

Author's response by Wenjie Wang et al.

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We greatly appreciate the time and effort that the Referees spent in reviewing our manuscript. The comments are really thoughtful and helpful to improve the quality of our paper. We have addressed each comment below, with the Referee comment in black text, our response in blue text, and relevant manuscript changes noted in red text.

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### **Anonymous Referee #1**

Line 30: "In addition, the slopes are equal to ...." 1) The wording: "In addition, ..." is awkward. 2) Why there is a range of slopes for  $j(\text{O}^1\text{D})$  and a single value for  $j(\text{NO}_2)$  is unclear at this point, and what the slopes refer to in the first place when the relationships are non-linear. 3) The slopes should be negative in any case. 4) The authors should take into account significant digits (throughout the paper). The precision of the data does not justify a statement "4.21-6.93". I would say "4.2-6.9" at the very most. 5) AOD has to be specified here, i.e. AOD (380 nm)?

Response: 1) I have removed the phrase "In addition".

2) There is a range of slopes for  $j(\text{O}^1\text{D})$  is because that the slopes varied at different total ozone column. Table 3 give different slopes for  $j(\text{O}^1\text{D})$  at different ozone column classes. For  $j(\text{NO}_2)$ , total ozone column has a negligible influence on the slope. The slopes are at AOD smaller than 0.7, where the relationship between  $j$ -value and AOD is close to linear.

3) Yes, the slopes should be negative in any case. Therefore, in Line 30, I changed "the slopes" into "the absolute values of slopes".

4) Many thanks and I have taken into account significant digits throughout the paper.

5) I have added AOD (380 nm) in line27.

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Line 32: "... larger than those observed in the Mediterranean." I would say: "... than those observed in a similar, previous study in the Mediterranean."

Response: Thank you and I have revised it in the manuscript.

Line 30: The absolute values of slopes are equal to  $4.2\text{--}6.9 \cdot 10^{-6} \text{ s}^{-1}$  and  $3.2 \cdot 10^{-3} \text{ s}^{-1}$  per AOD unit for  $j(\text{O}^1\text{D})$  and  $j(\text{NO}_2)$  respectively at SZA of  $60^\circ$  and AOD smaller than 0.7, both of which are larger than those observed in a similar, previous study in the Mediterranean.

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Line 33: "...have a stronger extinction on ..." Please reword.

Response: I have changed into it "...have a stronger extinction effect on ...".

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Line 38, 39: "...  $j(\text{NO}_2)$  by 24.2% and 30.4% for summer and winter, ...  $j(\text{O}^1\text{D})$  by 27.3% and 32.6%..." 1) The meaning of these numbers is unclear. I assume they refer to some kind of seasonal mean of the photolysis frequencies that needs to be specified. 2) The precision implied by three digits is misleading.

Response: Thank you and I have revised it.

Line 39-42: According to the parametric equation, aerosols lead to a decrease in seasonal mean  $j(\text{NO}_2)$  by 24% and 30% for summer and winter, respectively, and the corresponding decrease in seasonal mean  $j(\text{O}^1\text{D})$  by 27% and 33% respectively, compared to an aerosol-free atmosphere.

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Line 42: "... the monthly average net ozone production is reduced by 25%." By looking at Fig. 10, I assume the 25% refers to a monthly mean daytime net ozone production that needs to be specified.

Response: Yes, the 25% refers to a monthly mean daytime net ozone production. I have specified it.

Line 44: The simulation results shows that the monthly mean daytime net ozone

production rate is reduced by up to 25% due to the light extinction of aerosols.

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Lines 54, 59, 63, 65, 66: Use consistent notations for O(3P) and O(1D).

Response: Thank you and I have revised it.

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Line 60: “....the only significant chemical source....”

Response: Thank you and I have revised it.

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Lines 68-71: Remove the symbol “S” in the brackets. It may be added as an index to “sigma” and “phi” but “S” is no variable like “lambda” or “T”.

Response: Thank you and I have revised it.

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Lines 78-79: “Scattering aerosols can enhance..., while absorptive aerosols reduce ...throughout the boundary layer.” These statements are unclear and certainly do not apply for all conditions.

Response: I agree with you that these statements are unclear and certainly do not apply for all conditions. Therefore, I add “Some previous studies showed that” in front of this sentence. Both the abstract of Dickerson et al. (1997) and the introduction of Flynn et al. (2010) have proposed this viewpoint.

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Line 91: “Therefore it is necessary to quantitatively evaluate the effect of aerosols on photolysis frequencies for the purpose of ozone prevention”. I would say: “... for a better understanding of ozone formation under highly polluted conditions.”

Response: Thank you and I have revised it.

Line 98: Therefore, it is necessary to quantitatively evaluate the effect of aerosols on photolysis frequencies for a better understanding of ozone formation under highly polluted conditions.

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Line 98: "... are compared with the observed value to test the simulation effect." Unclear: If radiative transfer models are used there are usually no measurements available. And what is the "simulation effect"?

Response: The expressed meaning is unclear and thus I simplified this sentence.

Line 105: The observed data of related influential factors of the photolysis frequencies are taken as the model's input to calculate the photolysis frequencies.

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Line 100: "... due to complicated environmental conditions...". Clarify.

Response: The complicated environmental conditions will influence the simulation of photolysis frequencies by radiative transfer model. Although AOD is easily acquired from ARONET and satellite, SSA, asymmetry factor, AE and the vertical profile of aerosols are not always available, and their observed values often had a large uncertainty. The complicated environmental conditions including relative humidity, temperature, planetary boundary layer height and emission characteristics all influence aerosol optical properties and thus influence light extinction of aerosols. Previous studies indicated that the aging of black carbon, mixing state of aerosols, the absorptive capacity of organic aerosols and the hygroscopicity of aerosols, all of which are determined by specific environmental condition, significantly contribute to the uncertainty of aerosol optical properties.

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Line 123-136: Use the term "photolysis frequencies" consistently throughout the text.

Response: Thank you and I have revised it.

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Lines 134, 135: Mind significant digits, see abstract by Li et al., 2011.

Response: Thank you and I have revised it.

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Line 143ff: “Our overall goal...” It should be made clear that this study was strongly informed by a similar work by Gerasopoulos et al., 2012 which is not adequately referred to in the Introduction.

Response: Thank you and I have added this part.

Line 153: The relationship between AOD and photolysis frequencies is adequately compared with previous study in the Mediterranean (Casasanta et al., 2011; Gerasopoulos et al., 2012).

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Line 155: The exact measurement period should be given here. Was it exactly four years?

Response: I have described the exact measurement period.

Line 161: From August 2012 to December 2015,  $j(\text{O}^1\text{D})$  and  $j(\text{NO}_2)$  were measured continuously at PKUERS site. The data of August 2015 are missed due to instrument maintenance.

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Line 163: What absorption cross sections and quantum yields were used to calculate the photolysis frequencies?

Response: The quantum yields of  $J(\text{O}^1\text{D})$  was taken from Matsumi et al.(2002), while the ozone cross section was derived from Daumont et al. (1992) and Malicet et al. (1995). For  $j(\text{NO}_2)$ , the quantum yields used was taken from Bass et al. (1976) and Davenport et al. (1978), while the cross section was derived from Jones and Bayes

(1973), Harker et al. (1977) and Davenport (1978). I have add these sentences in the Manuscript.

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Line 163: I assume the  $j(\text{O}^1\text{D})$  were calculated temperature dependent according to Eq. 1 (and a statement in lines 302-304). That should be clearly stated. However, is it useful, if a common parameterization as a function of AOD is later used for summer and winter  $j(\text{O}^1\text{D})$ ? There may be arguments to include temperature but the influence should be mentioned and quantified in Section 3 (see below).

Response: The  $j(\text{O}^1\text{D})$  were calculated temperature dependent. I added the sentence “Measured temperature are used to retrieve ozone absorption cross section and photodissociation quantum yield.” in line 188. I have evaluate the impact of temperature on  $j(\text{O}^1\text{D})$  by calculating the ratio of  $j(\text{O}^1\text{D})$  to  $j(\text{O}^1\text{D})$  at temperature=298K (Figure S2). The result indicates that temperature changed  $j(\text{O}^1\text{D})$  by no more than 21%. In addition, the determination coefficients of fitted parametric equations are larger than 0.95, indicating the influence of temperature is relatively small.

Line 447-457: For  $j(\text{O}^1\text{D})$ , both of  $\text{O}_3$  column and temperature affect  $j(\text{O}^1\text{D})$  significantly. Figure S1 presents the dependence of  $j(\text{O}^1\text{D})$  on ozone column at low AOD level ( $\text{AOD} < 0.3$ ) and SZA of (a)  $30^\circ \pm 1^\circ$  and (b)  $60^\circ \pm 1^\circ$ , respectively. Ozone column ranging from 270 to 400 DU leads to  $j(\text{O}^1\text{D})$  reducing about 50%. In order to evaluate the impact of temperature on  $j(\text{O}^1\text{D})$ , we calculated the ratio of  $j(\text{O}^1\text{D})$  at measured temperature to  $j(\text{O}^1\text{D})$  at temperature = 298K ( $j(\text{O}^1\text{D})/j(\text{O}^1\text{D})_{\text{T}=298\text{K}}$ ) (Figure S2).  $j(\text{O}^1\text{D})/j(\text{O}^1\text{D})_{\text{T}=298\text{K}}$  varied from 0.82 to 1.03 indicating that temperature changed  $j(\text{O}^1\text{D})$  by no more than 21%. Therefore, temperature played a minor role in changing  $j(\text{O}^1\text{D})$  compared with ozone column. As a result, when we fitted the relationship among  $j(\text{O}^1\text{D})$ , AOD and  $\cos(\text{SZA})$ , the effect of ozone column is considered but temperature is not considered.

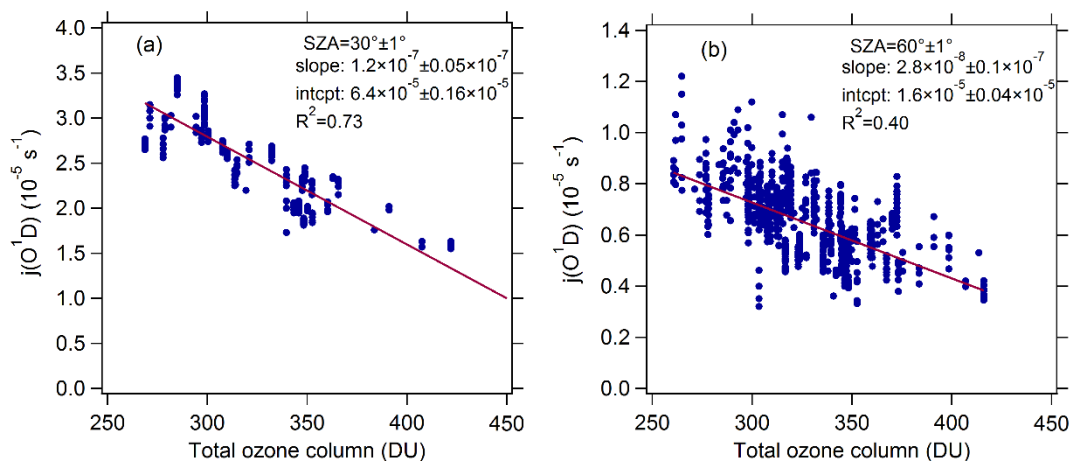


Figure S1. Dependence of  $j(\text{O}^1\text{D})$  on AOD (380nm) at low AOD level ( $\text{AOD} < 0.3$ ) and SZA of (a)  $30^\circ \pm 1^\circ$  and (b)  $60^\circ \pm 1^\circ$ , respectively.

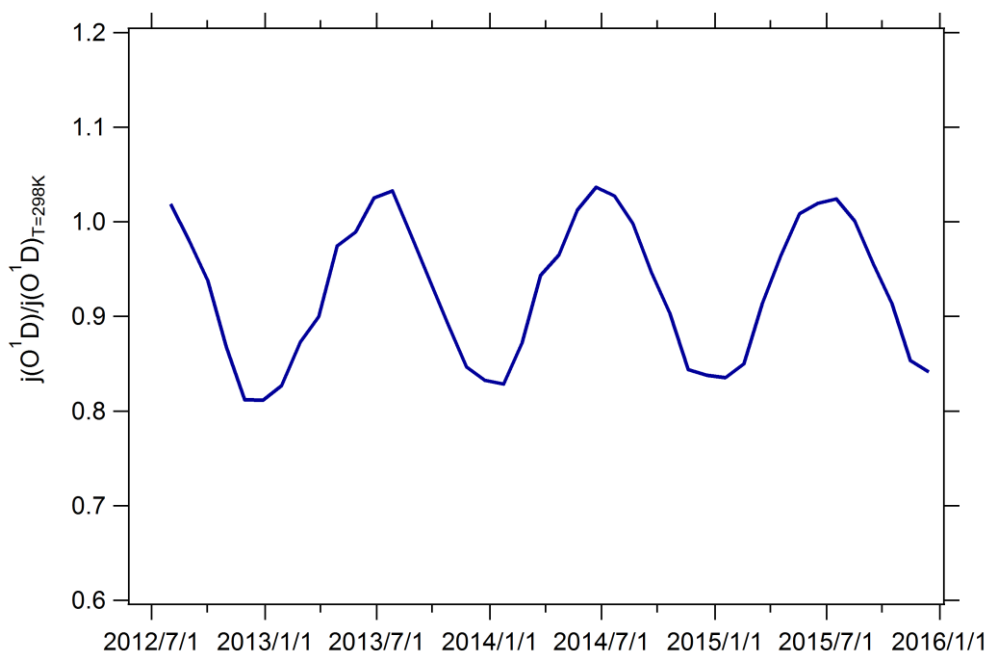


Figure S2. The time series of the monthly mean ratio of  $j(\text{O}^1\text{D})$  to  $j(\text{O}^1\text{D})_{T=298\text{K}}$  ( $j(\text{O}^1\text{D})/j(\text{O}^1\text{D})_{T=298\text{K}}$ ) from August 2012 to December 2016.

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Line 164: “Shetter and Müller, 1999”

Response: Thank you and I have revised it.

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Line 169: The studies by Shetter and Müller, 1999, and Hofzumahaus et al., 1999 describe double-monochromator based instruments with somewhat different properties. The authors should state what type of detector was used and how regular calibrations of the instrument were performed during the four-year period. Moreover, it is unclear if the 10% uncertainty comes from the calibration uncertainty or is attributed to the uncertainties of absorption cross sections and quantum yields.

Response: (1) The double-monochromators for wavelength separation and successive measurements with single detectors (e.g. photomultipliers) upon scanning the wavelength. This is excellent for stray light suppression which is important in the UV-B range (e.g. Shetter and Müller, 1999; Hofzumahaus et al., 1999). Drawbacks are the comparatively long time periods to complete the wavelength scans ( $\geq 30$ s) and the use of motor-driven optical components which may cause stability problems under field measurement conditions. Our method uses single monochromators and detector arrays (e.g. photodiode arrays) for simultaneous measurements covering the whole range of relevant wavelengths. This method has the advantage of high time-resolution and stability because no movable parts are involved. (2) The detector is a  $2048 \times 64$  pixels photodiode array detector. (3) The 10% uncertainty is associated with the quartz receiver and stray-light effects.

Line 177: The actinic flux was measured using a spectroradiometer and the photolysis frequencies were calculated from the absorption cross section and quantum yield of each species (Shetter and Müller, 1999). The spectroradiometer consisted of a single monochromator with a fixed grating (CARL ZEISS), an entrance optic with a  $2\pi$  steradian (sr) solid angle quartz diffusor and a  $2048 \times 64$ -pixel photodiode array detector. The spectral measurements were performed with a wavelength resolution of 2 nm, covering a wavelength range of 290-650 nm (Hofzumahaus et al., 1999). The measured spectra were corrected for dark signal and stray light. A 1000 W NIST traceable irradiance standard (Oriel) was used for calibration under laboratory conditions (Bohn et al., 2008). For  $j(\text{O}^1\text{D})$ , the quantum yield was taken from Matsumi et al. (2002), while the ozone cross section was derived from Daumont et al. (1992) and Malicet et al.



(1995). Measured temperature was used to retrieve ozone absorption cross section and quantum yield. For  $j(\text{NO}_2)$ , the quantum yield was taken from Bass et al. (1976) and Davenport et al. (1978), while the cross section was derived from Jones and Bayes (1973), Harker et al. (1977) and Davenport (1978). The calculated photolysis frequencies had a time resolution of 10 s and an accuracy of  $\pm 10\%$  including uncertainties associated with the quartz receiver and stray-light effects.

**Reference:**

Matsumi, Y., Comes, F.J., Hancock, G., Hofzumahaus, A., Hynes, A.J., Kawasaki, M., Ravishankara, A.R., Quantum yields for production of  $\text{O}(^1\text{D})$  in the ultraviolet photolysis of ozone: recommendation based on evaluation of laboratory data. Journal of Geophysical Research 107 (D3), 4024. doi:10.1029/2001JD000510. 2002.

Daumont, D., Brion, J., Charbonnier, J., Malicet, J. Ozone UV spectroscopy I: absorption cross-sections at room temperature. Journal of Atmospheric Chemistry 15 (2), 145-155, 1992.

Malicet, J., Daumont, D., Charbonnier, J., Parisse, C., Chakir, A., Brion, J. Ozone UV spectroscopy. II. Absorption cross-sections and temperature dependence. Journal of Atmospheric Chemistry 21 (3), 263-273, 1995.

Bass, A. M., Ledford, A. E., and Laufer, A. H., Extinction coefficients of  $\text{NO}_2$  and  $\text{N}_2\text{O}_4$ , J. Res. Nat. Bureau Standards, 80A, 143-162, 1976.

Davenport, J.E. Determination of  $\text{NO}_2$  photolysis parameters for stratospheric modelling, FAA Report No. FAA-EQ-7-14, 1978.

Jones, I. T. N. and Bayes, K. D., Photolysis of nitrogen dioxide, J. Chem. Phys. 59, 4836-4844, 1973.

Harker, A. B., Ho, W., and Ratto, J. J. Photodissociation quantum yields of  $\text{NO}_2$  in the region 375 to 420 nm, Chem Phys. Lett. 50, 394-397, 1977.

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Line 175: "... close to the PKUERS site" should be specified in km.

Response: Thank you and I have specified the distance.

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Line 184: “This wavelength (380 nm) was chosen as it is more representative of  $j(\text{NO}_2)$ ”  
Why wasn’t the AOD at 340 nm used as well to estimate AODs more representative for  $j(\text{O}_1\text{D})$  (around 300 nm), e.g. by the Angstrom equation? You can argue with better comparability with Gerasopoulos et al. , 2012 but that should be made clear.

Response: Thank you and I have added the sentence “Additionally, at this wavelength we can better compare with the results of Gerasopoulos et al. (2012).”

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Line 185: SSA measurements during a period of one month are hardly representative for four years. Since the AOD-SSA relationship becomes important later to explain the steep decrease of  $j$ -values with AOD, I wonder why AERONET based SSA are not consulted for the whole period.

Response: We didn’t use AERONET based SSA because that: (1) AERONET based SSA have a large uncertainty than that by ground based instruments; (2) There are only 10-20 data of SSA in a month for most months, which is much fewer than AOD data. The following figure is the relationship between AOD and AERONET based SSA during 2012-2015. There is a slight positive correlation between AOD and AERONET based SSA.

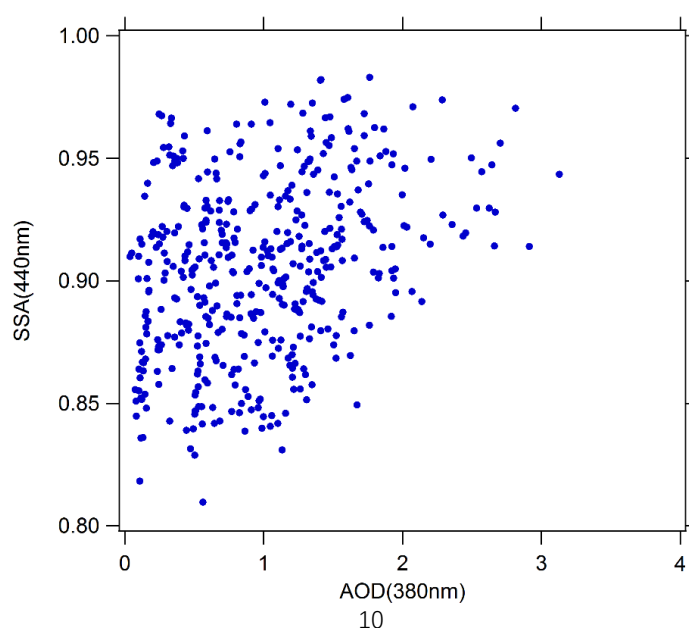


Figure 1. The relationship between AOD and AERONET based SSA.

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Line 191: The source of the ozone column data should be specified and a citation included.

Response: Thank you and I have added the website of the ozone column data and related citation.

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Line 198: "... under cloudless conditions." Was there an additional cloud screening performed or was any period marked cloud-free by AERONET taken? Because of the distance between the sites there were certainly some cases when clouds were present at PKUERS and no clouds at the AERONET site? Moreover, to assess the importance of this work, it would be interesting to learn what fractions of daytimes were identified as clear-sky during the four years. This could be included in Table 2 for the different seasons.

Response: (1) The cloudless conditions are identified according to the presence of AOD data in AERONET since AOD data is unavailable under cloudy conditions. However, we didn't have additional cloud screening procedure. (2) I agree with you that the distance between the sites will cause some cases when clouds were present at PKUERS and no clouds at the AERONET site, which may disturb our analysis of the relationship between AOD and photolysis frequencies. Due to the close distance between the two sites (6.4km), this influence maybe relatively small. (3) Many thanks. I added daytime clear-sky fraction for the different seasons in Table 2.

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Line 203: "Global irradiance" is a different quantity than actinic flux.

Response: Thank you and I have changed "Global irradiance" into "actinic flux spectra".

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Line 204: Explain “AE”. Were the AE taken from AERONET, was a constant AE used, or was AE set to zero to simulate with a wavelength-independent AOD? This is important later for the model measurement comparisons in Fig. 5.

Response: AE (380/550nm) are taken from AERONET and the mean value of 1.3 during August 2012 - December 2015 is used in TUV model.

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Line 205: Were mean Earth-Sun distances used in the calculations or were time, date and location specified? If not, were the measured j-values scaled to a common mean Earth-Sun distance?

Response: Mean Earth-Sun distances was used in the calculations of TUV model and measured j-values were scaled to the mean Earth-Sun distance.

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Line 205: Were the same absorption cross sections and quantum yields used to calculate j(O<sup>1</sup>D) and j(NO<sub>2</sub>) from TUV-derived spectra? What temperatures were used?

Response: Measured temperatures were used to calculate the absorption cross sections and quantum yields of j(O<sup>1</sup>D). The absorption cross sections and quantum yields of j(NO<sub>2</sub>) at 298K were used since they are influenced negligibly by temperature.

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Line 230-234: Equations E2, E3 are not self-explaining. At least give a citation where these formulas are rationalized and explain “θ”.

Response: Many thanks. I have gave a citation and explained “θ”.

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Lines 239-278: “In order to evaluate the extinction capacity...” The motivation to look into the relationship between PM<sub>2.5</sub> and AOD should be made clearer and the results shown in Fig. 1 and Fig. 2 should be reassessed. Obviously, PM<sub>2.5</sub> is not a good proxy to estimate AOD. Moreover, the summer-winter differences in the slopes in Fig.

1 are probably explainable by the different heights of the boundary layers alone and there is no basis to speculate about seasonal differences of aerosol optical properties unless you consult AERONET data. My conclusion of Sect. 3.1 and the first paragraph of Sect. 3.2 would be that PM<sub>2.5</sub> is not suitable to estimate AOD (and consequently, PM<sub>2.5</sub> data are not used in the remainder of the text). On the other hand, did you check the relationship between PM<sub>2.5</sub> and e.g. j(NO<sub>2</sub>) directly? I assume it looks much poorer than the relationship between j(NO<sub>2</sub>) and AOD which would confirm the assumption that AOD is a more relevant parameter.

Response: (1) I have revised the motivation to look into the relationship between PM<sub>2.5</sub> and AOD. Compared with AOD, PM<sub>2.5</sub> is a more common proxy to evaluate the level of particulate matter pollution in spite that AOD is a more closely related parameter of photolysis frequencies. As a result, we attempted to analyze the quantitative relationship between PM<sub>2.5</sub> and AOD to evaluate the influence of PM<sub>2.5</sub> on AOD and thus on photolysis frequencies. (2) I agree with you that PM<sub>2.5</sub> is not a good proxy to estimate AOD due to multiple interference factors including relative humidity, planetary boundary layer height, aerosol type, aerosol size distribution, aerosol distribution in the vertical direction. Zheng et al. (2017) studied the influential factors for the relationship between PM<sub>2.5</sub> and AOD in Beijing during 2011-2015. He found that in addition to RH and PBLH, aerosol components (scattering or absorptive) and size (coarse mode or fine mode) also influenced the slope of PM<sub>2.5</sub> vs AOD significantly according to SSA, AE and FMF data of AERONET. Since Zheng et al. (2017) have studied this question in detail in the same region and the same period, I think that our work needn't analyze SSA, AE and FMF data of AERONET again. Instead, we just compared the slope of PM<sub>2.5</sub> vs AOD of our study with other study in Beijing and other cities of North China. I agree with you that the conclusion is that PM<sub>2.5</sub> is not suitable to estimate AOD due to the large uncertainty. (3) I have checked the relationship between PM<sub>2.5</sub> and j(NO<sub>2</sub>) directly, it is true that there is much poorer correlation than the relationship between j(NO<sub>2</sub>) and AOD.

Line 273-279: Compared with AOD, PM<sub>2.5</sub> is a more common proxy to evaluate the

level of particulate matter pollution in spite that AOD is a more closely related parameter of photolysis frequencies. As a result, we attempted to analyze the quantitative relationship between  $PM_{2.5}$  and AOD to evaluate the influence of  $PM_{2.5}$  on AOD and thus on photolysis frequencies.

Line 307-308: Consequently, using  $PM_{2.5}$  to estimate AOD has a large uncertainty due to multiple interference factors.

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Line 249, Table 2: Table 2 should be mentioned in Section 2.1, not here. Please consider significant digits in Tab. 2 and specify season periods in the caption.

Response: I have revised it.

Line 215-218: In addition, meteorological parameters such as temperature, relative humidity, and pressure were simultaneously observed at the site. Table 1 presents  $O_3$  column concentration, temperature, relative humidity, daytime clear-sky fraction and respective standard deviation for different seasons.

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Figs. 2-3: Specify what is shown here. Averages, medians? The periods defined as “spring”, “summer”, “autumn” and “winter” should be defined clearly somewhere. Are the  $PM_{2.5}$  data in Fig. 2b also from clear-sky days only? Specify “AOD (380 nm)” in the caption of Fig. 2.

Response: (1) Figs. 2-3 show mean value. I have specified it. (2) Thank you, I have define “spring”, “summer”, “autumn” and “winter” in Line 285-287. (3) The previous  $PM_{2.5}$  data in figure 2b are from all-sky days. I have revised Figure 2b using  $PM_{2.5}$  data from clear-sky days.

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Lines 285-292: 1) What do the stated differences in photolysis frequencies refer to? Mean daily maxima? Please specify. 2) What are the uncertainties of these differences? 3) Does the TUV-derived difference refer to aerosol-free conditions? 4) What role plays

the temperature, certainly lower in Beijing during the winter compared to conditions in Crete?

Response: 1) The stated differences in photolysis frequencies refer to mean daily maxima and I have specified it in the manuscript. 2) I have added the uncertainties of these differences. 3) Yes, the TUV-derived difference refers to aerosol-free conditions and I have specified it in the manuscript. 4) I have added the analysis of the influence of temperature during winter.

Line 334-339: Additionally, we know that the temperature is lower in Beijing during the winter compared to conditions in Crete. The measured mean temperature in Beijing during winter is equal to  $0.53 \pm 4.2$  °C (Table 1). When we consider the temperature in Crete is 10 °C higher than in Beijing, the lower  $j(\text{O}^1\text{D})$  of Beijing than Crete is  $5.5 \times 10^{-7} \text{ s}^{-1}$ , which is also not able to compensate the  $j(\text{O}^1\text{D})$  gap between the two regions during winter.

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Fig. 4: Specify in the caption what the full lines show. Averages, medians? What AOD bin size was used? Indicate AOD (380 nm).

Response: The full lines are fitted by exponential function. The coefficients of determination ( $r^2$ ) vary from 0.5 to 0.7. I have specified it in fig. 4.

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Fig. 5: Add standard deviations to the measured values. Otherwise the relevance of the differences compared to the model calculations cannot be assessed. Specify the ozone column range of the measured data in the caption. Indicate AOD (380 nm) for the measured data and  $\text{AOD} \neq f(\lambda)$  for the model calculations (if that applies).

Response: (1) I have add standard deviations to the measured values. (2) The ozone column range for  $j(\text{O}^1\text{D})$  is 330-360 DU. I have specified it.

Line 319-326: Here the question again arises, what AE was used in the TUV calculations, what temperatures and if the annual changes in Sun-Earth distances were considered.

Response: We used the mean AE=1.3, temperature = 298K in TUV. In addition, we used the mean Sun-Earth distance in TUV.

Line 377-381: The observed  $j(O^1D)$  is at temperature of 288-308K and ozone column of 330-360DU. AE(380/550nm) = 1.3, ozone column = 345 and Temperature = 298K are used in TUV model for all simulations. Mean Earth-Sun distance was used in the calculations of TUV model and measured j-values were scaled to the mean Earth-Sun distance.

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Lines 327-341 and lines 341-347: These sections are too speculative without consulting AERONET data. As already mentioned, the 1-month data in Fig. 6 is probably not representative for the average aerosol over the four year measurement period.

Response: As mentioned above, there is a slight positive correlation between AOD and AERONET based SSA during 2012-2015. As the AERONET based SSA data have a large uncertainty, we are not sure if the result should be added into the manuscript.

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Lines 357-361 and Table 3: Consider significant digits.

Response: I have revised it.

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Figure 7: What do the full lines show?

Response: The full lines are fitted by exponential function.

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Line 397: Equation E5 should appear here.



Response: I have revised it.

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Lines 397-406, Tables 5 and 6: 1) If ozone columns have no significant influence on  $j(\text{NO}_2)$ , why does Tab. 5 give four different parametrizations for four different ozone column ranges? A single parametrization should be given here to make things easier for readers who want to use these formulas. 2) What is the nature of the error limits of the parameters  $a_1$ - $a_6$  and are they of any relevance to estimate the quality of the parameterizations? Please note that for  $j(\text{NO}_2)$  most parameters vary more strongly if different ozone column ranges are compared than indicated by the errors of the parameters. So these errors have no relevance and pretend an accuracy that is not real. 3) Did you systematically test if simpler parameterizations give satisfactory results as well by taking out single parameters? 4) For  $j(\text{O1D})$  the parametrization appears arbitrary: parameters show no clear trend with ozone column although this would be expected even for an empirical formula. It would be more convincing to use a parameterization that contains SZA, AOD and ozone columns in a single formula. 5) Given that the data were probably (i) not normalized to a common Sun-Earth distance, nor (ii) to the same temperature; (iii) the AOD (380 nm) used does not apply strictly to the  $j(\text{O1D})$  wavelength range, (iv) only 30 DU wide ranges of ozone columns were merged, and (v) cloud-screening cannot be perfect, the obtained  $r^2 > 0.95$  is remarkable, also compared to Tab. 3 and 4, and should be rationalized.

Response: 1) I agree with you that a single parametrization should be given here. I have revised it. 2) The error limits refer to 95% confidence bounds of these parameters. When we used a single parametric equation for all data of  $j(\text{NO}_2)$ , the error limits get lower significantly than that of different ozone column classes. 3) Yes, I try using different functions to fit the relationship, and it seems that the quadratic polynomial form gave the best fit, which reflects the nonlinear relationship between AOD and  $j$ -values and considers the combined effect of the AOD and SZA on  $j$ -values. 4) I agree with you that it would be more convincing to use a parameterization that contains SZA, AOD and ozone columns in a single formula. I have revised it in this part. 5) All of the problems

of the data you have mentioned is true. Even if there are these problems in the data, I acquired a remarkable  $r^2$  ( $r^2 > 0.95$ ), indicating the fitted results is relatively rationalized.

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Lines 407-409: What do the percentage reductions refer to? See also abstract and conclusions.

Response: The percentage reductions refer to seasonal mean values under clear-sky conditions. I have revised it.

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Line 411: "... and lower SSA in winter" Was not shown.

Response: I have removed "... and lower SSA in winter".

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Lines 431-433: As mentioned above, this statement is not justified and the use of PM<sub>2.5</sub> would most likely lead to no improvement of estimated  $j(\text{O1D})$  or  $j(\text{NO}_2)$  unless you can show it directly.

Response: I have remove this part.

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Line 460, Figure 8: I assume what is shown in Fig. 8, and the 25% reduction stated in the text, refer to mean daytime ozone productions. Please specify time period.

Response: Yes, it refer to mean daytime ozone productions and I have revised it.

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Figure 9: How were the data shown derived, i.e. what periods of time do single data points represent?

Response: Single data points represent daytime hourly mean values.

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Figure 10: Indicate in the caption that the data represent mean values over a period of one month (or n clear-sky days) in August 2012.

Response: I have specified it in the caption.

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Figure 11: In the caption refer to Table 7 to explain the meaning of day A and day B.

Response: I have specified it in the caption.

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Table 7, 8: Mind significant digits.

Response: I have revised it.

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