Response to Reviewers' comments

"Effects of clouds on the UV Absorbing Aerosol Index from TROPOMI" by Maurits L. Kooreman et al.

Reviewer #1

We thank the reviewer for his/her careful reading and for the comments and suggestions, which have improved the manuscript.
Below we give in *blue italic* the reviewer's comment, in black our response, and in purple the changed text in the manuscript.

General comments: This manuscript presents a generally well written study on the effect of clouds on the UV AAI. The presented structural features can be also found on next generation satellite instruments such as Sentinel-4, GEMS, TEMPO. Therefore, I recommend this study for publication in AMT after minor revisions.

We thank the reviewer for the support for this study. The relevance for the three next generation geostationary UV-VIS spectrometers (GEMS being already in orbit) is now added to the outlook in the Conclusions section:
The results of this study are relevant for the future UV-Vis spectrometers with high spatial resolution, like Sentinel-5 on Metop-Second Generation and the three next generation geostationary UV-VIS spectrometers GEMS, TEMPO, and Sentinel-4, all of which will have an AAI product (Kim et al., 2020).

Specific comments:

1. Line 24-26: Please provide some references.

We have added textbook and review paper references on aerosols and their radiative effects.

2. Line 41: Please provide some references.

We have added references on the role of surface albedo in aerosol retrievals.

3. Line 55: I recommend to add one more simple sentence related to physical meaning of using positive AAI for ALH retrieval. For example, only for scenes with a positive AAI value, the ALH retrieval is performed because AAI gets positive for high AOD and absorbing aerosols. For low AOD and scattering aerosols, the TOA reflectance sensitivity to ALH gets lower.

Thank you for this suggestion. Modified text:

Only for scenes with a positive AAI value, the ALH retrieval is performed, because the AAI gets positive for increasing amounts of absorbing aerosols, and especially for elevated absorbing aerosols (De Graaf 2005). Negative AAI values are mostly associated with scattering aerosols and clouds.

4. Line 74: Please add degree symbol here.

Added:

… that 178° E< η < -140° E, where ...

5. Line 76: As I know, there are two products of UVAI in the TROPOMI. One is 354/388, and the another is 340/380. Are there specific reasons for choosing 340/380? For the heritage of GOME?

The 354/388 pair was chosen for OMI because of instrument design reasons; this pair is - in addition to the 340/380 pair - also available for TROPOMI for reasons of continuity with OMI. The 340/380 pair is chosen here because of the TOMS heritage (Herman et al., 1997) and the GOME, SCIAMACHY and GOME-2 heritage. Moreover, we do not expect a significant difference in results between the two wavelength pairs.

Modified text:

We use the TROPOMI offline level 2 AER_AI aerosol index product from the 340\,\unit{nm} / 380\,\unit{nm} pair, version 1.2.0, rather than the 354\,\unit{nm} / 388\,\unit{nm} pair, because of continuity with TOMS, GOME, SCIAMACHY, and GOME-2 data. However, we do not expect a significant difference in results between the two wavelength pairs.

6. Line 77: Please provide this QA value meaning.

This was also asked by Reviewer 2, comment #2.
The meaning of the qa_value is now addressed accordingly. Added text:

This quality assurance value is a continuous quality descriptor, varying between 0 (no data) and 1 (full quality data). The value is changed based on observation conditions and retrieval flags and users are recommended to at least ignore data with qa_value < 0.5 (for details see Apituley et al. (2018)).

7. Line 77: I think the word 'Scene albedo' is very important and key word in this manuscript. Scene albedo in this study means, 'Lambertian Equivalent Reflectivity (LER)' or 'Rayleigh Corrected Reflectance (RCR)' in general. But the author probably did not use the 'LER' expression and using 'Scene albedo' because there is a Lambertian assumption for LER. I recommend to add clear physical meaning for 'Scene albedo' , for example, as an expression for Rayleigh corrected reflectance in section 3.1 (or at some appropriate location).

We agree that the scene albedo is a key quantity in this paper; see A_sc in Eqs. 2-4, which give the definition of AAI. The scene albedo is indeed the Rayleigh corrected scene

reflectance, for some model of the boundary (surface) below the atmosphere. The default model of the scene is a Lambertian reflector. There the scene albedo derives its name from and is then equivalent to the “LER”. Later on in the paper we present a non-Lambertian scattering cloud model. Also for that non-Lambertian scene the scene albedo is derived as the Rayleigh corrected scene reflectance.
Often the term “LER” is also used for the minimum scene albedo of the scene for a certain time period in order to avoid clouds. Here we use scene albedo for the actual scene reflectance.

Added text in Section 3.1, below Eq. 3:
The scene albedo is the Rayleigh-corrected scene reflectance, for an assumed model of the boundary below the atmosphere, where the default is a Lambertian model.

8. Line 81: ‘values. An’ → ‘values. An’

These are identical. If you refer to the gap between the words, we checked it to be a single space.

[...] AAI values. An example [...]

9. Line138: Please add additional explanation about exact position. "intersection of scanline 3600 and ground pixel 0, scanline 600 and ground pixel 0".

We have added your suggestion:

[...], positive AAI features are found near the orbit edges, especially near the orbit start and end, located at the intersections of ground pixel 0 and scanlines 600 and 3600.

10. Fig4: Could you please change the figure with higher resolution, or make the character bigger? Line 142: It is not defined. Please describe as scene albedo.

We have increased the font size and adapted the text:

[...] the mean orbital distribution of the AAI is shown for scenes with few to no clouds, where the retrieved scene albedo A_{sc} is small ($0.0 < A_{sc} < 0.2$) and for cloudy scenes ($0.6 < A_{sc} < 1.0$).

11. Line 144: Could you please add exact place of scanline and ground pixel for each case? Other readers may not understand this clearly.

We have added location details for the described features:

The cloudless scenes show surface effects, such as the sunglint (the blob at ground pixel 130 and scanline 2200) and mildly higher AAI at more extreme viewing angles. The cloudy scenes show that clouds play a large role in systematic AAI offsets. Especially the cloud-bow is a prominent feature (centered at ground pixel 280 and scanline 2200, with a radius of approximately 800 scanlines), but we also see enhanced AAI values at more extreme viewing angles.

12. Line 159: I recommend to describe the following sentence, otherwise it could mislead the reader. "The effect of aerosol on the backscattered radiation in the near-UV (320-400 nm), where the ozone absorption is weak and does not affect the interaction between the aerosols and the molecular atmosphere."

It should be clear that in practice ozone absorption is not interfering with aerosol absorption and scattering, since ozone is mainly in the stratosphere and aerosol is in the troposphere. Furthermore, ozone absorption is very weak in the region 340-380 nm where the AAI is determined, although especially for long paths it should be corrected for.

To take up the suggestion of the reviewer to avoid misunderstandings we added the following text:

The correction for absorption by ozone, which mainly resides in the stratosphere, can be done accurately, since in this part of the UV (340-400 nm) ozone absorption is weak and does not affect the interaction between aerosols and the molecular atmosphere.

13. Fig. 12-14: Have you ever seen these effect as a function of mean cloud fraction? The author has explained BRDF effect through the manuscript (e.g., Line 199, Line 202, Fig.6, Discussion part), and also mentioned at Line 199 that 'much smaller Rayleigh optical thickness, and causes a strong impact of surface BRDF on the TOA reflectance'. But, I guess the relative portion of surface reflectance from TOA reflectance might be still small at 340 nm and 380 nm especially for the clear sky vegetation (land) region, so that we could assume BRF = LER at those two wavelengths (usually over land). Actually this study are investigated over the Pacific area, so if the LCM, SCM model work well for the small cloud fraction region over Pacific ocean, that could be due to bright ocean surface reflectance. In general, the surface reflectance over the ocean would be bright compared to the land surface reflectance due to water-leaving radiance (including the effect of chlorophyll, CDOM) at 340, and 380 nm.

There are three points in this reviewer's remark, which we discuss below.

(i) *Fig. 12-14: Have you ever seen these effect as a function of mean cloud fraction?*

The features in the AAI visible in the multi-orbit transects in north-south and east-west directions (Figs. 12-13) are for the main part due to clouds and only for a smaller part due to ocean effects (especially sunglint). This can be clearly seen in Fig. 5, where the AAI distributions for cloudy scenes (right panel) and cloudfree scenes (left panel) are shown separately. The strongest positive AAI features occur for the cloudy scenes, except for the sunglint feature for the cloudfree scenes. There are however strong negative AAI features in the cloud free ocean scenes. This is an interesting result, which we did not mention before.

Modified text:
When discussing Figs 12-13 we refer to Fig. 5 for the separation of ocean surface and cloud effects (see below).

We have added a remark on ocean BRDF when discussing Fig. 5 in Sect.2.3:
The area of strongly negative AAI north and south of the sunglint (around scanlines 1000 and 3000) might also be related to ocean anisotropy (BRDF).

(ii) ".. *the relative portion of surface reflectance from TOA reflectance might be still small at 340 nm and 380 nm ".*

The question of the reviewer is whether the relative portion of surface reflectance in the UV is not too small to bring about an effect on the AAI. Indeed, in the UV the surface reflectance (LER) is typically only about 0.08 for ocean, and even smaller, about 0.04-0.05, for vegetation (Tilstra et al., 2017). In the paper we selected only orbits over the Pacific ocean, so apart from the ocean surface only clouds can contribute to the reflectance. Clouds are as bright in the UV as they are in the visible, so their reflectance may go up to 0.8-1.0. Therefore, in partly cloudy scenes the impact of clouds will surpass that of the ocean, except in the glint region. In cloudfree ocean scenes, the AAI features are due to the BRDF of the ocean surface. In principle, as discussed in Sect. 3, it is not the absolute value of clouds, ocean or land reflectance that counts for the AAI, but the differences in reflectance between bright and dark regions of the BRDF. For ocean surface this BRDF difference can be very large, namely between the specular sunglint and the dark ocean water.

We have added a remark in Sect. 4.2.2, when discussing Fig. 13, on the role of the ocean surface effects:
The north-south transect over the sunglint shows a steep behaviour, from strongly negative AAI values around scanline 1000 to positive AAI values at the sunglint around scanline 2200, and then again strongly negative AAI values around scanline 3000. The east-west transect over the sunglint in Fig.12 shows a similar behaviour. This steep AAI behaviour was earlier mentioned when discussing the cloudfree part of Fig. 5. This behaviour is probably due to ocean BRDF effects, because outside the sunglint region the ocean is very dark (see e.g. Gatebe et al., 2005).

We have further added a paragraph on the ocean albedo effect in the Discussion of Sect. 5:
For the data analysis, we selected orbits over the Pacific ocean, so apart from clouds only the ocean surface can contribute to the reflectance. At 380 nm the ocean surface albedo is typically only about 0.08 (Tilstra2017). Therefore, in (partly) cloudy scenes the impact of clouds will surpass that of the ocean, except in the glint region. As discussed in Sect. 3, in the absence of absorbing aerosols it is not the absolute reflectance of clouds, ocean or land reflectance that counts for the AAI, but the difference in reflectance between bright and dark regions of the BRDF of the scene; for the cloudfree ocean this means that the BRDF difference between sunglint and non-sunglint regions determines the AAI.

(iii) *In general, the surface reflectance over the ocean would be bright compared to the land surface reflectance due to water-leaving radiance (including the effect of chlorophyll, CDOM) at 340, and 380 nm.*

The reviewer suggests that the AAI effects over the ocean could be due to water leaving radiance, affected by chlorophyll and CDOM. In principle, Chlorophyll and CDOM do have a color in the UV, which could impact the AAI. However, we think that these effects are small, since we are looking at the open ocean, where CDOM and chlorophyll content is very small.

We added a remark in the Discussion, Sect. 5:

We note that possible spectral effects of dissolved matter and chlorophyll in the ocean are not expected, since the content of these constituents is very small in the open ocean.

14. Fig 10: Please add (a), (b) on the figure and caption.

See response to comment (15).

New caption:

AAI sensitivity to errors in surface albedo for the LSM method (dash-dot), LCM (dashed) and SCM method (solid), for HG clouds (blue) and Mie clouds (red). The green line shows the reference (expected) surface albedo of the scene. The horizontal axis shows the actual (real) surface albedo with the panel (a) showing the sensitivity for low albedo scenes with As=0.05, and panel (b) for high albedo scenes with A_s=0.9. The scene has an effective cloud fraction of 40%.

15. Fig 10: Also, there are many Surface Albedo (As) terms in this thesis, so little bit confusing. So In this Fig 10. the x-axis Surface Albedo +0.05(left) or + 0.9(right) corresponds to Surface Albedo in left term of equation (6), or equivalent to Surface Albedo in left term of equation (5), right? And, the x-asis Surface Albedo is actually, d(Surface Albedo) or delta(Surface Albedo), right?

We edited the figure and modified the caption of Figure 10 to clarify the points mentioned in reviewer comments (14) and (15)

Reviewer #2

We thank the reviewer for his/her careful reading and for the comments and suggestions, which have improved the manuscript.
Below we give in *blue italic* the reviewer's comment, in black our response, and in purple the changed text in the manuscript.

General Comments:
This study investigates the cloud effects on the AAI with three models: the traditional Lambertian Scene Model (LSM), and two cloud models (i.e., Lambertian Cloud Model (LCM) and Scattering Cloud Model (SCM)) of IPA assumption, primarily at large scales by aggregating TROPOMI data over the Pacific Ocean where absorbing aerosol effect is essentially negligible. Sensitivity studies of the cloud height and surface albedo with a series of scenarios are also conducted through RT simulations.
Strength: this paper presents the first systematic investigations of the cloud effects on the AAI using TROPOMI data of unprecedented high spatial footprints in UV by making comparison of the performance from three models (LCM, SCM, and LSM).
Weakness: results are inconclusive due primarily to the instrumental degradation and calibration issue in current L-1b data. It is difficult to evaluate the performance of the three models with measured radiances of a calibration problem. This limit of the study will be repeatedly pointed out in specific comments below, even though it is beyond the scope of this study. As stated in conclusions, extra future works still remain for improving the operational TROPOMI AAI product including the surface effects.
Overall, this paper is well written and provides useful information of the cloud effects on the AAI for aerosol community. It is appropriate for publication with minor revisions.

We thank the reviewer for the compliments and support of this study.

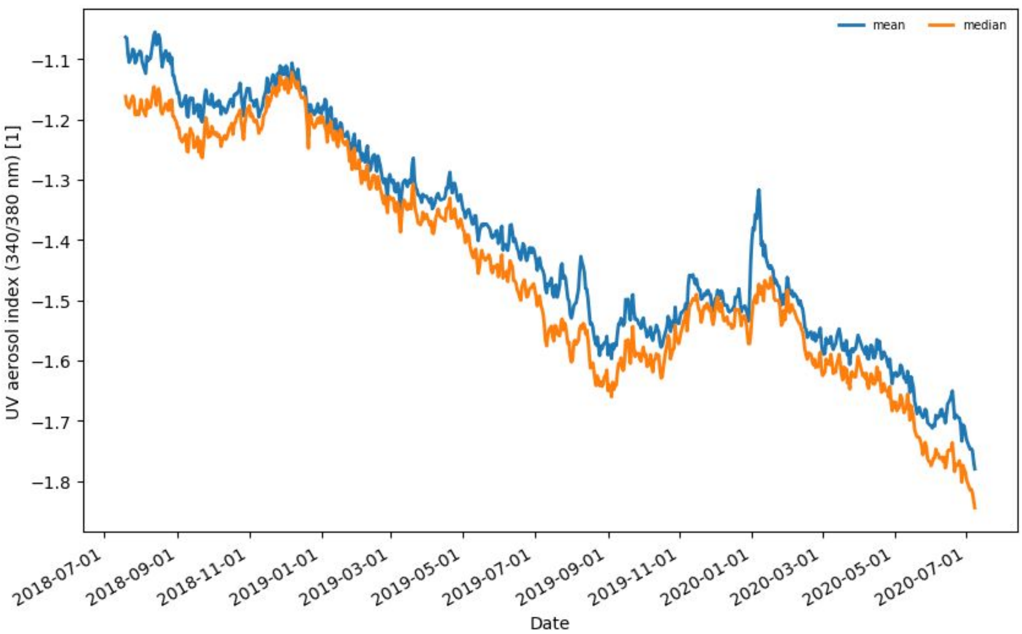
The reviewer mentions the degradation of TROPOMI and the current calibration of L1b data as a weakness of this study. We do not agree. Although calibration and degradation is an important issue when using the AAI from TROPOMI as an absolute threshold for detecting absorbing aerosol events and trends, it is not relevant for this paper. In this paper we consider relative AAI values, namely differences in AAI as a function of viewing angle and solar zenith angle, and differences in AAI of scenes with and without clouds, for one month of TROPOMI data. For these differences the absolute value of the AAI does not matter. In the submitted manuscript we discuss the degradation and the fact that it does not affect the study (Sect. 2.3, line 122ff). We cite:

"The range of values in Fig. 4 immediately shows us that AAI values are mostly negative. This is partly due to the scattering effect of clouds, but also due to a degradation in the TROPOMI irradiance data. Due to the degradation, the 340/380nm wavelength pair AAI has dropped by about 0.5 units over the period of 20 months. After sensor commissioning early 2018, the global average AAI was much higher (-0.2). An update of the level 1b product includes an irradiance degradation correction, which will alleviate the problem. Activation of

the version 2.0.0 level 1b processor is foreseen for late 2020. As the degradation is expected to be independent of viewing geometry, it does not affect the relative AAI values of the orbital features of interest in this work. "

There it is mentioned that the degradation in AAI is about -0.5 in a period of 20 months. This degradation makes all the AAI values shift downward, irrespective of solar and viewing geometry. So the large-scale AAI features over the orbit, e.g. the cloud bow and other BRDF effects, still remain, as well as the small-scale features due to clouds. Therefore, the results of this study can still be used for conclusive results on the three models LSM, LCM and SCM regarding the solar and viewing angle dependent features.

We should also mention the L1b calibration offset in the current data which causes a downward AAI shift. As an illustration of the TROPOMI degradation and calibration offset, the figure below shows the daily global mean and median AAI for the current two-year period of the operational mission, from July 2018 to July 2020. The temporal behaviour is a combination of the L1b degradation and the seasonal cycle of the AAI due to changing solar geometry (see e.g. Tilstra et al., 2012). In addition, there is an offset due to day-1 calibration. So to bring the TROPOMI AAI into agreement with other satellite instruments, one can shift all values upward. But the orbital features in the AAI will remain.



We expect that the L1b calibration of TROPOMI will be improved soon, as this is needed for use of the AAI for event and trend detection and for retrievals of aerosol optical thickness and aerosol single scattering albedo from TROPOMI UV L1b data.

To clarify the manuscript, we updated the text in Sect. 2.3:

… also due to a radiometric calibration offset and degradation in the TROPOMI irradiance data.

To further clarify the point that an overall shift of the AAI due to L1b calibration does not impact this study, we added the following text to the Discussion in Sect. 5:

The current TROPOMI L1b calibration bias (including degradation) causes a negative shift in the AAI. However this shift is independent of solar and viewing geometry. So the calibration bias reduces the AAI at a global level, but it does not change the differences in AAI. Therefore it does not impact our study of the orbital distribution of the AAI.

Specific Comments:

1. Page 1, lines 18 -19: I have not seen any comparison result of the performance of AAI in terms of footprints sizes (e.g., fine TROPOMI vs. coarse Suomi NPP/OMPS) in this paper. The authors need to discuss such topics in discussion or future plan before stating in abstract.

We agree, so we removed this sentence from the abstract.
New text:
The BRDF effect presented here is a first step - more research is needed to explain the small scale cloud effects on the AAI.

2. Page 3. Line 77: describe "qa_value" scheme how to derive this quantity.

The meaning of the qa_value is now addressed accordingly:
This quality assurance value is a continuous quality descriptor,varying between 0 (no data) and 1 (full quality data). The value is changed based on observation conditions and retrieval flags and users are recommended to at least ignore data with qa_value < 0.5 (for details see Apituley et al. (2018)).

3. Page 4, Figures 1 and 2: provide regional information of the maps (both longitudes and latitudes ranges) and TROPOMI orbital number.

The longitude and latitude of the image centers are already provided in the captions. Additionally we have provided the orbital numbers.

Fig 1: This scene was observed on 30 August 2018 (orbit 4563).
Fig 2: [...] of a cloudy scene over the southern Pacific Ocean (57.3° S, 120.4° W) on 30 August 2018 (orbit 4562).

4. Page 5, lines 120 -125: mostly negative AAI values imply that instrumental effects (not only time dependent degradation but also absolute calibration at 340 and 380nm) appear to be far larger than cloud effects.

As shown above, the calibration offset plus degradation in AAI is about -1 to -1.5, depending on the period. The cloud effects in TROPOMI AAI are much larger: (i) as shown in Figs. 1 and 2, the small-scale cloud effects are up to 6 and larger, namely the differences between the red high values and the blue low values; (ii) as shown in Fig. 3, the large-scale effects are up to 3, namely the differences between the high and low values; (iii) in simulated data in Fig. 7 the cloud effects are around 6. Most importantly, however, as mentioned above in the response to the General comments, we consider in this paper the orbital features (differences in the AAI) due to viewing and solar angle variation - an absolute shift of the AAI by a fixed amount does NOT affect these features.

5. Page 10-11. Section 3.2 describes the physical principles and interpretation of the Lambertian surface model-based AAI in terms of BRDF concept. In reality, surface BRDF behavior and its effect on the AAI can be far more complex than a simple interpretation of the diffuse to direct light ratio and difficult to say the signs of AAI. Reconsider the change of the title of section 3.2 since it did not show any real surface BRDF effect on the AAI.

Indeed, Sect. 3.2 is explaining the principle of the effect of anisotropy (which can be described by a BRDF) on the AAI which is based on a Lambertian surface. We think that this principle is very important, also for complex BRDF functions. Although the figure may be simple, the fact that anisotropy of surfaces and clouds affects the AAI in the UV has many implications.

We changed the title of Sect. 3.2 to:
Effect of anisotropy on the AAI

We added a new figure showing the BRDF of Mie scattering clouds for the three optical thicknesses that were used in Fig. 7 for the AAI simulation: COT=1, 8 and 32. We hope this clarifies the relation between BRDF and AAI, as a detailed illustration of Fig. 6 with a realistic BRDF.
New figure: Fig. 7a.

6. Page 21, line 410: sun glint features are due to the Fresnel reflection over the ocean surface under clear sky condition.

We added this explanation to Sect. 3.2, where we discuss sunglint:
" in the sunglint, …due to Fresnel reflection at the ocean surface under clear sky condition."

We further added text on the effect of the sea surface in Sects. 4 and 5, in response to Reviewer #1 comment 13.

7. Page 21, all plots in Figure 12 show large negative AAI values (except sun glints) and a strong cross-track dependence regardless of regions and models, which is not consistent with the results in Torres et al (2018). As stated in many places of this paper (page 2, lines 33-34; page 5, lines 117-119; page 26, lines 477-479), an optimal AAI retrieval would mean *that clouds give a neutral AAI, i.e., close to or equal to zero, especially from the statistics of averaging many orbits in this study. The results here indicate that the instrumental effect appears to be far larger than the improvements to forward modeling with cloud models.*

As discussed above, the TROPOMI instrumental effect causes an overall downward shift of the curves in Fig. 12, but does not not change the features and the shape of the cross-section curves. We note that there are large differences between our TROPOMI data and the OMI data of Torres et al. (2018):

- OMI has a 16 x larger pixel size than TROPOMI. Small details visible in TROPOMI AAI are washed out in OMI AAI data (as they are in GOME-2 PMD AAI data).
- Similarly the angular resolution of TROPOMI is much smaller than OMI (and GOME-2 PMD).
- The TROPOMI data are only for clouds over ocean; in Torres et al. aerosols and land effects are present.
- The TROPOMI data are for all latitudes; the OMI study has a limited coverage of latitudes.

We note that there is a cross-track dependence in the Mie UVAI data of Torres et al. (2018), visible in their Fig. 12 for large solar and viewing angles over ocean close to Antarctica. This is a BRDF feature present in all satellite AAI data.

Regarding the statements on a neutral AAI for clouds: in Sect. 1 and Sect. 2 we give the traditional view of the AAI: it should give zero for clouds. This is true when averaged over large areas and large angular ranges. The insight of this paper as given to us by the TROPOMI high resolution data is that an optimal AAI retrieval providing neutral AAI in the absence of aerosols is only possible if one knows the exact BRDF of the underlying scene, being land, ocean, or cloud, or a mixture.

8. Page 23, Figure 14, difficult to read legends and labels. Other figures also need to be improved with increased font size and legends.

We increased the figure size. Depending on Copernicus markup we will increase font size if required.

9. Page 24, Figure 15 shows mostly negative AAI values due to a calibration problem.

We agree. Improved TROPOMI L1b calibration will remove the negative bias in the AAI and will shift the entire histograms in the positive direction - but it will not change the width.

10. Page 26, line 463: never shown any real BRDF results.

Thank you for the suggestion to show BRDF results; see also point 5. A new figure, Fig. 7a, showing the BRDF of clouds has now been added to Sect. 3.2.

11. Page 26, lines 473-474: difficult to conclude it because of the instrumental degradation and calibration issue.

We do not agree. See our response to the General comments.

12. Page 28, lines 528-536: I disagree. This is not a "subjective" or "preferential" choice issue but a scientific issue. The current TROPOMI AAI product should be further investigated and improved with more effective physical models for absorbing aerosol studies by minimizing other effects such as clouds, surface, and instrument.

The choice of the appropriate physical model for particular use of the AAI is of course a topic of scientific debate. We changed the word "subjective" to "matter of choice".

13. Page 28, line 540. Other sensors (e.g., Suomi NPP/OMPS-NM AAI and DSCOVREPIC AAI) are also capable of detecting such huge smoke plumes at large scales. Clarify an unprecedented "sensitivity" of TROPOMI.

We agree that other satellite instruments can also detect these plumes. We changed the last sentence into:
" … can be detected by TROPOMI with unprecedented sensitivity to small details at high spatial resolution."