Response to Reviewer #2

We thank the reviewer for their detailed comments on the manuscript. We have addressed these comments as described below. All reviewer comments are presented in italic font while the author responses are displayed in standard font. Specific text that was added to the updated manuscript is provided in blue text.

General comments:

The paper is intended to describe validation of UV, IR, and UV+IR ozone retrievals based on TROPOMI and CRIS data and the TOPAS retrieval algorithm. The validation data sets are TOLNET (lidar) and ozone-sondes. I have three main concerns with this paper. Firstly, it is not clear how this paper contributes to our understanding of the TOPAS CRIS/TROPOMI ozone retrievals over a previous paper (essentially from the same group) by Mettig et al. (AMT 2022) as well as un-cited work by Malina et al (AMTD). The paper indicates that they use the "full capabilities" of the TOLNET data sets but I could not find what this means or how it advances the validation of these retrievals relative to what is describe in Mettig et al. The paper suggests that a key result is that using UV+IR radiances improves the ozone retrievals over use of UV or IR radiances alone; however this result is already well known and well described in other papers (some cited, some not). My second main concern is that while the paper is well organized, it is poorly written with numerous non-quantitative statements, and with essentially no context, introduction, or comparisons to other work within most of the results component of the paper. Thirdly, much of the paper appears to be repetitive with respect to the Mettig et al. paper.

To get through review, I would ask that the authors spend more effort in describing what is new and different about this paper relative to the Mettig et al. paper; likely this would also help in shortening the paper as material that appears in Mettig et al. need not be restated in the submitted manuscript. Adding context to each of the results sub sections and how what is presented is similar/different to previous work would also improve the writing.

We thank the reviewer for their suggestions on how to improve our manuscript. We have made substantial changes to the text in the revised manuscript to address these concerns. The main improvements are listed below:

- Numerous sections in the updated manuscript have been revised to compare the results from our study to the two other TROPOMI/CrIS O_3 profile validation studies from Mettig et al. (2022) and Malina et al. (2022). Furthermore, the comparison to other multi-wavelength O_3 vertical profile retrievals, using different satellite sensors, is now discussed and the appropriate references are provided (e.g., Landgraf and Hasekamp, 2007; Worden et al., 2007b; Cuesta et al., 2013, 2018; Costantino et al., 2017; Colombi et al., 2021; Malina et al., 2022; Mettig et al., 2022). For example, in Sect. 4 of the revised manuscript our results are compared to Cuesta et al. (2013) which used UV+IR retrievals from GOME-2/IASI, which was not done in detail in Mettig et al. (2022): "Applying different combinations of UV+IR joint wavelength retrievals (e.g., GOME-2+IASI) also displays improvements compared to UV-only products in the troposphere similar to that determined in this study (e.g., Cuesta et al., 2013, 2018). Cuesta et al. (2013, 2018) demonstrated how GOME-2+IASI retrievals show high accuracy compared to ozonesondes in the lowermost troposphere and displays a clear capability to capture PBL

O_3 enhancements. This differs from the results of this study which suggest that TROPOMI+CrIS UV+IR joint wavelength retrievals still struggle to reproduce large PBL O_3 enhancements due to limited lowermost tropospheric sensitivity. The reasons why GOME-2+IASI displays the remarkable capability to retrieve lowermost tropospheric enhancements compared to the results from TROPOMI+CrIS is not immediately apparent. There are differences in the retrieval algorithms, a priori input data sets, and the spectral resolutions of the UV and IR sensors applied. Comparing our results to Cuesta et al. (2013) shows that DOFs are higher in the troposphere and in the 0-2 km agl column (>33% higher) in GOME-2+IASI retrievals compared to TROPOMI+CrIS which would explain some of the differences in capabilities to retrieve lowermost tropospheric O_3 enhancements.".

- Multiple sections of the revised manuscript were updated to explain how our study expands upon Mettig et al. (2022) and Malina et al. (2022). In the introduction we state: "This study builds upon Mettig et al. (2022) to demonstrate the full capability of TOLNet (6 of the 8 systems that were available for the first year of TROPOMI observations) to validate satellite O_3 retrievals at multiple vertical levels in the troposphere. This study applies all available TOLNet systems with spatial coverage throughout the US and in the Netherlands, compared to the small subset of 2 lidar systems used in Mettig et al. (2022), to conduct a more robust validation of the UV-only TROPOMI, IR-only CrIS, and UV+TIR TROPOMI/CrIS O_3 profile retrievals. Furthermore, the only other study to validate TROPOMI/CrIS UV+IR retrievals (Malina et al., 2022) did not apply any ground-based lidar observations. Finally, this study conducts a detailed statistical analysis of satellite O_3 profile retrievals at various vertical levels of the troposphere and investigates the capability of these retrievals to reproduce anomalous atmospheric composition with large impacts on air quality (e.g., stratospheric intrusions, lowermost troposphere pollution events) which was not conducted in past studies validating TROPOMI/CrIS UV+IR retrievals in the troposphere (Mettig et al., 2022; Malina et al., 2022).". Demonstrating the capability of TOLNet to be used as a satellite O_3 validation data set has not yet been proven in the literature and is a major objective of this study. The importance of this is expanded upon in the introduction: "Demonstrating the capability of TOLNet to sufficiently validate satellite O_3 profiles is vital as TOLNet is the primary validation data source for validating TEMPO O_3 products in the troposphere. To-date, no studies have validated satellite data with TOLNet beyond 1 or 2 individual systems instead of the entire network (8 total lidar systems) (Mettig et al., 2022; Sullivan et al., 2022).".

- Our study now demonstrates the similarities and differences between the validation conducted here and the results from Mettig et al. (2022) and Malina et al. (2022). Even the similarities such as demonstrating the improvement in tropospheric O_3 profile retrievals when using UV+IR wavelengths compared to UV-only, while it has been shown in past studies, is important to prove that TOLNet can be used to validate satellite data as accurately as those that applied ozonesondes. We attempted to emphasize this objective in the original manuscript but have added additional text to help highlight this point such as that implemented into the results section of the updated manuscript: "The agreement in the validation statistics of TROPOMI UV, CrIS IR, and TROPOMI/CrIS UV+IR retrievals determined in this study when using TOLNet-AK and those using primarily ozonesonde data (Mettig et al., 2022; Malina et al., 2022) demonstrates that TOLNet is a sufficient validation source for satellite O_3 profile retrievals in the troposphere." and "It is important to note that TOLNet and ozonesonde validation statistics are generally consistent given the fact that ozonesondes are a highly-accurate and commonly-applied satellite validation data source. This suggests that TOLNet is a sufficient validation data source of tropospheric O_3 profile retrievals from satellites.".

- Besides using all the available TOLNet systems, which was not done in either Mettig et al. (2022) and Malina et al. (2022), we focus on chemical environments which are critical for air quality and tropospheric composition which can be challenging to retrieve from space (i.e., stratospheric intrusions, PBL O_3 enhancements) which was not done in Mettig et al. (2022) and Malina et al. (2022). This is emphasized in the updated manuscript: "This analysis of complex atmospheric environments important for air quality using idealized retrievals, produced with known O_3 profiles convolved separately with different retrieval AKs, in this study expands on past studies that have evaluated TROPOMI/CrIS retrievals (Mettig et al., 2022; Malina et al., 2022). It is important to understand the extent to which TROPOMI, CrIS, and TROPOMI/CrIS joint satellite retrievals, which rely on different wavelengths, can accurately retrieve typical and anomalous structures of O_3 in the troposphere.". In addition to this, our study conducts a very detailed validation of satellite O_3 profiles at multiple vertical levels of the troposphere which was not done in Mettig et al. (2022) and Malina et al. (2022). This is now emphasized in the updated manuscript: "This TROPOMI/CrIS validation at multiple layers in the troposphere allows for more detailed interpretation of the capability of these satellite vertical profiles to retrieve middle- to lower-tropospheric O_3 in comparison to other recent TROPOMI/CrIS validation studies (Malina et al., 2022; Mettig et al., 2022).".
- The reviewer states that many non-quantitative statements were made in the original manuscript. No examples were given so it was not immediately clear what they were referring to. However, we have gone through and added quantitative information in many sections of the updated manuscript to address this reviewer comment.

I next have a few specific comments just for the abstract and more general comments / questions about the paper thereafter.

Abstract:

(first paragraph) It is already well known that use UV+IR radiances to estimate ozone increases sensitivity (vertical resolution), relative to UV and IR alone; it is therefore not clear why this first paragraph in the abstract is needed.

This first paragraph was shortened slightly to remove the opening statement about the increased sensitivity in UV+IR retrievals compared to UV- and IR- only retrievals. However, the quantitative statement about the degree of increase in the troposphere, and the improved ability to retrieve high O_3 conditions in the upper/mid and lowermost troposphere, were retained.

Line 40: What are the "tropospheric systematic bias requirements"? Is there a source?

This statement in the abstract was updated to read "…meet the tropospheric systematic bias requirements defined by the science teams for the TROPOMI and CrIS sensors".

Line 41: If the averaging kernel and prior (observation operator) were applied to the TOLNET profiles before comparison than the a priori is removed from the comparison; therefore this should *not be a source of systematic bias unless you can show that non-linearities in the inversion make the choice of prior affect the inversion.*

The reviewer is correct. The bias in the magnitudes and shape of the a priori O_3 profile only biases the retrievals in comparison to raw observations. The statements about the a priori bias impacts when compared to convolved observations have been removed from the updated manuscript and this sentence now reads: "The primary drivers of systematic bias were determined to be solar zenith angle, surface albedo, and cloud fraction.".

Line 47: "random bias"? Please clarify.

This sentence has been updated in the revised manuscript to: "Random errors, representative of uncertainty in the retrievals and quantified by root mean squared errors (RMSE),…" to reflect how random errors are quantified.

Line 52: If TOLNET was sufficient why also use ozonesonde data. Also what does sufficient mean?

In order to show that TOLNet was sufficient, or has the capability to be used, for validating satellite O_3 profile retrievals in the troposphere, besides the fact these lidar data have been validated in past research and shown to be highly accurate as discussed in the manuscript, it is important to see whether TOLNet results in similar validation statistics compared to the well-known satellite validation data source from ozonesondes. The final sentence of the abstract has been updated to read: "TOLNet was shown to result in similar validation statistics compared to ozonesonde data, which are a commonly-used satellite evaluation data source, demonstrating that TOLNet is a sufficient source of satellite O_3 profile validation data in the troposphere which is critical as this data source is the primary product identified for the tropospheric O_3 validation of the recently-launched Tropospheric Emissions: Monitoring of Pollution (TEMPO) mission.". The similarities in validation results determined in this study, compared to other past TROPOMI/CrIS validation studies, which primarily used ozonesondes, are important to demonstrate the capabilities of TOLNet to validate a satellite product. As noted above, we attempted to emphasize this objective in the original manuscript but have added additional text to help highlight this point such as that implemented into the results section of the updated manuscript: "The agreement in the validation statistics of TROPOMI UV, CrIS IR, and TROPOMI/CrIS UV+IR retrievals determined in this study when using TOLNet-AK and those using primarily ozonesonde data (Mettig et al., 2022; Malina et al., 2022) demonstrates that TOLNet is a sufficient validation source for satellite O_3 profile retrievals in the troposphere." and "It is important to note that TOLNet and ozonesonde validation statistics are generally consistent given the fact that ozonesondes are a highly-accurate and commonly-applied satellite validation data source. This suggests that TOLNet is a sufficient validation data source of tropospheric O_3 profile retrievals from satellites.".

Other comments

There are far more ozone-sondes available than just the ones listed in Table 1. Why do you not use them?

We use a small subset of ozonesonde data as it is important to intercompare the validation statistics from TOLNet and ozonesondes separately (as mentioned above this was a goal of this study). In order to do this, we must use observations from both sources taken close to the same locations and time periods. This is further explained in Sect. 2.2 of the revised manuscript with the following statement: "Ozonesondes have been used extensively to validate satellite O_3 vertical profiles in past research (e.g., Worden et al., 2007a; Kroon et al., 2011; Verstraeten et al., 2013; Huang et al., 2017; Malina et al., 2022). In addition to the fact that TOLNet lidar data has been shown to be highly accurate (Leblanc et al., 2016, 2018) as discussed above, this study intercompares the validation statistics from spatially and temporally collocated TOLNet and ozonesonde observations to demonstrate the capability of TOLNet to be used for validating satellite O_3 vertical profiles.".

Equations 1 and 2 are inconsistent with Equation 3. Equations 1 and 2 indicate that the retrieval is linear with respect to concentration or VMR. Equation 3 suggests either a log or fractional value is estimated; additional explanation is required.

In accordance with a comment from Reviewer #1, the way we present Eq. (3) has been corrected in the revised manuscript. Furthermore, the following sentence was added for explanation: "The TOPAS retrieval is conducted with relative deviations from the X_a, therefore the $\boldsymbol{AK}$ is converted appropriately as explained in Mettig et al. (2021)".

There is another paper on this subject by Edward Malina that is not cited. The authors should take a look at this paper and describe what is different with their approach and results relative to those in Malina et al.

Joint spectral retrievals of ozone with Suomi NPP CrIS augmented by S5P/TROPOM

Malina, E., Bowman, K. W., Kantchev, V., Kuai, L., Kurosu, T. P., Miyazaki, K., Natraj, V., Osterman, G. B., and Thill, M. D.: Joint spectral retrievals of ozone with Suomi NPP CrIS augmented by S5P/TROPOMI, EGUsphere [preprint], https://doi.org/10.5194/egusphere-2022-774, 2022.

We thank the reviewer for pointing us to this paper. We now reference the work by Malina et al. (2022) and compare our results to this study throughout the revised manuscript. In addition to the comparison of our results to Mettig et al. (2020) and Malina et al. (2022), the following text was added to summarize the similarities and differences of the approaches of the studies of Mettig et al. (2022) and Malina et al. (2022): "Multiple recent studies have combined UV+IR wavelength retrievals from two newer satellite sensors TROPOMI and CrIS to retrieve tropospheric O_3 vertical profiles (Mettig et al., 2022; Malina et al., 2022). The combined UV+IR TROPOMI/CrIS O_3 profile retrievals from Mettig et al. (2022) were evaluated in the troposphere for a full-year between 2018-2019 using a small sample (2 lidar systems which are also part of the Tropospheric Ozone Lidar Network (TOLNet)) of ground-based lidar remote-sensing observations from the Network for the Detection of Atmospheric Composition Change (NDACC) and ozonesondes (i.e., World Ozone and Ultraviolet Radiation Data Center (WOUDC) and the Southern Hemisphere Additional Ozonesondes (SHADOZ)) and demonstrated that the combined UV+IR retrievals were more consistent with observations compared to the UV-only product. Malina et al. (2022) also evaluated a full-year (between 2019-2020) of combined UV+IR TROPOMI/CrIS O_3 profiles using correlative satellite retrievals and ozonesondes and further showed that combined UV+IR retrievals were more accurate in the troposphere compared to UV- and IR-only products. Mettig et al. (2022) and Malina et al. (2022) both combined TROPOMI and CrIS retrievals; however, applied different retrieval algorithms, a priori input data, and portions of the spectral bands from each satellite, thus the validation results differed to some degree which is discussed in the current manuscript.".

Line 105.. missing Worden et al. GRL 2007 reference where this is first discussed.

This reference has been added.

Section 3: Combinations of UV and IR have appeared in several papers over the last decade. How do the results appearing in each sub-section compare to this prior research (answer, you are essentially getting what is expected based on this prior research).

We agree with the reviewer and have done our best to compare our results to past research throughout the revised manuscript.

Figure 4: Are the UV, IR, and UV+IR, retrievals consistent (especially Figure 4c). Use Rodgers and Connor 2003 (not cited) to determine if purple, red, and blue are consistent or if differences in the troposphere are driven by attributable systematic errors (e.g. albedo, clouds) or because there is a lack of sensitivity in the troposphere.

I believe there was some confusion about what is presented in Fig. 4 and discussed in Sect. 3.2 of the original manuscript. This figure shows example TOPAS retrievals produced with TOLNet lidar profiles convolved with AKs and a priori profiles from UV-only (blue), IR-only (red), and UV+IR (purple) retrieval information. The figure caption of Fig. 4 has been updated to read "Example TOPAS retrievals produced from TOLNet lidar profiles convolved with AKs and a priori profiles from UV-only (blue), IR-only (red), and UV+IR (purple) retrievals at the location of RO_3QET (34.73 °N, 86.65 °W) from the surface to 40 km asl for the case studies of: a) clean/background conditions, b) PBL pollution enhancement, and c) stratospheric intrusion." to better clarify this point. The legend in this figure also now states "True convolved -" instead of "Retrieval" in the updated version of the manuscript. Finally, to avoid any other confusion about this, we have made other small changes to Sect. 3.2 of the updated version of the paper and refer the reader back to Sect. 2.4 where we describe how example retrievals are calculated.

The AKs and error covariance matrices for each TOPAS retrieval are produced with a radiative transfer model that only accounts for known uncertainties (i.e., noise in the retrieval) and do not reflect the impact of clouds/aerosol/albedo/etc. Therefore, all the differences seen in Fig. 4 between the three retrievals are driven by the differences in sensitivity to O_3 in the troposphere.

Line 340: where are these requirement thresholds described?

The accuracy requirements for CrIS are described in Table 5.2.8 of the referenced document in the original manuscript (JPSS, 2019). The link to the supplemental portion of this document has been corrected in the revised manuscript (and below in the Reference section). The table was misunderstood when we referenced the lower (±10%) and higher (±20%) bias requirements. The statements about the higher bias requirements have been removed from the revised manuscript.

Line 395: This approach makes no sense.. if you do not apply the observation operator, then there will be a bias from the combination of prior and sensitivity.

We assume the reviewer is referring to the paragraph between Line 396-404 in the original manuscript. We compare observations (i.e., TOLNet and ozonesondes) convolved with retrieval AKs and a priori profiles (AK-convolved) and the actual observations (raw). We focus the validation of the satellites using the AK-convolved observations which is well-described in the manuscript. A similar approach of comparing TROPOMI/CrIS UV+IR satellite retrievals against ozonesondes without the satellite observation operator being applied was also conducted in Malina et al. (2022) and Mettig et al. (2022). The comparison of satellite data to raw observations in the troposphere is important as this allows for the understanding of how the satellite retrievals are able to replicate actual O_3 values, not just the capability of the spaceborne sensors. This has been emphasized with the additional statement added to the updated manuscript: "While observations convolved with the observation operator is the primary validation data source, comparing the three retrievals to TOLNet observations not convolved with the retrieval AKs (hereinafter TOLNet-raw) is also important to understand how the satellite retrievals reproduce actual atmospheric composition in the troposphere.".

Line 427: This is an interesting statement about TOLNET "A major advantage of using TOLNet for validation of satellite O3 profile retrievals is the ability to make observations at different vertical levels of the troposphere over an entire day or more. " However, it is not obvious how this capability is used for the comparisons.

The ability of TOLNet to make high vertical and temporal resolution O_3 observations through the troposphere for many consecutive hours (TOLNet can make multi-day continuous observations) is a unique feature of these lidar systems. This aspect of TOLNet will be vital for validation of TEMPO, and other future geostationary sensors over the United States, retrievals of hourly O_3 profiles throughout the entire day. To better clarify this and emphasize the importance of TOLNet observations for this study, we added the following text to this section of the revised manuscript: "While TROPOMI and CrIS are polar-orbiting systems which only retrieve O_3 profiles once per day, the observations throughout an entire day are vital for validating geostationary sensors such as TEMPO. However, the high vertical resolution and accurate O_3 observations from TOLNet are applied here to robustly validate satellite retrievals at multiple layers of the troposphere.". Our study conducted a very detailed validation of satellite O_3 profiles at multiple vertical levels of the troposphere which was not done in Mettig et al. (2022) and Malina et al. (2022). This is now emphasized in the updated manuscript: "This TROPOMI/CrIS validation at multiple layers in the troposphere allows for more detailed interpretation of the capability of these satellite vertical profiles to retrieve middle- to lower-tropospheric O_3 in comparison to other recent TROPOMI/CrIS validation studies (Malina et al., 2022; Mettig et al., 2022).".

Section 3.3.3. Again, what is different about these comparisons and conclusions versus Mettig et al. and Malina et al.?

As mentioned in response to earlier comments, we now have included more comparison to our results and those from Malina et al. (2022) and Mettig et al. (2022) throughout the revised manuscript.

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