

## ***Interactive comment on “Export flux of unprocessed atmospheric nitrate from temperate forested catchments: A possible new index for nitrogen saturation” by Fumiko Nakagawa et al.***

### **Anonymous Referee #2**

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This manuscript is worth publication in Biogeosciences even though some major revisions are necessary. The paper presents temporal variations NO<sub>3</sub> concentrations and isotopic signatures (15N, 18O, 17O) in streams and soils of three forested watersheds in Japan. Stream discharge rates and total atmospheric NO<sub>3</sub> deposition rates were also measured and used to calculate the daily and annual watersheds export fluxes of nitrate (total, atmospheric, and “mineralized”). Using concentrations and  $\delta^{17}\text{O}$ , the authors show that there is a 4-months lag-time for NO<sub>3</sub> originating from soil nitrification to reach the stream through groundwater transportation. They found that the proportion of atmospheric NO<sub>3</sub> deposited (Matm/Datm) that leaves the watershed was positively related to the amount of total nitrate exported by the streams, which I think is

C1

rather intuitive. This said, they demonstrate that nitrate loss in N-saturated catchments was due to a reduction of its assimilation by plant and microbes. Finally, they suggest that the ratio Matm/Datm (or the Catm in stream water) could be used as an indicator of watershed nitrogen saturation particularly useful in humid climate. Although I think the paper brings new and interesting data, I identify three main points that challenge how much we can trust the conclusions. First, the watersheds (KJ, IJ1 and IJ2) have extremely different size (3.84, 298, 108 ha respectively) and there are no information concerning their geology and soil depth/quality which are important parameters to interpret stream discharge and nutrient flow. Second, stream discharge is estimated with a different method for each stream (section 2.2). Third, atmospheric nitrate deposition rates were also obtained with different approaches in the three watersheds (section 2.4). Finally one can wonder how much data variation is carried by each of these points and how this influences the results and their interpretation. My guess is that this should be at least acknowledged, analysed and discussed thoroughly to convince the reader.

Abstract: L21: I suggest you give the annual fluxes of nitrate instead of concentrations.

L23-25: It is misleading to compare, in the same sentence, 17O-excess in KJ soil water with those of KJ, IJ1 and IJ2 stream water. Add “in KJ” after “+0.1‰ to +5.7‰”.

L25: “was groundwater nitrate”. I would also remove the end of the sentence “which buffered the seasonal. . .” as it is confusing.

Introduction:

P2-L5 (and throughout the paper): Do not use the word “enrichment” when you simply mean “increase”, especially in a paper dealing with isotopes for which “enrichment” is usually used as in P4-L7.

P2-L25: There are other processes that should be mentioned : DNRA for instance.

P3-L2-3: Remove the end of the sentence “, including [ . . . ] catchments.”, it does not

C2

bring new information.

P3-L4: composition. . .has been (no plural)

P3-L8: "has survived"

P3-L13: remove "nitrate including"

P3-L21-22: this sentence is useless.

I miss some clear hypotheses at the end of the introduction.

Section 1.2 is Materials and Method.

P4-L6: atmospheric nitrate is not only produced by photochemical reactions.

P4-L13: specify this is Equation 1

P4-L24: Specify this is Equation 2

Here you use  $C_{atm}$  and  $C_{total}$  for the first time. In the rest of the manuscript they represent either mean daily or mean annual concentrations which is confusing. Please use different codes for daily and annual.

Experimental section

KJ, IJ1 and IJ2 have very different size. Here (and probably in the discussion) you should somehow reassure the reader that it is not a source of bias in your interpretation.

P5-L15: Remove "continuous" or replace by "weekly" as samples were collected once a week.

P5-L25: "2500 mm" seems a lot as the average precipitation in Shibata is 1263 mm.

P6-L9: "3300 mm" seems a lot. Actually if I do the maths with your figures I get 1500 mm for 10 months/yr.

P6-L23: I am not sure *Clethra* sp. and *Ilex* sp. can be considered as trees.

C3

P7-L1: You do not use water  $T^{\circ}$ , pH, alkalinity and EC in the paper. Please remove. In fact, maybe it could bring new insights in your data set and interpretation.

Section 2.2: Discharge is estimated with different methods for each stream. This is a source of variation for your results and you should acknowledge that somewhere in the discussion to convince the reader you took this issue into account in your interpretation. There is the same risk of discrepancy with the way atmospheric nitrate deposition rates were obtained (section 2.4).

Section 2.3: what SLS and SMS stand for? You need to explain why you decided to sample next to the stream and 20m upland, and why at 20cm and 60cm depth.

P7-L22: "between December and March" Section 2.5: I am surprise samples were not acidified prior storage.

P9-L15: remove ", the procedure [. . .]. Approximately"

P10-L22: You mention  $NO_2$  as a possible source of variation, what about  $NH_4$ ? Section 2.7: I suggest you give the units for each Equation.

P12-L1: replace "obtain" by "estimate"

Results

P12-L16-21: You can remove this paragraph which does not bring new information compared to the figures legends.

P12-L25: Could you add ranges or values to  $F_{total}$  and  $F_{atm}$ . Same thing for IJ1 and IJ2.

P14-L20-22: You can remove this sentence which does not bring new information compared to the figures legends.

Discussion

Specify to which category (catchment groundwater, through flow) belongs the water

C4

sampled in SLS and SMS.

P16-L25: Add to Fig 4 legend that the hypothetical mixing line is reported.

P17-L1: replace “buffered by” by “diluted in”

P18-L22: “This delay time reflects the magnitude and flow of the nitrate. . .”

P19-L23-24: A figure showing the relationship between Matm and stream NO3 conc. would be welcome.

P20-L3: replace “N” by “NO3”

P20-L4: replace “nitrogen” by “nitrate”.

P20-L8: Fig. 9 does not show that “Matm/Datm ratios are stable during the progress of nitrification in forested soils”

P20-L17-22: this paragraph is not clear enough. In particular the last sentence compare “stream nitrate enrichment due to nitrogen saturation” with “stream nitrate enrichment due to artificial processes”, which I do not quite understand. Artificial processes (e.g. fertilizers, leguminous fields. . .) are responsible for N saturation. Please clarify.

P21-L24: “. . .by 6 and 20 times respectively in accordance. . .”

Fig.2: Underneath RW1 and RW3 there are distances (120m and 40m respectively), are they elevation a.s.l.?

Fig1 and Fig.2: Please use the same kind of map for both figures.

Fig.4: Please specify what the line stands for?

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