Author comments in response to review
"Solute specific scaling of inorganic nitrogen and phosphorus uptake in streams"

We thank the two reviewers for valuable insights that have significantly improved our paper. Below we explain how we modified the manuscript in light of the reviewers' suggestions. In addition, one particular suggestion regarding the length scaling of rivers initiated a substantial change in the paper's content. Given this intellectual contribution, we have invited Denis Newbold to expand upon this topic in the paper and to be included as a co-author. We recognize the uniqueness of this situation; we were pleased that he accepted this offer. In summary, we feel that the revised manuscript is much stronger than the previous version. Below we detail our revisions in response to reviewer comments:

Please note: Reviewer comments are itemized in roman font, while *author replies are in italics.*

Reviewer 1
This paper is an interesting analysis of scaling of nutrient uptake in streams. The analysis is generally sound and the paper is nicely written and easy to follow. I think this paper is well suited to Biogeosciences and it has the potential to be quite important in helping move the field from the reach scale at which nutrient uptake studies are conducted to predictive, larger scale modeling efforts. I do see several areas where the analysis needs to be strengthened and reconsidered. Below I highlight a few 'major' issues followed by other miscellaneous comments.

Scaling relationships: You have decent relationships for NH4 and SRP (r2 = 0.57), but the relationship for NO3, while statistically significant, is extremely weak (r2 = 0.13). You mention there was more variation for it than for the others, but it pretty much gets swept under the rug in the discussion. I'm not convinced there is much of a relationship here considering there is such a poor correlation between Sw and Q/w. At a minimum you need to consider why the relationship was so weak for NO3 and what the implications are.

We agree that the relationship of Q/w with NO_3 S_w is weak in 2 senses: r^2 is low and the confidence interval (CI) for slopes is wider than for the other solutes. Here we focus our interpretation based on the values and uncertainty estimates of the slopes of the allometric regressions. We think that the low coefficient of determination is due to in background NO_3 concentrations driving variation in uptake. We have added the effect of concentration to the paper (explained in more detail below).

Reference vs. Altered systems: This is an interesting comparison, but there are several problems in this analysis. The r2 values are extremely low for nitrate and SRP-altered making it questionable as to whether there is even a relationship worth pursuing. In addition, comparing the regression of reference and altered P is misleading because you have a much larger range in Q/w for reference (_0.02-10) vs. altered (_0.5-10). If you are going to compare the two for SRP you should use only the overlapping ranges of Q/w (you should probably do the same for NH4 as well).

NO_3: The low r^2 does not hide the fact that the CI of the values of the intercepts do not overlap, showing that for this data set, altered streams have longer S_w than the reference streams. Low r^2 means that something else in addition to Q/w influences NO_3 uptake length. We hypothesize that land use is one of these factors and likely background solute concentration is another. We address this point in more detail below in the context of the concentration comment.

SRP. Good point. We re-ran the analysis using reference streams that had Q/w in the same range as the altered streams, and as before, the parameter estimates were not significantly different from each other. We added discussion of this point to the text in the results.

NH_4. Again, we re-ran the analysis using reference streams that had Q/w in the same range as the altered streams. As before, the slope was not different, but the intercept was significantly higher. We have added this point to the text.

Uptake/metabolism: The analysis and integration of metabolism and uptake is not particularly compelling. In the introduction you hypothesized that N and P would be decoupled because P is subject to abiotic sorption and N is more tightly tied to metabolism. That seems fairly reasonable except for the tendency for NH4 to adsorb. In the results you state carbon demand drove inorganic N uptake. You can show that N uptake was correlated with metabolism, but you can't show causation. More importantly, while the correlation was statistically significant for nitrate, the r2 value (0.04) was low to the point of being a largely irrelevant relationship. Ammonium was better (r2=0.27), but still not great. In the discussion, the metabolism argument focuses on NH4 and ignores NO3. Based on your initial hypothesis NO3 should be best predicted by metabolism because it's the least likely to be influenced by abiotic sorption, so why ignore it? Is it because the relationship is poor? In the end, the metabolism issue is a rather muddled mix of weak analysis of the data and what appear to be pretty weak relationships. I wonder if part of the issue is that you're stacking uptake velocity (uptake efficiency) against metabolic rate (basically O2/carbon flux per unit area and time). Presumably the relationship would be better as uptake rate vs. metabolic rate as they are measuring the same thing – nutrient demand to support metabolic demand. A second issue may be the integration of ER which may partially result in nutrient demand (e.g. for bacterial/fungal growth) but is perhaps more coupled to nutrient mineralization rather than uptake. GPP should be more directly linked to nutrient uptake rate as they both build biomass.

Based on this reviewer's comment we have deleted the metabolism analyses from the paper. We do this for 3 reasons, 1), we agree with the criticism above, 2) the effects of metabolism were better covered in other studies, and 3) the effects of metabolism on P uptake is based on only 25 studies which is far less than the number of studies for N *solutes.*

Coherence in the introduction and discussion: To some extent the discussion didn't really follow well from the introduction. In the discussion you set up the isometric vs. allometric scaling models, but there is nothing about this in the introduction to set it up. I think the paper would read better if you set this up better in the introduction. Perhaps you could set this up better when you talk about Wollheim's constancy assumption.

We appreciate the observation and suggestion and we added text in the introduction linking the constancy assumption with isometric and allometric scaling.

Nutrient concentration: A sleeper in all this is nutrient concentration which you pretty much disregard. On p. 6676 l. 10 you note the assumption of constant biological demand (U? Vf?) relative to concentration is needed for isometric scaling. Was concentration related to specific discharge in your data? Did it have any role in what's going on? You note the issue in just one sentence in the discussion (p. 6682 l. 15) but that's it.

This is a very good point that contributed significant subsequent revisions to the manuscript and further data analyses. We addressed the effect of concentration on S_w for NO_3 in Hall et al. 2009b, and skipped over it here. We combined concentration and Q/w via multiple linear regression (as far as we know there is no such thing as multiple SMA regression, so the parameter estimates are not really comparable). For all 3 solutes, concentration increased S_w and increase r^2 of the model. We put this information in the text and added a table with the parameter estimates. For NH_4 and SRP, there was no relationship between concentration and stream size. NH_4 concentration increased with stream size; this increase has the effect of lengthening S_w with stream size, yet NH_4 scaled isometrically. The important finding is for NO_3, where adding concentration improved model fit and was itself a strong predictor of S_w.

Miscellaneous comments: p. 6674 l. 21 – 'removing' is a term that gets (mis)used all the time especially in regards to uptake vs. net retention and/or true removal (e.g. the arguments over Cardinale's recent Nature paper). Would it be better so simply stick with uptake here and elsewhere since that's technically what your data are?

We have received conflicting advice over the years on whether to write removal or uptake. 'Uptake' implies that we know where the molecule goes, whereas 'removal' simply means that it has been removed from the water column, thus we think it is a more conservative word. On the other hand, we understand how removal can also imply 'gone from the stream' which of course may not happen at all except in the case of denitrification. In no way do we want to imply 'removed from the stream' forever (e.g., permanent sink). We prefer the usage of uptake and have gladly changed it throughout the paper. We did leave the word removal when defining uptake length so not to define uptake with the same word.

p. 6675 l. 2 and 6 – vf probably measures abiotic as well as biotic demand in most spiraling studies.

Yes, for sure for SRP and NH_4. Deleted biotic and added chemical next to biological.

p. 6676 l. 14 – how did you test for differences in slope and intercept? Perhaps a bit more detail.

We added some detail.

p. 6676 l. 24 – note the range in Q/w here

Yes, and we re-ran the stats with a limited range (as described above in detail)

p. 6676 l. 26 – start new paragraph for metabolism

Done.

p 6679 l. 11 – contrasting N and P uptake – You compared NH4 and NO3 independently to P to contrast N and P uptake. Did you try combining NH4 and NO3 to make this simpler? You could calculate Sw-N by combining Sw of NH4 and NO3 proportionally to their concentrations. Considering both forms of N can/will be used alongside P demand during uptake it makes sense to combine them. Also, you again ignore the r2 values on these which are really weak (Is the r2 = 0.082 for the top panel correct?). I think the real story here is th*at there simply is very little relationship between uptake of the two forms of N and uptake of P. That has been shown before.*

We agree that these r^2 values are very low. but low r^2 does not make the relationships non-meaningful. Rather, we believe it says that something besides Q/w controls the ratio of N to P uptake, which is something we do not dispute. We do not intend to use a relationship such as these to predict N:P uptake vs. stream size (that would be nearly useless), but rather to corroborate the findings from Fig 1, i.e. as stream size increases uptake of P decreases relative to N.

We like the idea of using DIN in addition to NO_3 and NH_4. The trouble is that practitioners rarely add NO_3, NH_4, and PO_4 to a stream as part of a single uptake experiment. Unfortunately, we were left with 18 points for DIN, vs. 57 points for NH_4 and NO_3, and thus statistical power was too low to detect a trend.

p. 6679 last paragraph – I don't see how you are pinning the variation in m on variation in b. You included variation in several components of eqn 10, including error in the slopes (a) from your SMA's. In the discussion (p. 6683 l. 8) you argue variation in m was solely due to variation in hydraulic geometry. Doesn't it also include the variation in the slopes of you SMA's and therefore include uncertainty in the average value of m? Perhaps you can tease out how strong the effect of the variation in 'a' was in calculating m.

We were unclear in saying that error in m *was due solely to that in the hydraulic geometry exponent* a*. We recast this sentence and explained that of all of the variables in eq 10,* a *was the most variable simply because the variability in shapes of channels. We also rewrote this paragraph based on comments from Denis Newbold.*

Discussion paragraph 2 – this analysis of the human influence seems jammed in here out of place

We moved this paragraph and combined with concentration effects.

p. 6682 l. 1 – why wasn't biological demand related to nitrate (you have previously shown it is for some streams. . .).

We added that point to the subsequent paragraph on why we expect NO3 to cycle differently than PO4. That makes this paragraph more parallel with the preceding paragraph on sorption.

p. 6682 l. 6 – differences in scaling relationships between SRP and DIN? Nitrate was the same as P (at least for the slope estimate, if not for the actual strength of the relationship) and you then note this a few sentences later. . .? It seems you need to do a better job separating out NH4 vs. NO3 in the discussion.

We recast the topic sentence to focus on NO_3.

p. 6682 l. 20 – Here you spend a paragraph arguing that your data suggest large streams and rivers are important sites for nutrient uptake based on your scaling relationships.
Two paragraphs later (p. 6684 l. 10) you explicitly note that larger systems are likely to be fundamentally different and that scaling relationships in smaller systems will not hold in larger systems. This needs to be better written to avoid the obvious contradictions.

The conclusion that rivers will be important sites of nutrient uptake hinges on the assumption that they are simply "big streams." We mention this assumption in both places identified above, and explicitly address it in the end of the paper.

Reviewer 2, *Denis Newbold*

This paper rather elegantly shows that, to a first approximation, spiraling length scales directly with river length while nutrient uptake measured as a mass transfer coefficient ("*vf*") varies relatively little throughout a basin. Individual nutrients--ammonium, nitrate, and dissolved reactive phosphorus (SRP)— each vary from this template in their own way. The analysis is framed in a theoretical context, tying in Hack's law and the power law relationships of channel geometry. I believe this is an important paper that has both theoretical significance as well as practical utility. I would like to see substantial revision. Most of my comments below are relatively minor and easily addressed. One that is noteworthy is that I think the interpretation of Alexander (2000) is incorrect—that their results (especially as updated in a 2008 paper)—are actually consistent with the result of this paper.

My major issue, fully elaborated below, is that your inferences about the nutrient uptake of large rivers, relative to small streams, are not clear and, worse, are not justified. I think you can easily clarify them and I suggest an extension of your analysis that would provide the needed justification.

I have a concern about using the word “remove,” because it obscures the notion of cycling and conveys the often wrong impression that nutrient once “removed” upstream does not reappear downstream. The spiraling literature hosts some confusion over this issue. I admit that the expression “to take up” has its weaknesses, especially in that it appears to exclude physical sorption. “Transfer” is also awkward. I suggest minimizing the use of “remove”, especially where it is potentially misleading. It is good that the first two invocations of “remove” are followed by “from the water column,” which constrains the interpretation. I also suggest that, early on, you add a clarification such as: “Nutrient uptake does not directly imply a reduction in downstream transport because of compensating nutrient regeneration. Uptake may reduce downstream transport temporarily during periods of benthic biomass accumulation, or on an ongoing basis through conversion to another dissolved form or to a particulate form, or in the case of denitrification by transferring nitrogen to the atmosphere.”

Both reviewers made this point and we have removed the word ‘removed’ except for “removed from the water column” when defining uptake. We added the caveat of uptake not equaling net removal, which Reviewer 1 also pointed out, and also note that there remains a bit of confusion in the literature regarding this terminology.

6673:2 remove period after “network”

Done.

6673:10 I suggest (request) changing the sentence to read:
Uptake length is estimated from the inverse of a first-order uptake rate of nutrients experimentally added to the water column of a stream, either as a tracer (Newbold et al., 1981), as a small increment to ambient concentration (Stream Solute Workshop, 1990).”

Done.

6673:21 I suggest removing “removing” by rewriting the sentence as: “The role of small streams in taking up dissolved nutrients has been well studied, with > 970 measurements of uptake length (Ensign and Doyle, 2006; Tank et al., 2008).”

Done.

6673:25 “initial evaluation of the degree to which large streams and rivers may regulate downstream transport of nutrients.” I find this problematic because it evokes the “removal” confusion, but if a clarifying caveat, such as I suggest above, has been presented prior to this, I have no fundamental objection.

We recast to:
“Considering how nutrient uptake scales with stream discharge allows an initial evaluation of the degree to which large streams and rivers may take up nutrients from the water column.”

6674:3 “Although there are much [many?] data”

Changed to "many".

6675:9 I suggest introducing the exponent, *a*, here: “…prediction of *SW* µ *(Q/W)a* where *a*=1.” Then also, in Table 1, identify the “slope” as the exponent, *a.*

Done.

6676:18 Clarify the citation to Vannote et al., perhaps by inserting, after “river,” , “i.e., along the river continuum”. As it stands, the uninitiated reader would have no idea why Vannote is being cited here.

Done.

6676:20 Why say "we predict"--isn't this what was just found? How about, "Based on the preceding analysis, nutrient uptake length (SW) will scale.. “

Done.

6678:10-11 It would be helpful to report the standard deviations for Hack’s constant, *h,* and for *c*, as you did for *b*.

We added this information.

6679:12 “SRP, thus” Run-on sentence (comma splice). Use semicolon or two sentences.

We added a semicolon and change thus to therefore.

6679:23- 6680:5. This paragraph is poorly written. Rewrite to resolve the following issues.
1. Topic sentence is not topic and is redundant on third sentence.
2. Rationale for Table 1 reference is unclear.

3. Second sentence is missing articles, “the”, and “a”
4. m is given as 0.79 once, then again as 0.8

1. We rewrote the topic sentence. 2. We deleted the reference to Figure 1. 3. We added the articles. 4. 0.8 is the correct value. We brought up the within-solute variation of m at the end of the paragraph and not the beginning or middle.

6680:11 Insert “length” after NH4.

Done.

6680:11-12. Define the distinction between isometric and allometric. E.g., insert (“i.e., *a*=1)” after “isometrically.” The second part of the sentence is stated imprecisely and could be written, “while uptake lengths for NO3 and SRP scaled allometrically (a>1), indicating a declining demand (*vf*) with increasing Q/w.

We made these changes.

6682:21 insert “(0.79-1.06)” after “1”.

Done.

6682:21-25 These lines refer to variation in *vf* with stream size and so belong in the previous section. The sentence about Ensign and Doyle (2006) begins with a statement about length, and is therefore appropriate to the section, but ends with a statement about uptake in large rivers, which belongs in the previous section.

We moved the v_f sentences to the first paragraph of the discussion.

6682:22 I suggest changing “remove” to “take up”.

Done.

6683:1-4. The paragraph started (previous page) by implying that the relation between *SW* and *L* is isometric (near 1), but in this sentence (line 2) it is allometric, while in the next sentence (line 6) we learn that for two of the nutrients it is isometric. You need to sort this out. It probably should be explicitly explained that the geomorphic parameters, *b*, *h*, and *c* combine to suggest an allometric scaling, which carries through to an allometric *SW -L* scaling for ammonium because its scaling with *Q/W* is isometric, but combines with a compensatory allometry in *SW* -(*Q/W*) for nitrate and SRP, to produce the isometry in *SW -L* for these nutrients.

We recast these sentences to make clear the difference in S_w vs. L for the different nutrient species.

6683: 34 “*SW* increases more slowly than does distance” lacks the necessary precision, because it could be true even under isometry. You might say “increases less than proportionately”.

We made this change.

6683:5 I suggest changing “remove” to “take up”.

Done.

6683:4-6 “This finding [m<1] suggests that large streams and rivers may remove as much or more nutrients than the well-studied small streams.” Perhaps it does “suggest” this, but it is not clear what is being suggested. On an areal basis (and assuming uniform concentration) the river takes up more ammonium, and about the same nitrate and SRP as the stream, but this is based on the vf and not the length scaling. On a per unit length basis, the river takes up more, but this is just because the river is wider. On a per unit length basis relative to downstream flux, the river takes up much less, because SW is longer. I think what you are trying to say is that a river takes up the same or more, relative to flux, *over the length of the river.* But you can’t really say this without saying something about how rivers are longer than streams. What’s missing is some way of scaling river length, specifically that length scales logarithmically. This concept is captured by stream order and Horton’s law of stream segment lengths. I suggest the following approach: Ensign and Doyle (2006), as you noted, showed that each order cycles nutrients about the same number of times, but they showed this using empirically derived order-specific SW . What you want is Ensign and Doyle’s result, but from a theoretical basis, tied to your allometry of SW. Here is how you might do it:
Ensign and Doyle (2006) calculated the number of cycles, ρ_i in a stream order, i, as:
$\rho_i = \mathbf{l}_i / SW_i$ (1)
where $\mathbf{l}_i$ is the mean channel length of streams of order i, and SW_i is the respective uptake length.
However, Equation (1) is an approximation because SW increases with stream length within the reach. For isometrically increasing SW, we can represent the incremental contribution to the number of cycles within a reach at stream distance, L, from the source, as
(*equation in .pdf of review*) (2)
where $\alpha = SW(L)/L$, and is constant due to the isometry. The number of cycles in stream order i can then be expressed more precisely as:

(*equation in .pdf of review*) (3)

where L_i is the average length of streams of order i, using Horton’s definition of stream order, i.e., measured from the source. The ratio L_i/L_{i-1} is Horton’s (1945) length ratio, which he showed to be uniform across stream order. Therefore, Eq. (3) shows that, for isometric scaling of uptake length, a nutrient cycles the same number of times in each order, as Ensign and Doyle (2006) empirically observed. Note that although the length ratio is expressed using Horton stream orders, i.e, using full mainstem lengths (as did Horton originally), the result applies to a Strahler-order segment, i.e., measured from the

confluence of two lower (Strahler) order segments. Measured across a drainage, the two systems yield nearly identical length ratios (Scheidegger 1968).
The result from Equation (3) allows you to state meaningfully that a large river takes up as much (or more, in the case of ammonium) nutrient than a small stream, relative to its upstream inputs.

We have effectively added this text in the discussion and expanded this idea based on Denis Newbold's contribution to the paper.

6683:13-23. Alexander et al. (2000) did not actually demonstrate that "most .. in small streams." They showed that a small percentage (<40%) of the N originating in small streams makes it to the Gulf while a large percentage ("some more than 90%) of N originating in or near large rivers reaches the Gulf. Alexander et al. (2008) updated the analysis, finding (p. 826) the small streams delivered more (a median of 57%) than reported in the 2000 paper. But the main point is that nitrogen originating in small streams was removed not necessarily in the small streams themselves, but throughout the network as it flowed to Gulf. Alexander et al.'s (2000, 2008) results are, in fact, consistent with a constant removal per stream order.
Suppose the removal rate was 7% per order. The Mississippi River is about order 11 (one order higher than represented by Table 5-2 of Leopold et al 1964 because their first-order was probably second order). Consider the minority of first-order streams that run through the entire hierarchy of first-to-11th order. Of the nutrient input to a first order stream that then passes through the full hierarchy of 11 orders, 45% would reach the Gulf. For a pathway that skips three of these orders, the delivery would be 75%. Many of the distal watersheds shown in Fig. 3A of Alexander (2008), except for the arid northwest fall into this delivery range. Alexander (2008, p. 825) stated, "The largest percentages (>90%) occur in the largest mainstem rivers and their tributaries." These would be nutrients that pass through only 1 or 2 orders. At our assumed loss rate 7% per order, the delivery of a nutrient passing through two full orders would be 87%. Thus, the assumption of a constant per-order loss rate is consistent with the results reported by Alexander and their results do not show that most nutrient uptake occurs in small streams.

We removed the comparison to Alexander et al. based on this comment.

Denis Newbold