**Comments to the author**:
Dear Amir Souri,

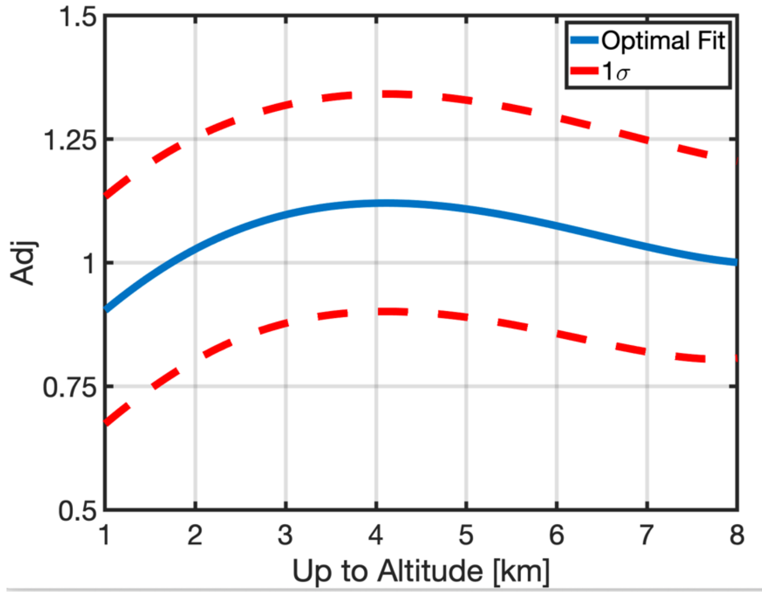
I'm pleased to accept your revised manuscript "Characterization of Errors in Satellite-based HCHO/NO2 Tropospheric Column Ratios with Respect to Chemistry, Column to PBL Translation, Spatial Representation, and Retrieval Uncertainties" for ACP subject to minor revisions as explained below.

The detailed replies to the comments made during the review are appreciated, and this discussion will be a valuable resource for the interested readers.

| Answer |
| --- |
| **We thank the editor for carefully reading our manuscript/replies and helping us bring our draft to a higher standard.** |

One aspect of the results remains confusing to me: In Figure 6, you show a correction factor between the columnar HCHO/NO2 ratio and the corresponding ratio for a column from the surface up to a certain height. For the lowest kilometre, this correction factor has a value of 0.6 However, when looking at Figure 5, I do not see any evidence in the median values for the need of such a factor. With the exception of the values in the highest layers (which have only a very small contribution to the columnar ratio observed from satellite, the ratio appears very stable between 4 and 6. Please explain why the correction factor is so large close to the surface.

| Answer |
| --- |
| **We are grateful for this comment because we realized that there was indeed a missing statement in our code to compute the adjustment factor. Although the violin plots represent the afternoon observations, we forgot to apply such conditions when calculating the centroids. As a result, the adjustment factor wrongly considered both morning and afternoon times. The gradients in the ratio tend to be steeper in the morning times (from 2 close to the surface to 8 in 5-6 km) resulting in smaller adjustment factors for areas close to the surface (See Jin et al., 2017). The new adjustment factor fluctuates closer to 1.0. Please note that we fit the second-order rational functions to the 25$^{th}$ and 75$^{th}$ percentiles to create the bounding polygon used for the centroid calculation.** |




**Because the contrast between morning and afternoon results is interesting, we decided to add new figures to the SI regarding this tendency.**

**Modifications**

**We replaced Figure 6 and Figure S12. We now see 19% standard deviation around the optimal fit as opposed to 27% error in the previous results (computed as the average of the deviation of the 1-sigma from the optimal fit throughout the altitudes). Therefore, we recreated Figure 13, 14, 15, and Figure S16. These changes did not impact the conclusion of our study.**

**We modified the error:**

"The standard error deviation of this conversion is around 19%."

"This data-driven adjustment factor exclusively derived from afternoon aircraft profiles during warm seasons in non-convective conditions had a standard error of 19%."

**We recalibrated the empirical equation to represent the adjustment factor:**

It is beneficial to model this curve to make this data-driven conversion easier for future applications. A second-order polynomial can well describe ($R^2$=0.97) this curve:

$$f_{\text{adj}} = az_t^2 + bz_t + c \qquad a = -0.01, b = 0.15, c = 0.78 \qquad (11)$$

**We added in the results and discussion:**

"The relatively low fluctuations in the adjustment factor around one suggest that under the observed atmospheric conditions (clear-sky afternoon summers), the columnar tropospheric ratios do not poorly represent the chemical conditions in the PBL region."

**We added new figures to the supplemental material:**

A caveat with these results is that our analysis is limited to afternoon observations because we focus on afternoon low-orbiting sensors such as OMI and TROPOMI. Nonetheless, Schroeder et al. (2017) and Crawford et al. (2021) observed large diurnal variability in these profiles due to diurnal variability in sinks and sources of $NO_2$ and HCHO and atmospheric dynamics. The diurnal cycle has an important implication for geostationary satellites such as Tropospheric Emissions indeed: Monitoring of Pollution (TEMPO) (Chance et al., 2019). Limiting the observations to morning time results in a smaller adjustment factor for altitudes close to the surface resulting from steeper vertical gradients of $HCHO/NO_2$ (Figures S13 and S14). This tendency agrees with Jin et al. (2017), who observed a large deviation from one in an adjustment factor used for the column-surface conversion in winter.

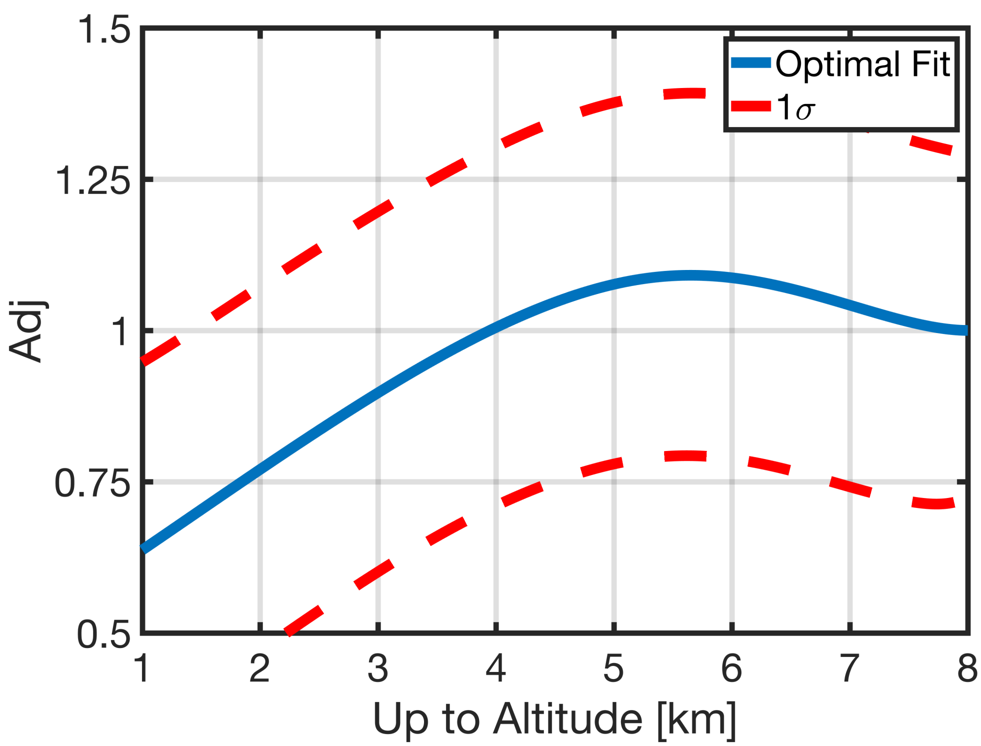



Figure S13. The adjustment factor defined as the ratio of the centroid of the polygon bounding 25$^{th}$ and 75$^{th}$ percentiles of the observed $HCHO/NO_2$ columns by the NASA aircraft between the surface to 8 km to the ones between the surface and a desired altitude. This factor can be easily applied to the observed $HCHO/NO_2$ columns to translate the value to a desired altitude streteching down to the surface (i.e., PBLH). Only observations made during morning are used.

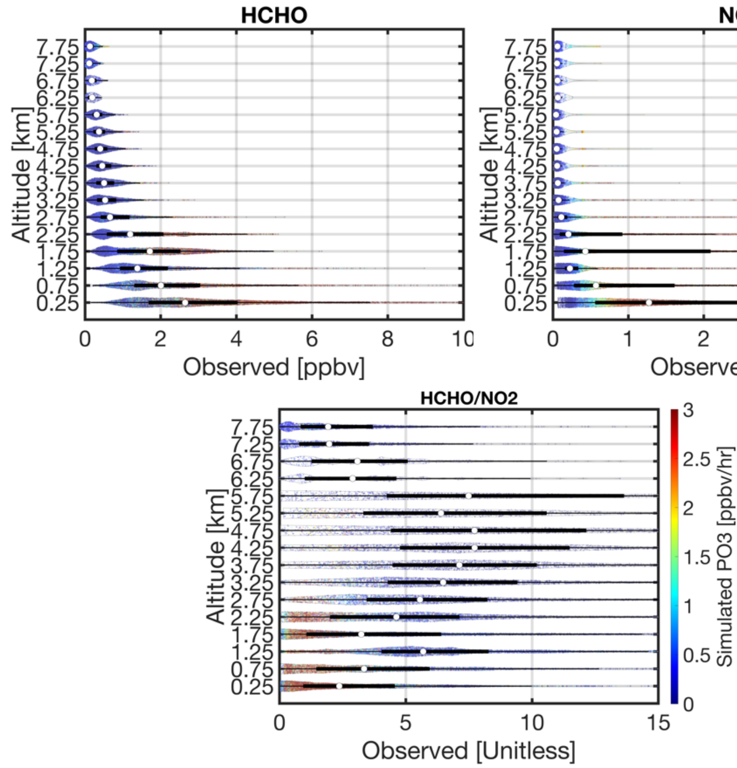



Figure S14. The violin plots of the morning vertical distribution of HCHO, NO2, and HCHO/NO2 observations were collected during DISCOVER-AQ Texas, Colorado, Maryland, and KORUS-AQ campaigns. The violin plots demonstrate the data distribution (i.e., a wider width means a higher frequency). White dots show the median. A solid black line shows both the 25th and 75th percentiles. The heatmap denotes the simulated ozone production rates.

**In conclusion we added:**

This behavior means that the ozone regime tends to get pushed slightly towards the VOC-sensitive regime near the surface for a given tropospheric columnar ratio. This tendency was more pronounced in morning times when the non-linear shape of FNRs was stronger.

In the discussion of the larger scatter of the HCHO values, I do not fully agree with your formulation on the absorption strengths of HCHO and NO2. The differential absorption which is used in DOAS is actually larger for HCHO than for NO2 in the UV. The NO2 differential absorption cross-section is much larger at visible wavelengths which is why NO2 retrievals usually are performed at wavelengths > 400 nm. An additional important factor is the smaller radiance and thus larger photon shot noise in the UV.

| Answer |
| --- |
| **We agree that for very small short wavelengths, the HCHO optical depth can become stronger than NO2; but we meant in the wider range of UV-ViS range, the optical depth of NO2 is much stronger. We agree that SNRs are larger for higher wavelengths providing higher fidelity information for $NO_2$ retrievals compared to HCHO.** |
| **Modifications** |

**We rewrote the sentence to:** "The distribution is slightly broader compared to that of $NO_2$, manifested in a larger standard deviation $4.32\times10^{15}$ molec./$cm^2$. This is primarily due to two facts: i) HCHO optical depths generally peak in the UV range (<380 nm), where the large optical depths of ozone and Rayleigh scattering result in weaker and noisier signals (Gonzalez Abad et al., 2019), and ii) the broader and stronger $NO_2$ optical depths in the ViS range (400-500 nm), where the signal-to-noise ratio is typically more outstanding, permit better quality retrievals."

In Figure 7, lowest panel I'm wondering what exactly is shown here - is that the mean of the 90 individual ratios, or is it the ratio of the mean values? Please clarify in the figure caption.

| Answer |
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| **It is the ratio of the mean values shown in other panels. We do not necessary have 90 ratios as clouds can substantially reduce the number of samples within the time frame.** |
| **Modifications** |
| **We added:**<br><br>"Figure 7. Oversampled TROPOMI total HCHO columns (top), tropospheric $NO_2$ columns (middle), and the ratio (bottom) at $3\times3$ $km^2$ from June till August 2021 over the US. The ratio map is derived from the averaged maps shown in the top and middle panels." |

There still are many sentences in the manuscript which to me as a non-native speaker appear to have language issues. I have marked some of them in the attached file; please check again.

| Answer |
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| **We gave the draft a very good read and adjusted poor grammar.** |

Best regards,

Andreas Richter