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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Twilight tropospheric and stratospheric photodissociation rates derived from balloon borne radiation measurements" *by* A. Kylling et al.

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Response to the referees interactive comments:

Referee 1:

Specific comments:

1. The manuscript has been changed such that the letter E is used for the irradiance and F for the actinic flux. The two were correctly treated in the paper, but unfortunately the same letter was used for two different quantities. At the end of

the Calibration section the following paragraph have been added to explain how the measured irradiance were converted to actinic flux.

> Each head measures the irradiance. To convert from irradiance to actinic flux the contributions from all heads are summed as discussed in section 2.1. Finally the angular response correction is performed as explained below in section 4.

2. No correction to the data from the up-pointing sensor have been made to account for possible shading or reflection effects from the balloon or the rest of the pay-load. As stated in the paper the instrument is located about 100 meters below the balloon itself. Furthermore, the closest instrument above the NILU-CUBE is about 10 meters away. The majority of the diffuse radiation is concentrated around the horizon for the solar zenith angles during the flight. As such the contribution from the zenith is small compared to the rest of the sky. This is so both for the up-pointing sensor and the total actinic flux. No assessement as been made for the possible reflection of solar radiation from the balloon. It is however noted that as the balloon ascends the balloon increases in size. Hence, if any contribution from possible reflection is of importance it should become more and more evident in the measured data as the balloon ascends. However, no such signal is clearly evident.

3. Both referees comment that aerosols may have a large impact on the actinic flux. As demonstrated by e.g. AndersonD1995 stratospheric aerosols loadings found in the period after major volcanic eruptions may severely change the radiation field. The present measurements were made during background levels of stratospheric aerosols. Hence, in the lack of simultaneous measurements of the aerosol amount and optical properties, it appeared justifiable to use a standard background aerosol model. Sensitivity studies were performed with larger stratospheric aerosol loadings using the moderate, high and extreme aerosol models

of Shettle1989. For the moderate case the measurements and model simulations agreed slightly worse than for the background case presented in the paper. For the high and extreme cases the model was more than 50% lower than the measurements for altitudes up to 20 km. Given the sensitivity of the model to the presence of aerosols it should clearly be possible to use the NILU-CUBE to measure the effects of various stratospheric aerosol loadings on the actinic flux. The description of the aerosol input to the model has been changed to the following to accommodate the comments from the referees.

> No information about aerosols composition and concentration was available for the flight. As the measurements were made during a volcanically quiescent period in the stratosphere it is justifiable to use a standard background aerosol model. The aerosol extinction profile was taken from the spring-summer background aerosol profile of Shettle1989. The surface visibility, which for this aerosol model affects the aerosol up to altitudes of 2 km, was set to 50 km. The Henyey–Greenstein phase function was used with asymmetry factor from the above spring-summer background aerosol model. Sensitivity studies with higher aerosol loadings representative for high and extreme stratospheric aerosol conditions gave poor agreement between the model simulations and the measurements.

For solar zenith angles $> 92°$ for the 312 nm channel and $> 93°$ for the 340 nm channel the signal measured by the NILU-CUBE is getting close to the noise level. This is also evident from Figures 5 and 6. Hence the worsening of the model/measurement agreement for solar zenith angles $> 93°$ may be attributed to increasingly noisy measurements. To investigate possible aerosol effects require measurements with a higher sensitivity. The following sentence have been added to the measurement section to emphasize this:

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For solar zenith angles larger than about 92° (93°) the signal in the 312 nm (314 nm) channel gets increasingly closer to the noise level and the measurements are thus less reliable.

The asymmetry factor from the Shettle (1989) aerosol model varies with wavelength and altitude. Hence, no single number may be quoted. The explanation as given in the text "The Henyey–Greenstein phase function was used with asymmetry factor from the above spring-summer background aerosol model", is thus as precise as it is possible to get. The same is of course true for the aerosol single scattering albedo.

4. The referee says that the "used filter #2 (Figure 4) is in fact a very poor approximation to the actinic spectrum of NO_2" and argues that the actinic spectra of NO_2 and $O(^1D)$ should be added to the figure. The filters used in the NILU-CUBE were not chosen to approximate the actinic spectrum of any photodissociation process. Indeed, the actinic spectrum of NO_2 extends all the way up to 420 nm, well above the range of the x-axis in Figure 4. The actinic spectrum of O_3 leading to $O(^1D)$ varies largely with altitude, solar zenith angle, temperature and ozone amount. To find a "representative" filter for this actinic spectrum is not possible. The purpose of Figure 4 is to document the spectral response of the two channels. To add two arbitrary actinic spectra to the figure will make the figure more difficult to read. Furthermore, it is not clear how the manuscript will benefit from adding these actinic spectra. We have thus not added the actinic spectra to the figure.

5. The cross section for NO_2 have been updated to Davidson1988 and the quantum yield $O(^1D)$ production to that reported by Matsumi2002. The references have been changed to reflect this. In addition the following discussion on uncertainties have been added

The accuracy of the NO_2 cross sections are estimated to be

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$\pm 5\%$ Davidson1988. Calculation of J$_{NO2 with the NO_2}$ cross section of Schneider1987 are about 4-5% smaller then if using Davidson1988. The most recent recommendations of the "tail" in the O(^{1}D) quantum yield have been given by Matsumi2002. The parameterization of Matsumi2002 give 15-18% larger J$_{O(}$^{1}D)$for solar zenith angles betwen$ 82° and $92°$ than using the O(^{1}D) quantum yield of Talukdar1998. This is larger then the differences ($< 2\%$) reported by Matsumi2002, however, they reported differences for the surface while the differences reported here occurr at altitudes above 13 km.

It is beyond the scope of this paper to provide a full overview of the uncertainties in the cross section and quantum yields of the photodissociation rates presented in the paper.

Technical corrections:

1. page 719, "colocated" changed to "collocated".
2. page 721, "nor the rest of the payload" changed to "or other parts of the payload"
3. It is assumed that the referee here actually means page 725 and not 721. As Bosch2001 did indeed also measure we have removed the following sentence from the conclusions "To the authors knowledge this is the first comparison of a radiative transfer model and UV measurements made at such large solar zenith angles throughout both the troposphere and stratosphere". The Bosch2001 reference is included later in the conclusions.
4. The axis have been labelled and the figure caption have been extended to better explain what is shown.
5. The last paragraph of section 4 as been moved to the conclusions section.

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Referee 2:

1. The referee here points to an important issue not considered in the manuscript. Namely, how the calibration factors will change in dependence of the spectral irradiance. As pointed out by the referee the spectral distribution at 312 nm depend on the solar zenith angle and altitude. We performed model simulations to investigate how the calibration factors change for the solar zenith angles and altitudes encountered during the flight. For the 340 nm channel the changes were less than 1% for all altitudes and solar zenith angles. For the 312 nm channel the model simulations gave changes less then 3% for solar zenith angles smaller then 88° or altitudes less than 19 km. The changes increased for larger solar zenith angles and for solar zenith angles between 90° and 94° the calibration factor was high by up to 18%. The measurements may be corrected for these changes in the calibration factor. This has been done in the revised manuscript. Figures 5, 7, 9, and 10 has accordingly been updated. The agreement between measurement and model simulations shown in Figures 7 and 9 have improved for the 312 nm channel, especially for solar zenith angles between 90° and 94°. The changes do not affect the discussion of the results or the conclusions of the paper. The following text have been added to the calibration section.

> The calibration factors depend on the spectral distribution of the irradiance. Model simulations were performed to investigate how the calibration factors change for the solar zenith angles and altitudes encountered during the flight. For the 340 nm channel the changes were less than 1% for all altitudes and solar zenith angles. For the 312 nm channel the changes were less then 3% for solar zenith angles smaller then 88° or altitudes less than 19 km. The change increased for larger solar zenith angles up to a maximum of about 18%, giving too large measurements values, for solar zenith angles between 90° and 94°.

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The measurements in the 312 nm channel have been corrected to account for these changes in the calibration factor.

2. Concerning aerosols, please see answer to question 3 by referee # 1.

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