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Technical Note: A comparison of two empirical approaches to estimate in-stream nutrient net uptake

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Abstract

To establish the relevance of in-stream processes on nutrient export at catchment scale it is important to accurately estimate whole-reach net nutrient uptake rates that consider both uptake and release processes. Two empirical approaches have been used in the literature to estimate these rates: (a) the mass balance approach, which considers changes in nutrient loads corrected by groundwater inputs between two stream locations separated by a certain distance, and (b) the spiralling approach, which is based on the patterns of longitudinal variation in ambient nutrient concentrations along a reach following the nutrient spiralling concept. In this study, we compared the estimates of in-stream net nutrient uptake rates of nitrogen (N) and the associated uncertainty obtained with these two approaches at different ambient conditions using a data set of monthly samplings in two contrasting stream reaches during two hydrological years. The rates calculated with the mass balance approach tended to be higher than those calculated with the spiralling approach but only at high ambient N concentrations. Uncertainty associated with these estimates also differed between both approaches, especially for ammonium due to the lack of significant longitudinal patterns in concentration. The advantages and disadvantages of each of the approaches are discussed.

1 Introduction

Understanding the relevance of in-stream uptake on nutrient loads has become an important question over the past decades due to the need to establish reliable nutrient budgets at catchment scale and to evaluate the impact of downstream nutrient export on coastal ecosystems (Behrendt and Opitz, 2000; Alexander et al., 2000; Wollheim et al., 2008). Several studies have shown that in-stream processes can have a significant influence on nutrient downstream transport, especially in headwater or relatively small-size streams (Alexander et al., 2000; Peterson et al., 2001; Bernhardt et al., 2003; Ensign and Doyle, 2006; Mulholland et al., 2008). Noteworthy, results from most of these studies are derived from estimates of gross nutrient uptake, which may overestimate the net influence of streams on nutrient downstream export because they do not consider processes associated to release of nutrients from biota to water column. Release processes (e.g. mineralization, nitrification, desorption), however, can be relevant especially in highly heterotrophic streams, which are common in headwaters (Battin et al., 2008). These processes may counterbalance to some extent nutrient uptake processes (e.g. assimilation, denitrification, and adsorption) in such streams or even result in a net downstream release of nutrients. For instance, Brookshire et al. (2009) found no significant longitudinal patterns in ambient concentration (i.e. net uptake rates ~ 0) in about 80% of a selection of stream reaches from several biomes, and concluded that in-stream processes may be commonly irrelevant for watershed nutrient balances because in-stream nutrient uptake is rapidly balanced by nutrient release. Nevertheless, Brookshire et al. (2009) also suggested that streams could act as a net sinks of nutrients (i.e. positive net uptake rates) under certain environmental conditions that favour denitrification or net biomass growth. Taking all this into consideration it becomes clear that, besides of characterizing stream ecosystems in terms of gross nutrient uptake rates, it is also important to estimate net nutrient uptake rates that provide more accurate information on actual nutrient export from a given stream reach and the relevance of in-stream processes at catchment scale.

In-stream net uptake rates integrate both uptake and release processes occurring along a reach, and can be positive (uptake $>$ release), negative (uptake $<$ release) or nil (uptake $\sim$ release). These rates have been commonly estimated using a mass balance approach considering changes in nutrient loads (i.e. nutrient inputs minus outputs) between two stream locations separated by a certain distance (Meyer and Likens, 1979; Bernhardt et al., 2003; Roberts and Mulholland, 2007; Niyogi et al., 2010). In this approach, values of nutrient fluxes from groundwater to the stream are needed to accurately estimate the net balance only associated with uptake and release processes (i.e. in-stream net uptake rates). When groundwater data are not available, a sensitivity analysis can be done using a range of potential values to examine the relevance of this nutrient source on estimated net nutrient uptake rates (Roberts and Mulholland, 2007).

Alternatively, in-stream net uptake rates can be estimated from patterns of longitudinal variation in ambient nutrient concentrations along a reach following the nutrient spiralling concept (Martí et al., 1997, 2004). Rather than assuming a linear trend of stream nutrient concentration between two locations, net uptake rates obtained with the spiralling approach are derived from the fit of ambient nutrient concentrations along the reach to an exponential equation (i.e. first-order reaction model). This approach integrates all the uptake and release processes occurring along the stream reach as well as the nutrient groundwater inputs, so that deviations from the fitted model can be treated as a measurement of the uncertainty associated to the processes occurring along the stream reach.

The aim of this study was to compare the estimates of in-stream net nutrient uptake rates and the associated uncertainty obtained from the mass balance and the spiralling approaches aforementioned. To do so we took advantage of a data set from monthly samplings along a longitudinal transect in two contrasting stream reaches during two hydrological years, which allowed us to calculate net uptake rates with both methods under distinct hydrological and environmental conditions. The advantages and disadvantages of each method are discussed in view of the results from this comparison.

2 Methods

Data for this study were collected in two forested headwater streams located in Catalonia (NE Spain): Santa Fe del Montseny and Fuirosos. The two streams have contrasted hydrologic regimes and dissolved inorganic N (DIN) concentrations (von Schiller et al., 2008). Santa Fe has permanent flow year round, whereas Fuirosos has intermittent flow regime with summer no flow periods of variable duration among years. During the study period, DIN was dominated by nitrate (NO_3) in both streams, but the concentration was higher and expanded a wider range of values in Fuirosos (mean $\pm$ SD = 368 $\pm$ 397 µg N l^{-1}, range = 35 to 1468 µg N l^{-1}, $n = 20$) than in Santa Fe (mean

$\pm$ SD = 125 $\pm$ 83 µg N l^{-1}, range = 12 to 321 µg N l^{-1}, $n = 25$). The concentration of ammonium (NH_4) was low (i.e. mean < 15 µg N l^{-1}) and showed no clear temporal pattern in both streams. More detailed information on the biogeochemical properties of these streams can be found in Bernal et al. (2005) and von Schiller et al. (2008).

Representative reaches of 140 m in Santa Fe and 80 m in Fuirosos were selected. We collected water samples for ambient concentrations of NO_3 and NH_4 and measured conductivity at eight locations along each reach. Discharge (Q, in l s^{-1}) was estimated based on a mass balance approach by conducting short-term constant rate additions of a hydrological tracer (i.e. NaCl) and using the time-curve conductivity data recorded at the bottom of the reach (Gordon et al., 2004). This method also allowed estimating variation of Q along the reach due to groundwater inputs. Wetted width (a, in m) was recorded at each sampling location and averaged to provide a value for the entire reach. Field samplings were conducted monthly from September 2004 until August 2006, except in Fuirosos during no flow conditions. A total of 25 and 20 longitudinal samplings were done in Santa Fe and Fuirosos, respectively. Water samples were analyzed for NO_3 and NH_4 concentrations following standard colorimetric methods (von Schiller et al., 2008).

We calculated net uptake rates (U, in µg N m^{-2} s^{-1}) of each DIN form (NO_3 and NH_4) for each sampling date using two alternative approaches: the mass balance approach and the spiralling approach.

The net uptake rates with the mass balance approach (hereafter referred to as U_{MB}) were calculated using ambient N concentrations from the sampling locations at the top and the bottom of the reach. We took into consideration the groundwater N inputs as an additional N source influencing the variation of N mass between the two sites following Roberts and Mulholland (2007). The U_{MB} was calculated using the equation:

$$U_{MB} = [(N_{top} \cdot Q_{top}) - (N_{bot} \cdot Q_{bot}) + (N_{gw} \cdot Q_{gw})]/(x \cdot a) \qquad (1)$$

Where N is the concentration of NO_3 or NH_4 measured at the top (N_{top}) and bottom (N_{bot}) of the reach and in the groundwater (N_{gw}) in µg N l^{-1}, and x is the length of the reach between the top and the bottom sites in m. Q_{gw} was calculated as the difference between Q_{top} and Q_{bot}. U_{MB} was estimated assuming that N_{gw} was equal to the average surface water concentration. Confidence intervals (CI, in µg N $\mathrm{m}^{-2}\,\mathrm{s}^{-1}$) of U_{MB} were calculated based on the uncertainty associated with N_{gw} by assuming that it could range from 0.5 to 2 times the average surface water N concentration (Roberts and Mulholland, 2007). The CI was then calculated as the difference between the upper and the lower limit of U_{MB}. We assumed that U_{MB} was not significantly different from 0 when the upper limit was positive and the lower limit was negative.

The net uptake rates from the spiralling approach (hereafter referred to as U_{SP}) were calculated using the longitudinal variation in ambient N concentration along the reach. Stream ambient concentrations were corrected by the longitudinal variation in ambient conductivity to account for groundwater dilution based on the method described by Martí et al. (2004). This method is an adaptation of the method used to estimate nutrient spiralling metrics using short-term nutrient additions (Newbold et al., 1981; Valett and Webster, 2006). From the longitudinal patterns in ambient concentrations, we estimated a net uptake coefficient per unit of reach length (k_w, in m^{-1}) by solving the equation:

$$N_x = N_{\mathrm{top}} \cdot (C_x / C_{\mathrm{top}}) \cdot e^{-k_w x} \tag{2}$$

Where N is the ambient concentration of $\mathrm{NO_3}$ or $\mathrm{NH_4}$, and C is the ambient conductivity in $\mu\mathrm{S\,cm}^{-1}$ at the top of the reach and at the downstream sites located x m from the top of the reach. U_{SP} was then calculated using the following equation:

$$U_{\mathrm{SP}} = (Q \cdot N_{\mathrm{avg}} \cdot k_w)/a \tag{3}$$

Where N_{avg} is the average of the N ambient concentration measured at the eight locations along the reach, Q is the average discharge along the reach, and a is the average wetted width. We estimated the CI of U_{SP} based on the k_w range obtained using the ±95% confidence interval of the regression between dilution-corrected ambient N concentration (i.e. ln N_x) and the downstream distance (x) derived from Eq. (2). The value of U_{SP} was not statistically different from zero when the regression was not significant (i.e. $p > 0.05$). The CI (in µg N m^{-2} s^{-1}) for U_{SP} was estimated as the difference in the U_{SP} values between the upper and lower 95% confidence intervals. We assumed that U_{SP} was not significantly different from 0 when the upper limit was positive and the lower limit was negative.

3 Results and discussion

The net uptake rates calculated using the mass balance and the spiralling approaches fell close to the 1:1 line for both DIN forms (Fig. 1). For NO_3, however, the values of U estimated with the mass balance approach (mean ± SD = 0.21 ± 1.19 µg N m^{-2} s^{-1}, $n = 45$) were on average two times higher than those obtained with the spiralling approach (mean ± SD = 0.10 ± 1.13 µg N m^{-2} s^{-1}, $n = 45$) (Wilcoxon paired test, Signed-Rank = 239.5, $df = 44$, $p < 0.01$). The discrepancy in the estimation of U between the two approaches emerged under high NO_3 ambient concentrations. We found that absolute differences between the U for NO_3 calculated with the two approaches augmented as stream NO_3 concentration increased (Spearman Rho coefficient = 0.71, $df = 45$, $p < 0.01$) (Fig. 2a, white squares). In fact, there were no differences between U_{MB} and U_{SP} for those cases in which stream NO_3 concentrations were $<\sim 300$ µg N l^{-1} (Wilcoxon paired test, Signed-Rank = 92, $df = 34$, $p > 0.05$). Consistent with the findings for NO_3 in which differences between the two approaches were small when stream N concentrations were low, the values of U for NH_4 (in all cases ambient concentration < 30 µg N l^{-1}) obtained with the two approaches showed no differences (Wilcoxon paired test, Signed-Rank = -60.5, $df = 44$, $p > 0.05$) and did not exhibit any consistent pattern in relation to stream ambient concentrations (Fig. 2b).

Furthermore, the mass-balance and the spiralling approach differ in a basic methodological assumption. While the mass balance approach assumes that the concentration of N changes linearly between the sampling stations located at the top and bottom of the study reach, the spiralling approach assumes an exponential change of concentration with distance downstream. Thus, assuming constant discharge along the reach (i.e. $Q_{gw} \sim 0$), it can be derived from Eq. (1) that U_{MB} is directly dependent on the difference in N concentration between the two sites (i.e., $N_{top} - N_{bot}$). Following the same assumption, and considering the simplest case with only two stations along the stream reach (top and bottom), it can be derived from Eqs. (2) and (3) that U_{SP} depends on the average N concentration as well as on the ratio in N concentration between the two stations (i.e. $N_{avg} \cdot \ln(N_{top}/N_{bot})$). Based on these assumptions, we used a sensitivity analysis to examine how the difference between U_{MB} and U_{SP} changed as a function of: (a) the average stream water N concentration and (b) the ratio in N concentration between the top and bottom stations (Fig. 3). Results from this analysis supported our empirical results, showing that the difference between both approaches tends to increase with increasing average N concentration. Moreover, our sensitivity analysis indicates that the effect of N concentration on the $U_{MB} - U_{SP}$ difference is magnified as the N_{top}:N_{bot} ratio increases.

Measurements of uncertainty differed between the mass balance and the spiralling approach for both DIN forms (Fig. 4). For NO_3, the confidence intervals calculated with the mass balance approach were larger than those estimated with the spiralling approach (Fig. 4a; Wilcoxon paired test, Signed-Rank = 249, $df = 44$, $p < 0.01$). The sensitivity analysis performed for U_{MB} indicated that the estimated rates could vary strongly depending on the N_{gw} scenario considered. The U_{MB} for NO_3 decreased on average 3.8 ± 9.7 times when the NO_3 concentration in groundwater was half that of surface water, and it increased 8.2 ± 19.3 times when the NO_3 concentration in groundwater was doubled with respect to surface water. In contrast, the U_{SP} for NO_3 varied only 2.7 ± 1.5 times due to the uncertainty associated with this approach. Despite of differences in the uncertainty associated with each approach, our results indicated either net in-stream uptake ($U > 0$) or release ($U < 0$) of NO_3 (i.e. non-equilibrium of net in-stream processes) in 40% and 51% of the cases for the mass balance and the spiralling approach, respectively. These percentages suggest a potentially high relevance of in-stream processes on NO_3 export, in contrast to findings from some previous studies (e.g. Brookshire et al., 2009).

In contrast to NO_3, the confidence intervals calculated for NH_4 with the spiralling approach were higher than those obtained with the mass balance approach (Fig. 4b, Wilcoxon paired test, Signed-Rank = −427, $df = 44$, $p < 0.01$). The uncertainty associated with groundwater inputs for NH_4 was low compared to that associated with NO_3. In relative terms, the U_{MB} for NH_4 decreased on average 2.2 ± 2.4 times and increased 1.1 ± 1.1 times when groundwater inputs were half and 2-fold those in surface water, respectively. For the spiralling approach, U_{SP} varied on average 5.9 ± 13.1 times due to the uncertainty associated with this calculation. The mass balance approach suggested non-equilibrium of in-stream processes (i.e. $U \neq 0$) for 75% of the studied cases. Contrastingly, according to the spiralling approach there was only a net change in NH_4 in 26% of the cases as a consequence of non-significant longitudinal patterns of NH_4 concentration in most of the cases.

Overall, results from this study showed discrepancies in the estimates of in-stream net uptake rates of DIN forms between the mass balance and the spiralling approach, in particular under high ($> 300\,\mu g$ N l^{-1}) ambient N concentrations. For lower ambient N concentrations, however, net uptake rates obtained with the two alternative approaches were similar. Roberts and Mulholland (2007) reported a small effect of groundwater inputs on N in-stream net uptake rates in the West Fork of Walker Branch (Tennessee, USA), where stream ambient DIN concentrations were low ($< 100\,\mu g$ N l^{-1}). We showed, however, that groundwater inputs could strongly influence estimation of net uptake rates when stream ambient concentrations are high, such in the case of NO_3 in our data set, or under large N_{top}:N_{bot} ratios. When that is the case, riparian groundwater samples may be collected to constrain the range of uncertainty in U associated with this nutrient source (Roberts and Mulholland, 2007). However, reliable riparian groundwater concentration measurements are difficult to obtain due to the high spatial variability (Lewis et al., 2006) and the potential difference between groundwater and the water found at the interface between the ground and surface water which really enters the stream (Brookshire et al., 2009). This limitation can be a disadvantage when

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using the mass balance approach under certain conditions, such as high stream nutrient concentrations, compared to the alternative spiralling approach for which additional information on groundwater sources is not required.

The spiralling approach is based on a whole-reach integrative measure of a longitudinal concentration trend across several sampling points, contrasting with the net change in nutrient loads between two sampling points considered by the mass balance approach. In addition, estimates of U by the spiralling approach are based on a first-order reaction model, which is likely to be more representative for in-stream nutrient dynamics than the linear model used by the mass balance approach (Newbold et al., 1981; Webster and Valett, 2006). We showed that when longitudinal patterns are uncertain, such as in the case of NH_4, the spiralling approach may be more reliable in estimating in-stream net uptake rates than the alternative mass balance approach which may tend to overestimate those cases in which there is net uptake or release.

Although the spiralling approach has been less commonly used in the literature, data sets of ambient nutrient concentration collected along stream longitudinal transects are often available from nutrient uptake studies using the nutrient addition methodology for which background nutrient concentrations are measured along the study reach (Webster and Valett, 2006; Ensign and Doyle, 2006). We encourage other researchers to profit from such sort of data to obtain reliable in-stream net uptake rates and to compare the two approaches across further systems. Furthermore, if data on both gross (e.g., from nutrient additions) and net nutrient uptake are available, rates of mineralization could potentially be inferred. This information is relevant to expand our knowledge on broad temporal and spatial patterns of net in-stream nutrient uptake rates, which could be based on different sources of existing data.

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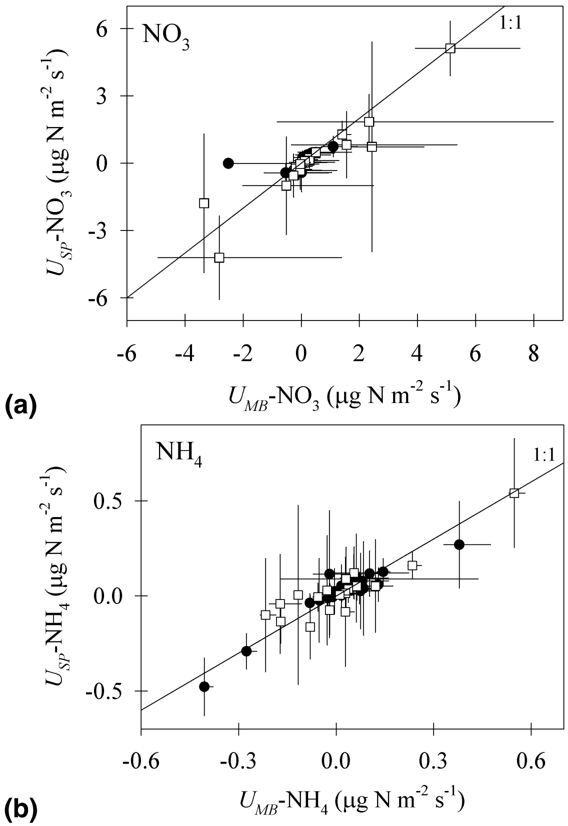

Fig. 1. Relationship between the net uptake rates estimated with the mass balance approach (U_{MB}) and the spiralling approach (U_{SP}) for **(a)** NO$_3$ and **(b)** NH$_4$. White squares and black circles correspond to values from Fuirosos ($n = 20$) and Santa Fe ($n = 25$) streams, respectively. Error bars are the confidence intervals estimated with the two approaches (see text for details). The 1:1 line is shown in each case.

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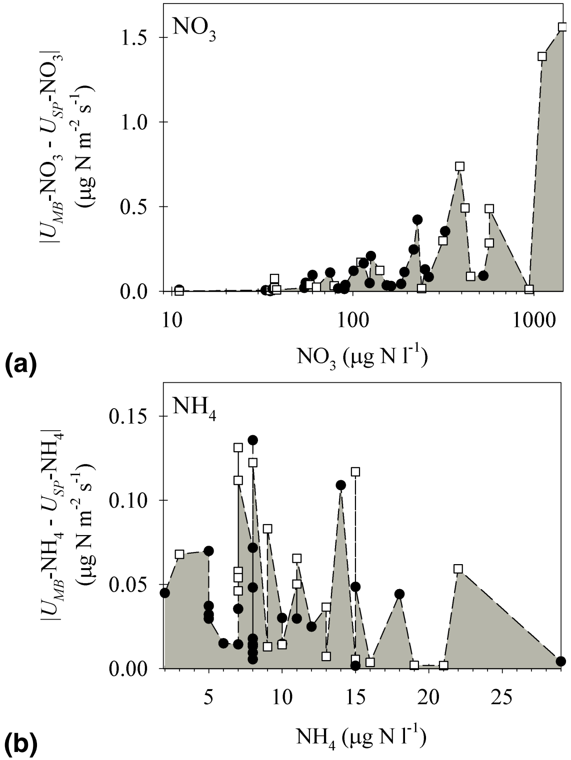

Fig. 2. Absolute differences between the net uptake rates estimated with the mass balance approach (U_{MB}) and the spiralling approach (U_{SP}) sorted as a function of average stream N concentration for **(a)** NO$_3$ and **(b)** NH$_4$. White squares and black circles correspond to values from Fuirosos ($n = 20$) and Santa Fe ($n = 25$) streams, respectively.

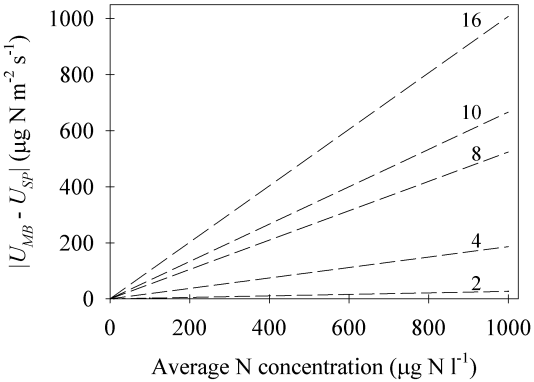

Fig. 3. Sensitivity analysis of the influence of the average N concentration and the proportion of N concentration between the top and bottom of the reach on the absolute difference between the net uptake rates estimated with the mass balance approach (U_{MB}) and the spiralling approach (U_{SP}). Constant discharge along the reach and changes between only two stations (top and bottom) were assumed. Numbers on each dashed line correspond to values of the proportion of N concentration between the top and bottom stations.

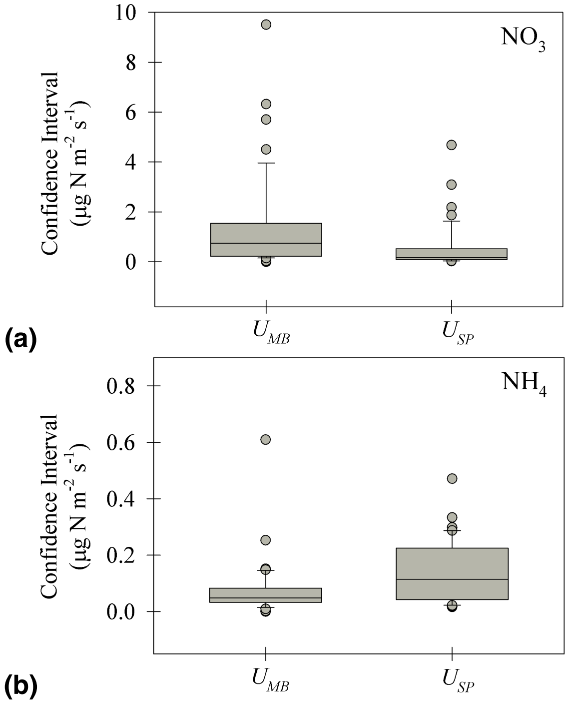

Fig. 4. Box plots summarizing the confidence intervals (CI) calculated for the net uptake rates using the mass-balance approach (U_{MB}) and the spiraling approach (U_{SP}) for **(a)** NO_3 and **(b)** NH_4. Data are from Santa Fe ($n = 25$) and Fuirosos ($n = 20$) streams. The centre horizontal line in each plot is the median value. Fifty percent of the data points lie within each box. The whiskers above and below the box indicate the 90% and 10% percentiles. Circles are outliers. See text for details on the calculation of the CI for each approach.