Anonymous Referee #1: Received and published 18 October 2016

Author responses are italicized in blue below the reviewer comments.

General comments:

The topic of the manuscript is suitable for the journal Soil. It reports on nitrate leaching from two soil horizons originating from sediments behind a milldam and from an older horizon buried under them. From the regional prevalence and from the high leaching rates observed, this study is certainly justified. The topic is very well explained in the introduction, which describes not only the specific questions of the study but also the geographic and historic context. The presentation of the site is extensive and very well written.

The main value of the experiment lies in the fact that such soil profiles have barely been studied in this manner. This value is, however, limited in the sense that it covers only one site. It is therefore not obvious to draw conclusions for other sites impacted by milldam sediments.

Response: We agree that the findings at BSR (just one site impacted by milldam sediments) may not be representative of all other sites similarly impacted by milldam sediments. Therefore, we have added a caveat in the results and discussion section bringing up this point, and include the comment that further research at multiple sites impacted by milldam sediments is required to better understand if the low NO_3^- retention we found in legacy sediments is specific to the BSR site or whether it is characteristic of all sites similarly impacted by legacy sediments.

The study was conducted on soil columns, which is an efficient way to measure leaching. Nitrate added to these columns was labeled with 15N, which allows to distinguish it from soil-derived nitrate. It would have been useful to add also a biologically inert tracer like bromide, to be able to distinguish between physical and biological processes. This would have especially been useful to ensure a better interpretation about preferential water flow, which may have been favoured by the mechanical extraction of the soil columns, as the authors note. A dye may also have been useful to answer this question.

Response: We agree that adding a biologically inert tracer to distinguish between physical and biological processes, or a dye to better explain the main flow regimes through the soil columns, would have been useful techniques to employ during this study. Unfortunately, we did not utilize these methods during our study, but we did add a comment in the conclusions section that data interpretation could be strengthened in the future if a co-tracer or dye is added when conducting similar soil column leaching/retention studies.

For the interpretation of the flow regime, the authors analysed the texture of the soil and estimated hydraulic conductivities. However, they do not describe the structure of the soil, nor if there were roots in the soil columns. In spite of the weight of the sediments, the buried, relict A horizon was found to have a high porosity, but it had a low hydraulic conductivity: this may be due to an horizontal structure within this horizon, with some less permeable layer. It is at least not likely that a former A horizon would be homogeneous over 30 cm in depth (even if it had been ploughed, then at that time certainly not so deep). Without a proper description of the structure, interpretations about the flow regime are difficult. It would certainly be useful to compare the measured water infiltration rates with those predicted by the parameters of table 3.

Response: The reviewer makes a good point that soil structure and the presence/absence of roots in the different soil horizons are important parameters that might impact our interpretations of different flow regimes. We added a description of both these parameters to the Soil Properties section of the text and drew on them to enhance our interpretations. However, we do not have measured water infiltration rates, which is why we needed to use the predicted parameters of Table 3.

The results about general soil properties are given after the results of nitrate leaching.
The contrary would certainly facilitate the presentation and discussion.

Response: We rearranged these two sections. Now we discuss the general soil properties prior to presenting the results of the nitrate leaching experiment.

Further, the question of N saturation (especially its kinetic aspect) would require a comparison of the applied amounts (approximately 0.5 g/m2 if my calculation is correct) with nitrification rates and with nitrate in atmospheric deposition. These rates are unfortunately not quantified here. The discussion about assimilation and remineralisation is essentially justified, but quantitatively it is likely to play only a minor role: it is less likely that N is assimilated and remineralised and nitrified and leached than just leached. The relative importance of direct leaching versus leaching after mineralization and nitrification should be discussed in the light of the tracer fraction in the leachate, i.e. the molar ratio of tracer N to total N in leached nitrate.

Response: We agree that to definitively determine the extent of N saturation in the system would require a much larger dataset, with a more complete inventory of inputs and outputs, which is beyond the scope of the current study. We focused on learning more about NO_3^- retention within the different soil horizons of BSR, and believe that our findings can best be interpreted using N saturation theory. As a first assessment we believe our hypothesis is reasonable, and acknowledge that more work is needed to quantitatively assert whether the low NO_3^- retention can be attributed solely to the system's N saturation status. We added this line of thinking to the text. We also agree that direct leaching is likely much more important than leaching after mineralization and nitrification (as we discussed in the text), but followed the reviewer's comments and added some quantitative data to back up this by looking at the tracer fraction in the leachate.

Finally, the discussion about a co-saturation of C and N is not convincing in this case. At the basis of this concept, there is the work of Cleveland & Liptzin (Biogeochemistry 85 (2007): 235-252), who give for such a co-saturation a C/N ratio of the soil near 14. Here, however, C/N ratios are around 9 or 10 (calculated from table 1). This means that the amount of C present in the soil would still give much "room" for N immobilisation. Further, a co-saturation is likely to be limited to soils which are well drained, have a sufficient pH and are warm enough. While the third condition is certainly fulfilled here, the other ones would need to be discussed: what is the pH in these soil horizons, and are there signs of anaerobicity that could hinder mineralisation?

Response: Prior findings at BSR showed that the relict A horizon appears to be inefficient at utilizing new C inputs (Weitzman et al., 2014). Thus, while the C:N ratio of the relict A

horizon may be near 10, instead of near 14, as Cleveland and Liptzin (2007) give as the critical ratio at which co-saturation is likely to occur, the finding that the soil horizon cannot utilize new C inputs suggests that it acts similarly to a C-saturated soil. For this reason we believe that our comment about co-saturation of C and N holds. We did, however, add a similar explanation to the text to address the above comment.

Details:

Introduction

Page 3, line 27: the abbreviation BSR should be defined here in the introduction (it is defined only later).

Response: We added the full definition for the BSR abbreviation as "Big Spring Run (BSR) in Lancaster, Pennsylvania" at this first introduction.

P. 3, L. 27: "created" is perhaps not the best wording for nitrate. And there is also nitrate which is not formed in the soil but brought by atmospheric deposition.

Response: We changed the wording from "created" to "produced in situ" to refer specifically to the NO_3^- that forms in the soil, as opposed to that which can accumulate via atmospheric deposition.

Material and methods

P. 6, L. 14: was the soil cultivated? If there were plants, how were they removed? Was there a litter layer, and if yes was it removed?

Response: We sampled the soil right next to Big Spring Run (<5 m from the edge of the high-cut stream bank; what is traditionally the riparian zone). The soil in this area was not cultivated and was characterized by tall grasses that were clipped close to ground-level (to a height of ~2.5 cm above the soil surface) to facilitate easier soil column extraction. Clipped grass and its associated roots remained in the surface soil columns for the leaching experiments. We added these details to the text.

P. 7, L. 12: the amount of water corresponds to 92 mm, which is high but not impossible for a single precipitation event.

Response: The amount of water corresponds to a rain event of ~370 mm (diameter of the cores = 20.32 cm, not the radius), which is more typical of an "extreme" rain event, as opposed to a normal precipitation event. Such an "extreme" rain event is more aligned with current climate change models that predict higher intensity rainfall events for the region, which could be followed by drier conditions.

P. 7, L. 28: "g soil-1" should be written with a parenthesis: (g soil)-1, otherwise it would be like only "soil" and not "g" is at the power -1.

Response: Parentheses were added around (g soil) to denote the correct unit raised to the -1 power.

P. 8, L. 16: atom% enrichment is not defined. This would be necessary to understand correctly the given equations.

Response: We realize that atom % enrichment can sometimes be confused with atom % excess, and could erroneously be interpreted to mean the enrichment above natural abundance. To avoid this confusion we chose to remove the word "enrichment" and instead use atom percent (shortened to atom %) throughout the paper. Here we define atom percent (atom %) as the absolute number of atoms of a given isotope (here ^{15}N) in 100 atoms of the total element, i.e. atom % $^{15}N = \left(\frac{^{15}N}{^{14}N + {^{15}N}}\right) \times (100)$. We included this definition to allow for more clarity.

P. 9, L. 8: multiplying by 100 is for a unit transformation (from a simple ratio to percent): such unit transformations are not an essential part of an equation and should be given only if it explicitly presented as such.

Response: We removed the "x 100" from the equation.

P. 9, L. 22: it would be useful to write in a couple of words what is the working principle of this software.

Response: The SWRC Fit program (Seki, 2007) performs non-linear fitting of soil water retention curves to a number of soil hydraulic models. The program automatically determines all the necessary initial conditions for the non-linear fitting, which allows users to simply input soil water retention curve data to obtain estimated hydraulic parameters. We added this explanation to clarify the working principle of the software, as suggested.

Results and discussion
P. 11, L. 23: "vice versa"

Response: We changed "vice-a-versa" to the wording "vice versa" as suggested.

P. 11, L. 29: better name a dimension by its proper name than indirectly by it units, i.e. better "abundance" than "atom%".

Response: We do not agree that "abundance" is the right term to use, but instead chose to keep atom %, which we defined earlier.

P. 12, L. 26: "too slow" is relative to the speed of leaching, i.e. the interpretation could as well be that leaching is too fast, especially in the case of preferential flow.

Response: This is a fair point. We clarified that the biological sinks may be "too slow" relative to the speed of leaching.

P. 13, L. 7: adsorption of ammonium is certainly also a reason why the concentration in solution are lower.

Response: We added that adsorption of ammonium to clay particles in the soil could also potentially explain why soil extractable concentrations of NH_4^+ were much lower than those of NO_3^-.

P. 14, L. 3: this repeats what has been written above.

Response: We removed the repeated explanation that organic matter content can impact the shape of the soil water retention curve.

P. 15, L. 2: quite a long sentence.

Response: We separated the one sentence into two sentences to reduce its original length and wordiness.

Tables and figures
Tab. 1: the legend of the figure mentions "concentrations" but the data are amounts per area, and it is not clear if these amounts are only for the 30 cm columns or if they are extrapolated for the whole corresponding horizon in the field.

Response: We replaced "concentrations" with the more accurate "mass per unit area" and clarified that the amounts in the table represent averages of the 30 cm soil core segments.

Tab. 2: it is astonishing that the relict A horizon has as much organic matter than the other horizons while it has (according to table 1) a much higher C content (but this may be related to the question above).

Response: Organic matter contents were reproduced from Weitzman et al., 2014. These correspond to different samples taken in a similar location. They may not correspond exactly to the soil columns, but the general trend should be similar.

Fig. 2: all the information in this figure is already contained in figure 1. It should thus be deleted.

Response: We believe both Figure 2 and Figure 3 are needed to understand the difference between 15N recovery vs. 15N retention. See longer explanation in response to Reviewer #2.

Fig. 4: what are "atm%" in the legend of the Y axis? Are these atom%? The graph would be easier to read if it would give a tracer fraction (as defined above).

Response: Atm% is atom percent. They should be interpreted in relation to the added 15N tracer which had a value of 60 atm%. We spelled out atom % in the figure and added the explanation about the tracer value to the legend for better clarification.

Conclusion

The manuscript discussed here starts very well and lets the reader hope for an exciting story. However, it shows then some limitations in the material and methods and ends with a discussion that misses some important aspects.

Response: As the first study to address the impact of legacy sediment effects on nitrate retention through the soil profile we are aware that there were some method limitations, and that more investigation is still needed. We have added these caveats to the conclusions, as discussed above, and have expanded the discussion to include more predictions about the long-term effects of restoration and other potential processes that may be limiting NO_3^- retention in the legacy sediment impacted soils.