improve the prediction of soil atmosphere CO2 emissions, and thisrequires accounting for deep soil 68 layers. 69 There is now significant concern about the contribution of soil organic carbon (OC) to future climate

70 change where a climate change driven acceleration of soil OC decomposition could represent a

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positive feedback on climate (ref e.g. Davidson and Janssens, 2006; Frey et al., 2013). Under our
temperate climate, temperature increase as well as summer drought would constitute potential climatic
changes (IPCC, 1990; 1992) which are supposed to increase OC turnover (e.g. Davidson and Janssens,
2006).

75 In addition to the role of soil mineralogy and microbial communities, rRecent studies highlight the 76 importance of soil bio-physical conditions that may vary substantially with time and across landscapes 77 (e.g. Dai et al., 2012). In addition to the combined effects of These studies have shown that, in addition 78 to the effects of soil moisture, temperature and OC quality, soil physical properties (e.g. gas diffusion 79 barriers) myay also exert an importanton-control on soil microbial activity and soil  $CO_2$  fluxes (e.g. Wiaux et al., 2014b), recent studies show the importance of physical controls on CO2 fluxes. 80 81 suggesting the role of such as gas diffusion barriers along soil profiles .(e.g.; Ball, 2013; Maier et al., 2011). Furthermore, or arguing that low decomposition rates of OC in soils may be because of there is 82 83 empirical evidence suggesting that physical protection (i.e. soil aggregates) rather than chemical 84 composition is a key factor controlling the long-term stability of OC in soils (e.g. Schmidt et al., 2011). 85 Schmidt et al.(2011) also argued that physical conditions may prevent decomposition of deep OC even 86 if this OC would be easily decomposable under optimal conditions. Furthermore However, othermost 87 process studies so far have focused on the soil surface layer while there is now increasing 88 awarenessindicate that subsoil OC represents an important C store that interacts actively with the 89 atmosphere (e.g. Rumpel and Kögel-Knabner, 2011). Recent studies (Rumpel and Kögel-Knabner, 90 2011) highlighted that deep soil OC is highly processed, and showed the need to consider C fluxes originating from deeper soil horizons. To elucidate the issue of soil atmosphere C exchangein the case 91 92 of deeply buried OC, in situ measurements of both OC stocks and CO<sub>2</sub>-fluxes along deep soil profiles 93 are needed. This Understanding the soil physical controls on soil CO<sub>2</sub> fluxes is thus particularly relevant 94 in landscapes with complex topography where buried OC indepositional areas represent a significant 95 part of the total OC storedeontributes substantially to soil C emissions(e.g. Van Oost et al., 2012;

96 Wang et al., 2014 and Wiaux et al., 2014a).

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| 97  | In a forest ecosystem, Goffin et al. (2014) showed that the upper first 30centimeters of a forestasoil                                     |   |
|-----|--|---|
| 98  | profile contribute substantially to the total surface CO <sub>2</sub> flux. However, to our knowledge, a vertical                          |   |
| 99  | partitioning has not been evaluated in agro-ecosystems or in systems with contrasting soil physical  |   |
| 100 | and/or chemical properties. Agro-ecosystems differ from forest ecosystems ason two main points.  |   |
| 101 | First, litter and Ap horizons in forest ecosystems display a specifically are characterized by both a high                                 |   |
| 102 | amount and quality of OC (e.g. Brahy et al., 2002; Goffin et al., 2014), while these horizons have   |   |
| 103 | disappeared in crop soils due to erosion, plowing, and export of plant residues (e.g.Wiaux et al.,   |   |
| 104 | 2014a). Hence, deep OC in forest soils may have a lower contribution relatively to surface CO <sub>2</sub> fluxes Mis en forme : Indice    |   |
| 105 | given that surface soil horizons enriched in fresh organic matter are supposemore likely to emit more                                      |   |
| 106 | CO <sub>2</sub> thatthanin-cropsoils in croplands. Second, rootsnetwork in forests is dense and difficult to remove Mis en forme : Indice  |   |
| 107 | when installing in situ measurement settings. However, rootsare known to largely contributeto  |   |
| 108 | respiration, creating interferences when measuring heterotrophic CO <sub>2</sub> fluxes as an indicator of OC                              |   |
| 109 | turnover (e.g. Davidson et al., 1998; Epron et al., 2006; Fiener et al., 2012). We argue that this latest                                  |   |
| 110 | issuecan be easily avoid in crop soils once vegetation has been removed. Hence, it seems highly  |   |
| 111 | important and appropriate to focus on the behavior of deep soil OC stocks specifically in agro-  |   |
| 112 | ecosystems. In addition, roots network in forests is dense and difficult to remove when installing in                                      |   |
| 113 | situ measurement settings compared to cropsoils. This creates interferences when measuring   |   |
| 114 | heterotrophic CO <sub>2</sub> fluxes as an indicator of OC turnover (e.g. Davidson et al., 1998; Epron et al., 2006;                       |   |
| 115 | Fiener et al., 2012).  |   |
| 116 | In this study, we aim to elucidate the role of physical controls on soil-atmosphere CO <sub>2</sub> fluxes and its                         |   |
| 117 | variation with soil depth for a cultivated soil. To that-this aim, we present a comparative analysis                                       |   |
| 118 | between two contrasting soil profiles along an eroded and cultivated hillslope. Based on a pPrevious                                       |   |
| 119 | studywork (i.e. Wiaux et al., 2014b), we expect differences in respiration along the topographical   |   |
| 120 | gradient, i.e. has shown that soil surface CO <sub>2</sub> respiration is highly variable along this hillslope, with Mis en forme : Indice |   |
| 121 | 30% more respiration at the downslope and 50% more at the backslope, relative to the uneroded  | _ |
| 122 | summit position. However, Why the some controlling factors have been identified, the role of soil  |   |
| 123 | physical controls and of the significance of subsoil OC contributions remain unkown.   |   |
|     |  |   |

The <u>specific</u> objectives of this study are <u>then</u>: (i) to quantify the relative contribution of soil surface and subsoil\_OC to CO<sub>2</sub> fluxes through a vertical partitioning of these fluxes; and (ii) to identify the role of soil physical properties using time-series of soil moisture measurements and gas diffusivity at different depths. The selected study site<u>is representative of a widespread agro-ecosystem of the Belgian</u> <u>loamy belt and</u> is characterized by two contrasting soils in terms of soil hydrological regimes and <u>soil</u> structure and is representative for the cultivated soils of the Belgian loam belt.

#### 130 2. Material and methods

#### 131 **2.1. Study site description**

132 The study was carried out in the Belgian loam belt along a cultivated hillslope of 150 meters length 133 (50.6669°N, 4.6331° W). The site has a maritime temperate climate, with an average annual 134 temperature of 9.7°C and an average annual precipitation of 805 mm. The slope percentage in the 135 backslope area ranges between 8.5 and 16%, with a mean slope of 12%. The slope percentage in the 136 convex shoulder area ranges between 4 to 8.5%, with an average of 6%. The field was plowed (0-30 137 cm soil surface layer) every year. Each year, manure and nitrate fertilization was carried out. The 138 previous crop rotation was winter wheat, maize and spring wheat. The study site has beendescribed in 139 detail in Wiaux et al. (2014a,b). For this study, we selected two measurement stations along the 140 hillslope: one at the summit and one at the footslope position. The soil is a DystricLuvisol type at the 141 summit and a Colluvic\_Regosol\_in the depositional area at the footslope (IUSS Working Group WRB, 142 2007; Wiaux et al., 2014a,b).

143 2.2. Soil physical and bio-chemical properties
144 In order to characterize the physical and bio-chemical properties of these The soil properties of these
145 two soil profiles, we measured have been characterized by Wiaux et al. (2014a,b): soil total OC, labile
146 OC and, soil porosity and soil water retention (SWR) curvesprofiles.

147 <u>Total andOC, laboile OC and soil porosity were already characterized by Wiaux et al. (2014 a,b) and</u>

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are illustrated in Fig. 1. Total C (i.e. the sum of organic and inorganic C) was analyzed using the dry

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| 149   | eombustion techniquean elemental analyzer(Variomax elemental analyzer, Elementar GmbH).  |                                     |
|---|--|-------------------------------------|
| 150   | Instrument precision for total C analyses is 0.05% C concentration. The samples were then treated  |                                     |
| 151   | with 1% HCl in order to remove inorganic CaCO3 and were analyzed again with the elemental  |                                     |
| 152   | analyzer. Soil OC concentration was then deduced from the difference between total carbon analyses   |                                     |
| 153   | before and after 1% HCl treatments.Stable OC was defined as the pool of NaOCl-resistant OC (Siregar  |                                     |
| 154   | et al., 2005). We quantified the stable OC by mixing 3 g of air dried soil with 30 ml of 6 wt % NaOCl  |                                     |
| 155   | (adjusted to pH 8). The NaOCI-treated soil was then washed (shaken and centrifuged) with de-ionized  |                                     |
| 156   | water until the solution was chloride free (i.e. no reaction with AgNO3 occurred). The samples were  |                                     |
| 157   | then dried at 105°C and homogenized before collecting a subsample for total C measurement by dry   |                                     |
| 158   | combustion. The labile OC pool was defined as the residual OC pool that was not resistant to NaOCI   |                                     |
| 159   | oxidation. Hence, this labile OC pool should be interpreted as easily mineralizable OC under ideal   |                                     |
| 160   | conditions where no other factors play a role in stabilization (e.g. anoxic environment, aggregation,  |                                     |
| 161   | <u>etc).</u>   | Mis en forme : Police :Non Gras     |
|   |  |                                     |
| 162   | The total percent $\langle \theta \rangle$ was already characterized by Wigny et al. (2014a b) and is illustrated in Fig. 2.   |                                     |
| 162   | The total porosity ( <sup>(a)</sup> ) was already characterized by Wiaux et al. (2014a,b) and is illustrated in Fig. 2   |                                     |
| 162<br>163  | The total porosity ( <sup>(III)</sup> ) was already characterized by Wiaux et al. (2014a,b) and is illustrated in Fig. 2<br><u>Porosity was</u> are illustrated in Fig. 1 and 2, respectively.   |                                     |
| 162<br>163<br>164   | The total porosity ( <sup>(a)</sup> ) was already characterized by Wiaux et al. (2014a,b) and is illustrated in Fig. 2<br>Porosity was are illustrated in Fig. 1 and 2, respectively.<br>We measured the total porosity ( <sup>(a)</sup> )it in the laboratory by weighing 100 cm <sup>2</sup> undisturbed soil cores both   | Mis en forme : Non Exposant/ Indice |
| 162<br>163<br>164<br>165  | The total porosity ( <sup>Ø</sup> ) was already characterized by Wiaux et al. (2014a,b) and is illustrated in Fig. 2         Porosity was_are illustrated in Fig. 1 and 2, respectively.         We measured the total porosity ( <sup>Ø</sup> ) <u>it</u> in the laboratory by weighing 100 cm <sup>3</sup> undisturbed soil cores both_         at saturation and after oven drying at 105°C for 48h. We deduced <sup>Ø</sup> from the mass of water needed to   | Mis en forme : Non Exposant/ Indice |
| <ol> <li>162</li> <li>163</li> <li>164</li> <li>165</li> <li>166</li> </ol>   | The total porosity ( <sup>Ø</sup> ) was already characterized by Wiaux et al. (2014a,b) and is illustrated in Fig. 2         Porosity was are illustrated in Fig. 1 and 2, respectively.         We measured the total porosity ( <sup>Ø</sup> )it in the laboratory by weighing 100 cm <sup>3</sup> undisturbed soil cores both at saturation and after oven drying at 105°C for 48h. We deduced <sup>Ø</sup> from the mass of water needed to fill sample pores. We calculated theair-filled porosity (ε) as the difference between <sup>Ø</sup> and volumetric  | Mis en forme : Non Exposant/ Indice |
| <ol> <li>162</li> <li>163</li> <li>164</li> <li>165</li> <li>166</li> <li>167</li> </ol>  | <ul> <li>The total porosity (<sup>Ø</sup>) was already characterized by Wiaux et al. (2014a,b) and is illustrated in Fig. 2</li> <li>Porosity was are illustrated in Fig. 1 and 2, respectively.</li> <li>We measured the total porosity (<sup>Ø</sup>)itin the laboratory by weighing 100 cm<sup>3</sup> undisturbed soil cores both at saturation and after oven drying at 105°C for48h. We deduced <sup>Ø</sup> from the mass of water needed to fill sample pores. We calculated theair-filled porosity (ε) as the difference between <sup>Ø</sup> and volumetric water content (VWC). We calculated average and standard deviation values on triplicate samples for</li> </ul>  | Mis en forme : Non Exposant/ Indice |
| <ol> <li>162</li> <li>163</li> <li>164</li> <li>165</li> <li>166</li> <li>167</li> <li>168</li> </ol>   | The total porosity ( <sup>Φ</sup> ) was already characterized by Wiaux et al. (2014a,b) and is illustrated in Fig. 2<br>Porosity was are illustrated in Fig. 1 and 2, respectively.<br>We measured the total porosity ( <sup>Φ</sup> )itin the laboratory by weighing 100 cm <sup>3</sup> undisturbed soil cores both<br>at saturation and after oven drying at 105°C for 48h. We deduced <sup>Φ</sup> from the mass of water needed to<br>fill sample pores. We calculated theair-filled porosity (ε) as the difference between <sup>Φ</sup> and volumetric<br>water content (VWC). We calculated average and standard deviation values on triplicate samples for<br>each depth.  | Mis en forme : Non Exposant/ Indice |
| <ol> <li>162</li> <li>163</li> <li>164</li> <li>165</li> <li>166</li> <li>167</li> <li>168</li> <li>169</li> </ol>  | <ul> <li>The total porosity (<sup>Ø</sup>) was already characterized by Wiaux et al. (2014a,b) and is illustrated in Fig. 2</li> <li>Porosity was are illustrated in Fig. 1 and 2, respectively.</li> <li>We-measured the total porosity (<sup>Ø</sup>)it in the laboratory by weighing 100 cm<sup>3</sup> undisturbed soil cores both at saturation and after oven drying at 105°C for48h. We deduced <sup>Ø</sup> from the mass of water needed to fill sample pores. We calculated theair-filled porosity (ε) as the difference between <sup>Ø</sup> and volumetric water content (VWC). We calculated average and standard deviation values on triplicate samples for each depth.</li> <li>The assessment of SWR curves was carried out following the commonlywidely used pressure plate</li> </ul>  | Mis en forme : Non Exposant/ Indice |
| <ol> <li>162</li> <li>163</li> <li>164</li> <li>165</li> <li>166</li> <li>167</li> <li>168</li> <li>169</li> <li>170</li> </ol>                           | The total porosity (♥) was already characterized by Wiaux et al. (2014a,b) and is illustrated in Fig. 2         Porosity was -are illustrated in Fig. 1 and 2, respectively.         We measured the total porosity (♥)itin the laboratory by weighing 100 cm3 undisturbed soil cores both_at saturation and after oven drying at 105°C for48h. We deduced ♥ from the mass of water needed to fill sample pores. We calculated theair-filled porosity (ε) as the difference between ♥ and volumetric water content (VWC). We calculated average and standard deviation values on triplicate samples for each depth.         The assessment of SWR curves was carried out following the commonlywidely used pressure plate technique: undisturbed soil samples were submitted to several increasing and discrete pressure values  | Mis en forme : Non Exposant/ Indice |
| <ol> <li>162</li> <li>163</li> <li>164</li> <li>165</li> <li>166</li> <li>167</li> <li>168</li> <li>169</li> <li>170</li> <li>171</li> </ol>              | The total porosity (♥) was already characterized by Wiaux et al. (2014a,b) and is illustrated in Fig. 2         Porosity was_are illustrated in Fig. 1 and 2, respectively.         We measured the total porosity (♥) it in the laboratory by weighing 100 cm3 undisturbed soil cores both_at saturation and after oven drying at 105°C for48h. We deduced ♥ from the mass of water needed to fill sample pores. We calculated theair-filled porosity (€) as the difference between ♥ and volumetric water content (VWC). We calculated average and standard deviation values on triplicate samples for each depth.         The assessment of SWR curves was carried out following the commonlywidely used pressure plate technique: undisturbed soil samples were submitted to several increasing and discrete pressure values inside a closed chamber, with a precise monitoring of soil water content for each pressure level  | Mis en forme : Non Exposant/ Indice |
| <ol> <li>162</li> <li>163</li> <li>164</li> <li>165</li> <li>166</li> <li>167</li> <li>168</li> <li>169</li> <li>170</li> <li>171</li> <li>172</li> </ol> | The total porosity ( <sup>∅</sup> ) was already characterized by Wiaux et al. (2014a,b) and is illustrated in Fig. 2         Porosity was_are illustrated in Fig. 1 and 2, respectively.         We measured the total porosity ( <sup>∅</sup> ) it in the laboratory by weighing 100 cm3 undisturbed soil cores both_at saturation and after oven drying at 105°C for 48h. We deduced <sup>∅</sup> from the mass of water needed to fill sample pores. We calculated theair-filled porosity (ε) as the difference between <sup>∅</sup> and volumetric water content (VWC). We calculated average and standard deviation values on triplicate samples for each depth.         The assessment of SWR curves was carried out following the commonly widely used pressure plate technique: undisturbed soil samples were submitted to several increasing and discrete pressure values inside a closed chamber, with a precise monitoring of soil water content (SWR) curves using We used | Mis en forme : Non Exposant/ Indice |

174 obtained the £100 and b parameters of the Campbell (1974) SWR model by fitting the model to the \_\_\_\_ Mis en forme: Non Exposant/ Indice

175 SWR observations (Moldrup et al. 2000).

#### 176 2.23. Monitoring of Monitoring of aterial and measurements techniques for soil

#### 177 CO<sub>2</sub>, water and temperature monitoring

178 We measured soil CO<sub>2</sub> concentrations using purpose<u>custom</u>-built soil CO<sub>2</sub> probes. The CO<sub>2</sub> sensor in 179 the probe is based on the CARBOCAP® Single-Beam Dual Wavelength non-dispersive infra-red 180 (NDIR) technology (GMM221, Vaisala corp., Vantaa, Finland). The analytical precision is-is function 181 of both the probe characteristicand the value of the observation. This can be calculated as the sum 182 of 1.5% of the measurement range added to and 2% of the observed value. The sampling head of the 183 CO<sub>2</sub> probe is a cylinder of 18.5 mm diameter and 40 mm long, covered with a PTFE 184 (polytetrafluoroethylene) membrane, enabling gas exchange and protection against water infiltration. 185 Since the GMM221 sensors were not designed for wet soil conditions, the sensors were encapsulated 186 into an additional perforated PVC tube, providing an additional protection against water (Fig. 1). This 187 tubing method is an adaptation of the technique presented by Young et al. (2009). We inserted these 188 tubes vertically into the soil, after creating boreholes with a diameter that equals the diameter of the 189 PVC tubes. This approach avoids the need to backfill the bore hole, which will disturb the soil 190 structure and diffusion process. Two rubber stoppers, one at 155 mm from the tube head, and another 191 at the top of the tube, prevented atmospheric air from penetrating into the gas sampling volume. 192 Petroleum jelly on these two rubber stoppers ensured a perfect air- and water-tightness and we verified 193 this under laboratory conditions before using the probes. We used a nylon membrane to avoid soil 194 particles entering the perforated tube and to limit further water infiltration.

195 We adjusted the concentration ranges of the CO<sub>2</sub> probe for each soil depth and for each slope position. 196 This allowed an optimal fit of the probes to the local concentrations. Each probe has to characterize 197 the entire range of values encountered during the seasons while at the same time, time; it should have a 198 sufficiently narrow measurement range to ensure measurement precision. At the summit position, 199 measurements ranged between 0-2 % at 12, 25, 45 cm depth and between 0-5 % at 85 cm depth. At the

| 200 | footslope position, measurements ranged between 0-5 % at 12 cm depth, between 0-10 % at 25 and 45                  |
|-----|--|
| 201 | cm depth and between 0-20% at 85 cm depth.   |
| 202 | To avoided vegetation growth and any autotrophic contribution to the soil respiration, we covered the              |
| 203 | measurement plots with a synthetic permeable geotextile during the complete measurement period. To                 |
| 204 | increase the quality of the soil CO <sub>2</sub> concentration data time-series, we removed observations where the |
| 205 | battery voltage was lower than 11.5 V. We also corrected soil profile CO <sub>2</sub> concentrations               |
| 206 | measurements for temperature variations using the empirical formulas described by Tang et al. (2003).              |
| 207 | This allowed removing the impact of temperature on the $CO_2$ reading of the $CO_2$ probe, since the               |
| 208 | CARBOCAP® technology is temperature dependent. The probe manufacturer (Vaisala corp., Vantaa,                      |
| 209 | Finland) provided probe specific parameters values for the correction formulas.                                    |
| 210 | We also obtained observations of surface CO <sub>2</sub> fluxes by means of a portable infra-red gas analyzer      |
| 211 | with an automated closed dynamic chamber (LI-8100A system, LI-COR, United-States), following                       |
| 212 | Davidson et al. (2002). The sampling design of these surface chamber CO <sub>2</sub> fluxes measurements on        |
| 213 | the same study site has been described in Wiaux et al. (2014 b).   |
| 214 | We monitored soil temperature using a thermistor probe (Therm107, Campbell Scientific Lt., UK).                    |
| 215 | Analytical precision is 0.4°C. We monitored soil volumetric water content (VWC) using Time                         |
| 216 | Domain Reflectrometry (TDR) probes. We used based on Topp's equation (Topp et al., 1980) to                        |
| 217 | determine VWC from the measured apparent dielectric constant measured. We used the                                 |
| 218 | parameterscalibratedinthe close vicinity of our study siteof the Topp's equation asidentified                      |
| 219 | by(Heimovaara, 1993; Garré et al., 2008; Beff et al., -(2013). In this study latter study, the Topp's              |
| 220 | equation was calibrated for an experimental field in the close vicinity of our study site, using the               |
| 221 | method of Heimovaara (1993) and following the protocol described by Garré et al. (2008).                           |
| 222 | We recorded water, temperature and CO <sub>2</sub> concentration profiles measurements with an automatic data      |
|     |  |

- logger (CR1000, Campbell Scientific Lt., UK), connected to a multiplexer (AM16/32, Campbell 223
- Scientific, Campbell Scientific Lt., UK). 224

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#### 225 **2.4. Overall sampling design**

In order to obtain an equilibrated soil environment around the soil VWC, temperature and CO<sub>2</sub>probes,
 we started measurements 1 month after the installation of the probes.We covered the measurement
 plots with a synthetic permeable geotextile during the complete measurement period. This avoided
 vegetation growth and any autotrophic contribution to the soil respiration.

The sampling design is <u>depicted shown</u> in Fig. 4. At each of the 2 slope positions, we measured soil VWC and CO<sub>2</sub> concentrations profiles with 3 replicates on each measurement depth (Fig. 4). <u>We</u> <u>averaged these triplicates, providing an average value for each soil depth and slope position. This</u> <u>allows to account for the spatial variability of VWC and CO<sub>2</sub> concentrations (Maier and Schack-</u> Kirchner, 2014), by extending the measurement footprint to an area of c. 5 m<sup>2</sup>.

235 We collected 18 VWC profiles measurement points (6 soil depths, 3 replicates) were collected, at each 236 of the 2 slope positions. We measured VWC was measured at depths of at a depth of 10, 25, 35, 50, 70 237 and 95 cm depths (Fig. 4). We measured the temperature at 4 soil depths (10, 25, 45, 85 cm) without 238 replicates (Fig. 4). We measured CO<sub>2</sub> concentrations was measured at a depths of 10, 25, 45 and 85 239 cm. We measured the Soiltemperatureat 4 soil was measured at the same depths (10, 25, 45, 85 cm) but 240 without replicates (Fig. 4). Soil temperature and VWC profiles were calculated using a linear 241 interpolation between the depth specific values within the profile. We kept the values constant 242 between the sampling point at the top of the profile and the soil surface. The estimation of CO<sub>2</sub> 243 concentration profiles calculation is described below in detailed in (section 2.5).

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In order to obtain an equilibrated soil environment around the soil VWC, temperature and CO<sub>2</sub> probes,
 measurements started 1 month after the installation of the probes. At the footslope position, hourly
 time-series of VWC, temperature and CO<sub>2</sub> concentrations were recorded from 12 May to 13 December

247 2012 and from 14 May to 22 November 2013. and At the summit position, measurements were

248 recorded for the period from the 2 June to 13 December 2012 and from the 14 June to 22 November

249 <u>2013 at the summit position</u>.

Mis en forme : Titre 3, Interligne : simple We also performed surface  $CO_2$  fluxes measurements with an infra red gas analyzer (IRGA) linked to a survey chamber at 16 dates (profile and surface sampling time was within a 30 minutes time interval). Note that the averaged values of  $CO_2$  concentration for each observation depth cover the same area as the IRGA chamber network located at the soil surface (Fig. 4). These reference surface  $CO_2$  fluxes allowed calibrating parameters of the soil gas diffusion model, ensuring the accuracy of profile  $CO_2$  fluxes (section 2.43).

256 We adjusted the concentration ranges of the CO<sub>2</sub> probe for each soil depth and foreach slope position. 257 This allowed an optimal fit of the probes to the local concentrations. Each probe has to characterize 258 entire range of values encountered during the seasons while at the same time, it should have a 259 nsuremeasurement precision summit position. 260 ranged between 0.7 % at 12 25 45 cm depth and between 0 at 85 cm depth. At the 261 ranged between 0.5 % at 12 cm depth, between 0-10 % at 25 and 45 262 depth and between 0-20% at 85 cm depth

We recorded hourly time series of VWC, temperature and CO<sub>2</sub> concentrations from 12 May to 13
December 2012 and from 14 May to 22 November 2013 at the footslope position and from the 2 June
to 13 December 2012 and from the 14 June to 22 November 2013 at the summit position. In 2012,
important parts of CO<sub>2</sub> measurements were not recorded as a result of sensors failures and/or the use
of an unsuitable initial measurement range of some sensors.

268 increase the quality of the soil concentration data time carias we removed observations where the 269 270 variations using the empirical formulas described by Tang et al. (2003) 271 This allowed removing the impact of temperature on the CO<sub>2</sub>-reading of the CO<sub>2</sub> probe, since the 272 CARBOCAP® technology is temperature dependent. The probe manufacturer (Vaisala corp., Vantaa, 273 Finland) provided probe specific parameters values for the correction formulas.

- 274 We averaged triplicate VWC and CO<sub>2</sub> concentrations data, providing an average value for each soil
- 275 depth and slope position. Note that averaging strategy allows to account for the spatial variability of

276 VWC and CO<sub>2</sub> concentrations (Maier and Schack Kirchner, 2014), by extending the measurement
 277 footprint to an area of c. 5 m<sup>2</sup>.

We calculated soil temperature and VWC profiles using a linear interpolation between the depth specific values within the profile. We kept the values constant between the sampling point at the top of the profile and the soil surface. We calculated the  $CO_2$  concentrations profiles by fitting Eq. 2 to the observations. We evaluated the performance of thisfitting by means of the regression coefficient ( $\mathbb{R}^2$ ). When the  $\mathbb{R}^2$  values were lower than a threshold value of 95%, we considered the  $CO_2$ concentration profile as unreliable and we did not retain the resulting  $CO_3$ fluxes in final analysis.

#### 284 **2.35**. Calculation of the CO<sub>2</sub> fluxes profiles

We calculated the CO<sub>2</sub>flux using Fick's first law of diffusion according to the gradient method (Eq. 1,
e.g. Maier and Schack-Kirchner, 2014):

287 
$$F_{CO_2} = -D_s \frac{\partial CO_2}{\partial z}$$
(Eq. 1)

where  $F_{CO_2}$  is the soil CO<sub>2</sub> flux [µmol m<sup>-2</sup> s<sup>-1</sup>], D<sub>s</sub> the diffusivity of CO<sub>2</sub> in soil [m<sup>2</sup> s<sup>-1</sup>], CO<sub>2</sub> the soil CO<sub>2</sub> concentration [µmol m<sup>-3</sup>], and  $\frac{\partial CO_2}{\partial z}$  the vertical soil CO<sub>2</sub> gradient (with "z" representing the soil depth).

| 291 | In order to calculate the vertical soil $CO_2$ gradient, we used suggest an -new-equation that accountings                              |
|-----|---|
| 292 | for some-curve concavity variations a double sigmoidal equation (Eq. 2), ). Variationsofin curve  |
| 293 | concavity in CO <sub>2</sub> concentration profiles hashave already been observed reported in the literature (e.g Mis en forme : Indice |
| 294 | Maier and Schack-Kirchner, 2014). which allows accounting for some curve concavity variations   |
| 295 | (Maier and Schack Kirchner, 2014): In this study, we built Eq.2 to consider this issue and improve the                                  |
| 296 | model fit to CO <sub>2</sub> concentration profiles. We evaluated the performance of this fitting by means of the Mis en forme : Indice |
| 297 | regression coefficient ( $R^2$ ). When the $R^2$ values were lower than a threshold value of 95%, we                                    |
| 298 | considered the CO <sub>2</sub> concentration profile as unreliable and we did not retain the resulting CO <sub>2</sub> fluxes in        |
| 299 | the final analysis.   |

300 
$$\operatorname{CO}_{2}(z) = 0.04 + A\left(\left(\frac{1}{1+e^{-\gamma_{1}z}}\right) + \left(\frac{1}{1+e^{-\gamma_{2}(z-d)}}\right) - \left(\frac{1}{2} + \frac{1}{e^{\gamma_{2}d}+1}\right)\right) (\operatorname{Eq.} 2)$$

where z is the soil depth [cm], d is the soil depth [cm] at which the sharpness of the curve changes due to a diffusion barrier,  $\gamma_1$  and  $\gamma_2$  [cm<sup>-1</sup>] are fitted parameters which characterize the sharpness of the curve, respectively above and below the soil depth d, and A [%] is a reference value used to define the fitted asymptotic value of the CO<sub>2</sub> concentration at infinite depth. We fitted the A, d,  $\gamma_1$  and  $\gamma_2$  parameters for each CO<sub>2</sub>-profile using the trust-region-reflective optimizational gorithm in Matlab ©. The derivative of Eq. 2 provided the CO<sub>2</sub>gradient ( $\frac{\partial CO_2}{\partial z}$ ) used in Eq. 1 to calculated the CO<sub>2</sub> fluxes...

308 The diffusivity of  $CO_2$  in soil,  $D_{s}(\underline{in} Eq. 1)$ , is a function of the diffusivity of  $CO_2$  in free air (varying 309 with temperature T and pressure, e.g. Davidson *et al.*, 2006) and of the gas tortuosity factor ( $\xi$ ) (Eq. 3):

310 
$$D_s = \xi \, 1.47 \, 10^{-5} \left(\frac{T+273}{273}\right)^{1.75}$$
 (Eq. 3)

whereξ depends on soil physical and hydrological properties.\_We used the Moldrup\_et\_al. (2000)\_\_\_\_ Mis en forme : Police :Non Italique
model (Eq. 4)which was showntoprovide the most accurate and preciseresults (Davidson et al., 2006;
Goffin et al., 2014);

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314 
$$\xi = (2\varepsilon_{100}^3 + 0.04\varepsilon_{100}) \left(\frac{\varepsilon}{\varepsilon_{100}}\right)^{2+3/b}$$
(Eq. 4)

315 where  $\xi$  is the gas tortuosity factor,  $\varepsilon$ [m<sup>3</sup> m<sup>-3</sup>] is the soil air-filled porosity, b[-] is the slope of the 316 Campbell (1974) soil water retention curve modelbetween -100 and -500 cm H<sub>2</sub>O water suction, and 317  $\varepsilon_{100}$ [m<sup>3</sup> m<sup>-3</sup>] is the soil air-filled porosity at a soil water potential of -100 cm H<sub>2</sub>O.

318  $CO_2$  fluxes, as assessed by the gradient based method, were calculated on an hourly time-scale, and 319 then integrated on a daily basis. Temperature, VWC, diffusivity and  $CO_2$  concentration values were 320 also averaged on a daily basis.

| 322 | diffusivity coefficient for the entire soil profile or for an entire soil layer. Contrary to Coffin et al.                                 |                                       |
|-----|--|---------------------------------------|
| 323 | (2014) and Maier and Schack-Kirchner (2014), we did not calculate the CO fluxes from each soil   | - <b>Mis en forme :</b> Indice        |
| 324 | slicebased on the difference of CO2-concentrations between the top and the bottom of soil horizons.  | Mis en forme : Indice                 |
| 325 | but we rather assessed a continuous profile of CO2-fluxes and production. We considered the vertical                                       | Mis en forme : Indice                 |
| 326 | distribution explicitly, and integrated Eq. 4 in the finite difference numerical solution of Eq. 1. In this                                |                                       |
| 327 | numerical integration, we used a depth increment of 0.1cm and constrained the surface $CO_2$   |                                       |
| 328 | concentrations with atmospheric CO <sub>2</sub> levels (i.e. $400 \text{ ppm}0.04\%$ ). In addition, and <u>Econtrary to Goffin</u>        |                                       |
| 329 | et al. (2014) and Maier and Schack-Kirchner (2014), we did not calculate the CO2 fluxes from each  |                                       |
| 330 | soil slice based on the difference of $CO_2$ concentrations between the top and the bottom of soil   |                                       |
| 331 | horizons, but we rather assessed a continuous profile of CO <sub>2</sub> fluxes and production.  |                                       |
| 222 | 2.6 Calibration of the gradient-based COs fluxes with surface direct observationst   | - Mis on formo : Titre 3 Interligne : |
| 332 | 2.0. calibration of the gradient-based Co2 nuxes with surface unect observations   | simple                                |
| 333 | at the soil surface  |                                       |
| 334 | We calibrated the diffusion model by adjusting the parameters related to the gas diffusion coefficient                                     |                                       |
| 335 | (i.e. b and $\varepsilon_{100}$ ) in such <u>a way</u> that calculated fluxes fit <del>punctual instantaneous</del> CO <sub>2</sub> fluxes |                                       |
| 336 | observations at 16 dates spread along the measurement period. This calibration ensures the   |                                       |
| 337 | consistency, and consequently the precision, of the calculated CO <sub>2</sub> fluxes. We obtained these                                   |                                       |
| 338 | observations by means of a portable infra red gas analyzer with an automated closed dynamic chamber  |                                       |
| 339 | (LI 8100A system, LI COR, United States), following Davidson et al. (2002). The sampling design of   |                                       |
| 340 | these surface chamber CO2 fluxes measurements on the same study site has been described in Wiaux   |                                       |
| 341 | et al. (2014 b)Comparing the gradient-based CO <sub>2</sub> fluxes with directly measured IRGA CO <sub>2</sub> fluxes,                     |                                       |
| 342 | we obtained a good precision of prediction with a $R^2$ of 92% for all soil types profiles together (Fig.                                  | Mis en forme : Exposant               |
| 343 | 5). This ensures the consistency (and consequently the precision) of the calculated fluxes. In addition,                                   |                                       |
| 344 | <b>T</b> the slope of the fit (i.e. 1.05 and 1.22, respectively in 2012 and 2013, Fig. 5) was used to correct the                          |                                       |
| 345 | calculated estimated fluxes and tofor ensure ensuring the accuracy. Thise comparison between   |                                       |
| 346 | gradient-based calculation and observation of observed surface CO2 fluxes, which allowed the   | Mis en forme : Indice                 |
| 347 | optimization of the calculated fluxes, is illustrated on Fig. 5.   |                                       |
|     |  |                                       |

In contrast to other studies (e.g. Pingintha et al., 2010; Turcu et al., 2005), we did not aggregate the soil

#### 348 **2.47**. Vertical partitioning of CO<sub>2</sub> fluxes

We partitioned the continuous  $CO_2$  flux profiles obtained usingEq.2into 10 slides of 10 centimeters along the soil profile. For each soil slide, we calculated the difference between the top and bottom fluxes. We divided this difference by the total  $CO_2$  flux (e.g. the value at the soil surface). This provides the relative contribution in terms of both  $CO_2$  production and transfer (in %) of each soil slide to the surface  $CO_2$  flux(e.g. Goffin et al., 2014; Maier and Schack-Kirchner, 2014).

In order to allow an easy representation of the temporal dynamic of this vertical partitioning, we averaged values on a semi-seasonal time-scale of one month and a half, representaing the beginning or the end of a season. Standard deviation values reflect the variability overtime during each semi-half season.

#### 358 **3. Results**

#### 359 3.1. Spatio-temporal analysis of measured soil variables

360 Fig. 56 to Fig. 10 respectively shows the spatio-temporal variation of soil temperature-and, moisture, whileFig. 6 shows the spatio temporal variation of CO2 fluxes, concentrations and diffusion. All these 361 362 values correspond to in-situ measurements during a 6 month period in 2013. Similar measurements 363 have been carried out in 2012 and display similar spatio-temporal trends (data not shown). Here, we 364 The focus is the observation of on the temporal dynamics of each of these measured variables, as well 365 as the shape of the spatial vertical distribution of these variables along the soil profiles. The 366 linkrelationship between these variables was previously analyzed in Wiaux et al. (2014b) and is not 367 deepened this is not further discussed here. It should be noted that the Comparisons of the profile 368 distribution at different dates or of temporal dynamics at different depths remainis done in a 369 qualitative mannerand have not been submitted to further statistical analyses.

During the observation period, the\_soil temperature (Fig. <u>56A</u>) <u>didseems tonot significantly differ</u>
betweenshows a rather similar evolution at the summit and the footslope, although higher temperatures
were observed at the summit profile for some shorter periods(e.g. day of year 180 to 220 where

temperatures areapproximately 2 to 3 °C higher). The mean daily surface temperatures at the soil
surface ranges between 4°C to 28°C at the summit, and between 4°C to 25°C at the footslope.

375 The space-time dynamics of the soil volumetric water content (VWC, Fig. 5B7) differ 376 substantiallybetween the summit and the footslope profiles. At the footslope, the observed soil VWC 377 at different soil depthsvaried in a narrow range(0.36 to 0.39 cm<sup>3</sup> cm<sup>-3</sup>). In contrast, soil VWC at the summitvariedbetween0.23 to 0.34 cm<sup>3</sup> cm<sup>-3</sup> for the plow layer (0-30 cm depth) and higher values 378 379 (approximately 0.539 cm<sup>3</sup> cm<sup>-3</sup>) were observed for the rest of the soil profile. The soil at the summit position wasthe wettest during the early spring and the late autumn and driest in the summer. At the 380 381 footslope, soil VWC reachedthe saturation level in the early summer after an important rainfall event 382 and then slowly decreaseduntil the early autumn and reachedsaturation again in the late autumn.

In contrast<u>InverselyIn contrast</u> to the VWC, and as expected given the physical dependence of
 diffusivity to soil water content (Eq. 4, section 2.5), the soil gas diffusivity (Fig. 6C8) reachedits
 maximum value in the summer at the summit while it was low at the footslope. Soil gas diffusivity
 was approximately 10 times lower at the footslope than at the summit.

The soil CO<sub>2</sub>concentrations at both the summit and the footslope\_increased gradually\_from spring to
late summer (Fig. 9a). Thereafter, concentrations dropped again and lowest valueswere observed in the
late autumn.

Theranges of  $CO_2$  fluxes obtained for thefootslope and summit profiles were very similar (Fig. 10a). However, their temporal distribution was different: the periods characterized by high  $CO_2$  fluxes didnot occur at the same time and had a different duration. More precisely, at the summit, peaks of  $CO_2$ fluxes appear at the early summer and disappear after one month, while at the footslope, peaks of  $CO_2$ fluxes appear at the early autumn and are 30% lower than at the summit but remain constant during two months. For all soil profiles,  $CO_2$  fluxes decreased with depth and reached null values at approximately30 cm depth at the summit and at approximately 15 cm depth at the footslope.

#### 397 3.2. Shape and variability of CO<sub>2</sub> concentrations and fluxes profiles

The observed soil CO<sub>2</sub> concentrations (Fig. 6Bb)increased\_with soil depth\_(Fig. 9b), from the atmospheric value of 0.04 % at the surface to concentrations which weretwo orders of magnitude higher at 100 cm depth (CO<sub>27</sub>(z) in Eq.2)(Fig. 6Bb). For the measurement period of 6 months considered here, CO<sub>2</sub>concentration values at 100 cm depth werethree to four times higher at the footslope position than at the summit position. In 2013, these values rangedbetween from 0.86 to 3.46 % at the summit position and betweenfrom 3.68 to 9.12 % at the footslope position.

404 The observed CO<sub>2</sub> concentration profiles (Fig. 9b) followed a double exponential trend (Eq. 2). This 405 particular model built in this study to represent soil CO<sub>2</sub> concentration profiles (Eq. 2) fits our 406 observations relatively well, with regression coefficients ranging between 97 to 100%. Thesecond 407 exponential curve startsapproximately at the middle of the profile, and is particularly pronounced at 408 the footslope, reflecting a shift of\_nearly\_4% CO2between 44 and 100 cm depth. Standard deviations 409 around averaged values of observed hourly CO<sub>2</sub> concentrations at each depth are given in Table 1. 410 The small-scale spatial variability is low relative to the mean values of  $CO_2$  concentrations, the only 411 exception being the footslope at 25 cm depth where the maximum standard deviation exceeded the 412 maximum mean value.

413 The  $CO_2$  fluxes (Fig. <u>6A10</u>) were calculated based on both  $CO_2$  concentrations and diffusivity. For all 414 soil profiles (Fig. 10a),  $CO_2$  fluxes decreased with depth and reached null values at c.30 cm depth at 415 the summit and at c. 15 cm depth at the footslope.

#### 416 **3.3. Vertical partitioning of CO<sub>2</sub> fluxes**

The distribution of the soil  $CO_2$  fluxes in the profile is illustrated in Fig. 7<u>11</u>. At the footslope,90 to 95 % of the surface  $CO_2$  fluxes weregenerated in the first tencentimeters of the soil profile. The soil layer between 10 and 20 cm contributed for only 5 to 10 %, and the deeper layers didnot significantly contribute to the surface fluxes. At the summit (Fig. 11a), the relative contribution of the different soil layers was more dynamic in time, with a contribution of the first tencentimeters of the soil profile

ranging from 80 % at the late spring, decreasing to 60 % in the early summer, and reaching 40 % from 422 423 late summer to the late autumn. At the summit (Fig. 11a), the first 30 centimeters of the soil profile 424 significantly contributed to surface fluxes. This contribution decreased with depth in the late spring and the early summer, but is homogeneously distributed with depth for the rest of the time. At the 425 426 summit (Fig. 11a), soil layers deeper than 30 cm depth sometimes contributed for up to 20% of the 427 total flux, especially in the autumn. Between 40 to 50 cm depth, and 80 to 90 cm depth, some negative contribution (i.e. CO<sub>2</sub> uptake) up to -20% is also observed. At the footslope (Fig. 11b), 90 to 95 % of 428 429 the surface CO<sub>2</sub> fluxes were generated in the first ten centimeters of the soil profile. The soil layer 430 between 10 and 20 cm contributed for only 5 to 10 %, and the deeper layers did not significantly 431 contribute to the surface fluxes.

#### 432 **4. Discussion**

433 4.1. Soil physical control on CO<sub>2</sub> emissions

434 The observed differences between the footslope and summit soil profiles, in terms of the temporal 435 dynamicsevolution of surface soil CO<sub>2</sub> fluxes between the footslope and summit soil profiles (Fig. 436 6A10), indicates that the controlling\_factors are not the same. At the summit, on one hand, the 437 dynamic evolution of surface soil CO<sub>2</sub> fluxes (Fig.  $6A_{10}$ ) clearly follows the temperature variations 438 (Fig. 5A<sub>6</sub>, maximum during the summer). At the footslope, on the other hand, the soil surface  $CO_2$ 439 flux was small even when temperature increased and remained relatively smallthroughout the summer 440 period (Fig. 6A10). This is most likely related to the high VWC values observed at the footslope\_(Fig. 441 **5B**7), as it is well known that VWC negatively impacts soil  $CO_2$  emissions (e.g. Webster et al., 2008b; Perrin et al., 2012; Wiaux et al., 2014b). More precisely, we suggest that <u>VWC is not the only factor</u> 442 443 controlling CO<sub>2</sub> emissions at the footslope, is not only VWC\_in itself but also that the difference between the VWC and the water saturation level of the soil pore spaces, i.e. the water-filled pore 444 445 spaces, also plays an important role. While the VWC at the footslope remained high throughout the 446 year, we observed that the soil surface  $CO_2$  flux dramatically increased when the air-filled pore spaces 447 becomes high enough, which is illustrated by the gas diffusivity exceeded exceeding a threshold value

| 448 | of c. 0.1 cm <sup>2</sup> d <sup>-1</sup> (i.e. from day 255 to 305 of year 2013, Fig. $6A_{10}$ ). Hence, we argue that the |  |
|-----|--|--|
| 449 | sometime occasionally s low $CO_2$ emissions at the footslope profileare related to the fact that a high                     |  |
| 450 | VWC, as described in the literature by the bimodal effect of VWC on CO <sub>2</sub> emissions (e.g. Davidson et              | Mis en forme : Indice                        |
| 451 | al., 1998; Perrin et al., 2004; Webster et al., 2008b; Castellano et al., 2011; Bauer et al., 2012; Wiaux                    | ( <b>Mis en forme :</b> Anglais (États-Unis) |
| 452 | et al., 2014b). Indeed, according to these authors, when exceeding a threshold VWC value is exceeded,                        |  |
| 453 | <u>VWC</u> both <u>this</u> : (i) strongly limits the transfer of biotic $CO_2$ along the soil profile, and (ii) reduces the |  |
| 454 | production of CO <sub>2</sub> in itself due to the lack of oxygen for the microbial community. In both cases, the            |  |
| 455 | lower CO <sub>2</sub> emissions at the footslope profile relative to the summit, are due to gas diffusion limitations        |  |
| 456 | (even indirectly in the case of oxygen lack), as also suggested by Ball (2013). This is stands in sharp                      |  |
| 457 | contrast to the summit profile where gas can easily diffuse throughout the year and along the entire                         |  |
| 458 | soil profile (Fig. <del>6C</del> 8).   |  |
| 450 | In the period preceding theimportant CO, emissions (i.e. from day 255 to 305 of year 2013. Fig                               |  |
| 439 | In the period preceding memportant $CO_2$ emissions (i.e. non day 255 to 505 of year 2015, Fig.                              |  |
| 460 | $6A_{10}$ , the soil CO <sub>2</sub> cannot move along the soil profile and accumulates within soil pores. This results      |  |
| 461 | in <u>entails</u> an increase of the $CO_2$ concentration increase both in <u>during</u> the early and the late summer,      |  |
| 462 | especially below 50 cm depth (Fig. 69), where a compacted soil layer appears (. This phenomenon is                           |  |
| 463 | particularly evident below the compacted soil layer between 40 cm and 50 cm depth. Based                                     |  |
| 464 | onaccordingsecto the porosity profile illustrated in Fig. 1), thThis suggests that for our footlslope soil                   |  |
| 465 | profile, which is a ColluvicRegosols, gas diffusion barriers strongly impact the $CO_2$ concentration                        |  |
| 466 | profile at the footslope. As a result of these gas diffusion barriers, 90 to 95% of fluxes occur from the                    |  |
| 467 | top soil (i.e. the first 10 cm) at this location (Fig. 11),This suggests that contributions of deep soil                     |  |
| 468 | layers would could be likely higher without theses diffusion barriers. This may occur in dry conditions                      |  |
| 469 | where even compacted soil layers can display a low proportion of water in pore spaces. For now, tThe                         |  |
| 470 | permanently high water content (Fig. 7), at least during the period of observations, measured at this                        |  |
| 471 | downslope location prevents the contribution of deeper soil layers. While this soil profile remains wet                      |  |
| 472 | all the time, the temporal dynamics of VWC and gas diffusion at the footslope (Fig. 7-8)in turn entails                      |  |
| 473 | acontrol the time-dynamic behaviorand hence the temporal dynamics of soil surface_CO <sub>2</sub> fluxes (Fig.               |  |
| 474 | 10). This-This is in agreement with recent studies (e.g. Maier et al., 2011; Schmidt et al., 2011; Ball,                     |  |
|     |  |  |

2013) that show that soil pore continuity and sizephysical properties are key to understand the
mechanisms regulating the soil gases emissions. Our study brings new insights in the current literature
by illustrating demonstrating the strong bridgelinkages between soil physical properties and CO<sub>2</sub>
emissions based on in-situ and depth-explicit observations. However, while further work on this topic
is still needed to better understand the specific part oprocesses controlling f-the-microbial inhibition

and of the gas transfer inhibition incase of soil diffusion barriers.

480

481 As a consequence, we argue that the significantly higher CO<sub>2</sub> concentrations observed at the footslope, 482 especially fordeeper soil layers, are probably not only related to the large amount of labile OC that was 483 found at this position (shown in Wiaux et al., 2014a,b), but more likely result from thelong term 484 accumulation (i.e. during periods with a very low diffusivity) of the CO<sub>2</sub> produced by the 485 mineralization of this important large labile OC stockduring periods with a verylow diffusivity. Maier 486 et al. (2011) showed that the CO<sub>2</sub> efflux (observed CO<sub>2</sub> flux resulting from all transfer and production 487 mechanisms together) can deviate in time from the instantaneous soil respiration (due to micro-488 organisms metabolic activity) due to because of the  $CO_2$  storage into soil pore spaces. Hence, we our 489 data suggest that at the footslope, soil physical properties are the dominant control onsurface  $CO_2$ 490 fluxes. In other words, while the footslope profile contains more labile OC in the subsoil relative to the 491 summit (Fig. 1, Wiaux et al., 2014a), there is a lowercontribution from the subsoil to the overall 492 respiration fluxes due to physical limitations (both low diffusivity and lack of O<sub>2</sub>).

493 In summary, we our study highlights that the mechanisms that govern soil surface CO<sub>2</sub> emissions are 494 highly variable in both space and time. On a well-drained soil at the summit of a hillslope, the 495 observed soil CO<sub>2</sub> emissions weredirectly related to soil microbial respiration and CO<sub>2</sub> production 496 (demonstrated ine.g. Wiaux et al., 2014b). However, at the footslope of the hillslope, which is 497 characterized by a different hydrological regime, we observed that the temporal dynamic of soil CO<sub>2</sub> 498 emissions were more closely related to physical transfer mechanisms: long periods of CO<sub>2</sub> production 499 and accumulation alternate with periods of important release at the soil surface. Indeed, even if When 500 considering a situation where gas diffusion is limited, -(and that consequently as a result, also oxygen 501 supply for micro-organisms is limitedlow), we argue that oxygen concentration in soil pore spaces is

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| 502 | not completely null. Hence, the remaining oxygen allows CO2 production through microbial  | Mis en forme             |
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| 503 | respiration, especially at the footslope due to the high amount of labile soil OC (Wiaux et al.,                                | Mis en forme :<br>Indice |
| 504 |   | Mis en forme :           |
| 504 | 2014b). This CO <sub>2</sub> then tirst accumulates under the soil diffusion barriers. This accumulated CO <sub>2</sub> is then | Mis en forme :           |
| 505 | later emitted when VWC decreases under a threshold value which allows a significant gas diffusion, as                           | Mis en forme :<br>Indice |
| 506 | suggested by Majer et al. (2011) and Ball (2013). The main implication of these observations is that if                         | Mis en forme :           |
| 500 | suggested by Maler et al. (2011) and Ban (2015). The main impleation of these observations is that in                           | Mis en forme :           |
| 507 | hydrologic regimes change and that footslope soils become drier (reaching moisture conditions                                   |                          |
| 508 | favorable for micro-organisms respiration and gas transfer), there is a large amount of potentially                             |                          |
| 509 | easily decomposable OC stored at depth that can suddenly decompose and be emitted to the  |                          |
| 510 | atmosphere.   |                          |
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#### 512 **4.2. Soil organic carbon storage in downslope deposits**

513 The soil respiration rate can be interpreted as an indicator of soil OC persistence (e.g. Gregorich et al., 514 1994; Wiaux et al., 2014a,b). However, a further analysis of what occurs along the soil profile is 515 needed to thoroughly answer the question of the persistence of OC. The vertical partitioning of the soil 516  $CO_2$  fluxes, as illustrated in Fig.711, shows that during the observation period, 90 to 95 % of the 517 surface  $CO_2$  flux originated from the first tencentimeters of the soil profile at the footslope. Given the 518 important amount of OC until up to 100 cm depth in our study site (Fig. 1, Wiaux et al., 2014 a), this 519 observation is not in agreement with the study of Goffin et al. (2014), whosuggested that the relative 520 contribution of asoil layer to the surface CO<sub>2</sub> fluxes is related to OC distribution along the soil profile. 521 However, while similarities exist in the physical controls and the method used to calculate the vertical 522 partitioning, the study of Goffin et al. (2014) reports on CO<sub>2</sub> production in forest soils-. Comparing 523 forest and crop soils is difficult because of the important part of theautotrophic respiration 524 comingoriginates from roots in forest while this can be easily avoided is less important in cropland 525 soils(e.g. Davidson et al., 1998; Epronet al., 2006; Martin and Bolstad, 2009; Webster et al., 2008b; 526 Goffin et al., 2014). Hence, in the case of forest ecosystems, the dense roots network in soilcreatesinterferences when measuring heterotrophic CO<sub>2</sub> fluxes, and this has been shown to explain 527

| 529 | (Goffin et al., 2014).In addition, the estimation of CO <sub>2</sub> production in forest soils areis more difficult as |
|-----|---|
| 530 | biased due to anincorrect description of CO2 transport inthis layer that neglects turbulent advection                   |
| 531 | needs to be accounted for (i.e. the predominance of non-diffusive transport in the litter layer, Goffin et              |
| 532 | al., 2014). All these elements preventingmake difficultany direct and quantitative comparison beween                    |
| 533 | forest and agro-ecosystems difficult. However, Wwe can although observe some qualitative similarities                   |
| 534 | between our observations and those of Goffin et al. (2014) in forest soils: (i) surface soil VWC values                 |
| 535 | and dynamics were shown to be a critical factor in accurately estimating topsoil CO <sub>2</sub> production, and        |
| 536 | (ii) the vertical distribution of CO <sub>2</sub> concentration increased with depth while CO <sub>2</sub> production   |
| 537 | decreased with depth.   |

an important part of the vertical distribution of CO<sub>2</sub> production along soil profiles in forest ecosystems

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538 In addition, the substantial contribution of the upper soil layers found here wasnotrelated to higher 539 temperatures (Fig. 5A6), contrary to what was suggested by Takahashi et al. (2004). According to the 540  $CO_2$  concentration and diffusivity profiles (Fig. 6C8), the relative contribution of the soil layers to the 541 surface CO<sub>2</sub> flux is more likely governed by soil physical controls (Ball, 2013) rather than by 542 biological production depending on thermal energy and OC substrate. Here, soil gas diffusivity 543 strongly decreases from 10 to 40 cm depth (where diffusivity is null) at the two slope positions, and 544 the profile of  $CO_2$  concentration displays no gradient between 10 and 40 cm depth, particularly at the footslope (Fig. 6A9). 545

Here, weOur data showed that despite the fact that the footslope profiles generates CO<sub>2</sub> fluxes which 546 547 exceed those observed at the summit position (demonstrated in Wiaux et al., 2014b), the contribution 548 of soil layers below10 cm depth is very small(Fig. 711). The OC in the top layer of the soil profile 549 (i.e.0-10 cm) contributed forc. 90% of the total  $CO_2$  flux at the footslope position (Fig. <u>117</u>). This can be explained by environmental conditions specific to this 0-10 cm layer playing in favor of both 550 551 microbial respiration and gas diffusion. There are no limitations related to both diffusion barriers and 552 access to the oxygen disappear close to the soil surface. Hence, the only impact of soil VWC on soil 553 respiration is its positive effect as it provides a more easy access forsoil micro-organisms to their OC 554 substrate, and to the enhancement of their metabolic activities by water (Akinremi et al., 1999;

Castellano et al., 2011; Herbst et al., 2008; Howard and Howard, 1993; Šimůnek and Suarez, 1993).
The combination of this high amount and high quality of soil OC (Fig. 1, as described by Wiaux et al.,
2014a) with this net positive effect of soil VWC results in a strong increase of microbial respiration
rates.

Finally, our results suggest that buried soil OC in colluvial deposits is effectively protected from mineralization below 10 cm depth, which corroborates the assumption of a long-term stabilization of buried OC in colluvial soils as suggested in the literature (e.g. Doetterl et al., 2012; Berhe et al., 2008, 2012a).<u>This also corroborates the notion of Schmidt et al. (2011) suggesting that deep soil OC may be</u> protected because of unfavorable physical conditions rather than substrate limitations.

#### 564 **5. Conclusion**

565 In this study, we evaluated the factors controlling soil carbon dioxide fluxes for two soil profiles along 566 a hillslope characterized by contrasting physical and chemical characteristics. At the summit position 567 of the studied hillslope, the time course of surface soil CO<sub>2</sub> fluxes was strongly related to soil temperature and maximum CO<sub>2</sub> fluxes were observed during the summer. Here, the observed soil CO<sub>2</sub> 568 569 emissions are directly related to soil micro-organisms respirationand associated to biotic CO<sub>2</sub> 570 production. In contrast, the higherlevels of water filled pore space observed at the footslope 571 profiles, strongly limited the transfer of biotic CO<sub>2</sub> throughout the soil profile and likely the transfer of 572 O<sub>2</sub> to deeper soil depths.Here,tThe soil surface CO<sub>2</sub> flux substantially increased substantially for 573 during limited amounts of timeshort periods when the gas diffusivity exceeded a threshold value 574 related to sufficient air-filled pore spaces. As a result, the time course of observed soil CO<sub>2</sub> emissions 575 was to a large extentexplained by physical transfer mechanisms: long periods of accumulation 576 alternate with shorter periods of important CO<sub>2</sub>release. The vertical partitioning of the soil CO<sub>2</sub> fluxes 577 for the footslope profiles showed that, during the observation period, 90 to 95 % of the surface CO<sub>2</sub> 578 fluxes originated from the first 10 centimeters of the soil profile. This study highlights the need to 579 consider soil physical properties and their dynamics when estimating and modeling soil CO<sub>2</sub> 580 emissions. This study highlights the need to include soil physical properties and their dynamics directly 581 oil OC models. The main implication is that When considering if changes in hydrologic regimes,

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Mis en forme : Police :(Par défaut) Times New Roman, 11 pt, Non Gras, Indice e.g. the <u>-change and that</u>-footslope soils become drier (reaching moisture conditions favorable for
 micro-organisms respiration and gas transfer), there is a large amount of potentially easily
 decomposable OC stored at depth that can suddenly decompose and be emitted result in an additional
 emission of C to the atmosphere.

#### 586 Author contribution

587 F.W. designed the experiments, and carried out the research. M.V., K.V.O. and F.W. analyzed the 588 results. F.W. wrote the main part of the paper and prepared the manuscript with contributions from all 589 co-authors.

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#### 775 Tables

 776
 Table 1. Range of standard deviation (S.D.) and averaged—mean\_values of triplicated measured hourly CO2

 777
 concentrations at each depth, both at the summit and at the footslope position. This range is indicated by minimum

 778
 (Min) and maximum (Max) values encountered along time (hourly time series) during the 6 months measurement

779 period. <u>NI means No Information (i.e. due to a lack of replicates to allow reliable mean and S.D.).</u>

| Summit position       |                    |                    |                 | Footslope position |                    |                    |                 |                 |
|-----------------------|--------------------|--------------------|-----------------|--------------------|--------------------|--------------------|-----------------|-----------------|
| Soil<br>depth<br>[cm] | Min<br>mean<br>[%] | Max<br>mean<br>[%] | Min<br>S.D. [%] | Max<br>S.D. [%]    | Min<br>mean<br>[%] | Max<br>mean<br>[%] | Min<br>S.D. [%] | Max<br>S.D. [%] |
| 10                    | 0.07               | 1.39               | 0.00            | 0.71               | 0.26               | 4.75               | 0.00            | 3.13            |
| 25                    | 0.06               | 1.83               | 0.00            | 0.68               | 0.30               | 3.93               | 0.00            | 5.32            |
| 45                    | NI                 | NI                 | NI              | NI                 | 0.12               | 3.96               | 0.00            | 1.96            |
| 95                    | 0.15               | 2.83               | 0.00            | 1.42               | 0.48               | 7.52               | 0.00            | 2.48            |

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781

#### 783 Figures



784

785 Fig. 1.Soil profiles (0-100 cm) of both soil total OC and labile OC pool concentrations [C%], at the summit and

786 footslope positions. Error bars indicate 1 standard deviation (n≥3).



788 Fig. 2. Soil porosity profiles at the footslope (plain line) and at the summit (dashed line) positions. Error bars indicate

789 1 standard deviation (n≥3). Continuous lines are linearly interpolated values.



791 Fig. 3. Description of the probes used for CO2 concentration measurements inside the soil.



792

Fig. 4.Schematic description of the experimental plot (sampling design) at each slope position showing how temperature, VWC, CO<sub>2</sub> concentrations and CO<sub>2</sub>fluxes probes collocate with each others. Probes have been inserted at different locations both vertically and horizontally. Consequently, all of them are not in the same plane (i.e. depth lines with axes labels on the right hand-side illustrate the foreground profile and depth lines with axes labels on the Pleft hand-side illustrate the background profile).







809 Fig. 6. Space-time dynamic of soil temperature at the summit (red) and the footslope (black) position in 2013: (a) time

- 810 series at different depths; (b) Profile at different dates.
- 811



813 Fig. 57. Space-time dynamic of soil temperature (A) and moisture (B) at the summit (red) and the footslope (black)





816 Fig. 8. Space-time dynamic of soil CO<sub>2</sub>diffusivity, at the summit (red) and the footslope (black) position in 2013: (a)

<sup>817 &</sup>lt;u>time series at different depths; (b) Profile at different dates.</u>



819

820 Fig. 9. Space-time dynamic of soil CO<sub>2</sub> concentrations, at the summit (red) and the footslope (black) position in 2013:





824 Fig. 10. Space-time dynamic of soil CO<sub>2</sub> fluxes, at the summit (red) and the footslope (black) position in 2013: (a) time

825 series at different depths; (b) Profile at different dates.



Mis en forme : Police :Calibri Mis en forme : Normal

827

828 Fig. 7. Depth distribution of the relative contribution to soilsurface CO<sub>2</sub> fluxes in year 2013 averaged by semi-seasons

829 (error bars represent the standard deviation of the time aggregation for each soil layer): (a) at the summit, and (b) at

830 the footslope position.