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Dr. Sébastien Fontaine
Editor of Biogeosciences

Title: Export flux of unprocessed atmospheric nitrate from temperate forested catchments: A possible new index for nitrogen saturation
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MS No.: bg-2018-251

Dear Dr. Fontaine:

Thank you very much for your e-mail on 1 Nov., 2018. We are very glad to hear that our manuscript was accepted with corrections. We have carefully studied the additional comments and corrected the manuscript. All the corrections have been listed in this letter. Besides, we have uploaded the revised manuscript.

Revisions from the previous submission to BG are as follows:

(P5/L1 in the previous/corrected MS) We added "‰", as suggested by referee #2.

(P7/L10 in the previous/corrected MS) We cited Aber et al. (1989) here, as suggested by referee #1.

(P8/L11 in the previous/corrected MS) We cited Aber et al. (1989) here, as suggested by referee #1.

(P8/L19 in the previous/corrected MS) We replaced "thrice" by "twice", as suggested by referee #2.

(P9/L4 in the previous/corrected MS) We added several words here to explain the details of the soil water sampling, as suggested by referee #1.

(P14/L25 and the reference list in the previous/corrected MS) We added a new reference here (Bourgeois et al., EST, 2018a), as suggested by referee #1. Due to the revision, we also replaced Bourgeois et al. (2018) in the previous MS (Bourgeois et al., Sci. Total Environ., 2018) by Bourgeois et al. (2018b).

Bourgeois, I., Savarino, J., Caillon, N., Angot, H., Barbero, A., Delbart, F., Voisin, D., and Clément, J.-C.: Tracing the fate of atmospheric nitrate in a subalpine watershed using $\Delta^{17}O$, Environ. Sci. Technol., 52, 5561−5570, 2018a.

(P19/L4 in the previous/corrected MS) We added several words here to emphasize that the "soil nitrate" corresponds to nitrate in the soil water samples (SLS20, SLS60, and SMS20), in response to the referee #2's comments.

(P19/L11 in the previous MS, P19/L12 the corrected MS) We removed "huge", as suggested by referee #1.

(P21/L4 in the previous MS, P21/L6 the corrected MS) We replaced "oxygen isotopic fractionation" by "^{18}O-enrichment", as suggested by referee #1.

(P24/L15 and P27/L12 in the previous MS, P24/L17 and P27/L15 in the corrected MS) We revised sections 3.5 and 4 to emphasize that, not only the M_{atm}/D_{atm} ratio, but also the stream nitrate concentration is the index of the nitrogen saturation stages, in response to the referee #1's comments.

(Table 1 in the previous/corrected MS) We added a row for the M_{atm}/D_{atm} ratios, in response to the referee #1's comments.

The answers to the referees' questions/comments are as follows:

#1> Section 2.3: Please expand a bit on your methods here. Explain for instance the
#1> conditioning of the samples (fridge, freezer, filtration) and if you use acid solution to stop
#1> any biological activity, etc.

We have added several words in section 2.3 to explain the details on the soil water sampling. The conditioning of the soil water samples, however, had been explained in section 2.5 (Analysis) in the previous/corrected MS, so that we did not add explanation on the conditioning here.

#1> Main comment:
#1> My main interrogation now is regarding your explanation of the difference in
#1> the Matm/Datm ratios between the studied catchments: you say that the difference is due
#1> to different biological assimilation rates, caused by different nitrogen saturation status.
#1> But couldn't the different biological assimilation rate be caused only by the difference
#1> in the vegetation in the forest for each catchment? The tree species abundances are different
#1> in KJ, IJ1 and IJ2 forests: could that cause the difference in the Matm/Datm ratios because
#1> the trees would have different metabolic rates? And in that case, the Matm/Datm ratio
#1> would not be an indicator of N saturation, but just of the retention capacity of a catchment?
#1> I think this needs to be at least discussed in your last section.

Please note that, not only the M_{atm}/D_{atm} ratios, but also the stream nitrate concentration is the index of the nitrogen saturation stages. If the differences in vegetation were responsible for the high M_{atm}/D_{atm} ratios in the studied sites (KJ site, especially) as you insisted, the differences in vegetation should be responsible for the elevated stream nitrate concentrations in the studied sites (KJ site, especially) as well. While there were many past studies on the elevated stream nitrate concentrations in the studied sites, as well as the other forested watersheds in the world as already presented in the MS, none of them had found significant correlation between stream nitrate concentrations and the differences in vegetation. That is to say, while the differences in vegetation could explain the variations in M_{atm}/D_{atm} ratios, it is impossible to explain the normal correlation between stream nitrate concentrations and M_{atm}/D_{atm} ratios in Fig. 8(a).

We revised sections 3.5 and 4 to emphasize that, not only the M_{atm}/D_{atm} ratio, but also the stream nitrate concentration is the index of the nitrogen saturation, in response to your comment.

#2> 1- Please give the units for each Equation.

We already answered to this comment during the discussion stage. Because the equations in this manuscript were general equations and no actual value was presented in the equations, there are no constraints on the units. That is to say, we can use any unit we want.

If we used "µmol L^{-1}" for C_{atm} and "L day^{-1}" for V and "m^2" for S, for instance, the unit of F_{atm} became "µmol m^{-2} day^{-1}" using equation (3). If we used "mg L^{-1}" for C_{atm} and "L yr^{-1}" for V and "km^2" for S, for instance, the unit of F_{atm} became "mg km^{-2} yr^{-1}".

#2> 2- In the discussion please specify to which category (catchment groundwater, through #2> flow) belongs the water sampled in SLS and SMS.

We already answered to this question during the discussion stage. We interpreted that the soil water sampled in SLS and SMS belongs to through flow.

In response to your question during the discussion stage, we already emphasized that soil nitrate represents nitrate in through flow (P19/L8) during the previous revisions. Additionally, we newly added several words in section 3.3 to emphasize that the soil nitrate corresponds to nitrate in the soil water samples (SLS20, SLS60, and SMS20).

#2> 3- A figure showing the relationship between Matm and stream NO3 conc. would be #2> welcome to support your statement P23-L20-21.

In response to the same request from you during the discussion stage, we had added Fig. 8(c) during the previous revisions.

We would like to thank you and referees for the helpful comments and suggestions. We trust that the revision is satisfactory response to the referees' comments. Thank you for your consideration.

Sincerely yours,
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Encl.
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