Reply to your comment (Referee #1).

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

> For example, Sebestyen et al. 2019 (ES&T) and the references it contains address a similar issue as this manuscript (e.g. Buda et al. 2009 and Sabo et al. 2016 both sampled storm events), and this manuscript could do a more throughout job of using those studies to help justify this study (in the Introduction) and then comparing/contrasting the results of this study to those studies in the Discussion. Similarly, oher studies (Burns et al. 2009; Barnes et al. 2010; Bostic et al. 2021) have addressed similar questions in non-forested systems and could be useful for helping to provide a broader context for the results that are presented in this manuscript.

Thank you for your advising. We would like to cite many of the suggested articles in the revised manuscript.

> Title: “Enriched” is a word that is often used incorrectly in the isotope literature to refer to increased values of the heavier isotope. Here I believe the authors use “enriched” to mean increased nitrate concentrations, which is might cause confusion given that this paper also talks about isotopic enrichment (e.g lines 66 and 303). One solution might be to simply delete “enriched” from the title and another solution might be replace “nitrate enriched” in the title with something like “elevated nitrate concentrations”

Thank you for your advising. We would like to revise the title as “Tracing the source of nitrate in a forested stream showing elevated concentrations during storm events” in the revised manuscript.

> Lines 2-3: This sentence implies that nitrate concentrations always increase in temperate forest streams everywhere. Is that true? If not, perhaps slightly adjust this sentence.

While many past studies reported increasing the stream nitrate concentration during storm events in temperate forest (e.g. Creed et al., 1996; Kamisako et al., 2008; Christopher., 2008), the decrease pattern (Christopher., 2008) or stable pattern (Shanley et al., 2011) of stream nitrate concentration during storm events also have been reported. Thank you for your advising. We would like to revise the sentence as suggested.

> For example, do some severely nitrogen saturated forests that show higher NO3 concentrations in baseflow than stormflow?

No. For example, the KJ forested catchment has been reported under severely nitrogen saturation (Nakagawa et al., 2018), both this study and past study (Kamisako et al., 2008) found significant increase in the stream nitrate concentration during stormflow than baseflow. Thank you for your advising.

> Line 5: Please tell the reader what time of year (winter, spring, summer, autumn) these storm events occurred.

Thank you for your advising. We would like to revise as suggested.

> Line 6: It might be helpful to insert "increasing" before "from" to help the reader understand that the "variation" nitrate concentration that was observed was primarily an increase in concentrations.

Thank you for your advising. We would like to revise as suggested.

> Line 14: I believe "($d^{15}N$, $d^{18}O$, and $C^{17}O$)" can be deleted without sacrificing meaning.

Thank you for your advising. We would like to revise as suggested.

> Line 26-27: Could the authors support this claim by calculating annual export of NO3-atm (and NO3-terr) using their concentration and flow data?

The annual export of unprocessed atmospheric nitrate in the stream can be calculated as 3.2 ± 0.7 mmol m^{-2} yr^{-1} by multiplying the average flow rate of stream and the average concentration of unprocessed atmospheric nitrate in the stream during the routine observation.
The annual export flux of unprocessed atmospheric nitrate relative to the annual deposition flux (M_{atm}/D_{atm} ratio), nitrogen saturation index, was estimated from annual concentration of unprocessed atmospheric nitrate in the stream, annual flow rate of stream, and annual deposition flux of atmospheric nitrate. In the forested catchment, the annual flow rate of stream and annual deposition flux of atmospheric nitrate can be considered as constant. The concentration of unprocessed atmospheric nitrate in the stream was 1.6 ± 0.4 μM, 1.8 ± 0.4 μM, and 2.1 ± 0.4 μM during the storm events I, II, and III, respectively, which have no significant difference with the average concentration of unprocessed atmospheric nitrate in the stream (2.2 ± 0.6 μM). Thus, the storm events have little impacts on the M_{atm}/D_{atm} ratio. Thank you for your advising. We would like to clarify this in the revised MS.

> Lines 26-30: Is this conclusion specific to the author's study site (or certain types of forests) or are they suggesting that is a more broad/general conclusion that applies to forested catchments everywhere?

We conducted the research at KJ forested catchments as an example of the nitrogen saturated forested catchment. The conclusion is only suitable for the KJ forested catchment at present. Further works should be needed to verify the conclusion in different forested catchments in the future.

> Line 33: "representative" of what? Please clarify.

Thank you for your advising. We will clarify this in the revised manuscript.

> Line 50: First, how are the authors using "overland flow" here and elsewhere (e.g. line 463) in the manuscript? My understanding is that overland flow is unlikely in areas that are not near channels or stream/riparian areas in forests except for unique situations, such as intense rain events or rain that occurs on frozen soils. Second, I don't believe either of the cited studies suggest that overland flow is a mechanism for direct suppler of atmospheric nitrate to stream water. As far as I recall, Kaushal et al. didn't show overland flow for their forested site and Sebestyen et al. talked about routing of NO3-atm along flow paths that allowed NO3-atm to bypass uptake/processing (but not specifically about overland flow).

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

> Line 72: Is beta completely constant or can it exhibit some variation around 0.5279? If so, does the variation affect the authors data analyses or interpretations?

Bostic et al. (2021) assumed the β could vary from 0.51 to 0.53. In the whole samples analyzed in this study (n=105), both the min and max value of the $\delta^{18}O$ was -3.3 ‰ and +7.7 ‰, respectively. Thus, the max range of deviation in the $\Delta^{17}O$ could be estimated to be 0.1 ‰ (Fig 1), which in accordance with our analytical standard error of $\Delta^{17}O$. As a result, our conclusion cannot be influenced by the variation of β. Thank you for your advising.

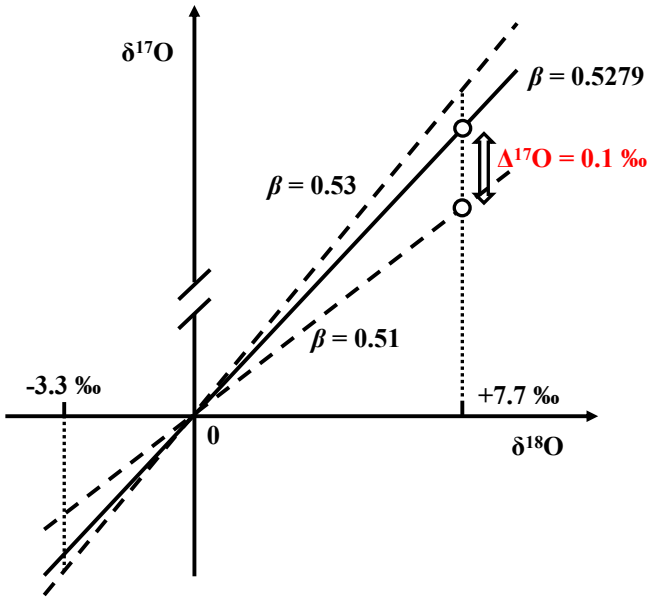

Figure 1. Schematic diagram showing the max variation of the $\Delta^{17}O$ in accordance with the variation of the β from 0.51 to 0.53 in this study.

> Lines 162-164: It seems like there would be potential for microbial alteration of the samples during the 1-2 weeks that they stayed in the field before being returned to the lab. Did the authors assess this?

We think the microbial alteration of the stream water samples during the storage period can be negligible.
(1) The sampler was set on the riverbank near a weir surrounded by ferns and other understory vegetation avoiding sunlight during the observation. In addition, the bottles are stored in a shaded space to minimize the microbial alteration of the samples. Besides, the automatic sampler (SIGMA 900, Hach, USA) has equipped with refrigerator to keep the samples in 4°C. (2) Kotlash and Chessman (1998) have assessed the storage effects of freezing, acidification, refrigeration and extended storage without refrigeration (6 days) on measured concentrations of nitrogen of different stream water samples, and found there was little difference in concentration of oxidized nitrogen (NO_3^- + NO_2^-) according to different treatment. (3) The concentrations of stream nitrate showed temporal variation in accordance with the variation in the stream flow rate during storm events (Figs. 3 and S1). As a result, the variation of the stream nitrate concentrations was primarily controlled by the flow rate instead of the microbial process. (4) The $\Delta^{17}O$ of stream nitrate is stable during the progress of such microbial processes (e.g., denitrification or assimilation). While the $\delta^{15}N$ and $\delta^{18}O$ of stream nitrate can be altered by the progress of partial removal through microbial process, the $\delta^{15}N$ and $\delta^{18}O$ of stream nitrate showed strong linear relationship between the reciprocal of concentrations, implying that the primary

process controlling both $\delta^{15}N$ and $\delta^{18}O$ was mixing. Thank you for your advising. We would like to emphasize this in the revised manuscript.

> Lines 184-185: How many "local laboratory nitrate standards" were used and what are their isotope values?
> Lines 205-206: What data were used to calculate the reported standard error of the mean for each isotope? For example, was precision determined from the lab standards, replicate samples, or something else?

In this study, we used three kinds of the local laboratory nitrate standards, which were named to be GG01 ($\delta^{15}N$ = –3.07 ‰, $\delta^{18}O$ = +1.10 ‰, and $\Delta^{17}O$ = 0 ‰), HDLW02 ($\delta^{15}N$ = +16.11 ‰, $\delta^{18}O$ = +22.20 ‰), and NF ($\Delta^{17}O$ = +19.16 ‰), which the GG01 and the HDLW02 were used to determine the $\delta^{15}N$ and $\delta^{18}O$ of stream nitrate, and the GG01 and the NF was used to determine the $\Delta^{17}O$ of stream nitrate. The standard error of the mean of the isotopic compositions ($\delta^{15}N$, $\delta^{18}O$, and $\Delta^{17}O$) was determined by repeated measurements of the GG01 (n = 3), ±0.17 ‰ for $\delta^{15}N$, ±0.25 ‰ for $\delta^{18}O$, and ±0.10 ‰ for $\Delta^{17}O$, respectively. Thank you for your advising. We would like to clarify this in the revised manuscript.

> Line 226: How was the error range "allowed"?

We estimated the uncertainty derived from the difference in the locality as 1 ‰. This was based on the standard deviation between the annual average $\Delta^{17}O$ values determined in four different monitoring stations located in the same mid-latitudes, in the past studies such as La Jolla (33° N; Michalski et al., 2003), Princeton (40° N; Kaiser et al., 2007), Rishiri (45° N; Tsunogai et al., 2010), and Sado (38° N; Tsunogai et al., 2016). Besides, we estimated the uncertainty derived from the seasonal difference in the $\Delta^{17}O$ values of atmospheric nitrate as 1.8 ‰, based on the standard deviation of six-month moving averages of atmospheric nitrate determined at the Sado monitoring station. Adding an additional 0.2 ‰ as a margin, we adopted 3 ‰ as the possible error for $\Delta^{17}O$ atm in the streams.

> Line 279: I believe "events" should be singular.

Thank you for your advising. We would like to revise as suggested.

> Line 353: I suggest inserting "primarily" or "likely" before "responsible" here and elsewhere that this conclusion is presented. The soil and stream data the authors are using come from different years as they describe on lines 318-342, so I think the conclusion on lines 351-354 should be considered tentative.

Thank you for your advising. We would like to revise as suggested.

> Lines 389-390: Please indicate which symbols indicate upland samples and which indicate riparian samples.

Thank you for your advising. We would like to revise as suggested.

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

Sincerely,
Weitian Ding

Reference

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