

Dear Referee #1

Thank you very much for your valuable comments on our manuscript. We would like to respond to each of your comments one by one.

Their simulation is based on a case where nitrate fluxes decrease with soil depth, which is the case if net nitrification is negative (nitrate consumption larger than gross nitrification). It would be very interesting to see what happens in the case of a positive net nitrification. This could be done with just one more simulation.

Thank you for your comment. Our simulation was done for the forested catchment reported by Hattori et al. (2019). While the deposition flux of NO_3^- was $7.0 \text{ kg of N ha}^{-1} \text{ y}^{-1}$, the leaching flux of NO_3^- was $2.6 \text{ kg of N ha}^{-1} \text{ y}^{-1}$ in the forested catchment, so that the influx of NO_3^- was higher than that of outflux. Thus, the NO_3^- fluxes always decreased with soil depth in our original simulations, shown by Figures 1c and 2c in the manuscript.

In response to your request, we made a new simulated calculation, in which NO_3^- fluxes increased with soil depth in the soil layers from 1 to 5 with an increasing rate of $0.44 \text{ kg of N ha}^{-1} \text{ y}^{-1}$ for each layer, while NO_3^- fluxes decreased with soil depth in the soil layers from 6 to 10 with a decreasing rate of $1.32 \text{ kg of N ha}^{-1} \text{ y}^{-1}$ for each layer (Table R1). While the newly estimated GNR ($19.1 \text{ kg of N ha}^{-1} \text{ y}^{-1}$) was comparable with that estimated for the forested catchment with the profile shown by Figure 1 ($13.0 \text{ kg of N ha}^{-1} \text{ y}^{-1}$), it was still significantly smaller than the GNR calculated by using Eq.6 ($83.6 \text{ kg of N ha}^{-1} \text{ y}^{-1}$). Such additional simulated calculation by changing the nitrate fluxes with soil depths further supports our conclusion that the GNR estimated from the $\Delta^{17}\text{O}$ value of stream nitrate in forested catchments was, to some extent, an overestimate of the actual GNR.

In addition, the present results imply that the most important parameter to determine total GNR (and thus total GDR + uptake) is the $\Delta^{17}\text{O}$ value of NO_3^- consumed in soil layers. That is, the depth profile of NO_3^- fluxes has little impact on GNR.

Table R1. $\Delta^{17}\text{O}$ values of NO_3^- , leaching flux of NO_3^- , total consumption rate of NO_3^- (GDR + uptake), and GNR in the simulated forested soil where the distribution of $\Delta^{17}\text{O}$ values of NO_3^- is heterogeneous. While the net nitrification from soil layer 1 to 5 showed positive values, the soil layer 6 to 10 showed negative values.

Depth layer	$\Delta^{17}\text{O}$ ‰	NO_3^- flux	GDR +uptake	GNR
		kg of N ha ⁻¹ y ⁻¹		
0	28.0	7.0	0.0	0.0
1	25.4	7.4	0.3	0.7
2	22.8	7.9	0.4	0.8
3	20.2	8.3	0.6	1.0
4	17.7	8.8	0.8	1.2
5	15.1	9.2	1.1	1.5
6	12.5	7.9	3.2	1.9
7	9.9	6.6	3.4	2.1
8	7.3	5.2	3.6	2.3
9	4.7	3.9	4.2	2.9
10	2.2	2.6	6.0	4.7
11	2.2	2.6	0.0	0.0
Total			23.5	19.1

In many soils, preferential water flow can be observed. In such cases, there is not a single, homogeneous nitrate pool per soil layer but nitrate that is more or less mobile along the flow paths and nitrate that is more bound within the soil matrix. The first is more prone to leaching and perhaps uptake, the second to denitrification. Simulating this would be a difficult task, probably out of the scope of the present article. Nevertheless, it would be useful if the authors would discuss this point

Thank you for your comment. As you point out, the leaching flux of soil NO_3^- in each layer is complex. Thus, we have added the following sentences in the manuscript (P10, L171-179).

Note that the linear variation in the leaching flux and $\Delta^{17}\text{O}$ values of soil NO_3^- used in the simulated calculations is just one of many possible variations in the forested catchments. It is impossible to decide whether the linear variation was realistic until the downward water flux, along with the concentration and $\Delta^{17}\text{O}$ values of NO_3^- , is determined for each soil layer. However, the simultaneous observations of the oxygen isotopes of soil NO_3^- and stream NO_3^- (Hattori et al., 2019; Osaka et al., 2010; Nakagawa et al., 2018; Rose, 2014) imply that the approximation of the $\Delta^{17}\text{O}$ values of the soil NO_3^- consumed through assimilation or denitrification to be always equal to the $\Delta^{17}\text{O}$ value of stream NO_3^- , shown in Fig. 2b, is unrealistic.

and especially if they could make recommendations on how to sample nitrate from the soil for $\Delta^{17}\text{O}$ determination: zero-tension lysimetry, tension lysimetry, centrifugation, extraction? I'm not sure if clear answers can be given with the present knowledge of soil nitrate transformations, but at least the question would deserve to be raised.

Thank you for your comment. We have recommended the sampling method of soil nitrate in the manuscript (P11, L191-194).

If we estimated the downward water flux at each soil layer, together with the NO_3^- concentration and $\Delta^{17}\text{O}$ value of NO_3^- in each soil layer using a tension-free lysimeter (Inoue et al., 2021), we could estimate the vertical change in the leaching flux of NO_3^- for each soil layer along with the $\Delta^{17}\text{O}$ value of soil NO_3^- in each layer.

L. 3: the word "eluted" is rather used for the what is done on purpose in the lab. In this case, for the process observed in the nature, a better choice would probably be "leached".

Thank you for your suggestion. We would like to change the "eluted" to "leached" in the revised manuscript.

Line 6: instead "nitrate metabolized", it would be better to write "nitrate that is consumed", first because as soon as it is consumed, it is no longer nitrate, and second because "metabolized" is rather used to indicate that it is incorporated into organic matter, which is not the case for the denitrification.

Thank you for your suggestion. We would like to change the “nitrate metabolized” to “nitrate that is consumed” in the revised manuscript.

L. 24: on the same idea: "consumption" instead of "metabolic".

Thank you for your suggestion. We would like to change the “metabolic” to “consumption” in the revised manuscript.

L. 27: "is negligible" is too general, better add "often".

Thank you for your suggestion. We would like to revise this in the revised manuscript.

L. 28: "by order of magnitude": do you mean "one" order?

We would like to revise the sentence to “the GNR often exceeds the net nitrification rate by several orders of magnitude.”

L. 21-29: very long sentence.

We would like to revise the sentence in the revised manuscript.

L. 31: it would be useful to explain shortly that the Δ anomaly is based on the δ of both ^{17}O and ^{18}O and that it is purposely defined so as to make it independent of mass-dependent fractionation.

We would like to add the information in the revised manuscript.

L. 31: in my opinion, "conservative" would be better than "conserved" (because it tends to be conserved but it is not always perfectly conserved).

Thank you for your suggestion. We would like to change the “conserved” to “conservative” in the revised manuscript.

L. 33: it seems strange to write "REmineralized" when it may be mineralized for the first time after centuries of N staying in the soil in the organic matter.

We would like to revise this in the revised manuscript.

L. 34-38, 53-57: long sentences.

We would like to revise the sentences in the revised manuscript.

L. 76: in this equation, some processes are denoted as subscript of NO_3^- (like deposition) while others are denoted for themselves (like GNR). GNR and GDR are usually expressed as a nitrogen rather than as a nitrate flux. As it is written, the equation lets it open. It would be better to explicitly express all rates either as N or as NO_3^- .

Thank you for your suggestion. Our simulation was done for the forested catchment reported by Hattori et al. (2019). Thus, the symbols used in the manuscript were in accordance with Hattori et al. (2019) as well. We would like to mention this in the revised manuscript.

L. 74-80, 85-90: it is not clear why the word "each" is always used for the catchments (not only here, in general in the text).

We would like to revise these in the revised manuscript.

L. 112-113: repeated usage of the word "limited".

We would like to revise this in the revised manuscript.

L. 116: which one of the $\Delta^{17}\text{O}$ is this? Or is it the difference?

The $\Delta^{17}\text{O}$ denotes the $\Delta^{17}\text{O}$ of stream nitrate. We would like to revise this in the revised manuscript.

L. 127-128: fine roots would be much more relevant than the total root biomass (with coarse roots obviously overrepresented close to the stem and thus close to the surface).

Thank you for your suggestion. We would like to add the information in the revised manuscript.

L. 146-148: it may be useful to explain this as a gradual uptake (consumption) of nitrate as water moves down the profile.

Thank you for your suggestion. We would like to add the information in the revised manuscript.

L. 152-160: these assumptions are obviously simplifications compared to real measurements, but they make sense for the demonstration. It would be interesting to test also the assumption of nitrate fluxes increasing with depth because of a positive net nitrification.

Thank you for your suggestion. We have simulated the positive net nitrification in the soil layers above.

L. 175-179: long sentence.

We would like to revise the sentence in the revised manuscript.

L. 220: as written, it is like anonymous reviewers would be named, which does not make sense.

We would like to revise the sentence in the revised manuscript.

Fig. 1, fig. 2: the soil does not float above water and therefore "soil layers" and "water layer" should rather be marked "unsaturated soil layers" and either "water-saturated soil layer" or "seepage water" (as these two are considered to exhibit the same flux).

We would like to revise the "soil layers" and "water layer" to "unsaturated soil layers" and "water-saturated soil layer" in the revised manuscript.

We would like to thank you for the helpful comments. We hope that our responses to your comments are satisfactory.

Sincerely,
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