

The Reviewers provided very constructive comments on the paper (Atmos. Meas. Tech. Discuss., 8, C3214–C3216, 2015), so we would first like to thank them for their efforts in improving the manuscript. The following are detailed responses to comments from Reviewer 2.

Anonymous Referee #2

The paper covers a very relevant subject with the validation of satellite observations of four important species by using vertically resolved observations. There are only a few studies showing vertically resolved observations of ammonia, and this study is one of the first to use the observations for the validation of satellite observed concentrations. The paper is easy to read and well structured. Both the satellite and airborne observations are explained well in the first paragraphs after which the authors go into more detail about the difference between the observations and possible causes. After the section covering the validation the authors show the usefulness of the observations by comparing them with modelling results from the GEM-MACH model which covers the local region surrounding the oil sands.

A few minor comments and edits:

1. The title reflects the validation effort but does not cover the modelling aspect of the paper.

We agree. The title has been changed to: “Tropospheric Emission Spectrometer (TES) observations of Ammonia, Methanol, Formic Acid, and Carbon Monoxide over the Canadian Oil Sands: Validation and Model Evaluation”

2. In general the validation is the strongest part of the paper and the comparison with the model, though being a good showcase of the use of the observations, feels like a separate paper/a bit of a distraction of the main message. The GEM-MACH is only mentioned in the last lines of the introduction. A small piece of extra explanation about the model and use would be helpful and improve the connection between the modelling and validation parts of the paper.

This is another good point. We included the model comparisons both as an example of the value of validated satellite observations (as pointed out by the Reviewer), plus we wanted to highlight the model comparisons to show the current status of the new GEM-MACH special oil sands observations as they are being used in the special JOSM project. This will also provide motivation for further analysis of NH₃. In the introduction we added, “In addition, GEM-MACH is also being used by Environment Canada to provide ongoing experimental air-quality forecasts at 2.5km resolution for a domain covering Alberta and Saskatchewan. The model is also being used for human health and acidifying deposition impacts of oil sands sources – hence obtaining accurate emissions of these trace gases are essential for the success of the model simulations., Here we demonstrate the satellite’s potential value in evaluating the model performance for these trace gases.”

3. In the text it is mentioned that the response time of the QCL system is 60 seconds. Is this not too slow with the airplane moving quickly over possible source areas and NH₃ concentrations varying quickly? And what would the possible bias be to the measured concentrations?

Yes, a 60 second response time is relatively slow compared to other gas measurements with the QCL. The reason being that NH₃ is very sticky and will stick to any surface on its way from the inlet outside the aircraft to the instrument (several meters of tubing, various valves etc.). All these surfaces have to approach an equilibrium with the air stream before the concentration reading inside the instrument will stabilize. That being said, this should not be important in regards to the profiles as: (i) The slow response time is due to a smearing-out of the signal from the inlet to the instrument. The integrated signal “under the curve” is still representative of the amount of NH₃ entering the system. In other words, any higher-frequency spikes are smoothed out but still represented; (ii) The satellite also integrates over a larger area. Therefore, resolving individual sharper spikes is not really important for the comparison.

4. All airborne observations within 120 kilometers are being compared with the TES pixels. What is the expected effect (a misrepresentation error) on the concentrations of the individual species? For example ammonia has a lifetime of hours up to a day. With low wind speeds it can take several hours for a plume of ammonia to bridge the 120km. Did you analyze the effect of a stricter spatial criteria?

This is a good question, and is related to questions 1, 2, and 3 by Reviewer 1 (please refer to these responses for details). The main point is that there is surprisingly little gradient observed during these flights, both temporally and spatially. Thus, we see very little difference applying different spatial criteria (up to including all the possible TES pixels).

5. As for the altitude comparisons – The differences between the observations are shown in the form of distributions at different pressure levels. It might improve the comparison by seeing also showing a figure with the airborne and TES observations in a simple scatter plot (maybe color coded with the different levels).

Plotting the values themselves in a scatter plot instead of showing just the differences is certainly a valid thing to do. However, we are reluctant to add more plots to each of the 4 species as there are already 15 figures in the paper, plus we do provide the TES magnitude values with the differences so the aircraft values can be obtained.

6. GEM-MACH evaluation: As mentioned fire and other natural emission sources are not included. However the timing of the observations is more or less in the middle of the Canadian fire season. Fires are one of if not the largest source of NH₃ in the north and transport of NH₃ in fire plumes is possible. NH₃ is emitted with a certain ratio to CO and both CO and NH₃ are underestimated. Will the fire emissions be included in the model in the future and if transport brings the plume this far down might it explain the difference between the model and observations? (I am not asking to include it now, just curious about the authors opinion).

Yes, we are going to include biomass burning in future simulations. Another component of this is that even though there are no “nearby” forest fires at the exact time of the observations there might be some lower concentration transport (nothing large as we don’t see it in the CO observations). The other related point is that there could have been some forest fires in the recent past that would have deposited ammonia into the soil/vegetation, which can then be released to the atmosphere later via bi-directional flux. As GEOS-Chem already has bi-directional

flux already implemented, we are presently setting up some new GEOS-Chem simulations that will have both biomass burning emissions and bi-direction flux capability so we will be able to study both the direct and indirect contributions of forest fires on the “background” ammonia in this region.

Edits:

Figure 1. Missing a ruler and indication of lat/lon position/grid.

We agree that there is not geolocation information provided. Since the plot is just for relative comparisons between the aircraft and the satellite we added that TES transect is ~240km in length to provide a geo context. Changed the Figure 1 caption to “...TES transect 5x8 km pixels (black polygons) spanning a total distance of ~240-km overplotted on Google Earth images.

Figure 14. Not seeing the Model (orig) Hidden behind a line?

Yes, we also noticed this in the last revision and added the to the caption, “...(note since the model gray line is smooth it is obscured by the mapped blue line on the plot).”

Table 1. Missing CO in the Table, estimates are given in the text, might be good as a summary.

CO was intentionally left out in the original table as we were highlighting the estimated errors from previous simulations, and CO has already been validated using observations, but we agree with the Reviewer that it would be good to include it in the table and added the following row.

CO	681	100-150	0 to -10%	~±10 to ±20%	Actual errors derived from comparisons with observations (Luo et al., 2007a, 2007b; Lopez et al., 2008)
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Page 9505 Line 7: CO is missing

We understand where you are coming from, but actually CO was left out purposefully in the sentence, “This study provides one of the first direct validations of Tropospheric Emission Spectrometer (TES) satellite retrieved profiles of NH₃, CH₃OH, and HCOOH through comparisons with coincident aircraft profiles.”, as CO direct validation have previously been performed, unlike NH₃, CH₃OH, and HCOOH.

Page 9511, Line 11, End of line “.” needed in between 2013 and These

Agreed and was corrected as suggested.

Page 9531 line 1, add Carbon Monoxide to the sentence

Good catch. “carbon monoxide” was added to the sentence as suggested.