

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

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We thank the reviewers for their constructive comments (repeated under RC1.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.

Interactive comment on “Underestimation of denitrification rates from field application of the  $^{15}\text{N}$  gas flux method and its correction by gas diffusion modelling” by Reinhard Well et al. Anonymous Referee #1 Received and published: 20 December 2018 Review of Biogeosciences [Manuscript #2018-495] Title: Underestimation of denitrifi-

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cation rates from field application of the  $^{15}\text{N}$  gas flux method and its correction by gas diffusion modelling. Dear Associate Editor, The manuscript in consideration sought to elucidate whether the field application of the  $^{15}\text{N}$  gas flux method underestimates denitrification rates and evaluate the possible reasons by using soil diffusion modelling. The topic is of particular interest to terrestrial biogeochemists attempting to constrain N cycling processes since denitrification is notoriously difficult to measure under field conditions. The authors significantly advance knowledge in this field of research by providing a first proof of denitrification rates underestimation due to subsoil diffusion and storage of denitrification products as stipulated previously by research published in this journal. RC1.1 Even though the authors provide strong indications that subsoil diffusion is indeed occurring during field application of the  $^{15}\text{N}$  gas flux method I am not convinced of the practical applicability of soil diffusion modelling for correcting this discrepancy. The significant difference between measured and modelled results suggests there is too many unknown factors (e.g. spatial variability of diffusivity) and that further assumptions (beyond the homogenous soil labelling of the  $^{15}\text{N}$  gas flux method) need to be introduced to model surface flux and subsoil diffusion and storage. I am wondering if the soil diffusivity assumptions (homogenous soil pore structure and water content, absence of stones, roots etc and constancy of diffusion and production rates) are actually introducing more bias than practical improvements to the traditional chamber method (e.g. depth of labelling, size of chamber, open or closed bottom and closure time).

AC1.1 Response: We agree that modelling of subsoil fluxes is associated with a variety of uncertainties which we explained in P 19 L 20-31: "The general agreement between measured and modelled increase in surface flux after closing the cylinder bottom is a first proof of our concept to quantify denitrification rates using surface fluxes and modelling. Reasons for the observed deviations between experimental and model results can be manifold, e.g., imperfect estimate of  $D_s$  by the empirical model (Millington & Quirk), spatial variability of diffusivity (Kuhne et al., 2012; Lange et al., 2009; Maier et al., 2017; Maier and Schack-Kirchner, 2014; Marrero, 1972) within the 10 cm layers

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for which  $D_s$  was determined, spatial variability of denitrification rates (Groffman et al., 2009), production of  $^{15}\text{N}_2$  and  $^{15}\text{N}_2\text{O}$  from possibly leached  $^{15}\text{NO}_3^-$  below the confined soil cores, and a possible shift in denitrification rates during the 6 hours between the two experiments with bottom open and bottom closed. A quantitative evaluation of the model by  $^{15}\text{N}$  gas flux experiments would be quite challenging since it would mean to assess all aforementioned uncertain factors and to include heterogeneity in the modelling. Future attempts are therefore necessary to improve model evaluation and check how our approach will perform under heterogenic conditions. But despite these uncertainties, the general agreement of model and measurements shows that our approach leads to improved denitrification estimates. “ But even uncertain estimates of subsoil fluxes would improve the outcome of the  $^{15}\text{NGF}$  in comparison with current practice (i.e. without taking subsoil diffusion and storage into account) as it would lower the bias in estimating denitrification rates. We can exclude that our approach could increase bias in view of the limited effect by varying diffusivity on relative subsoil diffusion and storage flux (see also AC 2.6 below). Severe overestimation of these quantities could only occur at high soil gas diffusivity, that means in dry highly porous soils . But these conditions are not relevant for our approach since denitrification is inhibited at high diffusivity.

Changes: We will address the points above in the extended discussion.

RC1.2 The authors suggest that previously published data could be corrected for underestimation by using their model with further parameterisation. This indeed would be something I would be very interested to see and particularly for more challenging soil types than arable land such as grasslands or forests. AC1.2 Response: Thank you for this suggestion. We plan to do this in follow-up studies Changes: In the conclusions we will mention that follow-up studies are needed to obtain further model parametrization to enable correction of previous published in situ  $\text{N}_2+\text{N}_2\text{O}$  fluxes covering all land use types

The manuscript is well structured and clearly written and it seems to me it is a first step

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towards the right direction for further improving field denitrification measurements. I therefore recommend that the manuscript is accepted for publication following a few minor corrections and clarifications detailed below: Minor comments: RC1.3 1. P1 Lines 18: End the sentence after total production and start a new one after it.

AC1.3 Response/Changes: will be done as suggested

RC1.4 2. P3 L9 and throughout: Please correct spelling of the word labelled throughout the manuscript. AC 1.4 Response/Changes: will be done as suggested

RC1.5 3. P3 L25&26: In Sgouridis et al. 2016 the labelled nitrate was applied via injections to the soil volume. Please correct the reference and replace with one that surface application was used. AC1.5 Response/Changes: will be done as suggested

RC1.6 4. P4 L16: Was steady state within the first 6 hours after the label application also measured or just modelled? In the next sentence the assumption stated is that gas production starts at constant rates after the label application. Is it therefore necessary to first establish steady state before applying the model? AC1.5 Response: Indeed the model can only yield correct values if fluxes are steady state at chamber closing. Hence, surface flux data collected immediately after labelling where activity and fluxes dramatically change over time could not be corrected exactly. Moreover, we did not determine steady state experimentally. However, measurements were conducted 5 days after labelling (section 2.3). Therefore, since this is 20 times the modelled steady state time, we can expect that near steady state was reached, even though we could not check this. But we realize that incomplete steady state, due to changes in activity and/or diffusivity, e.g., following precipitation, could be an issue. We will include this in the discussion. Changes: In the discussion we will add a statement that further uncertainty could arise from incomplete steady state, e.g., following precipitation and thus decreasing diffusivity, increasing moisture and change in the labelled volume, and that this effect should be evaluated in follow-up studies. RC1.6. P10 L22: Reference is repeated twice. AC1.6 Response/Changes: this will be corrected

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RC1.7 P11 L26: It would have been useful if measurements with or without closed bottom cylinder and varying labelling depths and lengths of cylinders were also taken. This could have shown whether the model predictions are true and if there is a significant difference in surface fluxes to justify the use of the model. Perhaps a combination of lower labelling depth, deeper cylinder and larger chamber would result in insignificant subsoil diffusion losses. AC1.7 Response: This is indeed planned for a follow up study Changes: In the discussion we will explain that the present study only compared constant depth of labelling and that the applicability of our approach for varying depth of labelling should be checked by future studies.

RC1.8 P19 L2: I agree that it would be a lot easier to apply the model under laboratory closed system conditions. However, pore space/headspace equilibration is relatively easier to achieve than attempting the soil diffusion modelling. The real challenge for the future application of the model would be to apply it under field conditions in more challenging soil types.

AC1.8 Response: We agree. Closed system was mentioned because there is also some effect that had not been taken into account until now. The challenge for the future application of the model would be to apply it under field conditions in more challenging soil types as was already addressed in the following section in page 19, where we discuss the potential and limitation of our approach for field studies. Changes: In the discussion we will deepen the discussion on future application of the model to apply it under field conditions in more challenging soil types.

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