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Interactive comment on "Inorganic carbon fluxes across the vadose zone of planted and unplanted soil mesocosms" *by* E. M. Thaysen et al.

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We would like to thank the reviewers for their thorough comments on our manuscript. In the following, we address each reviewer comment separately and suggest changes to the manuscript to be implemented if given a chance for revision.

Reviewer 1 (C. Gough)

Specific comments, elaborating on above: 1) Mesocosm treatments are unbalanced and seem to have been haphazardly applied (Pg. 4257, Lns 18-24). Why was the harvest schedule of 1 through 3 different from 4 and 5? Why was the sample size so low?

REPLY: We recognize the statistical limitation of having only two replicate mesocosms

for most treatments, and it is mentioned in the MS on pg. 4258, l. 18. The sample size of two was considered sufficient due to the good agreement between the estimated and measured DIC percolation, documented for unplanted mesocosms in Thaysen et al. (2014), that proved high reliability of the mesocosm system and the applied sampling procedures. The variance in planted mesocosms was investigated using the first set of mesocosms, i.e. mes. 1-3. Here we allowed for an additional mesocosm, i.e. a total of three, due to the additional biological variation that the presence of plants was expected to exert. As we found that mesocosms were very similar (see figs. 2-4 in MS and supplementary material) we continued with only two planted mesocosms (mes. 4 and 5).

We further agree that an optimal data set would have contained two growth trials of the same length. The different harvest schedule resulted from the occurrence of nutrient deficiency in the plants in mes. 1-3 around day 50. For these mesocosms nutrients were applied to the soil prior to experiments and mesocosms were irrigated with milli-q water. In the second barley growth trial (mes. 4-5) this problem was successfully avoided using nutrient irrigation, allowing for continued investigation of inorganic C fluxes under barley growth. Recognizing the heterogeneity in our experiments we considered only presenting data up day 50 after sowing for both growth trials. However, we believe that the good agreement between mesocosms in the two growth trials (e.g. Fig. 4A, B and D), and the important exponential rise in the ER at prolonged growth (mes. 4 and 5) justifies the display of the full, heterogeneous data set.

Suggested in-text revision: Please see our answer to question 10 by reviewer 2.

2) Much of the text is superfluous (e.g., Pg 4256, Lns 5-15; Pg 4262-4265; Pg 4270, Lns 3-23); the document reads like a thesis reporting numerous details; it's great the authors are so detail oriented in places, but this detracts from a focus.

REPLY: We agree to remove the section on the impact of plants on CO2 fluxes in the introduction (please also see our answer to comment 2 by reviewer 2 for a shortened,

more structured introduction). We do not agree that the whole section on modeling methods is superfluous, nor the section on soil CO2 production and diffusivity in mesocosms. The modeling methods are important for understanding the achieved effort on this part. The soil diffusivity and CO2 production were directly used in our simulations. Our soil diffusivity studies further indicated a large difference in the diffusivity of planted and unplanted mesocosms, contributing to our current understanding of CO2 dynamics in the vadose zone of soil with different land use.

We suggest shortening the MS and increasing its focus by:

1. Moving our data on the temporal variation of the pCO2, alkalinity, VWC and temperature (previous Fig. 2) to the supplementary information. These "raw data" were used to calculate the DIC percolation, soil CO2 production and inorganic C speciation; the latter of which provided important results on the controls on vadose zone DIC in acidic soil. But they are in themselves not essential to for the focus of the study, i.e. the quantification of upward and downward transport of inorganic C. With removal of Fig. 2 section 3.1 can be omitted. To account for the accompanied loss of the detailed view on the inorganic C dynamics in the mesocosms, we suggest a revision of Fig. 3 to include one more day (see figure 1 at the end of this answer).

2. Omitting the cropland C balance sections, i.e. section 2.4, section 3.5 and pg.4275, l.19 to pg. 4276, l.7 in the discussion.

3. Mentioning the cumulative CO2 emissions to the atmosphere and groundwater from former section 3.5 in continuation of the descriptions of the results in Fig. 4 (inorganic C fluxes in and out of the mesocosm) and removing Fig. 5. The corresponding discussion section on page 4275 may be reduced by lns.7-10 to: "The cumulative DIC percolation flux was small relative to the cumulative Rs. Results from planted mesocosms (1.6–2.0%) were higher than the global emission flux partitioning (0.26%) (Raich and Potter, 1995; Kessler and Harvey, 2001) but lower than a 2.5% fraction reported for an onion field (Sawamoto et al., 2003). The relatively higher ratio between cumulative DIC

percolation flux and the cumulative Rs in unplanted mesocosms resulted from lower cumulative Rs in unplanted soil that was further decreased by periods of net uptake of CO2 (Fig. 4a). " In a revised Fig. 4C we intend to recalculate DIC percolation flux to the same units as the gaseous emissions (i.e. μmol CO2 m-2 s-1) to ease comparison of the magnitudes of the upward and downward CO2 emission (see figure 2 at the end of this answer).

3) The following primary conclusion is not supported (Pg 4253, lns 23-25): "Our results indicate no change of the cropland C balance under elevated atmospheric CO2 in a warmer future climate, in which plant biomass and soil 25 pCO2 are expected to increase." The experiments manipulated neither mesocosm climate nor CO2.

REPLY: We recognize that this conclusion was a little bit overstated and would delete it from a revised version. See our answer question 16, reviewer 2, for a suggestion to a revised conclusion.

4) Modeling results are scantly described. The model setup is well explained, but the justification for presenting simulation results for only a single mesocosm over a short duration is unclear (Pg. 4260-4261, Lns 25-8).

REPLY: We dedicated more than 1 page to a careful step-by-step description of the modeling results and underlying geochemistry in Fig. 6A-I so we do not quite agree with the statement that modeling results are scanty described. Due to the similarity between replicate mesocosms and the lengthiness of the MS we chose not to include results for another planted mesocosm, nor the unplanted mesocosms. Modeling of the post-harvest CO2 fluxes requires programming of a separate set of equations into the model. This work has been commenced but is not finished. The reviewer is right in his critics that the shorter duration for modeling constitutes a limitation to the MS. The reason we did not simulate for a longer period is because the watering scheme unfortunately was not noted down several occasions towards the end of the experiments, making modeling of the soil water content and connected CO2 fluxes very difficult.

Suggested in-text revision: Pg.4261, l.1: “Due to the similarity in the CO2 dynamics between replicate mesocosms (Figs. 1-3), only simulation results one mesocosm from the second barley growth experiment (days 15-56) are presented herein.”

5) The authors only dedicate a single paragraph to model interpretation, making its relevance and context uncertain.

REPLY: We agree that the model interpretation received comparably little attention in the MS. We suggest partly accounting for the imbalance between the interpretation of experimental and modeling studies by cutting down on the general length of the discussion (i.e. by leaving out sections on the cropland C balance and speculations on how it is affected by elevated CO2, as mentioned earlier), and partly by dedicating a little more text to the model interpretation, see below. Suggested revision of the modeling paragraph: “Modeling of the CO2 fluxes using different explanatory scenarios was a valuable tool for identifying the key processes behind the observed evolution in the dissolved CO2 fluxes. Scenario 1 (root nutrient uptake coupled to cation exchange) was in accordance with previous demonstrations of slight soil alkalinization when nitrate is the dominating inorganic N species (Marschner et al., 1991; Weligama et al., 2010). However, our modeling of the nutrient uptake remains somewhat uncertain due to lack of data for actual nutrient uptake. A control of Al(OH)3(a) on the soil solution was unlikely but other buffering processes such as the dissolution of small pieces of lime (Fig. S3) may be possible. Soil pCO2 and Rs were hardly affected by any buffering process, despite the low subsoil pH that caused portioning of DIC towards H2CO3* and perhaps some degassing to CO2(g) (Fig. 3 in discussion MS). This is in accordance with existing knowledge that mineral reactions affecting the carbonate system have only marginal effects on the pCO2 (Kuzyakov, 2006).”

Due to the critics raised by reviewer 2 in question 11, we further suggest to add a discussion of our model with regards to reality and the impact of some applied simplifications on our simulations:

“The modified SOILCO2 model made use of several simplifications of reality, one of them being the linkage of the gammas to the RMI without inclusion of a bulk soil microbial respiration. However, due to the dense root growth in the A horizon (see Table 3, Fig. 1 and section 4.1) plant root independent microbial respiration was probably negligible in the A horizon. The exponential decline in the microbial respiration with soil depth by the fs(z) (Eqs. 3-5) further implied that the gammas in the C horizon was small compared to the topsoil. Hence, for the experimental conditions of this study, the omission of a plant-root independent microbial respiration in our model seems justified."

6) Pg. 4266, Lns 7-18. I find the C budget methods unclear and therefore potentially problematic. Likewise, how was GPP estimated from above and belowground plant biomass unless autotrophic respiration was added back in somehow (since GPP = NPP + Ra)? Most importantly, I don’t understand why/how the net ecosystem C balance is calculated as the difference between NEE and aboveground biomass. Consider that annual NEE minus total (above and belowground) NPP equals heterotrophic respiration; I’m not certain how the terms the authors incorporated in their calculation derives a “net C balance” for the ecosystem.

REPLY: Measurement of the net ecosystem carbon balance (NECB) should involve the following components (Smith, Lanigan et al. 2010) :

NECB= NEP−D−F−H−VOC−CH4−E+I Eq. 1

where D= C loss to the ground water via dissolved inorganic carbon, dissolved organic carbon and particulate organic carbon, F= C loss to fires, H= C loss by harvest, VOC= C loss to volatile organic compounds , CH4= C loss to methane emission, E= C loss by erosion, I= C gain due to fertilization with manure, re-deposition of eroded sediments from elsewhere, and deposition of dissolved carbon in water.

Carbon losses by erosion, as volatile organic compounds, and due to microbial produced methane (CH4) may be neglected from the cropland C balance (E, VOC and

CH4 term in Eq.1) (Ceschia, Beziat et al. 2010). The same may be done for C gains by deposition of dissolved carbon in rain and fog (part of I term in Eq. 1) because the gains from the latter source are likely to be very small (Smith, Lanigan et al. 2010). Since there was no fire in our experiments, and no C-fertilizer was applied, the F and I term could further be omitted from Eq. 1, reducing it to:

NECB= NEP−D-H Eq. 2

Because NPP= GPP – plant respiration, and NPP ïĆż plant biomass (Hall, Scurlock et al. 1993), NEP= plant biomass – ecosystem respiration (ER). The NECB hence becomes: NECB= biomass-ER−D-H Eq. 3 Eq. 3 represents essentially what we wrote in the MS on pg. 4266, lines 13-19, considering we inserted NEE=GPP-ER into NECB=NEE– D-H+I (Kindler, Siemens et al. 2011), used the biomass as a proxy for GPP and set the I term in Eq. 1 to zero (see above). Hence, the two ways of approaching the calculation of the cropland C balance essentially lead to the same formula for calculation of the NECB, namely Eq. 3. However, we recognize that our approach was a bit more unconventional than usual practice, and the connected uncertainties were perhaps not described clearly enough, (e.g., taking the biomass as a proxy for the GPP is significant underestimation, as pointed out correctly by both reviewers). In addition, we calculated the NECB without leaching losses (D term) and merely mentioned this in a dependent clause (pg. 4266, line 14) which may confused the reader. We would however like to mention at this point that other publications of estimations of the NECB rely on subtraction of the heterotrophic respiration from the net primary production (NPP) (Tate, Scott et al. 2000), i.e. neglecting both the “D” and “H” term in Eq. 2 , which is even more simplistic than the here used approach.

Suggested revision: As we wrote in our answer to question 2 by reviewer 1 we will omit C balance calculations from a revised version of the MS.

Why was the C balance only calculated for mesocosms 4-5? What is meant by the following?: “Post-harvest CO2 fluxes from mesocosms 1–3 were considered, but marked

as indicative due to a shorter growth period.”

REPLY: The C balance was calculated for planted mesocosms 4-5 and unplanted mesocosms. We did not calculate the C balance for planted mesocosms 1-3 during growth because we were trying to get the best estimate of the yearly C balance, which is likely affected by the growth period of barley. Since the latter was longer in mesocosms 4-5 and therefore closer to field conditions, these mesocosms were most suitable for the estimation. The comment “Post-harvest CO2 fluxes from mesocosms 1–3 were considered, but marked as indicative due to a shorter growth period.” builds on the above. The point we wanted to make here was that the post-harvest fluxes in mesocosms 4-5 were probably greater than in mesocosms 1-3 as a longer growth period implies a higher root biomass that can stimulate microbial degradation after harvest.

Suggested revision: As we wrote in our answer to question 2 by reviewer 1 we will omit C balance calculations from a revised version of the MS.

7) Pg 4269, Ln 20-23. Again, I do not understand this measure of ecosystem C balance: “The net ecosystem C balance of the mesocosm system (i.e. the amount of C captured in total biomass minus the ER minus the harvested aboveground biomass, but excluding leaching losses) was 1 to -10.8 mol C m-2 during 78 days of growth and 60 days of post-harvest”. Why not simply use NEE, which is the C balance (GPP – ER)? And, are the units expressed per day?

REPLY: For a detailed explanation of the applied method please see our answer to question 6 above. We did not simply use NEE because the harvested biomass and the DIC loss to the groundwater constituted significant C losses in our experiment (H and D term, respectively, Eq. 1). In fact, the C balance was estimated to highlight the importance of the DIC flux for the total soil C loss. The units were expressed on a cumulative basis, i.e. for 78+60 days for planted mesocosms (see MS pg. 4269, line 23: “The net ecosystem C balance of the mesocosm system was 1 to -10.8 mol C m-2

during 78 days of growth and 60 days of post-harvest")

Suggested revision: Again, we will omit C balance calculations from a revised version of the MS.

8) Pg 4275, Lns 19-21. Previously in the results the net ecosystem C balance was expressed on a per day basis (I think), and here on a per year basis; yet, mesocosm fluxes were not measured for an entire year, so how were annual estimates derived? Also, why is there such a large range in net C balance among mesocosms? Also, the sign convention (negative for C gain by ecosystem) presented by the authors is not the same as that for European croplands. Pg. 4276-4277.

REPLY: In our answer to question 7 we clarify the time period that the data for net ecosystem carbon balance refers to. Our data account for one cycle of plant growing and harvest, which is general practice in temperate areas, and allows for a rough comparison with measured annual C balances. Hence, we did not up-scale from the growth cycle to a year. We realize that this should have been stated somewhere in the text.

Suggested in-text revision: As mentioned in our answer to question 2 we suggest removing the C balance sections from a revised version of the MS.

9)Pg. 4266, Lns 22-23 The authors say that "the selected data are representative for all replicate mesocosms". Then why not show means for each treatment?

REPLY: As stated in the MS on pg. 4258, lns 18-20, we gave ranges for our measurements (C budget figure) or displayed data for all mesocosms (e.g. Fig. 4) since there were only two replicates for most treatments, limiting a meaningful calculation of the means and standard variation. However, in the suggested shortening of the MS (see our answer to question 2 by reviewer 1) we agreed to display the means in Fig. 3 (Fig. 1 at the end of this answer) which is a schematic over the temporal variation of the inorganic C species in the different treatments. We think this is justifiable since all raw data used for the calculation are provided in Fig. 2 and Fig. S1 (now suggested Fig.

S1 and S2).

10)Pg. 4268, Ln 14. The authors should specify that negative NEE indicates uptake by mesocosm ecosystem.

REPLY: We agree with this. Suggested in-text revision (pg.4268, Ln 14): "The negative NEE, i.e. ecosystem CO2 uptake, increased with plant age . . ."

11)Pg 4272, Ln 12. "we have quantified the inorganic C dynamics". Why not drop "inorganic since biomass was also characterized?"

REPLY: The focus of the study is on quantifying and understanding inorganic C dynamics in the vadose zone. Dissolved organic C losses, and the soil organic C content were not monitored, and the above suggestion would hence be an overstatement. Sections on the cropland C balance will be omitted in a revised version of the MS and the focus on inorganic C dynamics should hence become clearer.

Reviewer 2 (Anonymous)

Specific comments: 1)The atmospheric residence time of CO2 occurs at many different scales from seconds to centuries or longer, but averages about 5 years as noted in a footnote. Footnotes are confusing, especially when the number 1 is in superscript after 'years', and would be better off incorporated into the text.

REPLY: We agree with reviewer 2 on that footnotes should be avoided. We find that the text in itself provides all needed information and suggest deleting the footnote. Hence, the suggested in-text revision is: "The residence time of DIC in groundwater is at least as long as the residence time of groundwater which may be in the order of hundreds to thousands of years (Kessler and Harvey, 2001). The atmospheric residence time of CO2 however may be as low as 5 years (Solomon et al., 2007; Archer and Brovkin, 2008), implying that even small changes in the C emission balance can have important effects on the atmospheric CO2 concentration (Schimel, 1995)."

2) The flow in the introduction can be made simpler. The first paragraph discusses the

global C cycle and atmospheric CO2, the second DIC, Rs and pCO2, the third DIC, the fourth pCO2, and the 5th diffusion. Note that pCO2 is the unifying concept for both DIC and Rs (at least its measurement as soil to atmosphere CO2 efflux). By discussing the importance of pCO2 first, the resulting fluxes follow. Such a reordering will also place paragraph 5, on the role of plants, in context; at the moment it is not well connected to the rest of the introduction section.

REPLY: We thank reviewer two for this excellent remark. Suggested revision of the second part of the introduction, starting from pg. 4254, l.24: "The soil partial pressure of CO2 (pCO2) and the soil CO2 efflux to the atmosphere, also referred to as the soil respiration (Rs), are a function of the combined CO2 production from microorganisms and roots, the soil gas diffusivity and a limited contribution from mineral reactions via the carbonate system (Kuzyakov, 2006; Trumbore, 2006). Soil temperature and moisture are the main abiotic factors controlling the biological production of CO2 (Schlesinger, 1973; Maier et al., 2011). Further factors such as the overall soil nutrient content, soil mineralogy, land use history and plant phenology also play an important role for the magnitude of the soil CO2 production (Lohila et al., 2003; Trumbore, 2006). Diffusion of CO2 in air is about 104 times faster than in water (Suarez and Šimůnek, 1993). Rain increases the Rs due to a stimulation of microbial respiration and/or due to displacement of CO2-rich soil air by advection (Lee, Nakane et al. 2002; Huxman, Cable et al. 2004). Frequent and heavy rain may eventually result in a high soil water content that lowers the soil gas diffusivity, leading to accumulation of CO2 in air-filled pores, i.e. higher pCO2 and a reduced Rs (Jassal et al., 2005; Zhang et al., 2010). Dissolved inorganic C is the sum of C in carbonic acid, H2CO3* (where H2CO3*= CO2(aq) + H2CO3), bicarbonate, HCO3-, and carbonate, CO32-. The concentration of DIC, [DIC], is closely linked to the pCO2 via Henry's law. In addition, the [DIC] depends on the soil solution chemistry because of the pH-dependent solubility of inorganic C species, and is as such largely influenced by processes that increase soil alkalinity, e.g. the weathering of carbonate and silicate minerals (Appelo and Postma, 2005; Walmsley et al., 2011). The DIC percolation flux to the groundwater can be described by multiply-

ing the [DIC] by the recharge flux (Appelo and Postma, 2005; Thaysen et al., 2014). Here, we measured upward and downward CO2 transport in both gas- and aqueous phases in unplanted and planted mesocosms to quantify the total CO2 emission from ecosystems. The mesocosms, established to simulate the top 0-80 cm soil profile of a barley cropland, were incubated under controlled environmental conditions, allowing a model-based investigation (HP1 module, Hydrus 1D) of the biogeochemical controls on CO2 production and transport in the soil profile prior to seeding, during growth and after harvest. Mesocosms have been shown to provide useful environments for conducting process-related research in unsaturated soil (Hendry et al., 2001; Thaysen et al., 2014). Reactive transport modelling may further increase the understanding of the coupled physical, chemical, and biological processes influencing CO2 transport within soils (Steefel et al., 2005). HP1 allows for the complex biogeochemical modelling of CO2 transport in the vadose zone by providing options for simulation of soil water content, root growth, root water and -solute uptake, as well as for gas diffusion and geochemical reactions of all possible chemical species, the latter being a novelty amongst vadose zone models. "

3)In paragraph 5, note also the important role of the flushing (advective transport) in the soil air space after rain. Many studies find an increase in Rs with rain, even in systems that are not water limited (e.g. Lee et al., 2002,http://onlinelibrary.wiley.com/doi/10.1046/j.1440-1703.2002.00498.x/full).

REPLY: Please see our suggestion for a revised introduction in our answer to comment 2) above.

4)On page 4256 line 22, are mesocosms only useful for studying unsaturated soils but not saturated soils? Please clarify in the context of Thaysen et al. (2014).

REPLY: Mesocosms were only tested for unsaturated zone processes (Thaysen et al. 2014) but are likely suited for process studies in saturated environments as well (e.g. wetlands, sediment studies). These kinds of perspectives were provided in the MS on

mesocosm construction (Thaysen et al. 2014). As the current MS deals with processes in the unsaturated zone, only, we see no need to mention at this point that mesocosms could function for other processes as well.

5)On page 4257 line 25 it is stated that frequency and amount of irrigation were adjusted to serve the need of plants while maintaining downward leaching. Could you please clarify? Is this more or less rain than a barley crop in Denmark commonly receives? Is it typical for rain events of this magnitude to occur this frequently (in the case of Denmark I would assume that this is probably the case). At any rate, please quantify the amount of water that the plants actually received instead of in the context of plant nutrient delivery (which I note should also be justified in the introduction: is this an experiment on plant and fertilization impacts on Rs and DIC, and if so why?)

REPLY: As stated, planted mesocosms were irrigated at 1-2 days intervals. Average irrigation amounts for the first barley growth treatment (mes. 1-3), the second barley growth treatment (mes. 4-5), the post-harvest treatment (mes. 1-3) and the unplanted treatment (mes. 6-7) were 5.3, 13.5, 4.6 and 4.4 mm d-1, respectively, but irrigation events up to 35 mm were applied during the experiments. This is more frequent and heavy rain than "typical" for natural rain events that account for most of the infiltration in Danish fields. The reason for the application of these high irrigation rates and frequencies was the high evapotranspiration rates in the mesocosms (>50% of the applied irrigation at all times in planted mesocosms, but typically 70-80%) which were caused by the $\sim$2-5 times higher plant biomass in mesocosms compared to the field situation, the relatively high soil temperature and by the high ventilation in the growth chamber. Regarding the plant nutrient delivery, this is not a study on fertilization impacts which is why it was not mentioned in neither introduction nor discussion. The reasoning behind the different fertilization regimes of the two barley growth trials was already given on pg. 4258, lns.7-9 in the discussion paper: "The different mode of fertilizer application in mesocosms 4–7 was implemented in order to avoid nutrient depletion of the soil during the longer duration of the experiment. " Nutrient depletion was observed at the end of

the first barley growth trial (mes.1-3) (see also our answer to question 1 by reviewer 1).

Suggested in-text revision to be inserted on pg. 4258, l.15: "Average irrigation amounts for the first barley growth, the second barley growth, the post-harvest and the unplanted treatments were 5.3, 13.5, 4.6 and 4.4 mm d-1, respectively. Irrigation amounts exceeded those in the typical field situation because soil temperatures and plant biomass in mesocosms were elevated, leading to higher evapotranspiration (see section 4.1)."

6)In section 2.2.2, did the transparent chamber impact the light environment in a meaningful way? In other words, was the magnitude of NEE decreased by a potential slight shading effect of the chambers?

REPLY: The chambers did not in themselves shade the plants since they were maintained clean and dry at any time. However, the large canopy size, exceeding the surface area of the mesocosm by approx. 4 times towards the end of the experiments, implied insertion of the shoots into the measurement chamber (as mentioned on pg. 4259, l. 16). Disturbance of the natural canopy shape by this action may have caused some additional mutual shading of the leaves that would lead to underestimation of the NEE. We suggest mentioning this in section 4.1 in the revised MS, see also our answer to question 15 below: "Some artificial increase in the ER and a decrease in the NEE can be expected to result from the application of the chamber for CO2 exchange measurements since the plant canopy covered a larger surface area than the surface area of the mesocosms (Fig. 1)."

7) In section 2.2.3, please clarify the meaning of 'Cells were removed when their inside pressure had increased to 1013 hPa.' What are 'cells' in this context?

REPLY: This sentence should not be read without the previous one, i.e. "Evacuated ZnS(Ag)-scintillation cells (V = 200 mL) equipped with a manometer were attached to the mesocosm gas sampling ports." Hence, in the mentioned sentence we are referring to the scintillation cells. In-text revision: "The scintillation cells were removed when their inside pressure had increased to 1013 hPa."

8) How is equation 1, an expression of Fick's Law, a simplified approach of Fierer et al.? Don't most flux-gradient methods for determining soil CO2 transport use a similar approach?

REPLY: The reviewer is right in the fact that Eq. 1 is an expression of Fick′s first law. In the MS it should have said "using a simplified approach of Fierer et al." since Fierer et al. modified the law as to account for a calculation of the CO2 production rate for each 20 cm interval of the soil in their work.

Suggested in-text revision (pg.4260, line 8): "... Resulting bulk diffusivities were then used in the modeling of the CO2 (Sect. 2.3), and to estimate the soil CO2 production rate, RCO2, from Fick′s first law of diffusion (e.g., Fierer et al. (2005))"

9)Why was Hydrus only used for mesocosms 4 and 5, and why are results for only mesocosm 5 presented? REPLY: Please see our answer to question 4 by reviewer 1.

Likewise, in section 2.4, why are CO2 budgets for only mesocosms 4 and 5 presented? Measurements exist to estimate CO2 budgets for the other mesocosms as far as I'm aware, aren't these reported on page 4275?

REPLY: For an answer on why the C balance was calculated for planted mesocosms 4 and 5 and not for mesocosms 1-3, please see our answer to question 5 by reviewer 1.

10)I also note that it's difficult for the reader to remember microcosms by their number rather than some abbreviation related to the treatment. The numbering scheme is arbitrary.

REPLY: We agree with this. Suggested in-text revision, Methodology, pg. 4257, l. 18-24: "The mesocosms were subjected to different treatments (Table 1). Two mesocosms remained unplanted (referred to as the unplanted treatment). These mesocosms were shown to exhibit low variability and high reliability (Thaysen et al. 2014). To investigate the additional variability introduced by the presence of plants, barley (Hordeum vulgare L. cv Anakin) was sown into three mesocosms. In these mesocosms CO2 fluxes were

investigated during growth (days 14 to 58 after sowing) and after harvest (days 58-117) (referred to as the barley growth treatment #1 and the post-harvest treatment, respectively). The agreement between mesocosms remained good (e.g. Fig. 4), allowing for a reduction to a sample size of two for planted mesocosms, also. In further two mesocosms the growth period of barley was extended to 100 days with monitoring up to 78 days after sowing (barley growth treatment #2). " Please also see our suggestion for a revised Table 1 in fig. 4 at the end of this response.

11)In equation 2, why are production from both soil microorganisms and plant roots multiplied by the plant root index? An explanation is given, but were other formulations tested? Might there be a plant root dependent and independent component of microbial respiration (i.e. S=gammas1 + (gammas2 + gammap)*RMI, where gammas1 is the root independent microbial contribution?)

REPLY: As the reviewer rightly points out, we gave an explanation for the multiplication of the CO2 production by soil microorganisms with the root index, namely priming. Basal microbial respiration (i.e. microbial respiration independent of plant exudates) is likely found outside the rhizosphere. In our mesocosms, the root mass was 3-4 larger than in the field, resulting in a very dense network of roots in the A horizon (see Fig. 1 and Table 3 in the MS), leaving very little chance for any patch of soil not being affected by exudates. Hence, the assumption that all of the microbial respiration was linked to the root mass was reasonable for the A horizon. For the C horizon we recognize that this assumption is somewhat simplistic. However, in our model (and in SOILCO2) microbial respiration is scaled with depth by an exponential function ("fs(z)", see pg. 4264, line 8, and Eqs. 3-5), that implements an exponential decline with depth in the soil (see fig. 3 at the end of this answer). From fig. 3 it is evident, that beyond 30 cm, fs(z) is small, regardless of the value of "a", reducing microbial respiration considerably. This implies that a separation of microbial respiration into plant root-dependent and -independent part in the C horizon would have had very little effect on the simulated pCO2 and Rs.

Suggested in-text revision, Discussion, pg.4277, l. 4: “The modified SOILCO2 model made use of several simplifications of reality, one of them being the linkage of the gammas to the RMI without inclusion of a bulk soil microbial respiration. However, due to the dense root growth in the A horizon (see Table 3, Fig. 1 and section 4.1) plant root independent microbial respiration was probably negligible in the A horizon. The exponential decline in the microbial respiration with soil depth by the fs(z) (Eqs. 3-5) further implied that the gammas in the C horizon was small compared to the topsoil. Hence, for the experimental conditions of this study, the omission of a plant-root independent microbial respiration in our model seems justified."

12) In equation 5, how was r (root growth rate) simulated? Also, note that both dots and the multiplication sign are used in different equations. Please use the multiplication sign for accuracy and consistency.

REPLY: We think the reviewer means Eq. 6 here as Eq. 5 does not contain the parameter “r”. On pg.4264, lns.13-17, we provide the information he asks for: “For our simulations, Rinit and r were set to 2 g DW and $2.4\times10-6$ g $s-1$, respectively, as calculated from the measured root biomass in a similar mesocosm experiment (13.7 g, Table 3) at 65 days after plant emergence (70 days after sowing), under assumption of linear root growth.”

Hence the calculation for the root growth rate, r: 13.7g/65 days =0.21 g day-1 0.21g day-1/ 86400 seconds day-1 = $2.4\times10-6$ g $s-1$ Because we assumed linear root growth with time, the root growth rate was constant.

Suggested in-text revision: “For our simulations, Rinit and r were fixed at 2 g DW and $2.4\times10-6$ g $s-1$, respectively. Assuming linear root growth, the root growth rate was calculated by dividing the measured root biomass at the end of a similar mesocosm experiment (13.7 g at 70 days after sowing and 64.5 days after plant emergence, Table 3) with 64.5 days of plant growth. The Rinit was then calculated from multiplication of the r with the number of days after plant emergence at simulation time zero (i.e. 15

minus 5.5 days).”

In the revised MS we will of course also change all dots to multiplication signs (Eq. 1 and 6).

13) On page 4266 how was GPP estimated using above and belowground vegetation biomass as proxies if autotrophic respiration is not estimated?

REPLY: Please see our answer to question 6 by reviewer 1.

14)Please quantify ‘fairly stable’ on page 4267.

REPLY: In the sentence following our description “fairly stable” we elaborate on what this means, i.e. “The topsoil and subsoil had VWCs in the ranges 20–27 % and 7–15 %, respectively, except for the mesocosm bottom (73–76 cm) where near saturation VWCs 10 of around 27 % were caused by water logging.”

Suggested revision: As we intend to remove Fig. 1 incl. VWC profiles from the MS if given a chance for revision, this whole section will be removed.

15)Section 4.1 is largely a collection of facts and is difficult to read; there is quite a bit to digest.

REPLY: We agree with reviewer 2 on that section 4.1 is a bit difficult to read. As mentioned in our answer to question 2 by reviewer 1 we suggest shortening the MS considerably by omitting, amongst others, Fig. 2. Hence, a part of section 4.1 may be omitted as well, see our suggestion: “4.1 Soil respiration: gaseous CO2 efflux The Rs and the pCO2 in unplanted mesocosms were generally in agreement with field studies on a range of different arable soils (Table 2a). In barley mesocosms, the ER and the pCO2 were generally higher than in published field studies but were in accordance with pot-grown barley in humic clay (Simojoki et al., 1991) (Table 2b). Higher respiration rates in mesocosms than in the field were probably a function of a larger plant biomass, as also observed by Simojoki et al. (1991). The aboveground- and root biomass was 3–4 times and 2–5 times higher, respectively, than the values reported from field studies

(Barraclough and Leigh, 1984; Xu and Zuma, 1992; Malhi and Gill, 2002; Lohila et al., 2003; Walmsley et al., 2011). Additional increase in the ER may have been caused by 3 to 5 ïĆřC higher day- and night time temperatures in the growth room than in the field (e.g. Kotroczo et al., 2008). Some artificial increase in the ER and a decrease in the NEE can be expected to result from the application of the chamber for CO2 exchange measurements since the plant canopy covered a larger surface area than the surface area of the mesocosms. The soil CO2 efflux to the atmosphere was about one order of magnitude higher in planted mesocosms than in unplanted mesocosms at peak time (Figs. 4a). As previously shown by Lee (1997) this revealed a strong impact of vegetation on CO2 dynamics in the unsaturated zone. Post-harvest Rs were higher than from unplanted soil, indicating a stimulation of microbial respiration by root-derived substrates (Kuzyakov, 2002). Respiration rates after harvest were within the range of previously reported Rs from sandy soil (0.5 μmol C m$-$2 s$-$1) (Heitkamp et al., 2012) and silty clay loam (1–11 μmol C m$-$2 s$-$1) (Moyano et al., 2007). The relatively high postharvest Rs and its rapid decline are in accordance with the high root biomass in mesocosms combined with a fast depletion of labile C from sandy soil (Heitkamp et al., 2012)."

16)It is unclear to me how the authors reach the conclusion that increased atmospheric pCO2 will not change the net C balance of croplands given the experimental treatment of barley planted in mesocosms. Elevated CO2 may increase plant growth, and root exudation and subsequent C losses may perfectly balance this enhanced C storage, or they may not. C storage in soil organic matter under different atmospheric pCO2 treatments was not explored, so any statement with respect to cropland C balance is premature. See also statements on the last paragraph of page 4277: these also need to be changed to reflect the findings of the study.

REPLY: We agree that our statements regarding the change in the C balance and emission portioning under eCO2 are overstated. Also the statement "In regions of high precipitation after crop harvest climate change may be mitigated by an increase in the

DIC percolation flux" was a little premature. Please see our suggestions for revisions below.

Suggested in-text revisions: Conclusion:

"The DIC percolation flux of $\sim$5 mmol C m-2 d-1 during barley growth was $\sim$1.6-2.0% of the Rs at increased plant biomass and elevated soil pCO2 compared to the field situation. After harvest, the magnitude of the DIC percolation flux was lowered to $\sim$2.5 mmol C m-2 d-1 but the importance of DIC percolation flux for the overall cropland CO2 emission increased to $\sim$6-7% of the Rs. At constant conditions of temperature and water content, the Rs was controlled by the production and diffusivity of CO2 in the soil, both of which were increased by plant growth. The DIC percolation flux was primarily controlled by the recharge flux and the pCO2 due to the low soil pH in the acidic soil of our study. Modelling suggested that nutrient buffering during root nitrate uptake dominated any mineral control on the soil solution in planted mesocosms. This study showed that the integration of experimental and modelling work is a powerful tool in advancing process-understanding of CO2 fluxes in the vadose zone. Our findings are important for improving our base understanding of CO2 partitioning in the vadose zone and may be included in the optimization of climate models. Further research is needed to outline the effect of different crops and soil amendments on the CO2 emission portioning of croplands.

Abstract, second paragraph:

"The average CO2 effluxes to the atmosphere from unplanted and planted mesocosm ecosystems during 78 days of experiment were 0.1$\pm$0.07 and 4.9$\pm$0.07 μmol carbon (C) m$-$2 s$-$1, respectively, and largely exceeded the corresponding DIC percolation fluxes of 0.01$\pm$0.004 and 0.06$\pm$ 0.03 μmol C m$-$2 s$-$1. Postharvest soil respiration (Rs) was only 10% of the Rs during plant growth, while the post-harvest DIC percolation flux was more than one third of the flux during growth. The Rs was controlled by production and diffusivity of CO2 in the soil. The DIC percolation flux was controlled

by the pCO2 and the drainage flux due to low solution pH. Modeling suggested that increased soil alkalinity during plant growth was due to nutrient buffering during root nitrate uptake."

17)Figure 7 is confusing. What do the numbers in the legend mean?

REPLY: The heading of the legend is "Time (days)", so the numbers indicate the number of days of the experiment.

Suggested correction of figure text: "Fig. 7. Measured (full lines) and simulated (dashed lines) temporal variation (expressed as days after sowing) of soil air pCO2 (A), alkalinity (B), pH (C), Al3+ (D), CaX2 (E), AlX3 (F), Ca2+ (G), and Ca2+ root uptake (H) in barley mesocosm 5. Geochemical reactions in simulations were described by root nutrient uptake and cation exchange (scenario 1)." For a suggested correction of the figure legend please see figure 5 at the end of this answer.

18)In Figure 8, can the large spikes resulting from irrigation and CO2 displacement be validated using measurements?

REPLY: Unfortunately we have only the measurements we present in the paper. It is possible, that some of the large spikes originate from CO2 displacement during irrigation. However, after handing-in this paper we discovered that there was a time-step dependency on the simulated Rs: At lower time step, the large spikes decreased in size. Hence, the spikes are numerical noise arising from non-complete fulfillment of the von Neumann stability criteria by the numerical solution.

Suggested revision of the legend in Fig. 8: "Measured and modeled time course of soil respiration (Rs) (A) and cumulative DIC percolation flux in barley mesocosm 5 modeled with root nutrient uptake and cation exchange (B). Small fluctuations in the simulated Rs around the baseline arise from diurnal temperature variations. Large fluctuations are numerical noise caused by the fact that the numerical solution does not fully obey the von Neumann stability criteria (Šimůnek, Jacques et al. 2006)."

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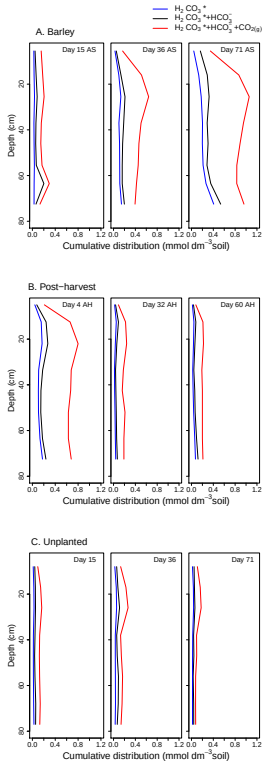

Fig. 1. Distribution of inorganic C between DIC species and CO2(g)

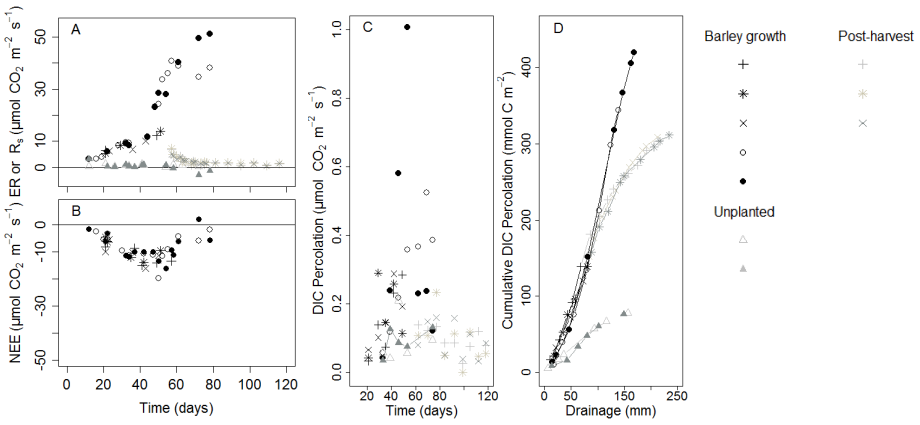

Fig. 2. Gaseous and dissolved CO2 fluxes from mesocosms.

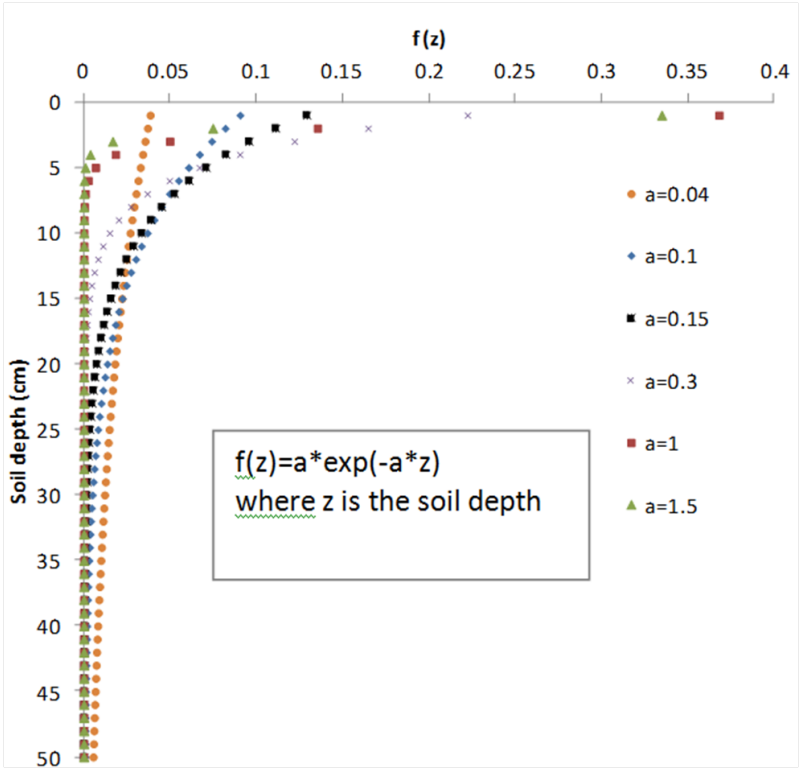

Fig. 3. Depth reduction of the microbial respiration at different values for the "a" parameter (see equation). A value of 0.15 cm was chosen in our study

Treatment	Duration of experiment (days)	Fertilization
Barley growth #1	58	Addition to soil prior to experiments
Barley growth #2	78	Nutrient irrigation
Post-harvest (post growth #1)	60	Addition to soil prior to experiments
Unplanted	78	Nutrient irrigation

Fig. 4. Overview over mesocosm experiments. The post-harvest experiment was the consecutive of barley growth experiment #1.

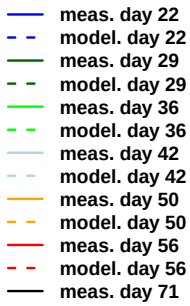

Fig. 5. Suggested revised legend for modelling figure.