Referee #1

Manuscript: ACP-2019-829 (Yu et al.) Title: The isotopic composition of atmospheric nitrous oxide observed at the high-altitude research station Jungfraujoch, Switzerland.

This manuscript presents measurements of the isotopic composition of N_2O obtained from a high-altitude European site – Jungfraujoch in Switzerland, using a recently developed QCLS coupled with a preconcentration unit. The system provided direct and individual measurements of four N_2O isotopocules at an ambient level of N_2O. From the extensive data sets covering the 5-year study period, authors attempt to derive seasonality and interannual trends in N_2O isotopic compositions and discuss them in combination with observed changes in N_2O mixing ratio. Overall, the writing and figures are clear, and the methodology maximizes the functionality of a high-quality dataset. I encourage the publication of this important work, with only a few minor considerations/edits suggested below.

1. LN 186: Sphinx observatory→ Sphinx observatory in the Jungfraujoch station

R: OK

2. LN 357-364: Authors determined annual growth rates of N_2O mixing ratio for all in-situ data from 2014 to 2018, with/without the 2014 GC-ECD data, and free tropospheric data only, respectively. Given their 1-sigma values, it seems there are some discrepancies between the entire dataset vs sub-sets of data. Authors did mention some about those discrepancies in lines 548-553. However, if authors thought that they are statistically significant, then additional explanations should be given here, rather than later.

R: Thanks for the suggestion. In section 3.1, we have further elaborated this: "This difference in N_2O growth rates is probably due to the limited data quality of GC-ECD, although a lower growth rate in 2014 compared to 2015-2018 cannot be excluded.".

3. LN 375-380: The observed, de-seasonalized trends of delta15N_SP for the whole dataset increased, while delta15N_SP trend showed a decrease when PBL-influenced air samples were excluded. So, authors stated that it implies an impact of local sources. Does it mean that the potential local sources have high delta15N_SP signals? What could it be? Based on the two-box model approach using the current data, authors determined the average isotopic signatures for anthropogenic sources were lower than those for the background troposphere (LN 394-397). If so, the local sources mentioned above could not be associated with anthropogenic sources?

R: The authors agree, that the increasing trend of $d^{15}N^{SP}$ observed between April 2014 and December 2018 at Jungfraujoch (Fig. 2) and the decreasing trend over longer timescales as derived with a two-box modelling approach using the EDGAR emission inventory (Fig. 6; original version) might look inconsistent.

However, it is noteworthy that, the deseasonalized trends of $\delta^{15}N^{SP}$ at Jungfraujoch were not statistically significant, with/without filtering for impact from planetary boundary layer (LN 375-377_Original version). The only significantly positive trend

of $\delta^{15}N^{SP}$ was found in the first phase (Table 1). Although mean $\delta^{15}N^{SP}$ values of N_2O sources according to EDGAR emission inventory are lower than that observed in tropospheric background (Table S2), a changing proportion of N_2O-emitting soil process, i.e. nitrification vs. denitrification, with $\delta^{15}N^{SP}$ values of 33‰, as compared to about 0‰ (Sutka et al., 2006), might rationalize this inconsistency. This shift in the isotopic signatures of anthropogenic sources, might be interpreted as a climate change feedback, as discussed in section 4.4. Similarly, Park et al. (2012) attributed an increase rate of 0.06‰ a^{-1} in $\delta^{15}N^{SP}$ in 2005 to 10% increase in the relative contribution of nitrification to global N_2O production since 1975. This is already discussed in section 4.3.

In the two-box model approach, the estimation of isotopic signatures for anthropogenic sources mainly depends on the measured current and predefined preindustrial N_2O mixing ratios and isotopic signatures (Table S1). As shown in Figure 6 (original version), the simulated trend of $\delta^{15}N^{SP}$ in the troposphere is negative, consistent with the lower $\delta^{15}N^{SP}$ for anthropogenic sources than for the tropospheric background. The current (insignificant) increase in $d^{15}N^{SP}$ at Jungfraujoch, might be evaluated with the two-box model approach in the future, if extended time-series of isotope data will become available (e.g. Prokopiou et al., 2017).

4. LN 405-409: Authors found that there were differences in seasonal patterns of all isotopes between the entire dataset vs. the second phase data. Authors then added that the seasonal variations for free tropospheric samples were similar to those for the whole dataset. Does it imply that the second phase patterns could more represent the PBL-influenced data?

R: As indicated in LN 401-409 (original version) and Figure 3, we found a significant seasonal pattern of $\delta^{15}N^{SP}$, with a summer minimum, for both the whole dataset and the second phase. For $\delta^{15}N^{bulk}$, a significant seasonal pattern was seen in the whole dataset but not in the second phase (seasonal variability > uncertainty). Hence, our results do not necessarily indicate that seasonal patterns were different between the entire and the second-phase data.

Air mass footprints suggest that, in 2017 (second phase), discrete sampling received less contribution from free troposphere than in the other years (Fig. 4b), possibly pointing to a stronger influence of PBL. However, this is not supported by *in situ* NO_y and CO measurements (Figure S6b; no clear difference), which has been suggested as a more effective indicator for free troposphere (Herrmann et al., 2015). Given the larger uncertainty in seasonality-analysis due to lower sampling frequency in the second phase (Section 3.4), it is difficult to draw the conclusion that such "insignificant" changes in seasonal patterns in the second phase are due to a stronger PBL influence.

5. LN 421-428: Authors seem to suggest strong exchange with the PBL in summer, based on the observed summer maxima in the monthly seasonal cycles for O_3 and NO_y mixing ratios. But it is not so clear that the summer maxima in O_3 and NO_y could support a stronger air mixing with the surface and thus a PBL impact on the seasonal

changes in N_2O isotopic compositions, because the maxima in O_3 and NO_y mixing ratio occur in summer most likely due to stronger sunlight.

R: We agree that O_3 alone may not be a good indicator for air exchange with PBL, as elevated O_3 concentration at Jungfraujoch can be due to air exchange with PBL and/or stratosphere. Therefore, the text in section 3.4 has now been revised. However, NO_y : CO used in this study has been previously tested to be an effective indicator for determining the age of air mass, i.e. to identify recently polluted air transported to Jungfraujoch from the PBL (Herrmann et al., 2015; Zellweger et al., 2003). In addition, air mass footprint analysis supports such pattern with lowest source sensitivity from free troposphere in summer (Figure 4).

6. LN 453-476: In the results section, authors analyzed the seasonal variabilities for not only the entire datasets but also the second phase data, but in the seasonality discussion, the seasonal patterns derived from the second phase data were not discussed, even though the second phase patterns might contains more the surface-influenced signals (see the comment #4). If authors decided not to consider the second phase seasonality, please add statements for the reason in the text.

R: For the second phase, the seasonal patterns of $\delta^{15}N^{bulk}$ and $\delta^{18}O$ were not significant, while $\delta^{15}N^{SP}$ showed a significant seasonal pattern similar to that for the whole dataset. Therefore, it was not specifically discussed. Nonetheless, we thank the reviewer for the suggestion and have added a few more discussion points in section 4.2, regarding the seasonal variabilities of N_2O isotopic signature in the second phase.

7. LN 488-505: Fig. 5 demonstrated that direct/indirect agricultural source contributes most to the N_2O enhancements, particularly in summer. Then considering peak N_2O fluxes and minimum of delta15N_SP observed in summer, does it suggest that the local agricultural activities enhanced N_2O production by "denitrification"? Are there any studies to support this result?

R: Yes, one isotopic study of N_2O emissions from Swiss grassland (Wolf et al., 2015) suggested that N_2O emissions in summer periods were mostly contributed by denitrification, given that high N_2O fluxes were associated with low $\delta^{15}N^{SP}$ values (below 5‰). This has been confirmed again in a recent study (Ibraim et al., submitted to Global Biogeochemical Cycles) showing that the $\delta^{15}N^{SP}$ of N_2O emitted from a managed grassland during a late summer was consistently within a small range of 0-10‰, regardless of soil water-filled-pore-space.

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