



Interactive comment on “Versatile soil gas concentration and isotope monitoring: optimization and integration of novel soil gas probes with online trace gas detection” by Juliana Gil-Loaiza et al.

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Received and published: 2 April 2021

Dear Dr. Nicolas Brüggemann,

We appreciate the constructive comments from reviewer 1 to improve the manuscript. This paper is the first of many steps to overcome the challenges of sample transfer on an online and real-time soil trace gas sampling system in soils. It highlights the need for innovating in soil probes, instrumentation, and sampling systems to detect soil trace gases in the subsurface with high sampling resolution.

C1

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Following, we will address each of the reviewer's comments and suggestions, those that required edits to the manuscripts will be addressed and will be noted in each response if pertinent for the editors' evaluation.

RC1: The importance of storage of gas in the air filled soil porosity on the link between soil concentration and net surface fluxes, and the dynamics of the air filled soil porosity, at short term (change in water content), long term (e.g. compaction), and also its spatial variation (e.g. local variation in bulk density), are not enough clearly stated and can be highlighted in the first paragraph of the introduction.

Answer: We agree with the reviewer's comment that the mentioned soil parameters are important in the soil trace gas dynamic. Based on the suggestion, we will modify the paragraph by adding the following (Line TBD): "Soil physical parameters play an important role in soil trace gas dynamics, where air-filled porosity influences soil trace gas concentrations and surface fluxes. Heterogeneity in soil structure and bulk density, changes in environmental conditions that alter soil water content (precipitation and temperature), and disturbances like soil compaction will change gas diffusivity and soil gas storage (Fletcher et al. 2007, Fujikawa and Miyazaki, 2005)."

Adding to bibliography: Fujikawa, Tomonori, and Tsuyoshi Miyazaki. "Effects of bulk density and soil type on the gas diffusion coefficient in repacked and undisturbed soils." *Soil Science* 170.11 (2005): 892-901

RC1: Tracing the fate of labeled gas in soil profile is also another promising approach that can be achieved by the development of such integrated monitoring tools. This can be mentioned in the second paragraph of the introduction.

Answer: Thanks to the reviewer for the suggestion and we agree that labeled stable isotopes and other gaseous tracers are interesting and promising tools that could be used with this integrated system to monitor and quantify consumption and production in soil.

Following the reviewer suggestion, we will add the following phrase to the second paragraph in the introduction paragraph to read as (Line TBD): "Previous studies used labeled ^{15}N - N_2O gas to determine consumption and production of N_2O in soil columns (Clough et al., 2006) and inert helium gas as a tracer to quantify in-situ real-time measurement of gas transport in the soil at different depths (Laemmel et al., 2017). Sub-surface isotopic tracer approaches will benefit from the development of an improved integrated systems that allow online quantification of trace gases in the soil as demonstrated by Laemmel et al., 2017."

Citations that would be included: Clough, T. J., et al. "Diffusion of ^{15}N -labelled N_2O into soil columns: a promising method to examine the fate of N_2O in subsoils." *Soil Biology and Biochemistry* 38.6 (2006): 1462-1468.

Laemmel, T., et al. "An in situ method for real time measurement of gas transport in soil." *European Journal of Soil Science* 68.2 (2017): 156-166.

RC1: The carrier gas (Ultra Zero Air) contains O_2 which diffuses to the soil. Is there a risk that microbial processes under study are altered by this change in O_2 concentration in the soil? Would it be better to use pure N_2 instead? Would it be possible to use a closed loop system rather than an open one? Can you discuss your choice for an open-one?

Answer: The reviewer makes good points about the sampling system. In this study, we used UZA as a carrier gas, but we recognize O_2 diffusing from the carrier stream during sampling could alter the soil gas environment if it was not already oxic, and we chose not to use N_2 that might affect the nitrogen isotopes we were measuring. We can address this suggestion by mentioning that the following should be considered in future studies: 1) biogeochemical implications of adding substrates, UZA in this study, to the subsurface and need to test inert carrier gases like He; and 2) criteria for recirculating sampling approaches to outweigh flow-through approaches for a given application. Regarding the flow scheme, we decided to evaluate an open flow-through scheme to

obtain an equilibrated gas sample directly from the probe. The return rate of the multiprobe system allows time for probe equilibration under the appropriate flow schemes before the sample is sent to the analyzer. The probe gas concentration reaches a steady-state value, that we characterize in the paper. To promote diffusive equilibration and also minimize advection across the flow, it is important to maintain constant, controlled, and matched flow rates. In closed-loop sampling, it is more difficult to control return flow without pressure perturbations from the inline pump, but it is feasible. In addition to decreasing cell volume demand by adding an insert, we reduced the TILDAS sample volume demand by adding in-situ dilution, which also helped to reduce concentration dependency and possible condensation in the sample lines. However, dilution in closed-loop systems would prevent the sample from achieving equilibration if diluent is added upon each sample pass, mitigating the inherent value of a closed-loop approach. For analyzers that have low volume demand and limited concentration dependence, we agree that closed-loop sampling is a great approach, e.g., GC analysis (Laemmel et al., 2017, <https://doi.org/10.1111/ejss.12412>), or additionally for sampling with low-cost sensors, e.g., for CO₂ (UMS CO₂ Multichannel Monitoring-System with BGLD-300/BGLD-30 SOIL GAS LANCE described in Jochheim et al, (2018) <https://doi.org/10.1002/jpln.201700259>).

A closed-loop scheme that requires simultaneous, continuous flow through all probes can make the system more complicated, costly, and complex for field deployment. Further, some analyzers (e.g., mass spectrometers) are destructive (PTR-MS ionizes molecules for analysis), preventing the sampling loop from being circulated. However, other soil gas sampling methods (e.g., online GC and low-cost sensors) using a closed-loop system continues to be promising approaches to decrease the impact on gas composition and chemistry during subsurface gas sampling. If the editor considers that these sentences will clarify our choice for an open-loop and highlight the need to continue improving soil gas sampling systems, we will add to the manuscript.

RC1: The control gas that were used for the test using silica filled column looks a bit

low compared to expected soil concentration. Would the results of the test be different with CO₂ or CH₄ concentration of 1% or more?

Answer: Diffusion across the soil probe membrane should be directly proportional to the concentration gradient. With ultra zero air as the carrier gas, and thus an end member, we would expect that higher soil gas concentrations than in the silica tests would move more quickly across the membrane and may equilibrate faster. As we show in Fig. 11, the probe flow rate is a controlling factor on probe equilibration in real soil, both for N₂O and CH₄, and that there is a plateau in equilibration at low flow rates (<20-30 sccm) in both silica and real soil. This is a very good suggestion for future work to test even higher controlled soil gas concentrations in the silica matrix.

RC1: Can you elaborate a bit more and give a definition of “backgrounds” (L257-261)

Answer: We agree with the reviewer, this clarification will increase understanding of the term, we will extend the definition on section “ 2.2.2 Novel laser spectrometer for N₂O and CH₄ isotopomers” to read as follows: “spectral backgrounds were recorded using the instrument auto-backgrounding routine. A sample spectrum is recorded with the instrument sample cell filled with UZA. This spectrum is used to normalize sample spectra, improving accuracy and sensitivity by accounting for changing instrument conditions and possible drift.”

RC1: You confirm that lower probe sampling flow rates allow more time to equilibrate than do high flow rates. Would it be possible to use the initial ‘pulse’ of high gas concentrations at the beginning of the measurement with high flow rates until the steady-state value is reached to recompute the initial concentration in the soil air (peak integration)?

Answer: The reviewer brings up a good point. We have considered integrating the initial pulse of gas concentrations to quantify fully equilibrated samples and overcome the partial equilibration at steady-state. Attempting to do this on flow-through data could be complicated because the full pulse peak is not observed due to the slow TILDAS turnover time, but drops only to the equilibration concentration. We have been

experimenting with flow schemes to transfer only the ‘plug’ of soil gas inside the probe, backed by the UZA carrier. However, evaluating that approach alongside the flow-through approach described here would be out of the scope of an already long paper. Thus, we describe it as a potential next step in Section 4.2.

RC1: Section 3 title is “results and discussion” but section 4 is “discussion”. Should section 3 be “results” only? Sometimes, delta is not well converted in the pdf and become a square instead

We thank the reviewer for the suggestion, we will change the typo error and delete “discussion” from Section 3 to reflect that it is “Results” only. Additionally, we will communicate with the editor to fix the incompatibility of the delta notation.

Interactive comment on Biogeosciences Discuss., <https://doi.org/10.5194/bg-2020-401>, 2020.

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