## Answer to reviewer 2

P10L16 reads: "For the multiple linear regression analysis, daily values will be used instead of anomaly values." This appears to be in disagreement with Eq. 6 and figure labels. UV has a unit W/m2, hence a dose rate. RAD has a unit W h/m2 (please convert to J/m2) Is Eq. 6 really a regression for daily UV-values? How can this be without reference to either the solar elevation angle or day of the year? Are not the anomalies addressed here? This has been addressed in the review stage before publication in ACPD. The answer to the reviewer has been repeated here:

**P10L16 reads: "For the multiple linear regression analysis, daily values will be used instead of anomaly values." This appears to be in disagreement with E.q. 6. and figure labels:** We do actually use daily values as input for the multiple regression model. Only for the change point analysis and the linear trend analysis, we used anomaly values.

UV has a unit W/m2, hence a dose rate. RAD has a unit W h/m2 (please convert to J/m2) There was an error in the unit of UV, which is actually expressed in J/m<sup>2</sup>. This error has been corrected throughout the manuscript. Global solar radiation values have been converted to J/m<sup>2</sup>. Is Eq. 6 really a regression for daily UV-values? How can this be without reference to either the solar elevation angle or day of the year? Are not the anomalies addressed here? Equation 6 uses the following information for the different parameters: UV: daily erythemal UV dose => the reference to the solar elevation is included in the global solar radiation term. Global solar radiation: daily total global solar radiation

Ozone: daily mean total ozone column value AOD: daily mean AOD value

To clarify this section, the following sentences have been added to the manuscript: 'As opposed to the previous analysis methods, the MLR is applied to daily values (instead of monthly anomaly values). For UV and global radiation, the daily sums are used, whereas for ozone and AOD, daily mean values are used.'

The next sentence on p23, 7-9, reads as if the authors have never been in this particular field of work. This remark is a rather trivial one.

This has been addressed in the review stage before publication in ACPD. The answer to the reviewer has been repeated here:

This sentence has been removed. Also, from the conclusion, the following sentence has been removed:

'For all seasons, the relationship between erythemal UV dose and TOC is negative, whereas the influence of global solar radiation and AOD is positive. The summer season is an exception as the regression coefficient of AOD is negative.'

Only the AOD is discussed. The single scattering albedo (SSA) is only discussed briefly. To the reviewers' opinion, an analysis of the AOD is quite limited without an elaborate discussion on the SSA, and on the size distribution of the aerosols.

As there are no measurements of SSA or size distribution available at Uccle, we are not able to discuss these parameters. We have recently installed an aethalometer and nephelometer at Uccle, so in the future their measurements can be combined to derive the SSA, which can then be studied together with the AOD. We agree that the size distribution would contribute to the understanding of what exactly will be the effect of aerosols at Uccle on UV. It would be interesting to know whether submicron particles ( $<< 1 \mu m$ ) or coarse particles ( $>1 \mu m$ ) dominate as an increase in UV radiation due to multiple scattering is less likely to be expected when coarse particles dominate (which scatter in the forward direction).

Ch 4.4.2 already states the following: "Both the aerosol composition, which determines if a mixture is rather scattering or absorbing, the aerosol amount, and the aerosol size distribution determine whether an increase in  $\tau_{aer}$  will lead to either an increase or decrease in UV irradiance. At Uccle there is not sufficient information on both parameters to unambiguously characterize the influence of Aerosol Optical Depth on UV irradiance."

#### Changes to the manuscript:

The last sentence will be changed a little and we added a new sentence:

"At Uccle there is **no information** on both parameters, **hence it is difficult to** unambiguously characterize the influence of Aerosol Optical Depth on UV irradiance. **Recently, a nephelometer and an aethalometer have been installed at our site in Uccle, so in the future, their measurements can be combined to derive the single scattering albedo value. This will shine a new light on the influence of the aerosols on the UV radiation at Uccle.**"

#### What means "adjusted" with respect to R2?

This has been addressed in the review stage before publication in ACPD. The answer to the reviewer has been repeated here:

Adjusted means that the sample size and number of explanatory variables have been taken into account for the calculation of the  $R^2$  value as opposed to the normal  $R^2$  where this is not the case. As additional variables are added to a regression equation,  $R^2$  will increase even when the added variables have no real predictive capability. The adjusted- $R^2$  is an  $R^2$ -like measure that avoids this difficulty and the value will only increase when the new variables have additional predictive capability. The adjusted  $R^2$  is defined as:

$$\hat{R}^2 = 1 - (1 - R^2) \frac{n - 1}{n - p - 1} = R^2 - (1 - R^2) \frac{p}{n - p - 1}$$

where p is the total number of regressors in the linear model (not counting the constant term), and n is the sample size.

This is explained in the sentence (p 13, line 22): 'The adjusted R<sup>2</sup> value is the measure for the fraction of variation in UV explained by the regression, accounting for both the sample size and the number of explanatory variables.'

Some extra information has been added:

'Compared to the  $R^2$  value, the adjusted  $R^2$  value will only increase if a new variable has additional explanatory power.'

Is "total column ozone" meant with "total ozone", ea. title of par. 2.3? Does that also applies to "...total and UV radiation can be either positive or negative." (p. 25, L29)? This has been addressed in the review stage before publication in ACPD. The answer to the reviewer has been repeated here:

Yes indeed, with 'total ozone' we mean 'total ozone column' or TOC. This abbreviation is now explained in the abstract and is used together with 'total ozone column' throughout the text. Concerning p.25, L 29: we made the following changes to this sentence: "...-total global solar radiation and UV radiation can be either positive or negative." (now p. 26, L13).

'TOC' is used only once; hence no need to introduce this abbreviation.

This was addressed in the review stage before publication in ACPD. However, according to the comments of reviewer 1, we decided not to use TOC but  $Q_{03}$  to represent total ozone column throughout the article.

The use of the abbreviation is not consequent. Please use one meaning for 'UV' and write the definition. Now it is in the title of the paper and in the abstract without out reference. This has been addressed in the review stage before publication in ACPD. The answer to the reviewer has been repeated here:

UV has now once been defined as UltraViolet (in the abstract, see answer above) and now only has this meaning throughout the rest of the text.

It appears that the authors refer to the regression coefficients (Eq. 7 - 10) as being trends. As what the reviewer understands from the paper, the seasonal variability of the seasonal means is described by Eq. 7 - 10. So where does the trend (which means linear change over time) come from? It might be that the reviewer completely missed the point here, but in that case, a better explanation should be given on what is actually done.

No trends were derived from the multiple regression coefficients! The signs of the regression coefficients are used to have an idea of the relation between UV and the different parameters.

What is also being discussed is the influence of the variation of the different parameters on the variation in UV, which represents the change in UV caused by a change in the different parameters.

## Change to the manuscript:

Added to Ch 4.4.2: (before "Changes in the variation of  $S_g \dots$ "):

"To determine the influence of the variation in the parameters on the variation in UV, the standard deviation of each parameter is multiplied with its corresponding regression coefficient,

which is then divided by the average  $S_{ery}$  value. This will give an idea of the magnitude of the influence of each parameter on UV. The results are given in table 10."

The caption of table 10 has been changed into: seasonal influence of the variation of  $S_g,\,Q_{O3}$  and  $\tau_{aer}$  on  $S_{ery.}$ 

The first line of the conclusion section is not what has been analysed in the paper; the paper does not support the remark that in recent years the focus of atmospheric research has shifted towards the variability of surface UV.

This sentence has been removed from the conclusion section.

#### Term 'Monthly anomalies' is used prior to an explanation.

This has been addressed in the review stage before publication in ACPD. The answer to the reviewer has been repeated here:

A sentence explaining the meaning of monthly anomalies is added to the introduction: P 5, line 29: 'Monthly anomalies are used to reduce the influence of the seasonal cycle on the analysis and are calculated by subtracting the long term monthly mean from the individual monthly means.'

A scatter plot showing the result of Eq. 6 versus the actual UV-measurements would help to grasp the idea.

This has been addressed in the review stage before publication in ACPD. The answer to the reviewer has been repeated here:

A new figure has been added to the manuscript showing a scatterplot of measured and modeled erythemal UV doses. This is now figure 7. Also, the old figure 8, which did not suit as a validation figure for the seasonal models, has been replaced by a new figure (now fig. 9 due to the addition of an extra figure) in which 4 new scatter plots are presented (one for each season). This figure will be discussed together with figure 10 to describe the seasonal models.

P26, L 2-3: "These small particles would enhance the multiple scattering and reflection of UV radiation, which in turn would increase the UV radiation observed at the surface of the Earth." The reviewer has strong doubts here. This would only apply if the source of radiation is located at the surface of the earth. The total transmission of solar radiation through the atmosphere is always reduced when more scattering particles are present, independent on their SSA and size.

Scattering of radiation (waves), deals with amplitude, while reflection is an amplitude-squared property of a scattering medium. Hence, writing "the multiple scattering and reflection of UV radiation" is a bit too compact.

We agree with the reviewer that the entire process of transmission of radiation through the atmosphere is very complex, but it is not the intention of this article to describe this whole

physical process in detail. It is true that the presence of any kind of aerosols normally reduces the amount of radiation that reaches the ground. However, if there were predominantly particles of size much smaller than the UV wavelengths (i.e. freshly formed particles, Aitken mode particles) and of high SSA, the UV radiation could be enhanced by the mulitple scattering by these aerosols, as long as the total amount of all aerosols does not exceed a certain (yet, in our study not possible to determine) threshold value, from which on extinction would dominate and the UV radiation would decrease. Thus, there are many variables (total amount, size distribution, composition, SSA), interacting with each other, which determine whether the atmospheric aerosol composition can enhance or reduce UV radiation:

## Changes to the manuscript:

We made the following changes to Ch. 4.4.2 of the manuscript to try and clarify this: (After "... if the increase in  $\tau_{aer}$  was caused by an increase in the amount of small scattering particles.":)

"If there were predominantly particles of size much smaller than the UV wavelengths (i.e. freshly formed particles, Aitken mode particles) and of high SSA, the UV radiation could be enhanced by the multiple scattering by these aerosols. However, when the amount of all particles exceeded a certain (yet, in this study not possible to determine) threshold value, extinction would take over and from this point, an increase in AOD would lead to a decrease in UV irradiance. Both the aerosol composition, which determines if a mixture is rather scattering or absorbing, the aerosol amount, and the aerosol size distribution determine whether an increase in AOD will lead to either an increase or decrease in UV irradiance."

## **Presentation details**

Axis labels are often too small or not easily readable. Different ways to label should be sought.

This has been addressed in the review stage before publication in ACPD. The answer to the reviewer has been repeated here:

This comment has been taken into account and all the figures (except fig. 1, 2 and 3) have been altered so that the axis labels are more easily readable. We will improve fig. 2 and 3 for the next stage of the review.

#### Fig2 to 4: a legend is missing or text should be added to the caption

This has been addressed in the review stage before publication in ACPD. The answer to the reviewer has been repeated here:

The caption of figure 2 has been changed:

'The black line represents the time series of monthly anomalies of total ozone column (1991-2013). The blue (dashed) line represents the (insignificant) negative trend before the detected

<sup>&#</sup>x27;The black line represents the detrended time series of monthly anomalies of erythemal UV dose (1991-2013). The red (dashed) lines represent the (insignificant) positive trends before and after the detected change point. The grey lines represent the mean before and after the change point. 'The caption of figure 3 has been changed:

change point and the red (dashed) line represents the (insignificant) positive trend after the change point. The grey lines represent the mean before and after the change point.' The following information has been added to the caption of figure 4:

'The blue lines represent the time series, whereas the red lines represent the trend over the 1991-2013 time period.'

Fig 8 can hardly be read as a validation figure; it does not convince the reader that a validation is carried out there. In addition, season borders are not as normally defined: winter does not end on Dec 31 as this figure suggests.

This has been addressed in the review stage before publication in ACPD. The answer to the reviewer has been repeated here:

This figure has been replaced by a new figure (figure 9) in which 4 scatter plots present the behaviour of the seasonal models (measured versus modeled erythemal UV values).

# Additional changes to the manuscript: (remarks from the quick reports before publication in ACPD)

You may want to consider and discuss what follows from the fact that the variables may not fully meet the distribution requirements of linear regression.

One of the assumptions of multiple linear regression is that the errors of a multiple linear regression should be normally distributed. Non-normal errors may mean that the t and F statistics of the coefficients may not actually follow t and F distributions and that the model might underestimate reality. However, as stated in Williams et al. (2013), even if errors are not normally distributed, the sampling distribution of the coefficients will approach a normal distribution as sample size grows larger, assuming some reasonably minimal preconditions. In this case, inferences about coefficients will usually become more and more trustworthy. As we have a rather large sample size in this study, we assume that the distribution of the coefficients approaches normality.

# Changes to the manuscript: Ch. 3.2.3: (After "Data from 2009 to 2013 will be used for validation of the model."):

"For the MLR analysis to produce trustworthy results, the distribution of the errors of the model should be normal. Non-normal errors may mean that the t- and F-statistics of the coefficients may not actually follow t- and F-distributions and that the model might underestimate reality (Williams et al. (2013)). However, as stated in Williams et al. (2013), even if errors are not normally distributed, the sampling distribution of the coefficients will approach a normal distribution as sample size grows larger, assuming some reasonably minimal preconditions. As we have a large dataset available at Uccle for the MLR analysis, we can assume that the distribution of the coefficients of the MLR model approaches normality."

+ New reference:

Williams, M.N., Gómez Grajales, C.A. and Kurkiewicz, D., Assumptions of multiple regression: correcting two misconceptions, Practical Assessment Research & Evaluation, Vol. 18, No. 11, ISSN 1531-7714, 2013.

Secondly, ozone column as such is taken as a linear independent variable although we know that the attenuation of radiation in media is not linear if Beer-Lambert law is true.

At our latitude, the variation in ozone throughout the year is rather limited. This is especially the case when we look at seasonal data, where the variation in ozone is the biggest during spring. Because of the rather small variation in ozone, we can consider ozone to be a linear independent variable between its limit values.

## Changes to the manuscript:

Ch. 3.2.3: after equation 3:

"Although the attenuation of radiation by ozone is not linear (according to the Beer-Lambert law), we consider total ozone column as a linear independent variable, based on the limited variation of this variable throughout the year and throughout the different seasons."

Thirdly, the independence of the explanatory variables is quite right tested in Ch. 4.4 and found satisfactorily low. However, p 26 states that the aerosol optical depth and the global solar radiation are linked to each other. Why was that not seen when testing the independence?

In literature, both parameters are sometimes related to each other (global dimming/brightening versus AOD), but at Uccle, there seems to be no relation between the two parameters. The parts where it was stated that the AOD and global solar radiation are linked to each other have been removed from the manuscript.

Page 14-15 has the text "the change point in the detrended time series is located around February 1998 (fig. 2). Since no calibration of the Brewer instrument took place around that period, it seems that the change point is not caused by known instrumental changes but rather by natural/environmental changes" which is confusing. Can you be sure that the instrument does not change or drift if it is left unattended and uncalibrated? Isn't the regular calibration rather needed to detect any drift and to remove it from the data? And don't you tell on page 6 that the instrument was calibrated on a monthly basis. Please rephrase something if I misunderstood.

This has been addressed in the response to the reviewer above.