

Interactive comment on “Long-path measurements of pollutants and micrometeorology over Highway 401 in Toronto” by Yuan You et al.

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1. Line 113-line 116: this needs slight elaboration – the stray light problem should be very minor and only from internal reflections (since the light is modulated before being sent across the open-path. This means ambient scattered radiation will not be modulated and thus not detected by the instrument).

Yes, the stray light influence on concentration retrieval is generally rather minor, but since it is possible to correct for this, we did. These sentences in the main text have been revised as follows: “This stray light spectrum accounts for radiation back to the detector from reflections by internal parts inside the spectrometer, i.e. not from the

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retroreflector array, and was subtracted from all the measurement spectra before performing further analysis. Stray light affected final mixing ratios by < 3 % in this study.”

2. The MALT program (which you say that your analysis is based on) can calculate the reference spectra directly from the HITRAN database using the ambient temperature measured at the same time as the spectra were recorded. Does the Bruker software really not allow this? This may only introduce uncertainties of 10% but it is an unnecessary added uncertainty, since you have accurate temperature measurements available.

This was a mistake in the manuscript; our analysis is based strictly on Bruker software, not MALT. The Bruker software, OPUS_RS, uses a non-linear fitting method. Reference spectra were fitted to the measured spectra using a model to calculate the instrumental line shape (ILS) (Harig et al., 2005) followed by a non-linear curve fitting method to retrieve concentrations of pollutants. In the OPUS_RS software, there is an option of setting up “temperature dependent reference files”. These files include either PNNL (5 °C, 25 °C, 50 °C) or HITRAN files at specific temperatures. Then the program takes the current temperature from a sensor (or temperature data file), and interpolates the high-resolution reference spectrum to the current temperature from those temperature-specific reference spectra which were included in “temperature dependent reference files”.

Harig, R., Rusch, P., Schäfer, K., Flores-Jardines, E.: “Method for on-site determination of the instrument line shape of mobile remote sensing Fourier transform spectrometers”, SPIE 5979, 432-441, 2005.

3. There are other uncertainties inherent to open-path FTIR measurements (like those that come from the HITRAN database and fitting errors). These are not mentioned in the text but should at least be referred to as existing even if a full uncertainty analyses is not given.

Yes, some uncertainties are associated with spectrum fitting errors; the thresholds of the correlation coefficients used in fitting analysis in each pollutant in Table 1 give an

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indication of this. There are also uncertainties dependent on environmental conditions. In this study, we have considered ambient temperature and pressure, as well as H₂O vapor as an interfering gas, in our retrieval analysis. We have stated that for pollutants such as NO and NO₂, water vapor interfered so much that we were not able to get good mixing ratio retrievals. The signal intensity differences related to the distance between spectrometer and retroreflector play a minor role on the detection limits of pollutants studied, since the distance in this study was near the optimal range for this instrument. These detection limits were updated in Table 1 and the text has been revised as follows: “Besides fitting errors and the effect of ambient temperature on the reference spectrum, other environmental conditions may also contribute to uncertainties, such as interference from ambient water vapor.”

4. Section 2.2 – at what time resolution are these calculations made? As mentioned in the main text, the LEDs were operated in the continuous mode. H , u^* and were calculated at a 1-minute resolution in this study.

To clarify, the main text has been revised: “Sensible heat flux (H), friction velocity (u^*) and Obukhov length (L) were calculated from scintillometer measurements at a 1-minute resolution in this study.” In addition, to make the manuscript more concise, we decided to move the theory of scintillometer to the Supplementary Material Section 2.

5. Line 212 – Isn't an estimate using WindTrax and CO mole fractions a “top-down” estimate? You then compare it to one based on traffic volumes – isn't this one “bottom-up”?

Yes, that was a typo and has been corrected. WindTrax and CO mixing ratio is a “top-down” estimate. A traffic-volume-based estimate is ‘bottom-up’.

6. Line 247: do they generally agree? The level of agreement is not quantified. From looking at the time-series the variations certainly seem to be well captured, but it would be good to give a correlation coefficient for this.

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We have done this analysis, and now we include Fig. S2a, CO_NAPS vs. CO_FTIR, in the supplemental material. The r^2 is 0.67 when the wind came across the highway to the NAPS trailer, and 0.57 when the wind came from other directions.

7. In fact some basic statistics for the model's skill level would improve the manuscript. The authors have used the “open-air” package and this has some great tools for quick evaluation of a model's performance against observations.

This is a good suggestion. We have added mean bias and root mean square error of comparison between CO measurement and CO model in the supplemental material, Fig. S2b and c.

8. Reading this discussion about model to measurement spatial differences begs the question as to why a comparison of model to open-path FTIR is not shown. This will suffer similar problems but should be much less than the in-situ observations.

In the supplementary material, we have included Fig. S2c on CO_model vs. CO_FTIR with statistical results, and Fig. S4 polar plot of (CO_model – CO_FTIR) vs. wind direction. In the comparison of CO_model and CO_NAPS, the slope when the wind came from the highway and the slope when the wind came from other direction was very different (0.72 vs. 1.20), indicating the strong spatial differences. In the comparison of CO_model and CO_FTIR, the slopes are much closer (1.20 vs. 1.09), indicating the sampling spatial difference is smaller when comparing path-integrated mixing ratio with the volume-averaged chemistry transport model. From the polar plot of CO_model – CO_FTIR vs. wind direction and speed, it also can be seen that positive differences (yellow) occur mainly when the wind is from other directions, and the dependence of (CO_model – CO_FTIR) on the wind direction is not that strong. This discussion is also included as a new paragraph in the main text, Section 3.2.1.

9. Line 297. Are the traffic volumes similar on the weekends? This is surprising and you have not actually stated that clearly before. Can you clarify?

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Yes, traffic volumes on the weekend were similar to weekdays except for the morning period (from around 6:00 to 11:00), as shown as the black dashed line in Fig. 5 (bottom). We also wrote “The median CO mixing ratio on weekends was close to that on weekdays, except for the early morning period.” And to make it clearer to readers, we have included the actual traffic volume number in this sentence: “On weekends, traffic volume increased more gradually during the morning until plateauing around 11:30 and on average remained high with about 21800 vehicles h⁻¹ until after 22:00.”

10. Line 323: is it worth showing the correlation plot at least in the supplementary data?

Agreed. A supplemental figure of linear regression of NH₃ and CO mixing ratio from FTIR has been added to the supplemental material (Fig. S5).

11. Line 332: Are you also assuming that traffic is the only source of CO emissions above background?

Yes, we assumed that highway traffic emission at this spatial scale is the only source of CO above background, to estimate traffic-related NH₃ emission. We observed a good linear relationship between mixing ratio of NH₃ and CO, shown in Fig. S5.

12. Line 438: Why only 3 days? I assume that these are the best steady wind conditions? Whatever the reason for the choice of these days, it should be stated briefly in the text.

We only picked three days because the dispersion model is not easily automated and requires lots of manual labour. We have revised that paragraph and added the following sentences to the text: “July 22 was chosen because the wind direction was steadily from northwest, and a traffic jam occurred for added interest. July 28 and 29 were chosen because they are two of the highest days for temperature and O₃ during this project.”

13. Line 529: “reasonable” correlations observed. You need to provide some actual

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statistics to back this up somewhere in the manuscript.

We meant to point out that wind direction affects the comparison, so we revised this sentence as follows: “Model results and measurement results are not expected to be directly comparable for all wind regimes, and comparisons can be better explained after separating wind directions.”

14. Consider changing “mixing ratio” to “mole fraction” throughout, as I believe this is now the preferred terminology.

Thank you. We have seen both terms. This is noted in line 53 in the revised manuscript.

15. Line 110: the “fraction” of the path is not actually a fraction but a distance, consider rephrasing.

This was rephrased into “the length of the path that was directly over. . .”

Please also note the supplement to this comment:

<https://www.atmos-chem-phys-discuss.net/acp-2017-328/acp-2017-328-AC1-supplement.pdf>

Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2017-328>, 2017.

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