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Interactive comment on "Accurate and precise quantification of atmospheric nitrate in streams draining land of various uses by using triple oxygen isotopes as tracers" *by* Urumu Tsunogai et al.

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Reply to your comment (Referee #2).

Thank you very much for your valuable comments on our manuscript. We would like to reply by responding to each of your comments and questions.

> While the topic is highly relevant and timely, and the data presented contain valuable information, the paper suffers severely from several weaknesses. Firstly, it is way too long, starting with the Abstract and ending with the Conclusions.

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We are very sorry but we do not understand what you want to say here. Compared with 20 other papers published lately in Biogeosciences, this paper could be classified as a shorter one. The style used, "starting with the Abstract and ending with the Conclusions", is the usual style for papers in Biogeosciences.

> The reader gets lost in the many detailed descriptions of results, while the description of methods is partly incomplete. For example, the method how the triple oxygen stable isotope analysis has been done is not described, even not briefly.

As presented in L9-11/P8, "The analytical procedures are the same as those detailed in previous research (Nakagawa et al. 2013; Tsunogai et al. 2014)" so we removed the details. However, we would like to add some brief descriptions on the methods in the revised MS, in response to your request.

> Also statistical and data evaluation methods are not described.

The measurement uncertainties in the isotopic composition of nitrate (delta15N, delta18O, and Delta17O) in each sample were presented in Section 3.1 (L24–25/P8). The uncertainties in the isotopic composition of the atmospheric nitrate end member (delta15Natm, delta18Oatm, and Delta17Oatm) were presented and discussed in detail in Section 3.1 (from L17/P11 to L2/P13) and 3.3 (L14–18/P14).

The uncertainties in both the absolute concentration of atmospheric nitrate (Catm) and the isotopic composition of the remineralized nitrate end member (delta15Nre, and delta18Ore) were simply calculated based on the propagation law of the errors, mostly derived from the errors in the values of delta15Natm, delta18Oatm, and Delta17Oatm in the equations (2), (6) and (7). We would like to emphasize this in the revised MS.

> But most importantly, the conclusions with respect to the effect of different land uses on the fate of atmospheric nitrate (which was the main motivation of the study) were based on many assumptions and uncertain values, especially by excluding more than two sources of nitrate (from atmosphere and from nitrification only).

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All the nitrate sources other than atmospheric, such as manure, sewage, and fertilizer, are classified to remineralized nitrate under the definition in this paper, because the oxygen atoms were derived from either terrestrial O2 or H2O through mass-dependent reactions, such as nitrification. We would like to clarify this in Section 1 to avoid misleading readers.

Based on the data of delta18Ore values around -2‰ (vs VSMOW), we concluded that either nitrification in soil or sewage effluent was the major source of the remineralized portion of the nitrate in the streams, as presented in Section 3.4. This is not an assumption because it is based on isotopic composition data.

> Furthermore, there were no statistical data provided that proved an unambiguous relationship between land use in the different stream catchments and the signature of nitrate in the stream water. This weakens the key message of the paper on the effect of land use and population density on the fate of atmospheric nitrate, and needs to be rectified before the paper becomes acceptable for publication.

All the statistical data on land use and population density in the catchments had been provided in the cited reference (Ohte et al., Water Resour. Res., 2010). We do not think the quality of the statistical data were too low to weaken the key message of this paper.

> Technical corrections can be found in the annotated pdf.

Thank you for the helpful suggestions. We would like to correct accordingly.

> Abstract, Results and Discussion, and Conclusions should be shortened significantly, focusing on the main outcome of the paper.

Compared with other papers published lately in Biogeosciences, this paper (9550 words text with 403 words abstract) could be classified as a shorter one. Besides, you and the other reviewer requested us to add much more information to this paper during revision. While we would like to try our best to shorten the revised MS as you

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requested, "significant" reduction might be unrealistic.

> It would be good if Lake Biwa were mentioned in the Abstract.

We would like to mention Lake Biwa in the revised MS.

> The number of references should be reduced to about 50 (from almost 80). The number of figures should be reduced to about 6.

Compared with the other papers published lately in Biogeosciences, 80 references and 8 figures were near average. Please tell us the basis for your estimation that 50 references and 6 figures are enough for this paper.

We would like to reduce the number of references in the revised MS as you requested, although 50 might be difficult. We would like to reduce the number of figures in the revised MS as well, although 6 might be difficult.

> Title: I suggest deleting “Accurate and precise” from the title, as it suggests a very high accuracy and precision of the data presented in the paper, which is not the case (the fraction of unprocessed atmospheric nitrate in relation to total nitrate in the stream waters of about 5% had a relative error of 10%, and average 15N and 18O values were assigned with an absolute error of +/- 10‰ which is really large).

Traditional quantification of atmospheric nitrate using the delta18O values of nitrate has been done in many past studies. Compared with the traditional method using delta18O values of nitrate only, our quantification of atmospheric nitrate using Delta17O was more accurate and more precise. Ohte et al. (2010) studied the same watershed using delta18O values of nitrate, for instance, but could not quantify the concentrations of atmospheric nitrate in the streams. Estimating the fraction of unprocessed atmospheric nitrate under a relative error of 10% is much better than the past studies. We would like to use the words "accurate and precise" to differentiate our results from those from past studies.

> Secondly, the reader might wonder why there is a differentiation between accurate

and precise, which occurs also several times in the paper, but which is never explained, also not the way how to achieve both high accuracy and precision.

These were explained in Section 1. We would like to emphasize this in the revised MS.

> Data evaluation and regression methods as well as statistics are not described.

The uncertainties in both absolute concentration of atmospheric nitrate (Catm) and the isotopic composition of the remineralized nitrate end member (delta15Nre, and delta18Ore) were simply calculated based on the propagation law of the errors, mostly derived from the errors in the values of delta15Natm, delta18Oatm, and Delta17Oatm in the equations (2), (6) and (7). We would like to emphasize this in the revised MS.

> The English should be checked by a native speaker.

The English of the manuscript was thoroughly edited by editage English editing service (http:// www.editage.jp/) prior to initial submission. We will have them edit the English again prior to submitting the revised manuscript.

> p. 2, l. 23-24: “important to primary production and thus eutrophication“: primary production does not in itself lead to eutrophication, but only a mismatch between primary production and heterotrophic consumption, usually induced by excess nutrient load. I suggest rewording to “important to primary production, and an excess of nitrate can lead to eutrophication downstream”.

We would like to make revision suggested.

> p. 2, l. 28-29: I would separate assimilation by plants and microbes and denitrification by microbes in two separate processes, as they are of completely different nature.

We would like to make the revision suggested.

> p. 3, l. 4-10: Here you cite 25 (!) references for one statement, overshooting by far. Please reduce to the 5-6 most important papers.

Most of the references cited here are cited again in later discussions. Our intention was to emphasize that using 15N/14N and 18O/16O ratios of nitrate had been conventional to determine the sources and behaviours of nitrate in stream water, especially for those who are not familiar with stable isotope traces of nitrate. In any case, we would like to reduce the number of citations, as suggested.

> p. 3, l. 10-12: As you use the bold statement ": : :can be quantified through a simple isotope mass balance approach", you should give ranges reported in the literature for the two isotope ratios for the different sources to allow the reader to assess the feasibility of the simple isotope mass balance approach.

Because the ranges were highly variable depending on the literature, it is difficult to specify a range here. Besides, we do not think such a simple approach to isotope mass balance is feasible for 15N/14N and 18O/16O ratios of nitrate as presented in the subsequent sentences. For this reason, we do not think such assessment of feasibility is essential.

> p. 3, l. 22-23: "the mixing ratios of unprocessed NO3-atm within total nitrate are minimum or uniform for whole or specific stream water samples": Meaning of this sentenceis unclear. Please reword.

We would like to reword the sentence, as suggested.

> p. 4, l. 6: "By using the Delta17O signature: : :": This term should be introduced and explained, not only by an equation, but also in words.

We would like to add the requested words.

> p. 4, l.12-13: "In addition, Delta17O is stable during the mass-dependent isotope fractionation processes within surface ecosystems.": Yes, but only if there is no oxygen exchange with the surrounding water, otherwise the Delta17O information gets lost. That is the reason why only UNPROCESSED atmospheric nitrate can be traced, not the further processing of atmospheric nitrate itself.

The oxygen exchange reaction between nitrate and water is unrealistic in the surface ecosystems (e.g. Böhlke et al., RCM, 2003; Kaneko and Kaneko & Poulson, GCA, 2013). Much higher T, as well as both lower pH and higher nitrate concentration, are needed for the progress of the oxygen exchange reaction between nitrate and water. The word 'unprocessed' is used to differentiate atmospheric nitrate from nitrate deposited as atmospheric N originally, but later assimilated by plants or microbes and then been remineralized again within surface ecosystems.

> p. 4, l. 14-15: "Therefore, although the atmospheric 15N or 18O signature can be overprinted by biogeochemical processes subsequent to deposition, Delta17O can be used as a robust tracer: : :": Again, also Delta17O can be "overprinted" by oxygen exchange, not only 15N or 18O of nitrate.

Again, the oxygen exchange reaction between nitrate and water is unrealistic (e.g. Böhlke et al., RCM, 2003; Kaneko and Kaneko & Poulson, GCA, 2013) at least in the watershed. If such oxygen exchange reaction between nitrate and water would be active, the delta18O values of remineralized nitrate should be much higher than observed.

> p. 5, l. 14: "NO3-atm is stable": I disagree. Nitrate from atmospheric deposition can and will be processed after deposition. Therefore, it cannot be considered as stable.

NO3-atm is stable DURING NITRIFICATION. What we discussed here was the variation in both NO3-atm and NO3-re during DURING NITRIFICATION. We would like to emphasize this in the revised MS to avoid misleading readers.

> p. 5, l. 18-20: "Moreover, we exclude the contribution of NO3-atm in the determined 15N and 18O values to estimate the corrected 15N and 18O values for accurate evaluation of the source and behavior of NO3-re.": Totally unclear what that means. Please explain more clearly.

Because nitrates were a mixture of NO3-atm and NO3-re, the raw delta15N and

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delta18O values of nitrate (= NO3-atm + NO3-re) were somewhat different from those of NO3-re. By using Delta17O values of nitrate, we could estimate delta15N and delta18O values of NO3-re by excluding the contribution of NO3-atm in the raw 15N and 18O values. We would like to add an explanation about this in the revised MS.

> p. 5, l. 21-22: “influences of flow stagnation into the lake on nitrate”: What influence is meant here? On nitrate concentration? On isotope ratios? On total amount?

We meant the influences on the concentrations of both NO3-atm and NO3-re, in streams. We have added an explanation about this in the revised MS.

> p. 6, l. 6-14: This paragraph should be moved to the end of the introduction as part of the motivation for the study.

This paragraph presented past study done in the watershed, not the motivation for this study.

> p. 7, l. 7: “To calculate the annual influx/efflux of nitrate via each stream: : : we used the sampling number n”: Unclear how the annual influx/efflux of nitrate was calculated using the sampling number. Please provide a more detailed description of the calculation.

The manner of calculation is presented later in Section 2.4. We have revised this part to avoid misleading readers.

> How were peak flow events after strong precipitation events or after snow melt(if there was) taken into account? Frequently, the solute composition of stream water is significantly altered during peak flow events, and the total annual discharge is often dominated by peak flow events.

As presented in L23/P6 (section 2.2), our estimates on concentrations of nitrate in the streams were based only on those during the base flow periods. The total annual discharge of water determined by Kunimatsu (1995) and cited as Qin in equation (9) in this study; however, includes those during peak flow events in the estimation. Because

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nitrate concentrations of streams during base flow periods almost represents that of subsurface runoff in humid temperate climate, as also presented in section 3.1 (L16-20/P12). Subsurface runoff was a major part of the stream flow, including that during peak flow events (e.g., Dincer et al., WRR, 6, 110-124, 1970; Sklash & Farvolden, J. Hydrol., 43, 45-65, 1979; Mcdonnel et al., WRR, 26, 445-458, 1990; McNamara et al., J. Hydrol., 206, 39-57, 1998; Kobayashi, J. Hydrol., 76, 155-162, 1985). Our estimates on annual nitrate discharge (DeltaNin, DeltaNout) included those during the peak flow events within the errors presented. This conclusion is also supported by other independent estimates on the annual nitrate discharge (DeltaNin, DeltaNout) in the lake (Kunimatsu et al., 1995; Tezuka, 1992; Yamada et al., 1996), based on more frequent measurements of nitrate in the streams.

> p. 8, l. 4-6: The principle of the method should be briefly described, despite the references.

We would like to add this in the revised MS.

> p. 8, l. 15: There is no mention of the method by which the 17O signatures of nitrate were determined. This need to be done here or above.

We would like to briefly present this in the revised MS.

> p. 8, l. 24: How do you define error here and elsewhere in the manuscript? Standard error of the mean? Standard deviation? Or else?

We used the standard error of the mean. We would like to clarify this in the revised MS.

> p. 9, l. 3-4: “showed NO2-/NO3- ratios of less than 5%; thus, the results were used with no corrections.”: How does that translate in the worst case to uncertainty of the nitrate isotope values?

Because NO2- concentrations were too low to determine the stable isotopic compositions, it is impossible to estimate the worst case uncertainty of the nitrate isotope

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values, in principle.

More than 90 % of the samples showed NO2- concentrations less than the detection limit (0.05 micro mol/L) and thus showing the NO2-/NO3- ratios less than 0.2 %. The NO2- concentrations in the samples that could have NO2-/NO3- ratios more than 1 % were also less than the detection limit. Because the NO3- concentrations also were low for the samples (as low as 1 micro mol/L), the possible maximum NO2-/NO3- ratios became 1-5%. As a result, we presented that "all samples showed NO2-/NO3- less than 5%".

The NO3–exhausted samples showing nitrate concentrations < 5 micro mol/L were found only in summer (June or August) when water flow rates were low. As a result, the isotopic compositions in these samples had little influence on quantifying the flow-weighted annual average isotopic compositions in each river in this study. As a result, we used the results without correction, as was also done in most of the stable isotope studies of nitrate in streams in the past. We have added an explanation about this in the revised MS.

> p. 9, l. 8: “flow-weighted”: There is no mention of flow measurements further up in the Materials and Methods section. This needs to be done, and the uncertainty of interpolating nitrate concentrations between four sampling dates only for a whole year needs to be addressed.

Most of the flow rate data used in the calculation was cited from a reference (Shiga Prefecture, 2015) as presented in L17-20/P9, so we did not mention the measurements in the Materials and Methods section of this paper.

As presented in Figure S2, our annual average nitrate concentration almost correlated with those determined in past. Besides, our annual average nitrate concentration in major inflow rivers also correlated with those determined by Shiga prefecture based on continuous monitoring at least every month (not presented in this paper). Based on the dispersion of the correlation as presented in Figure S2, we can estimate that

the uncertainty in the annual average concentration in each river is around 10 micro mol/L. Because the annual average nitrate concentrations almost correlated with those determined in other studies; however, the uncertainty in the total flux (DeltaNin) estimated from equation (13) must be minimum. Rather, uncertainty in alpha in (9) must be much larger for the total flux (DeltaNin) so that we did not take into account the uncertainty derived from interpolating nitrate concentrations between four sampling dates for a whole year.

> p. 9, l. 19: "For small streams with no data for the flow rate, we used a small and stable flow rate of 0.1 m3/s for fn.": For how many of the 33 streams was that the case?

These estimates were applied for 13 streams. We would like to clarify this in the revised MS.

> p. 9, l. 21f.: The calculation of the 15N and 18O values of remineralized nitrate with a two end-member mixing model with atmospheric nitrate as second end member falls short of taking into account also other sources of nitrate, e.g. fertilizer or sewage water.

We did take into account the other sources of nitrate as the source of remineralized nitrate at this point. All the nitrate sources other than atmospheric, such as manure, sewage, and fertilizer, are classified to remineralized nitrate under the definition in this paper, because the oxygen atoms are derived from either terrestrial O2 or H2O through mass-dependent reactions, such as nitrification. We would like to clarify this in Section 1 to avoid misleading readers.

> p. 10, l. 17: What is a "clear normal correlation"? Please specify.

We would like to add the rˆ2 value in the revised MS.

> p. 11, l. 6-7: "The present results imply seasonal and regional changes in the delta18O/Delta17O ratios of tropospheric ozone and in the OH radical.": Are there any references that back up this assumption?

None of which we are aware.

> p. 11, l. 8: “On the basis of both the temporal variation in the depositional flux of NO3-atm: : :”: No temporal/seasonal variation of the depositional nitrate flux has been described further up, and in Fig. 3c there is no clear seasonal pattern of the depositional nitrate flux visible, in contrast to the Delta17O values.

The temporal variation in the depositional flux of NO3-atm was presented in Fig. 3(c). We would like to clarify this in the revised MS.

The seasonal patterns are indicated in the isotopes. We did not claim that we could find clear seasonal patterns in the depositional nitrate flux.

> p. 11, l. 18-19: “additional corrections could be needed“: Were they required? And if yes, how exactly were these corrections done?

They were presented in subsequent sentences, until L25/P12.

> p. 11, l. 19-20 and 24: What do you mean with “NOx oxidation channel”? Pathway?

Yes, we mean a NOx oxidation pathway.

> p. 12, l. 3: “correct for difference in arrival frequency “: What do you mean with “difference in arrival frequency? Please rephrase in an understandable way.

We would like to rephrase this part.

> And has it been corrected for in the present work?

This was presented in subsequent sentences, until L25/P12.

> p. 12, l. 9-12: This statement is too vague and weak. It needs to be backed up with literature, or it should be abandoned.

Why? Four of the four annual observations in mid latitudes reported to the present (La Jolla, Princeton, Rishiri, and Sado) coincided within 2‰ mutual differences.

> p. 12, l. 10: “by allowing an appropriate range of errors presented later”: This “range of errors should be specified here at its first mention.

We would like to add this in the revised MS.

> p. 12, l. 14-18: The residence time of atmospheric nitrate could vary significantly between your different catchments with different land uses. How do you know whether the residence time was similar in all of your catchments to that of forested catchments reported elsewhere?

They were already discussed in the references presented.

> p. 12, l. 21: “we used the obtained Delta17Oavg”: At this stage it is not clear how the Delta17Oavg was obtained.

They were presented on P11. We would like to clarify this in the revised MS.

> p. 12, l. 22-23: “: : :by allowing the error range of 3.0‰ considering the whole factor change of Delta17Oatm from Delta17Oavg.”: What does that mean? Please describe in an understandable way. Why exactly 3.0‰ and not 2‰ 1‰ or any other value?

We estimated the uncertainty derived from the difference in the locality as 1 per mil, based on the standard deviation between the annual average Delta17O values determined in four different monitoring stations located in the same mid-latitudes in the past (La Jolla, Princeton, Rishiri, and Sado). Besides, we estimated the uncertainty derived from the seasonal difference in the Delta17O values of atmospheric nitrate as 1.8 per mil, based on the standard deviation of 6-month moving averages of atmospheric nitrate determined at Sado monitoring station in this study (the six months corresponded to the minimum residence time of water in the watershed). Adding a further 0.2 per mil for a margin, we adopted 3 per mil as the possible error for Delta17Oatm in the streams. We would like to add an explanation about this in the revised MS.

> p. 12, l. 28-30: “As a result, while using the 15Navg and 18Oavg values as 15Natm and 18Oatm, we assumed much larger error range on the values; i.e. 10‰ for both 15N and 18O.” Unclear, how this error was determined. Please describe in more detail.

The possible errors of both delta15Natm and delta18Oatm were originally set as twice

the enrichment factor during assimilation of nitrate (ca. +5 per mil; Granger et al., 2010). As a result, nitrate concentration must be reduced to 1/e^2 the original when delta values increased +10 per mil from the original. If the atmospheric nitrate concentration were reduced to 1/e^2 of the original, it is very difficult to detect. Of course this estimation is less reliable, so that we verified its appropriateness later in Section 3.3. We have added an explanation about this in the revised MS.

> p. 13, l. 10-12: “The spatially continuous variation in the values: : : imply that the values may represent land use changes in each catchment area.”: The annual average values of 15N and 18O vary by 10‰ at the most. Given the uncertainty range of these values of +/- 10‰ (as stated on p. 12, l. 30), how do you want to discern any significant differences here, not to mention to derive any statements about land-use effects on the processing of atmospheric nitrate in the different catchments?

The possible errors (10‰ were that of atmospheric nitrate, not remineralized nitrate. Because atmospheric nitrate occupied only about 5% of total nitrate on the average, the variation range was reduced to 0.5‰ much smaller than the regional variations.

> p. 14, l. 24: “determined recently“: By whom? No reference provided.

The references were provided in the subsequent sentences, from L24/P14 to L28/P14. We would like to rewrite this part to avoid misleading readers.

> p. 15, l. 2-4: “We concluded that the 18O value of NO3-re produced through nitrification in the temperate watershed having 18O(H2O) values of -7.8 ± 1.0‰ was -2.9 ± 1.2‰ and that we should use such a low 18O value: : :”: Did the soil and/or stream water have this delta18O(H2O) values of -7.8 ± 1.0‰ If yes, please make this clear in this sentence. If not, then the basis for this conclusion is not clear.

The delta18O value was the average of the delta18O(H2O) value of the streams studied in this paper. We would like to clarify this in the revised MS.

> p. 15, l. 8: “Although the Delta17O values of nitrate were stable during the biogeo-

chemical processing": Again, if nitrate is biogeochemically processed, then also the Delta17O gets lost.

Not just "during the biogeochemical processing" but "during the biogeochemical processing such as partial removal through assimilation or denitrification (L8-9/P15)". We would like to clarify this in the revised MS.

> p. 15, l. 13-16: "We concluded that the range of isotopic fractionations : : : was generally small": The basis of this conclusion remains unclear. Please explain in more detail.

If such isotopic fractionations were significant for the portion of atmospheric nitrate in total nitrate, the data should be plotted on the 18O-enriched side in Figure 6, especially for those enriched in atmospheric nitrate (i.e. those showing high Delta17O values). We would like to emphasize this in the revised MS.

> p. 15, l. 16-19: "This result also supports our assumption in section 3.1 such that the actual 15N and 18O values of NO3-atm in each stream water sample : : : correlate with the 15Navg and 18Oavg estimated at Sado-seki monitoring station within an error of +/-10‰Ź": This refers to the previous sentence, which does not report a result but a conclusion, the basis of which remained unclear. That is, the statement made in section 3.1 has been based on very weak grounds.

"The statement made in section 3.1 has been based on very weak grounds" was the reason we verified the appropriateness here in Section 3.3.

> p. 15, l. 30-31: ": : :responsible for the positive correlation between the 15N values of total nitrate and population density.": Was this correlation significant? I could not find any statistical information.

We would like to add the rˆ2 value (0.64) in the revised MS.

> p. 17, l. 1-3: ": : :the slight deviations in the reported 15N and 18O values from our results can be explained by the following factors: : :": Could also different sources of

ammonium for nitrification could have played a role (soil, sewage water, fertilizer)?

If your comment meant the contribution of 15N-poor ammonium for nitrification on the "sewage water" nitrate reported in this study, it could be possible. However, please note that what we insisted here was the observed differences in the values of delta15N and delta18O were insignificant. The explanation for the observed differences presented here (contribution of NO3-atm and progress of denitrification) was just one the most possible explanation. The detailed discussions on the differences should be done elsewhere, adding the Delta17O data of sewage water nitrate.

> p. 18, l. 17f.: It is unclear whether this snow signal of atmospheric nitrate could be captured by the sampling design of only four samplings per year.

In the watershed studied, monitoring both the nitrate concentration and the flow rate has been done for the major streams, periodically at least every month. This is the reason we chose March, June, August and November for the samplings. One of the reasons we chose March was to capture the snow signal season (usually from February to April), when the flow rates were at maximum every year.

> p. 23, l. 3: “The estimated annual average Delta17O value of inflows, +1.3‰ : : :”: Unclear, where this value comes from. Please explain.

The annual average Delta17O value of inflows was estimated using equation (15). We would like to clarify this in the revised MS.

> p. 23, l. 4: “: : :average mixing ratio of NO3-atm within total nitrate of 5.1+/-0.5%...”: This value shows only up here and in the abstract, but it is unclear how and when it was calculated.

The annual average mixing ratio of NO3-atm within total nitrate was estimated from the annual average Delta17O value of inflows (+1.3‰ using equation (2). We would like to clarify this in the revised MS.

> p. 23, l. 6-7: “: : :the remainder of the nitrate was of remineralized origin (NO3-

re) likely produced through nitrification within the catchments: : :": Again, what about direct input of nitrate via fertilizer and/or sewage water without remineralization?

As already presented, all the nitrate sources other than atmospheric, such as manure, sewage and fertilizer, are classified to remineralized nitrate under the definition in this paper, because the oxygen atoms are derived from either terrestrial O2 or H2O through mass-dependent reactions, such as nitrification. We would like to clarify this in Section 1 to avoid misleading readers.

> p. 23, l. 25: "Lake Biwa also acts as a net sink for fixed N": The question is what happens with the processed nitrate? Very likely most of it is denitrified and lost to the atmosphere as N2O and/or N2. Thus, the statement that Lake Biwa acts as a net sink for fixed N is questionable.

Why ?? The fixed-N (= nitrate + ammonium + organic-N, etc.) removal to the atmosphere as N2 through denitrification is no doubt a sink for fixed-N, because N2 is not fixed-N anymore. Because the total fixed-N outflux via streams was less than total fixed-N influx via streams in Lake Biwa as presented in L10-27/P23, Lake Biwa did act as a net sink for fixed-N.

> Table 1: This table should also include the dominating land use in the respective catchment.

We would like to add this in the revised MS.

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: Drs. Takanori Miyauchi, Takuya Ohyama, Daisuke D. Komatsu, Fumiko Nakagawa, Yusuke Obata, Keiichi Sato, and Tsuyoshi Ohizumi

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