Reply to comments by Referee #2

Summary:
This paper characterizes the CCD-SR instrument that deploys on the HALO aircraft and continues the discussion of actinic flux measurements and calibrations using Metcon CCD-based spectrometers. The determination of spectral and wavelength assignment accuracies and sensitivities are discussed and a new technique for removing stray light was demonstrated to improve UV-B spectral accuracy and account for low limits of detection. Cutoff wavelengths are determined for each measured spectra based on radiative transfer modeled fluxes to reduce noise in photolysis products and examine stray light impacts. Ground-based comparisons with low-stray light instrumentation show strong agreement to near the detection limits. The resulting data are shown to be of sufficiently high accuracy for studies of actinic flux and calculated photolysis frequencies. This work is of value, well written and recommended for publishing after addressing the comments.

Reply: We thank the referee for the positive evaluation of the text. All comments (italic font) will be addressed in the following.

General comments:
P3 L3-4: The authors contend that "upward radiation in the UV range can often be neglected" at surface sites. Upwelling jNO2 is often 4-10% to the total photolysis and can be much greater. The upward UV radiation can rarely be neglected (though it can be estimated from the downwelling). This sentence should be dropped or adjusted.

Reply: We agree that the statement was too sloppy as was also noted by referee #1. We will clarifiy this and use the following statement instead:

"In contrast to ground-based operations where measurements of upward radiation in the UV range may be dispensable under conditions of low ground albedos, aircraft deployments require separate measurements in the upper and the lower hemisphere."

P12, L24-27: The authors may have some luck in that they found linear stray light structure in the UV-B in 4 of the 5 tested spectrometers (with the exception of 45853). The paper's entire stray light analysis relies on a linear-based subtraction. However, non-linear stray light response is not fully controlled for in the manufacturing and is determined by testing after production. Thus, I think they should state explicitly that determination of the stray light spectral structure is required to assess if a linear regression is appropriate.

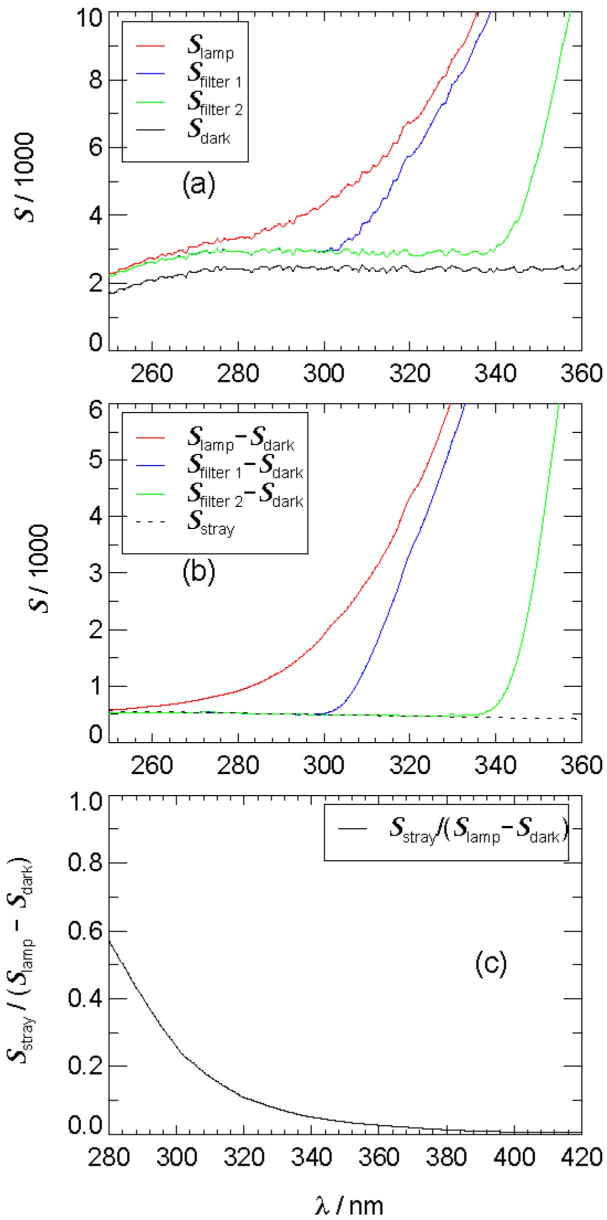
Reply: We agree that this should be stated more explicitly. Fig. 5 will be revised as shown in FigD2.1 below. Similar plots will be shown in the Supplement for all instruments. The corresponding paragraph was revised and extended following page 12, line 22:

"In panel (c) of FigD2.1 the fraction of stray light signals is plotted for the atmospherically most relevant wavelength range 280–420 nm. With increasing wavelength and lamp signals, the importance of stray light quickly diminishes to below 5% above 350 nm. Accordingly, uncertainties of the extrapolations become unimportant. Linear extrapolations of stray light signals were preferred in this work because they were most accurate in a range <350 nm

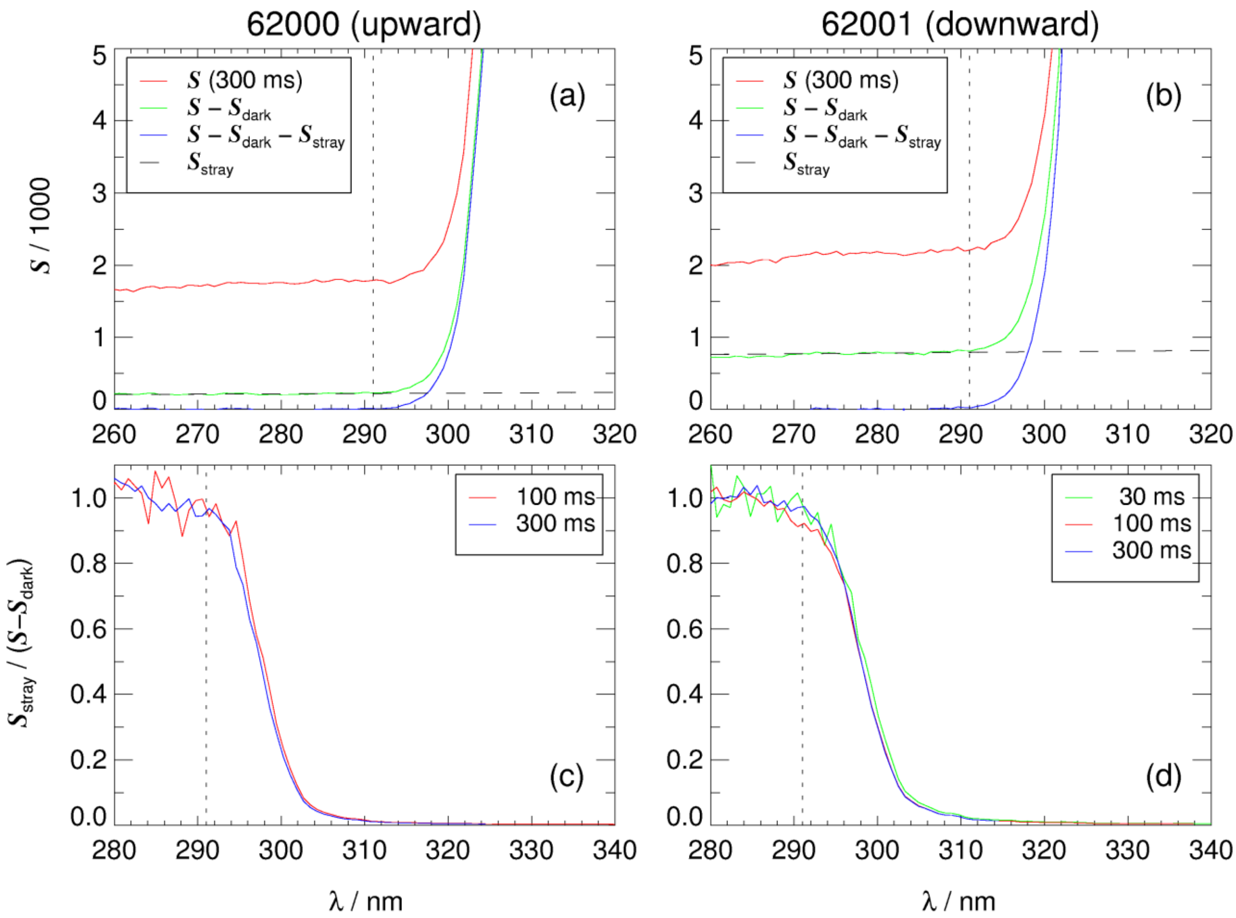
where the determination of D_λ is strongly affected by stray light. A modification of this procedure was only necessary for the oldest instrument 45853 where a stronger wavelength dependence and a leveling-off of the stray light induced signal around 340 nm was observed. For 45853 the stray light level was generally increased compared to the other instruments.
Plots as in FigD2.1 can be found for all instruments in the Supplement using the same typical optical receiver/fiber combination for direct comparison. If for other instruments a linear fit or a linear extrapolation of stray light signals towards longer wavelengths turns out to be insufficient, other functions should be tested to obtain an optimum description. For example, for 45853 a second-order polynomial was used for the extrapolation of stray light signals. This polynomial had the same slope at 300 nm as the linear approximation (270-300 nm) but was allowed to smoothly level out around 340 nm. Further cutoff filters with longer edge wavelengths were used to exclude significant spectral structures in the stray light signal at longer wavelengths. However, the spectral shape of the stray light signal without filter may differ from that observed with a filter below its cutoff wavelength because the filter can remove a substantial part of the radiation responsible for stray light. For that reason it is important to use filters with short cutoff wavelengths like a WG320 to get reliable results in the most affected wavelength range."

In the section on field measurements we come back to this issue. Also Fig. 10 will be revised as shown in FigD2.2 below. The corresponding paragraph was revised and extended following page 19, line 31:

"Expectedly, downward stray light signals in panel (b) were greater than upward stray light signals in panel (a). On the other hand, as shown in panels (c) and (d) of FigD2.2 the signal fractions caused by stray light are comparable for upward and downward measurements. Moreover, they quickly diminish with increasing wavelengths (≤1% above 320 nm). Accordingly, the uncertainty related with the linear extrapolation of stray light signals to wavelengths of up to 650 nm is insignificant. Tests with various cutoff filters confirmed that there were no significant stray light induced structures in the investigated spectral range. Of course, it cannot be excluded that for other instruments such structures exist or that stray light signals have to be approximated non-linearly. In these cases modified, instrument specific extrapolation procedures have to be developed, as already pointed out in Sect. 2.2.3."



FigD2.1: Example signals obtained during laboratory calibrations of instrument 62001 with two cutoff filters at Δt=1000 ms and close lamp distance. The wavelength range relevant for the determination of stray light signals is zoomed in. Saturation occurred around 400 nm for this integration time. (a) Total signals of lamp radiation with no filter (S_{lamp}), of lamp radiation with WG320 filter ($S_{\text{filter 1}}$), of lamp radiation with WG360 filter ($S_{\text{filter 2}}$), and dark signals (S_{dark}). (b) After subtraction of dark signals, stray light signals were estimated by linear regressions in a range 270-300 nm and extrapolated over the whole spectral range (dashed lines) before final subtraction. (c) Signal contribution of stray light in the atmospherically most relevant wavelength range. Similar figures for the other instruments are shown in the Supplement.



FigD2.2: Examples of flight raw data and evaluations of instruments 62000 (lower hemisphere, left) and 62001 (upper hemisphere, right). Data were obtained during a HALO flight on 20 Dec 2013 17:30 UTC over the North Atlantic (15.0N, 55.9W, 13.2 km) under conditions with few scattered low-lying clouds (solar zenith angle 47°, ozone column 245 DU). In panels (a) and (b) different colors indicate the evaluation steps: raw data (red), background subtraction (green) and stray light subtraction (blue). Stray light signals (dashed black lines) were determined by linear regression of background corrected signals in a range 270 nm to 291 nm (cutoff wavelength). In panels (c) and (d) the contributions of the inter- and extrapolated stray light signals are shown for the integration times eventually used in the displayed, most effected and atmospherically relevant wavelength range >280 nm.

P19, L13-15: The lookup tables are appropriate for clear-sky determinations of cutoff wavelengths, but no mention is made of the impacts of clouds and aerosols. Extinction by clouds and aerosols can greatly reduce the in situ measured flux and cutoff wavelength would be higher than expected from the table. In these cases, the variability in cutoff could be larger than 2 nm (as mentioned on P18 L5-6). Ideally the measured spectra could be evaluated for the cutoff, in concert with the model. The measurements are often most interesting in these complex atmospheric conditions and a discussion is needed here.

Answer: The idea behind the cutoff wavelengths is that they should set a safe lower limit to the wavelength range where significant flux densities can be expected and a safe upper limit for the range in which stray light can be determined. By choosing simulations under clear sky conditions and selecting downward radiation we ensure that under all conditions "real" cutoff wavelengths will be equal or greater than those in the lookup tables. We do not want to exhaust the method by approaching most realistic cutoff wavelengths for each spectrum

but rather stay on the "safe side". On page 19, line 14 we'll include the following paragraph to clarify this:
"Even though the cutoff wavelengths were derived from simulated clear sky downward actinic flux densities, they will be applied in the following under all conditions as well as for upward actinic flux densities that are typically much lower. The data from the lookup tables were taken as safe lower limits for convenience. An unaccounted presence of clouds, for example, would shift cutoff wavelengths towards slightly greater values which is non-critical for the data analysis."

P19-22: Section 3.1.2: A summary of the actinic flux total uncertainties was expected here. Uncertainties were discussed for the calibrations, but what are the measured uncertainties? What is the impact of the improved UV stray light determination that comprises much of the analysis in this paper and how does it improve over previous evaluations (Jakel et al., etc) . Also, what is the impact of stray light near detection limits where the uncertainty due to stray light determination is much larger (as a function of SZA, ozone, atmospheric conditions).

Reply: We'll try to answer all these questions in a revised version of section 3.2.

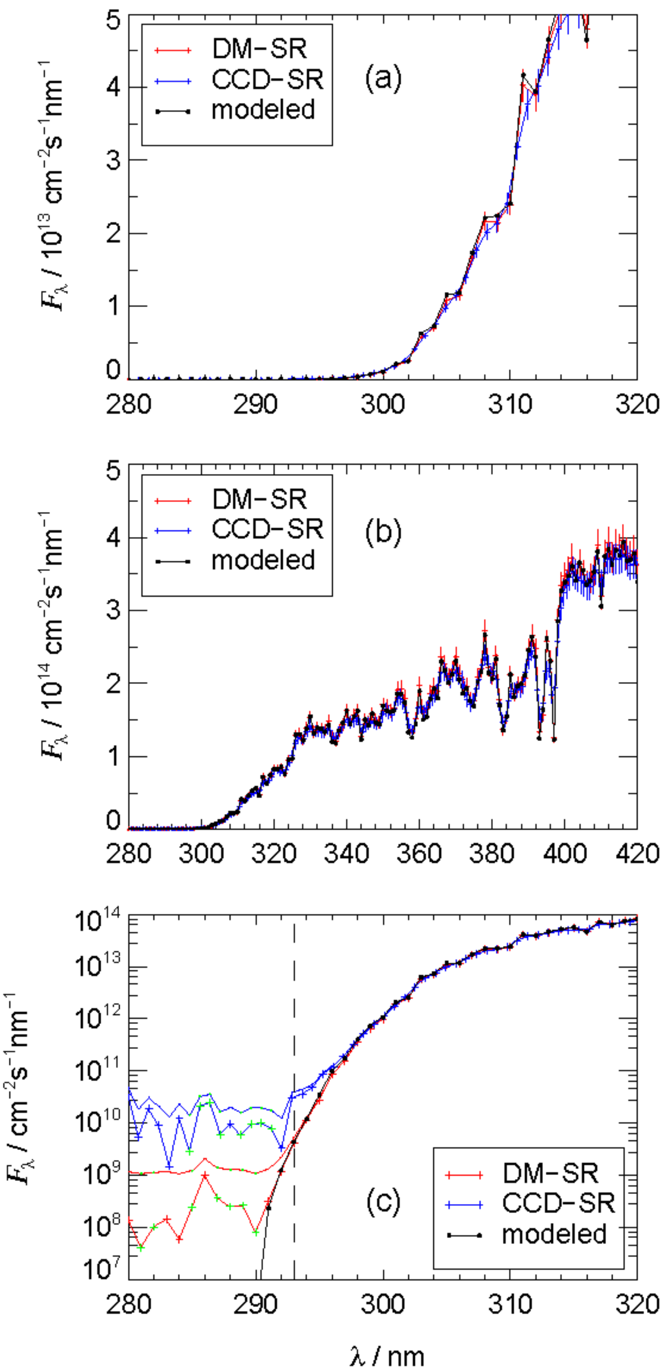
(1) As a prerequisite, measurement precisions were simulated based on spectra from a radiative transfer model and the signal dependence of instrument noise obtained in laboratory measurements. Please refer to the answer of point 29 of referee #1 and tables TabD1.1, TabD1.2 shown there.

(2) In the previous version of section 3.2 on page 23, line 30, the following statement was made: "A closer look at the CCD-SR residual noise during day and night reveals that while in the dark the noise is consistent with the F^{NE} obtained in the laboratory measurements (Fig 9), it is increased by a factor of up to 2–3 during daytime. This is attributed to additional noise induced by stray light." This statement was wrong because data were compared in a too wide wavelength range, still affected by atmospheric radiation.

(3) Section 3.2 was split up into three subsections. The first is dealing with actinic flux density spectra:

"FigD2.3 shows examples of actinic flux density spectra obtained simultaneously with the DM-SR and instrument 62001. The spectra were selected for stable, clear-sky conditions to avoid deviations caused by DM-SR scanning operations. The CCD-SR spectrum is a 10 s average obtained with a maximum 300 ms integration time. Minor optical receiver specific corrections (≈2% in this case) were already included for both instruments (Lohse and Bohn, 2017). Panel (a) shows the expected sharp increase of actinic flux densities in the UV-B range that is reproduced similarly by both instruments. Also shown is a radiative transfer model spectrum from the set of spectra produced to derive the cutoff wavelengths for ground measurements. The spectrum was selected for the indicated ozone column and solar zenith angle. In panel (b) the comparison is extended to the complete spectral range covered by the DM-SR. Generally good agreement is obtained except for sharp spectral features that are resolved more accurately by the reference instrument because of a smaller FWHM. The agreement of the modeled spectrum with the DM-SR is better because in the model calculations a matching FWHM of 1 nm was used. Error bars in panels (a) and (b) correspond to total uncertainties of 5–6% for both DM-SR and CCD-SR based spectral calibration uncertainties plus a 2% uncertainty from the optical receiver corrections. Additional uncertainties from instrument noise and stray light effects are invisible in panels (a) and (b).

Panel (c) of FigD2.3 shows the increase of UV-B spectral actinic flux densities in a semi-logarithmic plot where more details can be recognized. Here also data below the cutoff wavelength (vertical line) that are usually set to zero for the CCD-SR are shown for comparison. In this wavelength range data scatter around zero as expected, albeit with different residual noise. For the DM-SR the noise is unaffected by stray light and similar to nighttime values corresponding to F_λ^{NE}≈1×10^9 cm^{-2} s^{-1} nm^{-1}. Thus the DM-SR noise is smaller by a factor of about 10 compared to the expected F_λ^{NE} of the CCD-SR in the 280–290 nm range (TabD1.1). The residual noise of the CCD-SR is accordingly greater. From the measured signals, F_λ^{NE}≈1×10^{10} cm^{-2} s^{-1} nm^{-1} was derived and this value was added to or subtracted from the CCD-SR data in FigD2.3. Instead of error bars which cannot be reproduced in this representation, envelopes are shown comprising color coded positive and negative values for both instruments. A comparison with the radiative transfer modeled data shows that these are reproduced to well below the cutoff wavelength by the DM-SR while for the CCD-SR the detection limit is reached slightly above the cutoff wavelength. It should be noted that the ≈6% uncertainties shown in panels (a) and (b) are invisible in panel (c).
Considering that 10 s averages are shown in FigD2.3 the F_λ^{NE} are in fact greater than expected by a factor of about two. The reason is that for this particular deployment the CCD-SR sensitivity was reduced by a factor of ≈0.6 because of an increased fiber length compared to the data shown in Fig. 6. Moreover, even though stray light signals were subtracted, they induced some shot noise in addition to the dark noise. This increased the total noise by about 25% under local noon and clear-sky conditions which is the maximum shot noise increase expected from stray light under all conditions.
The subtraction of stray light signals (≈600 around noon) led to no appreciable increase of the noise even though these signals correspond to actinic flux densities of 1.5×10^{12} cm^{-2} s^{-1} nm^{-1} around 300 nm. It can be concluded that the interpolated stray light signals below the cutoff wavelength are well within 1% of the true values. If a corresponding additional uncertainty is assumed for all wavelengths, total F_λ uncertainties range between 0.06 × F_λ + F_λ^{NE} + 1.5×10^{10}cm^{-2} s^{-1} nm^{-1} around noon and F_λ^{NE} after sunset when stray light and atmospheric signals vanish (TabD1.1). The maximum stray light contribution to total uncertainties is therefore comparable to that of the CCD dark noise which only plays a role for very low values of F_λ below ≈10^{12}cm^{-2} s^{-1} nm^{-1}. The 1% assumption obviously is a rough estimate which may have to be adjusted for other instruments. In any case, the stray light signals approximately follow a cosine dependence on SZA and their relative importance increases with increasing SZA, at least for short wavelengths around 300 nm. Moreover, stray light signals are significantly greater in the presence of direct sunlight, i.e. the uncertainty would be lower for example for the upward F_λ shown in FigD2.2 or in the presence of clouds. Generally, the stray light influence on F_λ uncertainties is difficult to assess because it depends on instrument properties, measurement conditions and the procedure how stray light signals are determined."



FigD2.3: Comparison of actinic flux density spectra obtained on the ground with a double-monochromator based reference instrument (DM-SR) (red) and instrument 62001 (blue). Measurements were made on 01 Aug 2013 at Jülich (Germany) under clear-sky conditions. The spectra were taken around 12:00 UTC at a solar zenith angle of 33° and an ozone column of 310 DU. Black data points show the results of radiative transfer calculations for the same conditions. The different representations emphasize the increase of actinic flux densities in the UV-B range in panel (a) and the dynamic range of data in panel (b). Green data points in the semi-logarithmic plots of panel (c) show negative values that were plotted at their absolute values to make them visible. Full lines with minimum symbol size indicate the corresponding upper or lower limits after addition or subtraction of instrument noise. The dashed vertical line shows the cutoff wavelength below which values of the CCD-SR were normally set to zero. Note the different spectral actinic flux density and wavelength ranges.

(4) The influence of stray light signals on the uncertainties of photolysis frequencies will be addressed in a new paragraph in the new section on photolysis frequencies:

"In order to assess the estimated uncertainty of the stray light signal subtraction on photolysis frequencies, $j(O^1D)$ and $j(NO_2)$ were calculated from the subtracted stray light signals on a clear-sky day assuming again a 1% uncertainty for the inter- and extrapolated values. For $j(O^1D)$ the resulting total uncertainties strongly depend on whether or not data below cutoff wavelengths are taken into account. Without the help of cutoff wavelengths, total uncertainties are $0.06\times j(O^1D) + 7\times10^{-7}\ s^{-1}$ around noon and $1\times10^{-7}\ s^{-1}$ after sunset (TabD1.2). With cutoff wavelength these numbers are $0.06\times j(O^1D) + 1.3\times10^{-7}\ s^{-1}$ around noon and $0.3\times10^{-9}\ s^{-1}$ after sunset. Again it should be noted that despite greater absolute values at small SZA, the relative importance of these additional uncertainties increase with SZA reaching maxima of 3% and 100% around SZA=85° with and without cutoff wavelengths. Thus, the use of cutoff wavelengths expectedly also helps to confine the stray light influence. For $j(NO_2)$ the corresponding values are negligible under all conditions (<0.2%)."

(5) With regard to the methods of stray light determination the following paragraph was included following page 25, line 33:

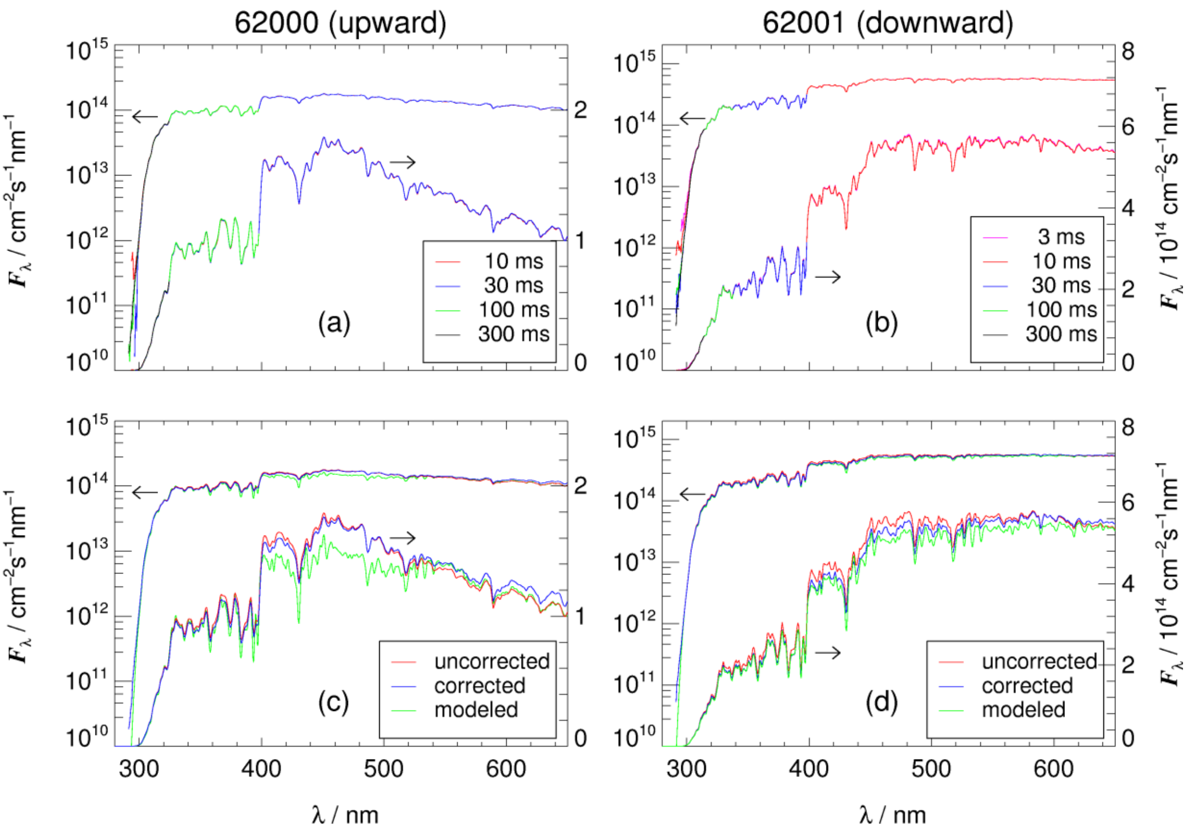
"However, for instruments without UG5 filter this procedure is no option because the spectral shape of the stray light is strongly influenced by radiation blocked out by a 700 nm filter. Moreover, the slope of the stray light signals during lamp calibrations was usually slightly negative while in the atmosphere they were typically slightly positive. For that reason laboratory measurements were not consulted to estimate the shape of atmospheric stray light signals also because the color temperature of the calibration lamps is much lower than that of the sun. This may explain why the stray light contributions in FigD2.2 diminish much faster than those in FigD2.1 and corresponding figures in the Supplement. Fitting the spectral dependence of measured stray light signals in a condition dependent range defined by the atmospheric cutoff wavelengths was therefore a manifest approach. Because the results were satisfactory no alternative procedures were systematically tested. The practice is believed to be widely transferable to spectral irradiance measurements."

P22, L17-18: I recognize the complexity of the optical uncertainties (or biases), but I do think a greater summary of the Lohse and Bohn, 2017 results would be appropriate here. They are critical to evaluation of the data and understanding the total measurement uncertainties. This would not hinder the more detailed and quantitative explanations I expect are in the separate publication.

Reply: In the revised version of the paper we will bring out more clearly that we are dealing with two separate aspects of the measurements. We'll rephrase the sentence in the abstract on page 1, line 18 to clarify this: "Because optical receiver aspects are not specific for the CCD spectroradiometers they were widely excluded in this work and will be treated in a separate paper in particular with regard to airborne applications."

And we'll add the following sentence on page 3, line 6 to explain this: "Since these corrections are complex and independent of the type of spectroradiometer, we attend to this difficulty...."

We agree that for an assessment of total uncertainties more information on receiver specific correction factors is important. We will therefore show corrected and uncorrected spectra in the revised version of Fig. 11 (aircraft data, see FigD2.4 below) and more explicitly state the (low) extent of the corrections for the presented ground based measurements. Because the discussion on uncertainties is already quite elaborate, more details on receiver related corrections under various conditions would be confusing.



FigD2.4: Evaluated upward (left) and downward (right) spectral actinic flux densities of the data shown in FigD1.5 in linear and semilogarithmic representations. Arrows point to the respective axes. In panels (a) and (b) spectra obtained with different integration times are plotted upon each other. Because already for 30 ms (a) and 10 ms (b) no saturation occurred, the data shown for the shortest integration time in each panel were not used for the final optimization of spectra. In panels (c) and (d) the spectra denoted as uncorrected (red) represent the optimized spectra of panels (a) and (b). Optical receiver specific corrections led to the slightly modified, corrected spectra (blue). Results of radiative transfer calculations for clear-sky conditions are denoted as modeled (green).

Technical comments:

P1 L13-14: Stray light is not strictly noise but rather a bias that varies with solar intensity. Reword.

Reply: We'll rephrase the sentence to "The spectra expectedly revealed increased daytime levels of stray light induced signals and noise below atmospheric cutoff wavelengths."

P1 L7: "1×10ˆ10 cm−2s−1nm−1", Units should be photons cm-2 s-1 nm

Answer: According to SI conventions qualifying terms like "photons" or "molecules" should not be part of a unit and in this case there is no risk it could be confused with any other unit. Referee #1 even asked us to remove "photon" from the name of the F-variable which we sometimes use to clarify the nature of the flux densities we are talking about. But we agree that this is not essential. To make sure there is no confusion we inserted a clarification following page 2, line 17 where the quantity is introduced: “F_λ is inserted in corresponding molecular units ($cm^{-2}s^{-1}nm^{-1}$)”.

P10, L5-6: I believe this should read “the SNR increases with the square root of the integration time and also improves at the shorter lamp distance”

Answer: We agree that singular sounds better. We’ll also change it in the lines above.

The following four points will be changed as recommended:

P10, L9: Expand PTB to Physikalisch-Technische Bundesanstalt

P11, Fig 5 caption: Change to “. . .stray light signals were also subtracted”

P12, L29: Remove “also”

P13, L11: Remove “also”

P22, L2: Remove “Evidently, also”