

The reviewer's comments are in bold type and our responses are in normal text. We thank this reviewer for his/her careful reading of our manuscript and for the comments. We have added a section to the supplementary material to describe the analysis process in more detail, and to show example spectra and fits.

1. The greatest shortcoming of the paper, however, is that there appear to be serious flaws in the data or data analysis, particularly the FTIR spectral analysis. Indeed, it appears that the FTIR data were not analysed at all, but simply taken “as is” from the system, and the IR data appear to have large systematic errors.

When writing this manuscript, we made the decision not to dwell on the minutiae of the FTIR analysis but to focus on the results. However, if more detail is required, we can certainly provide this. Quite a significant amount of work was done on optimizing the fittings and quantifying concentrations of pollutants. We have now included a section in the supplemental material to describe the FTIR spectral analysis by the OPUS_RS software in more detail. Examples of a measured spectrum and model fit spectra along with residuals for CO, O₃, and NH₃ are also shown in Fig. S1. To summarize briefly, the software's analysis algorithm is based on an iterative nonlinear fit of the measured IR spectra in compound-specified spectral windows. High-resolution spectra of the target and interfering gases, as well as baseline functions are fitted to those spectral windows in the measured spectra, making use of the known instrument response, the instrument line shape to meet the spectral resolution of the measurement, i.e., 0.5 cm⁻¹ in this study. More detailed descriptions and spectrum examples are now given in the supplemental material.

2. With regards to novelty of the study, the crux of the paper is to collect FTIR measurements over an urban highway. Such works have been previously reported – see for example work by Bishop et al., Stremme et al., Colman et al., Grutter et al. and Chaney et al, as well as by other analytical methods (for example, the seminal paper by Stedman et al., the tunnel study by Popa et al and the newly released paper by Haugen et al.). That is to say, there exists significant literature regarding traffic emissions, and the present authors need to cite more of these studies, and also need to cite other studies that use open-path FTIR to measure similar compounds from other sources

We believe our study contains several novel aspects: first, the measurement site is the busiest segment of the busiest highway in North America, as mentioned in the Introduction. Secondly, we conducted measurements for 16 days continuously. Thirdly, what we measured was pollution in the open ambient air over a highway from a real-world, un-manipulated mix of vehicle types. Our study is different from previous tunnel studies, because the air mass is more confined in the tunnel. Mixing processes and solar radiation conditions in the tunnel are different than in the open environment. We also observed and quantified additional pollutants, such as ammonia, formaldehyde, methanol and hydrogen cyanide, for which not much literature exists. These species are not well understood, and their levels can be different at different regions and countries due to different vehicle emission control regulations, fuels, and technologies. Another unique point of this study is that we combined detailed measurement of turbulent mixing (not only wind speed and direction) at the site simultaneous with pollution measurements from the FTIR, to show the role of turbulent mixing on the local air quality. We thank the reviewer for suggesting additional relevant background papers, and they have been cited in the revised manuscript.

3. To add a more unique aspect to their study, we suggest that the authors i) emphasize more the aspect of monitoring near such a large motorway (e.g. perhaps there are atmospheric reactions / products only seen due to higher order rate constants requiring very high CO or O3 levels?), and ii) present a more detailed analysis of the instrumental comparison, i.e. the “shoot-out” between the measurements collected from the FTIR and from the NAPS.

We believe there are many unique aspects to this study. As we have stated in the manuscript and explained in point 2 above, one unique aspect of this study is that we combined direct measurements of mixing ratios of gas-phase pollutants from such a busy segment of highway with detailed micrometeorological and turbulence measurement to show that not only traffic emission rates, but also turbulent mixing conditions in the surface layer play an important role on the accumulation and build-up of air pollutants over this highway. We also observed several interesting pollutants, such as ammonia, formaldehyde, and hydrogen cyanide, which have important implications to atmospheric chemistry and population health, and for which in-situ measurements over a busy highway have not been commonly reported. Another unique aspect is our evaluation of a “top-down” approach to calculate the emission rates of a few primary pollutants with a dispersion model.

The “shoot-out” aspect will be discussed below points 4, 5, 6, 7, and 8.

4. We believe (at least some of) the data as reported are not correct. In particular, looking at the CO plot in Figure 3, the trends for the two instruments follow one another in a nearly identical manner with overall very similar diurnal profiles. If the NAPS were as sensitive to wind direction as the authors purport, then its corresponding diurnal profile would not manifest the same diurnal trend as the FTIR: The NAPS profile would better reflect the wind direction dependence, yet the NAPS values never trend down as the FTIR values goes up (or vice versa). That is to say, at no time is there an obvious anti-correlation seen in the data. Furthermore, CO is a very well mixed gas and the NAPS instrument (Figure 1) is physically located in the middle of the FTIR optical path. Since the path for the FTIR includes both the highway and a stretch of land greater in length than the highway, the averaged CO mixing ratio obtain by FTIR over the entire path should be similar to the NAPS value, but possible lower due to mixing in cleaner adjacent air. Inspection of Figure 3 directly points to this – while the magnitudes and offsets differ (significantly!!), both the NAPS and FTIR data maxima and minima track each other very well, having the same diurnal patterns. However, the fact that the CO mixing ratio values (in terms of offset and amplitude) do not agree at a quantitative value suggest that either the data or data analysis is likely incorrect and it is in this regard we criticise the paper.

The long-path FTIR provides a path-integrated concentration, whereas the NAPS trailer measured concentration at a point located at the south edge of the highway. Therefore, concentration measured by the NAPS trailer was more dependent on the wind direction than concentration obtained from the FTIR. But we must keep in mind that even a point measurement has a footprint. The footprint covers more highway when the wind comes from the north, and covers much less highway (but more parking lots, buildings, a park and golf club), when the wind was from the south. The polar plot of the (CO_FTIR – CO_NAPS) in Fig. 4a already clearly shows the dependence of the difference between the two measurements on the wind direction. We have stated that when the wind direction was from highway to NAPS trailer, the difference was smaller, and when the wind was from other directions, the difference

was greater. When the wind was from the south, the NAPS trailer measured less footprint covering the highway, but still measured some traffic pollutants. Therefore, the NAPS measurements of CO (primary pollutant) still demonstrated a similar diurnal cycle on weekdays as the FTIR measurements. There is no reason to think that CO_NAPS and CO_FTIR should be exactly anti-correlated, since both instruments sample overlapping plumes.

The FTIR measures a path-integrated concentration, and its footprint was not completely independent of wind direction. The dependence is weaker compared to the NAPS trailer. As the reviewer has noticed, the path of our set up included “a stretch of land”, and this stretch of land was downwind when the wind came from north. Therefore, the downwind length of the FTIR path was longer when the wind came from north. To illustrate the different footprints the FTIR picked up with different wind directions, a plot based on the geometry of the path is shown below:

Portion of the measurement path of the FT-IR catching CO emitted from the highway, depending on the wind direction

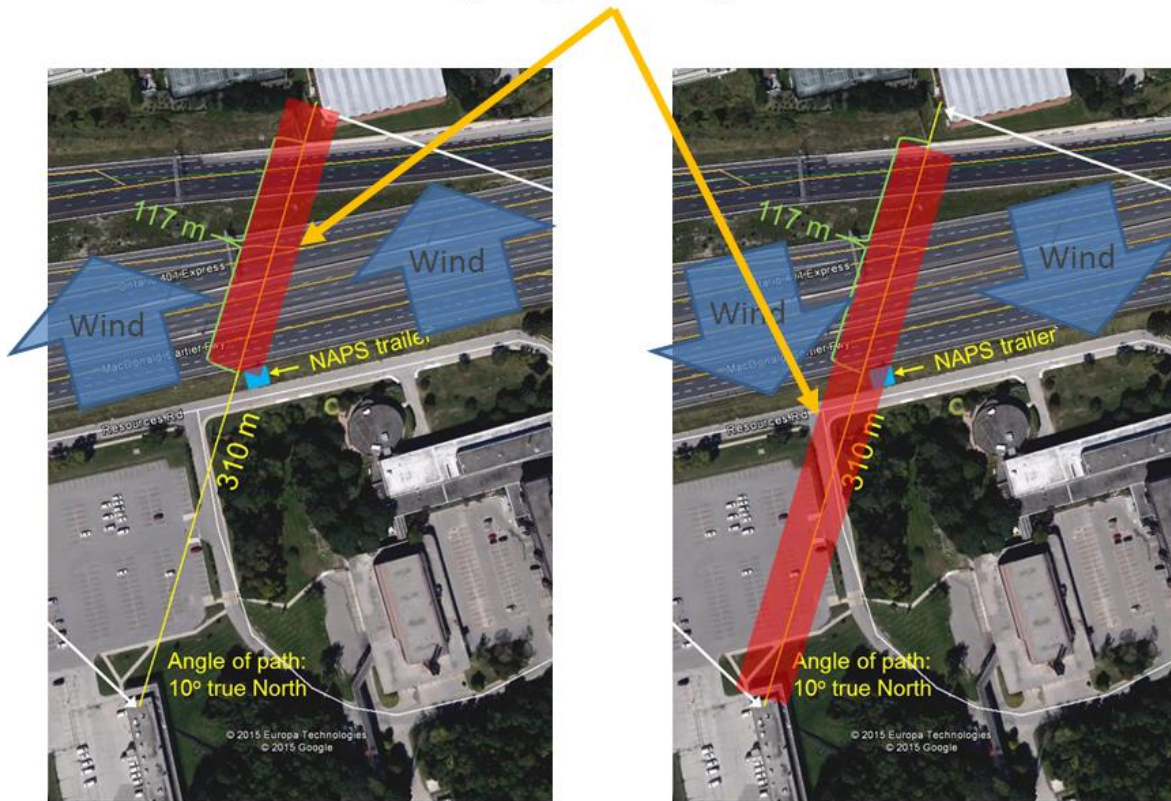


Figure: Dependence of the footprint of the FTIR path measurements on wind direction.

Regardless of wind direction, the FTIR path always picked up some pollution emitted from the highway, and this segment of highway always has traffic (daily minimum is around 3500 vehicle per hour at around 2 to 4 a.m.). So the FTIR measurement never actually sees mixing ratios at the level of urban background air. The NAPS trailer observed a lower pollution level than the FTIR when the wind came from the south because the footprint does not include the highway. This explains the offset between minimum levels of

CO_FTIR and CO_NAPS. Again, CO_NAPS and CO_FTIR were not expected to be anti-correlated at any time or wind direction.

In the supplementary material Fig. S1, we have included examples of a spectrum, and spectral fitting of CO, O₃, and NH₃, at 16:01:45 on July 29, when the wind came from south and a significant offset was observed. These plots of fittings do not show any significant residuals or problems.

We also have checked that NAPS measurement of CO had zero calibrations four times each day. The average of the zero readings was 12.6 ppb with a standard deviation of 7.4 ppb. So the NAPS measurement of CO did not have significant offset and we estimate the uncertainty of CO_NAPS to be 22 ppb (three times of the standard deviation during zero calibration).

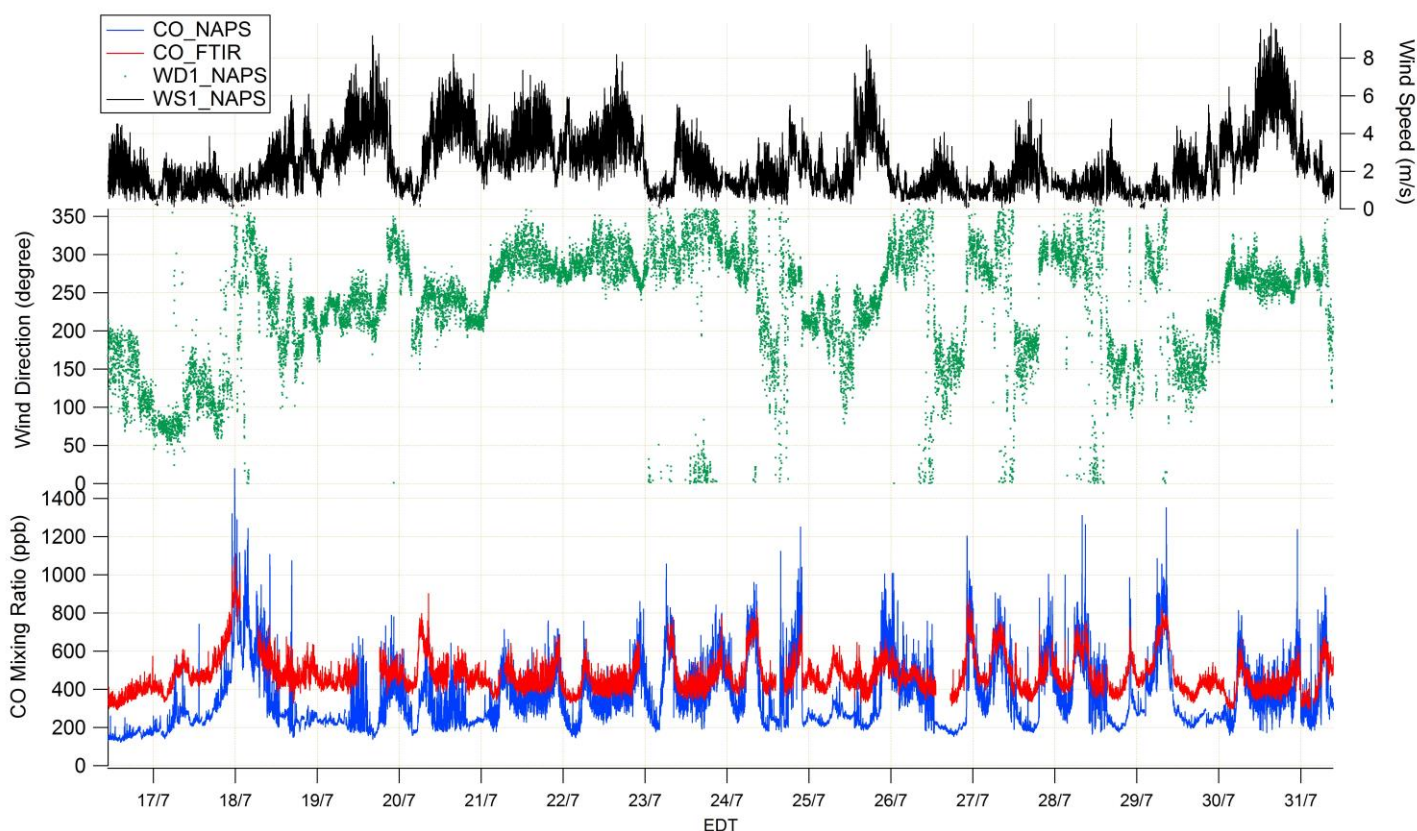


Figure: Time series of CO_FTIR, CO_NAPS, wind speed and wind direction.

5. Specifically, if one observes the CO plot in Figure 3 focusing only on off hours (weekends and during the deep night-time hours), one can draw horizontal lines through the mixing ratio minima for the two methods. While this reviewer is limited to a pencil and paper for the analysis, for such lines “urban minimum baseline” mixing ratio we find the minimum value for the FTIR is ~380 ppb and for the NAPS data the off-hours minimum ~190 ppb, almost exactly a factor of 2.0 difference for this “clean air” baseline.

As stated above, there really was no urban minimum baseline for the FTIR CO mixing ratio, since it always picked up pollution from the highway, even at night, with a minimum of 3500 vehicles passing by per hour. Therefore, over this extremely busy highway, the concept of “off hours” does not apply. For NAPS, if the wind came from the south, it could get some significantly lower levels than the FTIR.

6. While one could argue that this is due to different dispersion / mixing, this is clearly not the case because: 1) the NAPS point source measurements are 1/2 the FTIR values (always lower), and the FTIR values should be greater, representing increased dilution of CO, i.e. mixed with cleaner air further from the motorway, and

This is not quite correct; the NAPS measurements were not always lower than FTIR. For example, 7/24 around 01:00, 7/26 around 03:00, and 7/27 around 0:30, NAPS and FTIR CO were very close. These periods may correspond to the “off hours” mentioned by the reviewer. However, we think there are no “off hours”, since this segment of highway always has traffic (daily minimum is around 3500 vehicle per hour at around 2 to 4 am). NAPS and FTIR CO were very close during these periods mainly because the wind came from north during these periods. It has been shown in the polar plot Fig. 4a.

7. 2) the overall diurnal variations track each other over the entire time interval. We suggest that the difference is likely due to either a systematic negative offset for the CO measurements via NAPS, or (more likely) a systematic large positive offset for the CO measurements with FTIR. It is unclear which of the two instruments is out of calibration, but we suggest the FTIR values are systematically offset and have the incorrect scaling factor, as nowhere in the paper is a calibration procedure reported. There are differences in the offsets for the other gases as well, but CO appears to have the highest. Furthermore, the mixing ratio range for CO from the FTIR is ca. 60% of the NAPS. Again, just by “eyeballing it” the data would appear to show a relative instrument response relation is of the order $Y_{FTIR} = (0.60) * Y_{NAPS} + 180$ ppb. Picking the EDT of 24/7-25/7 (CO from Figure 3), the minima and maxima values for the NAPS are ~ 220 and ~900 ppb, respectively, which is a range of ~ 680 ppb. While the minima and maxima values for the FTIR are ~ 400 and ~780 ppb, respectively, which is a range of ~ 380 ppb or 56% the dynamic range of the NAPS.

We agree with the reviewer that the FTIR CO “offset” seems suspicious; this is something that bothered us from the beginning. Because of this, we have expended significant effort to ensure that there are no biases introduced by our spectral analysis. An example fit is shown in Fig. S1. Interference by H₂O was investigated and found to be insignificant; and different spectral windows suitable for CO were investigated (2068 – 2198 cm⁻¹, with correlation coefficient threshold = 0.7, the difference is 14 to 26 ppb (25 to 75 percentile)), but all led to the same result. After eliminating the possibility of instrumental biases, the most logical explanation remaining is that the higher offset in the FTIR CO is due to the persistent traffic throughout the night, which always affects at least part of the FTIR optical path.

The issue of the slope of 0.6 (rather than 1.0) is again explained by path-integration vs. point measurement. Due to the proximity of NAPS to the highway, larger maxima in CO_NAPS are to be expected, while the CO_FTIR is always an average that includes diluted air.

We have revised the text in the manuscript that describes how we came to the conclusion that this CO offset between FTIR and NAPS appears to be real:

“A major contributing reason for the differences of CO mixing ratios between the FTIR and the NAPS is that the FTIR and the NAPS were not sampling the exact same air, i.e., the measurements represented different footprints. The FTIR measured the air along the path across and above Highway 401, which always included some pollutants emitted from traffic. In contrast, NAPS numbers represented point measurements beside the south edge of the highway. Therefore, CO mixing ratios measured by the NAPS trailer were more dependent on the wind direction than mixing ratios obtained from the FTIR. When the wind was from the south and towards the highway, the NAPS trailer was mostly blind to the highway; when the wind was from the north, it was immediately downwind it. Therefore, CO mixing ratios from the NAPS are expected to be lower than mixing ratios obtained from the FTIR when the wind is from the south and towards the highway.

The path-integrating approach of the FTIR also has a dilution effect since a significant fraction of the path is not above the source (i.e. the highway). Therefore, the CO mixing ratios obtained from the FTIR should be less than CO mixing ratios from NAPS during winds from the highway towards the NAPS trailer. The polar plot in Fig. 4a clearly shows the dependence of the CO mixing-ratio difference between the FTIR and the NAPS on wind direction. When the wind came from the north over the highway towards the NAPS trailer (above the dashed line), CO mixing ratios from the FTIR were close to or lower than mixing ratios from the NAPS. When the wind was from the south and towards the trailer (below the dashed line), the CO mixing ratios from FTIR were higher than CO mixing ratios from NAPS.”

8. Ozone has less of an offset, but the scaling factor is greater. Using the same EDT as above, the dynamic ranges for NAPS and FTIR are 48 and 20 ppb, respectively, which correspond to the FTIR being 42% the range of NAPS. Clearly, there is (are) a systematic flaw(s) present that biases the results by factors of 1.7 (CO) and 2.4 (ozone) Again, since the measurements do not agree, it is unclear what the mixing ratios are for CO and ozone at this certain location at this specific point in time.

If one calls the difference between point measurements and path-integrated measurements a systematic flaw, then we agree. They cannot be directly compared in the sense of a cross-calibration; differences are due to the fundamental difference in the measurement itself, and as previously discussed, the observed differences can be explained by the differing footprints, point measurement right next to the highway vs. path-integrated/diluted measurement incorporating 1/3 highway and 2/3 parking lot, which, depending on wind direction, will contain highway emissions or not.

The offset of O₃ is mainly due to the different footprint. As already shown in polar plot Figure 7 (a), the difference is small when the wind came from the north, and the main offset occurred when wind came from the south. A time series plot of O₃ with wind direction and speed is also shown below. Both polar plot and time series show that the big “offset” occurred when the wind came from around 120 to 220 degrees. The two measurements were close during July 21 to 24 when the wind was steadily from north. We also admit, as indicated in the updated Table 1, that we have less confidence in the accuracy of the FTIR measurements of O₃.

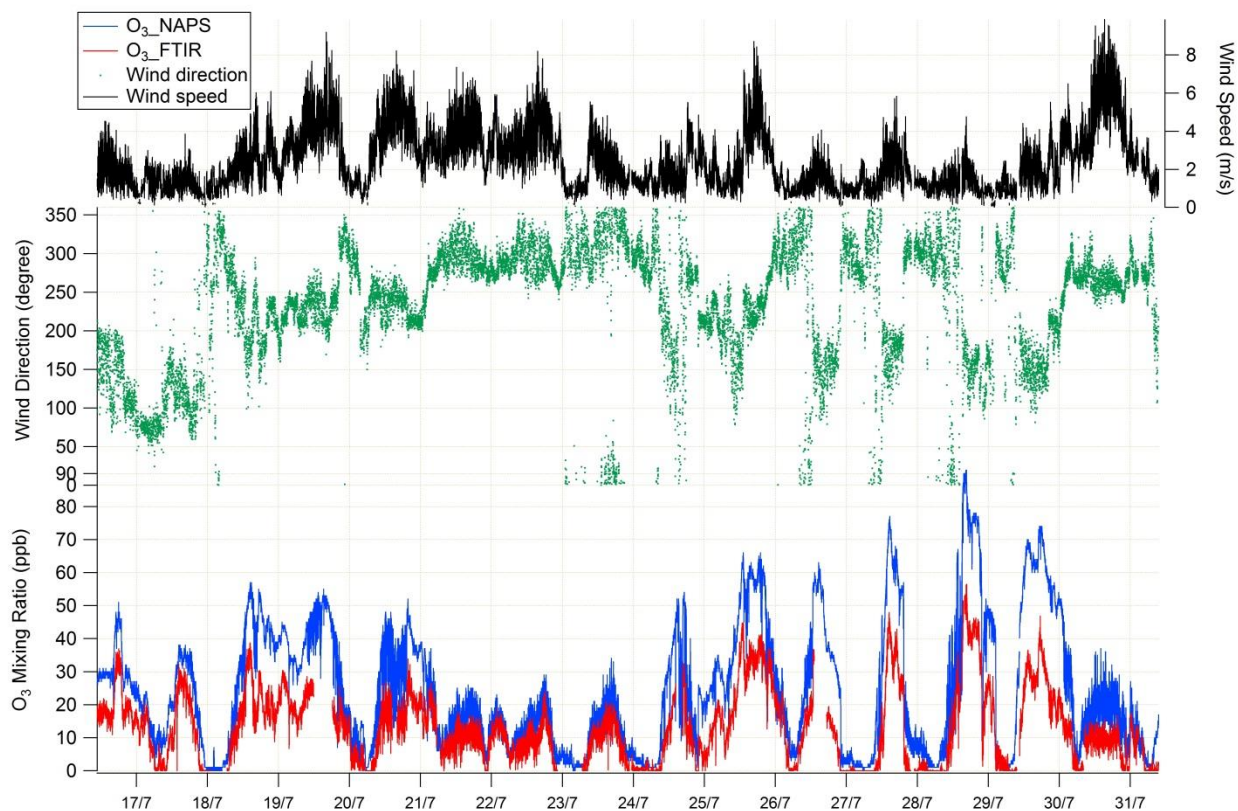


Figure: Time series of O₃_FTIR, O₃_NAPS, wind speed and wind direction.

9. In the paper, there are no explicit data evaluation plots that show what the classic least squares fit looks like relative to the measured spectrum as well as the associated residual plot. It would appear that there was minimal to none of the “hands on” analysis of the FTIR spectra, and as if the results of the Bruker FTIR software (OPS) were used without inspection or vetting. There may thus be miscalculations present that initially went unnoticed. FTIR spectral analysis of gaseous mixtures is not yet a fully automated procedure, but is an interactive process that is subtle and prone to mistakes, interferences, etc. There are several more sophisticated gas analysis programs that may be used to independently confirm or refute the results calculated by OPS, and actual evaluation of the spectra is required.

As already mentioned in our response to comment point #1, when we wrote the manuscript we made the decision not to get into all the details of the FTIR analysis but to focus on the results. To address the reviewer’s concerns, we have included a section in the supplemental material to describe the FTIR spectral analysis by the OPS software. Supplemental text and Fig. S1 have been included to show details of fitting analysis and example spectrum, fitting results, and residual plots. The selected example plot is a spectrum taken at 16:01:45 on July 29, when the reviewer observed a big “systematic offset” of CO and O₃. The wind was from the south. Fig. S1 shows reasonable fits and small residuals of CO, O₃, and NH₃.

10. Furthermore, while authors did correct the raw mixing ratios for temperature and pressure, it appears that the temperature/pressure corrections need to be processed on the reference spectra as well, prior to the fitting. In other words, each reference spectrum (from HITRAN or PNNL) need to be scaled to the correct temperature/pressure then used for quantification. For example, the high resolution reference spectra need to be deresolved to the same resolution of the measured spectra, which in this case is 0.5 cm⁻¹. In the paper, it was not evident if the HITRAN or PNNL reference data were correctly fit to the instrument parameters and instrument lineshape (ILS). It is critical to create the same (instrumental) lineshape and resolution for the fit. Reference spectra may be deresolved using a Gaussian, Lorentzian or Voigt profile, and after deresolving them it is a good idea to check the FWHM to confirm the resolution.

Actually the ILS fit was indeed performed and is included in the quantitative evaluation. We have now included ILS fit information in the supplemental material. Each reference spectrum (from HITRAN or PNNL) was converted to a spectrum with the same resolution (0.5 cm⁻¹ in this study) by the ILS model in OPS software. We did not calculate new reference spectra based on the actual temperature before fitting, but we provided conservative estimations (8.9 % for NH₃, 4.2 % for CH₄, 8.3 % for CO, and 4.1 % for CO₂) of temperature related uncertainty of the retrieved mixing ratios in the range of 5 to 50 °C, which is much wider than the range of difference between ambient temperature and 296 K during our study. We also corrected the raw mixing ratio from FTIR due to the change of air density with change of ambient temperature and pressure.

11. Finally, it appears that one or both instruments were used without any calibration. In order for the scientific community to have confidence in the values obtained, it is important that the instrument(s) undergo some sort of on-site calibration. For the FTIR a wavenumber calibration is necessary and e.g. uses a small gas cells containing NH₃ are used for this purpose; other compounds such as H₂O, CO or CO₂ can also be used.

A wavenumber/line position calibration was performed by using the spectrum fitting analysis of H₂O from the ambient air. In the Bruker software, OPUS_RS, this process is included in the ILS function, as described in the supplementary material. A gas-cell calibration is of course desirable but is not nearly as straightforward as it sounds, since specialized cell windows are required, making this an expensive proposition.

12. Abstract Pg. 1, sent.21: In previous studies, emission factors have units of g kg⁻¹, here the emission factors have units of g km⁻¹. Please explicitly define the emission factor that you are estimating somewhere in the manuscript.

One sentence has been added to the text in Section 3.7 indicating the units of emission factor reported in the references were gram mile⁻¹ and gram km⁻¹, and they were converted into gram km⁻¹. The references in Table 2 also reported emission factors in grams per distance, and they were all converted into grams km⁻¹ to compare to our results. In the last third paragraph in Section 3.7, one sentence was added to explain again that the emission factors results shown in Table 2 were calculated from the emission rate estimates obtained from WindTrax. These emission factors were calculated to compare with previous reported values.

13. Pg. 1 sent 29: There are more pollutants associated with motor vehicles that are not listed here. (Please see Review Article)

Yes, thank you. We have written it out more clearly (such as semi- and low-volatile organic compounds, aromatics and PAHs) and cited more references.

14. Pg. 2 sent 40: change “of” to “in”

It has been corrected.

15. Pg. 2 sent 60: change “spectrometry” to “spectroscopy”

It has been corrected.

16. Pg. 3 sent 65: remove “however” from the sentence.

We keep “however” here, since we are contrasting our study to those in the previous paragraph.

17. Pg. 3 sent 85-86: remove “which were” from the sentence

It has been removed.

18. Pg. 3 sent 87-88: please state what NAPS measures

This sentence has been revised in more specific: “to conduct an in-depth comparison of a range of pollutants (CO, O₃ and NO_x) measured by both the path-integrating FTIR instrument and the in -situ station.”

19. Pg. 3 sent 89: change “in-situ” to “in situ”

It has been corrected.

20. Pg. 3 sent 95: the objectives have already been done (paper by Griffith et al. and Yokelson et al.).

The description of objective point 1 has been revised: “1) to evaluate the capabilities of the long-path FTIR spectroscopy for quantifying the mixing ratios of gaseous pollutants in a heavily polluted open urban traffic environment for a length of time sufficient to cover a range of environmental conditions (16 days)”

Experimental

21. Pg. 4 sent 104: The Global temperature between 1200 and 1500C is its varying state, however, it will not be in its varying state during the measurements.

The exact temperature is not well known, so we decided to leave it out.

22. Pg. 4 sent 105: This is called a bistatic configuration.

Thank you. It has been noted in the revised manuscript.

23. Pg. 4 sent 116: Please add a reference to the end of this sentence. For example: Akagi, S.; Yokelson, R. J.; Burling, I.; Meinardi, S.; Simpson, I.; Blake, D. R.; McMeeking, G.; Sullivan, A.; Lee, T.; Kreidenweis, S., Measurements of reactive trace gases and variable O₃ formation rates in some South Carolina biomass burning plumes. Atmos. Chem. and Phys. 2013, 13 (3), 1141-1165.

We don't think this reference is supportive to that sentence, so we added this reference to the biomass burning sentence in line 65 in the revised manuscript.

24. Pg. 4 sent 119: Please add references to the previous studies that used absorption features in spectral window for analysis.

Yes, I have added a column for references to Table 1.

25. Pg. 4 sent 122: Please add reference for the HITRAN database

I had the reference at line 56, and I added the reference here again this time.

26. Pg. 4 sent 123: Please add reference for the PNNL database

I had a reference at line 57; I added this reference here again this time. I also included Johnson et al. (2010).

27. Pg. 5 sent 126: How were these values adjusted for temperature/pressure? The reference spectra need to be adjusted to the correct temperature/pressure and used in the fitting process. From this sentence, it appears that the reference spectra were not corrected, but instead the reference spectra were used as is to calculate the mixing ratios, which were then adjusted for temperature/pressure.

The reference spectra were not corrected by ambient temperature in the analysis shown in this manuscript. Mixing ratios were calculated by reference spectrum from two databases at 296 K and 298 K, and then were corrected by the density of air depending on ambient temperature and pressure. The effect of temperature on the reference spectrum is small for the range of ambient temperature in this study. We estimated the uncertainty due to the difference between the ambient temperature and reference spectrum in a conservative way, as described in section 2.1 and supplementary material.

28. Pg. 5 sent 131: cite PNNL and HITRAN databases.

They are cited here now. They were already cited in line 56 and 57 in page 2.

29. Pg. 5 sent 135: This is a huge uncertainty for greenhouse gases. For example, is the CO₂ 400? Or 440 ppm?

As we have stated in the text, the maximum uncertainty of 10 % is a conservative estimation because we used a much wider temperature range than the actual ambient temperature range. In addition, we did not report any CO₂ mixing ratio in the manuscript.

30. Pg. 5 sent 146: sensible heat flux. . . what is this?

Sensible heat flux is the turbulent flux of “heat” across a warm-to-cold air gradient. It is a component of the atmosphere/surface energy budget (net radiation = sensible + latent heat + soil heat flux). It is typically measured using eddy covariance, but in this case, we are calculating it using similarity theory, based on the scintillometer data. (Stull, 2003)

Stull, R.B.: An Introduction To Boundary Layer Meteorology, Kluwer Academic Publishers, Netherland, 670 pp., 2003.

31. Pg. 7 sent 174: does this make a difference?

The location of the weather station was written out here to describe the source of solar radiation data. 9 km is close enough at the relevant time scales.

32. Pg. 7 NAPS measurements: please provide type of analysers used at the NAPS station.

Information on the NAPS analyzers has been added to Section 2.3 as follows: “The CO analyzer [Model 48iTrace level-Enhanced, Thermo Fisher Scientific, USA], operates based on infrared absorption and gas filter correlation; the NO_x analyzer [Model 42i Trace Level, Thermo Fisher Scientific, USA] on chemiluminescence; the O₃ analyzer [Model 49i, Thermo Fisher Scientific, USA] on UV absorption; the SO₂ analyzer, [Model 43i, Thermo Fisher Scientific, USA] on UV fluorescence.; and the PM_{2.5} analyzer [Model SHARP 5030, Thermo Fisher Scientific, USA] on light scattering and beta attenuation. Meteorological parameters including air temperature, pressure, relative humidity, and wind speed and direction, were monitored using a WXT520 weather station [Vaisala, Finland].”

Results and Discussion

33. Pg. 9 sent 225: the measurements are off and so what is the point of the study?

As we have explained in points 3, 4, and 5, the offset between two measurements can be explained. They agreed well when wind came from north (highway). The main points of this study have been explained in our responses to comments 2 and 3. Text has been added in lines 89-95 on page 3 in the revised manuscript.

34. Pg. 9 sentence 244 add references to this sentence.

Yes, these references have been included now.

35. Pg. 9 sentence 247 “generally agree with each other, but with a significant offset. . .” what does this mean? The language used here is vague and does not tell us anything.

I have revised this sentence in the text to be more specific: “As shown in Fig. 3, many mixing ratios peaks of CO from the FTIR and the NAPS matched well, and mixing ratios generally correlated with each other

(Fig. S2 a), but with a significant offset and amplitude difference when the wind came from the south (more detailed comparison in the next paragraph).”

36. Pg. 10 paragraph 1/looking at figure 3: CO from FTIR has an offset of 390 ppb and the NAPS has an offset of 190 ppb?

Please see our response above in points 4, 5, 6 and 7.

37. Pg. 12 sent. 320-321: Please cite the studies that you are referring to here. Throughout the paper whenever referring to studies, please cite them.

Yes, now I have cited there again. I meant the two references/studies discussed in line 321-330.

38. Pg. 12 sent. 331: Fig 6 should be Fig 5?

Yes, thanks. We have corrected it into Fig. 5 in the revised manuscript.

39. Pg. 13 sent 364: Here you are not comparing the FTIR results (due to water interference) to the model, yet FTIR is mentioned here.

Yes, the reference to the FTIR path has been removed.

40. Pg. 13 sent 369: remove “and a” from the sentence.

Sorry, a typeset mistake. Should have been “significant decrease in the middle of the day and a secondary peak...” It has been revised: “... reaching a peak over 100 ppb from 6:00 to 8:00 followed by significant decrease in the middle of the day and a secondary peak between 20:00 and 23:00.”

41. Pg. 17 sent. 471: Here it states the differences, and the range is large, yet you state that it “within the range of estimates”. Cite some of those ranges to support your claim.

The ranges have been cited there.

42. Pg. 18 sent. 498: change “are in range” to “in the range”

It has been corrected.

43. Pg. 18 sent. 499: change “of” to “the”

It has been corrected.

44. Pg. 19 sent. 529: change “comparable” to “compare”

This sentence has been revised to: “...model results and measurement results are not expected to directly compare for all wind regimes, and comparisons can be better explained after separating wind directions.”