

## **Answer to reviewer 1**

On p.11, line 1-2: "Monthly means are only calculated for months with at least 10 individual daily values". This leaves the question why the minimum is 10 days and why not 15, 20, or 25, and what the effect is of accepting a monthly mean with no more than 10 or 15 days of data, eventually only covering the first or the very last part of the month while the long term monthly mean used in calculating the anomaly probably represents the entire month. During a month of a large change in solar elevation or other factors the lack of up to 66% of the days may cause a bias. Is it beyond the scope of this work to study the added uncertainty due to the missing days?

The decision to use a minimum of 10 days for the calculation of monthly means was made because of the low frequency of AOD data. For UV, global radiation and ozone, there is enough data to use a minimum value of 20 or even 25 days. For AOD however, only 5 monthly mean values remain for the entire time period when using a minimum of 20 individual daily values. This is of course not enough to determine any kind of reliable trend.

The table below shows how many monthly values are available (out of a possible max of 276 months for each parameter when using a minimum of 10 or 20 daily values.

	# months with min. 10 daily values	# months with min. 20 daily values
Erythemal UV dose	268	235
Global radiation	276	276
Total ozone	276	274
<b>AOD</b>	<b>92</b>	<b>5</b>

We recalculated the trends for UV, global radiation and ozone, using at least 20 individual daily values and this did not significantly change the trend. But the reviewer is correct in stating that using a lower number of individual daily values causes an uncertainty in the calculated trend (at least for AOD) due to values not being equally distributed over a month. (We looked at the distribution of the daily AOD values and in 55% of the cases, the values were evenly distributed.) However, we prefer to have an AOD trend based on 92 monthly values instead of one based on only 5 values. To study the added uncertainty due to the missing days would mean to make assumptions on AOD for those days. As atmospheric aerosol properties are, however, in general very variable, such an exercise would be too speculative in our view.

### **Changes to the manuscript:**

Added to Ch. 3.2: (after 'Monthly means are only calculated for months with at least 10 individual daily values.')

“For  $S_{ery}$ ,  $S_g$  and  $Q_{O_3}$ , accepting monthly means with only 10 daily individual values does not have an impact on the calculated trends, as respectively 85%, 99% and 100% of the months consist of more than 20 individual daily values. For  $\tau_{aer}$  however, the number of available monthly mean values is dramatically reduced (from 92 to only 5 remaining values) when only accepting monthly means based on 20 individual values. There is a risk in accepting months with only 10 daily values, as those days could be concentrated at the beginning or end of a month,

which could bias the calculated trend. However, the benefit of using 92 instead of 5 monthly mean values for  $\tau_{\text{aer}}$  trend calculations outweighs this potential bias.”

P.15, Ch.4.1.1. leaves many questions. It may be unclear for the reader firstly why a linear trend can cause a change point in the time series (a), and secondly why the instrument was not calibrated in early 1998 although Ch.2.1. suggests that it was calibrated every month (b).

a) A change point is detected when there is a significant change in the mean before and after a certain point in the time series. If there is one clear, statistically significant, trend present in the time series, a significant change point will always be found in the middle of the time series, because at this point in the time series, the change in the mean will be large enough to be significant. This is why it is necessary to subtract this trend from the time series in order to find a change point (other than the one that was caused by the presence of the linear trend).

#### **Changes to the manuscript:**

We have added the following sentence at the end of Ch. 3.2.2 to clarify this:

“When there is a clear and large enough, statistically significant trend present in the time series, this automatically leads to the detection of a change point in the middle of the time series as, at this point, the change in the mean is large enough to be significant. In this case, it is necessary to detrend the time series, i.e. subtract the general trend from the time series.”

b) There is indeed a monthly calibration, however, the calibration constants don't necessarily change from one month to the other. We revised this part a little bit as we meant to say that there was no change between the calibrations.

#### **Changes to the manuscript:**

The last sentence of Ch. 4.3.1. has been changed into: “**Since there was no change in the calibration constants of the Brewer instrument around that period**, it seems that the change point is not caused by known instrumental changes but rather by natural/environmental changes.”

On p.16 the sentence "No ozone calibrations were performed around 1998, so the change point has no known instrumental cause" is confusing. Can the instrument not drift or change by itself and, if not, then why is any calibration ever needed? A calibration, and not the lack of it as suggested in the text, in general would ensure that the data are fine. The abrupt change seen in Fig.3 would suggest an instrumental change because our first assumption is that the atmosphere only changes slowly. Are there any further facts telling that the instrumental change can be ruled out?

We meant to say that there is no shift in the calibration of the instrument, hence there is no cause to believe some instrumental change led to a change point. It is of course always possible that the instrument drifts by itself, but this is checked by the internal lamp tests performed at regular times. If these tests detect a drift, this is corrected for. So this rules out an instrumental cause for the observed change point.

#### **Changes to the manuscript:**

Added at the end of Ch.2.3:

“Internal lamp tests are performed on a regular basis to check whether the instrument itself is drifting. When instrumental drift is detected, it is corrected for.”

Also, the following sentence at the end of Ch. 4.3.3.:

“No ozone calibrations were performed around 1998, so the change point has no known instrumental cause.”

has been replaced by:

“There was no change in the calibration constants of the Brewer instrument around 1998, so the change point has no known instrumental cause.”

P.19-20, Ch. 4.3.1. is discussing the trends in UV observed at other sites. Here the recent work by Eleftheratos et al (2014) could be included if relevant. However, the concept of "UV" remains unclear in this chapter. Probably it is not the same as in the analysis of data from Uccle, i.e. the daily or monthly dose of erythemal irradiance. If this is the case, then you could discuss whether the different trends listed are truly comparable. Perhaps one or two observations per day at SZA = 60 or 65 degrees do not represent the daily sum. Or do they, in a trend analysis? You may suggest this problem to be analyzed in a later study.

At Uccle, we use daily doses, which includes all effects (such as those from clouds), whereas using a fixed SZA does not cover this. As such, the reviewer is correct when stating that the trends are not truly comparable. This could indeed be analysed in a later study.

The work of Eleftheratos et al. (2014) is very interesting, but as it is focused on high latitude sites, we have decided not to include it in section 4.3.1.

### **Changes to the manuscript:**

Added to Ch. 4.2.1: (after “... falls within the range of trends reported in literature.”):

“However, for the comparison of these trends, it has to be taken into account that not all trends in Table 6 are calculated in the same way as the one at Uccle. At Uccle, trends are based on monthly anomalies which are essentially calculated from daily doses. As such, all effects such as those from clouds are included in our analysis. Some of the studies from Table 6, report trends at a certain fixed solar zenith angle, which does not cover the same range of effects as the daily sum does and thus, the trends may not be truly comparable. The possible effect of a different concept of UV could be subject of a later study.”

On p.23 and on, Ch. 4.4. it is to be remembered that in Eq.(6) the  $S_g$  was derived from 10-minute and 30-minute data. The resulting modelled erythemal daily dose then has a much better time resolution than the measured UV dose. If the time resolutions were the same, the regression should probably be better. The largest outliers in the lower panel of Fig. 8 are likely to be a result of varying cloudiness that is poorly monitored by the Brewer. In your **future** work you may want to experiment by re-sampling  $S_g$  for the times of the Brewer UV scans only to get a better correlation coefficient than 0.96 (p.24, line 6). Also cf. the discussion by den Outer et al, 2005: UV radiation in the Netherlands: Assessing long-term variability and trends in relation to ozone and clouds. J.Geoph.Res., 110, D02203, doi:10.1029/2004JD004824 (2005).

Thank you for this useful comment! We will keep this in mind for future analysis.

On p.15, Ch. 4.1.2., please, state whether the trend in global solar radiation was positive or negative, and give the value, too.

The trend in global solar radiation was positive. The value is given in section 4.2.2 (+4 %).

**Changes to the manuscript:**

Ch. 4.3.2:

“Similar to the erythemal UV dose time series, there is one general **positive** trend present, which explains the detection of a change point near the middle of the time series.”

The measurements at Uccle started at about the same time as Mt. Pinatubo erupted. What is its expected effect on the time series? To what extent does the observed recovery of ozone actually show the return to the stratosphere of the pre-Pinatubo time and to what extent the influence of the regulations of the Montréal Protocol? If this further analysis is beyond the limits of this work, it could be mentioned both in the analysis and in the conclusions (p.28), perhaps in the abstract, too.

Thank you for this interesting comment! We decided to calculate the trends for the time period after the Pinatubo eruption (1994-2013) (as was done by Eleftheratos et al. (2014)) and compared them with the trends for the entire time period. The results are presented in the table below:

	<b>1991-2013</b>	<b>1994-2013</b>
<b>Erythemal UV dose</b>	6.91% (+/-1.54%)	7.20% (+/-1.83%)
<b>Global radiation</b>	4.29% (+/-1.31%)	4.36% (+/-1.64%)
<b>Total Ozone Column</b>	2.61% (+/-0.44%)	2.52% (+/-0.50%)
<b>Aerosol Optical Depth (320nm)</b>	-7.61% (+/-4.51%) ( <i>not sign.</i> )	-4.32% (+/-5.05%) ( <i>not sign.</i> )

Apparently, for Uccle, there is no big change in the calculated trends for the period with (1991-2013) and without (1994-2013) the Pinatubo eruption. As a result, we can conclude that the observed recovery is much more a result of the regulations of the Montréal Protocol than it is a result of the return of the stratosphere to pre-Pinatubo time.

**Changes to the manuscript:**

Added to Ch. 4.1.3: (after “..., it seems that ozone has been recovering over the past 10 years.”):

“Removing the Pinatubo period (1991-1993) from our analysis, does not change the trend in ozone significantly, which means that the observed recovery in ozone is not so much related to the return of the stratosphere to pre-Pinatubo time, but that it is more likely a result of the regulations of the Montréal Protocol.”

And also in the conclusions section:  
(after “..., following the regulations of the Montréal Protocol.”):

“The trend in the ozone time series at Uccle does not seem very affected by the eruption of the Pinatubo, which took place in June 1991.”

On p.18, line 5-8, the finding that the minimum values of global solar radiation have a large trend is most interesting. The conclusion "...this could mean that the cloud properties (such as cloud optical depth) changed over the past 23 years" may be too careful. Instead, you could probably say that "the cloud properties, i.e. their amount and/or water content, must have changed". The last sentence of the chapter "However, this is difficult to prove without direct information or measurements on cloud amount and/or properties" could be removed.

The suggested changes have been applied to the manuscript.

On p.30 you quite right state that "What is seen in reality (i.e. an increase in erythemal UV dose accompanied with an increase in TOC and a decrease in AOD) is not always what is represented by the models". The significance of this sentence can hardly be overemphasized and should be brought into the abstract, too.

### **Changes to the manuscript:**

Added to the abstract: (After "...mean absolute error of only 6%":)

“However, the seasonal regression models do not always represent reality where an increase in erythemal UV dose is accompanied with an increase in TOC and a decrease in  $\tau_{\text{aer}}$ . In all seasonal models, solar radiation is the factor ...”

On p.30 the discussion on which of the three independent parameters shall be included in the regression model does not sufficiently underline the fact that the regression is valid for one site, and perhaps one period of time, only. Moving it to another place or time is probably less hazardous if all the three parameters are included.

### **Changes to the manuscript:**

Added at the end of the conclusion:

(After “Total ozone column however, does seem to be a more important factor in capturing the variation in erythemal UV dose and cannot be discarded from the regression models.”:)

**“It has to be kept in mind that the regression models are only valid for Uccle, which means that for other sites, it might be necessary to include all three parameters in the regression models.”**

A sentence has also been added at the end of section 4.4.2:

“The developed regression models are only valid for Uccle. For other sites, it might be necessary to include all three parameters in the regression models in order to explain the observed variation in erythemal UV dose.”

The language is probably fine but reading the text suffers from the excessive use of parentheses. Please, consider opening them as much as possible or just leaving out in case of self-evident or inessential information.

This comment has been taken into account and the majority of the parentheses have been opened.

The following suggestions are made:

On p.2, line 8, the words “(without any known instrumental cause)” is something we all expect as a default and need not be mentioned.

This is removed from the text.

As always, the text could be more compact. E.g. on p.3 it says "Including TOC however, is justified as the adjusted R2 increases and the MABE of the model decreases compared to a model where only global solar radiation is used as explanatory variable" while it could be put shorter :”Including TOC however, is justified to increase the adjusted R2 and to decrease the MABE of the model”.

This has been changed in the text.

On p.5, line 20, please, replace"for a long time period of 23 years" by "for a time period of 23 years".

This has been changed in the text.

On p.6. the first paragraph may not be needed in this detail. It could be sufficient to state "The cloud screening algorithm (De Bock et al., 2010) was improved by making use of the sunshine duration data and by assuming that the variability of the AOD..."

This comment has been taken into account and the paragraph has been shortened as follows:

“The initial cloud screening algorithm (as described in De Bock et al., 2010) did not perform well and it was clear that improvements were needed. The improved cloud screening method makes use of sunshine duration data...”

Moreover, in several places the use of two different symbols for one physical quantity may be confusing. To be logical you may want to use one symbol for each quantity and replace TOC by QO3, AOD by  $\tau_{aer}$ , etc. throughout the text.

This has been adjusted throughout the text.

The text is scientifically sound except for one mishap in the sentence on p.4, lines 12-14 saying “In principle, long term trends in UV irradiance can either be inferred from direct measurements (from ground or space) or reconstructed based on proxy data such as total ozone and sunshine duration”. While satellites cannot make any direct measurements on the surface of the Earth, you could revise the text e.g. by saying “Physically, UV trend can only be detected from direct measurements on Earth. Reconstructed data can be based on proxy data such as the abundance of ozone, solar irradiance, sunshine duration, or regional reflectivity of the earth-atmosphere system measured from the space.”

This has been changed in the text according to what the reviewer proposed.

On p.5: "Clouds induce more variability in surface UV irradiance than any other geophysical factor" is perhaps missing the words "...besides the solar elevation".

This has been changed in the text according to what the reviewer proposed.

Ch. 4.1. is utilizing the results given in Ch.4.2. Should the order of presenting the results be changed, i.e. the trends first and then the change point analysis?

The two paragraphs have been moved throughout the entire manuscript so that the trends are discussed before the change point analysis.

P.19, lines 22-23 say "...the stations with comparable latitude to Uccle (45–55N, stations in blue in Table 6), the trends in UV range from –2.1 to +8.6% per decade". Two comments: Firstly, the downloaded pdf copy does not show anything in blue, and secondly, Hoher Sonnblick at 47.05N suggests a trend of 14.2%/decade.

As it is clear from the manuscript which stations have comparable latitudes (by stating that we look at the stations between 45 and 55N), we decided not to present those stations in blue in table 6. We have removed this sentence from the manuscript. Also, the reviewer is correct, Hoher Sonnblick at 47.05N has a trend of 14.2%/decade.

The sentence referring to this in Ch. 4.2.1 has been adjusted:

“... the trends in UV range from –2.1 to +14.2% per decade.”

The list of references is impressive but you may want to add the following two:

P.4 line 17: Lindfors et al, 2007: A method for reconstruction of past UV radiation based on radiative transfer modeling: applied to four stations in northern Europe. J. Geophys. Res., Vol. 112, D23201.

P.4 line 23 and in Ch. 4.3.1.: Eleftheratos et al, 2014: Ozone and Spectroradiometric UV Changes in the Past 20 Years over High Latitudes, Atmosphere-Ocean, DOI: 10.1080/07055900.2014.919897

Thank you for the suggested references, they have both been added to the manuscript.

**The figures and tables are clear and the following two comments are given:**

Fig.2 and 3: the unit of the y-axis is missing.

The unit has been added to both figures.

Fig.4, 8, and 9: the axis labels and the scale could be larger for a more easy reading.

The figures have been adjusted to improve the readability.

**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 vs 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.