

First of all, we would like to thank the reviewer for this positive assessment of our manuscript, the constructive and helpful suggestions. Point-to-point responses are given below. The original comments are black in color, while our responses are in blue. All the mentioned line number are referred to the revised manuscript.

Major comments

The new method is based on the aerosol optical properties, which is summarized from the measurements. So I suggested the authors can present some evidences of theoretical estimation with the forward RTM to enhanced the principle basis of the method somewhere, even in the supplementary materials.

R: Thanks for your great suggestion. In order to enhance the principle basis of this method described in the manuscript, we used radiative transfer model of SCIATRAN to simulate O₄ DSCDs in UV and Visible bands under conditions with different aerosol optical properties. As listed in Table R1, 11 different aerosol scenarios were simulated in total, in which case 1 is the default case to represent haze condition. Case 1 to 7 describe the aerosol scenarios of gradually increase of scattering properties with a fixed σ_{abs} of 0.050 km⁻¹, which cause the growths in both extinction and SSA. Case 8 to 11 present another process of the gradually increase of haze with more absorbing aerosols under the condition that σ_{sca} are fixed on 0.250 km⁻¹, which consequently result in an increase extinction but decrease of SSA.

Table 1. Simulation-based correlation information between O₄ DSCDs at 360.8 and 477.1 nm under conditions with different aerosol optical properties.

Aerosol information					Slope	R ²	Intercept
No.	σ_{abs}	σ_{sca}	σ_{ext}	SSA			
1	0.050	0.075	0.125	0.6000	0.9560	0.9968	0.5516
2	0.050	0.125	0.175	0.7143	0.9117	0.9859	0.3178
3	0.050	0.250	0.300	0.8333	0.8089	0.9087	0.1438
4	0.050	0.350	0.400	0.8750	0.5672	0.8842	0.3861
5	0.050	0.500	0.550	0.9091	0.5649	0.9800	0.2305
6	0.050	0.700	0.750	0.9333	0.4519	0.9447	0.2603
7	0.050	1.000	1.050	0.9524	0.4875	0.9979	0.1654
8	0.010	0.250	0.260	0.9615	0.6754	0.7963	0.3948
9	0.025	0.250	0.275	0.9091	0.7682	0.9138	0.2353
10	0.075	0.250	0.325	0.7692	0.8051	0.9007	0.1407
11	0.100	0.250	0.350	0.7143	0.8446	0.9063	0.1011

Then, we did the linear-regression analysis for the simulated UV and Visible O₄ DSCDs under different aerosol conditions. As shown in Figure R1, the slope and R² between UV and Visible O₄ DSCDs illustrate that:

- (1) Case 1-7 show an exponential trend in Figure R1. The fitting slope decrease accompanied with the increase of extinction coefficients and SSA if the condition of absorption coefficients are determined.
- (2) Case 8-11 show a linear trend in Figure R1. The fitting slope will decrease

together with the decrease of extinction coefficients and the increase of SSA when the condition of absorption coefficients are determined.

- (3) The correlation coefficients are high (R^2 are mainly greater than 0.90) for all the simulation results. As shown in case 8-11, R^2 decrease accompanied with the decrease of the correlation slopes. This conclusion need to be further supported by more detailed simulations.

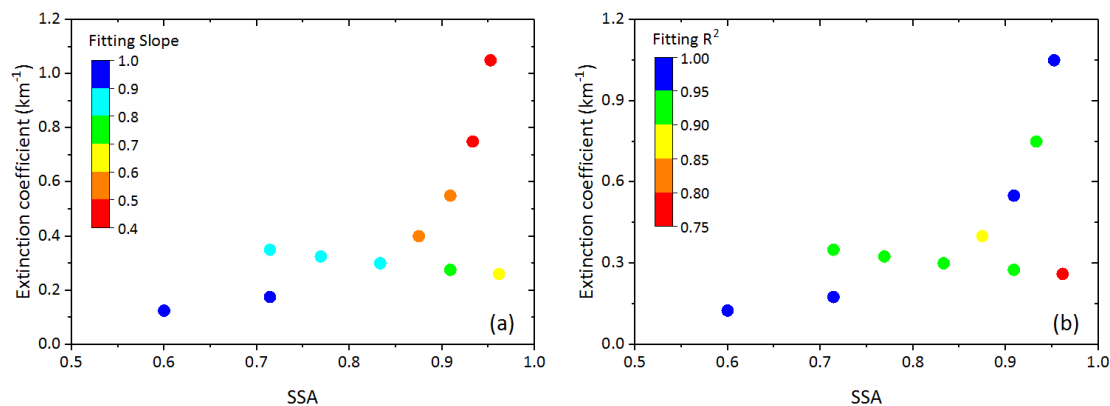


Figure R1. Correlation information (fitting slope and R^2) of the linear regression analysis between the simulated O_4 DCSDs at 360.8 and 477.1 nm under conditions of different aerosol optical properties in the simulation sensitivity studies.

The forward RTM simulation results could demonstrate that the O_4 absorptions (the value of UV and Visible O_4 DSCDs, the corresponding linear-regression slope and R^2 between them) could greatly reflect the variation of aerosol optical properties, which present the theoretical evidences to some extent and enhance the principle basis of the proposed method. Moreover, the simulation results are consistent with the conclusions in the manuscript. The more detailed simulations in the future could provide the better quantitative relationship to the aerosol properties even more.

In addition, we have added this section to Discussion and the Supplement. Please refer to 333-339 in the manuscript.

Minor comments

(1) P4, Sect. 2.2 & P5, Fig. 1 => Please provide the basic information of the measured spectrum in the fitting example, which can be help to evaluate the performance of spectral analysis better.

R: The Fig. 1 in manuscript presents the typical spectral fitting of O_4 DSCDs in UV and Visible bands, and the corresponding measured spectrum were collected at 09:57:29 (SZA (Solar Zenith Angle) = 66.67°, SAA (Solar Azimuth Angle) = 48.40°, ELE (Elevation Angle) = 10°) and 09:42:29 (SZA = 68.27°, SAA = 49.29° and ELE = 5°) on 22 November 2016, respectively. We have supplemented the measured spectra information in the Fig. 1.

Besides, the completed spectral fitting were shown in Fig. R2 here.

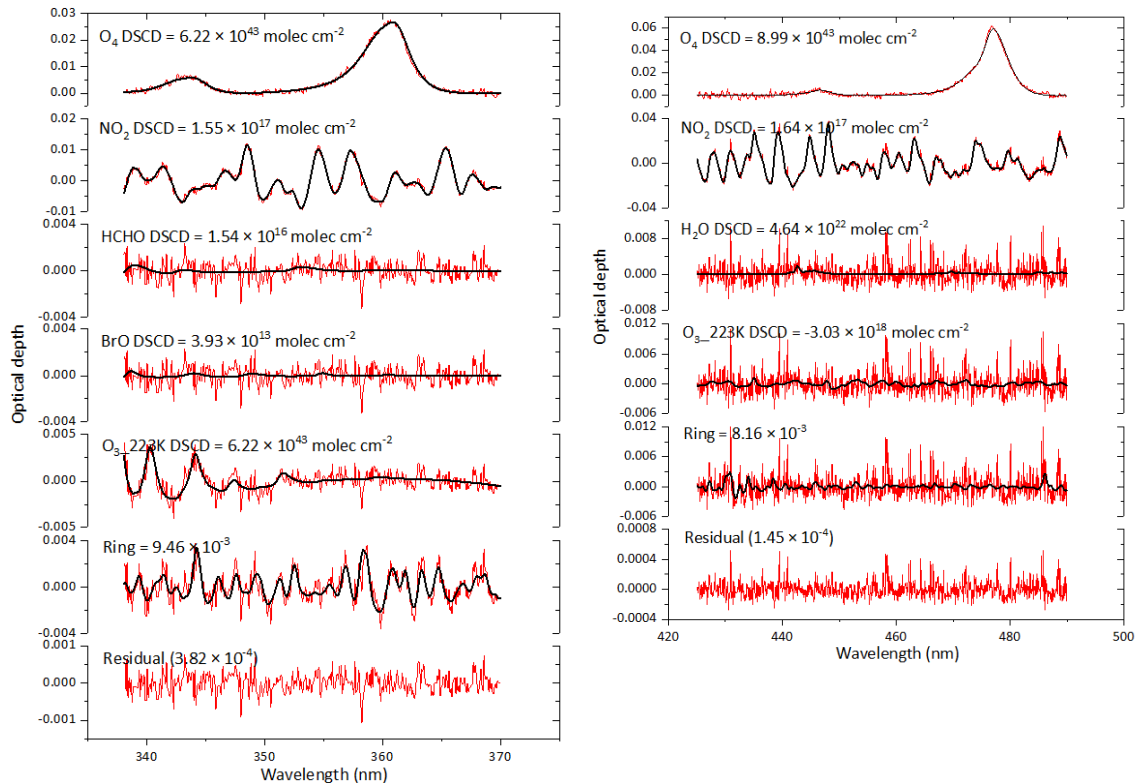


Figure R2. Example of DOAS spectral fitting of O_4 DSCDs in UV (left) and Vis (right) band. Black lines represent the absorption signal and red lines represent the sum of the absorption signal and the fit residual. The example spectrum used to retrieve UV and Visible O_4 DSCDs were obtained at 09:57:29 and 09:42:29 on 22 November 2016, respectively.

(2) P8, L161-165 => Any special consideration for using different RH for clear days, non-haze days ($RH < 80\%$) with haze days ($RH \leq 80\%$)?

R: We apologized for this mistake of typing. In fact, we use $RH < 80\%$ to determine haze and heavy-haze days. In other words, RH all should be $< 80\%$ for clear, non-haze, haze and heavy-haze days. We have corrected this in the revised manuscript.

(3) P10, Fig.3 => Besides the discussion about the correlation coefficient, could the authors give some explanations of the changes in slopes among different weather types? Obviously, the slope in clear and non-haze days are much larger than those in haze and heavy-haze days. Why?

R: The oxygen collision complexes O_4 vertical profiles is well known and nearly constant in the atmosphere, the observed O_4 absorption can serve as an indicator for the atmospheric distribution of photon paths (Wagner et al., 2004; Frieß et al., 2006). The O_4 differential slant column densities (DSCDs) measured by MAX-DOAS are mainly attributed to the photon paths. Since the existence of aerosol can change the light path a lot, the variation of aerosol vertical profiles will be the main factor influencing the photon paths in a cloud-free sunny day, which will be further reflected in the observed O_4 DSCDs.

The path lengths from the effective scattering event to the telescope are dependent on

wavelengths. The path length in visible ranges is obviously longer than that in ultraviolet ranges. In clear and non-haze days, the path length is slightly affected by aerosols. However, the significant increasing of aerosol extinction coefficients in haze and heavy-haze days will have a large effect on the reduction in light path lengths. The reduced light path lengths are thought to result in small O_4 DSCDs. If these light path lengths are sufficiently shortened to penetrate hazy atmosphere, the measured O_4 DSCDs have large uncertainties and may lose sensitivity to vertical distributions of aerosol load (Lee et al., 2011). That could be the reason for the correlation slope in clear and non-haze days are much larger than those in haze and heavy-haze days. Moreover, as shown in Figure R3, the simulation results could also greatly told us the correlation information (fitting slope and R^2) between O_4 DCSDs at 360.8 and 477.1 nm could changes under conditions with different aerosol optical properties. We also added the aerosol information on four different weather conditions in Figure 3 of the manuscript. The measured results and simulation results show good consistency on these four weather conditions of clear periods, light-haze periods, haze periods and heavy-haze periods, especially for heavy-haze periods.

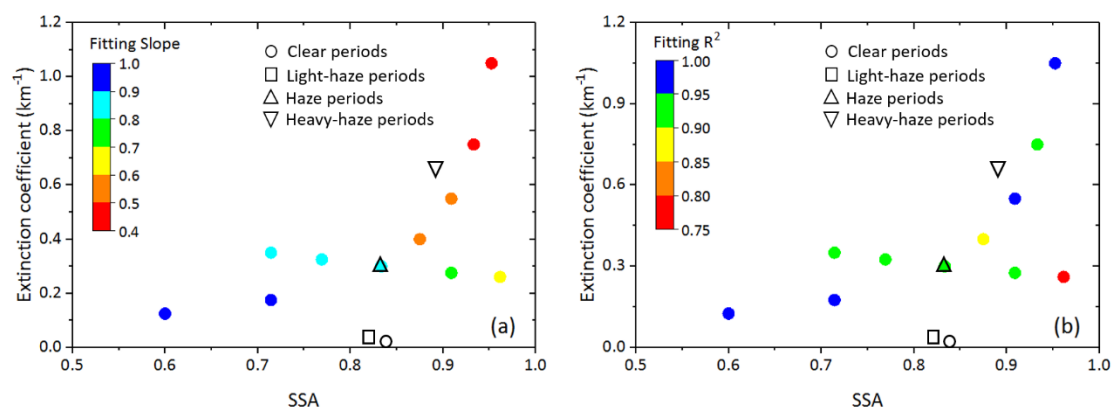


Figure R3. Simulation sensitivity studies of the correlation information (fitting slope and R^2) between O_4 DCSDs at 360.8 and 477.1 nm under conditions with different aerosol optical properties.

(4) P12, Fig.4 => The scat. and abs. changed around 09:05 and 12:00, while the correlation relationship analysis use the break point of 10:00 and 12:00. Why they are different in time? Moreover, why the authors choose the index of variations of scat. instead of abs.?

R: The Figure 4 in the manuscript, we could find the scat. and abs. have lowest values at 09:05 during the time periods of 08:00 to 11:00. However, we have more focused on the change rate (v_{sca} , a_{sca} and a'_{sca} as defined in the manuscript) of scattering and absorption coefficients. The change rate of scattering coefficients (v_{sca} , a_{sca} and a'_{sca}) could be better to help us to understand the relationship the O_4 DSCDs at different wavelength bands and the variations of σ_{sca} and σ_{abs} . For example, the change rate of scattering coefficient at 10:00 is larger than that at 09:05.

We also try to choose the variation of absorption coefficients to identify the break point, but we found it could not identify all the break points as good as the variation

of scattering coefficients. Therefore, we choose the index of variation of scattering coefficients instead of absorption coefficients.

(5) P13-14, Fig.5&6 => The empirical relationships between measured O₄ absorptions in different bands and characteristics of AOPs were mainly concluded from the statistic plot of Fig.5 and Fig.6. I have concerned that the some of the factors (e.g. correlation R² and VIS/UV in haze days, as well as scat. and abs.) have wide value range even cover some cases of other weather conditions. How to obtain the precise and accurate the correspondence between O₄ absorptions and AOPs under different weather conditions?

R: Thanks for your kindly suggestions. As shown in Figure R1 and R3, the fitting slope and R² have different values under conditions with different aerosol optical properties (scattering and absorption coefficient and the corresponding SSA information). Moreover, the corresponding values of O₄ DSCDs in UV and Visible ranges are also different under different conditions. Therefore, it will be a joint decision based on the values of O₄ DSCDs in UV and Visible ranges and the fitting slope and R² between them. This will help us to accurate the correspondence between O₄ absorptions and AOPs.

(6) P16-17, Sect.4 => For the validation, the authors classified the observational period segment into the different weather conditions, however, no further AOPs information, e.g., ADOs, scat. and abs., were inferred and achieved. Is the sentence in line 321(“The σ_{scat} , σ_{abs} and AOD are mainly located at 200-900 Mm⁻¹, 20-60 Mm⁻¹ and 0.9-2.5 under haze and heavy-haze conditions, respectively.”) a conclusion of measurement results or inference from O₄ absorptions?

R: We are very sorry that the description may cause some misunderstanding. The description has been updated as following:

Furthermore, the time series of in-situ σ_{sca} , σ_{abs} and MAX-DOAS retrieved AOD are shown in Fig.7 (c) and (d). According to the empirical relationships summarized above, the σ_{sca} , σ_{abs} and AOD should be mainly located at 200-900 Mm⁻¹, 20-60 Mm⁻¹ and 0.9-2.5 under the haze segment of 09:00-11:00 of 25 November. Simultaneously, the in-situ measured σ_{sca} , σ_{abs} and MAX-DOAS retrieved AOD during the above same periods are ranged in 588.30-730.77 Mm⁻¹, 58.19-67.63 Mm⁻¹ and 1.39-2.22. The inferred results are in good agreement with the measured results. It indicates that the concluded empirical relationships can be used as the criterion to accurately determine the ranges of aerosol optical parameters of σ_{sca} , σ_{abs} and AOD.

We have also supplemented this information in Line 323-329 in the manuscript.

Technical comments

(1) P2, L34: need to be developed

R: Please refer to Line 34.

(2) P2, L83: February of which year?

R: It should be February 2017. Please refer to line 82.

(3) P6, L143: growth => increase

R: Please refer to Line 14-141.

Reference

Frieß, U., Monks, P. S., Remedios, J. J., Rozanov, A., Sinreich, R., Wagner, T., and Platt, U.: MAX-DOAS O₄ measurements: A new technique to derive information on atmospheric aerosols: 2. Modeling studies, *J. Geophys. Res.*, 111, D14203, doi:10.1029/2005JD006618, 2006.

Lee, H., Irie, H., Gu, M., Kim, J., and Hwang, J.: Remote sensing of tropospheric aerosol using UV MAX-DOAS during hazy conditions in winter: Utilization of O₄ Absorption bands at wavelength intervals of 338–368 and 367–393 nm, *Atmospheric Environment*, 45, 5760-5769, 10.1016/j.atmosenv.2011.07.019, 2011.

Wagner, T., Dix, B., von Friedeburg, C., Friess, U., Sanghavi, S., Sinreich, R., and Platt, U.: MAX-DOAS O₄ measurements: A new technique to derive information on atmospheric aerosols—Principles and information content, *J. Geophys. Res.*, 109, D22205, doi:10.1029/2004JD004904, 2004.