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Discussions

Interactive comment on "Comparison of flux gradient and chamber techniques to measure soil N$_2$O emissions" *by* Mei Bai et al.

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Received and published: 13 December 2018

Dear Dr. Flechard, Re: Revision of manuscript Number: AMT-2018-90, Title: Comparison of flux gradient and chamber techniques to measure soil N2O emissions

We thank the positive feedback on the manuscript. We have addressed the comments thoroughly, our response to every issue raised is given point by point in blue text below.

The reviewer is aware of the conclusions that should count two aspects: 1) quantitative footprint analysis, 2) recalculate the chamber fluxes using non-linear algorithms.

Authors agree with the reviewer. We conducted a footprint analysis using the Windtrax dispersion model to examine the contribution of area outside our plot to the slant-path FG fluxes. We examined Site 2 (the smaller plot) and considered a wind direction which

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would be the worst-case scenario for a footprint (wind from NE). We also considered the atmospheric stability (1/L) and surface roughness (z0). We estimated the footprint of the slant-path configuration, and what fraction of the surface flux occurred within the plot boundary. Figure A shows the Estimated flux footprint fraction at Site 2 increased from stable to unstable conditions at z0 = 0.1 m. Figure A This result suggests the OP-FTIR flux estimates at night-time could be contaminated by emissions outside the plot (up to 40% contaminated). However, the contamination decreases to < 10% during the day time.

The results are summarised in the revised manuscript, see on page 7-8, Line 166-188 and Figure 2. "The FG flux measurements correspond to surface emissions within a flux "footprint". The footprint generally extends upwind of the concentration sensors, but its spatial size varies with wind conditions. A concern of this study is that the FG footprint extends beyond our plots, and the calculated emission rates are "contaminated" by emissions occurring outside the plot. This possibility was investigated by modelling the FG footprint for our smaller Site 2, where the contamination concerns are greater. The WindTrax dispersion software (thunderbeachscientific.com) was used to simulate the OP-FTIR slant-path setup, and calculate the fraction of the FG measured flux occurring within the Site 2 plot. We looked at the wind direction that results in a short fetch (NE), and looked at different atmospheric stability conditions and roughness lengths. The results for z0 = 0.1 m (representative of the plot) are shown in Figure 2. We concluded that during stable night-time conditions the FG emission calculations for Site 2 maybe contaminated by up to 40% by outside fluxes. This may result in either over- or under-estimation of Site 2 emissions depending on the emission rate outside the plot. In unstable daytime conditions the contamination potential falls to $0 - 10\%$. Contamination at Site 1 will not be as serious due to the larger fetches. The main objective of our study is to compare chamber and FG emission estimates. We looked at periods with concurrent measurements from the two techniques, and hourly flux ratios of QFG/Qchamber measured between 10:00 and 13:00 are compared. Because the comparison took place during the day when conditions were generally unstable,

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the FG contamination potential is low (and will be ignored). The contamination potential does highlight a concern with micrometeorological measurements, that a large measurement footprint may extend outside the study area and result in measurement errors."

2) For our chamber analysis we added an alternative estimation of the rate of dC/dt, using a non-linear monomolecular model (Bolker, 2007). This follows the equation: CN2O = a0 + a1 (1 − exp (−a2/a1 * t)), where CN2O is the mole fraction of N2O, a0 is the intercept corresponding to N2O mole fraction at time t = 0, a1 is the horizontal asymptote at t = +∞, a2 is the slope (dC/dt) at t = 0, and t is time after the chamber is chamber placement time (h). We plotted dC/dt estimated from both the original linear regression and non-linear monomolecular model in Figs. B, C. The chamber fluxes using the non-linear dC/dt estimate is 1.15 times higher than that using linear dC/dt estimate. This improved the geometric mean QFG/Qchamber_non-linear to 1.22 (confidence interval of 0.99 to 1.49). We still concluded that chamber measurements are underestimating the flux, but the use of a non-linear calculation of dC/dt decreases the disagreement to 22% compared to that of 40% using linear regression model. We also added this discussion in the revised manuscript, see page 11-12, Line 269-284. "To examine the potential bias in N2O emissions when dC/dt is estimated with a linear regression model, we also calculated the results using a non-linear monomolecular model (Bolker, 2007). The monomolecular model is one of the simplest saturating function and follows (Eq. 4): CN2O = a0 + a1 (1 − exp (−a2/a1 * t)) (4) where CN2O is the mole fraction of N2O, a0 is the intercept corresponding to the N2O mole fraction at time t = 0, a1 is the horizontal asymptote at t = +∞, a2 is the slope (dC/dt) at t = 0, and t is time after chamber placement (h).

Chamber fluxes calculated using the non-linear dC/dt (Qchamber-non-linear) were 1.15 times higher than Qchamber estimated using linear regression. Comparing the concurrent fluxes of QFG and Qchamber-non-linear, we found the geometric mean of QFG/Qchamber-non-linear to be 1.22 (confidence interval of 0.99 to 1.49). Using

10,000 bootstrap re-samples (Efron and Tibshirani, 1994), we computed 10,000 potential mean fluxes from the non-linear model, of which 9540 of the means were greater than 1, and 460 were lower than 1. This result suggests the use of the non-linear dC/dt calculation has resulted in better agreement with the FG estimates." Figure B

Special comments Title. Agree with the reviewer, and the title has been changed. P5, L109. We calculated the rate of dC/dt using a simple linear regression model. Following the reviewer's suggestion, we calculated the dC/dt using a non-linear monomolecular model. We added couple of paragraphs in the revised manuscript, see page 11-12, Line 269-284. Refer to above text. And Conclusions on page 12, Line 295-297. " Our results showed that soil N2O emissions measured by FG and static chambers (linear dC/dt) were statistically different, with fluxes from FG being on average 40% higher. Using a non-linear calculation of dC/dt in the chambers decreased the disagreement to 22%."

P6, L140. We have provided the precisions on page 6, Line 144-145. "OP-FTIR system measures multiple gas concentrations (N2O, CH4, NH3, CO2, CO and water vapour) with high precision (N2O < 0.3 ppb, CH4 < 2 ppb, NH3, 0.4ppb, CO2, 1 ppm, CO, 0.1 ppb, and water vapour < 5%)." P7, Line 170-171 and Fig.2. The background N2O flux is about 0.6 mg N2OâĂŠN m-2 h-1 prior to manure application. In this study, the topsoil (0−15 cm) had high N contents: 52 mg NH4+−N kg−1 and 118 mg NO3−−N kg−1 before manure application. Therefore, the high background N2O flux is reasonable. P8, Line 192-193. We agree with the reviewer. We made the changes to be "agreement with the FG measurements in terms of the long-term background exchange patterns...". See page 9, Line 222. P10. Line 237. As suggested, we calculated the chamber fluxes using the rate of dC/dt estimated using non-linear monomolecular model. The changes can be seen on page 11-12, Line 269-284, and Conclusions, Line 295-297. P10, Line 247-248. We agree with the reviewer. As mentioned previously, we conducted a footprint analysis using Windtrax dispersion model, and added the results at page 7-8, Line 166-188 and Figure 2. We also removed the statement "In

addition, concerns about FG arise if the fetch…occurring upwind of our sites." P 11, Line 249-250. The reviewer is correct. We have addressed this comment and added the footprint analysis results on page 7-8, Line 166-188. Refer to the above text.

P11. Conclusion, Line 263-265. In the Conclusions, we added a sentence "Our results showed that soil N2O emissions measured by FG and static chambers (linear dC/dt) were statistically different, with fluxes from FG being on average 40% higher. Using a non-linear calculation of dC/dt in the chambers decreased the disagreement to 22%." see page 12, Line 295-297. Our study supports the statement that chamber measurements underestimate the fluxes. See page 13, Line 301-303. "However, the relationship we observed, together with other reports on the biases created by chamber calculation procedures, supports an interpretation that our FG emission calculations were accurate and in this instance the chamber measurements were biased too low."

Technical Corrections P1. Abstract. Agree with the reviewer, the changes have been made. P1, Line 12. Agree. Have added "aerodynamic" and removed "micrometeorological". See page 1, Line 13, and P3, Line 59. P2, Line 42. Agree. "manually operated" has been added to "static chambers", see page 2, Line 43. P2, Line 44. Agree. Change has been made to be" The temporal variation issue can be addressed by…", see page 2, Line 45. P3, Line 59-60. We agree with the reviewer. Change has been made to be" an estimate of the turbulent diffusivity", see page 3, Line 61. P4, Line 86. Agree with the reviewer. The sentence has been changed to be "The average daily minimum and maximum temperature were 6 and 33ïČřC, respectively." See page 4, Line 87. P5, Eq. 1. Agree. We have modified the equation, see page 5, Line 107-113.

"Qchamber = KN2O (273/T) (V/A) dC/dt (1) where Qchamber is the gas flux (μg N2O$-$N m-2 h-1); KN2O is 1.25 (μg N μL-1) according to the ideal gas law, where KN2O = P m/R T0, and P is air pressure (at 1 atm), m is molecular mass (28 g mol-1), R is gas constant (0.0821 L atm K-1 mol-1), and T0 is 273 K; T is the air temperature within the chamber (K); V is the total volume of headspace (L); A is a surface area

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inside the chamber (m2); and dC/dt is the rate of change in mole fraction of N2O in the chamber (μL L-1 h-1) determined by linear regression model. The N2O mole fraction is provide by GC, in ppm." P6, Line 126. Because the path height is a function of x, when "kappa" is integrated with x, there is an implicit integration with z. Therefore, there is no error in the formula and the statement. P7, Line 153-154. Agree. The sentence has been modified as "…at a frequency of 10 Hz." See page 7, Line 160. P7, Line 155. Agree. The sentence has been changed to be "Atmospheric stability parameters of friction velocity (u*), surface roughness (z0) and Obukhov stability length (L) were calculated from the ultrasonic anemometer data." See page 7, Line 162-163. P7, Line 168. We agree with the reviewer. The subtitle has been changed to be "The Flux gradient fluxes". See page 8, Line 197. P9, Line 200. We agree with the reviewer. The subtitle has been changed to "Comparison of the two measurement techniques". See page 10, Line 229. P10, Line 235. We agree with the reviewer. The sentence has been changed to be "Several researchers have reported that chamber flux calculation procedures introduced large uncertainty in N2O emissions flux calculation", see page 11, Line 264.

References Bolker, B., 2007. Ecological Models and Data in R. Princeton University Press, The United States of America. Efron, B. and Tibshirani, R.J., 1994. An intruduction to bookstrap. Chapman&Hall/CRC, Boca Raton, London, New York, Washington, D.C., 456 pp.

Please also note the supplement to this comment:
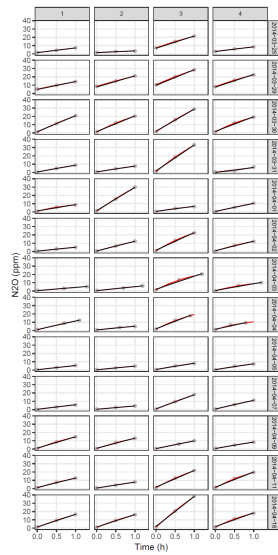https://www.atmos-meas-tech-discuss.net/amt-2018-90/amt-2018-90-AC2-supplement.pdf
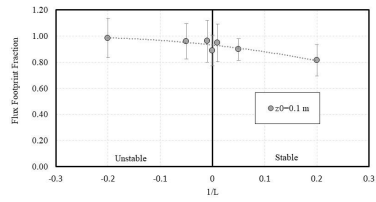
Fig. A Estimated flux footprint fraction at Site 2, plotted versus atmospheric stability (the reciprocal of the Obukhov length L). The model results are for a roughness length $z_0 = 0.1$ m.

Fig. 1.

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Fig. B the N₂O concentration (ppm) changes with time (hour) at site 1. The slope of the regression (dC/dt) using linear regression (black) and non-linear monomolecular model (red).

Fig. 2.

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