

Review on “The isotopic composition of atmospheric nitrous oxide observed at the high-altitude research station Jungfraujoch, Switzerland”

This manuscript described the 5-year observations of nitrous oxide (N_2O) mixing ratios and their isotopic compositions at Jungfraujoch using laser spectroscopic technique for the first time. The long-term observations of N_2O isotopocules allow the authors to characterize the integrated isotopic signatures of anthropogenic sources that have been emitted since the industrial revolution and to identify the main processes governing the seasonality of N_2O . The authors utilized a two-box model and a Lagrangian particle dispersion model to characterize the isotope signatures of anthropogenic sources that contribute to the atmospheric increase of N_2O concentration. The unique observations of N_2O isotopocules in the middle of the European continent and the interesting interpretation of data makes worth publication. Notwithstanding, there are several hazy spots in the manuscript which needs to be revised in order to avoid any confusion.

Major issues:

1. Application of a two-box model assumes the data obtained at Jungfraujoch to represent the variability of N_2O mixing ratios and its isotopocules in the troposphere. This appear to contradict to the use of footprint model to characterize the isotopic signatures of the anthropogenic sources in the European continent. This is demonstrated in Table 2 and 3 that the isotopic signatures of the anthropogenic N_2O are different. In the text on the lines from 626 to 635, the authors ascribed it to the different isotopic signatures of N_2O source emissions in the model. However, as shown in Table 2 and mentioned in the text (on the line of 612), the single spot observation won't be representative the global scale of atmosphere, but would represent the regional characteristics of N_2O . The long-term trends of N_2O isotopocules listed in Table 1 also support that the observation at Jungfraujoch does not represent the tropospheric variability of N_2O . Contradict to the global trends of isotopocules shown in Figure 6, the observations of $\delta^{15}\text{N}^{\text{SP}}$ and $\delta^{18}\text{O}$ are positive trends at Jungfraujoch. In view of these contradict aspects revealed in the observation and the model, the isotopic signatures of the anthropogenic source will not help understand the contribution of anthropogenic source to the increase of atmospheric N_2O . I would suggest limiting the data interpretation in regional scale.
2. The long-term observation at one station allowed seasonal variation to be explored. The authors argued the minimum N_2O concentration observed in late summer is driven by STE which is also evidenced by the enrichment of ^{15}N in the N_2O driven by the photochemical destruction in the stratosphere. On the other hand, $\delta^{15}\text{N}^{\text{SP}}$ and $\delta^{18}\text{O}$ did not seemingly synchronize the STE event, which, the authors argued, the N_2O emitted from the soil overwhelms the effect by STE. If these two processes govern the seasonality of the atmospheric N_2O , I would suggest quantifying how to compete these two processes along the year at Jungfraujoch.

Minor issues and technical comments:

1. L 52: The publication year of Tian et al. (2018) is 2019.

2. L 171: “gas chromatography” should be “gas chromatograph” in the context.
3. L 170 – 184: Since no references are given, I suggest describing the analytical methods in detail including the calibration of the system for the analysis of N₂O, CO, NO_y, and O₃ mole fractions perhaps in the section of Supporting Information.
4. L 217: Have you tested the mole fraction dependency of the isotope ratios of N₂O? Here, the amount of N₂O for the QCL is 45 ppm. However, Mohn et al. (2010, 2012) concentrated ambient air to > 60 ppm of N₂O.
5. L 218: I think the citation of Harris et al. (2017) should be Harris et al. (2014).
6. L 236: What are the matrix gases in CG1 and CG2 standards?
7. L 253: In Figure S2, the scattering of isotope ratios in the second phase look larger than that in the first phase, particularly for δ¹⁸O. Is it statistically insignificant?
8. L 313 – 314: T_{PI} and T_{PD} should be replaced to τ_{PI} and τ_{PD}.
9. L 353: It’s misleading. Fig. S3 shows the agreement improved since the year 2015 when GC-ECD was replaced to OA-ICOS, NOT in the second phase.
10. L 358 – 361: Provide the ground that the N₂O growth rates of 0.880±0.001, 0.993±0.001, and 0.93 are in agreement. Statistically they are different each other unless standard deviation of the global growth rate of 0.93 (by NOAA) is larger than ~0.02.
11. L 361: Add the literature (WMO, 2018) next “NOAA (0.93 nmol mol⁻¹ a⁻¹)”.
12. L 362 – 364: The annual growth rate, 0.813±0.027 is not lower than the value 0.858±0.002 within 2 standard deviations.
13. L 376: The authors indicate the insignificant increasing trend of δ¹⁵N^{SP} and δ¹⁸O. However, their standard deviations do suggest significant increase of them within 1 sd. It needs to be clarified.
14. L 383 – 391: It needs explanation why the trends of δ¹⁵N^{SP} and δ¹⁸O during the first phase is one order of magnitude larger than that in the second phase.
15. L 438: I would suggest moving Fig. S7 onto the main text as it is the unique visualization to illustrate Lagrangian footprint of isotopic signatures of the sources.
16. L 442 – 451: The section 4.1 does not seem to benefit the main theme of this manuscript. It rather makes the manuscript loose. Analytical quality has already mentioned in the section 2.4 Data analysis (see the lines 246, 252 – 253) and the excellent analytical repeatability for δ¹⁵N^{SP} by QCL is well described in Mohn et al. (2014).
17. L 458: Decock and Six (2013) does not describe the STE process at all. Is it an error in citation?
18. L 459: Add superscript “bulk” next ¹⁵N.
19. L 461: Comparing Figure 3(a) in Toyota et al. (2013) with Figure 1a here, it does not look “almost identical”, but perhaps comparable. The monthly mixing ratio of N₂O at Jungfrauoch is at maximum in June while in April at Hateruma Island, Japan.
20. L 464: What are the underlying mechanisms?
21. L 511: Provide the regression coefficients in Figure S8.
22. L 514: δ¹⁵N^{bulk} in Figure S7 is not particularly high in spite of potential influence of STE. It needs to be clarified.
23. L 537: Add minus sign before 0.06.
24. L 558 – 559: Rahn and Wahlen (2000) do not provide clear evidence on the influence soil water vapor to oxygen isotope in N₂O, but they speculated. Thus, it would appropriate to write “... assuming that ...” instead of “... given that ...”.

25. L 605: The authors' argument is not clear here. Based on the isotopic signatures of the anthropogenic N₂O, long-term observation at Jungfraujoch indicates the significant contribution of denitrification process in soil while the results from Park et al. (2012) or Prokopiou et al. (2017) favor nitrification process in soil. This is clearly contradicted each other.
26. L 617: Figure 6 shows that $\delta^{15}\text{N}^{\text{bulk}}$ from Jungfraujoch are higher than any other values including Park et al. (2012) and even Toyota et al. (2013). Thus, this sentence does not help explain why $\delta^{15}\text{N}^{\text{bulk}}$ of the anthropogenic N₂O from the observation at Jungfraujoch is higher than the value by Park et al. (2012).
27. L 618: It is impossible to mention trends of $\delta^{15}\text{N}^{\text{SP}}$ as the data is too scattered. In addition, $\delta^{15}\text{N}^{\text{SP}}$ at Jungfraujoch shows positive trends, too (Table 1).
28. L 652: What do the authors mean the "higher-frequency temporal variation" for $\delta^{15}\text{N}^{\text{SP}}$ and $\delta^{18}\text{O}$? Is it relevant to soil emission? Please state it clearly.
29. L 656: Table 2 clearly shows the isotope signatures from Jungfraujoch differ from the values obtained at other sites, opposite to the statement here.