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Interactive comment on "Salinization alters fluxes of bioreactive elements from streams and soils across land use" *by* S.-W. Duan and S. S. Kaushal

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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.

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-). 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.

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 in the discussion to emphasize land use, and keep "across land use" in the title.

Abstract: This section is a concise summary of the paper.

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.

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 main-point 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 the information regarding type of site as the name of each site, when we reported the data in results and discussion, or in Tables or figures.

There are a few additional points to address in this section that could improve repeatability. 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".

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 accepted the advice of the reviewer and plotted salinization effects against land use gradient – using impervious surface cover as an index.

Discussion: This section is an interesting and comprehensive interpretation of the data. 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.

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.

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.

Page 7414, Line 24: What is the origin of the stream water? Specify that it matches

the sediment collected at each site.

Reply: This paragraph has now been deleted because it is repetitive and unnecessary.

Page 7438, Table 1: Which NLCD year? Reply: We have added the year (2006) of NLCD in the legend of Table 1.

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.

Page 7417, Line2: Supplier/brand of NaCl?

Reply: We have deleted Supplier/brand of NaCl.

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.

Page 7418, Line 6: Subtract nitrate/nitrite?

Reply: We have added sentence that "that NO3-N/NO2-N concentrations were almost entirely NO3–N(>99 %), and we therefore referred to this fraction as NO3-N throughout". So, it is not necessary to make change here.

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.

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.

Figure 1: Nice graphic!

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.

Figure 6: Improve these by using 2-digits for R2 values to make the graph less crowded or report R2 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 MSExcel to make the figure more attractive or modify the default Excel settings.

Reply: To make the graph less crowded, we do not report R2 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.

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".

Page 7420, Line 10: Typo: higher

Reply: We have changed this typo.

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

Reply: We have corrected this error.

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”.

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: Good suggestions. Actually, we cannot consider these data as outliers, they were much higher values that other sites. We have now used different Y scale for this site, and the values can be shown. Because there were not outliers, they are part of the calculations.

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.

Figure 4: Align panel letters.

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

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

Reply: We have now changed this error.

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).

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

Reply: We have changed this figure to Figure 1, and use impervious surface cover as X- axil, and this change (number) is not necessary.

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.

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.

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.

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

Reply: “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, $p < 0.05$).".

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

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 remived.

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.

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. 5and 6), and sulfate reduction may not be dominant in free-flowing streams."

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.

Page 7431, Line 21: "our work suggests"

Reply: We have corrected this error.

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

Reply: Text justification has been removed.

Page 7437, Line 9: Italicize or underline species.

Reply: The species has been Italicized.

Please also note the supplement to this comment:
http://www.biogeosciences-discuss.net/12/C5578/2015/bgd-12-C5578-2015-supplement.pdf

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Interactive comment on Biogeosciences Discuss., 12, 7411, 2015.

Figure 1

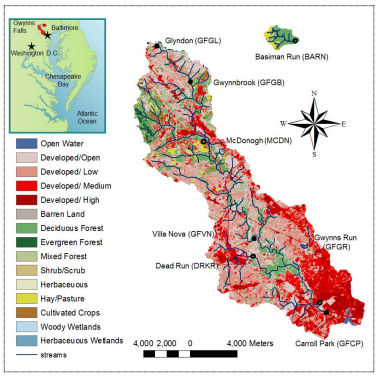

Fig. 1.

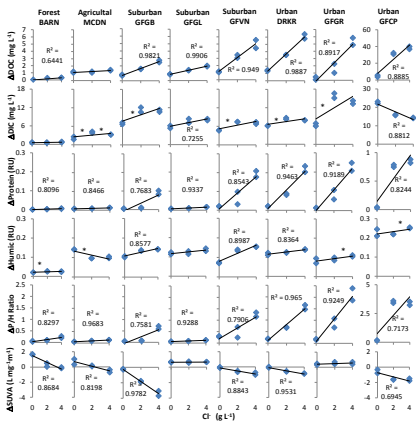

Fig. 2.

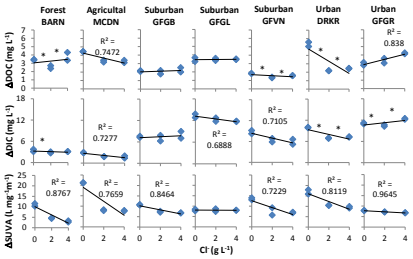
Forest BARN
Agricultal MCDN
Suburban GFGB
Suburban GFGL
Suburban GFVN
Urban DRKR
Urban GFGR
ΔDOC (mg L-1)
ΔDIC (mg L-1)
ΔSUVA (L mg-1m-1)
R² = 0.7472
R² = 0.838
R² = 0.7277
R² = 0.7105
R² = 0.6888
R² = 0.8767
R² = 0.7659
R² = 0.8464
R² = 0.7229
R² = 0.8119
R² = 0.9645
Cl- (g L-1)

Fig. 3.

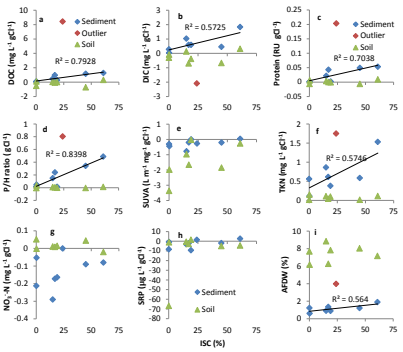
a
Sediment
Outlier
Soil
R² = 0.7928
DOC (mg L-1 gC-1)
b
R² = 0.5725
DIC (mg L-1 gC-1)
c
R² = 0.7038
Protein (RU gC-1)
d
R² = 0.8398
P/H ratio (gC-1)
e
SUVA (L m-1 mg-1 gC-1)
f
R² = 0.5746
TKN (mg L-1 gC-1)
g
NO3-N (mg L-1 gC-1)
h
Sediment
Soil
SRP (ug L-1 gC-1)
i
R² = 0.564
AFDW (%)
ISC (%)

Fig. 4.

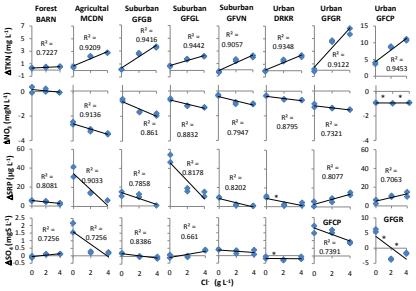
Forest BARN
Agricultal MCDN
Suburban GFGB
Suburban GFGL
Suburban GFVN
Urban DRKR
Urban GFGR
Urban GFCP
ΔTKN (mg L-1)
ΔNOx (mgN L-1)
ΔSRP (µg L-1)
ΔSO4 (mgS L-1)
Cl- (g L-1)

Fig. 5.

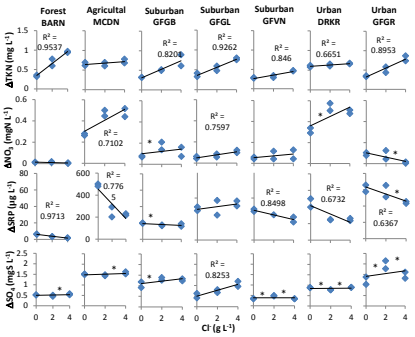
Forest BARN
Agricultal MCDN
Suburban GFGB
Suburban GFGL
Suburban GFVN
Urban DRKR
Urban GFGR
ΔTKN (mg L-1)
ΔNOx (mgN L-1)
ΔSRP (µg L-1)
ΔSO4 (mgS L-1)
Cl- (g L-1)

Fig. 6.

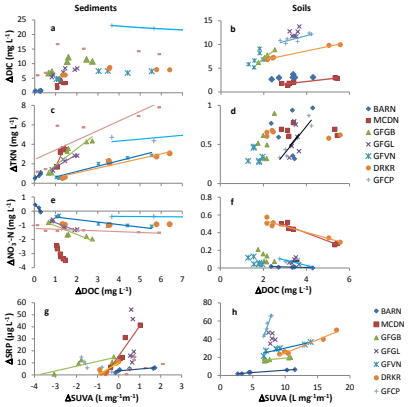
Sediments
Soils
a
b
c
d
e
f
g
h
ΔDIC (mg L⁻¹)
ΔTKN (mg L⁻¹)
ΔNO₃⁻-N (mg L⁻¹)
ΔSRP (µg L⁻¹)
ΔDOC (mg L⁻¹)
ΔSUVA (L mg⁻¹m⁻¹)
BARN
MCDN
GFGB
GFGL
GFVN
DRKR
GFCP

Fig. 7.

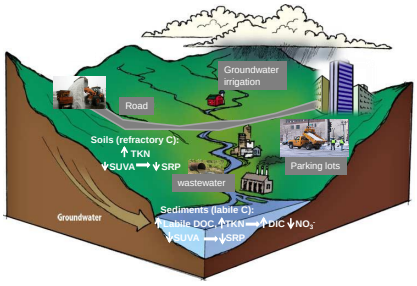
Groundwater irrigation
Road
Soils (refractory C):
↑TKN
↓SUVA
↓SRP
Parking lots
wastewater
Groundwater
Sediments (labile C):
↑labile DOC
↑TKN
↑DIC
↓NO₃⁻
↓SUVA
↓SRP

Fig. 8.