

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 criticism

(1)The approach to infer aerosol loading condition from the O₄ absorptions is too far from quantitative, although authors mentioned to develop a lookup table to retrieve aerosol optical properties in the future. Aerosol loading information in the level of conditions defined in this study (clear, non-haze, haze, heavy-haze, ect) can be easily told from human eyes. Therefore, the aerosol information inferred from the method of this study has no scientific value.

R: Thanks for your comments. As shown in Figure R1, the retrieval of aerosol extinction coefficients profile from MAX-DOAS measurements is usually performed based on Optimal Estimation Method (OEM) or look-up table method in the previous research (the green box) (Frieß et al., 2016; Beirel et al., 2019). Consequently, the yielded information is the vertical profile of aerosol extinction coefficient without any further specific scattering or absorbing coefficient. However, the research of this manuscript (the red box) is mainly concentrated on the identification of absorption and scattering coefficients directly from the O₄ absorptions with the empirical look-up table, rather than the aerosol loading information. In this way, the a priori information of aerosol for RTM simulation can be avoided, which contains considerable uncertainties in current OEM and look-up method.

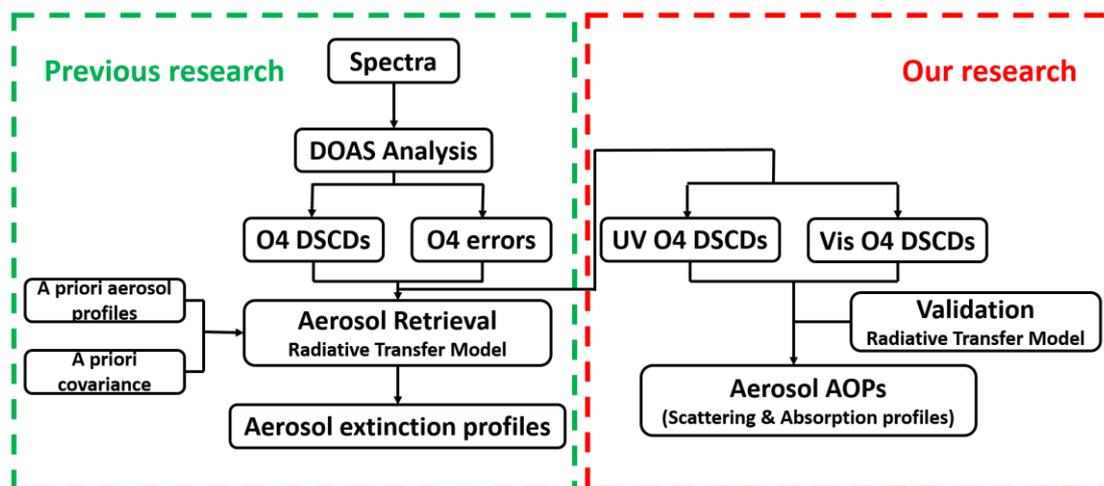


Figure R1. Aerosol profile inversion strategy.

In order to enhance the principle basis of this proposed method, we have used radiative transfer model of SCIATRAN to simulate O₄ DSCDs in UV and Visible bands under conditions with different aerosol optical properties as a validation for the established empirical relationship. As listed in Table 1, 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 O_4 DSCDs at 360.8 and 477.1 nm under conditions with different aerosol optical properties.

	Aerosol information				Slope	R ²	Intercept
	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

The linear-regression analysis for the simulated UV and Visible O_4 DSCDs under different aerosol conditions were also listed in Table 1. As shown in Figure R2, the slope and R² between UV and Visible O_4 DSCDs illustrate that:

- (1) Case 1-7 show an exponential trend in Figure R2. 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 R2. 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 (R² are mainly greater than 0.90) for all the simulation results. As shown in case 8-11, R² decrease accompanied with the decrease of the correlation slopes. This conclusion need to be further supported by more detailed simulations.

We also added the aerosol information on four different weather conditions in Figure R2. 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.

Finally, it can be expected that the vertical spatial-resolved of aerosol scattering and absorption information can be retrieved by using O_4 DSCDs at different elevation angles, although we could only obtain these information at surface using the O_4 DSCDs at elevation angle 1° now. The accuracy of the determination for aerosol optical properties can be improved when the look-up table is replenished and refined in the future.

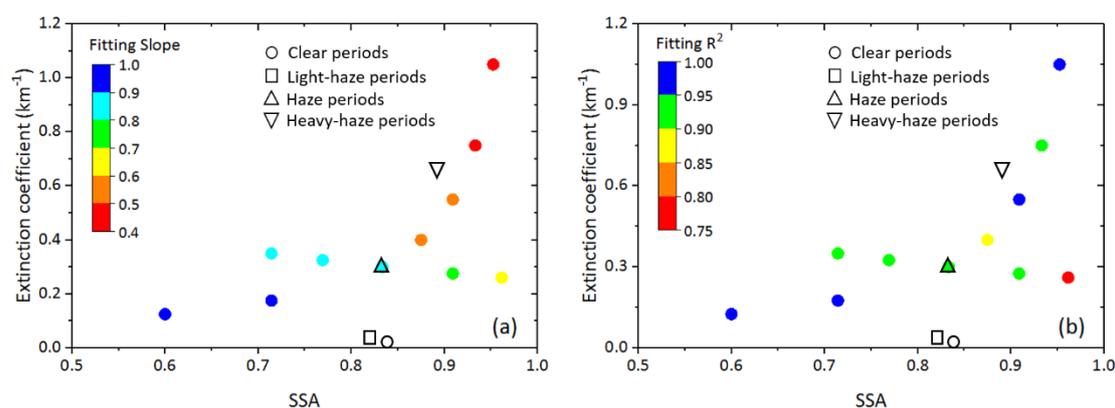


Figure R2. Simulation sensitivity studies of the correlation information (fitting slope and R^2) between O_4 DCSDs at 360.8 and 477.1 nm under conditions with different aerosol optical properties.

(2)Physically, the study methodology is not as much as sound. The O_4 absorption derived from the MAX-DOAS observations are indeed contaminated by aerosols. So in their method, authors intend to use an aerosol-impacted O_4 absorption data to infer the aerosol properties. I think a better way to get information from the MAX-DOAS observation is to retrieve aerosol information (if there is aerosol information) along with the O_4 absorption. Or from another perspective, to study the impact of aerosols to the retrieval accuracy in the O_4 absorption.

R: As shown in Figure R1, the current methodology to retrieve aerosol extinction vertical profile based on MAX-DOAS observed O_4 absorptions is mainly based on the OEM inversion strategy as indicated in the green box of Fig. R1. Considering the uncertainties on the a priori information in the stagey and the single yielded extinction coefficient, our research is aiming to obtain more detailed aerosol optical properties (with absorption and scattering vertical profiles) directly from the O_4 absorptions without the RTM simulation (inversion strategy is described in the red box).

Since the oxygen collision complexes O_4 vertical profile is well known and nearly constant, 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). Therefore, the retrieved O_4 differential slant column densities (DSCDs) at different elevations can provide information about the impact of aerosol scattering on photon paths. By combining measurements of the O_4 absorption with radiative transfer model simulations, ground-based MAX-DOAS has been used in previous studies to determine atmospheric aerosol vertical extinction profiles and optical depths (e.g., Irie et al., 2008, 2009; Li et al., 2010; Clémer et al., 2010; Hartl and Wenig, 2013; Hendrick et al., 2014; Vlemmix et al., 2015; Frieß et al., 2016). Moreover, the UV and Visible O_4 DSCDs are used to retrieve aerosol extinction information independently.

In previous, our group have carried out several researches to retrieve aerosol extinction profiles using O_4 DSCDs, which also show the good agreements with external simultaneous measurements, such as Lidar, tethered-balloon observations

(e.g. Xing, et al., 2017; Tan et al., 2018). However, there are still some uncertainties on the retrieval based on Optical Estimation Method (OEM). So we try to establish the new method using O_4 absorptions in UV and Visible ranges to directly get aerosol scattering and absorption information in the study.

(3) I am not convinced with the change speed, acceleration, and the change rate of the acceleration of diurnal scattering coefficient (equations 2-4) for describing the aerosol property change. These variables only make the trivial diurnal analysis more complicated.

R: In order to illustrate the rationality of using the change speed (v_{sca}), acceleration (a_{sca}) and the change rate of acceleration (a'_{sca}) of diurnal scattering coefficients, we selected two examples (February 21 and 11, 2017) to show the evidences.

The example described in Figure R3 is also discussed in the manuscript. We could find the scattering (σ_{sca}) and absorption coefficients (σ_{abs}) have significant variations based on the calculated v_{sca} , a_{sca} and a'_{sca} at $\sim 10:00$ and $\sim 12:00$, respectively, while the time-indicated O_4 DSCDs seems to be three segments with higher correlation coefficients by the break point of 10:00 and 12:00 (in Figure R3 (a) and (c)).

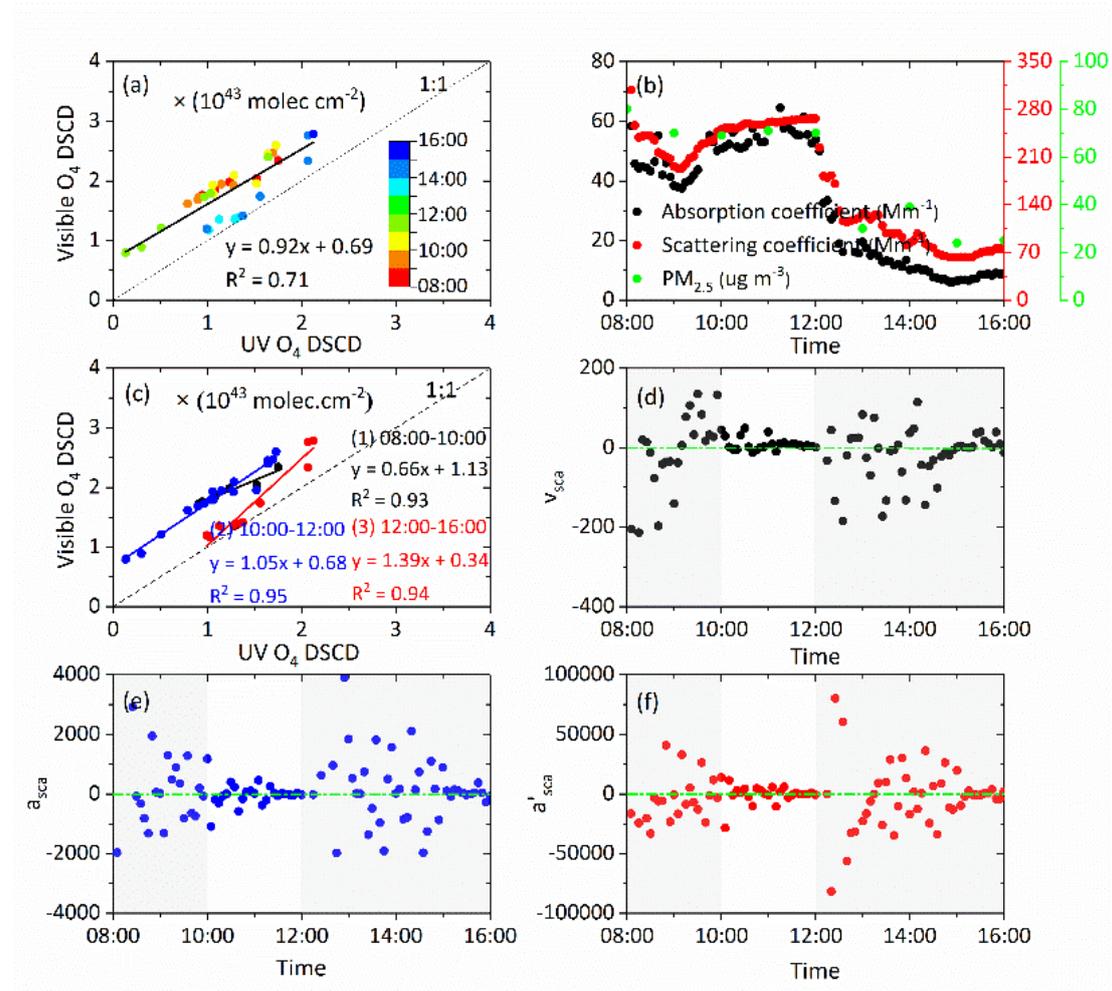


Figure R3. An example day on February 21, 2017: (a) the correlations between O_4 DSCDs

at 360.8 and 477.1 nm. The colorbar represents time sequence. (b) shows the time series of aerosol scattering and absorption coefficients. The correlations information between O_4 DSCDs at 360.8 and 477.1 nm during 08:00-10:00, 10:00-12:00 and 12:00-16:00 were shown in (c). (d) to (f) shows the time series of v_{sca} , a_{sca} and a'_{sca} of scattering coefficients, respectively.

Another case in Figure R4, there are no dramatic changes on σ_{sca} and σ_{abs} during the day. The σ_{sca} decreased slowly in the morning, kept in a stable level during 13:00-15:30, and then increased fast, which presented with three segmental periods (Fig. R4(b)). If we calculated v_{sca} , a_{sca} and a'_{sca} simultaneously, there is only one break point at 15:30 (Fig. R4(d)-(f)). Consequently, the time-indicated O_4 DSCDs of these two segmental periods have higher correlation coefficients. It also indicates aerosol scattering and absorption properties could be mainly manifested in the variations of O_4 absorptions at different wavelength bands.

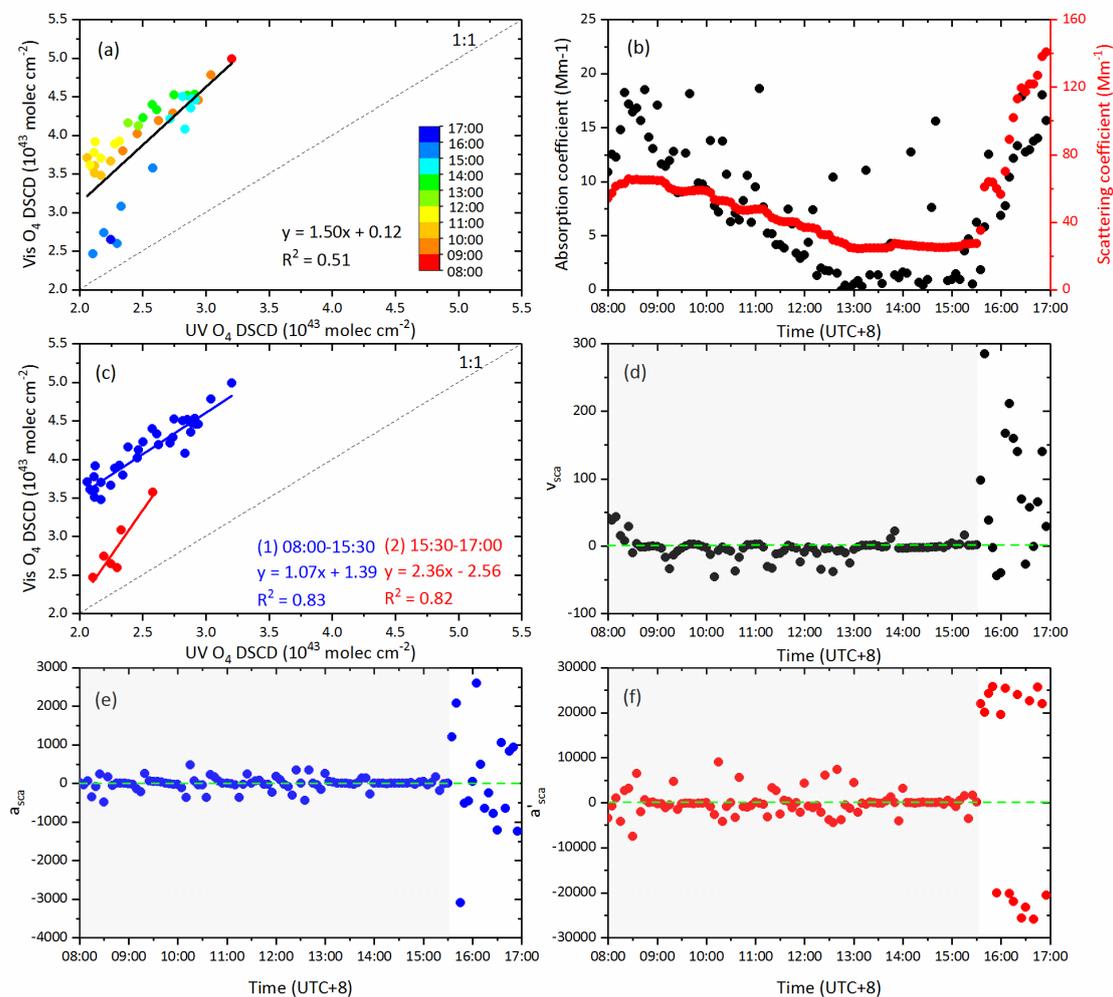


Figure R4. An example day on February 11, 2017: (a) the time series of O_4 DSCDs at 360.8 and 477.1 nm. (b) the correlations between O_4 DSCDs at 360.8 and 477.1 nm. The colorbar represents time sequence. (c) shows the time series of aerosol scattering and absorption coefficients. (d) to (f) shows the time series of v_{sca} , a_{sca} and a'_{sca} of scattering coefficients, respectively.

It can be concluded from Fig. R3 and R4 that the variations on the linear regression analysis between O₄ DSCDs in UV and Visible ranges could better reflect the variations of aerosol scattering and absorption properties. Moreover, the values of O₄ DSCDs are mainly dependent on the light path lengths. It will be difficult to find the break points of scattering coefficients, if we only rely on the values of O₄ DSCDs. Therefore, the above three parameters (change speed, acceleration and the change rate of the acceleration) can help us find the break points of scattering coefficients more accurately, for example in the Fig. R4, the proposed three index can better find the break point than the variation of σ_{sca} itself.

In addition, the calculation of the change speed, acceleration and the change rate of the acceleration are carried out automatically by the coded program, which will not become complicated in practical.

(4)The presentation quality needs improvement. The paper has many grammar issues. I try to catch them in the technical comments below.

R: Thanks for your kindly suggestions. We have improved the presentation quality in the manuscript.

Specific comments

(1)It seems different datasets have different sampling times. It is not clear based on what time length the data are aggregated and compared. Please clarify this.

R: The scattering and absorption coefficient data, the main analytical data, measured at Peking University Urban Atmosphere Environment Monitoring Station (PKUERS, 39.9892°N, 116.3069°E) all have the sampling time of 5 minutes. However, the scattering and absorption data, the data used for validation, measured at Gucheng, Hebei province (39.1382°N, 115.7163°E) have different sampling temporal resolution. The sampling of scattering and absorption coefficient are 1 minute and 1 hour, respectively. We have also clarified in the manuscript. Please refer to line 128 and line 306-307.

(2)Line 160 – 165: the weather conditions are called clear days, ... rainy days. Does it mean that all the data are analysis with on a daily based? If yes, it may be not appropriate, because different weather conditions can happen within a day. If no, these categories should not be called xx days.

R: All the data are analyzed with the hourly averages instead of a daily based. We will change “xx days” to “xx periods”.

(3)I would suggested change “non-haze” into “light-haze”, as “non-haze” means clear. And this condition has an average AOD of 0.35; calling light-haze is more proper.

R: We have corrected them in the whole manuscript according to your suggestion.

(4)Why the elevation angle = 1 degree is chosen for O₄ DSCD used in this study? Please clarify it in the article.

R: Most of scattering and absorption observations were carried out by in-situ instruments at the ground surface. The ground surface aerosol also attracts more attentions due to its closer interactions with ecosystem and human health. Meanwhile, the O₄ absorptions at elevation angle 1° could well reflect the near-surface aerosol information. We only have the in-situ measurements for scattering and absorption coefficients at the ground surface at this moment. Therefore, this study is mainly to focus on the near-surface aerosol optical properties.

Although it is difficult to measure scattering and absorption vertical profiles using in-situ instruments, we are attending to use air ship-based in-situ instruments to measure aerosol scattering and absorption vertical profiles in the near future, which can provide the completed profiles of aerosol scattering, absorption and extinction coefficients. Therefore, we would like further to use O₄ absorptions at multiple different elevation angles to study aerosol scattering and absorption coefficients at different heights. It will also could help us to improve the lookup table.

Technical comments

Line 24: I don't understand the meaning of "O₄ Differential Slant Column Densities (DSCDs) at UV and visible bands varied in the order of ...". Do you mean the magnitude of their correlation coefficients decrease I the order of ...?

R: No, here we mean that the absolute value of O₄ Differential Slant Column Densities (DSCDs) at UV and visible bands all varied "in the order of ...". The DSCDs are usually to denote the absolute value of the results of spectral analysis.

40: aerosols and Aerosol Optical => aerosol loading and Aerosol Optical

40 – 41: Different aerosols behave => Different aerosol types behave

43: heat the air contributing to => heat the air, and contributes to

44: profile causing => profile, causing

R: Please refer to Line 40-44.

48: Please note the AE often refers to Angstrom Exponent.

R: We will use AEC and AE represent Aerosol Extinction Coefficient and Angstrom Exponent in our manuscript, respectively.

49: Angstrom => Angstrom Exponent

51: atmosphere in vertical. => atmosphere.

52: SSA could represent => SSA is defined as

R: Please refer to Line 49-52.

55: the four general aerosol types of biomass burning aerosol, urban-industrial aerosol, dust aerosol and aerosol of marine origin are exhibiting => different aerosol types (such as biomass burning, urban-industrial, dust and sea-salt aerosols) exhibit

R: Please refer to Line 55-56.

63: pronounced SSA => SSA

R: Please refer to Line 61.

71: with maxima at => around

R: Please refer to Line 71.

73: O4 absorptions can be yielded by the DOAS method and further the aerosol vertical profiles at four different wavelength bands (xx). => O4 absorptions in four bands (xx, xx, xx and xx nm) can be estimated, and aerosol vertical profiles can be further derived.

R: Please refer to Line 73-74.

90: Science (CAMS => Science building (CAMS

R: Please refer to Line 89.

95: were used to cover => are used to cover

R: Please refer to Line 94.

93: was equipped with => is equipped with

R: Please refer to Line 96.

106: in UV => in the UV; in visible spectral interval => in the visible

R: Please refer to Line 105.

113: filtered => filtered out; measurements remains for the further discussion. => measurements remained

R: Please refer to Line 111-112.

Table 1: The second line is confusing; No asterisk mark can be found for the table footnote.

R: The asterisk mark at bottom of Table 1 has been removed. We have added the statements about I_0 correction in the text body. Please refer to Line 109-110.

124: several times only during the daytime and only works on non-rainy days => about every 15 minutes during non-rainy daytime.

R: Please refer to 123.

133: Please indicate the distance between Beijing Airport and your site.

R: Please refer to 133.

141: always appeared when PM_{2.5} concentrations increased obviously, and the corresponding AOD also have a significant growth. => coincided with significantly high PM_{2.5} concentration and high AOD.

143: have a significant growth => increases dramatically

R: Please refer to Line 140-141.

143: gray area => gray areas

143: particles pollution => particulate pollution

R: Please refer to Line 142.

150: decreased faster and declined to => decrease sharply to

150: during a shorter while => within a shorter period

R: Please refer to Line 150.

153: are up to => are

154: greater than => over; all the wintertime => the entire wintertime

R: Please refer to Line 153-154.

176: decreased about => decreased by about

R: Please refer to Line 175.

189: at UV => in the UV

R: Please refer to Line 188.

Figure 3: please indicate the time is Beijing Time (UTC+8)

R: We have made the corrections. Please refer to Figure 3 in the revised manuscript.

248: weather types => weather conditions

R: Please refer to Line 250.

255: on the O4 => for the O4

255: at UV band => in the UV band; at visible => in the visible

R: Please refer to Line 257-258.

306: weather type => condition

R: Please refer to Line 310.

325: table. to retrieve => table to retrieve

R: Please refer to Line 332.

330: at UV and visible wavelength bands => in the UV and visible bands

R: Please refer to Line 341.

335: heavy-haze days to. => heavy-haze condition

R: Please refer to Line 346.

341: correlation slope => linear-regression slope

R: Please refer to Line 352.

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