Author Comment for BG-2015-331 “Nitrogen export from a boreal stream network following forest harvesting: seasonal nitrate removal and conservative export of organic forms” by Schelker, J., Sponseller, R., Ring, E., Högbom, L., Löfgren, S., Laudon, H.

This second Author Comment will address specific comments and changes made on the manuscript. Please note that a detailed discussion of the main comments of the referees is given in the first Author Comment (described here again in blue color).

Yours sincerely for the authors

J. Schelker

Reviewer #1

This study quantified nitrogen removal by the river system draining a third order water-shed in Sweden. Nitrogen loading is elevated because of clearcutting in this primarily forested watershed. Because significant deforestation has occurred recently, loading into the headwater streams has increased. The amount of nitrogen entering the en-tire river network can be estimated based on the proportion of the watershed that has been clear cut (all in a similar time frame), and fluxes that are characteristic of forested and clear cut catchments. This modeled estimate of loading can then be compared to fluxes measured at the mouth of the watershed, and the difference is due to nitrogen retention by the watershed. The study found that DON is not retained, where a signifi-cant proportion of nitrate is net retained (from 30 to 100%). Highest retention appears to occur following spring snow melt, lowest during the winter, and intermediate during the summer growing season. Retention was not related to flow conditions. Results indicate that increased export from small catchments due to clear cutting can be re-tained by the river network, buffering the impact in larger rivers and downstream water bodies.This is an interesting study and well written manuscript. Overall, I believe that the analysis is sound. A few issues need to be addressed however to strengthen the paper.

Dear Dr. Wollheim, thank you very much for this overall positive evaluation. We will do our best respond to your comments and to address your concerns.

I was surprised that removal in this relatively small network is so high. I think it is important to report the surface area estimate of the river network. In addition, there are lakes in the watershed, which likely increase significantly the surface area of surface waters. The lake in the mainstem in particular could contribute to the high removal. What is the surface area of the lakes, and their residence time?

Yes, we have would like to add the information (see Table 1) of the estimated river surface area in an additional table that will be added to the site description.

Site Name	Short Name	Catchment Area	Proportion Clear-Cut*, 2004; 2011		Wetland Area	Total Stream Length	Lake Area*	Stream Surface Area	Total Aquatic Area
Unit		[ha]	[%]		[ha]	[m]	[m^2]	[m^2]	[m^2]
Balån River 1 Outlet	BA-1	2291	2%	11%	337	37521	87829	185738	273567
Balån River 2	BA-2	868	5%	18%	88	15754	6590	19249	25839
Southern Reference	RS-3	156	0%	3%	4	2195	0	2195	2195
Southern Clear Cut	CC-4	41	0%	56%	3	1650	0	660	660
Northern Catchment	NO-5	40	0%	33%	5	1386	0	554	554
Northern Reference	NR-7	24	0%	16%	4	835	0	334	334

* estimated from satellite data

Table 1, catchment characteristics of the Balsjö catchments.

Unfortunately we have no detailed knowledge of the depth of the lake located between BA-1 and BA-2. Thus, we are not able to estimate the mean residence time. However, from other work we know that similar ponds in this landscape are commonly very shallow, i.e. don't exceed a depth of 2-3 m, but there is not really more here that we could add to the manuscript.

In addition we have now also acknowledged the possible contribution of wetlands and lakes in DIN removal in the discussion section [L. 1459ff]

The high removal estimates hinge on the loading estimates from the two clear cut catchments. One of these (CC-4) had much higher loading estimates than the other one (NO-5) and this was attributed to riparian buffer in the latter removing the inputs. The mixing model uses the average of these two catchments and I believe assumes the average applies to all cleared land in the entire watershed. The issue here is that this amount is based only two catchments with very different loading estimates. If the catchment with smaller increases is more representative, then the estimate of removal by the river system would be an overestimate. Is there any additional data available to assess which of the catchments is more representative (or whether an average is)? If riparian removal is inferred as the reason why the second catchment does not have as high response to clearcutting, are there any data on what proportion of clear cuts maintain the riparian zone? Another way to address this uncertainty, is to look at the range in watershed removal by looking at two scenarios, one where all clear cuts have the low response, and the second where all clear cuts have the high loading response.

It is absolutely correct that the model results are strongly dependent on the clear-cut (CC) loading estimates. More specifically, the main question is how the clear-cuts in the landscape can be represented by the measured data from the two somewhat contrasting CCs. Here we would like to describe a few properties of the CCs in this study.

First, the difference between the two harvested catchments is not only that a forest buffer strip was kept, but also that CC-4 has a much smaller riparian zone than NO-5. The riparian zone within NO-5 has likely been a small peatland that was drained by 'shovel and spade' sometimes in the past with several meters of peat soil surrounding the stream (Schelker et al., 2013a). This peaty riparian zone is present almost along the full length of the stream; large parts of the stream bed also consist of peat. In contrast, the CC-4 stream drains mostly a more 'upland' like catchment with well-developed podzol soils. It does have some riparian zone, though with less peat. This

streambed is more sandy, underlain by low permeable glacial till, that limits high vertical hyporheic exchange.

Second, the properties of the harvests outside of our two experimental harvests, but inside the stream networks drainage area have riparian areas of mixed character. Some of the harvests appear to be more of the upland type, some may have more extensive peat-rich riparian zones. Figure 1 below shows a map indicating the locations of wetlands combined with the satellite clear-cut data. To us, this map indicates the mixed character of both types of harvests, represented by our experimental harvest in the landscape.

Third, even though the foresters claimed that in most of the other clear-cuts, a riparian buffer zone was left intact, we have evidence from harvested areas within the network, but outside the experimental treatments, that harvests reached almost all the way to the stream and that severe soil damage was caused in locations very close to the stream (see Figure 2). Such damage of riparian soil from forestry machines originating from, for example crossing the stream, was also present within the CC-4 catchment (were stream crossings were done on purpose), but not within the NO-5 catchment and may be an additional factor for the difference in the response of the two treatments. Furthermore, narrow buffer strips were often found to be subject to wind through. We would also add this information to the methods section of the manuscript.

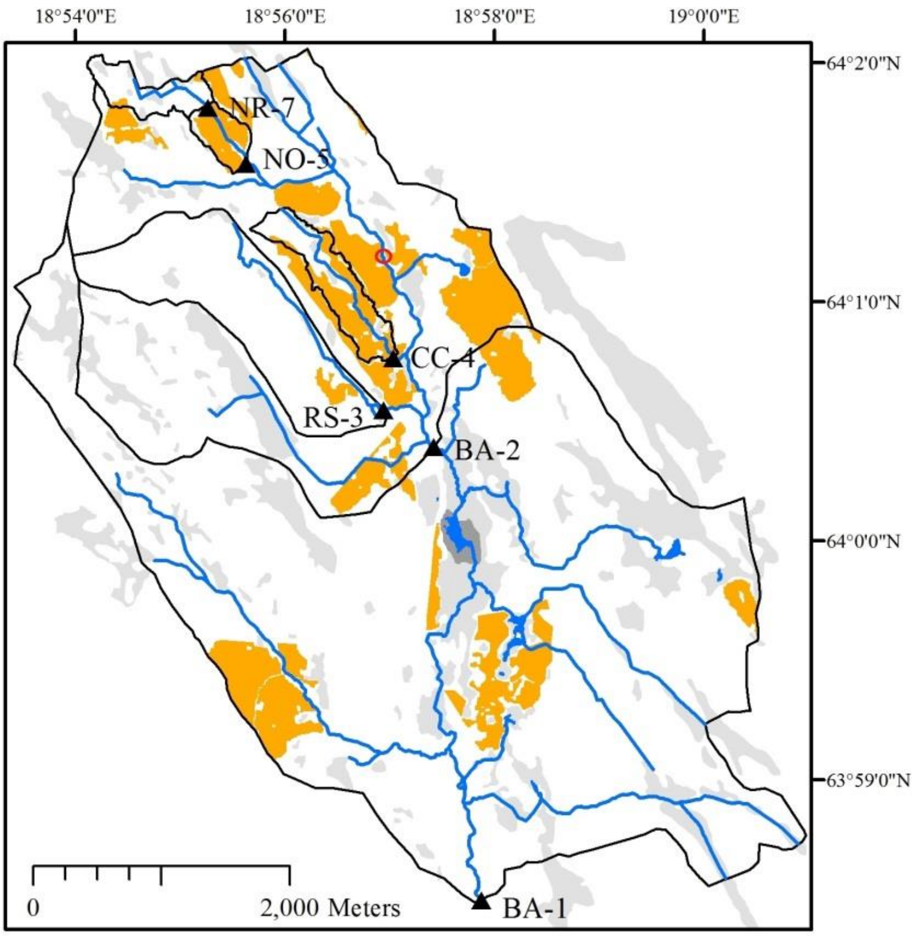


Figure 1, overlay of wetlands (grey shading), and forest harest (yellow) within the Balsjö stream network. The red circle denotes the location where Figure 2 was taken.

Figure 2, riparian soil damage in several locations within the Balsjö stream network, but outside the experimental harvests of NO-5 and CC-4 (approximate location of pictures given in Fig. 1).

Finally, the idea of using scenarios of each of the treatments to represent the CC end member in the mixing model was one we had developed previously, but decided against it. In short, the dataset we compare with contains at least one catchment that we know would not behave like this definition of the end member, i.e. if we would assume all CC areas to respond like NO-5, there would be at least the CC-4 catchment that we know does not follow this behavior. Considering that both harvest also account for an important fraction of the total drainage area of BA-1 and BA-2 that was harvested (CC4 for 9% and 15%; NO-5 for 5% and 9% of BA-1 and BA-2, respectively), applying the scenarios as suggested and comparing these to the measured values at the downstream sites would simply be wrong, as they cannot represent the physical system.

Another reason for not adopting the approach of different scenarios was given by the difficulty of presenting the resulting amount of model output data in a clear and concise manner. The effective amount of data to present would be threefold, which would make most of our plots difficult understand.

Overall we conclude that we are simply limited on more detailed data that could guide us on better choices of the input loading assumptions. Our current approach to estimate the loadings of clear-cuts as simple averages of the available data provides us with what we believe is the most robust estimate. We would therefore argue for keeping these assumptions. Furthermore, we argue that the use of model scenarios will be at the cost of clarity. We thus suggest to not introduce them into this study.

We have now also added a small section where we discuss the implications of these assumptions, and at least one scenario to the discussion section [L. 1428ff]

Greater confidence in the mixing model would be gained if there is also a conservative solute that responds to clearing, and then mixes throughout the network, where there is no removal. I believe that Shelker et al. 2014 may have this data. Use of a conservative tracer would also address the representativeness of the watersheds. I suppose the DON serves as a conservative tracer based on the result, but a priori this was not expected, whereas a solute like chloride would be conservative.

Yes, this is a good point. Figure 3, below provides this information for dissolved silica and chloride, both used as conservative tracers. We would also like to include this figure in the revised manuscript.

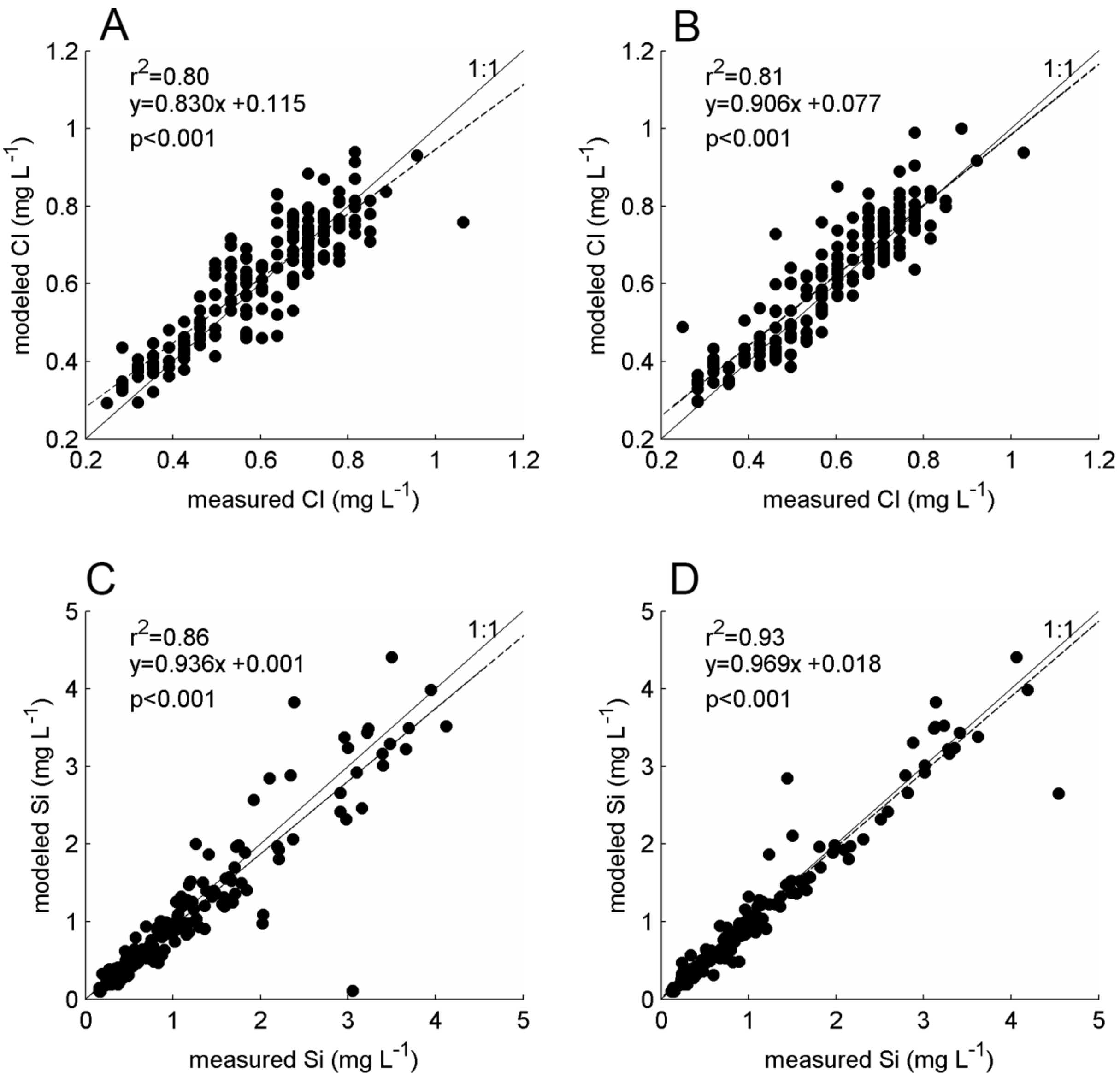


Figure 3, comparison of modelled and measured Cl and Si concentrations for BA-1 (panel A and C), and BA-2 (B and D).

These plots may be also seen in comparison to similar plots of the suggested model scenarios. The two figures (Figure 4 and 5) below present these. Panels A-C present the mixing model for the site BA-1, Panels D-F for the site BA-2. Thereby are the panels A and D the scenario with the highest load (CC-4, scaled to 100%), B and E, the 'average' scaled concentration load (as also shown above and used in the manuscript) and panels C and F the low loading scenario (response as NO-5, scaled to 100%).

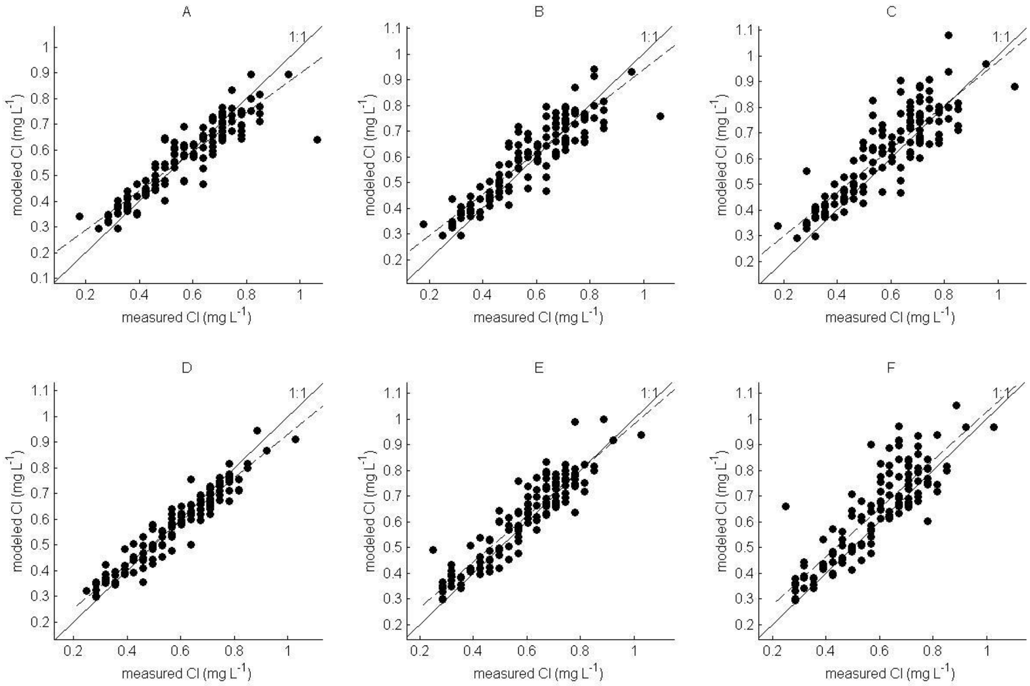


Figure 4, comparison of modelled and measured Cl concentrations for BA-1 (panel A to C), and BA-2 (D and F) for the three different loading scenarios.

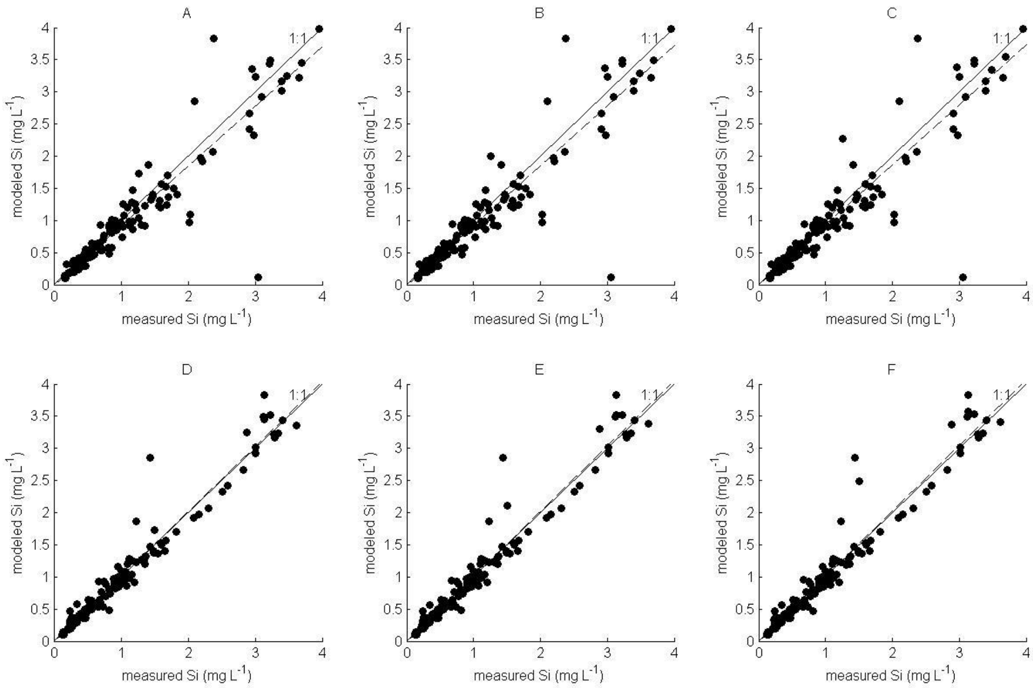


Figure 5, comparison of modelled and measured Si concentrations for BA-1 (panel A to C), and BA-2 (D and F) for the three different loading scenarios.

Overall we believe that the mixing models of Cl and Si using the different scenarios show the best overall visual fit (Figure 4, B and E, as well as Figure 5, B and E) and the smallest systematic deviation for the 'average scenario'. These observations lead us to conclude that our choice of the loading scenarios as the average concentrations, scaled to 100% is reasonable.

Although DON retention (or lack thereof) using the model is reported, no results on DON response to clear cutting are shown or presented in the results. I think this is important to include (perhaps adding a panel to Figure 2).

Yes, we fully agree to this point. We have added DON concentrations to the former Figure 2 (Figure 6, below) as a separate panel. Also, we are now presenting concentrations of NH4 and NO3 instead of only NO3, as requested by the other reviewers. These results will also be briefly presented in the revised results section.

Please see changes of the results, Figure 3, as well as discussion section.

Some more discussion of the mechanisms that contribute to the removal efficiency patterns (both over time and vs. flow) would also strengthen the paper. It is not clear what the mechanisms are so that the snow melt period would have the highest retention. Flows are high and temperatures are cold which should lead to low retention. Transfer to the hyporheic zone, riparian habitats, or groundwater is suggested very briefly. But could these explain such high losses, and why during spring only? The U term would incorporate net losses to these areas. If U is higher because there is more DOM or it is more labile why is there no DON retention then (or at least conversion of DON to DIN), especially when DIN supply is limited. What about light coming through the riparian canopy? Is it high and canopy cover low, so more primary producer uptake of nitrate during spring? If clear cut removes riparian this could be a mechanism contributing to temporary removal at least - but how common is riparian clearing. And why a more important factor in the spring? For Q to not be a factor means that as Q increases so does the uptake rate (or uptake velocity) in order for retention to remain high. A plot of uptake or uptake velocity over time or vs. flow would help to evaluate this.

When discussing the seasonal pattern of U, it should be remembered that others have shown very similar patterns before, such as high uptake in the spring (Roberts & Mulholland, 2007). Furthermore, we are somewhat limited in our data, so that not all mechanisms can be explicitly ruled out or confirmed. We will try to be more specific and less speculative in our revised version of the manuscript.

We have now also rearranged the discussion section and provide a better discussion of the seasonal patterns. Also, the results regarding NH4 have been moved to the results section.

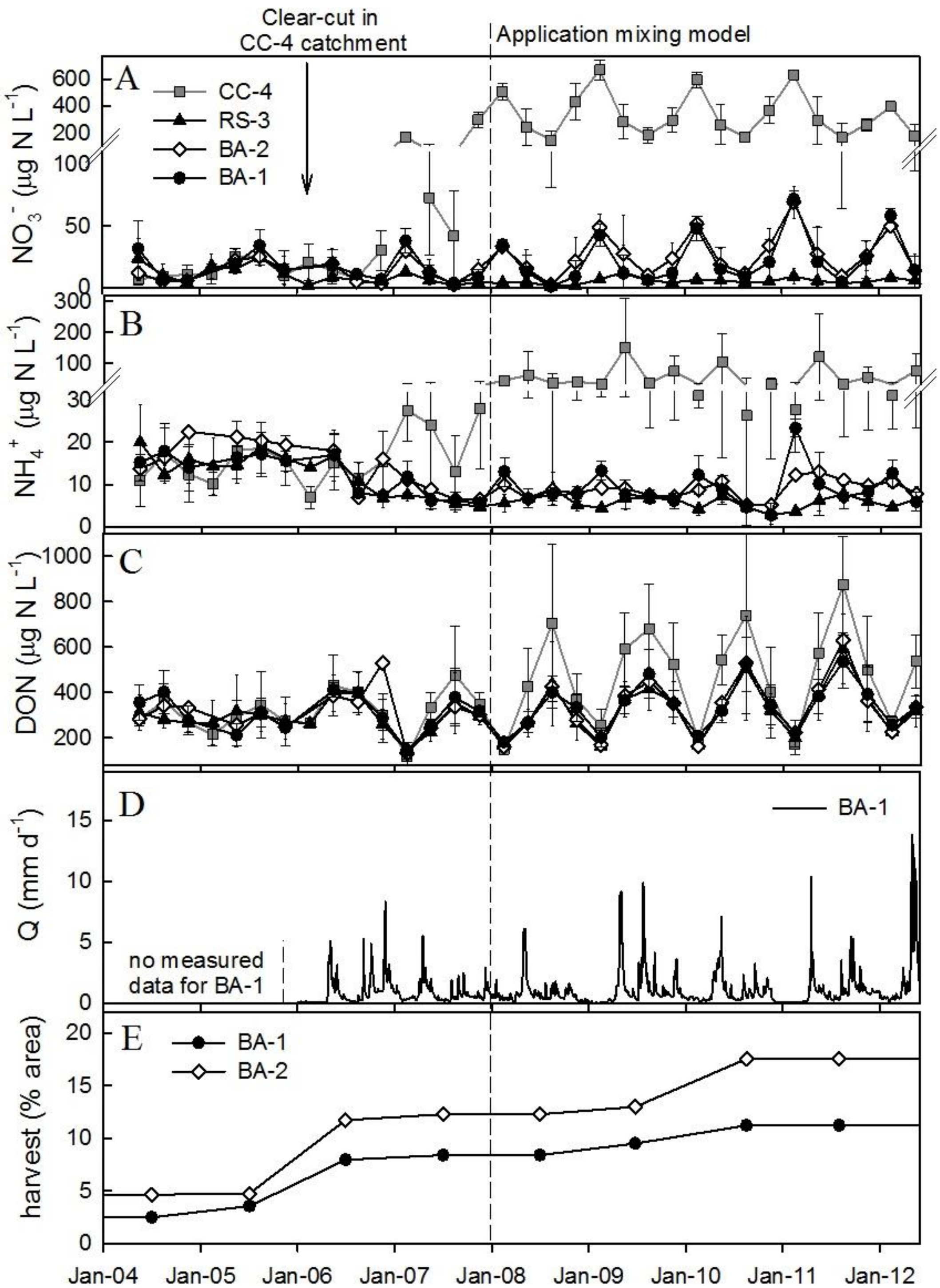


Figure 6, revised version of former Figure 2 of the manuscript.

Specific Comments

Equation 1. Units are confusing because of the use of mm/d (have mm, L, m2). Please use consistent units throughout (I suggest m), and make sure easy to see that all units cancel out correctly. Concentration units with equations given in mg, but data given in micrograms. Please be consistent.

Yes, thank you for pointing this out. The units need of course to be mm and not mm/day and also µg/L instead of mg/L. We have revised the units accordingly (see Eq. 1).

What is size of the small catchments? What length of stream is above the sample site in these watersheds? Could some removal already have occurred at sample location?

Yes, good point - we added the new Table 1 with characteristics of the stream network to the manuscript. Also, as there is some length of stream above the sample locations in all four 1st order catchments, there could of course be some removal prior to sampling. However, as we have no detailed data on this, we simply have to assume this effect to be minor.

The scaling to 100% harvested assumes (equation 2) assumes linear relationship between % harvest and concentration. Should state this explicitly.

Yes, we added a specific statement. Please also see our previous Author Comment where we also discussed this assumption for Reviewer #3.

12071.5-6. Unclear what the values in parentheses mean. Negative values are confusing. I understand the negative value is used because it is removing N from the water column, but areal uptake should be reported as a positive value.

Yes, absolutely right. We revised the numbers and present them now as positive uptake numbers for DIN.

12075.6. Denitrification is a dissimilatory process.

Yes, thanks for pointing this out. We removed the term 'assimilatory'.

12075.11. Dissimilatory reduction to ammonium (DNRA) is also a dissimilatory process, but seems unlikely in this site. Mostly occurs where there is low OM, and very high N (much higher than here). This discussion seems too speculative. If keep, then add refs on this process from the literature.

We removed this section.

12075.18. Should include more evidence of high DOC in this catchment if want to make this point. Seems too speculative.

We added a statement.

Figure 1. Hard to see basin boundaries. Make darker lines.

Yes, we have revised figure 1 accordingly.

Figure 2. Really hard to tell the lines apart. Especially important to see BA1 and BA2. Can't tell the two lines apart in bottom panel (symbols too small).

Yes, we have also revised figure 2 with additional data and larger symbols.

Figure 4. Points are very small so hard to tell them apart. So it is hard to make sense of what is happening. Not clear what points are (observed). Make points bigger. Add the seasonal demarcations so can tell evaluate result about high retention during spring, etc.

We have also updated Figure 4.

Figure 5. Uptake should be in positive units.

Yes - revised.

Reviewer #2

GENERAL COMMENTS:
This study evaluates nitrate and dissolved organic nitrogen removal along the river network of a boreal catchment in Sweden that has been altered by forest harvests. This is an important scientific question given current forestry practices in boreal regions and predictions of increased forest use in the near future.
The manuscript is generally well written and within the scope of the journal Biogeosciences. However, there are some issues that could be addressed to improve the manuscript.

Thank you for your review and for agreeing with us, that this manuscript is well in the focus of Biogeosciences. We will do our best to address the concerns and to improve the manuscript.

- The model used to estimate N removal would benefit from estimates of uncertainty. The general assumptions made are considerable. Some apparently smaller assumptions like using the average concentration of CC-4 and NO-5 to calculate Charvest seem dangerous without considering measurements of uncertainty or running different scenarios. Moreover, the model could be better explained to the reader. A figure may be helpful in this sense. For instance, it is unclear how dilution is accounted for. It seems that nitrate removal efficiency should be calculated with the flux rather than with the concentration. The area of stream network used to calculate U should be reported.

Whereas we generally agree with the statement that the model would benefit from uncertainty estimations, we would like to clarify that the model as such is not any type of model for which a 'fitting' is performed, but only a mass balance of the measured data. This means that typical tools to evaluate the uncertainty of model parameterizations (such as for example, GLUE, Beven (2008)), cannot be used, as there are no free model parameters to choose.

Instead the model uncertainty will be present in three forms. First, structural model uncertainty (i), that is, if the mechanisms that are assumed (such as conservative mixing, closed mass-balance etc.) are reasonable. Second, there is uncertainty relating to the spatio-temporal representativeness of the input datasets (ii). Third, there is the general data uncertainty (iii), such as uncertainties in Q estimates and chemical analysis.

We argue that the uncertainties of (i) are small, which appears to find also agreement by reviewer #2. This corresponds also to follow Occam's razor and is further our model hypothesis, as we could

reject the model, if the model would have not performed well for conservative tracers (see comment to reviewer #1). Similarly, we argue that the rather extensive dataset and the replicated character of the sampling (two end members were sampled per treatment, data from 2004-2012 which will allow for some time for space substitution) will minimize the uncertainties of (iii).

Thus, what likely remains as the largest driver of model uncertainty, and in our case also of the conclusions drawn, is the uncertainty related to (ii), also explicitly criticized by reviewer #1 and #3. As we stated in our extensive response to reviewer #1, we have developed the assumptions for the clear-cut loads on what we considered the most robust approach, given the available data. As we also pointed out, the use of different scenarios is difficult. Instead we would like to include the graphs for Si and Cl mixing to argue for the validity of our model assumptions. We hope this comment addressed these concerns regarding model uncertainty.

Also, to clarify here: All calculations are done as mass fluxes, that is, concentration times discharge, as explicitly stated in the methods section. This means, that simple effects such as dilution by higher runoff from CCs will be accounted for in the model.

- Given the availability of nitrate, nitrite and ammonium data and the fact that ammonium seems to be almost as important as the other forms, I suggest that the authors redo the their calculations to estimate the dynamics and removal of dissolved inorganic nitrogen (DIN) rather than nitrate. In this way, the manuscript would cover all dissolved nitrogen forms (inorganic=DIN and organic=DON), which according to the manuscript also represent most exported nitrogen because particulate nitrogen seems very low in these streams. Moreover, there will be no need of speculation on the processes that convert nitrate to ammonium or vice versa.

This is a very good and reasonable point. We have revised our calculations so that we now calculate the fluxes and removal of the DIN pools rather than just NO3. These new results will be incorporated into the revised manuscript.

- The study design seems not justified well enough for the objectives of the manuscript. For instance, it is unclear why those catchments were chosen and why the catchments were differently harvested. The authors should explain it more clearly.

There a few paired catchment studies in boreal regions and, in fact, to the best of our knowledge non that would provide and as detailed, multi-year record of water chemistry that is needed to close the mass balance so that DIN removal can be calculated in the way we do it here. The differences in the harvests, the choices of sampling locations etc. were mainly of practical matters (Schelker et al., 2012, Schelker et al., 2013a). Also, the study design was not only designed for the purpose of this very study and is by far not perfect. We will try to address this comment in an additional sentence or two.

Furthermore, the interested reader will find several references in the methods section (Löfgren et al., 2009, Schelker et al., 2013b) that describe the Balsjö-experiment in more detail. Thus we see little need for changes here.

- The discussion seems too speculative in some parts, especially when it refers to processes and mechanisms that have not been measured in this study to explain some of the observed patterns. The authors could tone down some sentences.

We assume the reviewer points towards some of the points within the discussion also raised by reviewer #1. If not, then we would be more than happy to get to know which additional sentences specifically need to be toned down.

- The use of some terms is confusing. The authors should clearly define the terms chosen and then use them consistently throughput the manuscript. For instance, the authors should clearly define what they consider the “stream network”, and then use terms like “in-stream”, “riparian”, “landscape” consistently. Another confusing use of terms occurs when using words like “uptake”, “removal”, “retention”, etc.

Thanks for pointing this out. We will do our best to be more consistent on these.

- The title could be improved to reflect more clearly the contents of the paper. It seems too long and confusing.

We did try to use the best title we could come up with. The current title is precise in i) describing the location, climatic region and ecosystem where the study is performed, ii) that the study will present the results of a harvest experiment and iii) what the key results of the analysis are, that is, the removal of NO3, but downstream transport of DON.

We find this title to be a good choice; however, we would be more than grateful to receive further suggestions on how the title can be further improved. For the moment, and as no additional advice on how to improve it is given, we suggest to keep it as is. Also, the title has not been criticized by reviewer #1 or #3.

As in the response to Reviewer #1, we state that we will revise the points listed as specific comments below within our revised manuscript.

SPECIFIC COMMENTS:

P12062 L17: “Landscape” here means river network or the whole catchment (including terrestrial ecosystems). Please clarify.

This is, in fact, not that easy to know. All we know is what approximately enters the stream and doesn’t make it to the outlet. However, we revised to ‘river network’.

L21: “Net removal” within the river network? Please specify.

In the network – revised.

L22-25: Unclear sentence. Especially the part that says “capacity and limitation of N-limited . . .” Please rephrase.

Yes. We revised the entire abstract again, including the last sentence. We gave it a clearer massage now.

P12063 L17: Some studies have. You could cite here Bernhardt et al. 2003, Riscassi and Scanlon 2009, etc. L18: I suggest adding “stream” or “river” before “network.

Revised to ‘ and few studies have explored how’… and cite Bernhard et al, 2003

P12065 L4: I think that the N limitation issue could be mentioned earlier in the introduction. Moreover, its consequences for this particular study should be explained. A hypothesis may emerge from here. L3-18: I miss some hypotheses and predictions here.

As experienced readers will know that boreal ecosystems are commonly N-limited (Vitousek & Howarth, 1991), we believe that we describe the N-limitation at a suitable location. Thus we argue to keep this as is. The implications are simple. Uptake may be higher and leaching lower than in P limited ecosystems – all this is covered in the manuscript.

P12066 L1: Why was the riparian buffer left intact in this catchment and not in the other?

This is part of the previous work done at this field site. As there are >15 publications from this site, we suggest the reader to follow some of the referenced works, such as the AMBIO special issue from 2009 (Löfgren et al., 2009) that we also cite at the beginning of this paragraph.

L7: It seems quite strange that the samples were analyzed unfiltered. Why? Did you make some tests to see the influence of not filtering on your DIN and DON estimates?

As we point out in the manuscript, most studies in the region use this assumption, as the particulate fractions are simply so low (Laudon et al., 2011). Saves time and effort.

P12068 L24: I understand that the efficiency can be set to zero but a negative value may also mean in-stream release of NO3 (i.e. negative U values).

This is correct. The main reason for U to be set to zero is that this mainly occurs in winter, when the flows are very low and the measurement of Q is often uncertain as it is affected by ice (up to 30-50cm of ice cover in mid winter are common in these streams). Then a built up of pressure under the ice can 'give' a slightly higher flow upstream than downstream, even if this is not really the case. In our model this would be a negative flux, also resulting in a negative Er value. For these occasions, we argue, it is better to set U to zero, rather trying to interpret uncertain data.

P12069 L16-19: It would be nice to see these different seasons depicted on the figures. This would allow the reader to follow results more easily.

We revised the figure, but found little space to add the seasons.

P12070 L9-17: The scale of the figure does not allow seeing most of the described patterns.

Yes, we have revised it.

P12071 L3-6: Why are U values negative? Net uptake values are usually positive if there is net uptake and negative if there is net release. I suggest changing it.

Yes, see also comment to reviewer # 1.

P12072 L8-16: Confusing paragraph. The supplementary figure is quite unclear and there is no figure legend or number. Unclear what is meant by upstream and downstream here and what the purpose of this paragraph is.

There was in fact a problem uploading the figure caption for the supplementary figure that explains what upstream and downstream is in this case. We will try to add the caption this time.

P12073 L11: Change to "zero or near-zero". L17: Did you try correlations with variables other than discharge?

Yes, we did some preliminary analysis with RAD (as a proxy for PAR), water Temp. and some DOM-quality indices derived from the Absorption spectrum. None of these variables gave any clear explanatory patterns, so we decided to not include these efforts. We may well be data limited for such an analysis.

L29: It would be interesting to see and integrated U for the whole year (in kgN) that could be compared to other variables in Table 1.

This is exactly what the difference between the modelled and the measured values in Table 1 (now Table 2) gives. U is calculated from the difference between the model and the measured solute fluxes, similar to all modelled concentrations.

P12075 L10-15: The effect of DNRA seems quite irrelevant here. I suggest removing these lines.

We agree, removed.

P12076 L14-19: These conclusions are ok, but they do not refer to consequences on stream network (in-stream) N removal.

Yes, this is correct. Within these last lines, we tried to apply the findings of the study not only to the basic research question of N cycling, but to also derive some suggestions for practical forest management. Thus we added these statements, which we believe are well enough founded on our results.

Fig. 2: In the first panel it is not possible to see the temporal trends of the sites other than CC-4. Maybe you could try to use a log scale or to add a new panel/figure. What does "estimated Q" in the second panel mean? Please explain.

We have revised the scale of the x-axis and added more data to this figure.

Fig.5: Strange to see U values as negative values. Also, I do not see the pairs of letters mentioned in the figure legend.¨

We have also revised this figure.

Anonymous Referee #3

General Comments

This is an interesting paper focused on how forest disturbances impact on stream water chemistry in boreal regions. The topic is relevant and the study fits perfectly within the scope of Biogeosciences. In general, the paper reads well and the introduction is well framed. I miss some information in the Study Site section such as the areas of the experimental catchments which can be useful to the reader for doing some back of the envelope calculations. The Methods section needs some extra work. The results are supported by a quite large amount of field and satellite data, though there are some results that need to be worked further. Rather than reporting patterns exclusively, the authors have brought the paper to a higher level by adopting a quantitative approach, which I mostly like. The discussion includes some results than need to be moved earlier in the text. Overall, I have some concerns with the applied mixing model in its present form. There are few other major issues that the authors need to solve before the paper can be published.

Thank you for your review. We hope to be able to address all the concerns and revise the manuscript accordingly.

The authors focus their study on nitrate because they argue that forest harvesting increase the mobilization of inorganic nitrogen, primarily nitrate. However, they indicate (in the discussion section) that the contribution of ammonium to the total inorganic pool in stream water is pretty high (from 20 to >50%). Therefore, by modeling only nitrate concentrations, the authors may be missing an important piece of information. I recommend showing more clearly nitrate and ammonium concentrations for the two periods of study (2004-2006 and 2007-2012). If changes in ammonium concentrations are small between the two periods, this would support the approach considered by the authors. Yet, if ammonium concentrations change substantially between the pre-harvest and post-harvest period, the authors should consider the possibility of calculating the mixing model for DIN rather than for nitrate to get a more complete picture of how forest disturbances translate downstream.

Yes, we have followed this advice and would like to provide the mass balance model for DIN, instead of only for NO3. Also, Figure 2 was revised to explicitly show NO3, NH4 and DON concentrations.

One of the major issues the authors need to deal with is the uncertainty associated with the mixing model calculations because the response to clear-cut differed tremendously between the CC4 and NO5 catchments. This issue cannot be overlooked by the authors and requires careful consideration. For instance, the concentration of the clear-cut end member (C_harvest) is characterized by averaging nitrate concentration for CC4 and NO5. Yet, results and conclusions could differ markedly from the ones presented here if authors would have used nitrate concentrations either from CC4 or NO5 alone. According to the authors, the distinct response between these two catchments may relay on the fact that riparian strips were kept in NO5 but not in CC4. If "leaving small (5-10 m) buffer zones along headwater streams is common practice" (12066.4), then one would expect that, on average, the mean response of the whole harvested area would be closer to NO5 than to CC4, being the later a more extreme scenario (savage clear-cutting without protecting riparian areas). By using the average of the two clear-cut catchments, the authors may be magnifying the "forest derived nitrate" and consequently the nitrate removal efficiency (Er) that is potentially attributed to in-stream processing.

Please find our earlier reply to reviewer #1s comment on this issue.

Another issue that the authors need to address is the implicit assumption that chemistry for the clear-cut and control end members is representative of the water draining through the whole harvested and uncut area within BA2 and BA1. Or in other words, that the chemical signature of groundwater entering to the stream outside the experimental catchments is similar to the stream water chemistry of the end members. I understand that this assumption is needed for applying the proposed mixing model, but the authors need to include this assumption explicitly in the paper and discuss the advantages and limitations of their approach.

That is an interesting point. First of all, it should be noted that these catchments are underlain by highly compacted till layers that have generally low hydraulic conductivities. Runoff generation is thus primarily from shallow saturated soil water entering streams laterally (Bishop et al., 2004, Bishop et al., 2011). Thus, and in contrast to other stream systems, contributions of deeper GW are considered minor, at least at the given spatial scale of this third order stream network.

The typical characteristics of deep GW from the underlying granitic bedrock in the region is that it is essentially free of nitrogen (DON, NO3, NO2, NH4; all normally all below detection limit). This is likely the result of a very low population density combined with a low pressure land use of forestry (for example in comparison to agriculture). Thus it appears reasonable to ask, if not deep GW inputs could have diluted the stream water causing an effect that would then be (mis-)interpreted as high NO3 removal. Such a dilution would be most likely found between the sites BA-2 and BA-1, as the small lake may be a location of GW upwelling. However, whereas such a mechanism appears generally plausible, there is little evidence for this. For example, it is very likely that the concentrations of the two conservative tracers Cl and Si (presented in response to reviewer #1), would have different concentrations in GW as in the surface water. As a result, systematic derivations of the results of the performed mixing models from the measured concentrations would occur. However, as we did not observe such derivations, we concluded that deep GW plays a minor role in modifying the water chemistry in this small stream network

Assuming a minor, negligible role of GW in the Balsjö catchment is also in agreement with our previous work (Schelker et al., 2014), as well as other work from the region that has evaluated this question in the face of DOC concentrations and found little GW influence in the till dominated regions above the highest coast line (Tiwari et al., 2014), but a stronger influence further downstream.

We have now also added this information to the site description.

The interpretation of the modelled results should be explained in the Methods section rather than in the captions of the Figures. For instance, explain how the differences between modeled vs measured concentrations were interpreted, or the reasoning of why Er and Q should be or should not be related to each other.

Yes, we have moved the explanation for the modelled vs. measured plots to the methods section and deleted the Er to Q explanation as it was not the right place.

Be consistent with the presentation of Figures and add letters to identify the different panels. The second panel in Figure 2 is not referred anywhere and it is not clear what the author mean by estimated and measured Q.

We have revised Figure 2. Also, we corrected the 'estimated and measured Q' issue.

Figure 3a and Figure 2b are redundant.

That is correct. The motivation to plot Q here again was to allow for quick comparisons of different Q conditions and Er.

However, we removed it now from the revised figure 3 (now figure 4).

The results in Table 1 are not included in the results section.

We are now referring to Table 2 (former table 1) in the results section of our revised manuscript.

Specific Comments

Introduction
12064.24 clarify what you mean by "these relationships"

We revised the sentence.

12065.13-18. The "questions" proposed by the authors are somehow interrelated because questions (i) and (ii) are focused on patterns, while (iii) refers to the involved processes and mechanism which lead to those observed patterns. Thus, I suggest some rewording for improving the strength of this final introductory paragraph.

We agree that these questions are interrelated but they do essentially reflect the goals of the study. We streamlined the text here so that there are now only two components – one addressing 'patterns' and the other addressing 'potential mechanisms'.

Methods
12066.20. Include some more quantitative information about the areas that were harvested within the different studied catchments.

Yes, we did. See new table 1.

12066.24 Include for which catchments C_modelled was calculated.

Yes, we revised: 'The concentration at the downstream locations BA-1 and BA-2 (C_modelled, in µg L-1) for each time…'

12065.24. Include drainage area for the 4 experimental catchments.

They are now presented in the new table 1.

12066.6. Indicate that water samples were also analyzed for chloride and silica (hydrological tracers) and that results on that were reported in a previous study (see later comment).

We added that samples were analyzed for these solutes. Also, we added the results to support our choice of model assumptions to the methods section.

12067.3-10. This info could be partially moved to the Study Site section; focus this section on the description of the mixing model.

This is the model description. We would like to keep it as is.

12067.17 According to eq. 1 “percentage” should be “fraction” and units would be “over 1” rather than in “%”. Otherwise the factor 100 should be included in eq. 1

Absolutely right - thanks for pointing this out. We revised it.

12067.25. The response to clear-cut differed tremendously between the CC4 and NO5 catchments. Thus, there is a substantial uncertainty associated to these calculations. There are several possibilities to deal with this problem. For instance, the author could consider either an upper and lower limit for C_modelled or different harvest scenarios (with and without keeping riparian strips).

Please see our earlier reply to reviewer #1 on this specific assumption.

12067.26. “. . .each scaled to 100% harvest using a scaling equation” Why the authors expect that C_harvest will increase linearly with increasing the harvest area (eq 2)? And by how much the results obtained would change if another ecosystem response (e.g. asymptotic) would be considered? The reasons behind this assumption are not clear, especially when reading later in the text that Q_harvest may not change substantially between a catchment harvested 88% or 100% (12068.12).

The assumption of a linear increase of C_harvest is of course critical, but needs to be made to be able to apply the model. Also there are at least some ‘good indications’ that suggest that this assumption is reasonable.

First, the concentrations of DIN of CC-4 and NO-5 scale linearly (Figure 7), if one plots them after the first of January of 2009, that is, after the harvest effects have stabilized a bit. To us this is suggestive that an increase in the harvested area will also cause an increase in DIN concentrations.

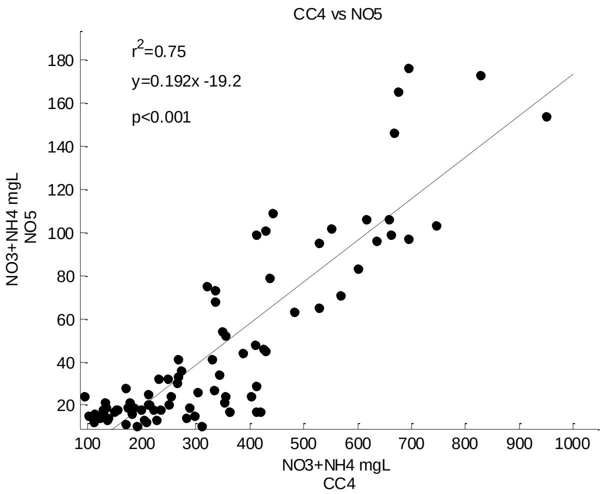


Figure 7, DIN concentrations of CC-4 vs. NO-5.

Second, the response of other solutes, such as DOC has been done using this very assumption in the past (Laudon et al., 2009, Schelker et al., 2012). Furthermore, other work on DOC concentrations along all the available Balsjö first-order streams also show a steady linear increase of the mean concentrations with increasing percentage harvesting (Schelker et al., 2014), at least for the range up to ~60% harvest.

Also, one should remember that the total percentage of the catchment area that is harvested within the catchments of the downstream sites BA-1 and BA-2 that are modelled within this study do not exceed 12% and 18% respectively (see new table 1 of the manuscript). We argue therefore that the assumption is reasonable.

Please also note, that we now state that we use this assumption explicitly.

12068.3 not clear what the authors mean by “reciprocal”.

We revised to ‘was defined as dj = 1/ Aj, that is the reciprocal…’. What a reciprocal is, is well defined in maths.

12068.4. Similar to C_harvested, the authors should consider some sort of confidence interval when characterizing the concentration of the control end member (C_control).

The variation of the control end member will be small as compared to the other variation.

12068.19. According to the results presented Er was calculated the other way around: (modeled – measured)/modeled.

This is correct. We corrected it.

12068.23. Values of Er < 0 could be indicating either in-stream nitrate release and/or groundwater inputs with higher nitrate concentrations than stream water. This information could be useful for discussing some of the obtained results. I recommend further considering this variable when working on the revised version of the mp.

As pointed out in one of the previous comments, there is very little nitrogen in GW in this region, commonly all below the detection limit.

12068.25. From here on, this info does not relate to the “Mixing Model”. Add a new subsection.

In fact, they do. To clarify we changed ‘calculated’ to ‘modelled’. However, we added a subsection of on ‘other calcualtions’.

12069.14-16. By doing so, the authors are also assuming that stream water chemistry for the clear-cut and harvest end members is representative of the water draining through the whole harvested and uncut area within BA2 and BA1. Or in other words, that there may be no longitudinal changes in groundwater chemistry entering to the stream. Is this assumption reasonable? Do the authors have some additional data throughout the basin area to support this assumption? Could changes in groundwater inputs along the stream partially explain the observed patterns?

As pointed out before, we found no direct evidence so far, that there is an important role of GW for water chemistry in this rather small stream network. This assumption may quickly need to be revised, if one moves further downstream, where deeper lakes and larger streams are included in the stream network.

We have now also added this aspect to the discussion section [L. 1459ff]

Results

12069.20 To improve the flow of this section, results could be divided in two subsections, one describing measured concentrations and fluxes; the other with the model results.

Yes, we created two subsections, one on the measured results and one on the modelled.

12070.15 This temporal pattern was also exhibited by CC4 but not for RS3 (as far as I can distinguish from the graph). You could reorganize these results in two paragraphs: the first focused on changes in concentration between 2004-2006 and 2007-2012 and the second focused on seasonal patterns.

Yes, we added this to the sentence.

12071.3-6. If U is the difference between modeled and measured fluxes, in-stream net areal uptake rates should be positive throughout the text. This would have more sense, since stream ecologists usually considered U> 0 when there is actually net nutrient uptake by stream biota.

Good point – we corrected this.

12071.3-6. Were the Us obtained for BA1 similar to those for BA2? And if not, why the bioreactive capacity of this stream may change along the longitudinal axis? The discussion of the paper would benefit if showing these results more clearly.

We are now presenting U values (for DIN) for both locations in the results section. However, one should keep in mind that these aquatic ecosystems generally have very low DIN concentrations and are likely often limited by DIN. As our U values are derived from the mixing model, they can be substantially lower between sites, simply because there wasn't more DIN available to be taken up. Longitudinal changes in the bioreactive capacity can thus not be evaluated by our net-uptake approach, as all we can essentially say is: 'Uptake was not higher than the U values, but could well be so, if more DIN would be available'.

Discussion
12071.8-17. The authors are right in that the marked response in CC4 was not observed downstream. Yet, it will be interesting to highlight the differential response exhibited by the two harvested catchments, especially because if riparian areas are usually protected against clear-cut, the response observed for CC4 may not be widespread.

Please see our previous response to reviewer #1. One may also consider our updated Figure 2 of the manuscript that now provides a better possibility for readers to visually examine changes of DIN and DON concentrations at BA-1 and BA-2 as a response to harvests.

12071.13 The results contained in Table 1 should have been introduced in the earlier section.

We do so now.

12071.18-24. These are results and should be moved to the earlier section.

Yes, we moved them.

12071.23. Clarify to which season you refer when saying 54 and 46%.

Thanks – we corrected this.

1207120-24. The contribution of NH4 to the total inorganic N pool is quite substantial, and thus, the authors could be underestimating the potential of in-stream processes to retain and transform DIN in these catchments. I wonder how different the results would be if considering DIN rather than nitrate alone. Could the authors provide some insights on that?

As stated before, we are now running the mass balance model for DIN.

12072.4. In this case, it may be clearer to refer to the years comprising the two periods than to “pre-treatment vs treatment”.

Yes, this is better.

12072.8-10. These are results; moved them to the earlier section.

We would like to keep them where they are, as the results as such are presented before, but need to be stated here again to go into the interpretation.

12072.11-16. What about NR7 and NO5? Did they show similar seasonal patterns than BA1, BA2? And if so, could one still say that “enhanced upstream inputs of nitrate in headwaters are translated downstream during the dormant season”?

We believe the revised figure 2 (now figure 3) makes this point more clear to the reader. So the answer is yes.

12072.21-22. I recommend to briefly comment on that already in the methods section. The good match between measured and modeled concentrations for hydrological tracers would give consistency to the mixing model. Note, however, that the fact that the model works well for chloride but not for nitrate, does not necessarily imply in-stream nitrate retention because groundwater entering downstream the experimental catchments could have similar chloride concentrations but different nitrate/ammonia concentration than groundwater upstream.

Good point. It is absolutely right, that the model could even converge on one conservative tracer, but still be conceptually wrong. However, as we have now added a graph showing the model performance of two conservative tracers, the chances for the model conceptualization to be wrong are rather small.

See newly added Figure 2. Also, we acknowledge the possible role of GW as a DIN sink in the discussion [L. 1459ff]

12073.11-13. or that the concentration of nitrate was higher in downstream groundwater inputs.

See earlier comments on low GW NO3 concentrations.

12073.16-23. Avoid repeating results or adding new results in the discussion section.

Yes, thank you – we will revise accordingly.

12073.21-23. The authors should be cautious and take into consideration the uncertainties associated to these calculations before claiming that ca.70% of the nitrate inputs were removed. I suspect this figure is far too big, likely because the actual approach magnifies the effect of CC4.

Please see earlier comment on the representativeness of the CC end member. According to our understanding an 'over magnification' would have been given, if we would have only used CC-4 as the sole definition of C_harvest.

One may also add here, that our revised calculations now include the entire DIN pool. The modelled removal is then already a bit lower with values around ~65%. This appears reasonable, given that the streams are N-limited during most of the year.
Also, we have added a short section to the discussion where we discuss these assumptions and their implications [L. 1429ff]

12073.24-29. According to Table1, the decrease in nitrate loads between BA2 and BA1 was <30%, which is a much lower number than the 70% proposed. Thus, and assuming that all nitrate retention was occurring within the stream channel and that there were no differences in groundwater inputs between BA2 and BA1, U values for this stream reach would be several times lower than 6 microg N/m2/min. How would the authors explain this shift in the in-stream bioreactive capacity along the stream?

We do not assume any change in bioreactive capacity along the stream, but simply close the mass-balance. Also, not all harvest are located upstream of BA-2, but some are also entering the network between BA-2 and BA-1 (see Figure 1 of the manuscript). May this be a reason for the inconsistency?

Overall we may add, that our additional table 1 will allow the interested reader to be able to make some back-of the envelope calculations with our data.

12074.11-26. These changes induced by forest harvest may be occurring only within the CC4 that (i) occupies a relatively small area of the BA2 and BA1 catchment and (ii) showed tremendous increases in nitrate concentrations. Thus I don't see how this explanation applies for patterns in BA2 and BA1.

Please find our previous comment on the representativeness of the CC end member.

12075.10-15. Too speculative. A more systematic analysis of the ammonium time data series will provide a clearer picture of whether seasonal changes in ammonium are terrestrially or stream derived. A table including ammonium and nitrate concentrations for the two periods could be useful.

We have added these concentrations to the revised version of Figure 2. We hope this allows the reader to evaluate this data.

12076.11. Not clear to which "two mentioned measures" the authors refer.

This needs clarification then also.

Revised.

Figures and Tables

Figure 2. The second panel is not referred within the main text. Based on the Methods section it is not clear what the authors mean by estimated vs measured Q.

We have revised the Figure, see above.

Figure 3. Differences between modelled and measured concentrations could indicate biogeochemical retention of the solute during transport downstream but also hydro-logical mixing with sources with different chemical signature. I recommend including the interpretation of the results in the Methods section where the authors can link the expected patterns to the assumptions underlying the model.

Yes, we have revised this.

Figure 4. Panel (A) is not introduced in the main text and it is redundant with Figure 2. If Er >0 means nitrate retention, then the differences is between modeled and measured concentrations nitrate concentration. Why did the authors explore the dependency of Er on Q? This should be explained in the Methods section.

We removed panel A.

Figure 5. Positivize U values. To avoid any confusion to the reader, highlight in the caption that this is a potential maximum value for in-stream uptake. The letters for statistical significance are not included in the figure. Show data for BA1; differences in U values between BA2 and BA1 can enrich the discussion by supporting (or not) the explanations given for in-stream nitrogen processing.

Yes, we revised the figure, with positive uptake numbers and added the letters. U values for both sites are now given in the results section, but as we pointed out in a previous comment, U Values result only form the mass balance and can not be interpreted as 'bioreactive potential'.

Table 1. This table and results therein should be included in the results section.

Done so.

Figure SS1. Please include the caption of this figure. Include data for the snowmelt period to be consistent with the data analysis throughout the mp.

Somehow the Figure caption got lost for this one. We will try to upload it correctly this time.

We will do our best to also address the comments below in our revised version.

Technical Corrections

12064.18. "photoautotrophic" rather than "autotrophic"

Yes, revised.

12067.7 or 2001-2011? (as in caption figure 1)

Yes, revised.

12068.26 Change “net uptake rates” by “net areal uptake rates” throughout the mp.

Yes, revised.

12069.7. change “treatment” by “clear-cut”.

Revised.

12069.15. Change “loss” by “export”.

Revised.

12069.24. Delete “buffer”

Revised.

12070.1 and 4. Which treatment? Clarify.

Revised.

12070.23. Change “nearly exclusively” by “usually”.

Yes, reads much better.

12072.4. Delete the “-“ sign.

Yes.

References

Schelker J, Öhman K, Löfgren S, Laudon H (2014) Scaling of increased dissolved organic carbon inputs by forest clear-cutting – What arrives downstream? Journal of Hydrology, 508, 299-306.

Tiwari T, Laudon H, Beven K, Ågren AM (2014) Downstream changes in DOC: Inferring contributions in the face of model uncertainties. Water Resources Research, n/a-n/a.

Schelker J, Grabs T, Bishop K, Laudon H (2013a) Drivers of increased organic carbon concentrations in stream water following forest disturbance: Separating effects of changes in flow pathways and soil warming. Journal of Geophysical Research: Biogeosciences, 118, 2013JG002309.

Schelker J, Kuglerová L, Eklöf K, Bishop K, Laudon H (2013b) Hydrological effects of clear-cutting in a boreal forest – Snowpack dynamics, snowmelt and streamflow responses. Journal of Hydrology, 484, 105-114.

Schelker J, Eklöf K, Bishop K, Laudon H (2012) Effects of forestry operations on dissolved organic carbon concentrations and export in boreal first-order streams. Journal of Geophysical Research, 117, G01011.

Bishop K, Seibert J, Nyberg L, Rodhe A (2011) Water storage in a till catchment. II: Implications of transmissivity feedback for flow paths and turnover times. Hydrological Processes, 25, 3950-3959.
Laudon H, Berggren M, Agren A et al. (2011) Patterns and Dynamics of Dissolved Organic Carbon (DOC) in Boreal Streams: The Role of Processes, Connectivity, and Scaling. Ecosystems, 14, 880-893.
Laudon H, Hedtjärn J, Schelker J, Bishop K, Sørensen R, Ågren A (2009) Response of Dissolved Organic Carbon following Forest Harvesting in a Boreal Forest. Ambio, 38, 381-386.
Löfgren S, Ring E, Von Brömssen C, Sørensen R, Högbom L (2009) Short-term Effects of Clear-cutting on the Water Chemistry of Two Boreal Streams in Northern Sweden: A Paired Catchment Study. Ambio, 38, 347-356.
Beven K (2008) Environmental Modelling: An Uncertain Future, Routledge: London.
Roberts BJ, Mulholland PJ (2007) In-stream biotic control on nutrient biogeochemistry in a forested stream, West Fork of Walker Branch. Journal of Geophysical Research: Biogeosciences (2005–2012), 112.
Bishop K, Seibert J, Köhler S, Laudon H (2004) Resolving the Double Paradox of rapidly mobilized old water with highly variable responses in runoff chemistry. Hydrological Processes, 18, 185-189.
Vitousek P, Howarth R (1991) Nitrogen limitation on land and in the sea: How can it occur? Biogeochemistry, 13, 87-115.

Title:

Nitrogen export from a boreal stream network following forest harvesting: Seasonal nitrate removal and conservative export of organic forms

Running title: Nitrogen export from a boreal stream network

Authors: Schelker, Jakob[1,2*]; Sponseller, Ryan[3]; Ring, Eva[4]; Högbom, Lars[4]; Löfgren, Stefan[5] and Laudon, Hjalmar[2]

Affiliations:

[1] Department of Limnology and Bio-Oceanography, University of Vienna, Vienna Austria.
[2] Department of Forest Ecology and Management, Swedish University of Agricultural Sciences, Umeå, Sweden.
[3] Department of Ecology and Environmental Science, Umeå University, Umeå Sweden.
[4] The Forestry Research Institute of Sweden (Skogforsk), Uppsala, Sweden
[5] Department of Aquatic Sciences and Assessment, Swedish University of Agricultural Sciences, Uppsala, Sweden.

* Corresponding Author:

Jakob Schelker, Department of Limnology and Bio-Oceanography,
University of Vienna, Althanstrasse 14, 1090, Vienna, Austria
email: jakob.schelker@univie.ac.at
mobile: (+43) 660 321 5081

Format: Primary research article

Keywords: Nitrate, Organic Nitrogen, Nitrogen Retention, In-Stream Processing, Denitrification, Boreal Forest, Forest Disturbance, Clear-Cutting, Scaling.

Abstract

Clear-cutting is today the primary driver of large scale forest disturbance in boreal regions of Fennoscandia. -Among the major environmental ~~impacts~~ concerns of ~~forest harvesting~~this practice for surface waters is the increased mobilization of ~~inorganic~~nutrients, such as inorganic nitrogen (DIN)~~, primarily as nitrate (NO_3^-)~~ into streams ~~surface waters~~. But ~~whereas~~ while ~~NO_3^-~~DIN ~~inputs~~ loading to first-order streams following forest harvest has~~ve~~ been previously described, ~~their~~ the downstream fate and impact of these inputs is not well understood. We evaluated the downstream fate of DIN and dissolved organic nitrogen (DON) inputs in a boreal landscape that has been altered by forest harvests over a 10 year period ~~to estimate the effects of multiple clear-cuts on aquatic N export~~. ~~in a boreal stream network.~~ The ~~S~~small first-order streams ~~showed~~ indicated substantial leaching of DIN, primarily as nitrate (NO_3^-)~~NO_3^-~~ in response to harvests with NO_3^- concentrations increasing by ~15 fold. NO_3^- concentrations at two sampling stations further downstream in the network were strongly seasonal and increased significantly in response to harvesting at the ~~medium~~ mid-sized, but not at the larger stream. DIN ~~Nitrate~~ removal efficiency, E_r, calculated as the percentage of 'forestry derived' ~~NO_3^-~~DIN that was retained within the stream network ~~landscape using~~based on a mass balance model was highest during the snow melt season followed by the growing season, but declined continuously throughout the dormant season. In contrast, export of ~~organic~~ DON from the landscape indicated little removal and was essentially conservative. Overall, net removal of ~~NO_3^-~~DIN ~~in the stream network~~ between 2008 and 2011 accounted for ~ ~~70~~65% of the total DIN~~NO_3^-~~ mass exported from harvested patches distributed across the landscape. These results highlight the capacity ~~and limitation~~ of N-limited ~~terrestrial and aquatic ecosystems~~boreal stream networks to buffer ~~inorganic N~~DIN mobilization that arises from multiple clear-cuts within this landscape. ~~meso-scale boreal watersheds.~~ Further, these findings shed light on the potential impact of anticipated measures

to increase forest yields of boreal forests, such as increased fertilization and shorter forest ~~rotations that~~rotations, which may ~~enhance~~increase the pressure on boreal surface waters in the future.

1. Introduction

Decades of research have shown that disturbance of forest ecosystems can lead to increased losses of inorganic nitrogen (N) from land (Vitousek et al., 1979; Likens and Bormann, 1995; Aber et al., 2002; Houlton et al., 2003), with potentially negative consequences for water quality in streams and rivers (Martin et al., 2000). Perhaps the clearest demonstrations of how forest disturbance influences terrestrial nutrient mobilization have used experimental harvests in small catchments to document changes in stream chemistry relative to undisturbed controls (Likens et al., 1970; Swank and Vose, 1997). While the magnitude and duration of response to harvest varies among studies (Binkley and Brown, 1993; Kreutzweiser et al., 2008), most have documented increases in stream-water nitrate (NO_3^-) concentrations. Such responses reflect the loss of plant nutrient demand (Boring et al., 1981), accelerated rates of soil N mineralization and nitrification (Holmes and Zak, 1999), and increases in hydrologic flux within the catchment (Hornbeck et al., 1997; Andréassian, 2004). By design, the majority of this research has addressed responses to forest disturbance at small spatial scales (e.g., catchments of first-order streams) ~~but~~ and few studies have explored how localized increases in nutrient concentration are translated downstream within fluvial networks (Bernhardt et al., 2003).

Whereas several recent studies have addressed the removal of inorganic N ~~at the~~within river networks ~~scale~~ (Helton et al., 2011; Wollheim et al., 2006; Worrall et al., 2012; Alexander et al., 2009), little has been done to investigate these processes ~~specific effects of forestry on nitrogen cycling~~ in boreal ~~stream networks~~landscapes subject to widespread and active forest management. A clearer understanding of how the enrichment of headwater environments through forestry is expressed at larger spatial scales (Futter et al., 2010) is important if policy makers are to consider the broader biogeochemical implications of forest management.

The degree to which surplus NO_3^- derived from forest disturbance is delivered to downstream receiving systems is determined by the balance between hydrologic transport and biological demand within multiple habitats at the terrestrial-aquatic interface (McClain et al., 2003; Seitzinger et al., 2006). For example, when forest harvesting leaves riparian buffer zones intact, plant nutrient uptake, immobilization by soil heterotrophs, and denitrification in streamside habitats can together greatly reduce the delivery of NO_3^- to streams (Laurén et al., 2005). The efficiency of riparian NO_3^- removal varies among studies (Ranalli and Macalady, 2010; Weller et al., 2011), and is determined, in large part, by topographic and soil properties that influence the rates and efficacy of denitrification through effects on hydrologic transport (Ocampo et al., 2006), soil/sediment redox conditions (Pinay et al., 2000), and depth of groundwater flow-pathways relative to biogeochemically active soil layers (Vidon and Hill, 2004; Groffman et al., 2002). Riparian N retention efficiency, and the mechanisms responsible, may also vary in response to changes in plant demand (Sabater et al., 2000), availability of labile carbon (C) to soil and sediment microbes (Starr and Gillham, 1993) and hydrologic forcing during floods that overwhelms biotic potential (Hill, 1993).

Where forest harvests extend to channel margins, or when retention of NO_3^- in riparian buffer zones is poor, surplus NO_3^- derived from disturbance is delivered directly to streams. Rates of nutrient uptake in streams and hyporheic zones can be rapid (Mulholland et al., 2008) and ~~retention~~ uptake of NO_3^- in headwater environments may reduce watershed exports in response to forest disturbance (Bernhardt et al., 2003; Riscassi and Scanlon, 2009). NO_3^- removal in streams may be linked to ~~uptake by photoautotrophic organisms~~immobilization by autotrophic and heterotrophic microbes, as well as to denitrification in hyporheic sediments (Harvey et al., 2013; Mulholland et al., 2008). The efficiency of this NO_3^- removal (i.e., the percentage removed per unit stream length) is determined by the strength of this biological demand relative to nutrient availability (Mulholland et al., 2008), and is further constrained by hydrologic factors that govern residence times in biological active zones (Wollheim et al., 2006). ~~The result of these relationships is that~~As a result, removal efficiency tends to be lowest during periods of high flow and/or NO_3^- flux (Alexander et al., 2009; Scanlon et al., 2010). Biological activity and associated nutrient demand in streams is strongly influenced by a variety of habitat factors (e.g., incident light, temperature, and organic matter availability) that vary seasonally (Roberts and Mulholland, 2007; Valett et al., 2008). These factors are also modified by disturbance in the surrounding landscape (e.g., through loss of canopy cover), with the result that in-stream retention of excess NO_3^- may itself change in response to harvesting (Bernhardt et al., 2003; Sabater et al., 2000).

In this paper we explore the potential for fluvial networks to remove NO_3^- derived from forest harvesting in a boreal landscape in northern Sweden, where N limitation of terrestrial (Högberg et al., 2006) and aquatic (Jansson et al., 2001) productivity is common. We compiled 10 years of data on clear-cuts performed in this landscape with 8 years of temporally coinciding stream chemistry data from a third-order stream network. The network includes a replicated paired-catchment harvesting experiment in the headwaters, plus several additional harvests (Figure 1). Enhanced NO_3^- loading to headwater streams (first-order) as a result of forest clear-cutting has been reported previously for this site (Löfgren et al., 2009). Thus, the study design and history of research in this landscape provide a unique opportunity to explore the downstream implication of forest harvesting. We use a simple modeling approach to ask: i) whether and how NO_3^- exported from recent (<10 yr) clear-cuts influences ~~downstream~~ water chemistry downstream within the same drainage system, ~~ii) how the strength of upstream-downstream connections changes seasonally, and iii)~~and ii) to what degree downstream patterns in nutrient concentration arise from simple dilution of upstream inputs *versus* biological uptake and retention in stream and riparian habitats.

2. Methods

2.1 Study Site

This study was performed in the 'Balsjö paired-catchment experiment' located in the boreal forest of northern Sweden (N 64° 1'37'' E 18° 55'43'') (Löfgren et al., 2009). The experiment consists of four first-order streams of which two were clear-cut harvested (clear-cuts = CC-4 and NO-5; controls = RS-3 and NR-7) in 2006 and two third-order downstream sites of different size (BA-1, size =22.9 km^2 and BA-2, size = 8.9 km^2, Figure 1). Clear-cutting at CC-4 was carried out to the stream bank, whereas a small, ~10 m wide ~~at each stream side~~, discontinuous riparian buffer was left intact on both side of the stream at~~at~~ NO-5. All clear-cuts in the network were performed as final-fellings for commercial purposes following environmental considerations according to the Swedish Forestry Act, interpreted and applied by the forest owner. Thus leaving small (5-10 m) buffer zones along headwater streams is considered common practice. However, field observations also showed substantial disturbance of riparian zones by forestry machinery crossing streams and by wind throw within narrow stream corridors. Together these impacts likely limit the effect of the environmental considerations for nutrient retention.

The Balsjö catchment is underlain by highly compacted till layers that have generally low hydraulic conductivities. Runoff generation is thus primarily from shallow saturated soil water entering streams laterally (Bishop et al., 2004; Schelker et al., 2013a~~; Bishop et al., 2004~~) Thus, and in contrast to other stream systems, contributions from deep groundwater sources are thought to be minor at the spatial scale of this third order stream network (Schelker et al., 2014).

2.2 Stream water chemistry

Concentrations of NO_3^-~~-~~, ammonium (NH_4^+) and dissolved organic nitrogen (DON), chloride (Cl), and dissolved silica (Si) were determined from unfiltered stream water samples. As fractions of particulate organic matter are generally very low in this landscape (<0.6%; see Laudon et al., 2011) we consider samples to represent dissolved solute concentrations. Samples were collected between 2004 and 2012 at one to two week intervals during spring, summer, and fall, and at four week intervals during winter low flow. Samples were frozen within 1-2 days after collection and analyzed using colorimetric methods at a SWEDAC accredited laboratory according to method SS-EN ISO 13395:1996 for NO_3^- (sulphanil amid method after cadmium reduction), according to Bran & Luebbe Method G-171-96 Rev. 1 (Phenate method) for ~~ammonium (~~NH_4^+~~)~~, and method SS-EN 12260:2004 for total N (combustion to nitrous oxide followed by chemiluminescence detection) (Löfgren et al., 2009). Thus, reported concentrations of NO_3^- equal the sum of nitrate and nitrite expressed as mass of N (µg N L^{-1}); DIN concentrations were calculated as the sum of NO_3^- and NH_4^+; concentrations of ~~for~~ DON ~~were calculated~~ as total N minus ~~inorganic N~~DIN. Analysis of Cl and Si are described in previous work (Schelker et al., 2014). Analysis uncertainty for NO_3^- were 5% for the concentrations range of 1-100 µg L^{-1} and 4% for 100-1000 µg L^{-1}; uncertainties for NH_4^+ were reported as 14 % for 3–20 µg L^{-1} and 8 % for 20-100 µg L^{-1}. Uncertainties for total N were 14% for 50-1000 µg L^{-1} and 8% for 1-5 mg L^{-1}.

2.3 Mixing model

We used a mixing model to represent the landscape mass-balance for NO_3^- and DON. This model assumes conservative mixing as well as conservative mass transport of water and solutes from two landscape end-members (EMs): clear-cuts and control forests (following Schelker et al. 2014). The chemistry at downstream stations (BA-1 and BA-2) can then be predicted from the simple mixing of the hydro-chemical signal from the upstream EMs. The percentage of clear-cut area of each sub-catchment was derived from high-resolution satellite images supplied by the Swedish Forest Agency combined with local ground-truthing (see Schelker *et al.*, (2014) for a full description). ~~This~~ These data comprise~~s~~ all clear-cuts from the past 10 years (~~2002~~2001-2011~~2~~, see also Figure 1). Similar to earlier work, we considered harvest prior to this period to have a negligible effect, due to their low spatial extent in the watershed (Schelker et al., 2014), and studies elsewhere in the boreal zone that suggest a ten-year time window within clear-cutting is likely to affect DIN exports (e.g. Palviainen et al., 2010). T~~Thus, t~~he remaining area of the catchment was assumed to constitute entirely uncut forest.

The concentration at the ~~a~~ downstream locations BA-1 and BA-2 ($C_{modelled}$, in ~~m~~µg L^{-1}) for each time step was modeled using the area specific mass export (Eq. 1):

$$C_{modelled} = (M_{harvest}\, A_{harvest} + M_{control}\, A_{control})\, {Q_{out}}^{-1}$$

with Q_{out} being the specific discharge (mm~~ day^{-1}~~) at the downstream site, M_i (~~m~~µg m^{-2}~~ day^{-1}~~) being solute mass export for the site i (i=*harvest*, *control*). M_i was calculated as $M_i = Q_i\, C_i$, with C_i (~~mg~~ µg L^{-1}) being the solute concentration and Q_i (mm~~ day^{-1}~~) being the discharge. A_i (~~%~~) was the ~~percentage~~ fraction of the total area that was harvested or acts as a control for the site i, respectively. This mass-balance model ~~allows~~ simulates~~ing~~ the contributions of clear-cuts versus control forests to downstream sites by considering ~~the~~ changes in solute concentrations and water discharge. When measured and modelled concentration are plotted against each other for each sampling date, comparatively higher modeled ~~than measured~~ concentrations (above the *1:1* line) ~~will~~ indicate a mass loss of the solute during transport downstream (and *vice versa*) assuming conservative mass transport and mixing.

A 100% harvested catchment did not exist in Balsjö and N leakage into first-order streams following clear-cutting may vary dependent on local factors, such as the presence of riparian forest buffers (Laurén et al., 2005), and was also observed to differ between the two harvested sites in Balsjö (Löfgren et al., 2009). Thus we calculated $C_{harvest}$ (~~m~~µg L^{-1}) in Equation 1 for each time step as the average concentration of CC-4 and the NO-5 northern catchment, each scaled to 100% harvest using a scaling equation. Assuming a linear increase of harvesting effects, ~~T~~this equation extrapolates the difference between observed concentration ($C_{obs,j}$, in µ~~m~~g L^{-1} with *j*=CC-4 or NO-5) and the concentration of the control forest EM, $C_{control}$ (~~mg~~ µg L^{-1}), to 100% harvest (Eq. 2).

$$C_{harvest,j} = C_{control} + \left(C_{obs,j} - C_{control}\right) d_j$$

The conversion factor, d_j, ~~is~~was defined as $d_j = 1/A_j$, that is, the reciprocal of the ~~percentage~~ fraction of the area harvested (A_j) for the site *j*. Furthermore, $C_{control}$, the concentration representing the control forest EM, was calculated as the average concentration of the two forested reference sites RS-3 and NR-7, that differ in terms of stand age and peatland coverage (Schelker et al., 2014; Löfgren et al., 2009).

Stream discharge (Q, in mm ~~day~~$^{-1}$) for each EM was determined using approaches described previously (Schelker et al., 2014). In short, Q was derived from waterlevel timeseries that were recorded hourly by two Trutrack WTH staff loggers at the sites NR-7, NO-5, CC-4 and BA-1 from which discharge was calculated using well established rating curves at V-notch weirs (Schelker et al., 2012). $Q_{harvest}$ was calculated as the difference between $Q_{NR\text{-}7}$ and $Q_{NO\text{-}5}$, a nested downstream catchment with 88% harvest that is assumed to represent a 100% harvest. $Q_{control}$ was set equal to $Q_{NR\text{-}7}$.

The~~se~~ definitions of Q have been validated in an earlier application of th~~is~~e mixing model, where it was shown that daily Q at BA-1 was modeled reasonably well and with minimal bias using the~~se~~ above assumptions (relationship of modeled vs. measured Q: r^2=0.77; slope=1.01; y-intercept=0.0001, see Schelker et al., (2014)). To further evaluate the representativeness and robustness of the mixing model, the two conservative tracers, Cl and Si were also modelled. A comparison of the modelled vs. measured concentrations (Figure 2 A to D) revealed modeled concentrations to scatter closely around the *1:1* lines with a slightly better fit for BA-2 than for BA-1 and no indications of systematic deviations. These results suggest the validity of the model assumptions for these two conservative tracers.

2.4 additional calculations

Inorganic ~~N~~nitrogen ~~ate~~ removal efficiency (E_r, *in %*) was calculated as the difference between ~~measured and~~ modeled and measured ~~NO_3^-~~ DIN concentrations divided by the modeled concentration. Thus, E_r equals the percentage of ~~NO_3^-~~ DIN that was removed between harvested areas and downstream sampling stations during transport, and this value approaches zero when DIN ~~NO_3^-~~ behaves conservatively in the landscape. If differences between measured and modeled [~~NO_3^-~~ DIN] were <0, E_r was set to zero.

Annual export of DIN and NO_3^- was calculated for each sampling station and year. Solute concentrations between the sampling occasions were interpolated linearly. Daily loads were calculated as the product of concentration and stream discharge and are expressed per unit catchment area. In addition, to compare against the observed DIN and NO_3^- export, modeled estimates of annual export were calculated for BA-1 and BA-2 assuming conservative transport of N from upstream sources. To further infer seasonal effects on N exports, seasons were defined as following: dormant season from November to the end of March, snow melt season from April to the end of May and growing season from June to the end of October of each year, respectively.

To evaluate whether in-stream processes could be responsible for the ~~calculated~~ modelled removal of N in the landscape, we calculated net areal uptakes rates (U; µg N m^{-2} min^{-1}) for ~~NO_3^-~~DIN as the difference between modeled and the measured mass fluxes of DIN ~~NO_3^-~~ divided by the total upstream stream surface area. Stream surface areas (Table 1) were estimated by linear interpolation from known transects within the network combined with a manual analysis of high resolution air photographs. These coarse estimates of U thus represent the net removal of DIN in streams that would be required to achieve mass conservation (an even mass-balance) in the landscape mixing model. Thus, these estimates also represent maximum potential rates as they assume that all uptake would occur within the stream boundaries and not within adjacent riparian soils.

Statistical analysis of differences in measured concentrations before and after ~~treatment~~ clear-cutting in the same stream, as well as between sampling sites were performed as two sample student *t*-tests, accounting for unequal variance. If data ~~was~~ were not normally distributed, a Mann-Whitney Rank Sum test was used instead for pairwise comparisons.

~~Annual export of NO_3^- was calculated for each sampling station and year. NO_3^- concentrations between the sampling occasions were interpolated linearly. Daily loads were calculated as the product of concentration times and stream discharge and are expressed per unit catchment area. In addition, to compare against the observed NO_3^- export, modeled estimates of annual loss were calculated for BA-1 and BA-2 assuming conservative transport of N from upstream sources. To further infer seasonal effects on N exports, seasons were defined as following:~~

~~dormant season from November to the end of March, snow melt season from April to the end of May and growing season from June to the end of October of each year, respectively.~~

3. Results

3.1 DIN and DON responses to harvest

Forest harvesting increased ~~NO_3^-~~ DIN mobilization into first-order streams. Average concentrations of NO_3^- (±SD) at the CC-4 catchment increased significantly (p<0.001) by more than 15-fold from 15.6 (±10.9; n=62) µg N L^{-1} before harvest to 261.0 (±170.4; n=151) µg N L^{-1} after the treatment (Figure ~~2~~3B). In the ~~buffer~~ NO-5 catchment ~~NO-5~~, the response to harvests was less pronounced but also significant (11.4 (±8.6; n=62) µg N L^{-1} before harvest and 25.9 (±35.3; n=151) µg N L^{-1} after, p<0.001). Average concentrations at the NR-7 control stream were 27.6 (±20.5; n=60) µg N L^{-1} in the early period of 2004 to 2006, ~~before harvest~~ and did not change significantly in the later period from 2007-2012~~after the treatment~~ (23.1 (±22.2; n=151) µg N L^{-1}). At the RS-3 control stream NO_3^- concentrations were also low, 12.3 (±9.2; n=49) µg N L^{-1} in the early period~~before harvest~~, but decreased significantly to 5.8 (±7.5; n=151) µg N L^{-1} ~~after the treatment~~during 2007-2012. Similarly, concentrations of NH_4^+ and DON increased in the CC-4 catchment following harvesting (Figure 3, C and D) from (14.7(±6.4; n=30) µg N L^{-1} to 61.8 (±79.9; n=151) µg N L^{-1} and from 324(±108; n=30) µg N L^{-1} to 484 (±239; n=151) µg N L^{-1} for NH_4^+ and DON, respectively. At the reference sites, NH_4^+ and DON remained at similar levels or decreased in the period after harvesting (Figure 3, C and D). In addition to concentration changes, stream runoff was substantially increased after harvest, which enhanced the relative contribution of clear-cuts *versus* control forests for downstream mass fluxes. Annual specific runoff of the CC-4 catchment after the harvest (2007-2012) was 518 (±128) mm whereas the northern control site (NR-7) had a lower average specific discharge of 355 (±88) mm.

At the BA-1 downstream site, NO_3^- concentrations ~~remained statistically similar~~showed no statistically significant difference between the periods of 2004-2006 (17.2 ±14.3 µg N L^{-1}; n=37) and 2007-2012 (17.2 ±18.9 µg N L^{-1}; n=151), even though the upstream area that was clear-cut increased from 2.5% in 2004 to 11.2% in 2011 (Figure 2). At the BA-2 site, where harvests ranged from 4.6% of the catchment area in 2004 to 17.5% in 2011, average NO_3^- concentrations increased ~~significantly~~ modestly (t-test, p=0.026) from 15.9 (±9.8; n=30) µg N L^{-1} during 2004-2006 to 21.3 (±19.1; n=151) µg N L^{-1} during 2007-2012. Similarly, NH_4^+ and DON concentrations at the downstream sites BA-1 and BA-2 increased slightly from 2006 to 2012 (Figure 3, C and D). Also, the contributions of NH_4^+ to the total inorganic N pool varied at both downstream sites between seasons. On average NH_4^+ accounted for 23% and 18% during the dormant season, for 45% and 39% during snowmelt and 54% and 46% of the inorganic N pool during the growing season for BA-1 and BA-2, respectively. Furthermore, NO_3^- concentrations at these downstream sites, as well as at CC-4 increased continuously throughout the winter period, with the highest values observed just prior to snowmelt ~~at the BA-1 and BA-2 sites~~. Annual DIN export was generally dominated by NO_3^- (Table 2) and was highest from the CC-4 catchment (1.28 - 1.83 kg N ha^{-1} y^{-1}), followed by NO-5 (0.10 - 0.17 kg N ha^{-1} y^{-1}), NR-7 (0.06 - 0.10 kg N ha^{-1} y^{-1}), and RS-3 (0.03 – 0.07 kg N ha^{-1} y^{-1}).

3.2 Mixing Model results

When modeled concentrations of DON and DIN ~~NO_3^-~~ at BA-1 and BA-2 were compared to the measured concentrations, distinct patterns emerged. First, modeled and measured DON concentrations correlated well (relationships: r^2=0.92, p<0.001 for BA-2 and r^2=0.72, p<0.001 for BA-1; see also Figure ~~3~~4). In contrast, relationships between modeled and measured DIN ~~NO_3^-~~ concentrations were significant, but explained little of the variability (r^2=0.~~25~~23 for BA-1; r^2=0.31 for BA-2) with modelled DIN concentrations usually ~~nearly exclusively~~ overestimating the measured ~~concentrations~~ values (Figure ~~3~~4). Similarly, a~~A~~nnual modelled DIN exports at the downstream sites were substantially higher than the measured export rates (Table 2).

Modelled DIN ~~NO_3^-~~ removal efficiency calculated as the fraction of DIN ~~NO_3^-~~ that was retained in the system showed a strong seasonal signal (Figure 5A~~4~~). E_r values above 75% were observed just after peak snow melt, with the exception of the snow melt of 2012. E_r then remained high (>75 %) during the summer of 2008, and stayed at intermediate-to-high levels (>50%) during the following summer seasons (Figure ~~4~~5A). Towards the end of the growing season, E_r decreased during all years and was followed by another distinct decline, often with values <40% throughout the winter (Figure ~~4~~5A). Furthermore, no significant relationships between discharge and E_r were ~~found~~ observed (Figure 5B and 5C~~4~~). DI~~Estimates~~ N removal in the network based on this modelling exercise yielded estimates of net retention (U) ~~of U at~~for BA-2 that ~~(Figure 56)~~ were significantly higher during snow melt (9.8 ~~9.9 (-43.1; -3.1)~~ µg N m^{-2} min^{-1}) ~~as compared to~~than the growing ~~season~~ (~~-4.7 (-18.8; -1.4)~~5.4 µg N m^{-2} min^{-1}) and ~~the~~ dormant ~~season~~ (5.3 ~~-5.6 (-14.5; -0.4)~~ µg N m^{-2} min^{-1}) seasons, respectively (Figure 6). ~~Similar~~ Estimates of U~~values~~ for BA-1 were lower, with 2.3, 1.1 and 0.8, µg N m^{-2} min^{-1} for the snowmelt, growing and dormant season, respectively.

4. Discussion

~~The observed changes in~~Increases in DIN ~~and NO_3^-~~ export in response to forest harvesting ~~in first-order streams~~are well documented (Jerabkova et al., 2011) and illustrate how ~~suggest that~~ terrestrial ecosystem disturbance can control~~s~~ N mobilization and delivery to ~~into~~ small streams. ~~The~~ In this study, ~~concurrent~~ increases in streamwater NO_3^- concentrations by up to ~15 fold, together ~~with significant increases in~~with elevated ~~stream~~ runoff~~, the latter primarily caused by low evapotranspiration in clear cuts during summer~~ (Schelker et al., 2013b), ~~are thus governing~~resulted in substantial increases in ~~NO_3^-~~DIN inputs to the fluvial network

(Table ~~1~~2). However, despite obvious effects of forest harvesting on ~~NO_3^-~~ DIN concentrations in first-order streams, only very subtle responses could be detected for the third-order streams within this same network, suggesting that significant DIN~~NO_3^-~~ retention occurred between the harvested areas in the landscape and downstream monitoring sites.

4.1 Network patterns in DIN concentration

At both downstream sites, and the CC-4 clear-cut catchment, concentrations of NO_3^- were higher during the dormant ~~season as compared to~~than ~~the~~ growing season (Figure ~~2~~3B). S~~.~~ ~~S~~imilar ~~ly, these~~ seasonal ~~variations~~ patterns were ~~also largely paralleled by~~observed for NH_4^+ concentrations (Figure 3C)~~(data not shown). However, contributions of NH_4^+ to the total inorganic N pool varied at both downstream sites between seasons. On average NH_4^+ accounted for 23% and 18% during winter low flow, for 45% and 39% during snowmelt and 54% and 46% of the inorganic N pool for BA-1 and BA-2, respectively~~. Overall, such seasonal variation in stream DIN~~inorganic N~~, and specifically stream NO_3^- concentration~~s~~, is common across Sweden (Sponseller et al., 2014; Löfgren et al., 2014) and is thought to reflect seasonal changes in terrestrial N demand (e.g. Mitchell et al., 1996). In contrast, NO_3^- concentrations at RS-3 did not show such a seasonal pattern, suggesting particularly low inorganic N availability and strong N-limitation persisting throughout the year (Stoddard, 1994). This hypothesis is further supported by the fact that average NO_3^- concentrations at this site decreased significantly by -6.5 μg N L^{-1} between the period from ~~pre-treatment~~ 2004 to 2006 as compared to ~~and~~ 2007 to~~-the treatment period~~2012, indicating that local factors, such as the presence of actively growing forest stands with dense riparian vegetation, resulted in particularly~~a~~ high ~~inorganic~~ terrestrial N demand and thus low stream concentrations at this site.

~~Seasonal~~ Temporal variation~~s~~ in NO_3^- concentrations at the CC-4 clear-cut stream during the dormant season (Figure 2) was closely~~ere~~ ~~related~~ correlated with~~closely with~~ temporal changes in NO_3^- concentration at ~~the~~ downstream sites (Supplementary Figure S1), indicating ~~a~~ temporal coherence in concentration change (*sensu* Kling et al., 2000) ~~within~~ across the stream network during this period. In contrast, temporal changes in upstream and downstream NO_3^- concentrations were not correlated during the growing season (Supplementary Figure S1). Overall, these observations suggest (i) a common seasonal control where NO_3^- retention in most catchments declines throughout the dormant season, (ii) that enhanced upstream inputs of NO_3^- in headwaters are translated downstream during the dormant season, and iii) that temporal nutrient dynamics at upstream and downstream reaches become uncoupled during the spring and the summer growing season.

4.2 Comparison of modeled and measured streamwater N

We found a close correspondence between modelled and measured DON concentrations, similar to relationships previously observed for dissolved organic carbon (Schelker et al., 2014), as well as the two conservative tracers, dissolved silica and chloride (Figure 2). This close relationship between observed and predicted concentrations is indicative of an approximately conservative downstream transport of DON in the network. These patterns provide additional support for the applicability of our mixing model in this landscape, and they are consistent with the idea that bulk DON is composed primarily of organic compounds of low bioavailability that are exported from landscapes without strong biotic controls (Hedin et al., 1995). For this reason, DON also often represents the major loss vector for N in catchments that are not subject to large anthropogenic inputs of DIN (Perakis, 2002; Kortelainen et al., 1997). ~~Moreover~~Importantly, DON exports at CC-4 also increased following harvesting (Figure 3D), ~~which~~a response which has been ~~also~~ reported elsewhere in Scandinavia (Smolander et al., 2001). While this ~~this~~response was more subtle than that observed for DIN, the conservative behavior of DON in the stream network suggests that it likely represents an important and largely unappreciated source of terrestrially derived N to downstream receiving systems (Rosén et al., 1996).

In contrast to DON, we observed generally p~~P~~oor relationships between measured and modelled ~~NO_3^-~~DIN concentrations at BA-1 and BA-2 (Figure ~~3~~4, data for BA-2 not shown). This mismatch most ~~are~~ likely ~~to~~ results from seasonal NO_3^- removal, a pattern illustrated ~~supported~~ by the temporal variation of E_r for both sites (Figure ~~4~~5). ~~In contrast, the relationships of modelled and measured DON concentrations are similar to those previously observed for dissolved organic carbon, as well as the two conservative tracers, dissolved silica and chloride (Schelker et al., 2014). These relationships are thus indicative for an approximately conservative downstream transport of DON in the network. Furthermore, these patterns provide additional support for the applicability of our mixing model in this landscape, as they are consistent with the idea that bulk DON is composed primarily of organic compounds of low bioavailability that is exported from landscapes without strong biotic controls (Hedin et al., 1995). For this reason, DON also often represents the major loss vector for N in catchments that are not subject to large anthropogenic inputs of inorganic N (Perakis, 2002; Kortelainen et al., 1997). Given that clear cutting led to increased DOC export from these same catchments (Schelker et al., 2014), and that DOC and DON are assumed to belong to the same organic matter pool and are thus often highly correlated in boreal catchments (Sponseller et al., 2014), losses of DON in response to harvesting may represent an important and largely unappreciated source of terrestrially derived N to downstream receiving systems (Rosén et al., 1996).~~

Low dormant season values of E_r suggest an ostensibly weak NO_3^- demand in cold, snow-covered streams and thus low strength of the biological sink within the fluvial network.

During this period a large fraction of NO_3^- entering the stream network was also exported downstream, which is exemplified by the upstream-downstream synchrony in nutrient concentrations observed during this period (S1) and the few wintertime occasions where E_r was near-zero. These occasions suggest that either (i) all NO_3^- was transported downstream (e.g. that NO_3^- transport was conservative) or (ii) that the downstream reaches of the stream network acted as source areas of NO_3^-. The latter has been previously hypothesized to cause discrepancies of reach scale N mass-balances (von Schiller et al., 2011).

Interestingly, E_r did not show a direct dependence on stream discharge at any of the downstream sites (Figure ~~4~~5), suggesting that N-demand rather than flow ~~(Hill, 1993)~~ and/or transient storage (Ensign and Doyle, 2006) were controlling ~~NO_3^-~~ DIN removal in the fluvial network. In addition, high removal efficiencies during spring and summer had substantial effects on overall annual net ~~NO_3^-~~-DIN ~~removal~~ uptake as estimated by the difference of modeled and measured annual DIN~~NO_3^-~~ exports. These estimates (±SD) showed that ~~71~~67(±~~4~~3)% and ~~67~~65(±~~10~~8)% of the ~~NO_3^-~~ DIN inputs to the BA-1 and BA-2 catchments were removed before reaching these monitoring stations ~~outlets~~ (Table ~~1~~2). These estimates are of course sensitive to how the clear-cut EM was represented in the mixing model. For example, if we assume that all clear cut areas would follow the less pronounced concentration response of NO-5 ~~,~~ then the average annual DIN removal would sum ~~up~~ to 22% for BA-1 and only 9% for BA-2, with the latter even acting as a source of DIN (+2%) during one year (2009). However, we consider this extreme scenario unrealistic for at least two reasons. First, several harvests in the drainage area of the stream network, but outside the experimental harvest of NO-5 and CC-4, showed substantial disturbance of riparian soils, for example from multiple stream crossings of forestry machines and from wind throw of trees in the riparian zone. These disturbances will likely result in a concentration response closer to that of CC-4, than that of NO-5. Second, the CC-4 clear cut is located within the BA-2 drainage area and represents an important fraction of the harvested area within this catchment (Tabe1). Thus the CC-4 harvest would itself not be correctly represented in this modeling scenario. Indeed, this omission gives rise to the hypothetical gain of DIN within BA-2 in 2009, which suggests a missing source of DIN in the catchment under this scenario. Regardless, further research characterizing the spatial and temporal variation in DIN runoff responses following harvests would lend more confidence to estimates of N removal based on this mass balance approach.

Our ~~Furthermore, our~~ estimates of net ~~NO_3^-~~DIN removal within this stream network suggest that, during most periods, reasonable levels of in-stream activity (i.e., net uptake) could account for the discrepancy between measured and modeled fluxes at downstream stations. Assuming that all ~~NO_3^-~~DIN retention was occurring within the stream channels, median values and interquartile ranges (~~10^{th}~~-25^{th} to ~~90^{th}~~-75^{th} percentile) of U for the BA-2 catchment were ~~-~~5.~~8~~4 (2.2; 10.4~~-21.9; -1.3~~) µg N m^{-2} min^{-1} for the entire year. Even lower rates of instream uptake would be sufficient to account for the differences between modeled and observed DIN at BA-1. ~~These~~ While these values fall well within the range of net uptake estimates made elsewhere for small streams (Bernhardt et al., 2003; Roberts and Mulholland, 2007; von Schiller et al., 2011), further efforts to directly quantify rates of DIN removal in boreal streams are warranted.~~.~~

As with E_r, estimates of U were significantly higher during snow melt as compared to the growing season and, interestingly, there was no significant difference in median values between growing and dormant seasons (Figure ~~5~~6). While other recent studies indicate the potential for high rates of nutrient uptake during the snowmelt period (Hall et al., 2009), these seasonal comparisons should be made with some caution as our estimates of net areal ~~removal~~ uptake do not account for losses that occur to the outside of the stream, as for example ~~losses to the hyporheic zone,~~in riparian habitats, embedded wetlands, lakes ~~, lakes,~~ and/~~-~~or ~~into~~ into deep groundwater aquifers~~zones~~. In particular, embedded wetlands and small lakes upstream of BA-1 and BA-2 (Table 1) are common features of boreal landscapes and may play a particularly important role in N removal at the scale of stream networks. Overall, these seasonal removal estimates are surprising, and ~~further~~more work is required to understand the hydrological and biogeochemical mechanisms underpinning these patterns.

Important mechanisms that control ~~NO_3^-~~DIN removal from stream water during the growing season are biological uptake by riparian vegetation (Sabater et al., 2000) and immobilization by in-stream ~~primary producers~~autotrophs and heterotrophs. These in-stream sinks may also change in response to forest harvesting, for example, if elevated light conditions foster increased photo~~-~~autotrophic production (Bernhardt and Likens, 2004). Indication that such increased in-stream ~~NO_3^-~~DIN demand during the growing season may also be present in the Balsjö stream network is given by ~30 fold greater summertime accumulation of algal biomass (*chlorophyll a)* onto ceramic tiles in the CC-4 stream as compared to RS-3 (R. Sponseller, unpublished data). Similarly, a recent survey of boreal streams (including CC-4 and RS-3) showed that heterotrophic biofilm respiration can be strongly N-limited and reported the highest rates of biofilm respiration at the clear cut stream of CC-4 (Burrows et al., 2015). However, ~~uptake~~ immobilization by autotrophs and heterotrophs ~~is~~does not necessarily result in ~~a~~ permanent removal of N from the stream, ~~ecosystem, because~~as a large portion of this ~~substantial amounts of N~~nutrient pool may be rapidly recycled ~~when~~ as ~~algal~~ biofilm material~~s~~ decay~~s~~ (Tank et al., 2000). Nevertheless, these observations highlight the importance of N as limiting factor in northern, boreal streams and ~~Similarly, a recent study found heterotroph microbial respiration in boreal streams to be strongly N-limited, with the highest observed heterotroph respiration rate (~70 µg O_2 cm^{-2} h^{-1}) in the CC-4 clear cut stream of this study (Burrows et al., 2015). This indicates the widespread N-limitation of biofilms in boreal streams and their~~ support the idea that these systems may ~~immediate~~ respond~~se~~ strongly ~~to higher~~to elevated ~~terrestrial~~ N loading~~s~~ following harvests.

An additional process that may account for the permanent removal of NO_3^- observed in this study and thus for the seasonal differences in U is denitrification (Mulholland et al., 2008). Environments that have been observed to favor the direct conversion of NO_3^- to gaseous N by denitrification are i) stream biofilms (Teissier et al., 2007), ii) stream hyporheic zones (Harvey et al., 2013) and iii) riparian sediments (Starr and Gillham, 1993). Furthermore, experimental studies have demonstrated that ~~the process of~~ denitrification is often found to be co-dependent on terrestrial NO_3^- inputs and bioavailable dissolved organic matter (DOM) as an electron donor (Baker et al., 1999). More specifically, *hot moments* of denitrification, that is, periods of ~~a~~ disproportionally high and ~~evanescent~~ short-lived ~~assimilatory~~ NO_3^- demand, can be generated by experimental additions of labile DOM (Zarnetske et al., 2011). Such enhanced demand in response to labile DOM inputs has further been shown to regulate uptake rates in streams ~~reaches~~ (Bernhardt and Likens, 2002) and hyporheic sediments (Sobczak et al., 2003). ~~Additional reach scale NO_3^- retention could also be linked to dissimilatory NO_3^- demand caused by the reduction of NO_3^- to NH_4^+. Such demands could also be causing the seasonally varying proportions of NH_4^+ of the total inorganic N pool. (Sgouridis et al., 2011)However, this processing does not represent a permanent removal of inorganic N from streams (Mulholland et al., 2008) as NH_4^+ may be re-oxidized to NO_3^- in downstream environments that favor nitrification.~~

Transferring this well-established process knowledge from the reach-scale to the network scale suggests that NO_3^- removal at the landscape scale may be dependent on a sufficient supply of labile DOM to all stream reaches within the network that are located downstream of harvests. Bulk DOM contributions in Balsjö have been observed to increase as a response to harvesting (Schelker et al., 2012) and ~~Research in~~ other ~~work~~studies in boreal headwater streams have~~s~~ shown that terrestrially-derived, low molecular weight DOM (e.g., ~~, commonly consisting of~~ free amino acids, carboxylic acids and carbohydrates), can achieve high concentrations during the spring snow melt (~~(~~Berggren et al., 2009). These terrestrial inputs have further been suggested ~~to be able~~ to support the microbial C demand of downstream aquatic ecosystems during a timeframe of days to weeks following the spring freshet (Berggren et al., 2009) – ~~times~~ a period when E_r was also highest in our study. Thus we ~~suggest~~ hypothesize that~~a~~ limitation of heterotrophic processes, such as denitrification and immobilization, ~~by~~ occurs via ~~the~~ restricted supply of bioavailable DOM from terrestrial sources during the dormant season as a plausible mechanism that inhibits ~~net NO_3^-~~DIN removal at the network scale. In turn, the ~~limited~~ restricted supply of ~~inorganic N~~DIN relative to bioavailable C during the other times of the year would then limit heterotrophic ~~turnover of DOM~~activities and foster efficient N removal in the network – a coupling that has been suggested previously for boreal streams (Berggren et al., 2007).

In summary our work agrees with earlier studies in that terrestrial ecosystem disturbance enhances DIN ~~and NO_3^-~~ mobilization into first-order streams (Likens et al., 1970) and that such increased NO_3^- concentrations can potentially be transferred downstream during some portions of the years (Alexander et al., 2007). The hypothesis that stream and riparian processing of NO_3^- may dampen the effect at downstream sites (Bernhardt et al., 2003) was supported during the snow melt, as well as during the growing season when rates of biological activity and supply of bioavailable C are likely to be high. During the dormant season, however, results suggest that limited ~~net NO_3^-~~ DIN uptake rates constrain the potential for ~~NO_3^-~~DIN removal within the fluvial network. Considering the ~~two mentioned~~ measures to increase forest production of either increased fertilization or shorter forest rotations (Egnell et al., 2011), we argue that both are likely to increase downstream export of ~~NO_3^-~~DIN, provided that instream removal rates ~~if the stream network's removal rates~~ remain the same as under current conditions. More specifically, shorter forest rotations would increase the frequency of disturbance due to harvesting and thus the periods where elevated leaching may occur.

Similarly, increased fertilization may enhance the risk of ~~NO_3^-~~DIN leakage into surface waters particularly during the dormant season (Binkley et al., 1999) when the biological demand for ~~inorganic N~~DIN is low within boreal stream networks.

5. Acknowledgements

Funding for this work was provided by the Swedish Environmental Protection Agency, EU Life (Forest for Water), CMF, Future Forests and the Formas (ForWater). We thank Peder Blomkvist, Viktor Sjöblom and Ida Taberman for help in the field and the laboratory.

6. References

Aber, J. D., Ollinger, S. V., Driscoll, C. T., Likens, G. E., Holmes, R. T., Freuder, R. J., and Goodale, C. L.: Inorganic nitrogen losses from a forested ecosystem in responseto physical, chemical, biotic,and climatic perturbations, Ecosystems, 5, 0648-0658, 10.1007/s10021-002-0203-8, 2002.

Alexander, R., Boyer, E. W., Smith, R. A., Schwarz, G. E., and Moore, R. B.: The role of headwater streams in downstream water quality1, JAWRA Journal of the American Water Resources Association, 43, 41-59, 10.1111/j.1752-1688.2007.00005.x, 2007.

Alexander, R., Böhlke, J., Boyer, E., David, M., Harvey, J., Mulholland, P., Seitzinger, S., Tobias, C., Tonitto, C., and Wollheim, W.: Dynamic modeling of nitrogen losses in river networks unravels the coupled effects of hydrological and biogeochemical processes, Biogeochemistry, 93, 91-116, 10.1007/s10533-008-9274-8, 2009.

Andréassian, V.: Waters and forests: From historical controversy to scientific debate, J. Hydrol., 291, 1-27, 2004.

Baker, M. A., Dahm, C. N., and Valett, H. M.: Acetate retention and metabolism in the hyporheic zone of a mountain stream, Limnology and Oceanography, 44, 1530-1539, 1999.

Berggren, M., Laudon, H., and Jansson, M.: Landscape regulation of bacterial growth efficiency in boreal freshwaters, Global Biogeochem. Cycles, 21, GB4002, doi:4010.1029/2006GB002844, 2007.

Berggren, M., Laudon, H., Haei, M., Strom, L., and Jansson, M.: Efficient aquatic bacterial metabolism of dissolved low-molecular-weight compounds from terrestrial sources, ISME J, 4, 408-416, 2009.

Bernhardt, E. S., and Likens, G. E.: Dissolved organic carbon enrichment alters nitrogen dynamics in a forest stream, Ecology, 83, 1689-1700, 2002.

Bernhardt, E. S., Likens, G. E., Buso, D. C., and Driscoll, C. T.: In-stream uptake dampens effects of major forest disturbance on watershed nitrogen export, Proceedings of the National Academy of Sciences, 100, 10304-10308, 10.1073/pnas.1233676100, 2003.

Bernhardt, E. S., and Likens, G. E.: Controls on periphyton biomass in heterotrophic streams, Freshwater. Biol., 49, 14-27, 10.1046/j.1365-2426.2003.01161.x, 2004.

Binkley, D., and Brown, T. C.: Forest practices as nonpoint sources of pollution in north america, JAWRA Journal of the American Water Resources Association, 29, 729-740, 10.1111/j.1752-1688.1993.tb03233.x, 1993.

Binkley, D., Burnham, H., and Lee Allen, H.: Water quality impacts of forest fertilization with nitrogen and phosphorus, For. Ecol. Manage., 121, 191-213, 10.1016/S0378-1127(98)00549-0, 1999.

Bishop, K., Seibert, J., Köhler, S., and Laudon, H.: Resolving the double paradox of rapidly mobilized old water with highly variable responses in runoff chemistry, Hydrol. Process., 18, 185-189, 2004.

Boring, L. R., Monk, C. D., and Swank, W. T.: Early regeneration of a clear-cut southern appalachian forest, Ecology, 1244-1253, 1981.

Burrows, R. M., Hotchkiss, E. R., Jonsson, M., Laudon, H., McKie, B. G., and Sponseller, R. A.: Nitrogen limitation of heterotrophic biofilms in boreal streams, Freshwater. Biol., in press, 10.1111/fwb.12549, 2015.

Egnell, G., Laudon, H., and Rosvall, O.: Perspectives on the potential contribution of swedish forests to renewable energy targets in europe, Forests, 2, 578-589, 2011.

Ensign, S. H., and Doyle, M. W.: Nutrient spiraling in streams and river networks, Journal of Geophysical Research: Biogeosciences, 111, G04009, 10.1029/2005jg000114, 2006.

Futter, M. N., Ring, E., Högbom, L., Entenmann, S., and Bishop, K. H.: Consequences of nitrate leaching following stem-only harvesting of swedish forests are dependent on spatial scale, Environ. Pollut., 158, 3552-3559, 2010.

Groffman, P. M., Boulware, N. J., Zipperer, W. C., Pouyat, R. V., Band, L. E., and Colosimo, M. F.: Soil nitrogen cycle processes in urban riparian zones, Environ. Sci. Technol., 36, 4547-4552, 2002.

Hall, R. O., Baker, M. A., Arp, C. D., and Kocha, B. J.: Hydrologic control of nitrogen removal, storage, and export in a mountain stream, Limnology and Oceanography, 54, 2128-2142, 2009.

Harvey, J. W., Böhlke, J. K., Voytek, M. A., Scott, D., and Tobias, C. R.: Hyporheic zone denitrification: Controls on effective reaction depth and contribution to whole-stream mass balance, Water Resour. Res., 49, 6298-6316, 10.1002/wrcr.20492, 2013.

Hedin, L. O., Armesto, J. J., and Johnson, A. H.: Patterns of nutrient loss from unpolluted, old-growth temperate forests: Evaluation of biogeochemical theory, Ecology, 76, 493-509, 10.2307/1941208, 1995.

Helton, A. M., Poole, G. C., Meyer, J. L., Wollheim, W. M., Peterson, B. J., Mulholland, P. J., Bernhardt, E. S., Stanford, J. A., Arango, C., Ashkenas, L. R., Cooper, L. W., Dodds, W. K., Gregory, S. V., Hall Jr, R. O., Hamilton, S. K., Johnson, S. L., McDowell, W. H., Potter, J. D., Tank, J. L., Thomas, S. M., Valett, H. M., Webster, J. R., and Zeglin, L.: Thinking outside the channel: Modeling nitrogen cycling in networked river ecosystems, Frontiers in Ecology and the Environment, 9, 229-238, 2011.

Hill, A. R.: Nitrogen dynamics of storm runoff in the riparian zone of a forested watershed, Biogeochemistry, 20, 19-44, 1993.

Högberg, P., Fan, H. B., Quist, M., Binkley, D., and Tamm, C. O.: Tree growth and soil acidification in response to 30 years of experimental nitrogen loading on boreal forest, Glob. Change Biol., 12, 489-499, DOI 10.1111/j.1365-2486.2005.01102.x, 2006.

Holmes, W. E., and Zak, D. R.: Soil microbial control of nitrogen loss following clear-cut harvest in northern hardwood ecosystems, Ecol. Appl., 9, 202-215, 10.1890/1051-0761(1999)009[0202:smconl]2.0.co;2, 1999.

Hornbeck, J. W., Martin, C. W., and Eagar, C.: Summary of water yield experiments at hubbard brook experimental forest, new hampshire, Can. J. For. Res., 27, 2043-2052, 1997.

Houlton, B. Z., Driscoll, C. T., Fahey, T. J., Likens, G. E., Groffman, P. M., Bernhardt, E. S., and Buso, D. C.: Nitrogen dynamics in ice storm-damaged forest ecosystems: Implications for nitrogen limitation theory, Ecosystems, 6, 431-443, 10.1007/s10021-002-0198-1, 2003.

Jansson, M., Bergström, A. K., Drakare, S., and Blomqvist, P.: Nutrient limitation of bacterioplankton and phytoplankton in humic lakes in northern sweden, Freshwater. Biol., 46, 653-666, 2001.

Jerabkova, L., Prescott, C. E., Titus, B. D., Hope, G. D., and Walters, M. B.: A meta-analysis of the effects of clearcut and variable-retention harvesting on soil nitrogen fluxes in boreal and temperate forests, Can. J. For. Res., 41, 1852-1870, 10.1139/x11-087, 2011.

Kling, G. W., Kipphut, G. W., Miller, M. M., and O'Brien, W. J.: Integration of lakes and streams in a landscape perspective: The importance of material processing on spatial patterns and temporal coherence, Freshwater. Biol., 43, 477-497, 2000.

Kortelainen, P., Saukkonen, S., and Mattsson, T.: Leaching of nitrogen from forested catchments in finland, Global Biogeochem. Cycles, 11, 627-638, 10.1029/97gb01961, 1997.

Kreutzweiser, D. P., Hazlett, P. W., and Gunn, J. M.: Logging impacts on the biogeochemistry of boreal forest soils and nutrient export to aquatic systems: A review, Environ. Rev., 16, 157-179, 10.1139/A08-006, 2008.

Laudon, H., Berggren, M., Agren, A., Buffam, I., Bishop, K., Grabs, T., Jansson, M., and Kohler, S.: Patterns and dynamics of dissolved organic carbon (doc) in boreal streams: The role of processes, connectivity, and scaling, Ecosystems, 14, 880-893, DOI 10.1007/s10021-011-9452-8, 2011.

Laurén, A., Finér, L., Koivusalo, H., Kokkonen, T., Karvonen, T., Kellomäki, S., Mannerkoski, H., and Ahtiainen, M.: Water and nitrogen processes along a typical water flowpath and streamwater exports from a forested catchment and changes after clear-cutting: A modelling study, Hydrology & Earth System Sciences, 9, 2005.

Likens, G. E., Bormann, F. H., Johnson, N. M., Fisher, D. W., and Pierce, R. S.: Effects of forest cutting and herbicide treatment on nutrient budgets in the hubbard brook watershed-ecosystem, Ecol Monogr, 40, 23-47, 10.2307/1942440, 1970.

Likens, G. E., and Bormann, F. H.: Biogeochemistry of a forested ecosystem, Springer-Verlag New York, Inc., 1995.

Löfgren, S., Ring, E., von Brömssen, C., Sørensen, R., and Högbom, L.: Short-term effects of clear-cutting on the water chemistry of two boreal streams in northern sweden: A paired catchment study, Ambio, 38, 347-356, 10.1579/0044-7447-38.7.347, 2009.

Löfgren, S., Fröberg, M., Yu, J., Nisell, J., and Ranneby, B.: Water chemistry in 179 randomly selected swedish headwater streams related to forest production, clear-felling and climate, Environ. Monit. Assess., 1-22, 10.1007/s10661-014-4054-5, 2014.

Martin, C. W., Hornbeck, J. W., Likens, G. E., and Buso, D. C.: Impacts of intensive harvesting on hydrology and nutrient dynamics of northern hardwood forests, Can. J. Fish. Aquat. Sci., 57, 19-29, 10.1139/f00-106, 2000.

McClain, M. E., Boyer, E. W., Dent, C. L., Gergel, S. E., Grimm, N. B., Groffman, P. M., Hart, S. C., Harvey, J. W., Johnston, C. A., Mayorga, E., McDowell, W. H., and Pinay, G.: Biogeochemical hot spots and hot moments at the interface of terrestrial and aquatic ecosystems, Ecosystems, 6, 301-312, 2003.

Mitchell, M. J., Driscoll, C. T., Kahl, J. S., Murdoch, P. S., and Pardo, L. H.: Climatic control of nitrate loss from forested watersheds in the northeast united states, Environ. Sci. Technol., 30, 2609-2612, 10.1021/es9600237, 1996.

Mulholland, P. J., Helton, A. M., Poole, G. C., Hall, R. O., Hamilton, S. K., Peterson, B. J., Tank, J. L., Ashkenas, L. R., Cooper, L. W., Dahm, C. N., Dodds, W. K., Findlay, S. E. G., Gregory, S. V., Grimm, N. B., Johnson, S. L., McDowell, W. H., Meyer, J. L., Valett, H. M., Webster, J. R., Arango, C. P., Beaulieu, J. J., Bernot, M. J., Burgin, A. J., Crenshaw, C. L., Johnson, L. T., Niederlehner, B. R., O/'Brien, J. M., Potter, J. D., Sheibley, R. W., Sobota, D. J., and Thomas, S. M.: Stream denitrification across biomes and its response to anthropogenic nitrate loading, Nature, 452, 202-205, 10.1038/nature06686, 2008.

Ocampo, C. J., Oldham, C. E., and Sivapalan, M.: Nitrate attenuation in agricultural catchments: Shifting balances between transport and reaction, Water Resour. Res., 42, 2006.

Palviainen, M., Finér, L., Laiho, R., Shorohova, E., Kapitsa, E., and Vanha-Majamaa, I.: Carbon and nitrogen release from decomposing scots pine, norway spruce and silver birch stumps, For. Ecol. Manage., 259, 390-398, http://dx.doi.org/10.1016/j.foreco.2009.10.034, 2010.

Perakis, S. S.: Nutrient limitation, hydrology and watershed nitrogen loss, Hydrol. Process., 16, 3507-3511, 10.1002/hyp.5078, 2002.

Pinay, G., Black, V., Planty-Tabacchi, A., Gumiero, B., and Decamps, H.: Geomorphic control of denitrification in large river floodplain soils, Biogeochemistry, 50, 163-182, 2000.

Ranalli, A. J., and Macalady, D. L.: The importance of the riparian zone and in-stream processes in nitrate attenuation in undisturbed and agricultural watersheds–a review of the scientific literature, J. Hydrol., 389, 406-415, 2010.

Riscassi, A. L., and Scanlon, T. M.: Nitrate variability in hydrological flow paths for three mid-appalachian forested watersheds following a large-scale defoliation, Journal of Geophysical Research: Biogeosciences, 114, G02009, 10.1029/2008jg000860, 2009.

Roberts, B. J., and Mulholland, P. J.: In‐stream biotic control on nutrient biogeochemistry in a forested stream, west fork of walker branch, Journal of Geophysical Research: Biogeosciences (2005‐2012), 112, 2007.

Rosén, K., Aronson, J.-A., and Eriksson, H. M.: Effects of clear-cutting on streamwater quality in forest catchments in central sweden, For. Ecol. Manage., 83, 237-244, 10.1016/0378-1127(96)03718-8, 1996.

Sabater, F., Butturini, A., MartÍ, E., Muñoz, I., Romaní, A., Wray, J., and Sabater, S.: Effects of riparian vegetation removal on nutrient retention in a mediterranean stream, J N Am Benthol Soc, 609-620, 2000.

Scanlon, T. M., Ingram, S. M., and Riscassi, A. L.: Terrestrial and in‐stream influences on the spatial variability of nitrate in a forested headwater catchment, Journal of Geophysical Research: Biogeosciences (2005–2012), 115, 2010.

Schelker, J., Eklöf, K., Bishop, K., and Laudon, H.: Effects of forestry operations on dissolved organic carbon concentrations and export in boreal first-order streams, J. Geophys. Res., 117, G01011, 10.1029/2011jg001827, 2012.

Schelker, J., Grabs, T., Bishop, K., and Laudon, H.: Drivers of increased organic carbon concentrations in stream water following forest disturbance: Separating effects of changes in flow pathways and soil warming, Journal of Geophysical Research: Biogeosciences, 118, 2013JG002309, 10.1002/2013jg002309, 2013a.

Schelker, J., Kuglerová, L., Eklöf, K., Bishop, K., and Laudon, H.: Hydrological effects of clear-cutting in a boreal forest – snowpack dynamics, snowmelt and streamflow responses, J. Hydrol., 484, 105-114, 10.1016/j.jhydrol.2013.01.015, 2013b.

Schelker, J., Öhman, K., Löfgren, S., and Laudon, H.: Scaling of increased dissolved organic carbon inputs by forest clear-cutting – what arrives downstream?, J. Hydrol., 508, 299-306, 10.1016/j.jhydrol.2013.09.056, 2014.

Seitzinger, S., Harrison, J. A., Böhlke, J. K., Bouwman, A. F., Lowrance, R., Peterson, B., Tobias, C., and Drecht, G. V.: Denitrification across landscapes and waterscapes: A synthesis, Ecol. Appl., 16, 2064-2090, 10.1890/1051-0761(2006)016[2064:dalawa]2.0.co;2, 2006.

Smolander, A., Kitunen, V., and Mälkönen, E.: Dissolved soil organic nitrogen and carbon in a norway spruce stand and an adjacent clear-cut, Biol. Fertil. Soils, 33, 190-196, 2001.

Sobczak, W. V., Findlay, S., and Dye, S.: Relationships between doc bioavailability and nitrate removal in an upland stream: An experimental approach, Biogeochemistry, 62, 309-327, 2003.

Sponseller, R. A., Temnerud, J., Bishop, K., and Laudon, H.: Patterns and drivers of riverine nitrogen (n) across alpine, subarctic, and boreal sweden, Biogeochemistry, 120, 105-120, 10.1007/s10533-014-9984-z, 2014.

Starr, R. C., and Gillham, R. W.: Denitrification and organic carbon availability in two aquifers, Ground Water, 31, 934-947, 10.1111/j.1745-6584.1993.tb00867.x, 1993.

Stoddard, J. L.: Long-term changes in watershed retention of nitrogen, in: Environmental chemistry of lakes and reservoirs, edited by: Baker, L. A., Advances in chemistry, 237, American Chemical Society, Washington D.C. , 223-284, 1994.

Swank, W. T., and Vose, J. M.: Long-term nitrogen dynamics of coweeta forested watersheds in the southeastern united states of america, Global Biogeochem. Cycles, 11, 657-671, 10.1029/97gb01752, 1997.

Tank, J. L., Meyer, J. L., Sanzone, D. M., Mulholland, P. J., Webster, J. R., Peterson, B. J., Wollheim, W. M., and Leonard, N. E.: Analysis of nitrogen cycling in a forest stream during autumn using a 15n-tracer addition, Limnology and Oceanography, 45, 1013-1029, 2000.

Teissier, S., Torre, M., Delmas, F., and Garabétian, F.: Detailing biogeochemical n budgets in riverine epilithic biofilms, J N Am Benthol Soc, 26, 178-190, 10.1899/0887-3593(2007)26[178:dbnbir]2.0.co;2, 2007.

Valett, H., Thomas, S., Mulholland, P., Webster, J., Dahm, C., Fellows, C. S., Crenshaw, C., and Peterson, C.: Endogenous and exogenous control of ecosystem function: N cycling in headwater streams, Ecology, 89, 3515-3527, 2008.

Vidon, P. G. F., and Hill, A. R.: Landscape controls on nitrate removal in stream riparian zones, Water Resour. Res., 40, W03201, 10.1029/2003wr002473, 2004.

Vitousek, P. M., Gosz, J. R., Grier, C. C., Melillo, J. M., Reiners, W. A., and Todd, R. L.: Nitrate losses from disturbed ecosystems, Science, 204, 469-474, 10.1126/science.204.4392.469, 1979.

von Schiller, D., Bernal, S., and Martí, E.: Technical note: A comparison of two empirical approaches to estimate in-stream net nutrient uptake, Biogeosciences, 8, 875-882, 10.5194/bg-8-875-2011, 2011.

Weller, D. E., Baker, M. E., and Jordan, T. E.: Effects of riparian buffers on nitrate concentrations in watershed discharges: New models and management implications, Ecol. Appl., 21, 1679-1695, 2011.

Wollheim, W. M., Vörösmarty, C. J., Peterson, B. J., Seitzinger, S. P., and Hopkinson, C. S.: Relationship between river size and nutrient removal, Geophysical Research Letters, 33, 2006.

Worrall, F., Burt, T. P., Howden, N. J. K., and Whelan, M. J.: The fluvial flux of nitrate from the uk terrestrial biosphere – an estimate of national-scale in-stream nitrate loss using an export coefficient model, J. Hydrol., 414–415, 31-39, 10.1016/j.jhydrol.2011.09.020, 2012.

Zarnetske, J. P., Haggerty, R., Wondzell, S. M., and Baker, M. A.: Labile dissolved organic carbon supply limits hyporheic denitrification, Journal of Geophysical Research: Biogeosciences, 116, G04036, 10.1029/2011jg001730, 2011.

7. Tables

Table 1: Catchment characteristics of the six nested Balsjö catchments.

Site Name	Short Name	Catchment Area	Proportion Clear-Cut*, 2004; 2011		Wetland Area	Total Stream Length	Lake Area*	Stream Surface Area	Total Water Area
Unit		[ha]	[%]		[ha]	[m]	[m^2]	[m^2]	[m^2]
Balån River 1 Outlet	BA-1	2291	2%	11%	337	37521	87829	185738	27356
Balån River 2	BA-2	868	5%	18%	88	15754	6590	19249	2583
Southern Reference	RS-3	156	0%	3%	4	2195	0	2195	2195
Southern Clear Cut	CC-4	41	0%	56%	3	1650	0	660	660
Northern Catchment	NO-5	40	0%	33%	5	1386	0	554	554
Northern Reference	NR-7	24	0%	16%	4	835	0	334	334

* estimated from satellite data

Table ~~1~~2: Measured and modelled annual DIN loads per unit catchment area from all six Balsjö catchments during 2008-2011. The percentage of NO_3^- of the total load is given in brackets.

Measured							Modelled*	
Site	BA-1	BA-2	RS-3	CC-4	NO-5	NR-7	BA-1	BA-2
Unit / Year	mg N m^{-2} yr^{-1}	mg N m^{-2} yr^{-1}	mg N m^{-2} yr^{-1}	mg N m^{-2} yr^{-1}	mg N m^{-2} yr^{-1}	mg N m^{-2} yr^{-1}	mg N m^{-2} yr^{-1}	mg N m^{-2} yr^{-1}
2008	6.1 (60%)	6.4 (66%)	3.1 (39%)	134.8 (79%)	10.2 (42%)	6.1 (58%)	20.6 (74%)	27.1 (76%)
2009	8.0 (56%)	13.1 (72%)	7.0 (54%)	182.9 (74%)	17.2 (54%)	9.3 (57%)	24.5 (67%)	31.0 (68%)
2010	6.5 (68%)	8.9 (70%)	3.5 (46%)	149.1 (81%)	12.2 (68%)	7.9 (67%)	18.9 (75%)	24.4 (77%)
2011	8.2 (63%)	11.2 (63%)	3.9 (37%)	128.3 (76%)	14.7 (69%)	9.6 (63%)	22.1 (71%)	30.6 (73%)

*assuming conservative mixing and solute transport.

8. Figure Captions

Figure 1: The ‘Balsjö Paired Catchment Experiment’ including the catchments RS-3, CC-4, NO-5 and NR-7, as well as the two downstream sites BA-2 and BA-1 that integrate the larger 22.9 km^2 Balsjö Stream Network. Areas harvested during 2001-2011 are shown as orange. Solid blue lines represent the stream network; solid blue areas show ponds with open water. Solid black lines indicate the catchment boundaries, black pyramids the location of water sampling.

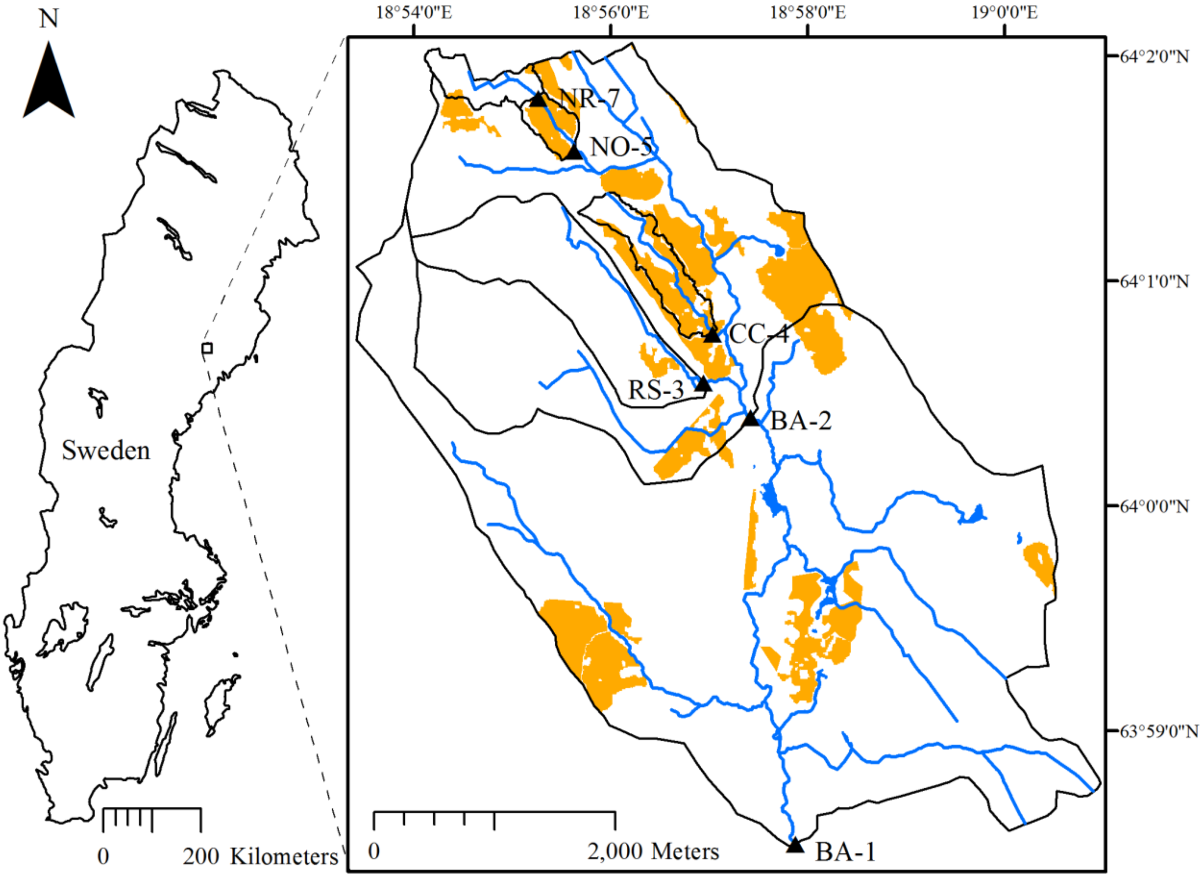
Figure 2: Comparison of modelled and measured Cl and Si concentrations for BA-1 (panel A and C), and BA-2 (B and D).

Figure ~~2~~3, First Panel: Trimonthly nitrate (NO_3^-) concentrations and standard deviations (whiskers) of two first-order streams, the clear-cut catchment (CC-4) and the reference south (RS-3), as well as for two third-order downstream sites BA-2 (size = 8.7 km^2) and BA-1 (size = 22.9 km^2). Second panel: discharge at the BA-1 outlet. Third panel: satellite derived percentage of catchment area that has been clear-cut harvested since 2001 within BA-2 and BA-1.

Figure ~~3~~4: Results of the mass-balance modeling approach for DON (left) and DIN~~NO_3^-~~ (right) for the downstream site BA-1. ~~Higher modeled than measured concentrations (above the 1:1 line) indicate a mass loss of the solute during transport downstream (and *vice versa*) assuming conservative mass transport and mixing.~~

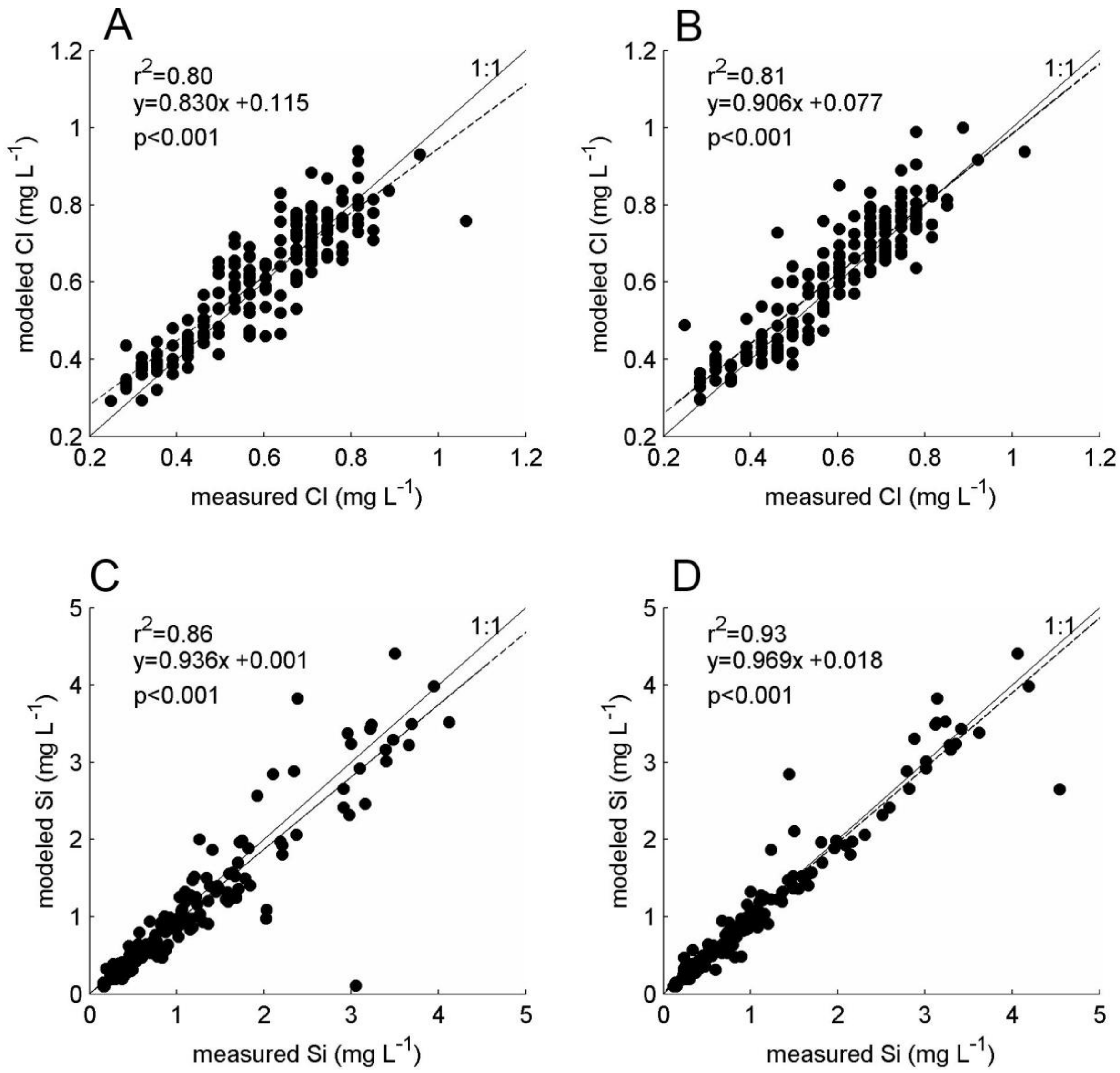
Figure ~~4~~5, Panel (A): Stream discharge (Q) and sample drawing at the BA-1 site. Panel (B): Seasonal variation in NO_3^- removal efficiency (E_r), ~~that is, the difference between measured and modeled NO_3^- concentration divided by the modeled concentration~~ for the two downstream sites BA-1 and BA-2; lines represent moving averages with n=5. Panel (C): E_r vs. Q for the BA-1 (left) and the BA-2 (right) catchment outlets.~~, respectively indicating little dependency of E_r on Q at both sites.~~

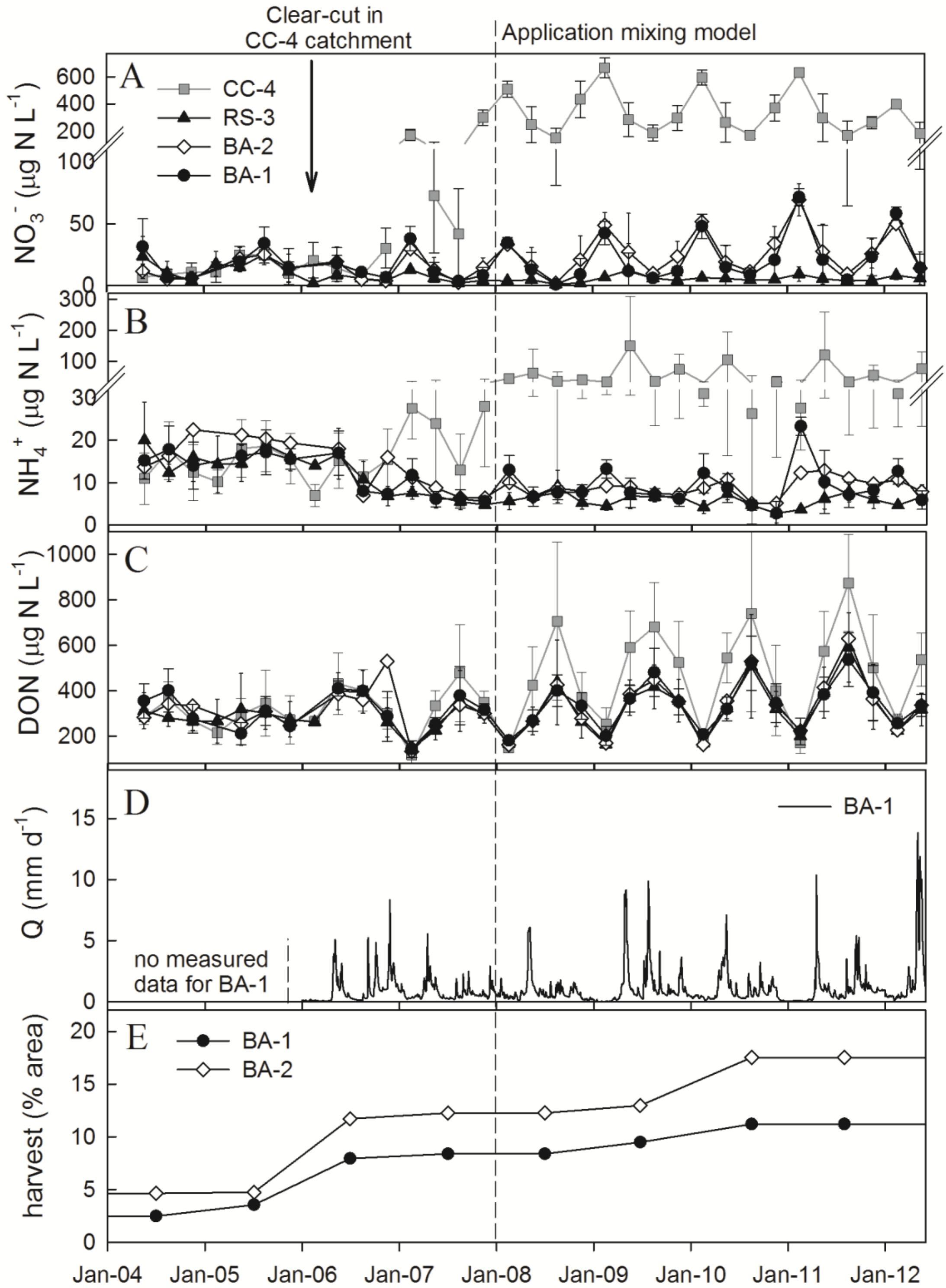
Figure 6~~5~~: Boxplot of the seasonal differences in net NO_3^- uptake rates (U) per unit stream area during 2008-2011 in the BA-2 catchment. Solid lines represent median values, boxes the 25th to 75th percentile range, whiskers the 90th to 10th percentiles and dots the 95th and the 5th percentiles. Pairs of letters indicate highly significant differences between seasons ($p<0.001$; Mann-Whitney Rank Sum Test). Values for BA-1 site are generally lower, but show similar seasonal differences.


N
Sweden
0
200 Kilometers
18°54'0"E
18°56'0"E
18°58'0"E
19°0'0"E
64°2'0"N
64°1'0"N
64°0'0"N
63°59'0"N
NR-7
NO-5
CC-4
RS-3
BA-2
BA-1
0
2,000 Meters

1859

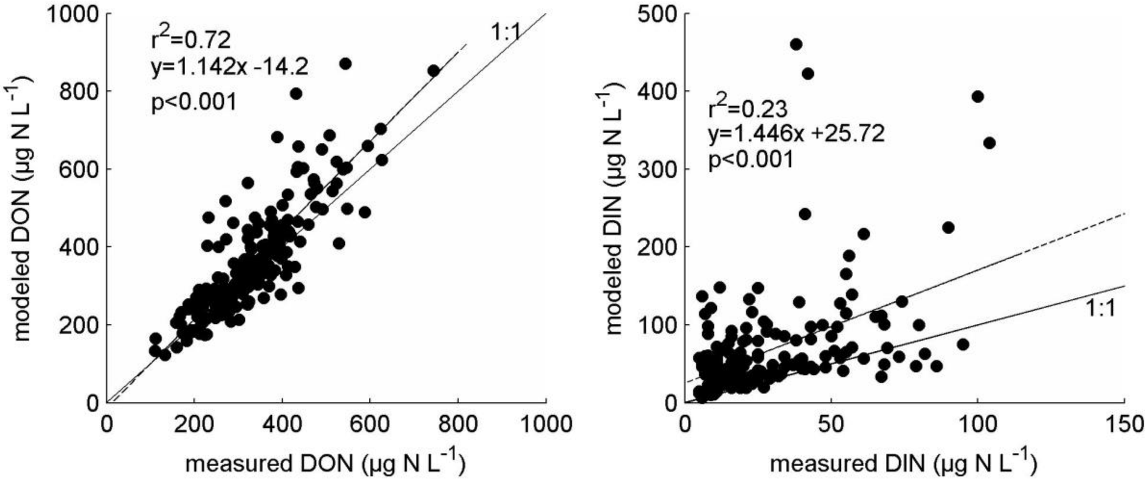
1860

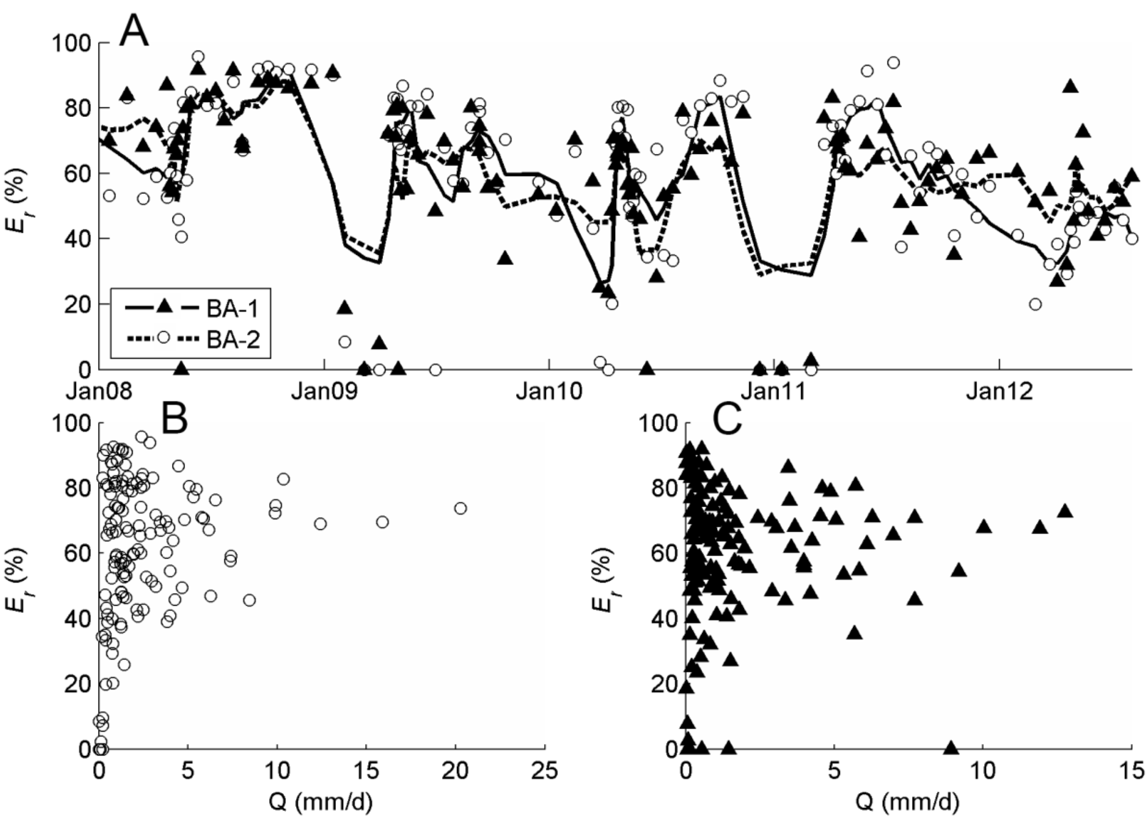

A
r2=0.80
y=0.830x +0.115
p<0.001
1:1
modeled Cl (mg L-1)
measured Cl (mg L-1)
B
r2=0.81
y=0.906x +0.077
p<0.001
1:1
modeled Cl (mg L-1)
measured Cl (mg L-1)
C
r2=0.86
y=0.936x +0.001
p<0.001
1:1
modeled Si (mg L-1)
measured Si (mg L-1)
D
r2=0.93
y=0.969x +0.018
p<0.001
1:1
modeled Si (mg L-1)
measured Si (mg L-1)

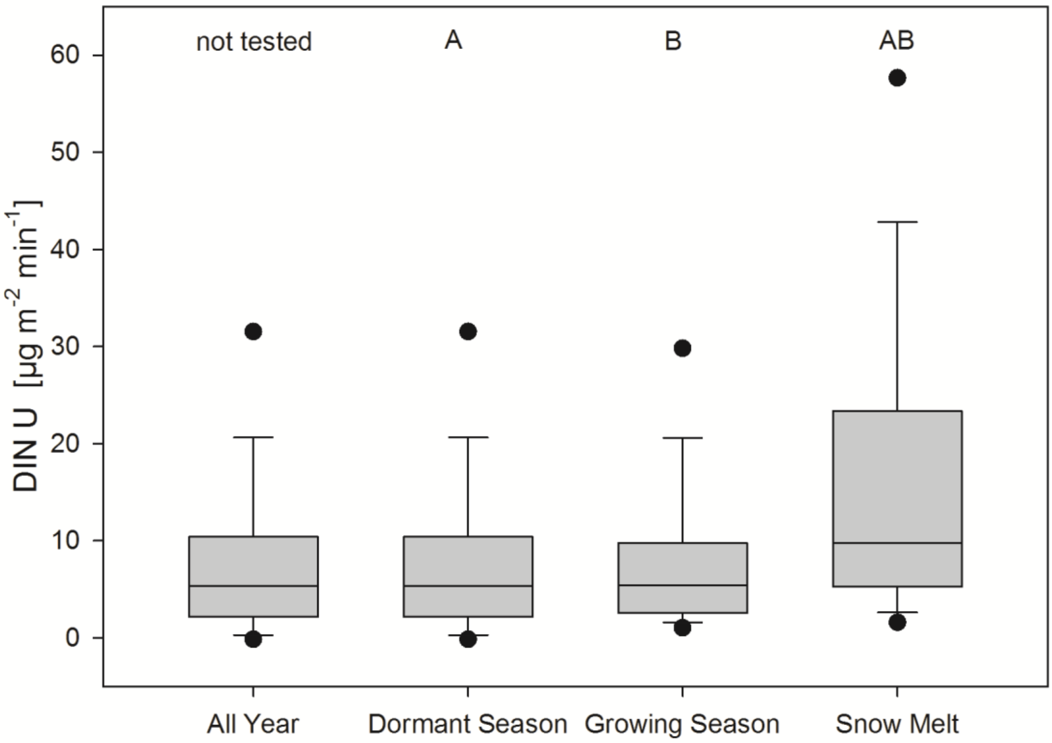

Clear-cut in
CC-4 catchment
Application mixing model
A
CC-4
RS-3
BA-2
BA-1
NO3- (µg N L-1)
B
NH4+ (µg N L-1)
C
DON (µg N L-1)
D
BA-1
Q (mm d-1)
no measured
data for BA-1
E
BA-1
BA-2
harvest (% area)
Jan-04
Jan-05
Jan-06
Jan-07
Jan-08
Jan-09
Jan-10
Jan-11
Jan-12

1863

1864


r2=0.72
y=1.142x -14.2
p<0.001
1:1
modeled DON (µg N L-1)
measured DON (µg N L-1)
0
200
400
600
800
1000
r2=0.23
y=1.446x +25.72
p<0.001
1:1
modeled DIN (µg N L-1)
measured DIN (µg N L-1)
0
50
100
150
200
300
400
500


A
B
C
E_r (%)
Q (mm/d)
BA-1
BA-2
Jan08
Jan09
Jan10
Jan11
Jan12


not tested
A
B
AB
60
50
40
30
20
10
0
DIN U [µg m-2 min-1]
All Year
Dormant Season
Growing Season
Snow Melt

1869