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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Actinic flux and O^1D photolysis frequencies retrieved from spectral measurements of irradiance at Thessaloniki, Greece" *by* S. Kazadzis et al.

S. Kazadzis et al.

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Response to the reviewer #1 comments

Note: a figure has been added so all figures were rearranged. All the comments to the present document concerning the figures are made according to the new manuscript. The number of each figure now is higher by 1.

1)Page 4195 (25): The chemical process O3 + hv -> O1D + O2 was added to the text to clarify that the photolysis frequency in interest corresponds to the above process. The term J(O1D) was defined at its first occurence, that is in the first sentence abstract and used J(O1D) from there on. The exact definition was made in the 6th line of the introduction, including the reaction equation (with "+ hv").

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2)Page 4193 (15): The definitions of radiance and the angles following equation (3) were added just after equation (2). The addends on the right hand side of equation (2) were corrected and reported as “the direct and diffuse components of the actinic flux”.

3)Page 4199 (7): The paper (Kazadzis et al., 2000) where the radiance measurements details are explained was added as reference.

4)Page 4199 (11): Kylling et al. and Webb et al are assuming a constant value of A(λ)=1.73 for overcast conditions. “Overcast conditions” was added to the text. The statement concerning the assumption of an isotropic radiance distribution where A(λ)=2 applies also for overcast conditions. This can be calculated mathematically when I is independent of the solar zenith and the azimuth angle.

5)A look up table of A’s (diffuse actinic to diffuse irradiance) as a function of wavelength, solar zenith angle, optical depth and ozone was produced using radiative transfer model calculations. Practically at each (spectrum) conversion of irradiance to actinic flux, the solar zenith angle, the optical depth and the total column ozone measurements were used to determine an A(λ) through this look up table.

6)Page 4204 (1): The absorption cross sections and quantum yields that are used in this work were specified. The temperature for which the calculations were made was also specified. According to the recommendation of the reviewer more recent functions were used for the absorption cross-section of ozone (Daumont et al.., 1992) and the quantum yield (Sander et al., 2003). The parameterization of the method (polynomials) was recalculated using the new functions.

7)Page 4204 (23). The polynomials that were used for the evaluation of the method that is described in this paragraph are extracted from the ratio J(O1D)/Jps versus global irradiance at 325 nm (E325) and not the J(O1D) itself. The sentence was erroneous and corrected, all polynomials that were used in this method are ratios of J(O1D)/Jps as a function of (E325), similar with the ones described in figure 6.

ACPD

4, S2199–S2203, 2004

8)Figure 7. The ratios of the retrieved and measured J(O1D) exhibit some vertical structure. The mean ratio and standard deviation was found 1.001ś0.026 using the 5 solar zenith angle degree binning. The first approach of the parameterization was done by 1 degree solar zenith angle resolution and the results of the comparison similar to figure 7 was 1.007ś0.024. The decision on using 5 degree binning was made based on the fact that the retrieved polynomials were calculated using bigger amount of data for each bin. In that case an uncertainty of the fitting process that could be introduced would be smaller. However, both approaches give similar results with differences much less than the scatter of the calculated to measured ratio results. For the scatter that is shown in figure 7 we believe that this could be due to differences of the radiation field distribution for a constant solar zenith angle and a given irradiance at 325nm. For such cases the actinic flux to global ratio varies due to the different principles of the two quantities. For example in figure 6 for the 25-30 solar zenith angle bin and for constant irradiance of 0.25 (W. m-2 .nm-1) the calculation of J(O1D)/Jps through the polynomials will be a single point while in the figure there is a group of points. A combination of small differences in the aerosol optical depth together with different type of aerosols (different single scattering albedo) could lead to such small deviations. The points that demonstrate big differences in figure 7 are biased by the fact that for some cases the spectrum is destructed by fast moving clouds, where at the moment of the irradiance measurement at 325nm the cloud situation could be different than the rest of the scan. In this case the (E325) value used in the polynomial approach will not be representative for the full scan. For such cases, using a spectral actinic instrument or this method, the J(O1D) measurement or calculation will represent an “average” situation of the sky for three minutes (that the measurement lasts) and not an “instant” value.

9) In the conclusions for the second method is reported: “No evident dependence of the method on aerosol optical depth or total ozone column was found, which makes is suitable for use under different atmospheric conditions”.

ACPD

4, S2199–S2203, 2004

One of the main advantages of the second method is that there is no need to know the cloud or aerosol conditions of each measurement. The use of (E325) as an independent parameter in the polynomials acts as an indicator of the radiaton field situation. The use of a model calculation for clear sky conditions will bias the retrieval, while the polynomials will not work properly for cloudy conditions. A use of a combination of a clear sky plus a cloud model could be used, but still the decision of the amount of cases that will be used for each condition (cloudy or not) has to be based to actual atmospheric parameters. The final attempt could be to distinguish each case and use a model value. This will make the method very complicated (for sites that can not have all the available information for the model inputs). We think that the calculated to measured differences are quiet small for attempting to introduce a more complex method with a different "philosophy" than the one presented. Based n the above the polynomials retrieved for Thessaloniki could be biased by the random atmospheric conditions used in the dataset, of the site itself. That is the reason that we decided to use the same polynomials for an area where the cloudy conditions are much more frequent (figure 9). The deviations presented in figure 9 could be an indication that the amount of data (figure 6) used for the polynomial retrieval are providing a statistically quiet acceptable dataset for its use also to different atmospheric conditions.

10) The parameterization was included in the paper in table 4.

11) The 5% accuracy of the Brewer instrument is reported as the uncertainty of the absolute measuring irradiance on the sky. The investigation performed at the laboratory was the following: The set up of the global irradiance calibration of the Brewer instrument was used and the two instruments have measured one after the other the lamp output in a fixed distance as reported by the calibration certificate of the lamp. The uncertainty of this relative lamp measurement by both instruments is less than 2% as: Ţ Most of the uncertainty sources that are reported in the references (Bais, 1997, Gardiner, 1997 6th page, 1st paragraph) do not exist. For example the angular response error (vertical beam in the dark room), temperature effects (temperature con-

ACPD

4, S2199–S2203, 2004

trolled dark room) the calibration lamp absolute uncertainty, transfer of calibration from the primary lamp to secondary standards e.t.c., are not included in the experiment. Ţ The remaining uncertainty is mainly operational (exact measuring of distances e.t.c.). The fact that the same results were observed (with deviations less than ś2%) earlier with different set up and lamps. (Experiment that took place at ECUV-Joint Research Center, Italy on May 2002) leads to the point that the difference in the calibration lamps of the two instruments is the one described in the text.

For the analysis and the times series presented the absolute scale of the Brewer instrument was based in the normal calibration procedure and lamps that are regularly used. Only for the evaluation of the results comparing the two instruments this lamp difference was taken into account, with the aim to minimize other sources of deviations and concentrate on the ones of the method presented.

12) Page 4208 (26) The sentence “The accuracy of the retrieved data using the two methods presented in this paper is comparable to that of chemical actinometers measuring JO1D (Shetter et al., 1996, Muller et al., 1995).” was deleted.

Technical corrections were taken into account in the new manuscript

Figure 6: The irradiance used in the (XX’ axis) submitted graph (figure 5 in the submitted document) was erroneous (wrong wavelength) . The figure was corrected.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 4191, 2004.