Response to Review comments (original comments in bold with responses following)

<u>General comments RC2</u>

1) The authors use long-term solute and hydrologic datasets in conjunction with a modeling framework to explore the dynamics of DOC and DIC release and transport in a high-latitude catchment.

We thank this reviewer for thoughtful comments that have helped us clarify the scope and presentation of this study.

2) The writing here is often vague and should be greatly edited to improve clarity and concision.

While I believe that some of the findings have very broad implications, the authors seem to have written this article with a very narrow and specific audience in mind. The general structure of the manuscript lacks traditional method/results/discussion sections. And while this may be a stylistic criticism, I feel that a reorganization of the text – so that a distinct methods, results, and more expansive discussion sections – would greatly improve the flow and readability of the manuscript.

We have expanded the introduction and discussion towards broader applications to demonstrate that this work has significance outside of permafrost environments (per the reviewer's recommendation). Further, we have broken the presentation up into traditional method/result/discussion sections to improve the readability of the manuscript.

3) This manuscript obviously builds off of the prior work published by Lyon et al. (2010). However, the manuscript could be improved by more clearly articulating how this approach presents a novel contribution or conceptual advancement based beyond this prior study. In the Introduction, the authors should be more clearly define what the Lagrangian approach is (Page 192) and what are its advantages (beyond flexibility) compared to alternative or previously published approaches.

The difference and novelty of this current approach over Lyon et al. (2010) is mainly that the work in Lyon et al. (2010) was conceived in a static sense with regards to *r,* with observation data from only one year. In this study we add the inter-annual variability of *r* by analyzing long term observations of carbon and can, thus, better explore the dynamics behind the fluctuations of solute export over time in response to altered flow conditions. We have added this articulation to the revised text.

4) The authors often invoke permafrost as possible control on the temporal and spatial dynamics observed and modeled in this study. However, there is no evidence or citation presented to support the presence/existence of permafrost in the study catchment. The authors cite Ridefelt et al. (2008) to acknowledge a "probable presence of permafrost" at higher elevations. Moreover, modeling framework is not designed or parameterized to account for changing soil thermal dynamics, permafrost configurations, or C pool sizes. It just happens that the study was conducted in a discontinuous permafrost region – but permafrost has very little to do with this study – at least as presented.

We acknowledge the lack of references on the status of permafrost conditions in the catchment. This is primarily due to a lack of direct observations. To better characterize the permafrost state in the catchment and region, we have introduced references to Brown et al. (1998) to account for the permafrost classification of the region. In addition, for the Abiskojokken catchment, permafrost presence (and change) has been documented through recession flow analysis by Lyon et al. (2009) and also regionally in northern Swedish catchments (Sjöberg et al., 2013).

Further, it is correct that the model framework presented is not designed to account for changes in the permafrost relevant parameters (such as soil thermal dynamics or permafrost configurations) in and of itself. This does not negate the model's mechanistically appropriate description of current conditions that allow it to form a basis for more detailed investigations. The characteristics assessed at the catchment scale and the solute transport framework presented in the current study can be coupled to models capable of representing permafrost-groundwater dynamics (e.g. Frampton et al. (2011) used to study evolution of flow pathways in varying permafrost conditions). This is precisely the advantage and benefit of such a solute transport framework. We have expanded to text to better reflect this connection between the framework presented and the specific case of permafrost.

5) I recommend including a table, similar to Table 1 in Lyon et al. (2010) that summarizes model parameters, values, data sources, etc. Also, it would useful to include a conceptual diagram that illustrates the stream tube and how the different parameters are influence solute concentration and export.

A table and two figures that summarize the parameters for the model as well as the observations have been added.

6) I also recommend expanding the discussion to put these findings in a much broader context, particularly in relation to C cycling at high-latitudes. Recent publications by Suzanne Tank in Global Biogeochemical Cycles explore landscape and source controls on DIC and DOC flux in arctic rivers. How might these findings presented here scale to larger catchments or to the region? What are the implications for DOC and DIC release under projected warming or future hydrologic conditions? Is there any evidence that permafrost was present at the beginning of the data record, but has since thawed?

The discussion has been changed, including the addition of references to Tank et al. (2012) and Olefeldt et al. (2012), to put the work in relation to other Arctic rivers. Thawing rates of permafrost in the Abiskojokken catchment has previously been studied by Lyon et al. (2009) and these have been introduced (per previous comment) into the text. This thawing over the past century highlights the need for dynamic modeling frameworks capable of representing flux changes across catchments and demonstrates the potential benefits of the modeling approach presented here over previous considerations in Lyon et al. (2010).

The following new text has been added into the discussion:

Across six large arctic catchments Tank et al. (2012) further found that annual DIC mass flux is positively correlated with discharge and with the presence of carbonate rock. DIC loading from the sub-arctic landscape at the scale of the Abiskojokken catchment, as such, appears to be associated with the diffuse and relatively slow DIC release from rather ubiquitous weathering across the entire catchment. Release time of DOC is likely to be enhanced under climatic warming, due to prolonged growing season and/or enhance breaking down of organic material. Release times of DIC from the ubiquitous weathering are not likely to change due to climate warming but transformation from organic to inorganic carbon has been found to increase with contact time with mineralogenic sediments (Kawahigashi et al., 2006).

The ubiquitous weathering interpretation of DIC results is consistent with recent work relating the easily-weatherable carbonaceous bedrock (specifically Ca/Na ratio) with DIC in streams across spatial scales for this region (Giesler et al., 2013). Furthermore, the interpretation of high DIC mass flux correlation with discharge is coherent with that wet years result in relatively greater increase in DIC export than in DOC export (Olefeldt et al., 2012). As such, the large average dissolution time scale (1/k >> average τ) relevant for weathering and its byproducts (i.e. for DIC) makes the mechanistic solution component Eq. (3b) predominant. The DIC concentration is then controlled at a similar stable level, as determined by average $k\tau$, throughout most or all the flow and transport pathways (stream tubes) through the catchment, as described in section 2.3. Not only k but also average τ are to large degree independent of the temporal fluctuations of discharge into the stream, because the whole flow and transport process through the soil-groundwater system to the stream, which determines average τ, is not much affected by (or correlated with) the most downstream discharge ($\overline{q_s}$) and its temporal fluctuations precisely at the stream interface.

Specific comments RC2

1) Page 191, Line 1 – Omit "are believed to"

This change has been made in the text

2) Page 191, Lines 3-4 – Should cite Harden et al. 1999 (Science) to support long-term C sink statement.

We could not find the Science paper that you refer, presumably you mean this paper:

Harden, J. W., J. M. Sharpe, W. J. Parton, D. S. Ojima, T. L. Fries, T. G. Huntington, and S. M. Dabney (1999), Dynamic replacement and loss of soil carbon on eroding cropland, Global Biogeochem. Cycles, 13(4), 885–901, doi:10.1029/1999GB900061.

Our statement refers to long-term in the geological time-scale, however Harden et al. (1999) is a good reference to put this work in a broader context relating the work to carbon fluxes in agricultural land, so we have referred to Harding et al., (1999) in the Introduction.

3) Page 191, Line 26 – Omit "While these phenomena can be described in such a straightforward manner"

This change has been made in the text

4) Page 192, Line 15 – Omit parentheses around "Lyon et al."

This change has been made in the text

5) Page 192, Line 25 – While seasonal frost is a definite, it's not clear that permafrost is present in the study catchment based on the evidence cited by the authors.

See answer for question no 4) in General comments

6) Page 192, Line 25-26 – Sentence stating, "we further link previous modeling components" – this is quite vague? What modeling components? How were the distinct and how are they now going to be coupled?

The text has been revised to clarify that the previous modeling components that are referred to are the characteristic travel time and release time.

7) Page 194, Lines 1-5 – Should provide genus and species for dominant vegetation types.

This change has been made in the text

8) Page 195, Lines 5-6 – The "arbitrary stream tube" needs to be more clearly defined and its function explained.

We have now added the two new Figures 2-3 that explicitly illustrate the flow and transport pathways (streamlines, trajectories), around and along which infinitesimal stream tubes are defined, and also added the following explanation directly after the sentence introducing equation (1), where the term stream tube is first used:

"The term stream tube is used to describe mass transport through an infinitesimal three-dimensional space centered around and along each of the hydrological pathways (streamlines, trajectories) of water flow and solute transport illustrated in Figs. 2-3. The terms stream tube, streamline, trajectory, hydrological pathway are defined and used similarly as in earlier work considering transport in steady groundwater flow by Cvetkovic and Dagan (1994), through linked unsaturated soil water-saturated groundwater flow system by Destouni and Graham (1995), through linked groundwater-stream system by Lindgren et al. (2004), and through multiple hydrologically and/or hydrochemically different parts of a whole catchment by Cvetkovic et al. (2012), to which we refer the reader for further mechanistic and physical explanation details."

9) Page 195, Line 22 – "DIC in solid, soil or aquifer" confusing as written. What is the difference here between solid and soil?

Solid is defined as the bedrock and soil is the unconsolidated material consisting of quaternary deposits. Solid and soil has been replaced by bedrock and unconsolidated material throughout the manuscript to avoid confusion.

10) Page 197, Lines 16-18 – Please describe further the "ensembles" discussed here. How does one stream tube differ from another in terms of parameters, etc.?

The ensemble of stream tube is referring to the collection of stream tubes with different lengths due to depth of flow paths and the distance to the stream. This should become clear with suggested figure (General comments no. 5) showing the conceptual model.

11) Page 198, Line 8 – Replace "exemplify" with better word choice

"Exemplify" has been replaced by "test"

12) Page 198, Line 18 – "under various conditions of relevance" – needs clarification and further explanation. Vague as written.

We have now extended the explanation so that it should clarify what "under various conditions of relevance" means. As follows:

"… Eqs. (3)–(5) look like under various conditions of relevance i.e. for DOC release and transport for a single stream tube with a single travel time τ = 1 yr and for an ensemble of stream tubes with variable (uniformly distributed) τ , or average τ = 1 yr ."

13) Page 201, Line 20, 22 – Typically, significance is evaluated at P< 0.05. If you're going to consider trends significant at P>0.05, this should be stated upfront in the methods section.

The methodology of the statistical analysis has been moved to the Method subsection *2.2 Observations and Analysis*. Further, in the Results section 3.2.1 Time series analysis of observed data p has been put in relation to α as follows The total annual water discharge: ($p = 0.078$; $p < \alpha$) and the annual flow-weighted average DIC concentration: ($p = 0.060$; $p < \alpha$).

14) In general, the section and subsection titles in section 5 are pretty vague and could be improved. For instance "Relating observed fluctuations to model implications" could be restated as "Relationship between observed and modeled C dynamics"

This change has been made in the text. Also, the Result subsection "*3.1 DOC and DIC release and transport"* has been changed to "3.1 *DOC and DIC release and transport dynamics"*

15) Fig 2 and 3 – Add DOC or DIC to y-axis labels

This change has been made in the Figures 2 and 3.

16) Fig A1 and A2 are replicates of each other. Figure A2 should show relationship between alkalinity and DIC.

This change has been made in the Appendix