

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 #3

General comments

This paper is describing a low resolution optical multichannel spectroscopic system for mobile column measurements using direct sun measurements in the infrared and UV/visible region. It includes a theoretical analysis of the infrared spectroscopic measurements and the ability to detect gases in the atmosphere. The paper also describe column measurements of NH₃, NO₂, and C₂H₆ and subsequent flux-estimates based on these measurements. It also includes a motivation of the importance to measure the above mentioned species. Even though solar infrared measurements have been used for decades to retrieve atmospheric columns of various species, within the network NDACC, Flux measurements using solar occultation technique has only been carried out by few researchers. Considering the large potential for such flux measurements, this paper provides a welcome and needed contribution to improving the common knowledge about this technique. The language is good and figure and tables clear. The paper gives a nice illustration how the solar occultation flux method can be applied and in particular focuses on its sensitivity towards measuring various species and in the application of ammonia measurements of cows. However, a few general points that need improvement are given below: First of all it is not stated so well what is the objective with this paper in terms of novelty and originality compared to what has been published elsewhere. The Solar occultation Flux method has been published in several scientific papers and reports, see papers below. It is also described in the US EPA handbook on Optical Remote Sensing for Measurement and Monitoring of Emissions Flux and it is also Best available technology in Europe for measurements of fugitive emissions, as part of the mineral oil BREF. It is also used operationally used to monitor fugitive emissions since more than 10 years. There is a short statement about the wind error in the conclusions and reference to earlier work but this could be improved. Even though the author has included some previous work the background description in the paper should be improved to better reflect work and applications that has already been carried out with the solar occultation flux method. Alos some more references to other work applying sky DOAS or direct sun DOAS for flux measurements should be included.

The instrument is similar to what has been used in other papers and also how the flux method is obtained. In my mind its originality therefore lies in the sensitivity calculation (structure functions) and in performing flux measurements of ammonia from cows with the per head analysis I think that what makes this papers unique should be much better stated in the introduction of the paper and also how the papers is organized.

Specific comments

Page Row 42 (Introduction): 1. Line 88: Include more background about how the solar occultation flux method is applied since more than 10 years fro measurements of fugitive emissions , NH₃ etc.

Thank you for the references. We certainly want to give credit where it is due, and in addition to Mellqvist et al. (2010) we now reference Johansson et al. (2014) and Kim et al. (2015). Despite 10 years of measurements there is little references in the peer reviewed literature. Most previous SOF work has focused on another topic (industrial complexes), which is not the topic of this work. There have been no previous mobile SOF measurements of NH₃, for example. This has now been noted in the introduction.

2. Line 115: Describe the focus of this papers better here

See our response to a short comment from Kang Sun. We have edited the ending of the last paragraph in the Introduction. It now reads as:

Previous satellite comparisons have used mobile in situ measurements of NH₃ to characterize the near surface NH₃ mixing ratio variability in the San Joaquin Valley in California, and compare with data from TES (Sun et al., 2015), and CrIS (Cross-track Infrared Sounder) satellites (Shephard and Cady-Pereira et al., 2015). To our knowledge there currently is no attempt to characterize the sub-satellite ground pixel variability using mobile VCD observations of NH₃ and C₂H₆. Mobile VCD measurements eliminate the need for assumptions about NH₃ and C₂H₆ vertical distributions.

3. Line 115: Strange language in sentence, pleas rephrase.

See our response to comment 2 above.

4. Line 130: I would like a brief (few sentences) explanation about the NO₂ system , since these measurements are carried out here.

See our response to reviewer #1 and #2. We have added a sentence on the precision and accuracy. For a detailed technical description of the NO₂ system, and comparison of the NO₂ VCDs with those measured by Car MAX-DOAS see Section 3.3: Field applications and Fig. S9 in Baidar et al.,2016.

5. Line 188: I would like a brief (few sentences) explanation about the NO₂ system , since these measurements are carried out here.

Same as comment #4.

6. Line 226: This appears to be somewhat difficult measurements with considerable risk for errors that you have not discussed. Please add something about this.

The authors have overcome these difficulties in conducting these measurements as is described in the text. We are unaware of similar characterizations for other SOF instruments. The results from our characterizations show a high quality of alignment, and no evidence for “errors” or bias impacting VCD measurements. This is transparent from the provided information.

7. Line 240: The usage of model wind when measuring close to cattle farms will add quite some uncertainties to the measurements. The author states a comparison between some wind measurements and the model at the lower layers, but in my experience this needs some more discussion and uncertainty of the errors.

We agree, and have expanded on our analysis of the error introduced by wind. See our detailed response to reviewer #1 for details.

8. Line 320: Define ONG storage tank

Thank you. In principle other components of the ONG production could also contribute. We have added "... (probably from the ONG storage tank).".

9. Line 354: Same as line 240. The fact that two consecutive days resulted in the same value does not prove anything about the uncertainty in the wind since there may be considerable systematic uncertainties. Please improve

See our response to reviewer #1.

10. Line 358: You claim 30%/sqrt(2) as error. Please explain. Appears small !

Same as comment #9.

11. Line 491: The section about ethane appears somewhat inconclusive. It needs improvement; I suggest by more comparison to other studies and better discussion about the uncertainty.

In comparison with NH₃, C₂H₆ has a longer atmospheric lifetime and therefore the background VCDs are much more enhanced. As discussed we had difficulties to determine emission fluxes for ethane and thus it may seem that this section is less conclusive than the section on NH₃.

Some References SOF method

Thank you for the references. The following sentence has been modified in the introduction:

Line 87: The Solar Occultation Flux (SOF) method [...] (Mellqvist et al., 2010; Johansson et al., 2014b; Kim et al., 2015; EPA Handbook 2011; European Commission 2015).

Line 88: The SOF method has been used on mobile platforms to measure ethene (Mellqvist et al., 2010; Johansson et al., 2014a; Johansson et al., 2014b, de Gouw et al., 2009), propane (Mellqvist et al., 2010; Johansson et al., 2014a; Johansson et al., 2014b), alkanes including C₂H₆ (Johansson et al., 2014b), C₂H₄ and C₃H₆ (Kim et al., 2011). Foy et al. (2007) used stationary SOF measurements of alkanes and NH₃ amongst others to evaluate a chemical transport model.

However, no reports of NH₃ currently exist using the SOF method on a mobile laboratory to quantify emission fluxes from agricultural sources.

Mellqvist, J., et al., (2010), Measurements of industrial emissions of alkenes in Texas using the solar occultation flux method, J. Geophys. Res., 115, D00F17, doi:10.1029/2008JD011682.

Included in the original paper

Johansson, J., et al., 2014a, Quantitative Measurements and Modeling of Industrial Formaldehyde Emissions in the Greater Houston Area during Campaigns in 2009 and 2011, Journal of Geophysical Research – Atmospheres, 2013JD020159R.

Added

Johansson, J. K. E., et al, (2014b), Emission measurements of alkenes, alkanes, SO₂, and NO₂ from stationary sources in Southeast Texas over a 5 year period using SOF and mobile DOAS, Journal of Geophysical Research, 118, doi:10.1002/2013JD020485.

Added

Johansson, J. K. E. et al. (2016), Optical remote sensing of industrial gas emission fluxes, Doctoral thesis, 162 pp, ISBN 978-81-7597-316-6, Chalmers University of Technology, Gothenburg.

We do not have access to this thesis. It does not seem to be available on the web.

Foy, B., et al., Modelling constraints on the emission inventory and on vertical dispersion for CO and SO₂ in the Mexico City Metropolitan Area using Solar FTIR and zenith sky UV spectroscopy, Atmos. Chem. Phys., 7, 781–801, 2007

Added

De Gouw, et al., Airborne Measurements of Ethene from Industrial Sources Using Laser Photo-Acoustic Spectroscopy, 2442 9 ENVIRONMENTAL SCIENCE & TECHNOLOGY / VOL. 43, NO. 7, 2009

Added

S.-W. Kim, et al., Evaluations of NO_x and highly reactive VOC emission inventories in Texas and their implications for ozone plume simulations during the Texas Air Quality Study 2006S, Atmos. Chem. Phys., 11, 11361–11386, 2011 NO₂ from stationary sources in Southeast Texas over a 5 year period using SOF and mobile DOAS, Journal of Geophysical Research, 118, doi:10.1002/2013JD020485

Added

European Commission, 2015, Best Available Techniques (BAT) Reference Document for the Refining of Mineral Oil and Gas: Joint Research Centre, Institute for Prospective Technological Studies, ISBN 978-92-79-46198-9 (PDF)ISSN 1831-9424 (online)doi:10.2791/010758, http://eippcb.jrc.ec.europa.eu/reference/BREF/REF_BREF_2015.pdf

Added

EPA Handbook: Optical Remote Sensing for Measurement and Monitoring of Emissions Flux, December 2011, Office of Air Quality Planning and Standards , Air Quality Analysis Division Measurement Technology Group , Research Triangle, North Carolina, 27711, editor Dennis K. Mikael, <http://www.epa.gov/ttn/emc/guidlnd/gd-052.pdf>

Added