

## Replies to Reviewer 1

General comments:

This paper appears underdeveloped in terms of biogeochemical background. Essentially it lacks of enough information to understand why authors draw up the hypothesis (H) currently stated in the paper. For example, the H1 that authors propose (regarding the implications of salinization across variable land use) could be discussed and re-considered. Otherwise, they need to argue it properly in the introduction. Something similar happens in relation to H2. As H2 currently states I cannot understand why authors suggest it.

**Reply:** We have now added two more paragraphs in introduction section to explain why we draw up our hypotheses (H) stated in the paper regarding effect across and land use, coupled biogeochemical cycles, and difference between sediment and soils.

**Second Reply:** polished and make it more organized. Now, we have provided enough information in introduction section on salinization and urbanization (H1), coupled biogeochemical cycles (H2), and difference in soil and sediment (H3).

Another failing point in this paper is that the results- almost the whole section- lack of statistical support which reduces the accuracy of the outcomes. For example, the effects of salinization at different levels and the influence of land use on salinization effects (key aspects in the study) are not supported by any statistical analyses. Also, pre- and post- snow stream water characteristics are not statistically compared (there is actually no error in the graphs showing these results; Fig.8- or at least, I cannot see it). If they did, statistical analyses should be better explained in the method section, properly linked with the research questions authors want to deal and then used to support their results (including figures).

**Reply:** We have now conducted much better statistical analysis to support our hypothesis. We used t-test and linear regression to support the effects of salinization at different levels, and use linear regression to examine the influence of land use on salinization effects. The part of pre- and post- snow stream water characteristics has been removed for the reason that will be given below.

**Second Reply:** polished and make it clearer. 4 types of analysis has been done to test our hypothesis: 1) the effects of salinization at different levels (linear regression plus t-test), 2) influence of land use on salinization effects (linear regression vs ISC), 3) difference between sediment and soil (t-test), and 4) coupling of N, P and P with DOC (correlation analysis).

On the other hand, certain level of disagreement between the salinization experiment and field observations can diminish the impact of the results. Field sampling are restricted to one sample occasion during pre and post snow, this last two days after snow smelt; the same duration for their experimental salinization in lab. Why did authors decide two days? Salinity impacts can take place at different moments; depending on the system (i.e. historical exposure). Also - as authors well recognize in the discussion pure NaCl used in lab and salts employed for deicers can have different effects, which ultimately weaken the use experimental observations to interpret ambient changes. I think the findings, while interesting, are incremental and do not lead to a better overall understanding of the problem of salinization in urban areas.

**Reply:** Because disagreement between the salinization experiment and field observations can diminish the impact of the results, we have removed this part of field observations.

**Second Reply:** These data have been removed.

Finally, authors should be consistent when reporting nutrient and elements (N, nitrate, NH<sub>4</sub>, ammonium, SRP, P, S, etc.). There is a general mixture throughout the paper that authors should avoid.

Reply: We have been consistent throughout the paper when we reporting nutrient and elements. When we report elements, we also reported forms of these elements with them.

Second Reply: Check again throughout the paper to differentiate elements and their forms. In method section (2.4 Chemical Analyses), we make it clear that nitrate, TDN, SRP, sulfate, DOC, DIC, are all the forms of bioactive elements.

Specific comments:

Title As title currently states seems authors also evaluated an effect on "terrestrial soils". The word "soils" sounds much more linked to terrestrial ecosystems rather than aquatic systems to me. Since authors ultimately study riparian soils, I recommended them just writing Salinization alters fluxes of bioactive elements from stream ecosystems across land use.

Reply: We have changed the title to "Salinization alters fluxes of bioactive elements from stream ecosystems across land use" as suggested.

Second Reply: Done last time.

### **Abstract**

Authors should incorporate in the aim that they evaluated the implications of % land use on salinization effects. If possible, a short sentence describing how salinity interact the way C and nutrients are processes would be appreciate to reinforce the justification of their study.

Reply: We have incorporated in the aim that we "evaluated the implications of percent urban land use on salinization". Before the statement of objectives, we've added a short sentence describing how salinity interact C, N and P biogeochemical cycles to reinforce the justification of our study.

Second Reply: Done last time.

Line 4: I would suggest to reword the sentence "The effects.....understood" to Although increased salinization has been shown to alter C and N dynamics in freshwater ecosystems, its effects on biogeochemical cycles are still not well understood.

Reply: We have reworded this sentence as suggested.

Second Reply: Done last time.

- Line 18. Authors should firstly say that the response to salinization varied between instream sediments and riparian soils. And then, they can explain that such differences could be attributed to organic matter.

Reply: We have now added a sentence to report differences in responses to salinization between in-stream sediments and riparian soils.

Second Reply: Done last time.

-Line 20: Authors say: "Results of the .....after a snow even". I would move this sentence to the part where authors describe their objectives.

Reply: We agree with the reviewer that this sentence should now be moved to the part where authors describe their objectives. In this version, this sentence is now deleted because we have deleted the whole section pertaining to field observations (for the reasons that we mentioned earlier).

Second Reply: Removed.

Section 3.6. Fig 5a and b should actually be Fig 8a and b

Reply: We have now changed Fig 5a and b Section 3.6. to Fig 8a and b. Because the field observation section has now been removed, this sentence is deleted.

Second Reply: Done last time.

## Introduction

Overall, introduction lacks of detailed mechanisms (chemical/ microbial) in which salinization would affect C and N fluxes. In the discussion they provide plenty of detailed information about mechanisms and author could recast some I would strongly recommend authors to describe with more detail the biogeochemical effects of increased salt on fresh water ecosystems. Salt (mainly Na and Cl) can chemically affect N and C through its effects on ion exchange but also through stress of microorganisms responsible of DOM and N cycling. There are plenty literature in Australian ecosystems evaluating the effects of secondary salinization (anthropogenic salinization; e.g. those from Nielsen et al.) - like the one that authors investigate in the present study. Authors may like to include in the introduction references (an idea): \* Nielsen et al. (2003), \*\*Kulp et al. (2007), \*\*\*Ardón et al. (2013) (See references at the end of my revision)

Reply: We have now added two sentences reviewing chemical/ microbial mechanisms in which salinization would affect fluxes of bioreactive elements. The three references recommended by the reviewer are also now included in the paper.

Second Reply: Done last time.

Line 7: Authors should include in their objectives the influence of % land use as a secondary aim (or even within the primary one since they have an Ho around land use implications).

Reply: We have now rewritten this sentence and include the influence of % land use as a main aim.

Second Reply: Done last time.

Line 10: What do authors base on to formulate Ho (1)? Could sediments and soils from rural or natural watersheds are more sensitive to salinization than those from urban areas where microorganisms could be already acclimated to live under salty conditions? In others words, could the historical exposure to salinization make less sensitive urban rivers than rural ones which rarely experience such a pressure? This alternative Ho sounds more reasonable to me from a microbial perspective. If authors hypothesize the (1) as it currently states in the paper, then they need strong background supporting it. Regarding Ho(2): is that an hypothesis? it sounds really ambiguous and a priori difficult to test. Also, there is no previous supporting information in the introduction to understand why authors present such Ho(2).

Reply: We agree with the reviewer that sediments and soils from urban watersheds may be less sensitive to salinization because of historical exposure. One the other hand, the opposite may occur because of more labile organic matter in the substrates of urban streams. So, we've changed Ho (1) to "1) the effect of salinization on soil leaching and sediment retention/release of bioreactive elements change with watershed urbanization". We've reworded the Ho(2) to make it clearer to understand - "retention/release of nitrogen, phosphorus, and sulfur in response to salinization can be abiotically and/or biologically coupled with carbon biogeochemistry." We have now added two paragraphs in introduction section to explain why we draw up the two hypotheses.

Second Reply: These questions have also mentioned in general comments. Done in last time. We also add a third hypothesis: "salinization effects on release/transformation of bioreactive elements vary between stream sediments and riparian soils". Otherwise, the story would not complete.

I would essentially recommend authors to re-consider their hypothesis and re-organize the last part of introduction (Line 7- 18) as following: i) Main aims ii) how authors approach their aims: describing basically (and shortly) their experimental and field approach iii) Main hypothesis (well reasoned in the introduction). iv) If possible, main predictions. Based on their hypothesis authors can predict some outcomes that they can rest in their experimental. For me, a hypothesis should explain observed facts. Here authors do not explain anything but rather are

simply tentative statements of what one hopes the research will show For example, can authors provide a key hypothesis of how salinity affect retention or release on stream bioreactive elements? For example, salinity may lead osmotic stress on microbial communities involve in NO<sub>3</sub> and NH<sub>4</sub> transformation (denitrification, nitrification, DNRA). According to that I would expect in my experiment significant changes in inorganic N concentrations as salinity increases.

Reply: We've followed the recommendation of the reviewer to re-organize this paragraph as: i) main aims ii) how we approach our aims, iii) main hypothesis. Because major predictions are hard to make, part IV is not present. We have now added two paragraphs in introduction section to explain why we draw up the two hypotheses.

Second Reply: We have added main predictions as reviewer suggested.

Yet data analysis and statistic should be clarified. Authors should better link their statistical analysis with their research questions. Also, was the 1-way ANOVA done per study site and type of habitat (in-stream sediments vs. riparian soils)? Please, clarify it. How did authors test that fluxes in urban watersheds are more sensitive to increased salinization than in rural areas? Did authors use Spearman correlations to deal with that?. Correlations do not involve cause-effect. Authors should either conduct linear models with % land use or include land use as a factor in the ANOVA analyses. Regression approach may be more appropriate in this case since authors have n=1 for both forest and agricultural sites.

Reply: All information mentioned above has added to Section of Data Analyses and Statistics  
Salinization effects: We performed linear regressions of sediment/soil biogeochemical fluxes with salinity across all sites, using data from 6 salinization experimental manipulations (3 salinity levels with duplicates). If the p-value was < 0.05 for the regression, we assumed that there was a significant salinization effect. Otherwise, differences between two salinization levels were tested using a t-test of two-samples assuming equal variances. Type of habitat (in-stream sediments vs. riparian soils): Differences in ash free dry weight, fluxes or salinization effect between sediments and soils were tested using t-test two-sample assuming equal variances. Urbanization effect: We calculate the slope of sediment fluxes (C, N, P and S), and regressed the slopes with impervious surface cover – an index of watershed urbanization.

Second Reply: These questions regarding statistical methods have been raised in general comments. We have polished this paragraph and make it clearer.

Did authors test the normality of data distribution? Please, specify it.

Reply: We've now clarified in the last sentence regarding normality of data distribution that "For linear relationships, Spearman's correlation was used in cases where assumptions of normality were not met".

Second Reply: Done last time.

I also drop here some comments that authors may want clarify in the methods:

2.2 Sample collection and processing: how many cm did you sample for surface sediments and top soils? How did you collect sediments and soils? did you use a core, shovel? Please, specify it. How many replicates of stream water authors collected to compare pre and post snow?

Reply: We have added more detail information in the methods section regarding sample for surface sediments and top soils. The field comparisons between pre and post snow have been removed based on poor experimental design (as discussed earlier).

Second Reply: Done last time.

2.3 Laboratory salinization: what about concentrations of Na?

Reply: Because pure NaCl was used, concentrations of Na should be the same as Cl-. So, we believe it is not necessary to report the data for Na concentration.

Second Reply: We have added one sentence “Molar concentrations of Na were assumed to be the same as Cl<sup>-</sup> because pure NaCl was used”.

When authors say they that experiments were conducted in duplicate, do they mean per study site? Are such duplicates either field replicates or analytical replicates?

Reply: We mean duplicate per study site and have added this information in the text. These duplicates did not include field replicates because we stated previously that we collected composite samples in the field. - when authors mean ambient temperatures:

Second Reply: Now the sentences reads as “All laboratory salinization experiments were conducted in duplicate per study site to account for analytical variability during laboratory salinization experiments”.

Is there any control for the riparian soil experiment as the one authors employed with sediment incubations?

Reply: Yes, we had controls for the riparian soil experiment. To clarify this, we've added one sentence “Deionized waters without soils were incubated at the 3 levels of salinization as soil-free controls.” Here we did not collected snow water to conduct riparian soil experiment, but used deionized waters assuming the minor difference between snowwater and deionized water would not affect our results.

Second Reply: Done last time.

## Results

In general, this section seriously needs to be supported with the proper statistical analyses. Also, if they do, they should write the exact p-value. Sometimes, p-values associated to statistical test can make the result marginally significant ( $0.05 < p < 0.1$ ). I believe showing exact p-values is worth (for example, Line 23:  $r^2 = 0.40$ ,  $n=8$  could have a p-value marginally significant). The same suggestion when reporting ANOVA results in any paper: F values, df and p-value should be shown.

Reply: We have now reported results of statistical analyses to the results section, and added p values to linear regression and correlation analysis. We have changed ANOVA to t-test two-sample assuming equal variances for different test, and added p values for the t-test, too.

Second Reply: Had done last time. Now, we have checked to make sure to add p value for every proper place of statistical analyses.

How authors calculated the ISC?

Reply: The ISC values were adapted from previous studies, and we have listed the literature in Table 1 where ISC values were reported.

Second Reply: Had done last time. Our sites are well-studied LTER site and ISC data are available in previous studies.

## Discussion

In general, discussion deals with a good literature review and provides valuable information that authors could also use to elaborate their introduction (especially that related to biogeochemical mechanisms). I suggest to authors to discuss their results alongside the support (or not) of their initial hypothesis or predictions. A brief paragraph at the beginning to this sections summing up their main findings would be appreciated.

Reply: We have discussed our results in each subsection alongside the support (or not) of their initial hypothesis. As suggested by the reviewer, we now begin with a brief sentence summing up our main findings. We prefer this way rather than using a paragraph at the beginning to this section.

Second Reply: Had done last time, but have checked again to make sure we did for every paragraph.

Figures and Tables:

Table 2: Authors should include stream sites in the proper column as well as including in the legend that study sites are organized from rural to urban land use. Also, the meaning for codes DOC, P=H, DIC, SUVA and SRP should be included in the legend.

Reply: We have makes these changes as suggested by the reviewer.

Second Reply: Had done last time.

## Replies to Reviewer 2

My main concern regards the methods of data analysis. There are three points in respect to this concern. First, because only one rural site is used ( $n = 1$ ), I do not believe that the authors cannot address their first hypothesis as stated. Second, results of the one-way ANOVAs are reported for only AFDM, yet magnitudes and trends of responses are reported in the results for all of the bioreactive elements measured; we are not informed as to whether these results are significant. And third, I would suggest using repeated regressions to analyze the data instead of ANOVAs. By setting up their study to sample across a gradient of urbanization, the authors have the opportunity to use this gradient (%ISC) in their analysis. This method of analysis could allow them to describe trends quantitatively. For example, in Figure 2, %ISC could be used as the explanatory variable for three separate regressions (one for each Cl concentration). With this approach, the authors could better support their conclusions about how salinization affects the dependent variables across a gradient of land use change because they could quantitatively compare slopes of the regression lines. Or they could use treatments as the explanatory variable and in the figures, shade points relative to their %ISC or color code by forest, agriculture, suburban and urban categories. After addressing these gaps in analysis and reworking the results and conclusions to reflect their findings, this manuscript will be a solid contribution to the literature.

Previous Reply: We agree with the reviewer that repeated regressions are more reasonable to analyze the data than ANOVAs, and have now changed the data statistical analysis as suggested. In particulate, we have now used linear regressing with %ISC as the explanatory variable to examine effect of urbanization. Our method is slightly different from his/her suggestion, however. We do not regress for dependent variables themselves but for their salinity effect (changes in their flux per unit Cl<sup>-</sup>). There are two reasons for this change. At first, we hypothesize that the effect of salinization on sediment/ soil fluxes change with land use, not the fluxes themselves. Secondly, if we use slope with salinity, there is only one data for each site and the analysis is easier. Otherwise, there would be 3 data for each site, making statistical analysis more complex. For salinization effects, we performed linear regressions of sediment/soil biogeochemical fluxes with salinity across all sites, using data from 6 salinization experimental manipulations (3 salinity levels with duplicates). If the p-value was  $< 0.05$  for the regression, we assumed that there was a significant salinization effect. Otherwise, differences between two salinization levels were tested using a t-test of two-samples assuming equal variances.

Second Reply: Had done last time. Here we emphasize:

**Point 1 and Point 3:** We agree that repeated regressions versus ISC are more reasonable than ANOVAs, and have use linear regression to replace one-way ANOVA.

We do not agree to plotting fluxes versus ISC but plotted the effect of salinity (or the slope of fluxes versus salinity) for two reasons:

1) We hypothesize that the effect of salinization on sediment/ soil fluxes change with land use, not the fluxes.

2) If we use fluxes, there would too many data and trend might be too much complicated. Now we use the slope, there is only one data for each site and the analysis is easier.

**Point 2:** Statistics for salinization levels and difference between soils and sediment  
Salinization levels – use linear regression. If not significant, use t-test for adjacent two levels.  
Difference between soils and sediment: use t-test.

Specific Comments:

Title: The title is informative, yet suggests that the results will be interpreted within a framework of land use and only a small section of the discussion emphasizes land use. If the analyses are changed as suggested above, the contents will more in the discussion to emphasize land use accurately reflect the title.

Reply: We've add more discussion in the discussion section to emphasize land use. For example, we add a subsection "Changes in salinization effects with watershed urbanization" in Discussion and put it in the first place. Thus, we still keep "across land use" in the title.

Second Reply: Had done last time. Change in salinization effect across land-use is one of the objectives of this paper, and we still keep it title.

Introduction: The introduction is short, but provides an appropriate framework for the study. To emphasize the importance of this study, the authors may also wish to include that salinization is difficult if not impossible to reverse, thus, remediation is unlikely. Further, they may want to list saltwater intrusion caused by sea-level rise as another cause of increased salinization that is relevant to this study.

Reply: We've listed saltwater intrusion (caused by sea-level rise) as another cause of increased salinization, and added a sentence stating that salinization is difficult if not impossible to reverse, thus, remediation is unlikely.

Second Reply: Had done last time.

Methods: The authors provide a detailed, clear methods section. Although the site names are specific to the Baltimore Ecosystem Study LTER, and thus used in many other studies, it would be easier for the reader to interpret them according to the mainpoint of the manuscript if the names relayed the type of site, for example "Forest", "Agriculture", and "Suburban 1" or an abbreviated version of those.

Reply: We have added in figures and tables the information containing both type of site and the name of each site. We also did so in text of results and discussion.

Second Reply: Had done last time.

How long were the samples stored before processing and experimentation? Was nitrite negligible? Were the response variables assessed for equal variance prior to statistical analysis? Were the data transformed? Did the data display normality?

Reply: All these information has now been added to method section. The samples were stored for 2 days before processing and experimentation, because we need some time for preparation. Nitrite was negligible because our analysis showed far low nitrite concentration relative to nitrate. The response variables were assessed for equal variance prior to statistical analysis. Our statistical analyses do not need data transformation.

For data normality, we reported "For linear relationships, Spearman's correlation was used in cases where assumptions of normality were not met".

Second Reply: Had done last time.

Results: As stated above in the Summary Comments section, the results could be improved by taking advantage of the land use gradient the authors sample across. It looks like there some very interesting trends, but without the statistics to support them, the conclusions are limited.

Reply: We have plotted salinization effects against land use gradient – using impervious surface cover as an index, and use statistics to show if the effect change across ISC%.

Second Reply: Had done last time.

Discussion: The authors could organize it a bit differently to match the hypotheses and objective stated in the introduction. I would also include some remarks about SRP at the agricultural site as this location was likely fertilized. S.C. Neubauer, M. Ardón, J.L. Morse, and A.M. Helton have published additional work that could inform the discussion.

Reply: We have reorganized this section to match the hypotheses and objective stated in the introduction. Yes, this location was likely fertilized, and thanks for providing the literature and we have now cited it in the paper.

Second Reply: We have made it clearer that the frequency of discussion section match the hypotheses and objective stated in the introduction. The first part is Changes in salinization effects with watershed urbanization (hypothesis 1), and then discuss Potential effects of increased salinization on DOC/DIC mobilization, and how it is different between soils and sediment (hypothesis 2 and 3). In the end, potential effects of salinization on N, P, and S leaching/transformation coupled with DOC biogeochemistry (hypothesis 2).

Technical Corrections: Check for consistency of “land use” or “land-use.” It varies throughout the manuscript.

Reply: We have changed “land-use” to “land use” to be consistent.

Second Reply: Had done last time.

Page 7414, Line 17-18: Rephrase to clarify “and improve water quality by benefitting our assessment and management of salt use”.

Reply: We have rephrased this sentence as suggested. Thanks.

Second Reply: Had done last time.

Page 7414, Line 24: What is the origin of the stream water? Specify that it matches the sediment collected at each site.

Reply: The whole paragraph has now been deleted.

Second Reply: Had done last time.

Page 7438, Table 1: Which NLCD year?

Reply: We have added the year (2006) of NLCD in the legend of Table 1.

Second Reply: Had done last time.

Page 7416, Line 18: For how long were samples stored before experimentation?

Reply: We have now stated that samples were stored for two days before experimentation.

Second Reply: Had done last time.

Page 7417, Line2: Supplier/brand of NaCl?

Reply: We have deleted Supplier/brand of NaCl.

Second Reply: Had done last time.

Page 7417, Line 7: Describe controls as sediment control or sediment-free controls (alternatively, sediment controls and water-only controls). What you refer to above seems to be the experiment, not the controls.

Reply: We have deleted the words “as controls” to avoid this misunderstanding.

Second Reply: Had done last time.

Page 7418, Line 6: Subtract nitrate/nitrite?

Reply: We have added sentence that “that NO<sub>3</sub>-N/NO<sub>2</sub>-N concentrations were almost entirely NO<sub>3</sub>-N(>99 %), and we therefore referred to this fraction as NO<sub>3</sub>-N throughout”. So, it is not necessary to make change here.

Second Reply: Had done last time.

Page 7418, Line 17: Keep tenses consistent throughout: “Basically, we used..” changes to “was used” to stay with the subjunctive tense.

Reply: We’ve changed the structure of this sentence as suggested, and tried to keep tenses consistent throughout the paper.

Second Reply: We’ve tried our best to use subjunctive tense.

Figures: Keep axis titles and keys consistent within and between figures. If you use L-1 (instead of /L) in the axis title, also use that notation for the key and all other figures. Also, indicate the statistical results. Which comparisons are significantly different?

Reply: We’ve changed “/L” to “L-1” throughout the paper (text and figures). We have now added \* and stated in figure caption that it means significant difference.

Second Reply: Had done last time.

Figure 2: Use Standard Error instead of SD.

Reply: We should have made such change. Now, we do not show Standard Error in this graph but all data.

Second Reply: Had done last time.

Figure 6: Improve these by using 2-digits for R<sup>2</sup> values to make the graph less crowded or report R<sup>2</sup> in figure caption instead. Specify linear or non-linear correlations (only panel ‘a’ looks non-linear) and explain this choice in the manuscript. Try a graphing program other than MS Excel to make the figure more attractive or modify the default Excel settings.

Reply: To make the graph less crowded, we do not report R<sup>2</sup> values in figure. In figure captions, we mention that “A line was added to the data only if correlation was significant ( $p < 0.05$ ). Correlation coefficients were not labelled but all  $> 0.67$ ”. We have added only linear correlations and removed all nonlinear correlations.

Second Reply: Had done last time.

Page 7420, Line 10: Indicate if  $\pm 1.9$  is Standard Error or Standard Deviation. Continue indicating this with each  $\pm$  throughout.

Reply: We have now added “mean  $\pm$  standard error, same below” to explain it is Standard Error. We have stated in method section that “Data are reported in mean  $\pm$  standard error”.

Second Reply: Had done last time.

Page 7420, Line 10: Typo: higher

Reply: We have changed this typo.

Second Reply: Had done last time.

Page 7420, Line 20-21: (increase of 1.2 . . . times) or (increased by 1.2. . . times)

Reply: We have corrected this error.

Second Reply: Had done last time.

Page 7420, Line 9-10: Indicate that data for calculating the 7.8 times higher DOC values at 4 g Cl/L are not shown since the graphs are for changes in DOC not absolute values.

Reply: "DOC values at 4 g Cl/L" was actually DOC releases, and there were shown in the Figure. Now, we have now changed "DOC values at 4 g Cl/" to "DOC releases at 4 g Cl/L".

Second Reply: No changes are needed, because the reviewers misunderstood "DOC values". "DOC values" are fluxes and shown in Figure, so we had changed "DOC values at 4 g Cl/" to "DOC releases at 4 g Cl/L".

Figure 2: Indicate outliers with \* and then explain this in the figure caption to remove clutter from the graph. What did you do with the outliers? Are they part of the calculations in page 7420 Line 10? Remove the replicate key and put the other key in a more prominent location (for example, above panel a. instead of within it).

Reply: We have now used different Y scale for this site (GFCP), and all the values can be shown.

Second Reply: Had done last time.

Page 7421, Line 3-4: Report results from similar studies in the Discussion instead of the Results section.

Reply: We have deleted this sentence here and removed it to Discussions Section.

Second Reply: Had done last time.

Figure 4: Align panel letters.

Reply: We've paid attention to panel letters alignment.

Second Reply: We've checked all figures to make panel letters are aligned.

Page 7421, Line 14: 1.6 times (not time)

Reply: We have now changed this error.

Second Reply: Had done last time.

Page 7422, Lines 9-21: Because your question is about general relationships of biogeochemical couples, I would suggest you analyze the sites as aggregate rather than looking for site-specific trends.

Reply: Although biogeochemical coupling between the fluxes of chemical species could be a common phenomenon, the relationships of biogeochemical couples likely varied site by site. The reason is that sediment and water chemistry were different at each site, and the controls for relationship between fluxes were thus different site by site. We still look for site-specific trends and use treatments (not site) as cases. Actually, we can see that there would be no correlation if we analyze the sites as aggregate (Fig. 7).

Second Reply: We still disagree with the reviewer and keep our initial approach for correlation analysis. We examine site-specific trends in order to test our 2nd hypothesis "retention/release of nitrogen, phosphorus, and sulfur in response to salinization can be abiotically and/or biologically coupled with carbon biogeochemistry". Actually, this question is not general relationships of biogeochemical couples, but site specific. For example, release of DOC from sediment can cause nitrogen retention in stream water, but effect may differ at forest site than

urban/agricultural site. The reason is that forest site is low in nitrate concentration; the same amount of DOC release has less effect at forest site than at urban site.

Figure 7: Use (n=1) instead of (1).

Reply: We have changed this figure to Figure 4, and use impervious surface cover as X- axis, and this change (number) is not necessary.

Second Reply: Had done.

Page 7423, Line 25: “there were no consistent changes”

Reply: The whole paragraph has been deleted in the new version of manuscript, and this change is now not necessary.

Second Reply: Had done last time.

Figure 8: Remove second column (Difference) as this is the same data as the first column just presented as a difference instead of the absolute values.

Reply: This figure and the whole section of pre and post changed have been deleted in the new version of manuscript for the reasons that we have mentioned above, and this change is now not necessary.

Second Reply: Had done.

Page 7423, Lines 19-20: If these changes are significant, report the statistics or use “Considerable” instead of “Significant.”

Reply: The whole paragraph has been deleted in the new version of manuscript for the reasons that we have mentioned before, and this change is now not necessary.

Second Reply: We have also done the same for many other cases for “significant”.

Page 7423, Lines 2-4: Report information about the correlation analyses (type of correlation, all  $p > 0.05$ ).

Reply: Initial sentence: “Salinization effects on DIC releases generally followed a similar pattern, but there was large variability at the urban sites.” This sentence has been moved to 4.1 - Influence of salinization on C fluxes and DOC composition across land use. We have reported statistical information about the correlation analyses - “Overall, our results suggest that the effects of increased salinization on sediment releases of DOC, protein-like fluorophore, TKN and DIC increased with impervious surface cover (ISC) – an index for watershed urbanization (Fig. 4; linear regressions, all  $p < 0.05$ ).”

Second Reply: Had done.

Page 7426, Lines 11-14: Do the results support this? Reply: “Due to larger fractions of humic substances, the: : .”

Reply: Because we did not measure humic substances, the results did not support this. Now, we have reworded this sentence to “In this study, although we did not measure humic substances, we observed much higher SUVA values in DOC leached from soils than from sediments. This suggested that soils were higher in humic substances. Probably, due to large differences in organic matter composition, effects of salinization on DOC leaching from sediments and soils were different”.

Second Reply: Had done last time.

Figure 6: Align panel letters (a,c,e). Figure 6a: Reason for using nonlinear patterns?

Reply: Panel letters has now been aligned. Nonlinear curves have now been removed.

Second Reply: Nonlinear curves have now been removed, but we still keep dash lines to link all points. We've also explained in legend the difference of solid lines and dash lines.

Page 7427, Line 9: You mean Figs.4 and 5 instead of Figs. 3 and 4? Page 7427, Line 22: Figs. 4 and 5

Reply: We have changed these errors.

Second Reply: Had done last time.

Page 7429, Lines 10-11: "stream sediments and soils" because Figure 5 refers to soils. Though, I am not convinced that there is a general trend of sediment sulfate release; Fig. 4d shows that for 0g Cl/L treatments, sulfate increased in just over half (5/8) of sites.

Reply: I agree with the reviewer that convinced that there is a general trend of sediment sulfate release. Now this sentence has been reworded as "However, our results show large variability in the effects of salinization on net sulfate release from either sediments or soils (Fig. 5 and 6), and sulfate reduction may not be dominant in free-flowing streams."

Second Reply: Had done last time.

Figure 9: Good conceptual figure. Try changing black text to white to see if it increases visibility.

Reply: We have now changed black text to white to increase visibility.

Second Reply: Had done last time.

Page 7431, Line 21: "our work suggests"

Reply: We have corrected this error.

Second Reply: Had done last time.

Page 7433, Lines 10-14: Remove text justification.

Reply: Text justification has been removed.

Second Reply: Had done last time.

Page 7437, Line 9: Italicize or underline species.

Reply: The species has been Italicized.

Second Reply: Had done last time.

Additional changes: Improved quality of Figure 2-7, and polished the text of the whole paper.