We thank the reviewer for the helpful comments and suggestions. In the following detailed response to all reviewer comments, the Reviewer Comment is in black, followed by our response in blue and modifications to the manuscript in green.

Response to Review #1

General comments

The authors give a detailed description about an instrument and its characterisics for Solar Occultation Flux (SOF) measurements in the UV-Vis and IR spectral regions built at the University of Colorado. They also show and discuss results from experiments where this instrument was used to measure the column densities and fluxes of ammonia (NH3), nitrogen dioxide (NO2), and ethane (C2H6) in various types of areas and loads of air pollution. The data was compared to literature and inventory data. The authors also used their findings to determine structure functions as a way to define trace gas variablity over distance which is a valuable contribition with respect to remote measurements from satellites.

The methods are described in a clear way and the results are sound and relevant. The structure of the text is well chosen and allows a good and fluent reading of the manuscript.

Specific comments

General: The abbreviations RD10 and RD11 are mentioned throughout the text supposely for ”research day” but specified first much later in the manuscript in form of a caption to Table 5. But this kind of naming makes the reader curious: How do the results of RD01 to RD09 look like? How many more RDs are there? Can the authors please motivate why only the results of these two days are presented.

We conducted a total of 16 research drives with different research objectives, quantifying emissions from feedlots being one of them. In this manuscript we chose to present the 2 research drives during which we drove around various feedlots; the exact drive track was the same on those two drives as shown in Figure 6. Data for all RDs is available from the FRAPPE data archive, and the information how to access the archive will be added in the Data Availability section of the final paper.

Line 319: The mobile SOF was deployed during 16 research drives (RDs) during FRAPPE. Here, we present data from two RDs that were conducted along almost identical drive tracks on consecutive days, and shared common scientific objectives. The drive track for the case study from 13 August 2014 is shown in Fig. 6 and similar to the drive track on 12 August 2014.

Lines 87 – 88: There are more sources that can be used to refer to earlier descriptions of the method. The SOF-method is described in an article by Mellqvist et al. (2005) and actually was patented by Galle and Mellqvist. I suggest to add a citation to this article.

We added several references. See comment on references from reviewer #3.

Lines 105 – 111: While the respective footprints for TES, IASI, SCIAMACHY, and OMI are given this information is not presented for ACE-FTS and MIPAS. Please add this information.

These satellites only conduct measurements in the upper troposphere, and do not relate directly to the near surface variability in VCDs investigated by our paper. We have added additional references to ACE-FTS and MIPAS.

Hoepfner, M., et al. (2016): First detection of ammonia (NH3) in the Asian summer monsoon upper troposphere, Atmos. Chem. Phys., 16, 14357-14369, doi:10.5194/acp-16-14357-2016.

Coheur, P.-F., et al. (2007): ACT-FTS observation of a young biomass burning plume: first reported measurements of C2H4, C3H6O, H2CO and PAN by infrared occultation from space, Atmos. Chem. Phys., 7, 5437-5446, doi:10.5194/acp-7-5437-2007.

Line 144: The authors mention that there EM27 FTS is customized. In which aspect is this EM27 FTS customized?

Our EM27 uses the fast-scan option, 24V power supply, TE cooling of detectors, sandwich InSb / MCT detector as is described on line 146ff. of the original paper.

Line 165 and Table 3: Based on which information were these a-priori profile paramaters chosen?

The a-priori profile of each species was taken from the WACCM model.

Line 253: How did the authors determine an uncertainty of 30 %. Has this number been arbitrarily chosen?

The authors conducted a sensitivity study on the model wind to investigate the sensitivity of the emission flux calculations to uncertainty in wind direction, wind speed and vertical extent of the plume. The paragraph from lines 245-253 has been modified:

Lines 245-253: The model wind was compared to measurements of wind speed and direction at the Boulder Atmospheric Observatory (BAO), observed at 10, 100, and 300 m above ground. The uncertainty analysis of the model wind speed and wind direction is based on the time window 16-22 UTC, which is the time spent on the research drives. The model wind did not exactly have altitude layers at 10, 100, 300 m to compare to BAO; therefore, the model wind was extracted at 3, 105, 325 m, respectively, which represent the values closest to the BAO tower altitudes. The results are shown in Figure S1 of the SI text.

The error component due to wind direction was actively minimized using the spatial information contained in the mobile SOF data. The wind direction is constrained by the direction of the plume evolution from the sites, and measurements of VCD column enhancements downwind. It was determined that the model wind direction for site 1 is representative of the actual wind direction, whereas for sites 2 and 4 the wind direction was corrected by 7/23 and 11/18 degrees for RD10/RD11, respectively. For comparison, the wind direction at BAO agrees to <40 degrees on 12 and 13 August 2014. To determine the effect the wind direction uncertainty has on the emission flux, the emission flux was first calculated using the model wind and then compared to the model wind corrected by direction. The wind direction uncertainty for site 2 results in an emission flux uncertainty of 9.3 ± 3.6 %, and 19.0 ± 8.6 % for site 4. For the three sites combined the wind direction uncertainty on the emission flux is 9.5 ± 7.8 %.

Based on the slopes in Figure S1 the wind speed has a percent error of 16.8 ± 1.2 %. There is little variability in the relative differences between measured and predicted wind speed with altitude at BAO. The model wind speed used to calculate the emission flux was determined by averaging over different altitudes within the PBLH, as indicated in the bottom panel of Figures 7 and S2.

Vertical plume dispersion determines which altitude to use for averaging the model wind speed. The PBLH varies from ~500–2,500 m from the time of driving around site 1 to site 4. The model estimates that most NH_3 is located in the lowest 500 m of the VCD. The error due to vertical variability in winds during RD10 and RD11 was 11.2 ± 8.3 %. This error falls within the error on wind speed, indicating that the emission flux here is not sensitive to the vertical plume extend.

The combined uncertainty of wind direction and wind speed on the emission flux is 18 % for site 1 during both RD10 and RD11, and dominated by the error in the wind speed. For site 2 the total wind uncertainty on emission flux is 17.8 ± 0.5 %, and for site 4 the uncertainty is 22.0 ± 3.4 %. Based on the evaluation of winds at BAO, and use of the corrected wind direction for each site, the uncertainty in the emission fluxes due to winds is 20%.

The uncertainties in Table 5, 6 and text due to wind were conservatively estimated as 30% in the original manuscript, and now replaced with the new finding of 20 % uncertainty in the revised manuscript.

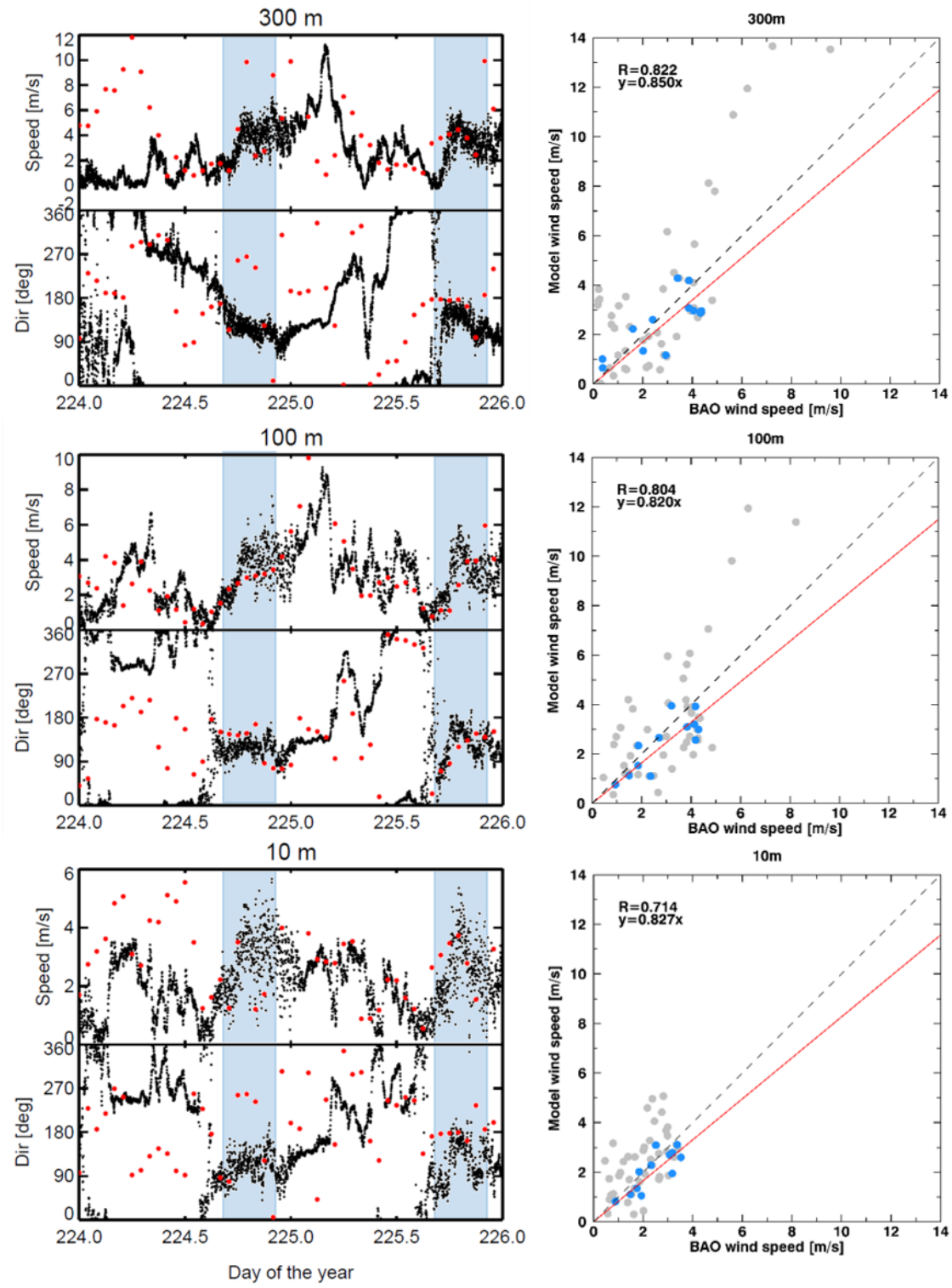

Figure S1: Comparison of wind speed and wind direction at 300 (top), 100 (middle), and 10 m (bottom). Left column: Black is BAO observed wind, red is modeled wind for 12 and 13 August 2014. The colored shading indicates the times of the RDs on both days (16-22 UTC). Right column: Blue indicates data from 16-22 UTC, gray is all data. The red line indicates the fit to blue data, the dashed line is the 1:1 line.

Section 3.1.1: The authors present their analysis of the precision and accuracy with respect to the measurements of NH3 and C2H6. Please add an equivalent analysis for the measurement of NO2.

Done

Line 280: The LOD and precision of NO2 from the DS-DOAS are 7 x 10^{14} and 3 x 10^{14} molecules cm^{-2}, respectively (Baidar et al., 2016).

Lines 287 and Table 4: The total error was calculated by arithmetic addition. Since the error sources can be regarded as statistically independent parameters, the root sum of the squares should be used. Why is the relative precision, expressed in percent, not calculated and taken into account for the calculation of the total error?

Thank you. The total error has been updated. The relative precision is accounted for in the total error in form of the fit uncertainty.

Section 3.1.2 and Figure 4 and lines 553 – 560: Why is a dependency of the ILS modulation efficiency with the azimuth and elevation angles expected? Don't the results presented in Figure 4 rather reflect the quality of the alignment within the SOF instrument? Further, only the effect of the ILS on the retrievals of NH3 and C2H6 is disussed here. Why is a according discussion missing for the retrieval of NO2?

We partially agree. The lack of a AZ and EA dependence demonstrates that the solar tracker is well aligned relative to the spectrometer, as well as good internal alignment of the FTS. This is an important distinction, as a dependency of the ILS modulation efficiency with azimuth and elevation angles carries potential to bias the field observations, where in particular the AZ varies by 360 degrees for each box. Ours is the only SOF instrument for which it has been shown that VCDs are free of such biasing to the best of our knowledge.

See above regarding the detailed evaluation of NO2 elsewhere. This paper is focusing both on results of NH3, C2H6 and NO2 emission fluxes from feedlots using the mobile SOF, as well as characterizing the performance of the FTS aboard the mobile SOF.

Figure 4: Is the scale on the x and y axes true for both unit circles?.

Thank you for pointing this out. The scale is only true for the outer unit circle, the inner unit circle has been downscaled to 80%.

Line 882: The black unit circles represent an ideal ILS modulation efficiency having a value of 1.000.

Line 358: Why did the authors assume here an error in the wind of 30 %/sqrt(2)? To my understanding, this is not an uncertainty which decreases with an increasing number of days of measurements, here 2 days, if it is that what was assumed.

The uncertainty of the model wind has been evaluated in more depth as described above including changes to the manuscript.

Line 377: Are the used O3 mixing ratios presented by Pierce (2016) applicable for the sites and times of the measurements of this study?

Thank you for the comment on the O3 mixing ratios. The values that were presented in the manuscript are from the daily summary for the specific days for the Ft.Collins-Greeley area. The more specific mixing ratios for the applicable time in the Ft.Collins-Greeley area are 50, 70 and 72ppb for RD10 at 1pm LT, and 66, 67 and 71ppb for RD11.

Line 377: On RD10 and RD11 O3 concentrations of 64 and 68 ppb (19 UTC and 18 UTC 1hr average, respectively) (Pierce, 2016) correspond to a NO lifetime of ~40s (66 ppbv O3). With wind speeds of ~4m/s NO was converted into NO2 over a distance of ~160 m (RD11).

Line 403 – 405: The connection of the higher C2H6 emissions in Weld County as compared to the ones in Boulder County with fracking is very interesting. I think it would also be very intresting to present some numbers here, at least a number for the order of magnitude of the difference.

The sentence has been modified to include numbers from all RDs:

Line 405: Using all 16 RDs, the median (minimum, maximum) VCDs in Boulder County and Weld County were 1.5 (0.5, 3.1) x 10^{16} molecules/cm^2, and 3.5 (1.0, 10) 10 x 10^{16} molecules/cm^2, respectively

Line 441: Because of the presented climatic dependency of the NH3 emissions, some indication would be helpful if the NH3 emission flux of 0.65 kg/km2 /hr suggested by the inventory is an average flux over the whole year or seasonally matches with the climate conditions during the conducted measurements. Maybe that could explain some additional contribution to the difference to the NH3 flux found in this study.

The information about the emission inventory was actually included in the original manuscript in lines 430-434. However, we have found a mistake in extracting the emission flux from the emission inventory that will be corrected in the revised manuscript. The peak emission flux comes from 20 UTC and is 13 % greater than the current reported 0.65 kg/km2/hr, and the 16-22 UTC emission average is 9 % greater. These changes are minor, and do not alter previous conclusions about the NEI11 comparisons. We have clarified the text to read: "[…] the emissions for July 2011 are given as hourly intervals, and do not distinguish between different source sectors. […] For Figure S4 the maximum emission flux from the diurnal profile in NEI 2011 was extracted (corresponds to 20 UTC, 0.73 kg/km2/hr), in order to make a conservative comparison with the measured emission rates. For comparison, the 16-22 UTC average NEI 2011 emission flux is 0.71 kg/km2/hr."

Line 492: I assume that the average C2H6 emission for site 1 should be presented here. In this case the uncertainty would be 19 kg/hr instead of 29 kg/hr.

We presented the average C2H6 emission with the propagated error.

Line 496: Could the authors give some more details about what kind of leaks they refer to?

We did not investigate the origin of the leaks further, and thus prefer to keep this generic statement. We note that the spatial mapping has some potential to identify leaks, but the elevated background variability in C2H6 provides some limitation here.

Line 535: The variability length scale for C2H6 is presented here as smaller than ~6 km. But in contrast to the cases of NH3 and NO2, no horizontal resolutions of the respective satellites have been presented above. I would suggest to do that for the sake of consistancy and completeness.

As the authors did not find clear information on the footprints of ACE-FTS and MIPAS which retrieve C2H6, we chose not to further compare these satellites here.

Technical comments

The use of capital letters in names and designations should be more consistent. One example can be found in the title ”The CU Mobile Solar…”, where the word ”Mobile” in the title is written with a capital letter ”M”. Below, in the text body, it is used with a small letter ”m”. Another expample can be found in line 138, but for this case also in Table 2, ”…Center-to-Limb ~~d~~Darkening (CLD)…”.

Thank you. We follow the reviewer suggestion. The word “Mobile” is now written as “mobile” in the title, and we changed the latter to “center to limb darkening” to be consistent with other literature.

The authors still use parantheses a lot which sometimes disturbs the flow of reading. I suggest to only use them for introducing abrevations, for uncertainties or alike in combination with averages values, and of course for citations. Otherwise I suggest to think in the following way: is it important information, then it should be part of the sentence; if not, then omit it. For example cross-references to figures, tables and sections can often be seperated by comma signs, e.g. ”…, see Figure 5.”. In this context I also noticed the use of parantheses as an indicator for different cases. An example of this can be found in line 111 ”…and Ozone Monitoring Instrument (OMI, 13 x 24 km2) (NO2) (Boersma et al., 2009).”. Rephrasing and using ”…for NO2…” instead of ”…(NO2)…” and would make reading more fluent. Examples like this can be found thorughout the manuscript.

We have implemented this proposed change in embedding most information into the sentences.

Often values and units are not separated by a blank space especially in the case of percentages, like in line 358 ”… taken as 30%...”. This is incosistent throughout the manuscript. I suggest to consistently use a blank space, e.g. ” ”… taken as 30 %...”.

We have added blank spaces to be consistent as suggested.

Many citations of personal communications are given. The authors are encouraged to complement these citations with a date.

Thank you, we added the following information:

Line 500: […] has been measured as 18.4 % (Alan Fried, personal communication, 2014), 11 % (Amy Townsend-Small, personal communication, 2014), and 10 % (Terra Yacovitch, personal communication, 2014).

Line 94: Please consider ”… around ~~an area source~~ a source area …”

We implemented the change.

Lines 247 and 320-321: The authors repeatedly use RD10 and RD11 to specify the days of the measurements. Unfortunately these are first introduced in the caption of Table 5 but not in the text. The authors are invited to introduce RD10 and RD11 accordingly for giving some consideration to the reader.

See comment to first specific comment. Changed “RD11” in line 247 to “13 August 2014”.

Line 104: Please consider"…to characterize the VCD variability of NH3, NO2 and C2H6 ~~VCD variability~~ on the spatial …".

We changed the sentence as suggested.

Line 123: Please consider" …Figure 1 shows ~~the~~ that photons along…".

The sentence has been corrected.

Line 167 – 168: Please provide a date alongside with the URL, e.g. "…/waccm, seen at 10 July 2016)…".

We now include "accessed on 14 October 2016" after the URL.

Line 171: Did the authors mean "by a factor of 100 ~~over~~ around" or "above"?

The sentence now used "around".

Line 175: What is meant by "contrasting" here?

We plotted these two quantities against each other and think the word contrast describes this.

Line 209: Please consider to replace "Each morning before…" by "In the mornings before…".

The sentence has been updated.

Line231, Eq(2): Please consider a blank space or multplication sign between VCD and vec(F).

We added spaces into the equation.

Lines 249 – 250: "…the wind direction varies a maximal 180°" it cannot be more than ±180°, so i suggest "…the wind direction varies up to 180°".

We changed the sentence as suggested.

Line 261: For the sake of clearity I suggest "…tool to quantify trace gas variability over horizontal distance…".

We added "horizontal" as suggested.

Lines 314 – 315: I suggest "There was a 0.5 % change in the retreived NH3 VCD and ~~0.0~~ no change seen in the C2H6 VCD."

We changed it to "no change" as suggested.

Line 384: Please consider "…indicating that ~80 % of NOx is ~~visible~~ abundent in the form of NO2."

The sentence has been modified as suggested.

Line 492: Please consider "We determined that the average C2H6 … is 63 ± 29 19 kg/hr."

The error remains as is 29 kg/hr, determined as sqrt(11^2+27^2).

Figure 3: Please consider a legend. It is not clear which percentiles you expressed with the black boxes and whyskers in case of the individual mobile SOF measurements. Similar for the red and blue graphs

where I assume that you present the mean value with some standard deviation, possibly with some factor.

The figure description now includes the following information.

Figure description: Box and Whiskers represent median, 25th, 75th, 5th and 95th percentiles for every 15 min. The VCD uncertainty on the mobile SOF and NCAR measurements, in red and blue respectively, is given as the 1-sigma standard deviation.

Figure 5: The modulation efficiency for an elevation angle of 65^{o} seems to be cropped away due to its offset.

That is correct, and we decided to leave the figure as is since the cropped away 65^{o} data does not alter the message and instead could lead to confusion if the OPD goes past 1.8 cm in the figure.

Figure 8 panels a, b, d: The names of the places of the underlying map are very small and difficult to read. Maybe some orientation can be provided written manually on top of the maps. Alternatively, some longitude and latitude coordinates on some representative axes would do. Further, an increase of brightness of the underlying maps would help to highlight the VCD data.

Thank you. We have added labels on the graph to include points of orientation.

Figure 8 panel e: I don't know how I should interprete the vertical, grey line at about 17:33. Further, in panel 3e, the letters for the cardinal directions seem to be shifted a bit too far to the right.

We have corrected panel e such that the letters are visible and all grey lines represent a change in vehicle orientation.