The editor's / reviewers' comment is in black, the author's response is in blue.

According to editor's and reviewers' comment, the structure of the manuscript has to be changed for better reading. As a guidance, we describe here the major changes:

- We used to describe SSA retrieval using radiative transfer simulations and SVR in parallel, which caused troubles in reading. Now in the revised manuscript, we separate the two methods thoroughly. Section 2 includes everything about the SSA retrieved by radiative transfer simulations, and Section 3 contains all information on SVR retrieval.
- There used to be 2 SVR models: one uses the OMAERUV-AERONET joint data set (UVAI, ALH from OMAERUV and AOD, AAOD from AERONET) to train the SVR model, we call it as the SVR trained by the original training data set; another uses the same training data but with adjusted ALH to replace the ALH in OMAERUV, we called it the SVR trained by the adjusted training data set. The adjusted ALH is using an intermediate SVR trained by TROPOMI ALH. Thus, there used to be 3 SVR models in the previous version manuscript. The SSA retrieved by the adjusted training data set is slightly better than that retrieved from the original training data set (OMAERUV-AERONET joint).
  The original purpose to adjust the ALH is because the OMAERUV ALH is not retrieval but is guessed either from CALIOP climatology or a priori assumptions from AOD retrieval. We used to adjust it with TROPOMI ALH to make it more like observations. But the SSA retrieved by the SVR with the original training data set is acceptable, meanwhile the adjusted ALH causes many confusions. Thus, in the revised manuscript, we have removed the process of adjusted ALH and the SVR trained by the adjusted training data set. There is only one SVR model in the revised manuscript, which is trained by the OMAERUV-AERONET joint data.
- We used to employ AEORNET version 2 inversion product to evaluate our SSA retrievals, and to construct the training data set for SVR method. According to Omar Torres's comment, we have replaced it with AERONET version 3 inversion product. The results and conclusions may change to some extent.
- We used to have only one case study in the manuscript as it was the only one available at that time. Now, we have searched through the recent half year since 2018 November and added cases as long as there are collocated TROPOMI UVAI and ALH, MODIS AOD and AERONET measurements available.
- We have included MERRA-2 aerosol reanalysis (Appendix C) as an independent reference to analyze the spatial variability of retrieved SSA in Section 3.6.3.

The structure of the revised manuscript is as follows:
Section 1 Introduction

Section 2 Experiment 1: SSA retrieval using radiative transfer simulations
Section 2.1 Radiative transfer simulation setup
Section 2.1.1 Aerosol models
Section 2.1.2 Inputs from satellite
Section 2.2 SSA retrieved by radiative transfer simulations

Section 3 Experiment 2: SSA retrieval using support vector regression
Section 3.1 Support vector regression
Section 3.2 Feature selection based on OMI and AERONET observations
Section 3.3 Preparing training and testing data sets
Section 3.4 SVR hyper-parameter tuning
Section 3.5 Error analysis
Section 3.6 Case applications
Section 3.6.1 California fire event on 12 December 2017
Section 3.6.2 Other case applications
Section 3.6.3 Spatial variability of retrieved SSA

Section 4 Conclusions
Appendix

Response to reviewer Omar Torres's comments

Summary

This manuscript documents a statistics-based approach referred to as SVR (support vector regression) to retrieve single scattering albedo using MODIS retrieved aerosol optical depth (AOD) and TROPOMI UV Aerosol Index (UVAI) and aerosol layer height (ALH) from TROPOMI radiance measurements in the Oxygen-A band.

AERONET ground-based aerosol observations and the 13-year satellite OMI aerosol record (OMAERUV product) are used to build a training data set. The OMAERUV component of the training data set consists of a sub-set of ancillary parameters as well as UVAI and ALH values assumed in OMAERUV for the simultaneous retrieval of AOD and SSA. The resulting training data set includes only UVAI and ALH values associated with high accuracy OMAERUV AOD/SSA retrievals as measured by the difference between collocated AERONET and OMAERUV reported parameters (not larger than 0.03 for SSA and better than 5% for AOD).

Two versions of the trained SVR algorithm were used to retrieve the SSA of an aerosol plume over the Pacific Ocean off the coast of Southern California on December 12, 2017. Retrievals were also carried out using a conventional radiative-transfer-based algorithm, referred to as RTM by the authors. Comparison of the three satellite-based retrievals to AERONET Version 2 retrieved SSA at 500 nm (University of California Santa Barbara site) shows that the three space-based inversions agree with the only AERONET ground-based measurement available within AERONET's stated uncertainty (±0.03). On the other hand, the spread of the three satellite based SSA retrievals over the AERONET site is 0.01.

The authors examined the resulting SSA spatial variability over the extent of the plume, and conclude that the results of the SVR retrievals that show higher homogeneity are more convincing than the RTM approach that shows more spatial variability.

Comments

The authors have not demonstrated that the proposed SVR algorithm performs better than conventional RTM-based approaches. Deriving conclusions on the suitability of a retrieval method based on just one independent measurement is scientifically dubious. The author's accompanying argument that the lower spatial variability of the SVR approach makes the result more convincing is purely subjective and lacks the backing of a rigorous error analysis. It also ignores the radiative and dynamic interaction between the aerosol plume and the atmosphere that could generate SSA heterogeneity over a plume stretching over hundreds of kilometers.

The authors should carefully build an evaluation dataset using as many AERONET observations as possible, to judiciously examine the SVR algorithm performance. The interpretation of spatial variability is certainly not easy. Perhaps, CTM-generated data could also be used for this purpose.

I see a problem with the use of the AERONET as both training and evaluation tool. Unlike the AOD, AERONET SSA is not regarded a 'ground truth' measurement. The SSA is the result of an inversion procedure that yields non-unique solutions, and can produce different answers as the inversion algorithm evolves. For instance, for the case study in this paper the AERONET V2 500 nm SSA value used for evaluation of the satellite retrieval was 0.960. In the recently released AERONET V3 data, the reported SSA for the same event is now 0.982. If a SVR operational algorithm is in place, does the algorithm needs to be re-trained every time a new version of the AERONET data becomes available?

Based on the above consideration I do not think this work is publishable in its current form. Additional specific comments follow.

We have added more case applications to present the capability using SVR algorithm to retrieve SSA in Section 3.6 of the revised manuscript, as long as there are collocated TROPOMI, MODIS and AERONET measurement available. In the end there are 9 collocated samples from total 5 cases.

We also have rephrased the discussion on comparison between SSA retrieved from radiative transfer simulations (Section 2.2) and that retrieved from SVR algorithms (Section 3.6) in order to make it more objective.

The CTM data might be an option. We have provided MERRA-2 aerosol reanalysis data (M2T1NXAER) and derived SSA by dividing total scattering AOD with total extinction AOD (averaged between 12:00-15:00 local time) in Appendix C. We also provide the range of retrieved SSA for each case (maximum SSA – minimum SSA) in Table 4. According to MERRA-2, although the plume pattern differences between satellite observations and model simulations exist, the MERRA-2 SSA heterogeneity of plume is at level around 0.1. This value is significantly smaller than the spatial variability of SSA retrieved in Experiment 1, while it is closer to the SSA variability retrieved from SVR.

Your concern on the use of AERONET is true. The supervised machine leaning algorithms have to be re-trained every time if the training data is changed, and the predicted results may also change accordingly. In our case, if the AERONET product is updated, then the SVR algorithm has to be re-trained. It is therefore important to ensure the training data set is of high quality. As your said, the SSA from AERONET inversion product is not 'ground truth', but in most cases, the SSA provided by other sources is compared with that from AERONET. AERONET SSA plays a role as a reference, even though it is not the true value.

Specific comments

Line 29: Equation (1) is not consistent with equation given in Herman et al [1997]. What is the meaning of λ and $\lambda 0$?

It is actually the same equation as the Eq(2) in Herman et al (1997) only with notations changed. We have added description: *where I_λ and $I_{\lambda 0}$ are the radiance at wavelength λ and λ_0 (reference wavelength). (line 34)*

Line 31: Such a SSA global long-term record derived from the information content of the UVAI already exists [Torres et al., 2007]. It is produced by inverting OMI observations at 354 and 388 nm (same wavelengths used in the UVAI definition) to simultaneously retrieve aerosol optical depth (AOD) and single scattering albedo (SSA) at 388 nm. The AOD/SSA retrieval approach by the OMAERUV algorithm is fully documented [Torres et al., 2007; Torres et al., 2013] and SSA retrieval results have been systematically evaluated by comparison to the global AERONET SSA record [Torres et al., 2013; Jethva et al, 2014] and to SKYNET [Jethva and Torres, 2019] and MFRSR [Mok et al., 2016] SSA retrievals. The author's disregard of the 15-year near UV SSA record in the literature review is rather puzzling.

Sorry for the confusion, but we are not saying that there is no method to retrieve SSA from UVAI or there is no long-term record of SSA that derived from UVAI. The message we want to deliver is that the SSA does not have abundant amount of data as UVAI. UVAI data has global coverage for over 4 decades, while SSA data availability is much less than that.

The sentence has been changed into: *It would be beneficial to derive aerosol absorption properties from the long-term global UVAI records, e.g. the single scattering albedo (SSA), which is the ratio of aerosol scattering to aerosol extinction. (line 37-39)*

Line 38: The label 'RTM-based method' is not appropriate. All atmospheric retrievals methods are one way or another based on radiative transfer calculations. The authors are referring to SSA inversions in the UVAI space that infer SSA by 'matching' calculated to observed UVAI. The listed references on this approach are mostly academic exercises, none of which led to algorithm development. While the UVAI parameter contains information on aerosol properties, it is also affected by land surface effects, ocean color, sub-pixel size clouds, gas absorption, etc. Thus, the direct UVAI to SSA conversion techniques is not an optimal way to extract aerosol absorption from the near UV measurements. It is best to use actual radiances.

Truly, all the atmospheric methods are based on radiative transfer calculations. We used to name 'RTM-based method' is to compare with 'SVR-based method', which is only applicable in this paper. In the revised manuscript, we call it as SSA retrieved from radiative transfer simulations.

We admit that fitting the radiances should be better than fitting the UVAI. Nevertheless, the radiance itself is also affected by surface conditions, clouds, atmospheric gases and aerosols. On the other hand, the UVAI only contains the information of aerosol absorption.

Line 47: Please mention the recently developed ALH retrieval capability from EPIC oxygen absorption bands to retrieve ALH of dust layers and carbonaceous aerosol layers over both ocean and land surfaces Xu et al., 2017, 2019]

We have added the work of Xu et al. (2017, 2019). (line 52-53)

Line 70: The discussion of SSA retrieval for this event should also include OMAERUV SSA results if available.

We have added the information of OMAERUV SSA if available, which is in Table 2 and Table 4.

The OMAERUV pixels are applied the same collocation method as that of TROPOMI (distance within 50 km and time difference within 3 hours). For the California fire on 2017-12-12, the OMAERUV SSA is 0.92$\pm$0.01, which is 0.06 lower than that of AERONET. Considering all case studies, there are total 6 OMAERUV samples collocated with AERONET, with half of them within difference of 0.03 (Figure 15).

Line 111: An UVAI threshold of 1.0 also excludes low altitude absorbing aerosol layers, and low AOD elevated layers.

Yes, indeed. We only focus on the aloft (strong) absorbing aerosol layers in case studies. The low AOD may contains low aerosol signal.

Line 114: Sensitivity of results of the assumed 50 hPA pressure thickness assumption should be discussed.

The depth of 50 hPa is used in the TROPOMI ALH retrieval algorithm. We have added explanation in the manuscript: *For the forward radiative transfer calculations, the input aerosol profile is parameterized according to the settings in ALH retrieval algorithm: a one-layered box shape profile, with central layer height derived from TROPOMI and an assumed constant pressure thickness of 50 hPa (Sanders and de Haan, 2016). (line 136-138)*

Besides, the effect of aerosol layer depth on retrieved UVAI is minor, please see the sensitivity study in Sun et al., 2018.

Line 130: The Herman et al [1997] assumption (spectrally flat As in the near UV) has been shown to be not generally valid. Is there a reason why the authors do not use the OMI-based Kleipool et al [2008, 2010] databases?

We used surface albedo at 388 nm of the OMI LER data (Kleipool et al., 2008, 2010) and set the same value for surface albedo at 354 nm based on Herman et al. (1997). Since this is proved to be not generally valid, we have re-run the simulation that use 354 and 388 directly based on Kleipool et al. (2008, 2010). Although in the radiative transfer calculation, due to the round-off, the results may not be significantly changed.

Line 131: In the description of the OMAERUV record, the authors list only the UVAI and ALH and omit the fact that both AOD and SSA are reported retrieved parameters. After all, the reason why the ALH is included in the OMAERUV product is that the inversion requires information on ALH. A more candid description of the OMAERUV product should read '....long-term UVAI, AOD and SSA with corresponding ALH....'

The part is moved to Section 3.2. The sentence has changed into: *We choose OMAERUV because it is currently the only product containing a long-term UVAI, AOD, SSA and corresponding ALH (Torres et al., 2007; 2013). (line 243-246)*

Line 142: Discuss the error bars and whiskers on Fig 2, particularly for the SSA. What are the implications of the expected diurnal variability?

This figure is no longer in the manuscript. This figure is used to provide an overview of AERONET version 2 inversion product availability for the first case we chosen. There was only one day in 2017 December that captures the plume generated by the fire event meanwhile there are TROPOMI UVAI and ALH available. But now we find more cases, thus it is no longer necessary to show this plot.

The diurnal variability of SSA may be caused by the changes of aerosol types, meteorological conditions (cloud contamination, wind direction, humidity, etc.), combustion phases (for biomass burning aerosols), as well as the measuring period, etc.

Line 144: The time difference between TROPOMI and AERONET observations on December 12, 2017 is about 2.5 hours. Discuss the implication of that time difference in the context of the AERONET results in Fig 2.

In the manuscript, we always use the time window of 3 hours to collocate AERONET and corresponding satellite measurements, as what is done in Jethva et al., 2014. The time window is used to exclude AERONET measurements during early morning or late afternoon meanwhile ensures there are sufficient records available. All records within the time window are averaged into one value, which implies that we accept the SSA discrepancy due to the time difference as long as the time difference is within 3 hours. Moreover, since there is only one record, it is almost impossible to determine whether this record is the truth or just an outlier.

Nevertheless, we have added description on the effects of the time difference: *Although our retrieved SSA seems better than that provided in OMAERUV, one should keep in mind that there is only one record for this event, the meteorological conditions, combustion phases and even the aerosol compositions may change during the 3-hour time difference. (line 183-185)*

Line 145: Both AERONET SSA and TROPOMI are results of inversions in which multiple solutions are possible. Thus, an inversion cannot be validated with another inversion. Use 'compare' instead of 'validate'. Use 'comparison' instead of 'validation' in all instances in the paper where the word 'validation' is used.

We have changed them accordingly through the manuscript.

Line 146: Use AERONET version 3 data in the construction of the training data set. There are significant differences between version 2 and 3 of the AERONET inversion product.

We have replaced the AERONET inversion product into version 3 for all the places in the revised manuscript where AERONET data is used.

Line 170. Provide the reasoning to conclude that the southern part of the plume is the most absorbing region. All it can be said, is that the largest UVAI is observed in that region, but AOD, ALH and spectral aerosol absorption exponent affect the magnitude of the resulting UVAI.

The sentence has been changed into: *The highest UVAI appeared at the south part of the plume, where both aerosol loading and aerosol layering are relatively high (AOD > 2 and ALH is over 2.5 km). (line 159-160)*

Line 187. Assuming constant refractive index for wavelengths longer than 388 nm is not a reasonable assumption. At longer wavelengths, the role of black carbon is more important. Discuss the implication of this assumption on the reported results.

Because the spectral dependence of the refractive index between UV and visible band is also not well-understand, and we do not know the exact compositions in the smoke plume. We thus assumed it is 'gray' within this range to keep it simple and only investigate the influences due to spectral dependence in the near-UV range. We thought in previous OMAERUV product the biomass burning aerosols are assumed to be spectrally flat (Jethva and Torres, 2011).

We have added the explanation: *As we only investigate the influence due to aerosol absorption spectral dependence in near-UV range in this study, aerosol absorption at wavelengths larger than 388 nm is set equal to that at 388 nm. (line 122-124)*

Line 193: The 'existing' MODIS AOD and TROPOMI ALH retrievals involve assumptions on particle size distribution (PSD) and aerosol single scattering albedo. Are the assumed PSD's in the two algorithms consistent? How about the complex refractive indices assumed in the AOD and ALH retrieval? Please list the values of those parameters and discuss the implication of inconsistencies if any.

The 'existing' measurements here indicates the source for training data set, which consists of OMAERUV UVAI and ALH, and AERONET AOD and AAOD. The MODIS AOD and TROPOMI ALH are only used in the case application phase (prediction phase), rather than in the training phase. We have noticed the inconsistency you mentioned due to the different aerosol models in two independent algorithms. The inconsistency itself is not the most interested to the SVR model, but the retrieval bias caused by the a priori aerosol models matters. As you also suggested in other comment, we have provided the error analysis of retrieved SSA due to the uncertainties of input UVAI, ALH and AOD in Section 3.5.

But for your request, we will answer your question in this document. There is inconsistency in aerosol models between MODIS/MYD04 product and TROPOMI/ALH product. The MODIS aerosol models can refer to Remer et al. (2006) and Levy et al. (2013). The global evaluation shows that over 66% Dark Target retrievals are within uncertainty envelop of $\pm$0.05+15%*$AOD_{AERONET}$, and the uncertainty of Dark Blue retrievals is $\pm$0.03+0.2*AOD_{MODIS} (Sayer et al., 2013).

The aerosol models in TROPOMI ALH product can refer to Sanders et al. (2016). Aerosols are characterized by SSA of 0.95 and Henyey-Greenstein phase function with asymmetry factor of 7. These values are the averages of long-term AERONET observations for all aerosol types. Retrieved ALH is insensitive to errors in SSA (even with error as large as 0.2). The bias due to SSA is usually smaller over ocean and lager over land, but generally meets the TROPOMI target accuracy of 50 hPa. The algorithm is robust over dark surface even with incorrect knowledge of the phase function. Over bright surface, however, the ALH bias depends on the aerosol loading. For AOD at 550 nm above 0.4, the bias is typically smaller than 50 hPa.

Moreover, there is a long-term downward trend in the magnitude of TROPOMI irradiance (Rozemeijer and Kleipool, 2018), which results in the degradation in UVAI. The degradation is around 0.2 since from August 2018 to June 2019 (Lambert et al., 2019)

We have added descriptions of the uncertainty of MODIS AOD and TROPOMI ALH: *The error the retrieved SSA due to the input features may come from the observational or retrieval uncertainties in each parameter. In our case, the typical UVAI bias requirement is at magnitude of 1 (Lambert et al., 2019). It is reported TROPOMI UVAI suffers from the long-term downward wavelength-dependent trend in irradiance (Rozemeijer and Kleipool, 2018). The detected degradation in* $UVAI_{354,388}$ *is around 0.2 since August 2018 (Lambert et al., 2019). The typical accuracy of TROPOMI ALH is 50 hPa, though in some situations the* *bias may over this value (e.g. low aerosol loading over bright surface) (Sanders et al., 2016). Depending on the retrieval algorithm the uncertainty of MODIS AOD is ±0.05+15%$AOD_{AERONET}$ (Dark Target algorithm) (Levy et al., 2010) or ±0.03+0.2AOD_{MODIS} (Deep Blue algorithm) (Sayer et al., 2013). (Deep Blue algorithm) (Sayer et al., 2013). (line 318-325)*

Line 206: Please describe in more detail the implied statistical analysis of 13 years of data involving the OMAERUV and AERONET data sets. What are the parameters being examined?

The parameters to be examined are: UVAI, ALH, measurement geometries, surface albedo and surface pressure in OMAERUV, and SSA, AOD and AAOD from AERONET.

In the revised manuscript, they are located at line 252-254: *The parameters in OMAERUV-AERONET joint data set for feature selection consists of UVAI calculated by 354 and 388 nm wavelength pair, satellite geometries, surface conditions and ALH from OMAERUV, and SSA, AOD and AAOD from AERONET.*

Line 208: The 13-year OMAERUV global dataset includes AOD and SSA, a record that the authors claim back on page 31, does not exist.

Sorry we are not clear about this comment as we only have 24 pages in the last version manuscript.

Line 212: I am totally lost here. The statement *'samples are excluded if the SSA difference between OMI and AERONET are larger than 0.03 or the AOD difference between OMI and AERONET is larger than 5%'* is incomprehensible. What OMI SSA/AOD are the authors talking about? Are these OMAERUV-retrieved values? Up to this point in the manuscript, the authors have not acknowledged the existence of such products. If these are indeed the OMAERUV SSA/AOD, then the authors have created a dataset consisting of the best quality OMAERUV AOD and SSA retrievals (as measured by the level of agreement with AERONET) to train the SVR algorithm. It is suggest that the description OMAERUV-SSA and OMAERUV-AOD be used (instead of the generic OMI-SSA or OMI-AOD) to avoid confusing the reader since there is a second OMI aerosol algorithm (OMAERO).
Sorry for the confusion. They indicate OMAERUV-SSA and OMAERUV-AOD. We have changed it accordingly in the revised manuscript.

Line 228: Add Torres et al 2013 reference to the CALIOP ALH climatology.

We have added it accordingly: *The best-guessed ALH in the OMAERUV is a combined information of CALIOP climatology or assumed ALH in the retrieval (if the CALIOP climatology is not available) (Torres et al., 2013). (line 246-247)*

Line 232: The UVAI height dependence was first documented [Herman at al., 1997; Torres et al., 1998] based on analysis of TOMS data.

Due to the content changes, this sentence is no longer applicable in the revised manuscript.

Line 235: If spectrally dependent AOD (354 and 388 nm) and ALH are indeed independently know, one should be able to retrieve the SSA at 354 and 388 nm via a direct RTM inversion of the 354 and 388 nm radiances (not the UVAI). This is the simplest RTM approach that would fully characterize the aerosol plume.

Indeed, the method mentioned here is true. But it is still inevitable to assume aerosol micro-physics (size distribution functions, refractive index) if one uses RTM approach. The reason we propose SVR method is to avoid such kind of assumptions and retrieve SSA from empirical measurements.

Line 241: Fig 7(c) is not mentioned in the discussion. Remove it if not needed. Otherwise, explain, or eliminate, the difference between UVAI OMI and UVAI OMAERUV in the z-axis label of figures 7(b) and 7(c).

This figure is no longer applicable. The purpose of this set of 3D plots is to show the difference between UVAI-AOD-ALH relationship in OMAERUV-AERONET and that in TROPOMI-MODIS (for California fire 2017-12-12 only) data set. But the TROPOMI-MODIS joint data is only for elevated absorbing aerosol layers.

We only present the relationship of parameters in OMAERUV-AERONET joint data sets in Figure 6 in the revised manuscript.

Line 245: As described the ALH adjustment sounds arbitrary. It looks to me the authors are just conveniently making up a convenient dataset. Please provide an understandable rationale for the creation of this ALH dataset.

We used to create an adjusted ALH for better SSA retrieval, as the ALH provided by the OMAERUV product is not retrieved by wither from CALIOP climatology or from a priori assumptions during the AOD retrieval. In the previous version manuscript (when AERONET version 2 inversion was used), the results indeed show a slightly better results compared with the results directly retrieved from OMAERUV-AERONET data set. But both results are within AERONET typical uncertainty (0.03). Besides, you commented that it would be over-interpretation if SSA difference is 0.03.

Consequently, we remove the ALH adjustment part in the manuscript, i.e., we only use OMAERUV-AERONET data set to train the SVR model. Please read the Section 3.2 in the revised manuscript. The SSA retrieval is acceptable, with 6 out of 9 AERONET-collocated samples fall within 0.03 difference and all the samples are within 0.05 difference. (Section 3.6)

Line 250 There is no mention in this work of the Oxygen-A band AOD that is simultaneously retrieved with ALH from TROPOPMI observations. Shouldn’t it be better to use the TROPOMI AOD rather than the MODISD AOD? That would eliminate possible implicit inconsistencies in aerosol microphysics.

It is planned that AOD is simultaneously retrieved with ALH in TROPOMI, but the AOD product has not been operational yet (thus not available).

Line 259: What does ‘rule of thumb ratio 70% versus 30%’ really mean? This all sounds arbitrary.

It just means the ratio is chosen based on experience. It is quite arbitrary, but normally the fraction of training data set is 60-90%, and the fraction of test data set is 10-40%.

Line 290: Figures should be described sequentially. From the description of figures 7(a) and (7b), the authors jump to Figure 5, and then back to Figure 7(c).

Figure 7 is no longer necessary thus we have deleted it as explained in the previous response.

Line 301: The MODIS AOD uncertainty needs to be taking into account and propagated in a sensitivity analysis of the SVR application. Over the US west coast, in particular, the AOD is subject to large uncertainty due to surface effects [Jethva et al., 2019].

The error analysis has been added in Section 3.5 in the revised manuscript. Figure 9 presents the sensitivity of retrieved SSA to the changes in UVAI, AOD and ALH.

Line 323: The difference of 0.01 between the two SVR applications has not statistical meaning, as they are both within the stated AERONET uncertainty of ± 0.03 for a single measurement. Any over-interpretation is just splitting hairs.

Thank you for the correction. We have modified our interpretation on the retrieved SSA. The comparison between retrieved SSA, AERONET SSA, as well as OMAERUV SSA is summarized in Table 4 and described in Section 3.6. The difference between SVR retrieved SSA and AERONET is always within

0.05, and 6 out of 9 collocated samples are within difference of 0.03. Compared with OMAERUV, the SVR retrieved SSA is in better agreement with that of AERONET.

Line 326: I disagree with this statement. No measurable improvement in performance is apparent from this comparison. The use of the adjusted ALH instead of the original OMAERUV ALH makes no statistically quantifiable difference whatsoever. A more systematic analysis using a large number of independently measured SSA values is required.

Thank you for the correction. But as the AERONET product has been changed into version 3, some of the results and conclusions are also changed accordingly, and more case studies has been involved. For details please see Section 3.6.

Line 328: In section 4.2, the authors try interpreting the lower spatial variability over the entire plume of the SVR retrieved SSA with respect to the SSA spatial variability resulting from the RTM-based approach, as an indication of better SVR accuracy. The north-south extent of the plume over the ocean is about 1000 km whereas the east-west dimension varies from about 200 to 700 km. For an aerosol plume this large, it is not unreasonable to expect spatial variability in SSA. The SSA of carbonaceous aerosols from biomass burning or wild fires is lowest near the source areas in the flaming phase of the fires. As the resulting smoke layer is transported downwind, it interacts with the surrounding air. Aerosol SSA may increase due to water uptake by hydrophilic particles. The resulting SSA homogeneity over the plume may therefore depend on the homogeneity of meteorological fields.

I do not think this study conclusively demonstrates that the described SVR technique yields more accurate retrieval than standard well thought out RTM approaches. Undoubtedly, however, the availability of TROPOMI ALH observations will improve the accuracy of retrieved aerosol absorption.

Thank you for the correction. The plume spatial variability of this size may exist, depends on the meteorological conditions, combustion phase, etc. But whether a variation from 0.69 to 0.97 is reasonable needs further investigations.

We have added an independent data from a chemistry transport model (i.e. MERRA-2) as a reference to compare with. The MERRA-2 data for each case study is provided in Appendix C. According to MERRA-2, the SSA spatial variability is at magnitude of around 0.1, which is smaller than that in Experiment 1. On the other hand, the spatial contrast of SVR-retrieved SSA is rather flat, with the largest difference from 0.07 to 0.13 (Table 4), depending on cases.

We have modified the explanation: *Depending on the combustion phase and meteorological conditions, heterogeneity in aerosol properties is expected for plume of this size. Nevertheless, whether such a large SSA difference 0f 0.38 (maximum SSA – minimum SSA, Table 2) is reasonable needs further investigations (discussed in Section 3.6.3). (line 192-195)*

We also have added a section to analyze the spatial variability (Section 3.6.3).

Line 345-347: The statement '*In cloud-free cases, it is expected that micro-physical properties of smoke particles within the plume should be similar over short time periods as they were originated from the same source and generated under the same conditions..'* is not always correct. The variability over a large smoke plume like the one used in this analysis may be important.

Thank you for the correction. We have modified our description into: *Depending on the combustion phase and meteorological conditions, heterogeneity in aerosol properties is expected for plume of this size. Nevertheless, whether such a large SSA difference 0f 0.38 (maximum SSA – minimum SSA, Table 2) is reasonable needs further investigations (discussed in Section 3.6.3). (line 192-195)*

Line 364: The statement that the proposed method based on the correlation between UVAI, AOD and ALH requires no a priori assumptions on aerosol micro-physics is incorrect. Implicit microphysics assumptions are involved in the use of MODIS AOD as well as TROPOMI ALH. The authors have ignored this fact, and treat the AOD and ALH as ‘given true values’, ignoring the fact that these parameters come out as the result of RTM-based inversions that assume particle size distribution, and complex refractive index over an extended spectral range. The results of a sensitivity analysis that propagates AOD and ALH retrieval uncertainties into the SVR method should be included in the paper.

Sorry for the confusion, when we say, ‘no a priori assumptions on aerosol micro-physics’, it indicates that we do not have to make such assumptions by ourselves as that in the Experiment 1 (to run radiative simulations. Aerosol models in Experiment 1 (or refer toTable 1) may trigger additional uncertainties in the retrieved SSA. By contrast, the SVR model retrieves SSA based on the given data set without additional assumptions from us.

In the revised manuscript, we have tried to emphasize that the SVR is free from the a priori assumptions made in the radiative transfer simulations in Experiment 1 as clear as possible. For example:

From our perspective, ML techniques can avoid making assumptions on poorly-understand aerosol micro-physics as that in the first experiment. (line 75-76)

An inappropriate assumption may lead to significant bias in retrieved SSA (Fig.3). On the other hand, SVR (and other ML algorithms) is applicable to solve ill-posed inversion problems by learning the underlying behavior of a system from a given data sets without such a priori knowledge on aerosol micro-physics. (line 200-202)

In the second part of this paper, we propose a statistical method based on the long-term records of UVAI, AOD, ALH and AAOD using an SVR algorithm, in order to avoid making assumptions on aerosol absorption spectral dependence over near-UV band. (line 407-409 The error the retrieved SSA due to the input features may come from the observational or retrieval uncertainties in each parameter. In our case, the typical UVAI bias requirement is at magnitude of 1 (Lambert et al., 2019). It is reported TROPOMI UVAI suffers from the long-term downward wavelength-dependent trend in irradiance (Rozemeijer and Kleipool, 2018). The detected degradation in $UVAI_{354,388}$ is around 0.2 since August 2018 (Lambert et al., 2019). The typical accuracy of TROPOMI ALH is 50 hPa, though in some situations the bias may over this value (e.g. low aerosol loading over bright surface) (Sanders et al., 2016). Depending on the retrieval algorithm the uncertainty of MODIS AOD is $\pm0.05+15\%AOD_{AERONET}$ (Dark Target algorithm) (Levy et al., 2010) or $\pm0.03+0.2AOD_{MODIS}$ (Deep Blue algorithm) (Sayer et al., 2013).*)*

The error analysis has been added in Section 3.5 in the revised manuscript. Figure 9 presents the sensitivity of retrieved SSA to the changes in UVAI, AOD and ALH.

Line 365: The statement ‘*a priori assumptions on aerosol microphysics is considered one of the major error sources in RTM-based method*’ is misleading. I should read instead ‘wrong *a priori assumptions* ..’

This content has been moved to Section 2.2. The sentence has been changed into: *The large variability in retrieved SSA (from 0.69±0.13 to 0.94 ±0.03) demonstrates that inappropriate assumptions on the spectral dependence of near-UV aerosol absorption may significantly bias interpretations of smoke aerosol absorption and should be carefully handled in forward radiative transfer calculations. (line 165-167)*

Line 368: ‘*Convincing*..’ is not an objective characterization. I was not convinced as stated in this review.

We have avoided using this term in the revised manuscript.