

We would like to thank the anonymous referee #2 for his detailed review of our manuscript and for his precious comments and suggestions, which have led to a clear improvement of our manuscript. Below, the referee's comments (RC2) are displayed followed by the authors' response, which are highlighted in grey.

General observations and detailed comments:

The paper by Ibraim and co-authors presents a novel technique to measure a suite of isotopic fingerprints of N₂O using a field-deployable device. Since N₂O-emissions and their isotopic ratios vary spatially and temporally to a large degree, such an instrument is very useful to allow extent our knowledge on the processes driving these N₂O emissions. The measurements presented in the paper demonstrate that the QCLAS instrumentation works well under field conditions and allows its implementation in further studies. This successful field deployment is certainly a central step after a yearlong construction and test phase in the laboratory and I congratulate the team for this effort. Likewise it is clear to the authors of this study that measuring the isotopic information of N₂O is just one step, and to interpret and exploit the data requires extensive other knowledge, ranging from meteorological boundary conditions, soil analyses, to interpreting the application of manure on the sampling site. As further outlined below, some of these non-measurement aspects of the paper should be improved to gain more clarity for the readers. Since the current measurement set up was not perfect, i.e. only nighttime measurements provide robust results, a lot can be learned from this measurement campaign for future field applications. At some places, the authors already suggested how these limitations could be overcome with a better design etc. I have the impression that these “lesson learned statements” could be extended to guide future measurement campaigns in this area. In general, the paper is clearly structured, the figures are mostly instructive and the text is written nicely. I therefore welcome this paper for final publication after my and the other reviewers comments are included in the final version.

RC2 Comment 1

In section 4.4.6 (page 13) you discuss the influence of the manure application (12th July) on the calculated source signatures of the emitted N₂O. A causal link between excessive nitrogen addition on subsequent N₂O emissions from the soil is to be expected and might be the case. However, looking at the flux time series in Fig. 3 it is equally clear that N₂O fluxes rise after intense rainfall events and this also fulfils expectation. The highest N₂O emissions within the entire study follow the strong rainfall event end of July - here without a manure application. However, the manure application on the 12th July is almost synchronous with the rainfall event; the farmer apparently waited for rain to apply the manure. My feeling is that the discussion in section 4.4.6 focusses too strongly on the manure application, while the more likely driver behind the rising N₂O emissions (intense rainfall) is not really discussed equally. A second argument for the “heavy rain hypothesis” is also the wide footprint of the isotope measurements. From Fig. 7 I get the impression that the largest fraction of the emissions stems from outside of the dashed rectangle where the site is located. Only 15 to 30% of the N₂O emissions came from the local field where the manure was applied, while the largest fraction comes from an area of a few km distance. It might be that the other fields in the surroundings were also fertilized at the same time by the farmers and thus the De-Fen site is representative, but this information is missing. I might be wrong, but you might gain additional insight to view the N₂O flux data and the isotopic signatures also from this heavy rainfall point of view (likewise WFPS) and a wider, more realistic regional footprint. In this respect, you might also put less weight on the NH₄ and NO₃ soil extracted solute data because these data might be too local for the footprint of the measured N₂O isotope signatures.

Authors' response: The authors agree that N₂O emissions and isotopic signatures are driven by both manure (substrate) addition and rainfalls (WFPS) and it might not be possible to disentangle both effects for the manure application on 12 July. The effect of WFPS is explicitly discussed in the section 4.1 ("N₂O fluxes and WFPS") as well as in the sections 4.4.3 and 4.4.5. Nevertheless, we acknowledge the reviewer's comment on the fact that the N₂O emissions strongly raised after rainfall events since each of the three major rainfall events on 24 June, 12 July and 23 July was followed by a clear increase of the N₂O emission rates, which is in agreement with literature (Peng et al., 2011; Liu et al., 2014). Unfortunately, for the two "rainfall – only" events no N₂O source signatures could be retrieved: around 24th June the observed data did not allow for significant Keeling plot analysis, while on 23rd July the measurement campaign was terminated and the fractional measurement data did not allow calculation of N₂O isotopic composition. We also agree that the footprint of the N₂O isotope measurements goes beyond the fertilized plot. For this reason, we added a number of statements to section 4.4.6:

Page 13 line 7: After the manure application on 12 July and rainfall events in the days thereafter a strong ...

Page 13 Line 9: ... in the isotopic composition of the applied precursors, by an enhanced fractionation due to higher substrate availability or changes in process conditions (e.g. WFPS, see sections above).

Page 13 Line 14: ... during nitrification may have been used as substrate for denitrification, given the increase in WFPS due to intensive rainfall events.

RC2 Comment 2

As suggested in your conclusions, it will be valuable in future measurement campaigns to also sample air from chambers that are more representative of the site. In other words, the precious isotope measurement time during the day or for meteorological situations that do not lead to sufficient N₂O accumulation in the boundary layer could be better invested.

Authors' response: In our second field study, which took place between July and December 2017 in Central Switzerland, we deployed automated flux-chambers at the day-time according to a specifically designed measurement schedule. This work is currently in preparation for publication.

RC2 Comment 3

Please mention the footprint shown in Fig. 7 earlier in the paper. It would be helpful for some readers (including me) to be aware that the actual footprint of the N₂O isotope data is more extended than what is visualized in Fig. 1.

Authors' response: We agree and we add the following sentence to the end of the section 2.1.1, where the De-Fen site is characterized:

Page 3 line 37: "[...] respectively (Raiffeisen Laborservice, Ormont, Germany). The average footprint area for N₂O flux and isotope measurements is given in Figure 7"

RC2 Comment 4

Page 1, Lines 33 and 37: (i) mentioning the sink term" while N₂O reduction acted as a major sink" may not clear to all readers. Does this refer to a consumption of N₂O produced in the soil itself or also for ambient atmospheric N₂O, i.e. a net sink to the atmosphere? (ii) "N₂O reduction

to N₂ largely dictated the isotopic composition of measured N₂O. “ Does this statement refer to all measured isotope ratios; this statement seems very general.

Authors' response:

- (i) Page 1 line 37: In the given context our statement referred to soil produced N₂O. We cannot exclude reduction of ambient N₂O but at least no net uptake of N₂O was observed. To emphasize this fact we added the following statement: "[...] source for N₂O, while N₂O reduction acted as a major sink for soil produced N₂O."
- (ii) The dual isotope mapping approach (Fig. 8) indicates that irrespective of the selected approach (SP versus $\Delta\delta^{15}\text{N}$ or SP versus $\Delta\delta^{18}\text{O}$) and scenario (1: reduction first, 2: mixing first) differences in SP to pure BD/ND were mostly controlled by N₂O reduction. We therefore would like to keep this statement.

RC2 Comment 5

Page 3, Lines 35: stick to one name for the management “cutting” vs moving (caption Fig. 4)

Authors' response: Done. "Mowing" was changed to "cutting" at the following positions: page 6 line 34, page 6 line 40 and caption of Figure 4 at page 21 line 9.

RC2 Comment 6

Page 3, Line 30: Site name: Could you use just Fendt as the site name in your paper rather than the awkward De-Fen (I know the acronym De-Fen is the more official in terms of the European Flux Database cluster).

Authors' response: We would like to keep “De-Fen” for consistency with other work related to this research site.

RC2 Comment 7

Page 3, Lines 37: (i) I am not a specialist for agricultural manures, but my understanding is that manure usually refers to animal feces (with the N mostly in form of urea) so I am confused by the ammonium N and referring to the Raiffeisen Laborservice; Do you mean inorganic fertiliser e.g. pure Ammonium sulfate? In any case, please specify this. (ii) Further, I wonder if it would have been worthwhile to obtain also the bulk N-isotopic composition of the two different kinds of fertilizers/manure. You put a lot of effort into measuring the spatial and temporal distribution of the $\delta^{15}\text{N}$ of soil-extracted nitrate while a value for the manure might be valuable as well for the input signature of $\delta^{15}\text{N}$ of soil NH₄

Authors' response:

- (i) The referee is correct, manure (and not ammonium sulphate) was applied. Raiffeisen Laborservice offers manure analysis (including analysis of N contents). Please see here: <https://www.raiffeisen-laborservice.de/biogas/analysen-guelle>.
- (ii) We agree that it would have been beneficial to get the manure analysed for the $\delta^{15}\text{N}$ content, especially for the interpretation of the event around 12 July. Unfortunately, we did not sample the manure that was applied at De-Fen for subsequent analysis.

RC2 Comment 8

Page 4, Line 14: This measurement-specific information seems not necessary here (“While. . .) and could be deleted.

Authors' response: we agree. The sentence "While NH_4^+ was converted to an indophenol complex, NO_3^- was reduced to nitrite to produce the diazo chromophore." was deleted.

RC2 Comment 9

Page 5, Line 2: sentence could be shortened: “Then the gas was dried using a Nafion dryer. . .). . .also, delete: overpressured (the 4.5 bar already indicate that)

Authors' response: The sentence was adapted as follows "Then the sample gas was dried using a nafion drier [...]". We would like to keep the term "sample gas" in order to emphasize the gas type. The term "overpressure", however, was deleted.

RC2 Comment 10

Page 5, Line 36: Given the complexity of the pathways, this correlation criterion is not a sound argument for a valid measurement as it discards 18 out of 30 values while accepting 12 only leading to a bias in the results.

Authors' response: The main reason for discarding many Keeling plot derived source signatures, e.g. in the period 10 June to 21 June, was the low accumulation of N_2O . However, since both reviewers commented on the selection criteria for valid source signatures, we adopted the criteria as stated in our comment 2 to reviewer 1.

RC2 Comment 11

Page 8, Line 32. You could end the sentence after the Toyoda citation and delete after “, but. . .” as this does not add much.

Authors' response: We would like to keep this part of the sentence to highlight differences between agricultural and suburban sites.

RC2 Comment 12

On page 8, line 35 you write: “At night, within a stable nocturnal boundary layer, vertical wind speeds and hence tracer transport are low, while lateral wind speeds can be high and constituents like N_2O can be transported over larger distances. As a result, N_2O emissions from other land uses or land cover may have contributed to the observed N_2O isotopic composition. To assess the possible influence of other land use / land cover. Please omit may and possible in these occasions where you actually know that more distant emission contribute.

Authors' response: We agree, done.

RC2 Comment 13

- (i) Table 1: unit for bulk density is not % but rather g/cm^3 ; pH is dimensionless
- (ii) Table: 2: to prevent confusion with the units, provide all values for the mole fraction in ppm, i.e. for T 0.329 ppm.
- (iii) Table 3: Event no (a.u)? did not get that for the three columns with SPKeeling and $\delta^{15}\text{N}$ and $\delta^{18}\text{O}$: one digit seems enough for the +- values, i.e. 1.9 instead of 1.91 for SP.

- (iv) Table 4: caption: better: Characterization of the lower and upper range for. . . first column header “Source signature” should read parameter or signature

Authors' response:

- (i) Done
- (ii) Done
- (iii) 'Event no. (a.u.)' was changed to 'Event number' and the number of digits was reduced to one as suggested
- (iv) We agree, done

RC2 Comment 14

- (i) Figure 3: Note, the x axis label for Fig. 3 and 4 and 5, 6 are all different (Date vs Datum in). Please select one for all, e.g. Date in 2016.
- (ii) panel c with precipitation: 80 mm per hour seems a very high value, please check.
- (iii) please rewrite sentence to omit, respectively: “Blue and red dashed lines refer to a cutting event and to a manure application, respectively.” to: The blue dashed line indicates a cutting event and the red line manure application. (similar as you write in caption Fig. 4).

Authors' response:

- (i) Done, 'Date in 2016 (dd.mm.)' was chosen for all
- (ii) Done
- (iii) Done

RC2 Comment 15

- (i) Figure 4: please explain the values given on the right side of the histograms
- (ii) zoom panel a: at around 20.7. there is a weird magenta dot within the background values (black dot)
- (iii) caption: please replace y-axis with axes (plural) general observation at Fig. 4: In Fig. 3 it becomes apparent that the heavy precipitation events (around 12.7. and 22.7.) that lead to a progressive reduction of the WFPS are strongly connected with two prominent N₂O fluxes.
- (iv) While the first heavy rain event (around 12.7.) is connected with the manure application, the second rainfall event happens without manure application. It would be worthwhile to add these heavy rainfall events also in Fig. 4 with lines or other markers.

Authors' response: We agree and we will make the following changes:

- (i) Done
- (ii) Done.
- (iii) Done.
- (iv) The precipitation mentioned by the reviewer occurred on 23 July. As the scaling of Fig. 3 and 4 are different, the rainfall event occurs at the end of the N₂O isotope measurements and did not show up in the results. We therefore prefer to not add a vertical line on the last day of this plot, because possible consequences of this precipitation are not seen in the figure due to missing data in the next days.

RC2 Comment 16

Figure 7: If possible adjust the colour legend to rounded numbers rather than $3.16e+02$, e.g. 0.5; 1; 5; . . . 300. Also, if possible, add the numbers (15, 30, 45 %) of the source sensitivities onto the isolines of figure itself (this is quicker than having this written in the caption).

Authors' response: the numbers in the legend of the Figure 7 are now written out.

Please also note the supplement to this comment:

<https://www.biogeosciences-discuss.net/bg-2018-426/bg-2018-426-RC2-supplement.pdf>

References

- Liu, X., Qi, Y., Dong, Y., Peng, Q., He, Y., Sun, L., Jia, J., and Cao, C. (2014). Response of soil N₂O emissions to precipitation pulses under different nitrogen availabilities in a semiarid temperate steppe of Inner Mongolia, China. *Journal of Arid Land*, 6(4), 410-422. doi:10.1007/s40333-013-0211-x
- Peng, Q., Qi, Y., Dong, Y., Xiao, S., and He, Y. (2011). Soil nitrous oxide emissions from a typical semiarid temperate steppe in inner Mongolia: effects of mineral nitrogen fertilizer levels and forms. *Plant and Soil*, 342(1), 345-357. doi:10.1007/s11104-010-0699-1