This document contains the responses to the 2 reviewers (response to the reviewer 1 starts in page 1 and response to the reviewer 2 starts in page 6), response to the editor (starts in page 8) followed by a marked-up version 3 manuscript (starts in page 12).

The reviewers' comments and questions are in **bold**. For each comment / question, the authors' reply / answer is 5 in black, and the corresponding modifications and line number is the manuscript version 3 are marked in blue colour.

#### (1) Author's response to Anonymous Referee #1

Thank you very much for your comments to our manuscript. The introduction has been trimmed. We also have revised 10 the entire manuscript. The following are the responses to your specific comments.

# Line 20-21: a difference in SSA of 0.06 is very big as far as aerosol remote sensing and climate applications are concerned! It is not "slightly" smaller.

We admit that a difference 0.06 is not a small value for SSA and we have deleted this word accordingly. This statement 15 has been rephrased into The retrieved mean  $\omega 0$  at 550 nm for the entire plume over the period from 26-30 January 2017

varies from 0.81 to 0.87, whereas the nearest AERONET station reported values in the range from 0.89 to 0.92 (line 21 - 23).

But it is comparable with previous research. Hu et al. (2007) used TOMS AAI to retrieve SSA, their analytical uncertainty is 15%. For the typical AOD level in our case ( $0.3 \sim 0.7$ ), the SSA uncertainty is  $0.02 \sim 0.06$  according to

20 Hu et al. (2007) research, which matches with our results.

Furthermore, as we mentioned in the abstract and stressed throughout the manuscript, the AERONET site and the plume we defined are not collocated (the AERONET site is in the city centre as mention in line 200, which potentially overestimates the SSA). This makes this kind of retrieving to be validate. Other concerns, such as the lack of aerosol layer height, uncertainty in MODIS AOD and AERONET itself, should also be considered.

25

# Also, missing what is the purpose of this study? And what are the conclusions that the reader should take out of this work?

This application attempts to quantify the aerosol absorption by retrieving SSA from satellite measured AAI. The conclusion is satellite retrieved AAI is a useful parameter to constrain forward simulation and to derive SSA. Although

30 currently we have a difference of 0.06 compared with AERONET, this discrepancy can be interpreted by the uncertainties in the inputs and AERONET itself as well as difference in measurement techniques (i.e. satellite vs ground-based measurements).

The purpose is presented in the last paragraph in Section 1 (line 59 - 61).

#### 35 Line 41: incorrect definition of SSA, it is not a ratio of radiation. This is too basic to be missed.

Thank you for the correction on the SSA definition.

We have rephrased to  $\omega_0$  is defined as the ratio of the aerosol scattering over the extinction (line 33).

# Line 43-44: No, POLDER does not measure the "aerosol polarized phase function". It measured polarized 40 radiation that can be linked after modeling to the aerosol phase function.

Thank you for the correction.

We have rephrased to More advanced sensors, such as the POLarization and Directionality of the Earth's Reflectances (POLDER), can retrieve  $\omega 0$  from a combination of multi-angular, multi-spectral observations of the polarized radiation (line 36-38).

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Line 53-54: This is not entirely correct. Eck et al (2013) demonstrated that MODIS retrievals are impacted by the variation of SSA in smoke. (Eck, T. F., B. N. Holben, J. S. Reid, et al. 2013. "A seasonal trend of single scattering albedo in southern African biomass-burning particles: Implications for satellite products and estimates of emissions for the world's largest biomass-burning source." J. Geophys. Res. Atmos. 118 (12): 6414-6432 [10.1002/jarrd 50500])

### 50 [10.1002/jgrd.50500])

It may be a misunderstanding. We are not saying aerosol absorption has no effect on AOD, we are just saying AOD is less sensitive to aerosol absorption. AOD could be large under either a very scattering case or a very absorbing case. The reference as you mentioned here states the effect of retrieving AOD from a constant pre-assumed SSA, and this effect is presented as the systematic bias of retrieved MODIS AOD from AERONET AOD. This statement stresses more on the AOD

55 retrieval bias is sensitive to SSA, rather than AOD itself. Because the major revision in the Section 1 Introduction part, this sentence is no longer available.

#### Line 60-61: not clear with what you mean that the AAI reduces the retrieval uncertainty. Uncertainty of what?

Using AAI, instead of AOD to constrain the inversion of aerosol properties retrieval can reduce the uncertainty of 60 retrieved aerosol parameters. For AAI, the uncertainties come from the measured reflectance. But for AOD, the uncertainties come from both the measured reflectance and pre-assumed aerosol types.

Because the major revision in the Section 1 Introduction part, this sentence is no longer available. The corresponding content is in line 48 – 49: The most important advantage of the satellite retrieved AAI is that it does not dependent on assumptions on aerosol types, while a-prior aerosol types are major uncertainties in aerosol parameter retrievals, such as T.

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# Line 66-67: this is a poorly phrased sentence and very confusing, the AAI in presence of aerosol is sensitive to aerosol height, SSA and concentration,... not SSA alone.

Sorry for the confusion. But we do not mean that AAI is only sensitive to SSA, just in terms of SSA, AAI is more root sensitive than AOD.

Because the major revision in the Section 1 Introduction part, this sentence is no longer available. The corresponding content is in line 53: Moreover, the near-UV AAI is by definition highly sensitive to  $\omega_0$ .

#### Line 98-99: incomplete/confusing sentence

75 Sorry for the confusion.

We have rephrased to The basic idea of the residue method is that in a pure Rayleigh atmosphere, the reflectance (or equivalently the radiance  $(L_{\lambda})$ ) decreases strongly with the wavelength (line 79-80).

Equation 2: what is the definition of deltaI(lambda)?, This equation is different than what other groups use as definition of AAI. Can you provide a reference where this equation is derived? It seems to me a minus sign is missing, also not clear where the delta is coming from? For example de Graaf et al., (2007) uses the standard definition of AAI. how does you equation related with the more commonly used equations? (de Graaf, M., P.

# Stammes, and E. A. A. Aben (2007), Analysis of reflectance spectra of UV- absorbing aerosol scenes measured by SCIAMACHY, J. Geophys. Res., 112, D02206, doi:10.1029/2006JD007249.)

85 We have added the definition of  $\Delta I_{\lambda 1}$  in line 88. We also have added that AAI calculation assumes a Rayleigh atmosphere at  $\lambda_2$ ,  $I_{\lambda 2}^{Ray}(a_s) = I_{\lambda 2}^{obs}$  (Herman et al., 1997) in line 87. The derivation from Eq.(1) to Eq.(2) is then not difficult (line 83). If it is still not clear to you, here is the derivation procedure:

According to the AAI definition:

$$AAI = -100 \left( log_{10} \left( \frac{l_{\lambda 1}}{l_{\lambda 2}} \right)^{obs} - log_{10} \left( \frac{l_{\lambda 1}}{l_{\lambda 2}} \right)^{Ray} \right)$$
(1)

90 , which can be re-written into:

$$AAI = -100(log_{10}(I_{\lambda 1})^{obs} - log_{10}(I_{\lambda 2})^{obs} - log_{10}(I_{\lambda 1})^{Ray} + log_{10}(I_{\lambda 2})^{Ray})$$
(2)

, the Rayleigh radiance is calculated by a surface albedo that satisfies  $(I_{\lambda 2})^{obs} = (I_{\lambda 2})^{Ray}$ , then Eq.(2) can be re-written into:

$$AAI = -100(log_{10}(I_{\lambda 1})^{obs} - log_{10}(I_{\lambda 1})^{Ray})$$
(3)

95 , reformed into:

$$AAI = 100(log_{10}(l_{\lambda 1})^{Ray} - log_{10}(l_{\lambda 1})^{obs})$$
(4)

$$AAI = 100 log_{10} \left( \frac{(\lambda_1)^2}{(l_{\lambda_1})^{obs}} \right)$$

$$\tag{5}$$

, here we define  $(I_{\lambda 1})^{Ray} = (I_{\lambda 1})^{obs} + \Delta I_{\lambda 1}$ , where  $\Delta I_{\lambda 1}$  can be explained as the change of radiance spectral dependency between a Rayleigh atmosphere and an observed atmosphere. Under cloud-free condition, the presence of absorbing 100 aerosols lead to a positive  $\Delta I_{\lambda 1}$ . The definition of  $\Delta I_{\lambda 1}$  is also mentioned at line 105 in the manuscript. Then Eq.(5) can be re-written into:

$$AAI = 100 log_{10} \left( \frac{(l_{\lambda1})^{obs} + \Delta l_{\lambda1}}{(l_{\lambda1})^{obs}} \right)$$

$$AAI = 100 log_{10} \left( \frac{\Delta l_{\lambda1}}{(l_{\lambda1})^{obs}} + 1 \right)$$
(6)
(7)

, which is the Eq.(2) in the manuscript.

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### Line 117: linear interpolation of what? What parameters are being interpolated? Please explain.

Linear interpolation of complex refractive index over spectral range from 340 to 675 nm.

We have rephrased into part into We obtain the size distribution function and complex refractive index at 440, 675, 880 and 1018 nm from AERONET, and apply the linear interpolation / extrapolation to derive the complex refractive index
over the spectrum from 340 to 675 nm, with spectral resolutions of 2 nm. Then DISAMAR uses above information to calculate the aerosol phase function P(Θ) and ω₀ over the full spectrum (line 103-106).

# Line 123: Aeronet Phase function data is not reported at 354nm , where does this come from? As it is , this is not correct.

115 The phase function at 354 nm is calculated by the radiative transfer model DISAMAR with AERONET constraints. We took size distribution function, and complex refractive index at 440, 675, 870 and 1018 nm from AERONET. We used

linear extrapolation method to extend the spectrum refractive index to 340 nm. Then the radiative transfer model used that information to calculate the phase function and SSA over the full spectrum (those are intermediate outputs). That is how the phase function and SSA at 354 nm comes. With those intermediate outputs (that carry information on aerosol
 types), DISAMAR can execute forward simulation of AAI.

We have not have a this work into We obtain the size distribution f

We have rephrased this part into We obtain the size distribution function and complex refractive index at 440, 675, 880 and 1018 nm from AERONET, and apply the linear interpolation / extrapolation to derive the complex refractive index over the spectrum from 340 to 675 nm, with spectral resolutions of 2 nm. Then DISAMAR uses above information to calculate the aerosol phase function  $P(\Theta)$  and  $\omega_0$  over the full spectrum (line 103-106).

#### 125

# Figures 3 and 4 cannot be interpreted because deltaI has not been explained/defined. No further reading of the manuscript since what I found until here warrants a rejection.

Please refer to the derivation of Eq (2)  $(I_{\lambda 1})^{Ray} = (I_{\lambda 1})^{obs} + \Delta I_{\lambda 1}$  in previous or line 88 in the manuscript.

### 130 Line 15: what max value was observed?

To your question, the maximum AAI observed by OMI for Chile 2017 wildfires over all the pixels of the 4 days is 5.80. The maximum median value is 4.05 and obtained on 29 January 2017.

### Line 18: what measurements/obs you are referring to? Radiances?

135 We are referring to the CALIOP backscatter coefficient measurements. We have rephrased into The simulated plume ascends to an altitude of 4.5-4.9 km, which is in good agreement with available CALIOP backscatter coefficient measurements (line 18-19).

#### Line 18-20: not clear what you want to say in the sentence starting with "Due to the relatively..."

140 To your question, the OMI observation is sparsely distributed which may contains geographical outliers that may not be the plume even it has AAI value larger than 1. Therefore, we applied an additional data quality control procedure with interquartile range technique. That is, calculate the difference between simulated and observed AAI, and remove the pixels that have AAI difference outside the interquartile range. This is detailed described in line 250-260.

We have rephrased into Due to the heterogeneity of the data that may contain the pixels outside the plume, an outlier detection criterion has to be applied (line 19-20).

#### Line 20-21: are these SSA values averages over the plume or specific pixels?

To your question, the retrieved SSA is the mean value for the entire plume over the period from 26-30 January 2017.

We have rephrased to The retrieved mean 600 at 550 nm for the entire plume over the period from 26-30 January 2017
varies from 0.81 to 0.87, whereas the nearest AERONET station reported values in the range from 0.89 to 0.92 (line 21-23).

#### Line 33: replace "bright surfaces" with "snow", I am assuming this is what you meant.

Thank you for correction.

155 Because the major revision in the Section 1 Introduction part, this sentence is no longer available.

Line 82-83: what are the locations of Pichilemu and Constitution? Are those forests ? Cities? regions? Please provide more details of the geographical setting. Was there a drought?

The location of Pichilemu and Constitución are two cities at the central of Chile as mentioned in line 62. The local
forestry industry (pine and eucalyptus) contributed a large fraction of the fire source in line 64. There was a drought as mentioned in line 61. All this information was actually mentioned in the version 2 manuscript.

**Line 84: figure 1 has very poor contrast when printed, please correct.** The original plot is from NASA. It has been adjusted now (line 515).

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#### (2) Author's response to the Anonymous Referee #2

Thank you very much for your comments on our manuscripts. The following are the responses to your comments. 200

1. On page 6 authors write: "the fine and coarse mode are derived separately from AERONET". This is not true. AERONET inversion algorithm primarily retrieves aerosol size distribution (ASD) in 22 discrete radius points. The separation in two modes is done after the inversion by finding inflection point in between two ASD peaks and then approximation each part of ASD by log-normal distribution. The parameters of these

- 205 approximations are provided as ASD parameters for fine and coarse modes. The detailed description of this procedure can be find at https://aeronet.gsfc.nasa.gov/new\_web/Documents/Inversion\_products\_V2.pdf. Therefore the averaging of SSA of two modes is not needed. The total SSA for the initially retrieved ASD (at 22 points) can be used instead. I am wondering how close averaged SSA to the total SSA is provided by AERONET. In addition, SSA for fine and coarse modes are not advised to use because retrieval in
- 210 implemented under assumption that complex index of refraction is the same for all the sizes. Sorry for the confusion. We are not indicating that the AERONET derives fine and coarse mode separately, but indicating that we obtained the two modes by finding the peaks in the size distribution function provided by AERONET.
- The radiative transfer model DISAMAR currently cannot directly apply bi-modal distribution function. Instead, we 215 built two modes individually, corresponding to the two modes taken from AERONET. For each mode, we assigned the optical properties (refractive index) provided by AERONET. Then DISAMAR calculates the phase function and SSA for each mode. Finally, we combined two modes into one weighted by number density fraction and extinction cross section.

We have rephrased the sentence into The fine and coarse mode particle size are derived by finding the two peaks of the
 log-normal distribution function provided by AERONET. The complex refractive index is assumed the same for both modes. Since bi-modal aerosol is not applicable in DISAMAR yet, we first calculate optical properties of two modes individually, then we externally combine the optical properties of two modes into a bi-modal aerosol with a fraction (line 210-213).

225 2. On page 1 authors mention that retrieved SSA (0.84) is slightly lower that AERONET value. I encourage authors to replace or remove word slightly because the absolute difference 0.06 is significant in terms of radiative forcing estimation.

Thank you for your suggestion. We admit that 0.06 difference for SSA retrieval is not minor, and 'slightly' is not properly used.

230 We have deleted 'slightly' accordingly, and rephrased into The retrieved mean  $\omega_0$  at 550 nm for the entire plume over the period from 26-30 January 2017 varies from 0.81 to 0.87, whereas the nearest AERONET station reported values in the range from 0.89 to 0.92 (line 21-23).

### 3. Page 2. "The foremost advantage of the AAI is its independence from assumptions on aerosol types, which 235 significantly reduce the retrieval uncertainty". This statement is confusing because the simulation of AAI is still

# dependent on aerosol type.

Sorry for the confusion. Indeed, AAI forward simulation (like what we do in this study) needs aerosol information, including aerosol loading, profile, micro-physics, etc. But AAI retrieval from satellite is independent of pre-assumed

aerosol types. It is calculated directly from the measured radiance (Eq.(1)). Here we want to stress that the AAI
 retrieval is independent of aerosol information itself, thus there is fewer uncertainties in the retrieved AAI.
 We have rephrased this sentence into The most important advantage of the satellite retrieved AAI is that it does not dependent on assumptions on aerosol types, while a-prior aerosol types are major uncertainties in aerosol parameter

- retrievals, such as τ (line 48-49).
  245 4. Page 3. What wavelength interpolation is used for? Index of retraction or aerosol optical parameters? To your question, we used linear interpolation to interpolate the AERONET refractive index from 440 to 1080 nm, and
- used the same technique to extrapolate the spectrum to 340 nm. With wavelength dependent size distribution function and refractive index, the radiative transfer model (DISAMAR) calculates the phase function and SSA for the full spectrum we specified (340 to 675 nm, with resolution of 2 nm) (those are intermediate outputs). With those intermediate 250 outputs (that carry information on aerosol types), DISAMAR can execute the forward simulation of AAI.
- We have rephrased this part into We obtain the size distribution function and complex refractive index at 440, 675, 880 and 1018 nm from AERONET, and apply the linear interpolation / extrapolation to derive the complex refractive index over the spectrum from 340 to 675 nm, with spectral resolutions of 2 nm (line 103-105).
- This is also mentioned in line 223-225 where we applied the same method for the case study: To constrain the spectral dependency of optical properties in the near-UV band, complex refractive index nr and ni in the UV band are linearly extrapolated using available data between 440 and 675 nm as mentioned in Section 2.2.
  In line 225-226 we mentioned the treatment for AERONET AOD and SSA (we use them for evaluating our results): Finally, the AERONET retrieved τ and ωo is also linearly interpolated to 550 nm.

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#### (3) Author's response to the editor

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Thank you very much for your comments on our manuscript. In the followings please find responses to your comments.

- Your manuscript # amt-2018-40 has received comments and suggestions from two anonymous reviews for which you have just submitted a response addressing their concerns. Thank you. Along the review process, I have read your paper and gave some thoughts on the research content. As I understood, the concept of the retrieving aerosol SSA in the near-UV region by constraining AOD quite resembles to the methods presented in Satheesh et al. (2008) and Gassó and Torres (2016). Both earlier studies, and the results of your own paper demonstrate that it is possible to retrieve aerosol SSA and layer height from the color ratio information (UVAI) in the near-UV given the AOD as an a priori and a pre-285 defined aerosol model. It was a surprise that your paper completely misses to discuss and cite these two papers relevant
- to the present study.

We have read the two research papers we missed out upon that you mention in the comment. We appreciate the studies you suggested here (the publication of Satheesh et al. was actually published in 2009). From our view, the goals and / or the methods of these two studies do not resemble to ours. Satheesh et al. (2009) provided a hybrid retrieval method by combing
OMI and MODIS measurements, and Gassó and Torres (2016) aimed to discuss the discrepancy between OMAERUV AOD and other independent measurements. But both studies did discuss the relation among aerosol layer height, aerosol concentration, and aerosol absorption, thus we have included them in the introduction part.

Satheesh et al. (2009) used a hybrid approach to retrieve aerosol layer height (ALH) and aerosol single scattering albedo (SSA).

295 They combined the OMI aerosol product (OMAERUV), which is sensitive to ALH and aerosol absorption, with MODIS's accurate aerosol optical depth (AOD), which is insensitive to ALH. Their study has a similar object as ours, that is to retrieve SSA from satellite measurements, but the method is not the same. Both studies retrieve ALH and SSA from given a priori aerosol models. Satheesh et al. used MODIS AOD as the parameter to constrain the operational OMAERUV retrieval, while we use the absorbing aerosol index (AAI). The role of AAI used in Satheesh et al. (2009) is a qualitive parameter to distinguish absorbing aerosols from non-absorbing ones.

Specifically, Satheesh et al. (2008) extrapolated MODIS AOD to the near UV band. Using this MODIS-produced AOD to constrain the standard OMI AOD inversion procedure (OMAERUV) allows to derive improved ALH and SSA (that specified in the LUT). In our study, we used MODIS standard AOD at 550 nm to constrain the radiative transfer calculation in the forward simulation, and used OMI measured AAI to constrain the backward retrieval of ALH and SSA. Furthermore, Satheesh

- 305 et al. (2009) only compared the SSA retrieved with the hybrid-algorithm with that retrieved with the standard OMI algorithm SSA. They did not validate it with measurements from other instruments such as AERONET. Satheesh et al. mentioned the difficulty of extrapolating MODIS AOD from visible band to 388 nm where OMI requires the AOD information for retrieval (especially difficult for small particles, e.g. biomass burning aerosols). They first applied a linear least square fitting (log-log scale) for AOD as a function of wavelength, then they improved the method by including
- 310 information on the AOD spectral curvature. The relation between UV and visible AOD may provide some clues to determine the relation between UV and visible SSA and refractive index, and it is worth to study in a separated research. Satheesh et al. also mentioned that measurements of aerosol absorption in the UV spectral range are rare, which makes it difficult to validate the retrieval results. We met similar difficulties in determining the spectral dependence of aerosol properties from the visible to the UV band. But most importantly, we did not notice that the use of MODIS AOD can eliminate the uncertainty of ALH
- 315 assumption. We have included this as one of the reason that use MODIS AOD to constrain the forward simulation.

Gassó and Torres (2016) compared the OMAERUV aerosol product with independent measurements (MODIS and AERONET) in order to: (1) assess the quality of the OMAERUV AOD retrieval over ocean (section 3); (2) estimate the impact of cloud contamination on their AOD retrieval (section 4); (3) demonstrate the effect of variations in aerosol concentration and

- 320 height on OMAERUV by comparing the operational OMAERUV algorithm and the OMI-MODIS joint algorithm presented in Satheesh et al. (2009) (section 5); (4) determine whether the assumed aerosol models that leads to discrepancies in AOD retrieval of section 5 (section 6). Gassó and Torres (2016) aimed to evaluate the influences of several factors on the AOD retrieval, rather than to retrieve the SSA from measured AAI (with MODIS AOD constrain).
- Through case studies, Gassó and Torres (2016) evaluated the hybrid retrieval method presented by Satheesh et al. (2009) via 325 case studies. At high AAI magnitude (AAI > 1.8), the retrieved ALH is comparable with CALIOP measurements, although their method is quite sensitive to small variations in the input AOD in case of low AAI. Although they seemed to discuss the interplay among AAI, ALH and AOD, the AAI was not used as the constraint to retrieved aerosol absorption. However, Gassó and Torres (2016) concluded that AAI alone cannot be used quantitatively if no ALH or AOD information is available. This is consistent with our finding that the accuracy of the retrieved SSA depends strongly on the uncertainties in AOD and ALH.

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In summary, although these two studies discussed the interplay among AOD, ALH, and aerosol absorption (SSA / AAI / refractive index), but the purpose / method differ from ours. We use the measured AAI as an additional constraint to retrieve SSA, which is not the case the other two studies. But both studies provided useful information in aerosol research that we can include in our manuscript. Here are the corresponding contents that we have included study of Satheesh et al. (2009) and Gassó and Tarma (2016).

335 and Torres (2016):

In section 1 introduction: But this aerosol absorption over near-UV is highly sensitive to the assumption on aerosol layer height. Satheesh et al. (2009) therefore used the  $\tau$  from MODerate-resolution Imaging Spectroradiometer (MODIS), which is independent of aerosol layer height, to constrain the OMAERUV retrieval. The validation showed that compared with operational OMAERUV algorithm, the retrieved aerosol height by the hybrid method is in a better agreement with air-borne

340 measurements, implying a potential improvement in aerosol absorption retrieval. This OMI-MODIS joint retrieval was also evaluated by Gassó and Torres (2016). They found under less absorbing condition, the hybrid method is sensitive to the variations in the input  $\tau$ , which is used to select the retrieved pair of aerosol layer and  $\omega_0$ . (line 43 - 49)

In section 3.1.2 MODIS and OMI aerosol optical thickness: Besides, the MODIS retrieved  $\tau$  is free from the uncertainty triggered by assumed aerosol profile (Satheesh et al., 2009). (line 187 - 188)

345 In section 3.2 Methodology: Previous research suggested AAI cannot be quantitatively used without  $\tau$  or z<sub>aer</sub> information (Gassó and Torres, 2016). (line 257 - 258)

In this inversion process, your study employs an aerosol model developed using the AERONET data. It was not clear in the Methodology section what kind of spectral dependence is assumed between the two near-UV wavelengths. A 350 number of studies, using laboratory measurements and satellite data, have shown that the carbonaceous aerosols generated from the open field agricultural burning and wildfires exhibit a strong wavelength-dependent of absorption (imaginary part of the refractive index) in the near-UV region. In our earlier study (Jethva and Torres, 2011, ACT), we learned that the relative spectral dependence in the imaginary part of the refractive index needs to be increased to 20% with reference to no spectral dependence (equivalent to black carbon) in order to obtain a better agreement in

355 AOD and SSA between OMI and AERONET. A strong wavelength dependence in aerosol absorption in the UV has been shown as a proxy for the amount of organics in the biomass burning smoke. The color ratio (UVAI) information in the near-UV strongly varies with the Absorption Angstrom Exponent, a parameter that quantifies the spectral dependence of aerosol absorption. Since your paper lacks to provide this essential information, it is hard to interpret the SSA retrievals presented in the paper. Perhaps, a significant disagreement of 360 ~0.06 in SSA between the OMI retrievals and that of AERONET reflects the issue described above that you should consider while revising your manuscript. Please verify the relative spectral dependence in absorption assumed in the aerosol model and discuss/mention in the revision.

Apology for the unclear explanation in the Section 3 Methodology and datasets. Indeed, the spectral dependence of aerosol absorption has a significant influence on the retrieval of SSA. As you mentioned in Jethva and Torres (2011), compared with 365 the gray aerosol assumption used in OMAERUV algorithm, an aerosol model with wavelength dependence can significantly reduce the bias in retrieved AOD and achieve better agreement of retrieved SSA with AERONET measured SSA (70% retrieved data is within +/-0.03 difference).

Therefore, one has to be careful when relating the aerosol refractive index in the UV to the one in the visible band. Jethva and Torres (2011) finally adopted a spectral dependence with a 20% increase in the 354 nm imaginary refractive index with respect

- 370 to the value at 388 nm (absorbing Angstrom Exponent around 2.5 to 3.0 for seven carbonaceous aerosol models in OMAERUV). It is an improvement compared to the operational application, where the aerosol properties are pre-assumed. However, aerosol properties evolve over time and thus also over space. Also, even for a specific aerosol type, the aerosol properties vary from one study to another. Thus, we prefer to directly use AERONET measurements to define the aerosol type rather than to use to one of the pre-defined aerosol types.
- 375 The corresponding content in the manuscript has been rephrased into: The spectral bands of the AERONET instrument at this site only cover the visible band. Due to the absence of observations, assumptions have to be made on the spectral dependence of aerosol properties to obtain their values in the near-UV band. The properties of biomass burning aerosol depend on the type of fuel, the procedure producing the smoke, the age of the smoke, and also the atmospheric conditions (Reid et al., 2005). Using measurements to constrain the input aerosol refractive index can reduce the uncertainties due to a-priori knowledge.
- 380 Our treatment on the complex refractive index is as following: (1) take the complex refractive index at visible band (440 to 675 nm) from AERONET measurements; (2) linearly extrapolate the complex refractive index to near-UV band. The real part nr for radiative transfer calculation is obtained in this step. A slight wavelength dependence of nr is found from the measurements; (3) for the imaginary part nr, we multiply it (for the entire wavelength from UV to visible) with a scaling factor as we treat it as a free parameter. By varying the value of the scaling factor, both the magnitude and the wavelength dependence of nr ic can change to meet the requirement of retrieval (line 234 243).
- We have now also included the retrieved refractive index in Table 2. In our case the spectral dependence between 354 and 388 nm is less than 5%, which is smaller than the 20% in Jethva and Torres (2011). We also added a plot to show the original AERONET measured (dashed line) and retrieved complex refractive index (scaled by a factor) in Fig.9. Although the scaling factors can amplify the spectral dependence, it is not very large due to the flat character of the original refractive index. We
- 390 have added an explanation for this: There are many sources accounting for this discrepancy. First of all, the nearest site Santiago\_Beauchef is not exactly in the primary biomass burning regions as mentioned in section 3.1.3. The AERONET site is located in downtown, where reflective urban or industrial aerosols may have been mixed with the smoke enhancing the ω<sub>0</sub>. This would also affect the complex refractive index used in radiative transfer calculation, since we use the AERONET measured refractive index to constrain the forward simulations. According to Table 2, the retrieved n reveals that the difference
- **395** between 354 and 388 nm is less than 5%. This small spectral dependence of  $n_i$  is mainly determined by AERONET measurements in the visible band (dashed lines), whereas the effect of the scaling factor is minor in this case (Fig.9). We thus find a much weaker wavelength dependence than in the Jethva and Torres (2011) study, where a 20% difference between the two UV wavelengths was applied to OMAERUV algorithm to achieve the result that 70% of the retrieved  $\omega_0$  differ less than  $\pm 0.03$  from the  $\omega_0$  from the AERONET measurements. A stronger spectral dependence of  $n_i$  between 354 and 388 nm would
- 400 allow simulations to reach a higher AAI while keeping  $n_i$  at a relatively low level, so would retrieve a higher  $\omega_0$  at 550 nm. The presence of non-absorbing aerosols weakens the spectral dependence (particularly in the UV spectral range) and the linear

extension would overestimate the aerosol absorption. In this situation, the uncertainties due to assumed spectral aerosol properties might compensate the measurement errors to some extent (line 313 - 326).

In our case, we use the AERONET measurement to constrain the spectral dependence of aerosol properties, but it might be 405 improved if observations from other observational aerosol properties in UV band would be available as constraint. This is summarized in the conclusion: This study proves the potential of utilizing OMI measured AAI to quantitatively characterize aerosol optical properties like  $\omega_0$ . Currently, it is challenging to retrieve and validate results without reliable aerosol profile information. In the future, the availability of daily global aerosol profile data, e.g. the L2 aerosol layer height product

TROPOspheric Monitoring Instrument on-board Sentinel-5 Precursor (TROPOMI) that is underdevelopment (Sanders and de 410 Haan, 2016), are expected to provide a stronger constraint on the forward calculation and to significantly reduce the uncertainty in the retrieved aerosol properties. Perhaps, more sophisticated assumptions on spectral-dependent aerosol absorption (e.g. steeper gradient of  $n_i$  in UV than visible band) have to be made and evaluated by other observational aerosol properties in UV spectral range, e.g. AERONET measured  $\tau$  in UV band, instead of only depending on measured refractive index in visible band (lie 381 - 388).

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#### Quantifying the single scattering albedo for the January 2017 Chile wildfires from simulations of

#### the OMI absorbing aerosol index

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Abstract, The absorbing aerosol index (AAI) is a qualitative parameter directly calculated from satellite measured reflectances. Its sensitivity to absorption by aerosol particles in combination with a long data record start in the late 1970's

- $\frac{445}{makes it an important parameter for climate research. In the first part of this study, a series of AAI sensitivity analyses is presented exclusively on biomass burning aerosols. Later on, this study applies a radiative transfer model (DISAMAR) to simulate the AAI measured by the Ozone Monitoring Instrument (OMI) in order to derive the aerosol single scattering albedo (<math>\omega_0$ ). The inputs for the radiative transfer calculations are satellite measurement geometry and surface conditions from OMI, aerosol optical thickness ( $\tau$ ) from the MODerate-resolution Imaging Spectroradiometer (MODIS), and aerosol micro-
- 450 physical parameters from the AErosol RObotic NETwork (AERONET), respectively. This approach is applied to the Chile wildfires for the period from 26 to 30 January 2017, when the OMI observed AAI of this event reached its peak. The Cloud and Aerosol Lidar with Orthogonal Polarization (CALIOP) failed to capture the <u>complete</u> evolution of the smoke plume, therefore the aerosol profile is parameterized. The simulated plume ascends to an altitude of 4.5-4.9 km, which is in good agreement with <u>available CALIOP backscatter coefficient</u> measurements. Due to the <u>heterogeneity of the data that may</u>
- 455 <u>contain the pixels outside the plume</u> an outlier detection criterion has to be applied The results show that the AAI simulated by DISAMAR is consistent with observations. The correlation coefficients <u>fall into the range between 0.85 and 0.95</u>. The retrieved mean  $\omega_0$  at 550 nm for the entire plume over the period from 26-30 January 2017 varies from 0.81 to 0.87, whereas the nearest AERONET station reported values in the range from 0.89 to 0.92. The difference in geolocation of the AERONET site and the plume, the assumption of homogeneous and static plume properties, the lack of the aerosol profile

460 information, and the uncertainties in <u>the inputs for radiative transfer calculation</u> are primarily responsible for this discrepancy.

#### 1 Introduction

Biomass burning aerosols are generated from combustion of carbon-containing fuels, either by natural or anthropogenic processes (Bond et al., 2004; IPCC, 2014). They are of great concern from the perspective of climate Kaufman and

- Boucher, 2002; IPCC, 2007; Koch and Del Genio, 2010; Huang et al., 2013; IPCC, 2014). The reported radiative forcing of black carbon (BC) produced by fossil fuel and biofuel is around 0.4 Wm<sup>-2</sup> (0.05 – 0.80 Wm<sup>-2</sup>) (Ramanathan and Carmichael, 2008; Bond et al., 2013; Huang et al., 2013), but this estimate is highly uncertain. Accurate measurements of the aerosol single scattering albedo (ω<sub>0</sub>) on a global scale can reduce the uncertainty in radiative forcing assessments (Hu et al., 2007), ω<sub>0</sub> is defined as the ratio of the aerosol scattering over the extinction. Currently ω<sub>0</sub> is mainly measured by ground-based
- 470 instruments (Dubovik et al., 1998; Eck et al., 2003; Petters et al., 2003; Kassianov et al., 2005; Corr et al., 2009; Yin et al., 2015). Satellite sensors, such as the POLarization and Directionality of the Earth's Reflectances (POLDER), can retrieve ω<sub>0</sub> from a combination of multi-angular, multi-spectral observations of the polarized radiation. By measuring the anisotropy of the reflected radiance for each ground pixel, POLDER is expected to determine the reflected solar flux more accurately (Leroy et al., 1997). Unfortunately, there is no continuous temporal coverage because the first two POLDER missions ended
- prematurely due to technical problems on the satellite level. The third POLDER mission only covered the period 2004-2014.

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Instead, satellite derived  $\omega_0$  is usually retrieved simultaneously with the aerosol optical thickness ( $\tau$ ) based on the pre-defined aerosol properties, such as the near-UV aerosol product (OMAERUV) of the Ozone Monitoring Instrument (OMI) (Torres et al., 2005; Torres et al., 2007), But this aerosol absorption over near-UV is highly sensitive to the assumption on aerosol layer height. Satheesh et al. (2009) therefore used the  $\tau$  from MODerate-resolution Imaging Spectroradiometer (MODIS), which is

- 595 independent of aerosol layer height, to constrain the OMAERUV retrieval. The validation showed that compared with operational OMAERUV algorithm, the retrieved aerosol height by the hybrid method is in a better agreement with air-borne measurements, implying a potential improvement in aerosol absorption retrieval. This OMI-MODIS joint retrieval was also evaluated by Gassó and Torres (2016). They found under less absorbing condition, the hybrid method is sensitive to the variations in the input τ, which is used to select the retrieved pair of aerosol layer and on.
- 600 Herman et al. (1997) first defined the near Ultra-Violet (UV) absorbing aerosol index (AAI), which provides an alternative methodology to retrieve 60% from satellite observations. The near-UV AAI, usually derived from the spectral range between 340 and 390 nm, is a qualitative measure of absorbing aerosols that was first provided by the Total Ozone Mapping Spectrometer (TOMS) on-board Nimbus-7 in 1979. Since then several instruments have contributed to the AAI data record, that now spans nearly four decades. This long data record is an important motivation for us to improve methods to derive quantitative aerosol information from the near-UV.

The <u>most important</u> advantage of the <u>satellite retrieved</u> AAI is <u>that it does not dependent on</u> assumptions on aerosol types. <u>while a-prior aerosol types are major uncertainties in aerosol parameter retrievals</u>, <u>such as t</u>. Ginoux et al. (2004) suggested that comparing model simulations with AAI from TOMS allows a better control of discrepancies because the only error source is the model. Further advantages of <u>AAI</u> are the low reflectivity of the Earth's surface and the absence of significant

- 510 molecular absorption over the near-UV range. Using this band can ensure the aerosol absorption is one of the major contributors to the total signal. Moreover, the near-UV AAI is by definition highly sensitive to  $\omega_0$ . Previous studies have proven the potential of the near-UV AAI from TOMS in aerosol properties retrieval. Torres et al. (1998) provided the theoretical basis of an inversion method to derive  $\tau$  and  $\omega_0$  from backscattered radiation. This method was validated by ground-based observations during the Southern African Regional Science Initiative (SAFARI) 2000 measurement campaign.
- The agreement of τ and ω<sub>0</sub> reaches ±30% and ±0.03, respectively [Torres et al., 2005]. Hu et al. (2007) retrieved global columnar ω<sub>0</sub> based on the AAI from TOMS with an average uncertainty of 15%.
  This study is inspired by previous research to quantify the aerosol absorption from AAI. We use the near-UV AAI provided by <u>OMI</u> on-board Aura, the successor of TOMS, to derive the aerosol properties of the Chile wildfires in January 2017. Triggered by a combination of long-term drought and high temperature, this series of fires occurring in central Chile
- 620 (Pichilemu 34.39<sup>°</sup>S<sub>2</sub> 72.00<sup>°</sup>W and Consititución 35.33<sup>°</sup>S, 72.42<sup>°</sup>W) was regarded as the worst wildfire season in the national history (The Guardian, 2017). The fires led to evacuations of the affected areas and caused massive losses of the local forestry industry (pine and eucalyptus forests) (NASA gov, 2017). The smoke plume was transported away from the source regions towards the tropical area in the Pacific Ocean by north-westward winds (Fig.1). In this study, we quantitatively retrieve the ω0 of this smoke by simulating the near-UV AAI from OMI with the radiative transfer model Determining
- 525 Instrument Specifications and Analysing Methods for Atmospheric Retrieval (DISAMAR). The aerosol inputs of DISAMAR includes the τ retrieved from MODIS on-board the NASA EOS Aqua satellite, and information on aerosol micro-physical parameters provided by AERONET. In the next section, we provide a brief introduction on the near-UV AAI and its sensitivity to various parameters. The retrieval methodology is described in section 3. In section 4, retrieved results and uncertainty analysis of Chile 2017 wildfires are discussed, followed by main conclusions in section 5.

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#### 2 AAI sensitivity studies based on DISAMAR

In this section, we first introduce the near-UV AAI. In the sensitivity analysis, we show that the AAI depends not only on aerosol parameters, but also on the surface conditions and the observation geometry. The sensitivity analysis in this study is only designed for biomass burning aerosols.

#### 2.1 Near-UV AAI definition

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The concept of the near-UV AAI was first conceived to detect UV-absorbing aerosols from the spectral contrast provided by TOMS observations, known as the residue method (Herman et al., 1997). The basic idea of the residue method is that in a pure Rayleigh atmosphere, the reflectance (or equivalently the radiance (L<sub>2</sub>) decreases strongly with the wavelength. The presence of absorbing aerosols will reduce this spectral dependency of L<sub>2</sub>. The change in this wavelength dependency is summarized as the AAI, which is calculated from the L<sub>2</sub> at the wavelength pair  $\lambda_1$  and  $\lambda_2(\lambda_1 < \lambda_2)$ :

$$AAI = -100 \left( log_{10} \left( \frac{l_{\lambda 1}}{d_{\lambda 2}} \right)^{obs} - log_{10} \left( \frac{l_{\lambda 1}}{d_{\lambda 2}} \right)^{Ray} \right),$$

(1)

The *obs* and *Ray* denote the <u>J<sub>k</sub> from the</u> satellite measurement and calculated using a Rayleigh atmosphere, respectively. The longer wavelength  $\lambda_2$  is treated as reference wavelength where the surface albedo (a<sub>s</sub>) is determined by fitting the

805 observed radiance, i.e.  $\int_{a_1}^{Ray} (g_{s_0}) = \int_{a_1}^{bbs} \sum_{a_2}^{a_1} F_{a_1}^{bbs}$  is done using an atmosphere containing only molecular scattering bounded by a Lambertian surface. The spectral dependence of the surface albedo is neglected thus  $\int_{a_1}^{Ray} \sum_{a_1}^{a_2} \sum_{a_2}^{a_3} \sum_{a_1}^{a_2} \sum_{a_2}^{a_3} \sum_{a_1}^{a_2} \sum_{a_2}^{a_3} \sum_{a_1}^{a_2} \sum_{a_2}^{a_3} \sum_{a_3}^{a_4} \sum_{a_4}^{a_4} \sum_{a_5}^{a_5} \sum_{a_1}^{a_5} \sum_{a_5}^{a_5} \sum_{a_5$ 

AAI =  $100 \log_{10} \left( \frac{\Delta I_{\lambda 1}}{I_{0}^{obs}} + 1 \right)$ 

(2)

Jt is advantageous to use Eq.(2) because the AAI can be simply interpreted as the ratio between the simulated and observed 810 radiance at  $\lambda_{1,x}$ 

#### 2.2 Near-UV AAI sensitivity studies

In this section, we present results from sensitivity studies, performed with the radiative transfer model DISAMAR. DISAMAR can perform simulations of the forward L spectrum in a wide spectral coverage (270 nm to 2.4 μm) and models scattering and absorption by gases, aerosols and clouds, as well as reflection by the surface (De Haan, 2011). It uses either

- 815 the Doubling-Adding method or the Layer Based Orders of Scattering (LABOS) for the radiative transfer calculations. In this study the latter one is used, because it is less computationally intensive (De Haan et al., 1987; De Haan, 2011), DISAMAR allows to apply several aerosol scattering approximations. Here we assume Mie scattering aerosols. The parameters to describe Mie particles and their corresponding values are listed in Table 1. Considering the Chile wildfires plumes, which were dominated by biomass burning aerosols, these sensitivity studies are specifically performed for
- 820 parameterized smoke aerosols, with only fine mode particles and weak linearly wavelength dependency of the complex refractive index (nr and ni). The default values refer to observations of the daily average on January 27 of the AERONET station Santiago Beauchef (33.46°S, 70.66°W). We obtain the size distribution function and complex refractive index at 440, 675, 880 and 1018 nm from AERONET, and apply the linear interpolation / extrapolation to derive the complex refractive index over the spectrum from 340 to 675 nm, with spectral resolutions of 2 nm. Then DISAMAR uses above information to
- s25 calculate the aerosol phase function  $P(\Theta)$  and  $\omega_0$  over the full spectrum. The corresponding  $P(\Theta)$  at 354 nm is presented in Fig.2. DISAMAR requires  $\tau_1$  to be defined at reference wavelength 550 nm. Surface parameters include a spectrally flat as and the surface pressure Ps. The aerosol profile is parameterized as a single layer box shape, with its bottom at  $z_{aer}$ - $\Delta z/2$  and top at  $z_{aer}$ + $\Delta z/2$ , where  $z_{aer}$  and  $\Delta z$  are the geometric central height and the geometric thickness of the aerosol layer, respectively. The whole sensitivity analysis is performed for cloud-free conditions. The wavelength pair of OMI (354 and

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	wavelength where the surface albedo ( $a_s$ ) is determined by fin	ting [159]
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	388 nm) is applied to compute the AAI. To make different sensitivities studies comparable, the AAI calculated in this		
940	section is normalized by the maximum value among each sensitivity study. Note that each sensitivity study always uses the		
	default settings listed in Table 1 unless different values are explicitly mentioned.		
	Aerosol optical properties are determined by micro-physics, such as the real and imaginary part of the complex refractive		
	index ( $n_r$ and $n_i$ ), and the particle size ( $r_g$ ). Fig. 3 shows the variation of the AAI, $\Delta I_{a,1} L_{a,1}^{obs}$ as well as of the optical		
	properties $\omega_0$ and the asymmetry factor g, as a function of the complex refractive and the particle size. The asymmetry factor		
945	g is the averaged cosine of the scattering angle $\Theta$ , weighted by P( $\Theta$ ). Fig. 3 shows that the effect of the complex refractive		
	index is dual. As shown in Fig.3 (a), an increase in the real part of refractive index nr directly enhances the magnitude of		
	$J_{a1}^{obs}$ whereas $\Delta J_{a1}$ reduces. This results in low values of the AAI, which correspond to a large $\omega_0$ (Fig.3 (b)). Under the		
	condition that measurement angle is $\Theta$ =150°, the declining g implies that more light is scattered in the line-of-sight of the		
	detector, thus the higher Lobs Conversely, the imaginary part of refractive index m, which is directly associated with 600, has		
950	an opposite influence, see Fig.3 (c) and (d). The particle size distribution has a more complicated influence on the AAI. As		
	shown in Fig.3 (e), the AAI first decreases and then increases, when rg is varied from 0.1 to 0.4 µm. The AAI primarily		
	follows the behaviour of $\Delta I_{d,1_{0}}$ whereas $\omega_{0}$ is continuously decreasing and g is continuously increasing		
	In addition to the micro-physics, the concentration and vertical distribution of aerosols also have a strong influence on the		
	wavelength dependency of the radiance $\Delta I_{\lambda 1k}$ As shown in Fig.4 (a), the AAI is positively correlated with $\mathbf{r}_{k}$ The AAI is		
955	highly sensitive to the aerosol vertical distribution (Herman et al., 1997; Torres et al., 1998; de Graaf et al., 2005). As the		
	aerosol layer ascends (Fig.4 (b)), more molecular scattering beneath the aerosol layer is shielded, which reduces $L_{11}^{obs}$ while,		
	increases $\Delta J_{act k}$ The relation between the AAI and $z_{aer}$ is almost linear. Fig.4 (c) shows that at the same altitude, the AAI		
	slightly increases with the geometrical thickness of the aerosol layer. The reason could be that a larger $\Delta z$ indicates the		
	coming sunlight has a higher possibility to be absorbed by aerosols, amplifying the absorption of the aerosol layer.		
960	The calculated AAI does not only depend on the aerosols themselves, but also on ambient parameters such as surface and		
	clouds. Although the near-UV AAI is capable to distinguish absorbing and non-absorbing agents (Herman et al., 1997) and,		
	even to retrieve aerosol information over clouds (Torres et al., 2012), the uncertainty triggered by clouds is relatively high		
	and therefore cloudy conditions are excluded in this study. Surface conditions are parameterized by Ps and as. It can be seen		
	in Fig.5 (a) that a decrease in Ps, or equivalently an elevated terrain height, leads to less Rayleigh scattering shielded between		
965	the surface and the aerosol layer. As a result, the AAI decreases significantly due to smaller $\Delta J_{a_{1}a_{2}}$ in agreement with a		
	previous study (de Graaf et al., 2005), According to de Graaf et al. (2005), increasing as has two counteracting effects. On		
	the one hand, it increases the amount of directly reflected radiation at the top of the atmosphere, namely a larger $\frac{I_{a1}^{obs}}{a}$ on the		
	other hand it enhances the role of absorption by the aerosol layer rather than the surface, namely a larger $\Delta I_{2,1}$ . Which effect		
	of $a_s$ is decisive depends on $P_s$ (Fig.5 (b)). When the aerosol layer is relative to the sea level ( $P_s = 1013$ hPa), the first effect		
970	dominates. However, a brighter surface compensates the loss of molecular scattering shielded by the aerosols when the		
	terrain height rises ( $P_s = 813 \text{ hPa}$ ), which makes the absorbing layer more detectable.		
	The AAI depends also on the Sun-satellite geometry. Here we provide the AAI as a function of the measurement geometries		
	for the default case with the relative azimuth angle $\Delta \phi = 180^{\circ}$ . As presented in Fig.6 (a), the AAI becomes very sensitive to		
	the geometries for zenith angles larger than 60°, which confirms previous research (Herman et al., 1997; Torres et al., 1998;		
975	de Graaf et al., 2005). This is mainly due to the significant growth of $P(\Theta)$ when $\Theta$ becomes smaller (Fig.2). It is thus		
	suggested that the OMI measurement with $\theta_0$ larger than this value should be removed due to large variations in the AAI. To		
	analyse the radiance behaviour as previously, we plotted the $L_{21}^{ous}$ and $\Delta L_{21}$ as a function, of $\Theta$ along the cross section,		
	respectively (Fig.6 (b)). It is noted that $I_{\underline{s}_1}^{\underline{o}_2}$ increases when $\Theta$ is larger than $\underline{90}^{\circ}_{\underline{s}}$ whereas the P( $\Theta$ ) decreases at this range.		
0.00	(Fig.2). The reason could be that the Rayleigh scattering has an increasing contribution to the radiance at those measurement		
980	angles (backscattering).		

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#### 3 Methodology and datasets

080 In this section, we first present the datasets used and their pre-processing, followed by the strategy to retrieve the aerosol  $\omega_0$  while constraining the simulated near UV AAI to correspond to the observed one,

#### 3.1 Datasets

#### 3.1.1 OMI and GOME-2 absorbing aerosol index

The TOMS near-UV AAI retrieval has been proven a robust algorithm and applied to successive sensors, such as OMI onboard Aura and GOME-2 on-board MetOp-A/B. GOME-2 has higher spectral resolution (0.2-0.4 nm) than TOMS, but the spatial resolution is rather coarse (<u>80×40 km<sup>2</sup></u>). In this study, GOME-2 measured AAI at wavelength pair 340 and 380 nm, (http://archive.eumetsat.int) is only used as an independent dataset to assess the potential bias of the OMI measurements. OMI combines advantages of both TOMS and GOME-2. It covers wavelengths from 264 to 504 nm with a spectral resolution of approximately 0.5 nm and has a much higher spatial resolution than GOME-2 of 13×24 km<sup>2</sup> [Levelt et al.,

- 090 2006 Since OMI was launched in 2004, the AAI retrieved from this instrument has been widely used in various applications. Kaskaoutis et al. (2010) employed the OMI measured AAI for regional research of the aerosol temporal and spatial distribution in Greece. Torres et al. (2012) utilized the advantage of near-UV AAI to detect aerosols over clouds. The OMI observed AAI was even used to evaluate the impact of surface dust loading on human health (Deroubaix et al., 2013), Buchar et al. (2015) validated the NASA MERRA aerosol reanalysis with the AAI retrieved from OMI.
- D95 In this study, the OMI level 2 product OMAERO (<u>https://disc.gsfc.nasa.gov</u>) is used to provide AAI retrieved at the wavelength pair of 354 and 388 nm, and the corresponding <u>viewing</u> geometry and the surface condition <u>when the measurements took place</u>. The samples are included in the radiative transfer simulation only if 00 is smaller than 60°, and if ground pixels are not contaminated by sun-glint, clouds, row anomalies of the instrument, etc. The simulation is only applied to ground\_pixels inside the biomass burning plume, which as AAI values larger than 1, for both OMI and GOME-2.

#### 100 3.1.2 MODIS, and OMI aerosol optical thickness

MODIS on-board Aqua/Terra is a sensor that was specifically designed for atmosphere and climate research. The combination of two satellites ensures daily global coverage. The spatial resolution ranges from 250 m to 1 km and it has 36 spectral bands in the wavelength range between 400 nm and 14.4 µm (Remer et al., 2005), MODIS employs separated algorithms for aerosol retrieval over oceans and land, respectively (Tanré et al., 1997; Kaufman and Tanré, 1998; Hsu et al.,

- 105 2004: Remer et al., 2005). Currently the  $\tau$  provided by MODIS is one of the most reliable datasets [Lee et al., 2009], with an estimated uncertainty of only 3-5% over ocean and 5-15% over land [Remer st al., 2005]. Besides, the MODIS retrieved  $\tau$  is free from the uncertainty triggered by assumed aerosol profile (Satheesh et al., 2009). As mentioned before, DISAMAR requires  $\tau$  at 550 nm. This study uses cloud-filtered  $\tau$  at 550 nm from the <u>Collection 6</u> level 2 product <u>MYD04 as the input</u> for radiative transfer calculation (<u>https://ladsweb.modaps.eosdis.nasa.gov</u>).
- 110 In addition, the  $\tau$  measured by OMI and AERONET are treated as a reference dataset to evaluate potential biases in MODIS. The OMAERO retrieval uses multi-spectral fitting techniques. The retrieved  $\tau$  is in good accordance with AERONET and is highly correlated with MODIS (Torres et al., 2007), with a correlation of 0.66 over land and 0.79 over the oceans (Curier et al., 2008), although it suffers from cloud contamination due to the relatively coarse spatial resolution of OML Due to the wavelength difference, the  $\tau$  measured by OMI at 442 nm has to be transferred to 550 nm using the Ångström exponent (ÅE)
- 115 <u>440 675 nm taken from AERONET at the time when OMI flies over the selected site. The AERONET dataset used in this study is introduced in the next section.</u>

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1	3.1.3 AERONET aerosol properties		Formatted: Font: 10 pt, English (US)	
	APPONITY is an example of standard strengthened and the standard strengthened in the strengthened and	$\langle - \rangle$	Deleted: micro-physical parameters	
1.70	AERONE I is an aerosol monitoring network of ground-based sun photometers. With standardized instruments, calibration,	$\langle \rangle$	Formatted: English (US)	
170	processing and distribution, AERONET provides a long-term global database for aerosol research and air-borne and space-	Ì	Formatted: Font: 10 pt, English (US)	
	borne measurement validation. The system takes two basic measurements. The g and ÅE are retrieved from the direct solar		Formatted	[146]
	irradiance measurements; the rg, P(O) Nakajima et al., 1983; Nakajima et al., 1996), wo (Dubovik et al., 1998), nr and ni		Deleted: The	
	Dubovik and King, 2000 are derived from multiple-angular measurements of sky radiance.	>	Formatted	[147]
	The AERONET site nearest to the fire sources of 2017 Chile wildfires is the Santiago Beauchef (33.46°S, 70.66°W),		Deleted: used as inputs for radiative transfer calcu	ulation
175	(https://aeronet.gsfc.pasa.gov). The dataset in use is version 2 level 1.5 product. To minimize the influence of temporal	$ \bigvee $	Formatted	[148]
1,0	difference, the peremeters of A EPONET measured elegent to the time of the OMI everyons of the site are used to complete	$\sim$	Deleted: nearest	)
	directione, the parameters of AERONET measured <u>closes (in the prince of the original strength</u> in site are used to simulate	N	Formatted	[149]
	the optical properties of Mile scattering aerosois in DISAMAR. Note that the level 1.5 dataset is not quality-assured, and the	ľ /	<b>Deleted:</b> inversionroduct. To minimize the infl	luence of
	location of this site is in downtown of Santiago City and close to major roads. The presence of scattering aerosols may bias	$\langle \rangle \rangle$	measured nealosestr	[150]
	the measurements of the plume,	$\left( \left( \right) \right)$	Formatted	[151]
180	The AERONET direct sun product provides $\tau$ and $\omega_0$ are used to evaluate the MODIS $\tau$ and retrieved $\omega_0$ , respectively. The		Deleted: when f the OMI was flying over	[152]
	AERONET measured $\tau$ is transferred to 550 nm using the ÅE in range 440 – 675 nm while the $\omega_0$ at 550 nm is linearly		Deleted: It is notable	
	interpolated from values between 440 and 675 nm.		Formatted	[153]
	The AERONET inversion product needs to be processed into the inpute required by DISAMAR. Firstly, a conversion from		Deleted: . These facts	
	The AERONET anversion product needs to be processed into the inputs required by DiSAMAR. This y, a conversion from	$\langle     \rangle$	Deleted: , which	
	the volume size distribution $V(r_v, \sigma_v)$ provided by AERONET to the number size distribution $N(r_g, \sigma_g)$ used in DISAMAR is	//	Formatted	[154]
185	required:	- // )	Formatted: Font: 10 pt, English (US)	
	$N(n, \sigma) = V(n, \sigma)^{-3} e^{-45\sigma^2}$ (4)	$\langle \rangle$	Deleted: measurements	
	$\mathbf{N}(I_g, \sigma_g) = \mathbf{V}(I_v, \sigma_v) \frac{1}{4\pi u_g^3} e^{-i\mathbf{x}} \mathbf{x}, \tag{4}$	_	Formatted	[155]
	The following relation between the geometric and volumetric mean radii (r, and r) and standard deviations ( $\sigma$ , and $\sigma$ ) is	7	Formatted	[156]
	assumed:			
	$\underline{\chi}_{g_{\lambda}} = \underline{\chi}_{p} e_{\lambda}^{-3\sigma_{g}^{2}}, \tag{5}$		Formatted	[157]
190	$\mathcal{G}_{\boldsymbol{\theta},\boldsymbol{k}} = \mathcal{G}_{\boldsymbol{\theta},\boldsymbol{k}}, \tag{6}$		Formatted	[158]
	The fine and coarse mode particle size are derived by finding the two peaks of the log-normal distribution function provided		Deleted: separately from AERONET	
	by AERONET. The complex refractive index is assumed the same for both modes. Since bi-modal aerosol is not applicable			
	in DISAMAP yet, we first coloulate enticel properties of two modes individually, then we externally combine the enticel		Beletede W. C. ( DIGAMAR)	
	in Disamark yet, we instructeduate optical properties of two modes menvioually, then we externally combine the optical		Deleted:ww first use DISAMAR to	[159]
	properties of two modes into a bi-modal aerosol, with a fraction;		<b>Deleted:</b> , and the optical properties ofnto a bi- aerosolswith a fraction in DISAMAR are calculated	modal ted by externally
195	$w_{r} = \frac{N_{f}(r_{g,f}\sigma_{g,f})}{(7)}$		mixing the derived modes with a fraction	[160]
175	() N[x_a, g_a, a) + N = (x_a, g_a, a), ()		Formatted	[161])
	$w_{e_{a}} = 1 - w_{f_{a}},$ (8)		Formatted	[162]
	Then the weights for calculating the total $\omega_{0}$ of the mixed aerosol are:		Formatted	[163]
	$w_{c} = \frac{w_{f}\sigma_{f}}{(2\pi)^{3}} \tag{9}$	1	Formatted	[164]
	Wifither,			[104]
	$w_{\sigma,c} = 1 - w_{\sigma,f_k},\tag{10}$		Formatted	[165]
200	Where the $\sigma_f$ and $\sigma_c$ are the extinction cross section of the fine and coarse aerosols. The expansion coefficients of the mixed			
	aerosol is weighed by the $\omega_0$ of the fine and coarse aerosols ( $\omega_{0,f}$ and $\omega_{0,c}$ ), respectively:			
	$w_{\omega_0,f} = \frac{w_f \sigma_f \omega_{0,f}}{w_f \sigma_f \omega_{0,f} + w_f \sigma_f \omega_{0,f}},\tag{11}$		Formatted	[166]
	A. A			
	$w_{\omega\alpha c_{\star}} = 1 - w_{\omega\alpha f_{\delta}} \tag{12}$		Formatted	[167]

The spectral bands of the AERONET instrument at this site only cover the visible band. Due to the absence of observations, assumptions have to be made on the spectral dependence of aerosol properties to obtain their values in the near-UV band. The properties of biomass burning aerosol depend on the type of fuel, the procedure producing the smoke, the age of the smoke,

- and also the atmospheric conditions (Reid et al., 2005). Using measurements to constrain the input aerosol refractive index can / reduce the uncertainties due to a-priori knowledge. Our treatment on the complex refractive index is as following: (1) take the complex refractive index at visible band (440 to 675 nm) from AERONET measurements; (2) linearly extrapolate the complex refractive index to near-UV band. The real part n<sub>x</sub> for radiative transfer calculation is obtained in this step. A slight wavelength
- 250 dependence of n<sub>r</sub> is found from the measurements; (3) for the imaginary part n<sub>k</sub>, we multiply it (for the entire wavelength from UV to visible) with a scaling factor as we set it as a free parameter. By varying the value of the scaling factor, both the magnitude and the wavelength dependence of n<sub>i</sub> can change to meet the requirement of retrieval.

#### 3.1.4 CALIOP backscattering coefficient

The CALIOP on-board CALIPSO, which was launched in 2006, provides high-resolution profiles of aerosols and clouds. It has three channels with one measuring the backscattering intensity at 1064 nm and the rest measuring orthogonally polarized components at 532 nm backscattering intensity (Winker and Omar, 2006). Due to the limited spatial coverage, CALIOP did not observe the <u>Chile</u> plume for all the cases for which we have OMI observations. We only use the total attenuated backscatter at 532 nm from level 1B Version 4.10 Standard data to evaluate the parameterized aerosol profiles (https://eosweb.larc.nasa.gov/project/calipso).

#### 260 3.2 Methodology

In this study, we employ the radiative transfer model DISAMAR to simulate the near-UV AAI from OMI and to derive the  $\omega_0$  for a specific case, i.e. the Chile wildfires in January 2017. We select the period from 26 to 30 January 2017 (28 January is excluded due to lack of data) when the AAI value reached its peak.

- The forward simulation consists of two major steps. First, DISAMR calculates the Mie aerosol optical properties with

   265
   aerosol micro-physical information taken from AERONET measurements (rg, nr and ni). As mentioned in Section 3.1.3, we set the spectral-dependent imaginary part ni as a free parameter to vary ω₀. Then, DISAMAR operates radiative transfer calculation with the calculated aerosol optical properties for a specific aerosol and environmental conditions.

   It is noted that the observed aerosol vertical profiles is limited for the Chile wildfires. Previous research suggested AAI cannot be quantitatively used without τ or z<sub>met</sub> information (Gassó and Torres, 2016). Instead, we implement the same
- 270 parameterization as in the sensitivity study to obtain the aerosol profile. Since the AAI dependency on Δz is minor (Fig.4 (c)), and to reduce the computational cost, Δz is set constant of 2 km based on the information from the CALIOP measurements of backscattering coefficient (β) at 532 nm (Fig.7). The z<sub>aer</sub>, to which the AAI is highly sensitive, is treated as an unknown, variable to be retrieved together with ω<sub>0</sub>.
- Consequently, with various combinations of z<sub>aer</sub> and n<sub>a</sub> a lookup table (LUT) of the calculated AAI is constructed with
   DISAMAR. It should be noted that for all ground pixels in the plume we assume the same aerosol microphysical properties as well as the same vertical profile. Pixels outside the plume may have had significantly different properties and this will affect the results. <u>But as shown in Fig.8</u> the distribution of OMI measurements is sparse in space, which implies that the dataset is quite sensitive to geographical outliers that may cause the heterogeneous properties of the plume, Consequently,
- we apply a data quality control procedure before retrieving or First, we manually remove the pixels that are geographically
  isolated from the main plume. Furthermore, we remove the potential outliers based on statistical tool. We filter the dataset
  using an outlier detection based on the interquartile range (IQR) of the AAI difference between DISAMAR simulations and
  OMI measurements. According to Tukey's fences (Tukey, 1977), an AAI difference falling outside range between Q1-1.5
  IQR and Q3+1.5 IQR may be regarded as an outlier and removed, where Q1 and Q3 are the first and third quartiles of the

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AAI difference, and the IQR is the range between Q1 and Q3. Only the data passing the outlier detection criterion is used to calculate the cost function  $(Eq.(\beta))$ :

DMSE -	$\left[\sum_{i}^{n} \left(AAI_{DSM,i}^{qualified} - AAI_{OMI.i}\right)^{2}\right]$
KMSE –	n,

B50 Here AAI<sub>i</sub> indicates the AAI for *i*th ground pixel of the selected OMI data; subscripts DSM and OMI indicate DISAMAR simulation and OMI observation, respectively. The combination of z<sub>aer</sub> and 600 that leads to the minimum residue is used to simulate the AAI.

Finally, the simulated AAI is compared with OMI observations. We also employ the independent data from GOME-2 on MetOp-A/B as a reference to <u>identify</u> the potential bias of OMI. Similarly, the  $\tau$  retrieved from OMI and AERONET serves

as a reference to that of MODIS. The estimated aerosol profile and <u>600 at 550 nm</u> are evaluated with independent observations from CALIOP and AERONET, respectively.

#### 4 Results and discussion

By applying the methodology described in the previous section, we quantitatively retrieved the aerosol profile and  $\omega_0$  of the Chile 2017 wildfires by AAI simulation. The OMI measurements of the plume are displayed in Fig.8 (a) – (d). The presented

- ground pixels are with AAI value larger than 1, and are free of cloud contamination, sun-glint and yow anomaly of the instrument. Fortunately, the remaining data is still able to capture the main plume features. It can be clearly seen that from 26 to 30 January, the plume produced by wildfires in the central Chile was transported by the south-easterly trade wind from the continent towards the lower latitude region of the Pacific Ocean. The plume travelled over <u>a distance of 3000 km during the period</u>.
- 365 The vertical movement of the plume is given by CALIOP backscattering coefficient measurements (β) at 532 nm (Fig.7). The CALIOP paths closest to the plume are marked by a black dashed line in Fig.7. If is noted that CALIOP did not always measure the plume, and may even fail to capture the elevated plume, e.g. on 26 January. The aerosol layer captured by CALIOP is distributed from 2 km to 6 km, with an average height at approximately 4-5 km. The ascent of the plume was driven by the heat generated by the fires and sunlight absorption, as well as the atmospheric vertical motions.
- Fig.8 (e) (h) show the AAI simulation selected by the data quality control mentioned in Section 3.2. The spatial distribution of the simulated AAI shows similar patterns as the OMI observations. Some data points that are geographically isolated from the plume, e.g. in case 26 and 30 January, differ strongly from what are observed inside the plume. Including these outliers in the optimization could bias the retrieved aerosol properties. This can also be seen in Fig.8 (i) (l), where the points passing the data quality control described in Section 3.2 are highlighted in red color. By removing the outliers, the average spatial correlation coefficient reaches 0.90.
- Table 2 lists the statistics of the qualified AAI data, in terms of the median, relative difference and RMSE. The median of measured AAI ranges from 2 to 4 during the research period. Except 26 January, the median of simulated AAI in other cases js in good agreement with the measurements, with relative differences within ±6%. The low RMSE confirms the high spatial consistency between the simulations and the observations. The majority of the simulated AAI of 26 January is negatively
- biased, which is reflected by the small slope without an intercept correction in Fig.8 (i). A systematic bias in the inputs might cause this result. In terms of  $\omega_0$ , both the AERONET measured and the AAI retrieved aerosol absorption become weaker with time (Table 2). Although the simulated and observed AAI are in good agreement, the difference in  $\omega_0$  is significant. The mean of the retrieved  $\omega_0$  at 550 nm for the whole period is 0.84, contrast to the AERONET measurements with a mean value of 0.90.

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	There are many sources accounting for this discrepancy. First of all, the nearest site Santiago_Beauchef is not exactly in the
425	primary biomass burning regions as mentioned in section 3.1.3. The AERONET site is located in downtown, where
	reflective urban or industrial aerosols may have been mixed with the smoke enhancing the way. This would also affect the
	complex refractive index used in radiative transfer calculation, since we use the AERONET measured refractive index to
	constrain the forward simulations. According to Table 2, the retrieved ni reveals that the difference between 354 and 388 nm
	is less than 5%. This small spectral dependence of $n_i$ is mainly determined by AERONET measurements in the visible band

- 430 (dashed lines), whereas the effect of the scaling factor is minor in this case (Fig.9). We thus find a much weaker wavelength dependence than in the Jethva and Torres (2011) study, where a 20% difference between the two UV wavelengths was applied to OMAERUV algorithm to achieve the result that 70% of the retrieved  $\omega_0$  differ less than  $\pm 0.03$  from the  $\omega_0$  from the AERONET measurements. A stronger spectral dependence of ni between 354 and 388 nm would allow simulations to reach a higher AAI while keeping  $n_i$  at a relatively low level, so would retrieve a higher  $\omega_0$  at 550 nm. The presence of non-
- absorbing aerosols weakens the spectral dependence (particularly in the UV spectral range) and the linear extension would overestimate the aerosol absorption. In this situation, the uncertainties due to assumed spectral aerosol properties might compensate the measurement errors to some extent. Furthermore, the AERONET inversion product is not error-free. The uncertainty of size distribution retrieval is minor for biomass burning aerosols (Dubovik et al., 2000), but under optically thick circumstances, when retrievals are quality-assured, the reported accuracy of complex refractive index is 0.04 for nr and
- 440 30%-50% for ni, respectively (Dubovik et al., 2002). It is also reported that AERONET tends to underestimate the absorption of biomass burning aerosols compared with in situ measurements (Dubovik et al., 2002; Reid et al., 2004). The uncertainty of  $\omega_0$  is 0.03 under high aerosol loading ( $\tau_{440} > 0.5$ ) and 0.05-0.07 under low aerosol loading (Dubovik et al., 2002; Holben et al., 2006).
- Apart from AERONET itself, information from other datasets could also bias our estimate of aerosol absorption. Among all the inputs, the parameterization of a one-layer box-shape aerosol profile could be the largest error source. Although the influence of  $\Delta z$  on the AAI is small (Fig.4 (c)), the AAI calculation highly depends on  $z_{aer}$  (Fig.4 (b)). As shown in Table 2, the estimated plume altitude varies from 4.5 to 4.9 km. As the black solid line indicated in Fig.7, the retrieved  $z_{aer}$  can capture the measured geometric vertical location of the plume. The zaer on 26 January seems overestimated because of the
- temporal and spatial difference. Concretely, CALIOP sampled the plume near the sources and close to the surface, while the 450 plume observed by OMI had been already elevated and transported to the open ocean. The lack of information on the real plume height makes it challenging to determine whether the plume height is responsible for the systematic bias in Fig.8 (i). Except for 26 January, zeer is in good agreement with what CALIOP observed. Although the retrieved aerosol profiles are convincing to some extent, one should keep in mind that CALIOP and OMI observations are not exactly co-located. Besides, the parameterized aerosol profile may fail to represent the spatial variation of the plume. Therefore, the uncertainty cannot be 455 directly determined due to the lack of validation observations.
- Among the four days for which we retrieved  $\omega_0$ , the value for 27 January is significantly lower than others. For this day the agreement with CALIOP is reasonable and also the CALIOP track is not far away from the OMI measurement. We therefore explore the effect of measurement biases in AAI and  $\tau$  on the retrieved  $\omega_0$ . We investigate the potential bias of these two datasets by plotting the histogram of the AAI measurement difference between GOME-2 and OMI (Fig. 10, (a)), against the  $\tau$
- measurement difference between MODIS and OMI (Fig. 10, (b), both are converted into 550 nm). It is clear that on 27 January, the AAI from OMI seems to be overestimated compared to GOME-2, Although the difference in wavelength pair choice for AAI retrieval, measurement time and condition, etc., could contribute to the AAI discrepancy between GOME-2 and OML exploring the difference between the two datasets is beyond the scope of this study, In aspect of input aerosol concentration, the  $\tau$  from MODIS could be potentially underestimated. Fitting a higher AAI with a lower input  $\tau$  leads to an
- 465 overestimation in aerosol absorption. <u>Here, we analytically quantify the impact of  $\tau$  for this specific case by systematically</u> enhancing the  $\tau$  of MODIS with a constant variation ( $\Delta \tau$ ) added to all <u>pixels</u>, with the AAI level and the aerosol profile

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remain unchanged. Fig.  $1Q_{4}(c)$  presents how the AAI RMSE and the estimated  $\omega_{0}$  respond to the enhanced  $\tau$ . It can be clearly seen that an increase in overall  $\tau$  level by 0.07 raises  $\omega_{0}$  to 0.84, and optimizes the AAI simulation to a RMSE less than 0.45. If we apply this  $\tau$  adaption, the retrieved  $\omega_{0}$  of 27 January becomes more consistent with the other days. Apart from the observational errors in AERONET, OMI and MODIS data, the assumption that the plume features are homogeneous could also result in the discrepancy between AAI retrieved and AERONET measured  $\omega_{0}$ . In reality, the plume altitude, the optical properties and even the chemical compositions could vary in space and time, while our simulations

610 cannot take into account those effects.

#### 5 Conclusions

Biomass burning is a major source of absorbing aerosols making a significant contribution to climate warming. Quantitatively characterizing the absorption by biomass burning aerosols is therefore important to reduce the uncertainty in assessments of global radiative forcing. Facing the lack of long-term  $\omega_0$  records, this study <u>explores</u> an approach to retrieve

- 615 000 based on reflectivity in the near-UV channel measured by OMI. Although AAI is not a geophysical parameter and depends on many <u>factors</u>, its independence from pre-defined aerosol types, its high sensitivity to aerosol absorption as well as its long-term data record, makes it an attractive <u>quantity to aerosol research</u>.
- We test the retrieval of ω<sub>0</sub> for the wildfires happening in central Chile in January 2017. After filtering the data from outliers, a high spatial correlation coefficients (0.85 to 0.95) reaches between the simulated and observed AAL. The retrieved aerosol
   profiles indicate the plume was elevated to height of 4.5-4.9 km during the research period. These results are in agreement
- with CALIOP measurements. This <u>average of the retrieved</u>  $\omega_0$  at 550 nm is approximately 0.84, which is 0.06 lower than that of AERONET retrieval. The retrieved  $\omega_0$  is <u>out of</u> the typical uncertainty in the  $\omega_0$  retrieved from AERONET (±0.03). The <u>sources for</u> discrepancy <u>includes</u>; the location of the AERONET site that may bias the measured  $\omega_0$  and the spectral <u>dependency of complex refractive index</u> the simplified parameterization of the aerosol profile; the observational errors in
- 525 the input aerosol micro-physics τ, as well as AAI; and the assumption of homogeneous and static plume properties, which ignores the plume evolution over space and time. We quantitatively analyze the uncertainty of τ for a specific case (27 January) when the estimated aerosol profile is in good agreement with the CALIOP measurements. An improvement in retrieved ω<sub>0</sub> can be seen by adapting the magnitude of aerosol concentration.
- This study proves the potential of utilizing OMI measured AAI to quantitatively characterize aerosol optical properties like
   630 ω<sub>0</sub>. Currently, it is challenging to retrieve and validate results without reliable aerosol profile information. In the future, the availability of daily global aerosol profile data, e.g. the L2 aerosol layer height product TROPOspheric Monitoring
   Instrument on-board Sentinel-5 Precursor (TROPOMI) that is underdevelopment (Sanders and de Haan, 2016), are expected to provide a stronger constraint on the forward calculation and to significantly reduce the uncertainty in the retrieved aerosol properties. Perhaps, more sophisticated assumptions on spectral-dependent aerosol absorption (e.g. steeper gradient of n, in
- 635 UV than visible band) have to be made and evaluated by other observational aerosol properties in UV spectral range, e.g. AERONET measured  $\tau$  in UV band, instead of only depending on measured refractive index in visible band,

#### Acknowledgements

This work was performed in the framework of the KNMI Multi-Annual Strategic Research (MSO). <u>The authors thank to</u> <u>NASA's GES-DISC, LAADS DAAC and ASDC for free online access of OMI, MODIS and CALIOP data. The authors also</u> thank to the <u>Centre for Climate Resilience Research (CR)2 at University of Chile (CONICYT/FONDAP/15110009)</u> providing the data of the Santiago Beauchef AERONET station.

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### Table 1. Parameters used in sensitivity analysis.

Parameters	Default value	Sensitivity range	Unit	
Geometric mean radius (rg)	0.15	0.1, 0.15, 0.2, 0.25, 0.3, 0.35, 0.4	μm	-
Geometric standard deviation ( $\sigma_g$ )	1.5		μm	
Real refractive index (n <sub>r</sub> ) at 354 nm	1.5	1.3, 1.35, 1.4, 1.45, 1.5		
Imaginary refractive index (n <sub>i</sub> ) at 354 nm	0.06	0.04, 0.06, 0.08, 0.1		
Aerosol layer geometric central height (zaer)	4.5	2.5, 4.5, 6.5, 8.5	km	
Aerosol layer geometric thickness(Δz)	1	0.5, 1, 1.5, 2	km	
Aerosol optical thickness (τ) at 550 nm	1	0.5, 1, 1.5, 2		
Surface albedo (a <sub>s</sub> )	0.05	0.05, 0.1, 0.5, 1.0	-	•
Surface pressure (P <sub>s</sub> )	1013	1013, 963, 913, 863, 813	hPa	•
Solar zenith angle $(\theta_0)$	30	0, 15, 30, 45, 60, 75	°	
Viewing zenith angle ( $\theta$ )	0	0, 15, 30, 45, 60, 75	°.	
Relative azimuth angle ( $\Delta \varphi = \rho - \varphi_0 + 180^\circ$ ).	0	$0, \pm 45, \pm 90, \pm 135, \pm 180$	Ŷ	



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Table.2 Summar	v of	retrieved	results	(applying	IOR	outlier de	etection).
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L	Date	2017-01-26	2017-01-27	2017-01-29	2017-01-30	
AAI	AAI median (OMAERO)	2.52	2.38	4.05	2.61	
	AAI median (DISAMAR)	2.17	2.48	3.81	2.49	
	Relative difference (%)	-13.88	4.20	-5.93	-4.60	
	RMSE	0.67	0.51	0.60	0.41	
Aerosol	Zaer [km]	4.9	4.5	4.7	4.7	
profile	Δz [km]	2				
<u>n</u> ;	<u>n<sub>i</sub> at 354 nm</u>	0.0395	0.0382	0.0388	0.0314	
	<u>n<sub>i</sub> at 388 nm</u>	0.0386	0.0366	0.0373	0.0306	
	Ni difference between 354	<u>2.33%</u>	<u>4.37%</u>	4.02%	2.61%	
	and 388 nm					
ω <sub>0</sub> at 550 nm	ω <sub>0</sub> (AERONET)	0.89	0.89	0.92	0.91	
	ω <sub>0</sub> (DISAMAR)	0.83	0.81	0.87	0.85	
	Relative difference (%)	<b>-6.74</b>	-8.99	-5.43	-6.59	

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The absorbing aerosol index (AAI) base	d on the near Ultra-Violet (near-UV) rem	ote sensing techniques is a
qualitative parameter that allows to retrie	ve aerosol optical properties with confide	ence
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$\mathbf{F}$ (10) (A $\cdot$ ) (1) (0)	(0.1) = 1.1 (110)	

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slightly smaller than the value of 0.90	measured independently by the AERON	ET instrument.
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Except for the observational errors, the	impact of remaining error sources on $\omega_0$	retrieval is difficult to quantify.
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They consist of fine particles (aerodyna	amic diameter smaller than 2.5 $\mu$ m) that l	have adverse impacts on the
environment and human health (Bäume	er et al., 2008; Adler et al., 2011). Bioma	ss burning aerosols
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), one type of absorbing aerosol, black car	bon (BC), can be considered as the	second important warming agent after
carbon dioxide. Absorbing aerosols heat t	he atmosphere primarily by interact	ion with solar radiation. They directly
absorb the incoming or reflected sunlight.	They are also able to reduce the ref	lectivity of the planet by depositing on
absorb the incoming or reflected sunlight. bright surfaces	They are also able to reduce the ref	lectivity of the planet by depositing on
absorb the incoming or reflected sunlight. bright surfaces Page 12: [20] Deleted	They are also able to reduce the ref	lectivity of the planet by depositing on 4/23/18 2:09:00 PM
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Quantifying the climate effect of absorb	ing aerosols is therefore important.	
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As a result,  $\omega_0$  is usually retrieved by forward simulations that are adapted to observational parameters. Many implementations have been done for ground-based network measurements (Dubovik et al., 1998; Eck et al., 2003; Petters et al., 2003; Kassianov et al., 2005; Corr et al., 2009; Yin et al., 2015), while relatively fewer applications to satellite instruments exist due to lack of validation (Lee et al., 2007; Ialongo et al., 2010; Eck et al., 2013). Moreover, a majority of those methods heavily depend on the aerosol optical thickness ( $\tau$ ), either in forward model simulations or in validation procedures. This makes the derived  $\omega_0$  subject to large uncertainties. The reason is that  $\tau$ retrieval requires assumptions on aerosol types, and the commonly used  $\tau$  that is retrieved in the visible band where the signal of bright surfaces is strong. Besides, the aerosol effect on radiance is inversely proportional to wavelength (Kaufman, 1993), and the sensitivity to  $\omega_0$  is not significant for most  $\tau$  measurements in the visible and infrared band (Kaufman et al., 1997).

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$\Gamma = 1.1 (110)$		

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, the sensitivity of $\tau$ in the visible band to $\sigma$	00 is lower over dark surfaces (Kau	ufman et al., 1997), while

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Empirical models were also developed to build connections between the AAI and parameters it depends on. Hsu et al. (1999) found a linear relation between the TOMS retrieved AAI and Sun-photometer measured  $\tau$  over regions with biomass burning and regions covered by African dust. Ginoux and Torres (2003) implemented an empirical relation between the AAI retrieved from TOMS with  $\tau$ ,  $\omega_0$  and surface pressure (P<sub>s</sub>) to characterize the dust aerosols. Although requiring less computational cost, applying these empirical models is either limited by specific conditions or subject to large errors. Thus, these methods have not been widely used.

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is valid. This The way the longer wavel	ength $\lambda_2$ is treated as reference wavelen	gth where the surface albedo $(a_s)$ is
determined by fitting the observed radia	ance, i.e. $I_{\lambda 2}^{Ray}(a_s) = I_{\lambda 2}^{obs}$ . This $a_s$ is also	b assumed used at $\lambda_1$ to compute $I_{\lambda_1}^{Ray}$ .
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Given size distribution function (rg), com	plex refractive index (nr and ni) at spe	ecific wavelengths and a certain
wavelength interpolation method, DISAN	MAR calculates the spectrally dependent	ent optical properties (e.g. $\omega_0$ and
phase function $P(\Theta)$ ) within the specified	wavelength range. In this study, we	use the linear interpolation and the
spectrum coverage from 340 to 675 nm.		
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The asymmetry factor g is the averaged	d cosine of the scattering angle $\Theta$ , weighted by P( $\Theta$ ).	
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As shown in Fig.3 (e) and (f), even with	a decreasing $\omega_0$ and an increasing g, or	alternatively a decreasing $I_{\lambda 1}^{obs}$ , the
AAI primarily follows the behaviour of	$\Delta I_{\lambda 1}$ . The significant reduction in the sp	bectral dependency of $I_{\lambda}$ overwhelms
the high reflectivity for small particles (	$r_g=0.1\mu m$ ).	
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corresponding to the selected $\Theta$ does not st	rictly follow the changes in $P(\Theta)$	
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the reason that the length of the light path the	rough the aerosol layer also varie	es with the measurement geometry.
Although the overall change in $P(\Theta)$ with a	n increasing $\Theta$ is negative, the lig	ht path within the aerosol layer also
decreases. Less absorption occurring in the	aerosol layer overwhelms the dec	crease in reflectivity for larger $\Theta$ ,
resulting in an increase in $I_{\lambda 1}^{obs}$ with $\Theta$ . [Sj1]		-
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I reconsider the reason, and also set (not included in the manuscript). It to direction is strong.	up a simulation under the saturns out the Rayleigh scatter	me condition but without aerosol ing at forward and backward
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burning regions as mentioned in section 3.1	.3. The $\omega_0$ measured by AERON	ET could increase as the result of
aerosol ageing. Specifically, the location of	the AERONET site is in the dow	ntown, where the more reflective urban
or industrial aerosols may mix with the smo	bke and enhance the measured $\omega_0$ .	Besides
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However, even with relative reasonable retrieval of  $z_{aer}$ , it is noted that the  $\omega_0$  retrieved on 27 January is significantly underestimated and biased from the mean level of other cases. This implies the existence of other error sources, such as the observational errors from the input  $\tau$  of MODIS and the AAI of OMI to be fit.

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This study proves the potential of utilizing OMI measured	sured AAI to quantitatively characterize	e aerosol optical
properties like $\omega_0$ . Even without direct observation of	faerosol profiles, this parameter can also	o be retrieved with quite
good confidence. However, apart from the observation	nal uncertainties, the current study is pr	obably somewhat
limited by the necessary assumptions of homogeneou	s and static plume properties, whose im	pact on retrieved $\omega_0$ is
difficult to quantify. In the future planned		
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work, a chemistry transport model is needed to describe the evolution of the plume properties in space and time. Moreover, also clouds should be taken into consideration in order to use the AAI observations over clouds, thus making the maximum use of the near-UV observations.

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