The CU 2D-MAX-DOAS instrument - part 2: Raman Scattering Probability Measurements and Retrieval of Aerosol Optical Properties

Ivan Ortega[1,2], Sean Coburn[1,2], Larry K. Berg[3], Kathy Lantz[2,4], Joseph Michalsky[2,4], Richard A. Ferrare[5], Johnathan W. Hair[5], Chris A. Hostetler[5], and Rainer Volkamer[1,2]

[1]Department of Chemistry and Biochemistry, University of Colorado, Boulder, CO, USA

[2]Cooperative Institute for Research in Environmental Sciences (CIRES), Boulder, CO, USA

[3]Pacific Northwest National Laboratory, Richland, WA, USA

[4]Global Monitoring Division, Earth System Research Laboratory, NOAA, Boulder, CO, USA

[5]NASA Langley Research Center, Hampton, VA, USA

Response to Reviewer 2; 12 April 2016

Black: Referee's comments
Blue: Author's reply
Green: sentence added/modified in the manuscript

We thank Reviewer 2 for the helpful comments and suggestions.

Ortega et al., describe derivation of aerosol optical depth and Henyey-Greenstein asymmetry factor from solar almucantar measurements (-170_ to 170_, 5_ steps) of Rotational Roman Scattering probability. The method applies DOAS technique to hyperspectral intensity measurements to derive differential RSP in 426 – 440 nm window and therefore does not require absolute radiometric calibration. The authors present radiative transfer simulations at 430 nm using Monte Carlo model to demonstrate sensitivity of the RSP to AOD, H-G asymmetry factor, aerosol profile, relative solar azimuth angle, and solar zenith angle. They conclude that RSP is independent of the aerosol profile and has low dependence on single scattering albedo and surface reflectivity. On the other hand, RSP has high sensitivity to total AOD and H-G asymmetry factor especially at small RSAA, small AOD and large SZA. Based on these simulations they develop a method to minimize difference between retrieved (426 – 440 nm) and simulated RSP at 430 nm. Direct sun spectrum is used as a reference Fraunhofer spectrum to minimize amount of RPS in the reference spectrum. RSP in the reference spectrum is derived from Langley plot analysis of the zenith and direct sun spectra. The method is applied to 2 days, one with low and one with high AOD, during TCAP filed campaign (1 July – 13 August 2012). The retrieved AOD are compared to co-located measurements by CIMEL, MFRSR, and HSRL-2. Reasonable agreement in diurnal variability is achieved between CU 2D-MAX-DOAS, CIMEL, and MFRSR. The method is well described and the paper is well organized. I recommend publishing the paper after some modifications.

Major comments:

1. One of the main assumptions of the method is that solar almucantar measurements of RSP are independent of aerosol profiles based on the simulations at SZA 35˚ and 70˚. This might not hold for all SZA, all G-H asymmetry factors and SSA, and especially more realistic aerosol phase functions. I would recommend expanding the sensitivity studies to aerosol profiles to include 20˚, 35˚, 70˚, 80˚ and 85˚ SZA for G-H asymmetry factors 0.64 and 0.72 and SSA 0.85 and 0.98.

We have conducted simulations to show the sensitivity of AOD and aerosol extinction profile shapes at SZA of 85˚ (EA = 5˚) as suggested – Similar as Figures 4 in the manuscript and Figure S3 in the supplement. The results of this sensitivity are shown in Figure R1. As can be seen the same effects are found: high sensitivity towards AOD and low sensitivity towards aerosol extinction profile. It is quite interesting that some differences are noticeable when the aerosol extinction is aloft.

The discussion of the sensitivity towards aerosol profiles and higher SZAs (low EAs) has been expanded based on a similar comment of reviewer #1 (please see Figure R4 and the discussion in our response to reviewer 1).

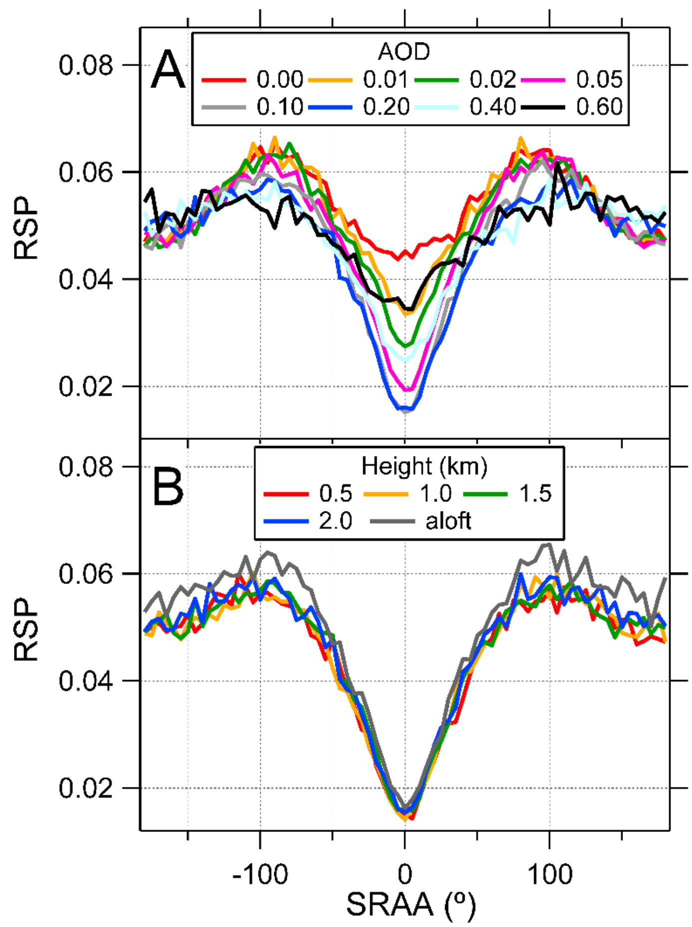

Figure R1. Sensitivity study showing that simulated RSP (430 nm) strongly influenced by (A) AOD, and insensitive to (B) aerosol vertical distribution. (A) AOD is varied, keeping aerosols homogeneously distributed (box profile) up to 1.5 km altitude. (B) The aerosol extinction vertical distribution is varied for a constant AOD of 0.2. The simulation is for SZA = 85˚, SSA = 0.98, g = 0.70, SA = 0.05.

Our statement were not correct for the ranges of SZA we discuss. We incorporated in the revised manuscript the next paragraph:

The sensitivity studies in Figs 3, 4, and in the supplement confirm that the RSP does not depend on the aerosol vertical distribution for SZA smaller than 80˚. Note that all of the azimuth scans here were conducted at solar EA, which for measurements at SZA < 80˚ corresponds to EAs of 10˚ or higher. For measurements at low EAs the RSP becomes slightly dependent on the aerosol vertical distribution (see Fig. S7 panel C).

We additionally incorporated Fig. R1 into Fig. S6 in the revised supplemental information. The range of 0.64 to 0.75 G-H have been shown in Figures 5 and same results are found at other SZA, which are captured by the look up table. As shown in section 2.4.2 the RSP does not show a significant variability among different SSA. Furthermore, in this study we use known SSA based on co-located observations.

2. Please discuss the effect of G-H phase function approximation on the AOD retrieval compared to a more realistic Mie phase function for different aerosol types?

We refer the Reviewer to our detailed response to Reviewer #1 (see comment A), and the new Section 3.3.3 that compares the HG phase function with Mie phase functions constrained by Aeronet observations.

3. I think that error estimation is overly optimistic especially at small SZA and small RSAA when dRSP are very small and "close" to the reference spectrum. The change in dRSP and its error do not change linearly with AMF especially for dAMF<0.5 from AMFref therefore the error in RSPref is larger then presented (0.0018). I think that more reasonable will be to either assume no RSP in the direct sun reference spectrum, or to model RSP with an RSP error equal to the RSPref itself currently derived in the paper (0.0044).

Reviewer #1 suggested a third method to quantify the RSP in the reference. We have applied this method (see revised Section 3.1), and found it supports the error bounds that we use and propagate in the paper.

We respectfully disagree that the error estimation is overly optimistic, and provide additional evidence on the robustness of the RSP fit and it's the error in RSPref below. The results support that the reported error is in fact estimated conservatively, and limited by the error in the determination of the RSP in the reference. The final error in the determination of AOD is calculated with the error propagation of the DOAS fit error and the error in the reference RSP.

Figure R2 shows RSP fit examples (top) and residuals (bottom) at SRAA of (A) 5˚ and (B) 140˚. On both examples the RSP fit error is smaller than the 0.0018 conservative error used in the manuscript. This presents additional evidence that the error in the RSP does not depend significantly in the SRAA (see also Figures R5 and R6).

Figures R3 and R4 shows the possible systematic errors quantified by changing the wavelength intervals and polynomial orders in a similar way as performed by Vogel et al. (2013) – Similar as Figure S1 and S2. From these figures it is clear that the difference with respect to the actual fitting settings are lower than 8% (RSP error of 0.0012) for SRAA of 5˚ and lower than 3-4% (RSP error of ~0.0015) for SRAA of 140˚. Hence, the fit uncertainty for RSP is lower than the 0.0018 reported in the manuscript.

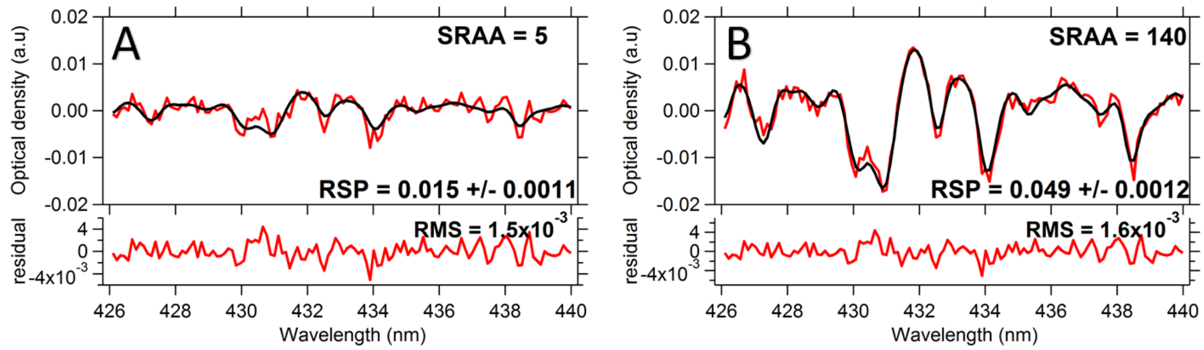

Figure R2. Same as Fig. 2 in the main text but for SRAA of (A) 5 and (B) 140.

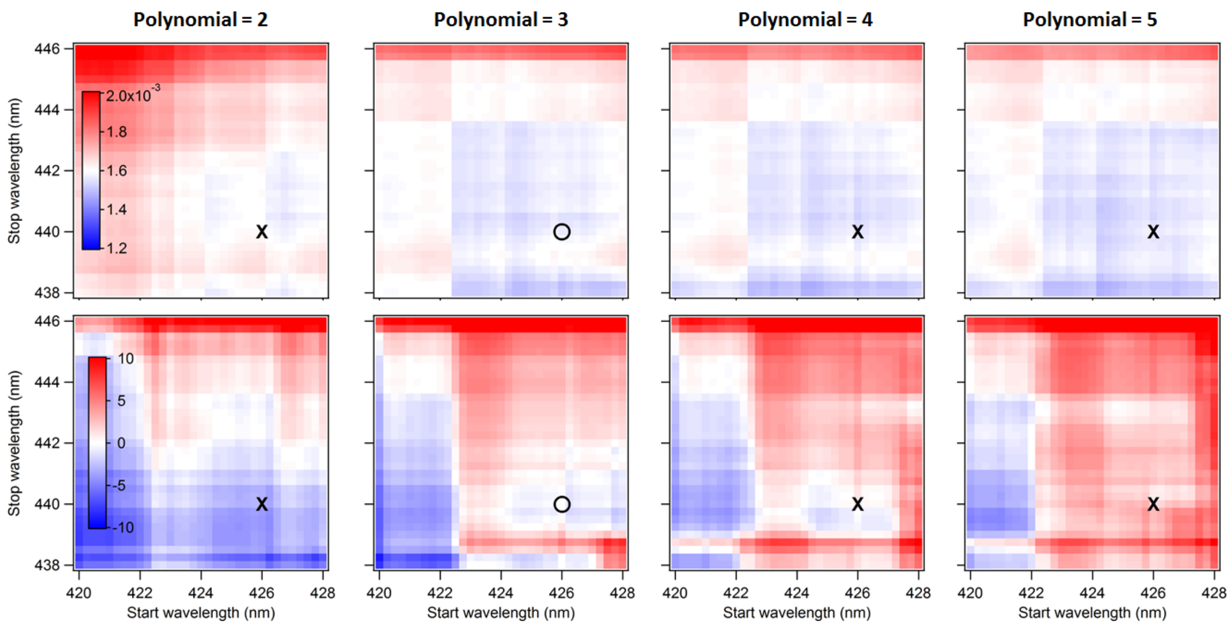

Figure R3. Same as Figure S1 but for SRAA = 5˚ and SZA = 60˚.

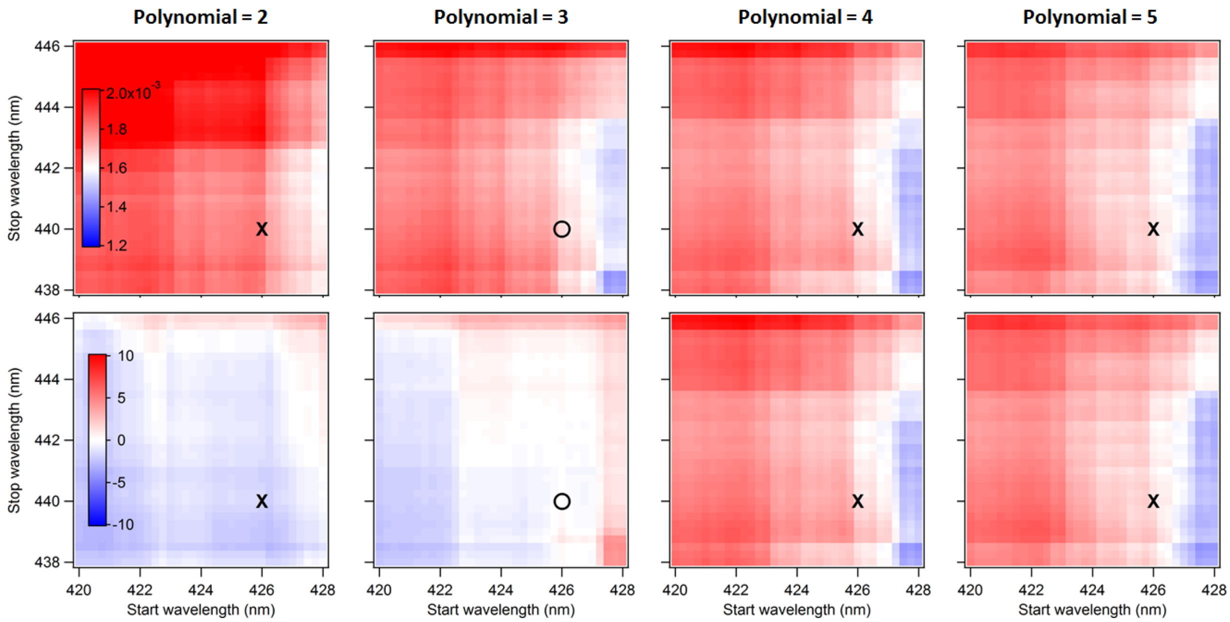

Figure R4. Same as Figure S1 but for SRAA = 140˚ and SZA = 60˚.

Figures R2-R4 have been added in the revised supplemental information.

4. Method limitations need to be better stated: e.g. small AOD (how small?), clear skies (what is the tolerance to clouds), homogeneous aerosol profiles (what is the tolerance to heterogeneity), instrument FOV, instrument stray light, instrument SNR, etc.?

The limitations of the method have been stated along the manuscript and in our dedicated section 3.7 “Context with literature: advantages and limitations”. In the revised manuscript we estimate the response in RSP to changes in AOD based on the change of RSP with respect to AOD. In order to quantitatively show the response through different sets of AOD a linear correlation have been calculated for small subsets of AODs and the results are shown in the table R1 in our response to reviewer #1.

Regarding aerosol inhomogeneity, we have seen that the RSP is quite sensitive to such condition as discussed in the results of Fig. 9B where the retrieval method is not applied (see section 3.6). However, further investigation is needed to study in detail this conditions and the effect of broken clouds using more sophisticated simulations, if possible 3D RTM. However, this is beyond the scope of this manuscript, and mentioned in the revised manuscript.

5. The field campaign lasted for over a month. Could the authors show all successful retrievals and show the linear correlations with other datasets based on all data not just 2 days?

There are several reasons for why be believe this is neither needed, nor a good idea. We have limited this study to two days for the following reasons: (1) We are unaware of a previous attempt to retrieve AOD and g using RSP. Clear and homogeneous aerosol conditions should be the starting point to establish any new method with credibility. (2) Weather conditions during the whole deployment was characterized by overcast conditions (Berg et el., 2015), hence limiting the number of days where the evaluation of the retrieval method is straightforward; this does not rule out that the method could not be applied to other days in the future. (3) The focus of this paper is the demonstration and evaluation of a new method, emphasize the limitations and benefits of the method including periods of large and small aerosol loading, and to validate the method with coincident independent measurements that do not operate under broken sky conditions (especially CIMEL and MFRSR – and also HSRL-2). (4) We are planning to apply the validated approach in future 2D-MAX-DOAS deployments, but there is currently a lack of suitable measurement techniques to evaluate the method under broken cloud conditions.

Minor comments:

Line 93: described by an asymmetry factor g Section 2.1: please describe the atmospheric conditions during TCAP in more detail (e.g cloud cover, aerosol types, vertical profiles). The authors probably have all the information to use Mie theory to calculate phase functions from other in-situ measurements.

In the revised manuscript we added more information about the general conditions during TCAP. We refer the Reviewer to our detailed response to Reviewer #1 regarding Mie calculations (first comment).

Line 127: I suggest moving the sentences “To further: : : (Holben et al., 1998)” after point (3).

We adopted this suggestion.

Line 154: What was the motivation to do almucantar scan at EA 45_. Have you analyzed these data?

We have not looked in detail at the almucantar scan at EA45. The advantages of evaluating azimuth scan at solar EA consist in the enhanced the sensitivity towards aerosol phase functions and minimizing the effect of aerosol inhomogeneity at small SZA. Using the EA of 45˚ would not provide fundamentally different information but could be used to check results and/or gain information of trace gases but this is beyond the scope of the manuscript. The sentence has been modified in the main text as follow:

The advantages of evaluating azimuth scan at solar EA consist in the enhanced the sensitivity towards aerosol phase functions and minimizing the effect of aerosol inhomogeneity at small SZA.

Line 163: Why the authors did not use the integrating sphere to scan the sun in azimuthal and zenith direction to determine the precise position of the sun? Pointing accuracy and precise knowledge of the instrument FOV is important to characterize contribution of external stray light into the system. Please provide a figure in the supplement showing measured FOV of the instrument.

The pointing accuracy and FOV of the 2D telescope have been characterized in detail in Ortega et al. (2015). We refer the reviewer to section 2.1.3 in that paper.

Line 180: Please clarify whether the authors use a single direct sun spectrum for the whole campaign, a single spectrum per day or for each solar almucantar scan its own DS spectrum. I believe it is crucial to have high pointing accuracy to minimize contribution of the scattered photons in the direct sunbeam measurement.

In section 2.2 we mentioned that direct sun spectra were collected for specific cloud free days. In the retrieval of the dRSP we use a single direct sun spectra under low AOD conditions (see section 2.3). The reviewer is correct, high pointing accuracy is important. As explained in section 2.3 the normalized intensities collected in every azimuth scan are used to calculate the pointing accuracy and all results shown are quality assured.

Line 197: Why did the users use Bogumil et al., 2003 NO2 cross section compared to Vandaele et al., 1998?

Thanks for catching this error. This is the wrong citation; we did use Vandaele et al. (1998).

Line 210: Could the authors show one figure with the dRSP error vs dSCD and one with dRSP error vs RMS, and one RMS vs RSAA for SZA 35˚ and 70˚ in the supplemental material?

Figure R5 shows the dRSP error vs dRSP and Figure R6 shows the dRSP error and RMS as a function of SRAA color coded by SZA. Figure 6 consolidates our conservative error reported in the manuscript and the small SRAA dependency. Both Figures have been added to the SI text.

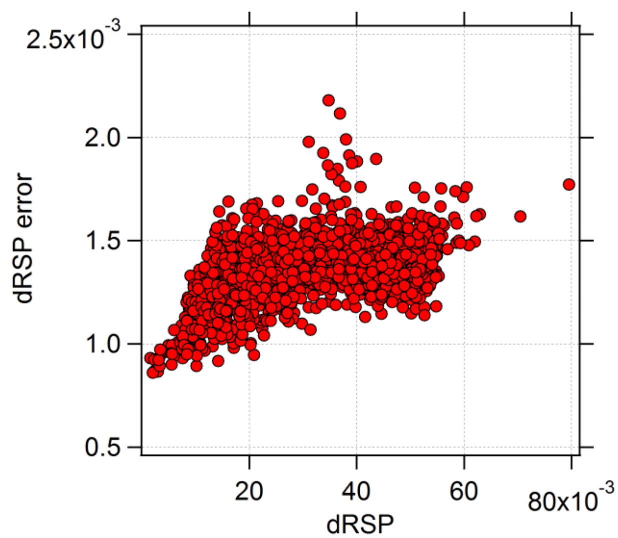

Figure R5. dRSP error vs dRSP on 22 July 2012.

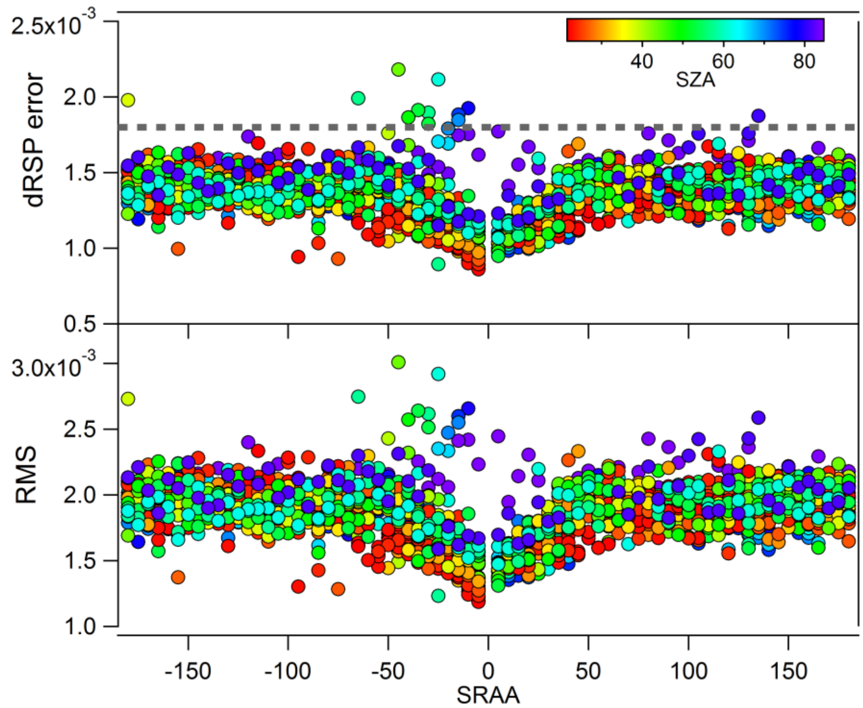

Figure R6. (top) dRSP error and (bottom) RMS vs SRAA on 22 July 2012. The gray horizontal discontinuous line represents the conservative error of 0.0018 reported in the manuscript.

Line 266: could you please specify the dates when these layers where present and the results of the AOD retrieval from the MAX-DOAS instrument? I would think that such layers indicate heterogeneity of the air masses around the observations site and potentially intervene with the retrieval.

Section 3.2 describes in more detail the aerosol inhomogeneity found in both days. The sentence has been modified slightly to point the reader out to section 3.2:

The elevated aerosol layers documented by Berg et al. (2015) during TCAP hence are captured, and do not present a limitation for this work. Section 3.2 describes in more detail the aerosol inhomogeneity on both days.

Section 3.2: Please explore the effect of aerosol inhomogeneity on the retrieval by performing RTM simulations. Section 3.2 describes the angular asymmetry factor but does not show how it impacts the retrieval at different SZA and AOD. The authors adopt AERONET almucantar screening at 20%. But it is not clear whether this is justified for RSP measurements.

The good agreement with methods that have a different field of view, and average over a different airmasses suggests that there is no limitation from aerosol inhomogeneity. We consider a systematic study that deals with aerosol inhomogeneities, their AOD and SZA dependence to be beyond the scope of this paper, which introduces a novel retrieval. We do mention about the use of 3D-RTM to assess inhomogeneous aerosol conditions and broken clouds in section 3.7.

Line 532: Fig 9 shows AOD430 = 0.6 at 14:00 LST.

Line 533: I am not sure I see this. SZA at 14:00 and 11:00 LST are about the same (30˚) while AOD at 14:00 is 0.6 at 11:00 is 0.3-0.4. Despite a smaller AOD (therefore larger dRSP) at 11:00 the retrieval failed. Looking at Fig S6 Asymmetry Factor Parameter is about 10% around 11:00 which might be the reason for retrieval failure.

We agree and meant to discuss it. The Asymmetry Factor Parameter (AFP) plays an additional important role on July 17 where values larger than 10% were identified. Section 3.2 discusses further the AFP. We believe both high AOD and high values of AFP are the reason for the retrieval to fail. The following sentence was updated in the revised manuscript:

On 17 July the AOD430 reached values of 0.6 at noon (Fig. 9B). The high AOD and the inhomogeneity identified with AFP values larger than 10% from 11:00 to 14:00 LST limited the retrieval of AOD and *g* from the 2D-MAX-DOAS.

References

Vandaele, A. C., Hermans, C., Simon, P. C., Carleer, M., Colin, R., Fally, S., M'erienne, M. F., Jenouvrier, A., and Coquart, B.: Measurements of the NO_2 absorption cross section from 42 000 cm^{-1} to 10 000 cm^{-1} (238–1000 nm) at 220K and 294 K., J. Quant. Spectrosc. Ra., 59, 171–184, doi:10.1016/S0022-4073(97)00168-4, 1998