

Shoma Yamanouchi et al.

General Revisions:

We thank the two reviewers for their helpful comments, which have enabled us to improve the manuscript. The reviewers' comments are in regular font below and our responses are in bold font. Line numbers in the responses refer to the revised manuscript with changes tracked. Also of note is that there was a minor bug in the trend analysis code; this was revised, and affected values were corrected (this only affected the 2σ confidence intervals from bootstrap resampling).

Reviewer 1

The approach to determine the observational footprint of the FTIR column measurements seems to be oversimplified. It is only based in correlating the data with the satellite observations at different spatial and temporal scales. The best correlation and slope is obtained with the most strict criteria (25km/20 min). A proper footprint analysis would require to take the wind fields within the considered time period in consideration, which is not done. Although this simple analysis gives some indication of the representativeness of ground-based measurement, it should not be claimed in the text that a proper observational footprint assessment has been performed.

All sentences claiming that the “footprint assessment” was performed were replaced by sentences that mention the representativeness of ground-based measurements.

A bias would be expected to be observed between the FTIR and in situ data just because the FTIR only measures during sunny conditions. NAPS data is collected regularly every third day. Moreover, NH₃ has a strong diurnal pattern that is not reported in this paper. While in situ data represents the average concentration within a 24 h period, FTIR data is available only during the day.

A brief discussion of this bias was added (Line 248).

The authors contrast the trends from the linear regressions from both data sets (TAO and NAPS) when outliers are and are not considered (L204). However, no mention or explanation is given for this source of bias given that NH₃ concentrations are probably expected to peak during warmer days and warmer hours. It would be interesting to compare both data sets only for coincident

measurement days and give a more comprehensive explanation of this additional source of bias.

The comparison analysis using only coincident measurements is shown in Figure 5a. A brief discussion on warmer days and higher NH₃ was added, along with an additional analysis to examine coincident FTIR and in-situ measurements and temperatures; on three occasions where simultaneous enhancements were observed in the FTIR and in-situ data (once in May 2014, twice in May 2016), the daily average temperatures were higher than the monthly averages (Line 248-259).

It seems that the comparison of both TAO and IASI data sets with GEOS-Chem is challenging due to the coarse resolution of the model. It is shown from the comparison of the ground-based data with the satellite observations that NH₃ presents high frequency variability in the region. It would then seem logical that the authors filter out the enhancements from the FTIR data, as done in the trend analysis, before correlating to the model data. The same could apply to IASI data since the enhancements observed within the large model domain are probably due to local emissions that are not well represented by the model. Figures 9a and b could then show the correlation and regression results as is, as well as from the filtered data sets.

This analysis was performed. Filtered FTIR measurements compared with GEOS-Chem resulted in $r^2 = 0.22$ and slope = 0.68 (when no filtering was performed, the values were $r^2 = 0.26$ and slope = 1.16). Comparisons of filtered IASI observations and GEOS-Chem resulted in $r^2 = 0.29$ and slope = 0.57 (when no filtering was performed, the values were $r^2 = 0.33$ and slope = 0.85). Corresponding plots were added (Figures 9c and 9d).

L28. The sentence is not accurate. The health impact of PM_{2.5} is strongly dependent on the chemical composition and the cited study does not take composition into account. In the context of this contribution, the PM containing ammonium salts are not the most hazardous and also those that contribute to smog are rather organic in nature. Please rephrase.

The sentence was reworded and another reference added here (Schiferl et al., 2014). We are not claiming that particulate matter forming due to ammonium salts are the most hazardous. Additionally, recent studies (e.g., Liu et al., 2019; Wielgosiński & Czerwińska, 2020) have shown that ammonium salts do contribute to smog as well as haze. The sentence was also reworded clarify this (Line 29-34).

L41. Referring to NH₃ being injected to the free troposphere, you may want to cite Hoepfner et al 2016 (www.atmos-chem-phys.net/16/14357/2016/)

The reference was added (Line 48).

L86. A citation or description for the camera and solar disk-fitting system of the solar tracker is missing.

Further details can be found in Franklin (2015, <http://hdl.handle.net/10222/64642>). This reference was added.

L90 Should say "... microwindows in the ... and ... spectral regions."

Fixed (Line 98-99).

L76. Was there any quality control and data filtering performed? Please describe. Same for the in situ data.

No filtering was done for the in-situ (NAPS) data, although all NAPS sites adhere to quality control/quality assurance guidelines set forth by the Canadian Council of Ministers of the Environment (see https://www.ccme.ca/files/Resources/air/Ambient%20Air%20Monitoring%20and%20QA-QC%20Guidelines_en%20SECURE.pdf for details). FTIR columns were retrieved to conform to NDACC standards. Archived species are filtered by RMS/DOFS ratio.

L110. No need to repeat (National Air Pollution Surveillance Program)

The repeated bit was a part of the citation for the data (link to an entry references section). This has been removed (Line 119).

L117. Define the IASI acronym.

This was previously defined in the introduction (Line 73).

L121. May not be clear to the reader what a 2 x 2 circular pixel is. Maybe a matrix of 2 x 2 pixels?

The sentence was replaced with "[a]t nadir, the field of view is a 2 x 2 matrix of pixels, each with a 12 km diameter (Clerbaux et al., 2009)" (Line 132-133).

L126. Indicate the overpass times of each satellite instrument

The 3 IASI instruments are onboard the Metop A, B and C satellites which are all in the same polar orbit. Measurements are then performed at 09:30 and 21:30 mean local solar time for the descending and ascending orbits. A sentence clarifying this was added (Line 128-130).

L155 What do "longer time series" refer to? The length considered in this contribution? Please specify.

This refers to the duration of the measured dataset. In the method outlined by Weatherhead et al. (1998), measurements that are highly auto-correlated require longer time periods to obtain trends (for any given confidence interval).

L165. If mirroring a value is the same as taking its absolute value, the readers might be more familiar with the second terminology. It may also be wise to mention that the average of the mirrored residuals include the positive ones.

The term "mirroring" was used here, as it was also used by Zellweger et al. (2009). The argument for using this terminology is that the residuals should have both negative and positive terms, and in this analysis, the positive ones were "replaced" by the absolute values of the negative ones. The positive residuals are not used, in order to reduce biases introduced by enhancements.

Fig4. Figure 4 b) seems redundant since no additional information is provided with respect to a).

This figure was included to better illustrate points made around line 237.

Fig5. It would seem sufficient to show the correlation plots a) and e) in this figure, while keeping the results of the different resampling periods in the text (L219-223) L300. A larger trend with outliers with respect to that obtained without them may not be conclusive when looking at the data availability of the TOA data series. Measurements seem to be performed more regularly in recent years so to me the increase in seasonal variability is more evident when comparing for example the standard deviations year to year.

The standard deviations of the TAO columns are in fact increasing (as discussed in Section 3.1). The conclusion was edited to re-iterate this point (Line 338).