*****Reviewer's comments are in black*****

*****Answers to the reviewer's comment are in blue*****

Comments from reviewer for UV aerosol retrieval paper

The authors would like to thank the anonymous reviewers for their time and effort to help significantly improve the manuscript.

RC1: 'Comment on amt-2021-307', Anonymous Referee #1

Thank you for expressing your concerns; we have significantly revised the abstract and introduction to improve clarity of the approach. We believe this will help to prevent any further misunderstanding.

This work present and iterative method to obtain lidar ratio at 292 nm, which is later applied, to Langley Mobile Ozone Lidar (LMOL) backscattered signal. Once lidar ratio at 292 is estimated, authors use the classical Klett method to obtain independent aerosol extinction and backscattering. It is well-known in lidar literature that Klett method cannot provide accurate estimates of extinction profiles because of possible variations of lidar ratio with height.

Nevertheless, the authors try to address an important challenge and provide an estimation of lidar ratio at 292 nm. Typically, backscattered lidars use co-located measurements of sun-photometry AOD for estimating lidar ratios (see for example MPLNET or EARLINET/ACTRIS retrievals). Currently, there are not many radiometric measurements that provide aerosol AOD at 292.

Answer: We use Fernald method for aerosol extinction retrieval. Instead of using the sun-photometer AOD to constrain the aerosol retrieval, the analysis in this manuscript uses the co-located HALO measurement to constrain the retrieval. The HALO provides aerosol backscatter, extinction, and lidar ratio profiles at 532 nm with high vertical resolution which is a more reliable constraint for the aerosol retrieval. This is because the sun-photometer is a column-only measurement (no profile) and also does not provide data at 292 nm. As a result, we use the iterative method to determine both the lidar ratio at 292 nm and AE between 292nm and 532 nm.

However, I do not rely in the approach presented by the authors. It might need further explanations. But as I understand they propose iterative variations of lidar ratio in LMOL system and provide different aerosol extinction profiles. The range of variation of lidar ratios is not enough because absorbing aerosol can present lidar ratios larger than 90, and OMI satellite retrievals demonstrated the importance that aerosol absorption might have extinction. On the other hand, I understand that they vary Angström exponent iteratively in HALO system to obtain equivalent aerosol extinction at 292 nm. They are ignoring possible effects of variations of

Angström exponent with altitude. If so I would rely more on Angström exponent measurements using sun photometry. Finally, for the evaluation they are using the same data that for the computation in the iterative method, which is not appropriate.

Answer: There are two reasons that we believe the range of variation of lidar ratio from 10 to 90 making sense. The first one is according to previous publication (Sasano and Nakane, 1984). The second one is the result that we get from our calculation. Most 292 nm lidar ratio calculations converge to values between 20 to 70 sr. That is why we selected the current lidar ratio range (10 sr –90 sr) to save calculation time.

The sun photometer cannot provide the variation of aerosol Angström exponent with altitude. The sun photometer only provides the aerosol Angström exponent for the total column aerosol, while LMOL lidar measures aerosol of lower part of troposphere. Aerosol Angström exponent derived from our method using the LMOL data and HALO data at the same altitude range, provides a more reliable result. In addition, the sun photometer also cannot provide Angström exponent between 292nm and 532 nm since there is no sun-photometer data available at Westport site.

As stated in the paper, we are not proposing a new iterative variation based on some variation of the lidar ratio. We are here comparing the LMOL retrieval, using several lidar ratio, to the HALO data. HALO is an HSRL lidar, which means that it can retrieve the lidar ratio and Angström exponent in an independent and reliable way (see refs cited). As a result, this represents the state-of-the-art to provide aerosol parameters at each altitude. In the present paper, we consider the HALO value as the ground truth, and we compare it to the Fernald method applied to LMOL. We have added an additional figure (Figure 1) and description at the beginning of the manuscript to help clarify the analysis approach.

With all these points I propose to evaluate the method with CCNY lidar for 355 nm and make intercomparisons with extinction coefficient at that wavelength computed by Raman methodology.

Answer: The aerosol AE between 355 nm and 386.7 nm still need to be assumed when 355 nm aerosol extinction was retrieved using Raman Lidar. HALO can independently obtain aerosol extinction and backscatter using Rayleigh and Mie signal without any assumption. So, it is better choice to evaluate our method with HALO data. Furthermore, quantitative CNNY comparisons to LMOL retrievals, which may be interesting, are spatially too far away to be useful for this study.

Section 4.2 does not provide any relevant scientific results. It only shows coherence in the vertical structures of aerosols, and for that it is not necessary to retrieve extinction coefficients. Moreover, the study-case selected to demonstrate the novel methodology must be different than that used for the validation.

Answer: The comparison of aerosol profile in section 4.2 is very important because it will show the difference between LMOL and HALO aerosol profiles when you select the optimized lidar ratio and Angström exponent. This intercomparison is important because it illustrates the ability of the LMOL aerosol retrieval to capture a consistent aerosol feature when compared to HALO HSRL aerosol data. And thus, can produce relevant data for campaign analysis in the relationship of aerosols to ozone features. It is important also because is a first study in the development of a new data product for LMOL.

Finally, section 5 only shows that lidar ratio is the most important parameter in the retrieval of backscattering and extinction, which is widely known in lidar community. What is necessary in section 5 is sensitivity test of the new methodology proposed by the authors, which can be done using synthetic data.

Answer: This section not only shows the lidar ratio being important, but uncertainties in O_3 are comparable and change as a function of height. We agree lidar ratio is a key factor in the retrieval of backscattering and extinction. The sensitivity of both lidar ratio and angstrom exponent is illustrated in section 3.2 and therefore believe an additional sensitivity analysis is not needed.

RC2: 'Comment on amt-2021-307', Anonymous Referee #2

Review of "Retrieval of UVB aerosol extinction profiles from the ground-based Langley Mobile Ozone Lidar (LMOL) system" by Lei et al.,

This paper describes an algorithm for the aerosol extinction retrieval out of the Langley Mobile Ozone Lidar (LMOL) as compared to 20 coincident flights with the NASA Langley High Altitude Lidar Observatory (HALO) 532 nm aerosol extinction product. This work also accomplishes the first known 292nm aerosol product inter-comparison between HALO and Tropospheric Ozone Lidar Network (TOLNet) ozone lidar.

In general, this paper would benefit from an additional proofreading.

Major Comments:

In general, this is a very technically developed manuscript. Many of the equations are first principles and well known in the lidar community. In general, substitution of these for graphical elements (flow charts, or signal processing chains) would improve the readability. This also allows the author to highlight sections that are new to this original research.

Answer: We agree and added a diagram as Figure 1 and descriptive text at the beginning of the paper to better illustrate our approach and improve the readability.

The added flow chart and description of flow chart are as follows:

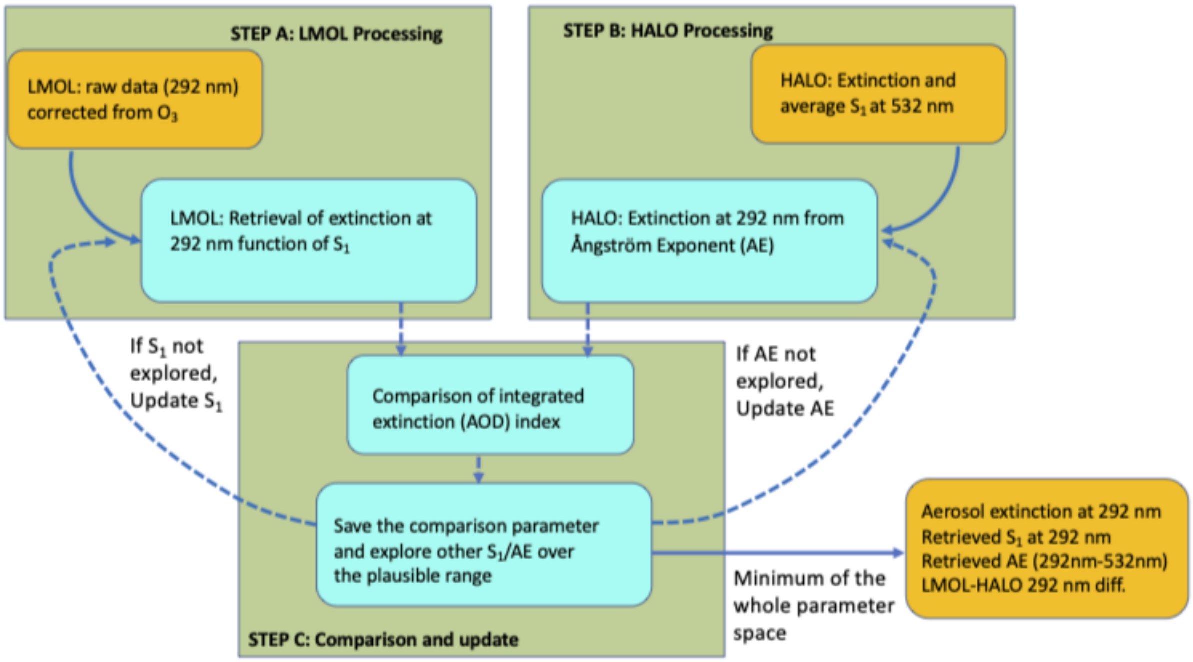

Figure 1. Flow chart for the approach used in this work. The cyan section corresponds to the processing needed for the retrieval of the optimal *(S_1 , AE)*

"To retrieve the S_1 and AE, an iterative method with 3 main steps was used as shown in Figure 1. The first step is the retrieval of the aerosol extinction at 292nm from LMOL. For that, the LMOL raw data are corrected from the ozone absorption. Then the Fernald method (Fernald et al., 1972, Fernald, 1984) is used with an empirical S_1 (which is modified in subsequent iterations to explore the parameter space). For the current study, the impact of the aerosols was low enough that an iterative correction to the O_3 density was not necessary to retrieve the aerosol extinction accurately; for dense aerosols layers, the method described in Browell et al., 1985 would have been used. The second step is the retrieval of the aerosol extinction at 292 nm from HALO. The conversion of the extinction from 532nm to 292nm is done by using an assumed AE which is also modified in subsequent iteration to explore the (S_1 , AE) parameter space. The third step is the comparison of the aerosol extinction from both instruments at 292 nm. The integration of the difference provides the partial aerosol optical depth (AOD) difference, refered later as the partial AOD index. Once the plausible (S_1 , AE) parameter space has been explored, there will be a minimum to the partial AOD index which points to the best (S_1 , AE) for the observed conditions. The LMOL aerosol extinction profile related to optimized S_1 and difference between the LMOL and HALO 292 nm aerosol profile related to the optimized S_1 and AE was also recorded for further analysis."

My major questions

Is this approach actually novel? The authors describe this as working from between 0.5 and 3.5km – does this indicate it may only work properly or is biased for aloft/transported aerosol layers? Please re-emphasize the importance of this work.

Answer: ***a little more wordsmithing***

- Yes, this approach is unique because it provides a way to obtain lidar ratio at 292 nm and get the 292 nm aerosol retrieval for LMOL system. It also provides the AE between 292nm and the intercomparison between the LMOL and HALO system.
- This work focuses aerosol retrieval between 0.5 to 3.5 km because the restriction of the lidar measurement which is lower at daytime because of the strong background.
- Capturing aerosol extinction between 0.5 to 3.5 km is very useful because it will help us to retrieve the planetary boundary layer (PBL) height and also help us to learn aerosol property in the lower part of troposphere. Furthermore, aerosol profiling information can still play an important role for model intercomparisons and satellite retrievals.
- The extinction in wavelength less than 300 nm is difficult to be retrieved using simple lidar techniques because the impact of O_3 absorption and lacking information of lidar ratio at those wavelengths. We proposed the new method to retrieve aerosol extinction at 292 nm using Fernald method with combing the profile of ozone and the profile of 292-nm elastic backscattering from LMOL. The selection of lidar ratio is very import for the retrieval of aerosol extinction. However, the lidar ratios less than wavelength of 300 nm for different aerosol type are rarely discussed according to previous research. So, HALO results were used to constrain LMOL retrieval to improve lidar ratio accuracy.
- Combing long-term measurements of HALO and LMOL, a database of lidar ratio for different aerosol type will be built and can improve LMOL aerosol extinction retrieval without relying on HALO measurements in future.

Can this method be extended to cases outside of when there were HALO overpasses? Otherwise this does not have as much appeal to the general audiences.

Answer: As mentioned above, combing long-term measurements of HALO and LMOL, a database of lidar ratio for different aerosol type can be developed < 300 nm and improve LMOL aerosol extinction retrieval without HALO in future.

A missed opportunity is using the ceilometer to compare with – this is a 24/7 measurement that is made very widely over the country. Then use the HALO data to act as a reference for the quality of the results in this specialized case – and then improve confidence in the ceilometer derived method.

Answer: Although it is not the focus of this work, we agree that applying similar HALO/ground-lidar analyses to ceilometer networks could prove useful and worth pursuing as a separate study. However, this is expected to be challenging due to the longer near-IR wavelength and related signal-to-noise limitations of these systems. As a result, we show a qualitative comparison with the Westport ceilometer that is shown in Figure 6 (Figure 7 in new version of manuscript). With respect to the overall approach described in this study, the ceilometer limits would not provide quantitative benefit since it is not deriving the lidar ratio as a function of height and then requires the a-priori assumptions that we are trying to avoid when comparing with LMOL.

The author described Canadian wildfire smoke, but I cannot tell clearly from any of the images 1) where the smoke resides, 2) how improved the retrieval is in these areas of smoke, or 3) what effects the data set has made to improving the remote sensing of the optical properties of the aerosols.

Answer:

(1) The measurement was impacted by the Canadian wildfire smoke and the active fire in southeastern United States. As we mentioned in introduction: "The August 28 case was shown in detail because the air quality exceedance during that day was probably caused by the impact of long-range transport of wildfire emissions (Rogers et al., 2020)". (We encourage the reviewer to check figure 2 and figure 4 for Rogers et al., 2020 paper)

(2) For the wildfire emission case, some cases are directly has smoke layer aloft, but some cases like the August 28 case the smoke is already mixed in the boundary layer and does not appear as a distinct layer. We can tell that our measurement was impacted by smoke according to previous publication (Wu et al., 2021; Rogers et al., 2020; Hung et al., 2020). The August LISTOS lidar, HALO, and AERONET data provided guidance on which days smoke was expected to be present.

(3) The optimized lidar ratio at 292 nm was smaller than lidar ratio at 532 nm, consistent with expectations but also yields the most appropriate quantitative outcome for the aerosol influenced by wildfire emission.

Minor Comments:

L55 – remove 'a lot'

Answer: We remove it.

L60 - Langley

Answer: We change it. Thank you for reminding us!

L64 "In this paper, the impact of the aerosols was low enough that an aerosol correction to the O3 density was not necessary; otherwise, an interative process would have been necessary (Browell et al., 1985)" – Suggest rephrasing this statement. This sounds like it basically voids the need for this work. Although the ozone correction to the signals may not reduce accuracy, the authors state the uncertainty in ozone is 10-20% - that must impact the uncertainty of the aerosol correction.

Answer: The objective of the present paper is to characterize the aerosols using the 292 data from LMOL, not specifically to correct the O_3 from aerosol impact. Ultimately, when the technique is validated, this would be a step; however, the validation of the retrieval is partially in the current paper. With the standard method for correcting the O_3 signal from the aerosol contribution, (1) O_3 should be retrieved from the raw signal, which (2) allows to correct 292 nm for (3) retrieving the aerosols extinction. From the aerosol, we could (4), correct the density of O_3. With that, we could go back to (2) for correcting the signal. The procedure repeats until convergence.

In the present paper, we have conditions that lead to a small correction of O_3 after the step (4), which in turns gives a negligible change in the aerosols computed in the initial (3).

This is what we meant by “aerosol correction to the O3 density”. We changed into “an iterative correction to the O_3 density is not necessary to retrieve the aerosols accurately”.

L66 – New paragraph for LISTOS

Answer: Yes, we give new paragraph for LISTOS instruction.

L105 - rather than ‘raw’ data, what is being analyzed? Range corrected-Elastic Backscatter Profiles at 292?

Answer: The O_3 corrected range corrected-elastic backscatter profile at 292 nm was analyzed.

L132 – LISTOS

Answer: We change it. Thanks for reminding us!

2.4 The Ceilometer located nearby LMOL – consider moving this up to 2.2 since it was co-located with 2.1.

Answer: That make sense. We move it.

L160 – Lidar ratio is introduced here but is frequently used in the text up until this point.

Answer: We define the lidar ratio at the first time we use it in the text (at line 51).

Section 3 could benefit from some sort of “flow chart” graphic. Or illustration of the changes in the corrected signals after certain steps.

Answer: We add the flow chart in the introduction part (figure 1 in new version of manuscript) which will be helpful to improve the readability of section 3.

Figure 1 caption - (a) August 28fternoon needs to be fixed. Are these derived vertically or for a column?

Answer: Yes, we fix it as "Augst 28 afternoon ...". It is vertically average value derived from HALO S1 profile.

Table 1- 521nm?

Answer: It is 532 nm. We changed it in new manuscript. Thank you for reminding us!

L280 – "These results show that by using the selected S1 and AE in Table 1, LMOL has the capability to retrieve aerosol extinction in 292nm with reasonable accuracy.", What is the estimated uncertainty of this retrieved aerosol component?

Answer: We could see the difference between the retrieved LMOL aerosol profile and the converted HALO aerosol extinction are less than 10 % when use the optimized lidar ratio and Angstrom exponent for August 28 afternoon case. The uncertainty of using lidar ratio other than the optimized value for retrieval was show in figure 3 (b) (figure 4 (b) in the new version of manuscript).

Figure 6 – Are you able to convert the ceilometer to 292nm? Please label in the plot title. Is the PBL height detection "in-house" for the ceilometer or the Vaisala standard product?

Answer: We think it is hard to convert the ceilometer to 292 nm because the ceilometer only provides the backscatter signal at 910 nm. We have added the wavelength information in the plot title for each curtain plot in figure 6 (figure 7 in new version of manuscript). The PBL height is from the Vaisala standard product which could obtained from the LISTOS archive data. We have added related information in section 4.2 in the manuscript.

Figure 7 - total uncertainty (blue) – should be black. Why is the analog Det Nois decreasing with altitude? Wouldn't you expect as the signal to be much higher compared to background noise values, that the uncertainty would increase? Is there a need to show both analog and photon counting here?

Answer:

- Yes, the total uncertainty is black. We changed it in the caption.
- I think we need to point to equation 15 (equation 16 in the new version manuscript) here: the detection noise is the coupling between the Udet, which increases with altitude and the differential of the retrieved value with the detection rate, which could be decreasing with altitude. We set up a value for the aerosols, fixes, at the higher altitude, then we go down from there. The uncertainties are therefore adding while going downwards and are considered stable at high altitude.
- We think it is better to show both analog and photon counting channel here because the analog and photon counting are used for retrieval for different altitude ranges.

Figure 7 In some cases in Photon Counting it looks like the uncertainty in using 60sr is less than using the technique applied in this paper. Is that the case?

Answer:

In the sensitivity study, we apply the uncertainty algorithm to a specific lidar ratio and with an uncertainty on that lidar ratio. If the retrieval equations were perfectly linear, the uncertainty of the retrieval (in percent) would be proportional to the uncertainty of the lidar ratio (in percent) and therefore constant with altitude. In that case, a lidar ratio of 60 +-40% or 35 +-40% would lead to the same uncertainty in the retrieved aerosol extinction.

This is obviously not the case: a change of the lidar ratio leads to a change in both the retrieved extinction and absorption, which could lead to uncertainties higher or lower depending upon the density of aerosols and the amount of aerosols above.

Please note that in the sensitivity study, we assume the 40% uncertainty, i.e. without knowing the optimized value of the lidar ratio which would fit better with the dedicated instrumentation. The error is therefore composed both of the possible bias (poorly known ratio) and the noise due to the change in altitude of the ratio.

Conclusions – rather than say 'good', please use specifics from the retrieval results. Consequently, further research is needed to characterize S1 AE at UVB wavelengths – what exactly is needed?

Answer: We add specific result in conclusion instead of using "good" or "very good". The new statement are as follows:

"The inter-comparison between HALO and LMOL aerosol products showed an agreement within 10% up to 3 km after the optimization method was applied in the case of August 28, 2018. The retrieved LMOL 292 nm aerosol was also compared with co-located ceilometer and CCNY aerosol lidar. It shows that LMOL could capture a consistent aerosol feature and mixing layer evolution. Error analysis shows that the uncertainty from O_3 and S_1 dominate the 292 nm aerosol retrieval and needs to be carefully considered in the retrievals of aerosol profiles of all the TOLNET Lidars. In cases when there is no HALO data, a-priori determinations from differing aerosol types based on this kind of analysis work will serve to provide reasonable S_1. Consequently, further research is needed to characterize S_1 and AE at UVB wavelengths: first, an effort should be made on determining the variation of S_1 and AE with altitude by carefully addressing the uncertainties in the HALO S_1 profile products; second, additional co-located LMOL/HSRL measurements should be done to evaluate S_1 and AE for different aerosol types (smoke, dust, marine aerosol, and pollutant aerosol). This characterization could ultimately enable the use of equipment with a better availability than an HSRL (examples of such equipment could be the MPLs) to provide the ancillary data necessary for the aerosol extinction retrieval."