

Referee 1

Authors have improved the manuscript after this revision, solving most of the weak points of the initial MS. Now the arguments done in the MS are easier to follow by readers. The paper has novelty and interest from a mechanistic point of view, especially due to the information obtained about the isotopic signal of N₂O emitted under denitrification conditions. Results confirmed that there were 2 different N pools in soil with different dynamic. Therefore, I recommend its publication in the current form.

R: We thank the reviewer for the comments that made the manuscript greatly improved.

Referee 2:

Nitrous oxide is a powerful greenhouse gas, the pathway of N₂O production is important to model and mitigate N₂O emissions, which would be changed due to differ in moisture contents. The authors seek to evaluate the roles of soil water filled pore space and nitrous oxide emission and its pathway. They established a laboratory incubation experiment in a relatively narrow range of moisture (70-100%), emissions of N₂O, N₂ and CO₂, as well as the isotopocules of N₂O, were measured. The authors found that denitrification as the main source of fluxes at the highest saturations, but nitrification could have occurred at the lower saturation, even though moisture was still high (71% WFSP). The results indicated that there are 2 N-pools with different dynamics: added N producing intense denitrification, vs soil N resulting in less isotopic fractionation. Lots of studies have reported the relationship between the moisture contents and denitrification and N₂O and N₂ emission, and the effects of spatial heterogeneity and source processes on GHG. Whilst, the study conducted in a relatively narrow range of moisture is rare. These results could contribute to model and mitigate GHG emissions from agricultural soils. Additionally, the authors have revised the paper according the first 3 reviewers, which improved the paper a lot. In my opinion, this is an interesting paper, although it has some small problems should be revised. Thus, it can be accepted after minor revision. My suggestions were shown as follows.

Line 121 and 22, the authors hypothesized that, even at high soil moisture, a mixture of nitrification and denitrification can occur. However, the authors haven't explain the base of this hypothesis.

R: we base this hypothesis on the fact that pockets of aerobicity as well of anaerobicity can occur at high soil moisture, mainly driven by soil respiration after application of N and C (using up O₂) and further recovery after nutrients are used becoming limiting (increasing aeration). Text has been added to explain this.

Line 183, the incubation experiment lasted 13 days. The detail of incubation term should be described, "one day before amendment application and 12 days after ...". Otherwise, the readers would be puzzled.

R: text amended as recommended

Line 268, in "Results" section, the results were described too much in details. In my view, the results which supported the conclusions could be described in details, whilst, the other results may be described in brief. In fact, it is difficult for the reads to remember so much information and to find the important information for the conclusions.

R: most of the details provided are the result of recommendations of previous reviews (5 in total before these three). For example:

Referee Lines 349 to 351: The results showed that the total N emission (N₂O+N₂) (Table 3) had a consistent decreasing trend, with decreasing soil moisture i.e. from 63.4 for SAT/sat (100% WFPS) to 34.1 kg N ha⁻¹ (71%

WFPS) for UNSAT/halfsat. I don't see a consistent decreasing trend. Only the driest treatment shows a lower emission.

Response: we have modified the text to reflect this properly: 'The results showed that the total N emission (N₂O+N₂) (Table 3) decreased between the highest and the lowest soil moistures i.e. from 63.4 for SAT/sat (100% WFPS) to 34.1 kg N ha⁻¹ (71% WFPS) for UNSAT/halfsat'

Also:

Referee: Line 367-369: The writing is confusing here; I cannot follow the meaning of this sentence.'

Response: These are the lines in the submitted pdf: "This is in juxtaposition with the situation when the N emissions are dominated by N₂ or N₂O is low, where the SP values of soil emitted N₂O were much higher (Fig. 3), pointing to an overall product ratio related to an 'isotopic shift' of 10 to 12.5‰."

We modified to (including previous sentence):

"The plot of the N₂O / (N₂O + N₂) ratio vs SP for all treatments in the first two days (when N₂O was increasing and the N₂O / (N₂O + N₂) ratio decreasing) shows a significant negative response of the SP when the ratio increased (Fig. 3). The regression suggests that when the emitted gaseous N is dominated by N₂O (ratio close to 1) the SP values will be slightly negative with values around -2 (Fig. 3), i.e. within the range SP range of bacterial denitrification. With decreasing N₂O / (N₂O + N₂) ratio the SP values of soil emitted N₂O were increasing to values up to 8 per mil."

Line 364 to 368, in "Results" just showing the results of the experiment. Thus, those sentences may be moved to "Discussion" section.

R: sentences have been moved as recommended

Line 385 to 389, move those sentences to Fig. 4 as a footnote.

R: text has been moved

Line 402, "2-pool dynamics" should be described.

R: this reflects the competition for soil N vs applied N as described in Lewicka-Szczebak et al. (2015). Reference is already mentioned in the paragraph, and we had said: 'Pool 1 representing amended soil....and added 'whereas for Pool 2 (amendment-free soil)...'

Line 434, the topic of different paragraphs might be used as the subtitle in "Discussion" section, which might be easy for readers to understand and to follow authors' thought. For example, in 4.1, "Nitrogen emission increasing with moisture contents" may be used as the subtitle of this section.

R: sub headings were added to help clarity

Fig. 1, take the figure of N₂O, N₂ or CO₂ together, and then they would be easily compared.

R: we have joined N₂O and N₂, left CO₂ separate as the figure becomes too busy and difficult to see.

Fig. 4, the icon of Fig. 4 should be described.

R: we have added text in the legend to describe the periods. We are not sure if the reviewer means something else.

Fig. 5, "BD", "ND" and "FD" should be described.

R: these are already explained in the legend: 'Endmember areas for nitrification, N; bacterial denitrification, D; fungal denitrification, FD and nitrifier denitrification, ND'.

Referee 3

The authors should emphasise the novelty of the combination of approaches used, i.e.

i) that the ms addresses the relationship between %WFPS and DIRECT measurement of both N₂O and N₂. (It does not solely focus on the well documented relationship between N₂O fluxes and WFPS), nor does it use the acetylene inhibition technique which has its own deficiencies - especially in fine textured soils and high soil moisture contents,

R: text re. moisture and acetylene was added in the introduction: 'They mostly rely on the knowledge of the effect of moisture on soil processes, whilst in our study, we combined direct measurements of both N₂O and N₂ with isotopomers of N₂O to verify the source processes.'

N₂ emissions were based on direct measurements from the incubated soils, avoiding methodologies that rely on inhibitors such as acetylene with limitations in diffusion in soil and causing the oxidation of NO (Nadeem et al., 2013).'

ii) it combines the measurement approach with N₂O isotopomer measurements to verify both the pool of N emitted as N₂O, and verify the dominant soil microbial processes responsible for the N₂O and N₂ fluxes, and

R: text has been added in the conclusions: 'This study combined direct measurements of N₂ as indicator of denitrification with isotopomers providing a measurement approach that verifies the source processes of N₂O emissions.'

ii) it uses a precise means of compacting the soil and generating the initial %WFPS treatments.

R: text was added in the introduction to enhance the importance of this: 'In addition, the packing of the cores in our study was of great precision increasing our potential to achieve reproducibility in the replicates where a mixture of aerobic/anaerobic pores might have occurred.'

The authors should also expand the discussion to address the assumptions made about the distribution of the added glucose and NO₃ within the soil profiles of the different treatments and subsequent interpretation of the results. I do not see this as a reason to prevent publication - but I do think that the authors could comment on this in the discussion a little more. It's a shame that this could not have been verified in a parallel set of soil cores using labelled C and N. Can the authors also comment on whether there was any leaching/loss of the glucose and nitrate additions from the soil 'cores'.

R: thanks to the reviewer for these useful comments.

We were not able to simultaneous labelling and emissions measurements of N₂O and N₂ at the time of this study, in fact we are not aware of any study which have ever done this before, but this is now a facility we have developed. We have however in the recent past used labelling techniques to help us determine the contribution of the added pool to the emissions. For example in Loick et al. (2016) we estimated that the proportion of N₂O emitted that derived from the added pool of N and C was ca. 85% under denitrifying conditions. However, this does not give us direct information of the amendment distribution. Also in Loick et al. (2016), we undertook preliminary tests to determine infiltration of the amendment using dyes, but it was difficult to be certain on where the amendment went.

However, in previous studies we assumed (for modelling purposes) multiple pools after N and glucose amendment. In Bergstermann et al. (2011) for example we presumed they occupied 10% of the core volume (pool 1), because this resulted in a good fit for measured and modelled N₂ and N₂O fluxes as well as d¹⁵N^{bulk} values. In the current study, we could assume that in the wettest treatment this (proportional) volume was smaller ie similar to the pore volume displaced by the added 5 ml of amendment since pores were almost completely filled with water. Furthermore, that it would have been the largest in the driest treatment where the amendment solution was able to infiltrate the partly saturated pore space and thereby increasing the water content in the infiltrated volume. With regards to leaching, it was minimal (< 0.5 mL water in the core) and so significant leaching of amendment can thus be excluded.

Text has been added to the manuscript in the discussion.