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Biogeosciences
Discussions

Interactive comment on "Underestimation of denitrification rates from field application of the ^{15}N gas flux method and its correction by gas diffusion modelling" *by* Reinhard Well et al.

Reinhard Well et al.

reinhard.well@thuenen.de

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We are grateful to the Editors for considering our manuscript. We thank the reviewers for their constructive comments (repeated under RC2.x), which revealed several gaps and options for improvement. They will be the basis for substantial improvement of the manuscript. In the following we answer all comments and declare how we will change the manuscript to fulfill the comments.

Anonymous Referee #2 Received and published: 30 January 2019 Overall the paper is quite relevant to researchers who have used or are planning to use the 15N labelling method to quantify identification rates in-situ. The researchers convincingly

show through modelling and field data that the impact of subsoil diffusion and storage fluxes have a significant impact on the estimated denitrification rates and thus have likely caused under reporting in the current literature.

General comments

R2.1 - My main reservation is regarding the applicability of the modelling more broadly for correction of field results. Parameters like diffusivity are notoriously difficult to estimate in the field, and therefore the discrepancy between model and measured as reported here may never be reconcilable.

AC2.1 Response: This was also addressed by reviewer 1 (RC1.1). We agree that exact prediction of subsoil diffusion is difficult, but determination of Ds in the subsoil is feasible with reasonable effort and can also be modelled based on bulk density and soil moisture. This will lead to Ds estimates accurate enough for subsoil diffusion modeling (see als AC2.6 below).

Changes: In the results we will add additional scenarios to illustrate the impact from uncertain estimates of diffusivity. In the discussion we will add a section to explain that (i) taking into account maximum uncertainty in subsoil diffusion modelling we demonstrate that worst case scenarios would still improve estimates compared to previous practice and (ii) that small scale heterogeneity in Ds had little or moderate effect on simulate subsoil diffusion. . ..". Moreover, we will supply references highlighting the accuracy of Ds measurement and modelling.

R2.2 - While i appreciate the difficulty of including water phase gas transport in a model, especially one with such a complicated isotopologue structure, I feel it should at least be discussed in the paper as another important factor. It would both contribute to pore space storage as well as isotopic fractionation although the latter may not be important given the label strength. AC2.2 Response: We agree, especially in case of N2O due its high solubility in water, there would be some storage which would have some impact on the change of fluxes following chamber closing due to the slow diffusion in the

water phase. This should be tested in follow up studies. Changes: In the discussion we will mention that water phase transport was not yet taken into account, but might have some relevance due low diffusivity in the water phase. We will also mention that this effect would be largest for N2O due to its high solubility in water and will add references were the impact of water phase gas transport is addressed with respect to CO2 flux modelling. We will also mention that water phase dynamics might be another explanation for the deviations between the N2O/(N2+N2O) ratios determined with bottom open and bottom closed.

RC2.3 - The model results are somewhat dense and difficult to digest - my concern is that someone who is not a modeller/gas diffusion specialist would get lost in the current brief narrative. Suggest being more verbose but for the benefit on enhanced clarity. AC2.3 Response / Changes: we are sorry for this. The model results will be explained more detailed.

RC2.4 - Section 3.1.2 requires significantly more explanation. I would have expected to see a more normal flux calculation as a proxy for production as is done with CO2 or CH4, however the fitting approach is applied here. Why did the authors not use a linear or exponential flux model as is commonly used for other gases. What do the parameters alpha and delta signify or what is their physical manifestation - are they related to chamber volume and surface area, cylinder depth, etc? Is this approach/equation commonly applied outside of this paper? AC 2.4 Response / Changes: we are sorry for this. We will supply adequate explanations.

- Discussion and conclusions

RC2.6 - If the modelling approach cannot be applied quite yet to correct the values, perhaps there should be a small table or histogram or similar of "likely errors" that may have been incurred in past experiments using this method. This would at least allow the community to make an educated guess on how far off our current estimates are from reality (and may allow some reconciliation across methods as well). AC 2.6 Re-

sponse: We agree that this is would be useful. We will generate such a table based on the scenarios we already included in the manuscript, and add additional new scenarios Changes: We will add a table showing % underestimation for our micro-plot and chamber geometry in dependence of Ds and chamber deployment time. We will discuss how results would change with differing depth in labelling or size of the micro-plots.

RC2.7 - Overall the flow of the paper could be improved, this is partly due to sections with poor sentence structure or run-on thoughts mostly in the introduction and discussion portion of the paper. AC2.7 Response: We are sorry for this and will evaluate and improve the flow of the paper and check sentence structures. Changes: We will work on flow and sentence structure as requested

Specific Comments

Page 2 RC2.8 - Line 9 - "to measure" should be "in measuring" or similar.

AC2.8 Response/changes: will be done as suggested

RC2.9 - Line 14 - gastight should be gas tight unless this is a brand of container

AC2.9 Response/changes: will be done as suggested

RC2.10 - Line 25 - suggest inserting several sentences explaining to the reader why in-situ measurements are important. Is there literature to cite comparing in-situ to lab incubations or similar?

AC 2-10 Response: Denitrification is complexly controlled by interaction of labile C, abundance and community structure of denitrifiers, pore structure, soil and root respiration, mineral N dynmics. Hence, it is difficult to keep conditions in the lab identical to the field where some conditions dynamically change due to climatic factors but especially due to the activity of plants. Field measurements are therefore needed for reliable determination of denitrification in ecosystems. There are numerous lab studies but few field measurements. To the best of our knowledge respective comparisons are still missing for unsaturated soils. We conducted such a comparison only for den-

itrification in shallow groundwater (Well, R., et al. (2003). "Comparison of field and laboratory measurement of denitrification and N2O production in the saturated zone of hydromorphic soils." Soil Biology & Biochemistry 35(6): 783-799.

Changes: The content of the response will be included in the introduction.

Page 3 RC 2.11 - Line 8 - some more detail around why we don't just measure these parameters instead of modelling them AC 2.11 Response: we are not sure if we understand this question correctly. Line 8 f reads: "Modelling diffusion of 15N2 + 15N2O produced in 15N-lableled surface soil could be used to estimate its accumulation in pore space and diffusive loss to the subsoil and thus to quantify denitrification from the sum of surface flux, subsoil flux and storage within the 15N-labelled soil volume." By modelling diffusion we mean: modelling the diffusive flux. But the diffusive flux can not be measured directly. But unfortunately we failed to mention that this approach would include measurement of surface flux.

Changes: We will reformulate this sentence as: "Modelling diffusive fluxes of 15N2 + 15N2O produced in 15N-labeled surface soil based on measured surface flux and diffusivity could be used to estimate its accumulation in pore space and diffusive loss to the subsoil and thus to quantify denitrification from the sum of surface flux, subsoil flux and storage within the 15N-labelled soil volume. "

RC2.12 - Line 14 - change amount to concentration AC 2.12 Response/changes: will be done as suggested

RC2.13 - Lines 18-29 - consider separating into bullets. AC 2.13 Response/changes: will be done as suggested

RC2.14 -Consider annotating figures with some of the details contained in lines 18-29 AC 2.14 Response: This will be done as suggested: Changes: Modified caption of Fig. 2a,b: Figure 2a: Increase in pore space concentrations of N2 evolved from the 15N-labelled pool after start of denitrification with open chamber when production of

15N–labelled N2 and N2O would start at constant rates, leading to accumulation of 15N-labelled gases and thus to build-up of concentration gradients to the surface and to the subsoil. Concentration trends following chamber closure are shown as dotted lines.

Figure 2b: Time course of relative fluxes of N2 and N2O evolved from the 15N-labelled pool after start of denitrification with open chamber showing increasing surface and subsoil fluxes while the storage flux decreases until steady state is reached. Trends of fluxes following chamber closure are shown as dotted lines.

RC 2.15 Page 6 - Figure 2b - What is the origin of the oscillation in the flux data Page 7

AC 2.15: Response: The oscillation is a numerical artefact, that affected only the simulation of the steady state dynamics. This problem is now solved Changes: We will replace the figure with a the results of the updated model

RC 2.16 Line 1- is the chamber here fully/homogeneously mixed? AC 2.16 Response: Yes, the model assumes a fully mixed chamber. Changes: we will add the information that the model assumes a homogenously mixed chamber

RC2.17 Line 18 - Is the atmosphere multi-layer? This isn't clear AC 2.17 Response /changes: We will explain this in the description of the model

RC2.18: Line 22 (and elsewhere) - NO3 is often used, but are there any chemical or biological processes modelled that convert NO3 to other species? If not then perhaps its best to clarify that gases are produced independent of NO3 transformation. AC2.18 Response: In the methods we explain that we assume a simplified process dynamics where in terms of N transformation only nitrate reduction by denitrification occurs with N2 and N2O as emitted products. Our estimation of subsoil diffusion is based on the assumption that N2 and N2O production stay constant long enough to reach steady state before chamber closure and also do not change during chamber closure. Other

nitrate transformations, e.g., microbial immobilisation, plant uptake or leaching, would only be relevant for our approach if they cause a rapid change in N2 and N2O production so that near steady state fluxes are not reached. To exclude a discussion on the potential impact of numerous pathways of nitrate transformations we will clarify in the methods that we address N2 and N2O production from 15N labelled nitrate pool via canonical (i.e. heterorophic bacterial) denitrification and that we assume relative constant rates so that near steady state is established. In the discussion we will briefly mention other nitrate pathways and possible impact on our results. Changes: We will add statements mentioned above in the methods and discussion sections.

Page 8

RC2.19 Line 10 - Is production constant with depth over the length of the collar? Page 11

AC 2.19 Response: yes Changes: we will add that production rates are assumed constant over the length of the cylinder

RC 2.20 - Line 11 - Clarify that these initial results are from the bottom open scenario

AC 2.20 Response: We are sorry that we did not make this clear enough Changes: We will update Table 1 to include all scenarios used in the manuscript. Moreover we will refer to these scenarios in the caption of all figures and tables were scenarios results are shown.

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