

Dear reviewer (Anonymous Referee #2),

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.

Reviewer suggested to add a discussion on uncertainty budget and calculate emission rates as a real-case scenarios.

[We agree with the reviewer. The initial manuscript described the measurements of side-by-side FTIR-laser comparisons and demonstrated excellent correlations between measured line-averaged mole fractions. Ratios of mole fractions in most cases were equal to ratios of release rates within 2%. The measurements confirmed the underlying principle that the released gases were equally dispersed by the atmosphere between source and measurement path. Scatter in measured ratios was larger than the measurement precision would imply and suggests that the random nature of atmospheric dispersion is the limiting factor to accuracy in the overall open path techniques, not instrumental measurement precision.](#)

[In the revised manuscript, following reviewer's suggestion, we added three sections:](#)

[1, the trial release experiments at Kyabram in July-August 2005 were also analysed using WindTrax to calculate gas source strengths from measured line average mole fractions for each gas. For uniform area sources, excellent recoveries were consistently obtained for N₂O with Windtrax-calculated mole fractions in agreement with measured mole fractions within 5%. For CH₄ the results were often affected by additional local sources of CH₄ \(local dairy herd\) and for NH₃ there was evidence of losses at the ground surface leading to reduced NH₃ recoveries.](#)

[2, We also added a study on measuring GHG emissions from a herd of grazing cattle \(353 cattle\) using side-by-side OP-FTIR and OPL with sonic anemometer data and WindTrax for dispersion analysis. This experiment assessed the usefulness of the WindTrax approach when the source is not strictly uniformly distributed \(grazing cattle for CH₄, scattered dung and urine patches for NH₃\). Realistic results were obtained \(average 290 g cow⁻¹ day⁻¹ for CH₄, and 0.3-0.8 μg m⁻² s⁻¹ for NH₃\), which was comparable to the results from our previous study. The CH₄ laser results for this trial are in good agreement with the FTIR results.](#)

[3, we added total uncertainty budget:](#)

[We want to compute the total uncertainty associated with the difference in mole fraction between upwind and downwind. There are three uncertainty sources: instrument precision uncertainty, fitting uncertainty, and absorption cross-section \(HITRAN\) uncertainty \(the latter two are fractional uncertainties and were taken from Paton-Walsh et al. 2014\). The measurement precision is in units of ppbv and so the fractional uncertainty that this represents will change with the trace gas mole fraction. The instrument precision uncertainty](#)

(δ) associated with upwind measurement is $1-\sigma$, and the uncertainty associated with downwind is also $1-\sigma$. We assume these errors to be independent. The instrument precision uncertainty in the difference in mole fraction between upwind and downwind is thus $\sqrt{(1-\sigma)^2 + (1-\sigma)^2}$. We then divide this value by the difference in mole fraction to recover the relative uncertainty due to instrument precision: $\sqrt{(1-\sigma)^2 + (1-\sigma)^2} / (\text{CH}_4_{\text{downwind}} - \text{CH}_4_{\text{upwind}})$. $\Delta\text{CH}_4 = \text{CH}_4_{\text{downwind}} - \text{CH}_4_{\text{upwind}}$. We then add in quadrature the relative measurement uncertainty due to instrument precision with the fitting and absorption cross-section uncertainties (also expressed in terms of relative uncertainty). The unit for precision ($1-\sigma$) is ppbv, and % for uncertainty.

For example, for CH_4 , when ΔCH_4 was as low as 20 ppbv, we have a relative uncertainty of 0.28 for the instrument precision, 0.02 for fitting uncertainty, and 0.05 for absorption cross-section uncertainty. The relative uncertainty propagated across these three components is: $\sqrt{0.283^2 + 0.02^2 + 0.05^2} = 0.288$ or 28.8%. In this case, total uncertainty is dominated by the uncertainty due to instrument precision. When the ΔCH_4 was increased to 50 ppbv or 100 ppbv, the uncertainty declined dramatically to 12.5 and 7.8%, respectively. However, for N_2O and NH_3 the uncertainty was not limited by the mixing ratio enhancement but likely attributed to absorption cross-section uncertainty.

Table 9. Total uncertainty budget

	CH_4	N_2O	NH_3
Measurement precision (ppbv)	4	0.3	0.4
Spectral fitting uncertainty (%)	2%	4%	2%
Absorption cross-section uncertainty (%)	5%	5%	5%
$\delta(\Delta \text{ trace gas mole fraction}^\ddagger)/\Delta \text{ trace mole fraction (%)}$			
$\Delta \text{ trace gas mole fraction (ppbv)}$			
20	28.3%	2.1%	2.8%
50	11.3%	0.8%	1.1%
100	5.7%	0.4%	0.6%
Total uncertainty (%)			
$\Delta \text{ trace gas mole fraction (ppbv)}$			
20	28.8%	6.8%	6.1%
50	12.5%	6.5%	5.5%
100	7.8%	6.4%	5.4%

$^\ddagger\Delta \text{ trace gas mole fraction} = (\text{trace gas mole fraction})_{\text{downwind}} - (\text{trace gas mole fraction})_{\text{upwind}}$

Minor comments:

- 1) please show the layout of the experiments at three experimental sights.

Agree with the reviewer. we have added the layout of the experimental site at Wollongong (Fig. 3) and commercial feedlot (Fig. 4).

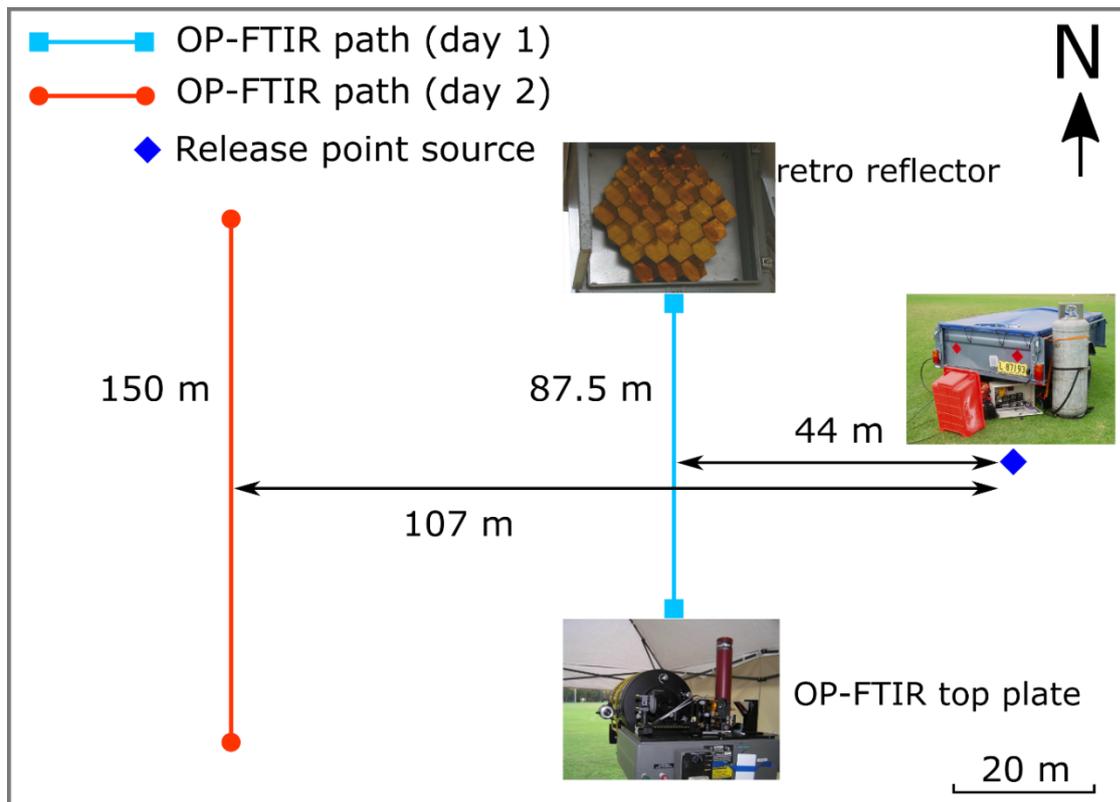


Figure 3: Point gas release sources and OP-FTIR path geometries (distances in m) at Wollongong August 2005. The OP-FTIR measurement path lengths at day 1 and 2 were 87.5 and 150 m (two-way path), respectively. Three ¼" tubes coming from three tanks (CH₄ (nature gas), NH₃ and N₂O) bundled together on a stake at the release height 1.28 m above ground level.

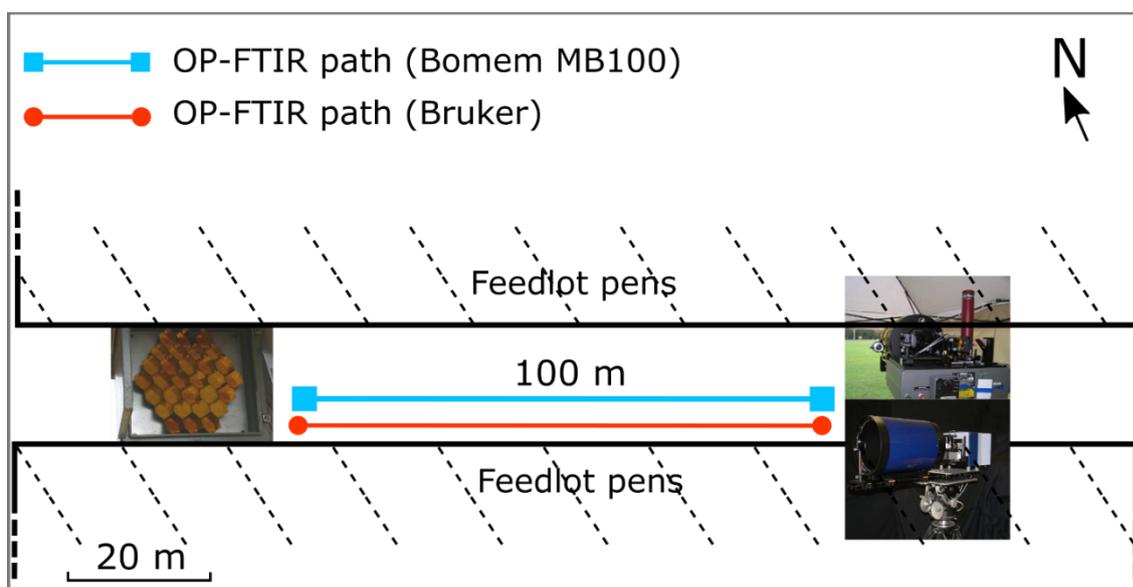


Figure 4: Two OP-FTIR (Bomem MB100 and Bruker) during side-by-side operation in a commercial feedlot in Victoria in February 2008. Each feedlot pen held approximately 100 beef cattle.

2) measurement data of CO and CO₂ is presented in e.g. 2.4.2, however, it is out of the scope of the paper. Please remove or elaborate more.

Agree. we have removed the section about CO and CO₂ in the revised manuscript.

3) L272, not shown data is important for the discussion on the instrument precision, please include in the main text.

Agree. we have added Figure 6 in the revised manuscript.

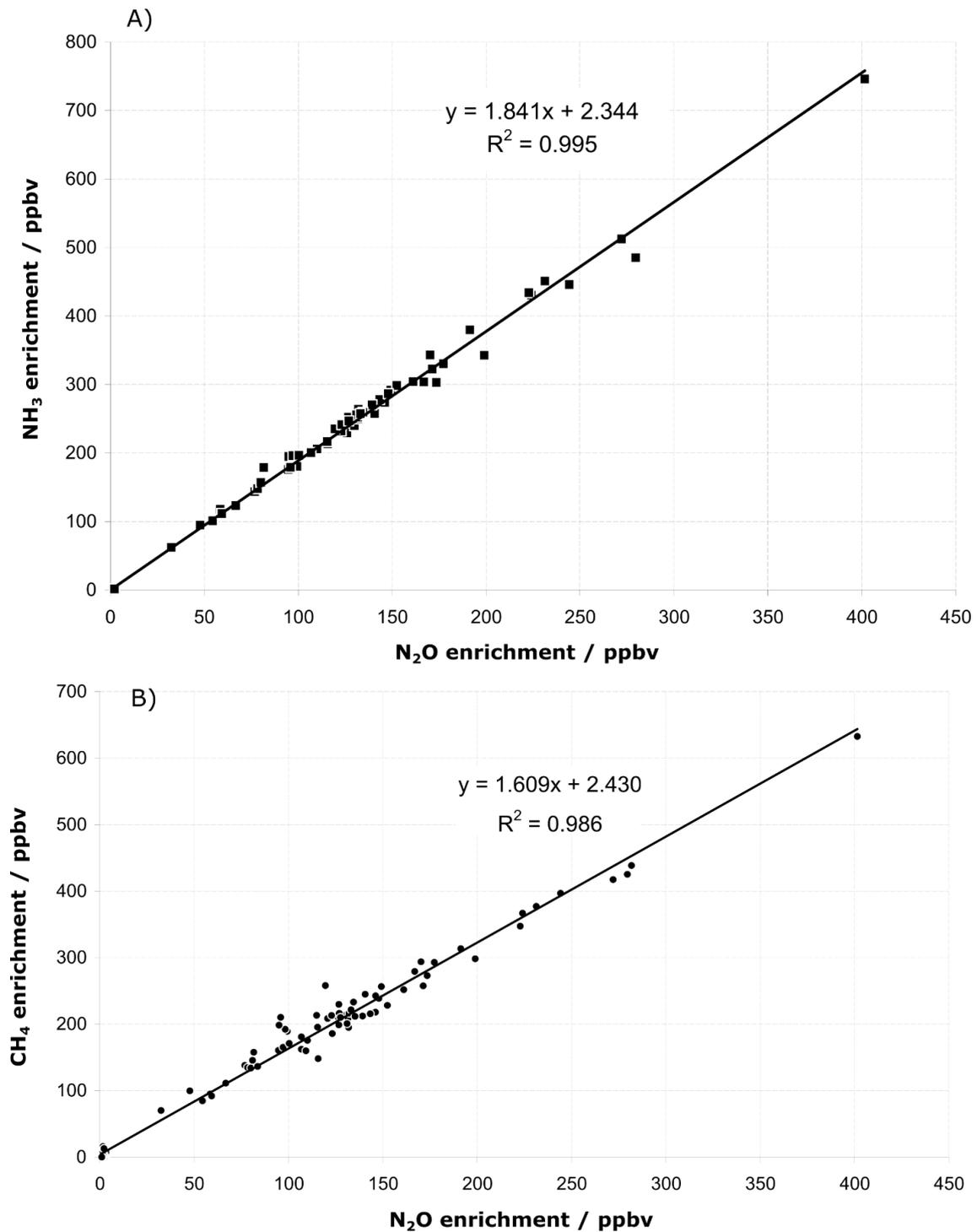


Figure 6: Regression/correlation analysis of the OP-FTIR measured enrichments in Figure 5 between 14:45 and 16:25 of NH₃ vs N₂O (A) and CH₄ vs N₂O (B).

4) L310 (data not shown), data can be included in the supplement.

Agree. we have added Figure 9 in the revised manuscript, please see page 20.

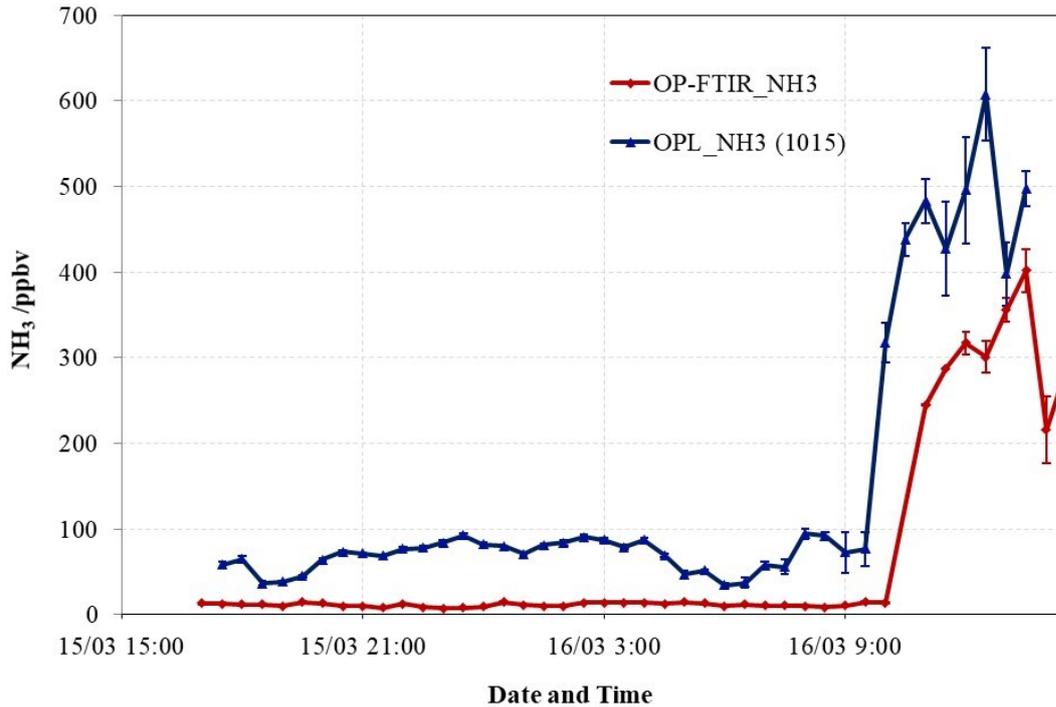


Figure 9: Thirty-minute averaged NH₃ mole fraction measured by OP-FTIR and OPL unit (1015) positioned side-by-side (path length = 148 m) at Wollongong site. Error bars denote the standard error of the thirty-minute means.

5) Figures 3-5, please include the Y - error bars.

We added standard errors (s.e.) to the five-minute averages of CH₄ and NH₃ mole fraction measured at Kyabram (Fig. 7) and s.e. to the thirty-minute mean of CH₄ mole fraction measured by OP-FTIR and OPL at Wollongong (Fig. 8). However, we did not add error bars in Figure 5 as each dot represent a single measurement (i.e., raw data) which makes it impossible to compute the standard error associated with each dot.

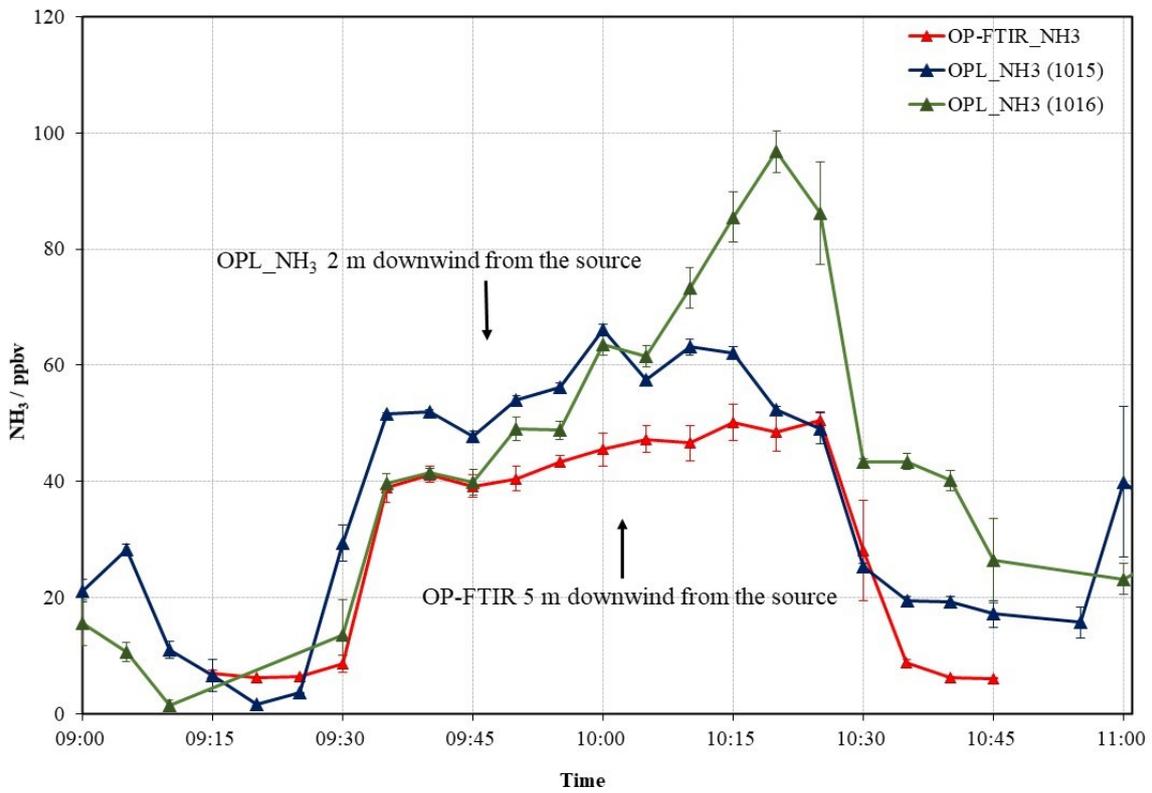
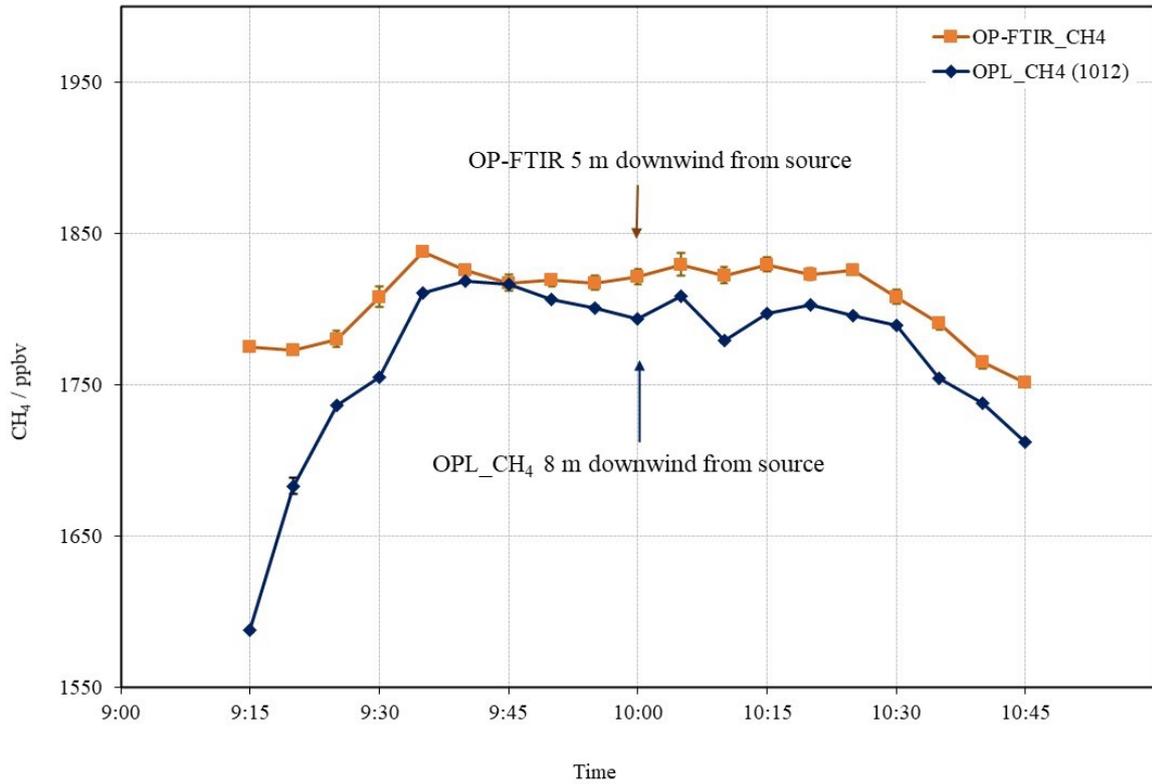


Figure 7: Five-minute averages of CH₄ (upper) and NH₃ (lower) mole fraction measurements from the OP-FTIR and OPL downwind of a ground-level grid source 40 × 15 m wide (path length = 125 m) at Kyabram on 3 August 2005 (T2). Error bars represent the standard error.

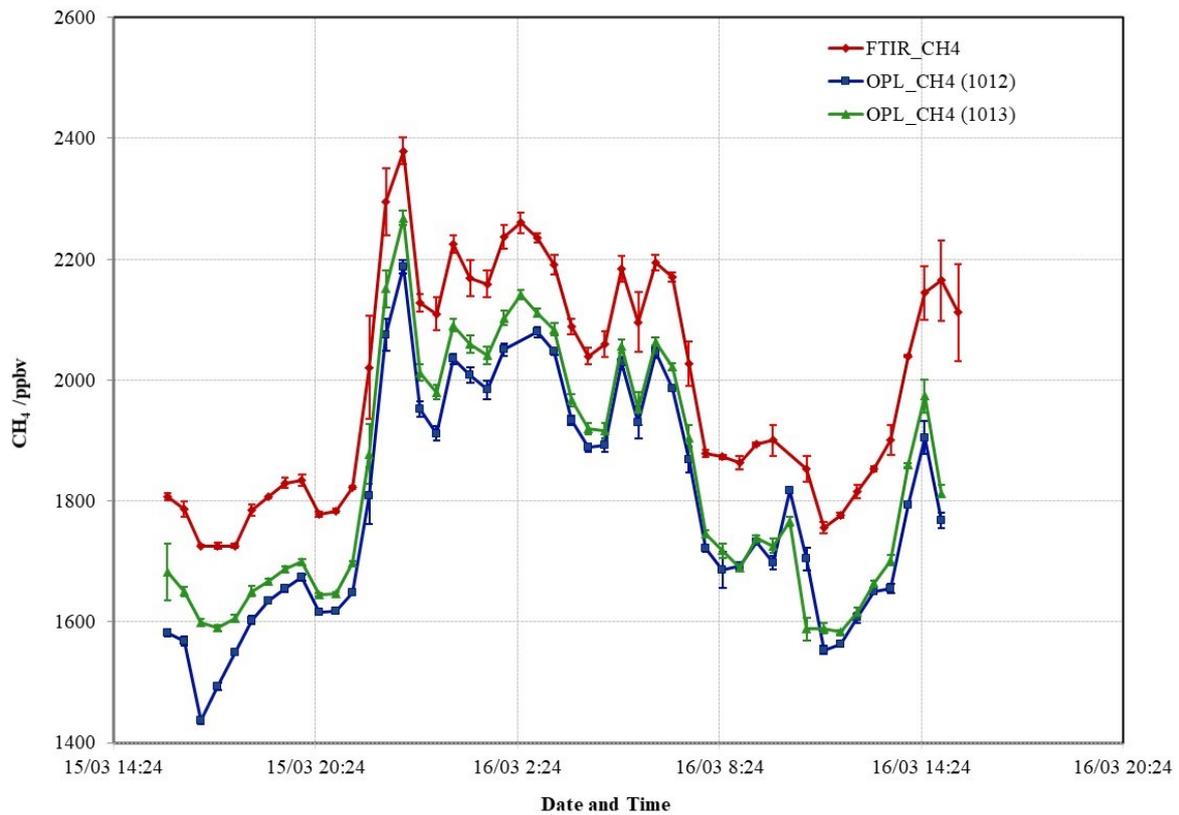


Figure 8: Thirty-minute averages of CH₄ mole fraction measured by OP-FTIR and both OPL units (1012 and 1013) positioned side-by-side (path length = 148 m) at Wollongong site. Error bars denote the standard error.