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Discussions

Interactive comment on "Quantifying the effects of clear-cutting and strip-cutting on nitrate dynamics in a forested watershed using triple oxygen isotopes as tracers" *by* U. Tsunogai et al.

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Thank you very much for your valuable and positive comments on our manuscript. We would like to reply to you by citing each of your comment/question.

> Comment 1: Annual atmospheric nitrate flux in the research period, i.e., 2003-2005, is estimated as the mean of 2008-2011. I doubt this is suitable, because that the inter-annual variability of atmospheric nitrate flux is large, varying from 15.1 to 20.9 (Table 2), not to mention if there is a long term trend in atmospheric nitrate abundance. So how well does the average of 2008-2011 represent the flux in each year of 2003, 2004 and 2005? I understand that the authors probably don't have data covering the

years of 2003-2005, but the authors at least can estimate the nitrate flux in each year of 2003-2005 based on the annual precipitation record in the research region, as the atmospheric nitrate flux is dominated by wet deposition.

Total deposition rate (wet + dry) of atmospheric inorganic N had been preliminarily monitored during 2003-2005 as well. The results have been presented in Fukuzawa et al. (2006) and thus cited them in P7435/L7 in our original manuscript. As presented in P7435/L6-8, the annual deposition rate almost corresponds with our total deposition rate of inorganic N, while the data coverage was scarce. As a result, we used the atmospheric nitrate flux determined during 2008-2011 based on the standard EANET protocol to our research period (2003-2005).

> Comment 2: when calculating the fraction of atmospheric nitrate in total nitrate leaching from the watershed, the authors treated D17O of atmospheric nitrate its annual mean of 26.2 ‰ This again is not suitable, as D17O of atmospheric nitrate varies seasonally, from ∼20 ‰ in summer to as high as 34.5 ‰ in winter in the research region. The annual mean is suitable only if in Equation (2) Catm, Ctotal, and D17O are all annual means, which is obviously not what the authors did. Seasonally or monthly average of atmospheric nitrate D17O should be used in Equation (2).

The mean residence time of atmospheric nitrate deposited onto the watershed should be much more longer. If the mean residence time of atmospheric nitrate was less than a month, the export flux of atmospheric nitrate should increase in response to significant loading event of atmospheric nitrate onto the watershed. While we found significant loading event of atmospheric nitrate via heavy rain in May and June 2003 and September 2004 in the watershed, we could not find any significant increase in the export flux of atmospheric nitrate in response to the events. Besides, the residence time more than a few years have been assumed for nitrate in shallow groundwater of a temperate forest catchment in Japan in past studies (e.g. Ohte et al., 2003). The annual mean Delta17O must be more preferable for calculating Fatm. Furthermore, even if we used monthly mean for calculating each Fatm, the final export flux of atmospheric

nitrate (Matm) was much the same.

> Comment 3: the spring nitrate concentration peaks in 2004 and 2005 (Figure 2). The authors suggested that they are due to spring snow melting, but why the flow rate is delayed compared to the nitrate peak? Presumably, snow melting will enhance nitrate concentration (atmospheric nitrate scavenged by snow) and water flow simultaneously.

Direct discharge of snowmelt to stream was responsible for the nitrate peak. Significant portion of snowmelt, however, seems to seep into ground, and mix with groundwater of the watershed. As a result, groundwater level in the riparian zone increased and thus the discharging rate of groundwater increased. Because nitrate concentration of groundwater was smaller than that of snowmelt, we can find the flow rate to be delayed compared to the nitrate peak.

> Comment 4: there should at least to be a short paragraph as the conclusion to wrap up the major findings/points of this work.

We would like to add "conclusion" in the revised manuscript as suggested.

> P7415, line 9-10: "atmospheric nitrate accounts for more than 50% of total nitrate exported...", this statement is misleading. As shown in Figure 3, every year only in a very small period (one sample point) has D17O of ∼14 ‰ when atmospheric nitrate is important.

We would like to add "at the maximum in spring" at the end of the sentence "Additionally, the high Delta17O values of +14.3‰ suggest that the direct drainage of atmospheric nitrate accounted for more than 50% of total nitrate exported from the forested watershed" in the revised manuscript.

> P7416, line 10: what is the difference between uptake by microbes and denitrification? Aren't they usually the same?

We classified assimilative nitrate reduction to "uptake by microbes (No. 3)" and dissimilative nitrate reduction to "reduction through denitrification (No. 4)". Because the

final products are essentially different (No.3 = fixed-N (ammonium or organic-N), No.4 = N2), we classified them into different category, with emphasis on the biogeochemical point of view.

> P7417, line 19: What does "natural" mean?

We added "natural" to differentiate from the tracer methods using 15N etc.

> I think it is "Isotopic composition", not "compositions".

We would like to revise as suggested.

> P7418, line 12: it is not "international standard", it's "standard reference material", i.e., VSMOW for oxygen and N2-Air for nitrogen.

We would like to revise as suggested.

> Section 2.3: I was curious about the analytical method. Why not measure d15N and D17O together after converting N2O to O2 and N2, instead of measuring d15N in N2O first?

The reason is the same with that we determined d18O from N2O monitoring, not from O2 + N2 monitoring as already presented in Komatsu et al. RCM Vol.22 p.1587-1596 (2008). Because the process converting N2O to O2 + N2 accompanied additional small mass-dependent fractionation, the precision based on O2 + N2 monitoring is worse than that of N2O monitoring.

> In addition, when measured d15N in N2O, were the final d15N result corrected for the interference of 17O?

To calculate d15N in N2O, we corrected for the interference of 17O, based on the equation (7) of Komatsu et al. (2008).

> Section 2.4: How the flux of nitrous acid and nitrogen oxides were used to determine the flux of nitrate? I think nitrate flux is estimated from the flux of particulate nitrate and

gaseous HNO3.

When we use the filter pack method to determine gaseous HNO3 concentration, a slight portion (ca. 0.5%) of nitrous acid (HONO) is gathered onto nylon filter to which most of gaseous HNO3 is gathered. As a result, we should determine nitrous acid simultaneously with gaseous HNO3 to correct for the contribution to nylon filter. Besides, a slight portion of nitrogen oxides (NO2) is gathered onto alkaline filter to which most of gaseous nitrous acid is gathered. As a result, we should determine nitrogen oxides simultaneously with nitrous acid as well to correct for the contribution. The detail is presented in the reference (Noguchi et al., 2007b).

> P7427, line 1-5: I don't agree with this argument, Because nitrate produced by nitrification in the soil will also have d15N values around -1 ‰ as shown in Figure 3 and Figure 4a. Just that d15N in the spring happens to be consistent with that of atmospheric nitrate illustrates nothing, as nitrate in the spring stream is still a mixture (about 1:1) of atmospheric nitrate and nitrate from nitrification. So ideally, d15N of nitrate in the spring stream should be different from that of atmospheric nitrate.

What we discussed here is that the d18O and d15N values of those with high Delta17O values were reasonable as those half of which is occupied by atmospheric nitrate. As you suggested, the d15N data implied that the average d15N values of remineralized nitrate must be around -1‰ (roughly from -4‰ to +3‰ as well. Still, nothing is contradicted with our conclusion that d15N values of high Delta17O values were reasonable as those half of which is occupied by atmospheric nitrate.

> P7429, line 12-20: there is no need to discuss samples with D17O around or lower than 0.2 ‰ as the authors already stated that the analytical uncertainty is 0.2 ‰ it is straightforward to treat these samples as zero.

This paragraph is for those not familiar with Delta17O.

> P7430, line 7-10, why high D17O will cause large errors in d18O?

We replied the same one to reviewer #1.

The value of d18Oatm (+87.1 per mil) used in the calculation shown in equation 4 was the average value of atmospheric nitrate. Differ from Delta17Oatm, d18Oatm values could have been altered within the forest ecosystem subsequent to deposition so that we should consider errors in the d18Oatm values, i.e. differences in the d18O values between the average of atmospheric NO3- and atmospheric NO3- actually contained in each sample.

The relationship between sample NO3- (sample A and B), NO3-atm, and calculated NO3-re on a Delta17O v.s. d18O plot were presented in the supplementary figure of this reply, together with +-15 permil errors in the d18Oatm value. While the errors in the calculated d18Ore values were small for those having the low Delta17O values (shown as the case of sample B in the figure), the errors in the calculated d18Ore values were large for those having the high Delta17O values more than +10 per mil (shown as sample A in the figure). As a result, we presented that high Delta17O will cause large errors in the estimated d18Ore.

We would like to add this explanation in the revised manuscript.

> P7430, line 10-26: I don't think the discussion in this part make sense at all. As stated by the author that d15Nre represents remineralized nitrate produced in the soil through nitrification. presumably, the process of nitrification should be more important in determining d15Nre. However, in this part the authors talked only the effect of dinitrification on d15N of residual nitrate. The residual nitrate could be also atmospheric nitrate, and which is not directly related to nitrate produced from nitrification. In addition, d15N of soil nitrate is determined by many factors, including d15N of atmospheric nitrate, nitrogen isotopes of ammonium and organic N in soil (i.e., the source of nitrogen) and the process of nitrification, and the last, the process of dinitrification. I agree it is complicated to discuss d15N, but if the authors decided to discuss d15N, it should be discussed adequately. Overall, I think the authors overstated the importance of den-

itrification in influencing d15N of soil nitrate, as the effects of other factors were simply ignored in this part.

> P7431, line 6-12: This paragraph is an example that nitrification is also important in determining d15N of soil nitrate.

Our conclusion is based on the linear correlation between d15Nre and d18Ore shown in figure 5. Because nitrification is the only process producing remineralized nitrate in soil, it must be important as well to determine d15Nre values, especially those showing low d15Nre and d18Ore values.

However, it is difficult to explain the significant correlation ($r^2 = 0.31$) from the isotopic fractionation during nitrification. Because the variation range of d15Nre was so large (figure 5), from -10‰ to +5‰ we concluded that the MAJOR process producing d15Nre VARIATION is denitrification, while the isotopic fractionation during nitrification could be responsible for the dispersions of data points around the correlation line in the figure 5.

> In addition, 1) if T-test was used to compare the summer values against other seasons, at least t-value should be given;

We would like to add t-values in the revised manuscript.

> 2) why active nitrification reduce denitrification? Aren't they both sensitive to temperature and enhanced at higher Ts? > 3) nitrification process also preferentially use lighter N (e.g., [Rafter et al., 2012]), which means that nitrification produced nitrate should have relatively low d15N, could this explain the summer low d15Nre?

Denitrification could increase as well. What decrease is RELATIVE progress of denitrification within whole remineralized nitrate pool in soil. We would like to make additional explanation in the revised manuscript, adding the reference.

> P7432, line 14-16: please to be consistent, the spring increase of nitrate in the stream is due to snowmelt or due to the strip-cutting, or both?

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Only due to snowmelt before the strip-cutting (2003) but due to both after the strip-cutting (2004 and 2005). These interpretations were presented in the final part of this section, from P7433/L25 to P7434/L5 in the original manuscript.

> P7433, line 8-9: How does nitrification in soil affect atmospheric nitrate in soil? Only denitrification has.

Of course nitrification cannot affect elution rate of atmospheric nitrate directly. We estimated that enhancement of assimilation rate was primarily responsible for enhancing the consumption rate of atmospheric nitrate in summer, as presented in P7433/L5. Active nitrification, however, could indirectly enhance the consumption rate of atmospheric nitrate, by enhancing assimilation rate. All these changes were summarized as "rapid turnover of nitrate in soils (in summer)", as presented in P7433/L5.

We would like to thank you for the helpful comments and suggestions. We trust that the answers are satisfactory responses to your comments and questions.

Sincerely, Urumu

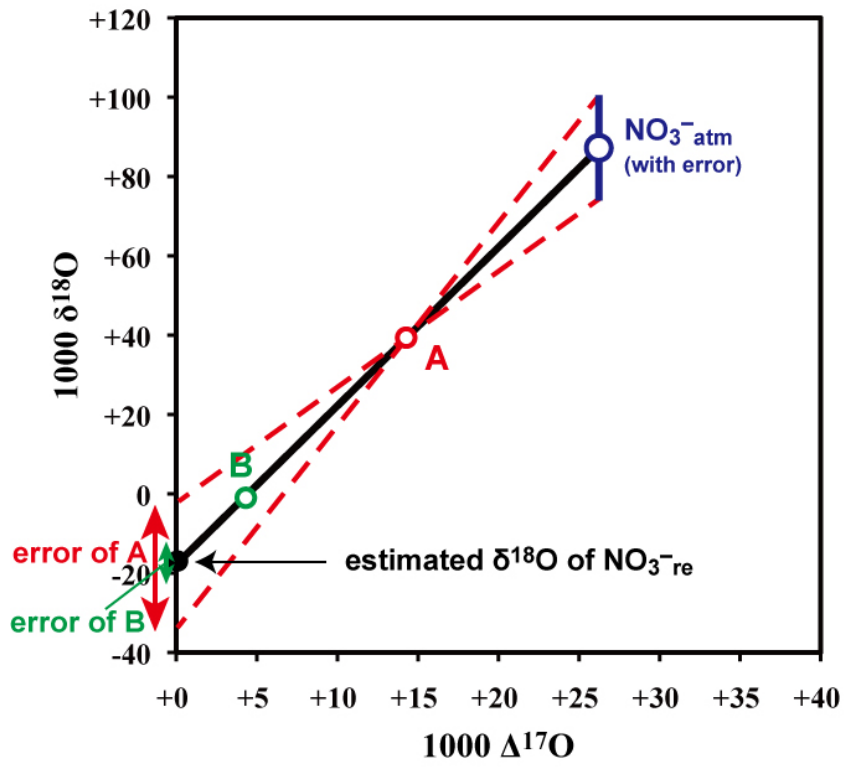Cc: D.D. Komatsu, T. Ohyama, A. Suzuki, F. Nakagawa, I. Noguchi, K. Takagi, M. Nomura, K. Fukuzawa, and H. Shibata

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Fig. 1. supplementary figure

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