Dear Editor,

Referee #3's comments are in italic and our answers to all comments of reviewer #1 are embedded in red. **Bold text** are manuscript additions and ~~strikethrough text~~ are manuscript deletions.

This manuscript does a very nice job evaluating NO2 and HONO from the Sheridan fire plume during FIREX-AQ using measurements from GCAS. This manuscript is worthy of publication after some minor revisions.

Major comments:

It would be helpful if the authors could provide a better motivation for Section 3.3. I am a bit confused. To be clear, I fully understand why there's a need to better understand uncertainties in the EMG method and/or develop a better method, but it's unclear to me exactly how Section 3.3 is doing this. Additional clarification is needed.

Please see a more detailed response about motivation for Section 3.3 below.

Errors in the wind speed and direction are a likely contributor to the errors in the EMG fit. From Figure 1 it seems that the rotated plume is not perfectly horizontal, partially due to meandering winds. This will lead to an artificial shortening of the derived lifetimes. In my more detailed comments, I suggested ways to look at this uncertainty.

We agree that plumes that are not perfectly horizontal will lead to artificially shorter derived lifetimes. Please see a more detailed response further below.

Also, some more discussion on the aerosol effects on the GCAS retrieval would be helpful. See the Introduction of Cooper et al., 2019 (https://agupubs.onlinelibrary.wiley.com/doi/pdf/10.1029/2019GL083673) for a great discussion on this. This is especially relevant for Section 3.4 when comparing to inventories. I am guessing that the shielding effect (aerosols "shielding" the sunlight from NO2 at lower layers) is more prominent than the enhancement effect (aerosols "enhancing" sensitivity to the GCAS measurement above the wildfire plume layer), and therefore would consistently underestimate NO2 and HONO emissions, but this is merely a hypothesis.

A discussion of the aerosol effects on the GCAS retrieval is described in more detail below. We thank the reviewer for providing an example of an aerosol discussion.

More detailed comments are below:

Line 87. What was specific altitude of the ER-2? It's relevant because it's important to know how much of the upper troposphere and lower stratosphere is being observed by GCAS. Also can you briefly comment here or elsewhere in the manuscript regarding how much NO2 and HONO is in the lower stratosphere that could have been observed by GCAS?

The specific altitude of the ER-2 was roughly 20.35 km during the sampling of the Sheridan Fire, so the aircraft was flying in the lower stratosphere. However, as our paper presents the enhanced HONO and NO_2 VCDs by removing the upwind background VCDs, and not the absolute HONO and NO_2 VCDs, the stratospheric component is removed.

The text has been amended as below:

"On this same day, the ER-2 aircraft flew over the fire in such a way to create a sweeping bowtie pattern **from an approximate altitude of 20.35 km**, where the fire plume was captured on the downwind side of the bowtie and the background air was captured on the upwind side of the bowtie…

We then subtracted from the entire scene the average HONO and NO_2 VCDs upwind of the fire**, called the background HONO and NO_2 VCDs, resulting in enhanced HONO and NO_2 VCDs solely from the wildfire. This process also removes any stratospheric component of HONO and NO_2**."

Line 133. What is the 588 mb pressure level referring to: the plane altitude or the mean plume height or both or neither? Can you clarify? And if it's not the mean plume height, what was the mean plume height? And did this vary by time of day?

The 588 mb pressure level refers to the plane altitude, but is also an approximation of the mean plume height. The DC8 performed a flight track above the plume, flying along the plume's length and back just after 17 August 2019 0 UTC. The DIAL instrument captured the plume at an altitude of roughly 4700 m. When the DC8 made cross-sectional tracks of the plume, its flight altitude was 588 mb. As the DC8 flew only two lengthwise tracks during the Sheridan Fire, we cannot determine how plume height varied by time of day,

To clarify the main text, we have amended it as follows:

"Altitude was converted to a pressure level by recording the DC-8 aircraft data at a time when the DC-8 was flying through the smoke plume**. The DC-8 plane's altitude and the plume's altitude were approximated to be at**~~, which resulted in~~ a pressure level of 588 mb."

Figure 1. As a sensitivity study, it may worthwhile to artificially rotate the plume even further, maybe another 20 degree clockwise to see what effect that has. It does seem that the plume meandered a bit, from southwesterly flow initially to more westerly flow 20 km downwind. I don't know if it's possible, but I would recommend rotating the 0-10 km section of the plume differently than the 10-60 km section of the plume. I think this may be a partial cause as to why the lifetimes from the EMG method are underestimated as discussed in Lines 345-350.

As mentioned above, rotating the plume even further will shorten the lifetimes due to a compressed line density, and enhance the emission rates since more emissions will be present in a compressed line density. In theory, there should be a wind direction that results in the maximum lifetime and minimum emission rate. This may prove to be a better wind direction estimate, however this method is not perfect either, namely, that nonlinear plumes will not have a clear, best rotation angle based on the wind direction.

We agree that our wind rotation method is not perfect and is made useless under situations where a wildfire smoke plume is not linear, and we acknowledge this limitation in Line 463 of the original submittal.

We do not think that the underestimation of lifetimes as discussed in Lines 345-350 is due to the wind direction. In this idealized 1-D model, there is only one direction for the emissions to go. In theory, the lifetimes should be at their peak characterization. However, it is because of the fast-shifting emission rates that the EMG fits underestimate the lifetime.

Lines 246 - 249. I'm not following the 3rd and 4th configurations. Is the motivation of this to better quantify the pulsing nature of wildfires? Or something else? One or two more sentences motivating these four sensitivity studies could be very helpful.

To clarify the motivations between each model configuration, the following text has been added to the manuscript:

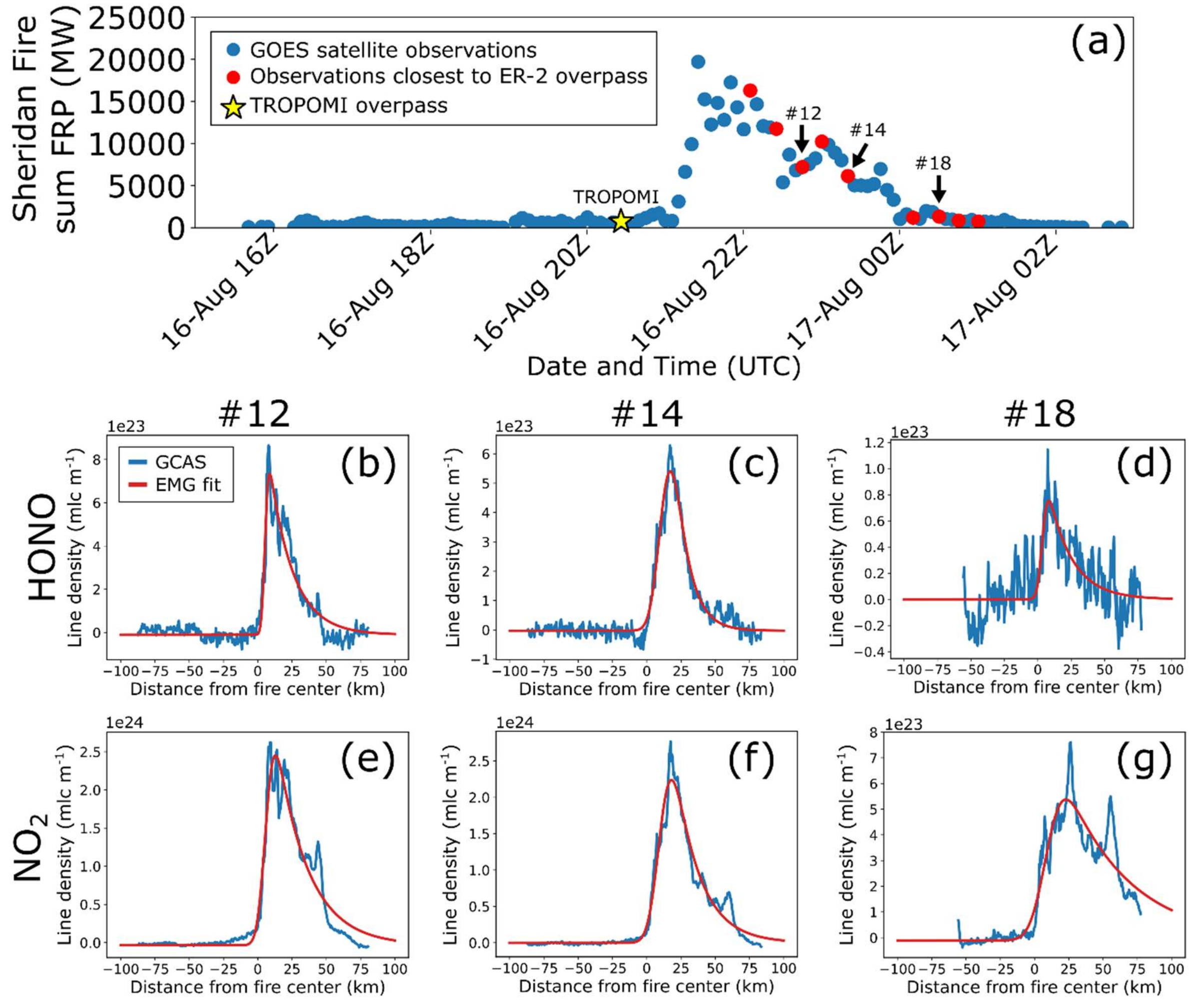
"In the first configuration, the emissions rate is kept constant for the entire model**ed** run time **and is the simplest model configuration**. In the second configuration, the emissions rate has a step-change halfway through the model**ed** run time, **adding complexity to the modeled emissions rate**. In the third configuration, the emissions rate is multiplied by the probability density function of a Gaussian with a mean of 5,000 s and standard deviation of 1,000 s**, as daily fire activity has been modeled with a Gaussian distribution previously (Andela et al., 2015)**. Finally, in the fourth configuration, the emissions rate is prescribed such that for every time step, the **total daily emission** rate is multiplied by the fractional FRP**, representing a data assimilation scenario since emission rates have been modeled as functions of FRP**."

The following references were added to the References section:

"**Andela, N., Kaiser, J. W., Van Der Werf, G. R., and Wooster, M. J.: New fire diurnal cycle characterizations to improve fire radiative energy assessments made from MODIS observations, Atmos. Chem. Phys., 15, 8831–8846, https://doi.org/10.5194/acp-15-8831-2015, 2015.**"

Figure 3a. Can you clarify x-axis to be 16-Aug 16Z, etc. It's not intuitive that the second number is the hour.

We have edited Fig. 3a as requested by the reviewer. The new figure within the manuscript is shown below:



Lines 319 - 351. I am not fully following Figure 5 and the associated analysis. I think more detail motivating this section is needed near Line 319. I think you are trying to better understand how the EMG method could better capture the pulsing nature of wildfires, but that's not explicitly said, so maybe I am misinterpreting. Also see comments regarding Lines 246 - 249 which are contributing to my confusion. The times (200 s 5000 s, etc.) are also confusing to me. Are you referring to how much time it takes on your local computer to do the analysis? Can you better define what these mean?

We have provided text to further motivate our idealized simulations in Section 3.3. That text is provided below:

"We found evidence of this behavior in Fig. 3e. **Additionally, if a fire subsides and dies out, but the plume is still being transported, the EMG fit would still assign a substantial emission rate to the fire. The EMG method in this situation disconnects the fire from the plume, as seen in Fig. 3g. These situations are complex and nonideal, and the EMG method is not suited for them. This then begs the questions of what situations the EMG method is suited for; how long a fire needs to burn before the EMG method provides an accurate result; how quickly the EMG method responds to a change in fire emissions; and how the EMG method stands against a simplified diurnal emission profile.**"

Additionally, we have realized that instead of model run time, we meant modeled run time, the virtual time within the simulations themselves. We have corrected all instances of “model run time” to “modeled run time” where appropriate. The first instance of modeled run time has been amended as below:

“In the first configuration, the emissions rate is kept constant for the entire model**ed** run time **and is the simplest model configuration.**”

Line 436. A discussion of any potential systematic biases of your MC diurnal 1-D method is warranted here. In particular, the effects of aerosols on the retrievals should be discussed. I am guessing the aerosols could be causing an underestimate in the GCAS retrieval, which would in turn cause derived emissions to be too small. Something to think about and include as discussion.

We have added the following brief discussion about aerosol impacts at the end of section 3.4:

“**…As discussed earlier, the VCD retrievals from the GCAS instrument are subject to uncertainties, primarily uncertainties in the AMF determination. One major source of AMF uncertainty is the presence of and the characteristics of aerosols in the retrieval columns (Cooper et al., 2019). Aerosols can increase the AMF through scattering; scattering can increase the light path or increase the radiance observed by remote sensing instruments. Aerosols can also decrease the AMF by shielding a compound below a layer from remote sensing instruments or absorbing aerosols can reduce the scattering back towards remote sensing instruments. Wildfire smoke is made up of both scattering and absorbing aerosols and can be present as dilute or concentrated plumes. With both HONO and NO_2 enhancements coinciding within the aerosol layer, we hypothesize that aerosols both shield HONO and NO_2 deeper within the plume and absorb sunlight. This will likely lead to an underestimation of HONO and NO_2 concentrations and therefore an underestimation of emission rates. Thus, the actual emissions may be more on par with the QFED and GFAS inventories…**”

The following references were added to the References section:

“**Cooper, M. J., Martin, R. V., Hammer, M. S., and McLinden, C. A.: An Observation-Based Correction for Aerosol Effects on Nitrogen Dioxide Column Retrievals Using the Absorbing Aerosol Index, Geophysical Research Letters, 46, 8442–8452, https://doi.org/10.1029/2019GL083673, 2019.**”