Dear reviewer (Anonymous Referee #1),

Re: Revision of manuscript Number: amt-2021-347, Title: Performance of open-path lasers and FTIR spectroscopic systems in agriculture emissions research

We thank your 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.

## Major comments

The introduction lacks the typical emission ratios of CH4, N2O, NH3, etc, in the agricultural fields. The authors use several cylinders to release these gases at known fluxes, but are these fluxes reasonable compared to the real cases?

Emission ratios of  $CH_4$ ,  $N_2O$ ,  $NH_3$  from agricultural fields can be varied. The release rates were point sources, not distributed as cattle or soil would be. The aim is to show that the known fluxes can be retrieved from the measurements, for all three gases. In this case it is permissible to have higher emissions than those typical in the field to minimise uncertainty due to background variability. We have added this in the revised manuscript on page 7, Line 140-142.

It would be very useful to add one section to use the measured VMR from two OP systems to calculate the emission rates. The potential readers are more interested in the fluxes calculation instead of VMR measurements.

Agree with reviewer's comment. we have added a section of flux calculation using backward Lagrangian stochastic dispersion (bLs) modelling (WindTrax) and compared it to the known release rate (recovery study). Please see the section 3.5, 3.6 herd of cattle emission study and 3.7 Windtrax sensitivity in the revised manuscript, page 21-23, the Figures A1-A7 and Table A1 are attached at Appendices. We also added a sentence in the Abstract "The mole fractions measured by OP-FTIR and OPL were also input into models of atmospheric dispersion (WindTrax) allowing the calculation of fluxes. Trace gas release recoveries with Windtrax were examined by comparing the ratio of estimated and known fluxes."

## Minor comments:

Line 31: NH3 is an important atmospheric gas, but it is not GHG

Yes, agree with the reviewer. It is worth to mention in the manuscript that ammonia ( $NH_3$ ) is an indirect greenhouse gas, because  $NH_3$  deposition can ultimately increase landscape emissions of nitrous oxide ( $N_2O$ ), a potent greenhouse gas (GHG). These "secondary offsite" emissions, generally referred to as indirect emissions, are an important component of the GHG budget of the agricultural sector. The change can be seen in the revised manuscript, pages 1-2, Lines 35-36.

Line42: "relies on" -> reply on.

Yes, agree with the reviewer. The change has been made to be "reply on", see page 2, Line 46.

Line 99: add information on why there are two pathlengths for some experiments. Yes, we added more information to explain the different path length. "The different path length was determined depending on the factors of wind conditions (direction and wind speed) and the distance between the path length and source area. Given the constant wind direction, the longer pathlength was needed when the measurement path was further away from the source so that the gas plume could pass by most of the OP measurement path." See page 8, Line 152-155.

Line 107: "and 1 atm pressure" -> and 1 atm. Yes, agree with the reviewer. "the pressure" has been removed in the revised manuscript. See page 5, Line 113.

Line 120: "the measurement path was 137 and 125 m (two-way path) for path 1 and 2, respectively." - > the measurement pathlengths were 137 and 125 m (two-way path) for paths 1 and 2, respectively.

Yes, agree with the reviewer. The change is made to be "above ground level" in the revised manuscript, page 6, Line 126-128.

Line 136: "wind conditions were such poor". Do you have the wind information? The poor wind means the wind direction is opposite? Or the wind speed is too fast? Yes, we agree with the reviewer. We added the information of poor wind conditions (E winds dominated) in the revised manuscript, page 7, Line 145.

Line 120. "above ground level". Other places use above the ground, please be consistent throughout the whole manuscript.

Yes, changes have been made in the revised manuscript.

Line 145-146 (The layout of the experiment is not shown here): Please add it too.

Yes, we agree with reviewer's comment. In the revised manuscript, we have added the layout of gas release trial at Wollongong on 3 August 2005 (Fig. 3), as well the layout of two OP-FTIR comparisons at a commercial feedlot in Victoria in February 2008 (Fig. 4). See pages 8, 9.



Figure 3: Point gas release sources and OP-FTIR path geometries (distances in m) at Wollongong August 2005. The FTIR measurement path lengths at day 1 and 2 were 87.5 and 150 m (two-way path), respectively. Three  $\frac{1}{4}$ " tubes coming from three tanks (CH<sub>4</sub> (nature gas), NH<sub>3</sub> and N<sub>2</sub>O) bundled together on a stake at the release height 1.28 m above ground level.



Figure 4: Two OP-FTIR (Bomem MB100 and Bruker) during side-by-side operation in a commercial feedlot in Victoria in February 2008. Each feedlot pen held approximately 100 beef cattle.

Line 156. First time mentioning CO2 here. It might be better to add some introduction in Section 1.

As suggested by Reviewer 2,  $CO_2$  is not relevant to this study, and we agreed to remove the section that was associated with  $CO_2$ . Please see the change in the revised manuscript.

Line 210: First time mentioning CO here. Same as CO2, it is good to add more information in Section 1. Same as above response, we have removed CO section in the revised manuscript.

Line 209-210: the three spectral ranges are recorded simultaneously or individually with specific optical filters?

The FTIR spectrometer measures the broadband IR spectrum simultaneously over the range 600-5000 cm<sup>-1</sup>. The three separate spectral regions (N<sub>2</sub>O (2130–2283 cm<sup>-1</sup>), CH<sub>4</sub> (2920–3020 cm<sup>-1</sup>), and NH<sub>3</sub> (900–980 cm<sup>-1</sup>)) are extracted from the broadband spectrum and analysed separately for each target species. Please see the changes in the revised manuscript pages 10-11, Line 229-232.

Line 234 : "14:45- 16:30" the time is local time or UTC? The time of measurement period was local time. We added "(local time)" in the revised manuscript, please see page 12, Line 282-283.

Line 235: "From 14:45-15:10" -> Between 14:45 and 15:10. Yes, agree with the reviewer. We made the change in the revised manuscript, please see page 12, Line 283.

Line 310: (data not shown). Please show it. We added the figure of comparing NH<sub>3</sub> mole fractions between two lasers and OP-FTIR (Fig. 9) on page 20.



## Figure 9: Thirty-minute averaged NH<sub>3</sub> mole fraction measured by OP-FTIR and OPL unit (1015) positioned side-by-side (path length = 148 m) at Wollongong site.

Line 322: what is "in stable conditions"?

Monin-Obukov length L,  $L \cong 0-10$  m. we added it in the revised manuscript, page 21, Line 420.

Line 323: "Carbon dioxide results from both FTIRs were lower than those of air samples by approximately 15%." Do you have a reason for that?

We have removed this statement as CO<sub>2</sub> is out of this study.