

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
AOD	92	5

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 τ_{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 τ_{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 Sg 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:

	1991-2013	1994-2013
Erythemal UV dose	6.91% (+/-1.54%)	7.20% (+/-1.83%)
Global radiation	4.29% (+/-1.31%)	4.36% (+/-1.64%)
Total Ozone Column	2.61% (+/-0.44%)	2.52% (+/-0.50%)
Aerosol Optical Depth (320nm)	-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 τ_{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 τ_{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.

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/m^2 , hence a dose rate. RAD has a unit $W h/m^2$ (please convert to J/m^2)

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/m^2 , hence a dose rate. RAD has a unit $W h/m^2$ (please convert to J/m^2)

There was an error in the unit of UV, which is actually expressed in J/m^2 . This error has been corrected throughout the manuscript. Global solar radiation values have been converted to J/m^2 .

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 erythematous 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 ($\ll 1 \mu\text{m}$) or coarse particles ($>1 \mu\text{m}$) 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 τ_{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 R² value as opposed to the normal R² 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^2 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_{O_3} 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 ...”):

“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_{O_3} and τ_{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 multiple 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 τ_{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 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:

‘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 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.

Relations between erythemal UV dose, global solar radiation, total ozone column and aerosol optical depth at Uccle, Belgium

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Abstract

At Uccle, a long time series (1991-2013) of simultaneous measurements of erythemal ultraviolet (UV) dose (S_{ery}), global solar radiation (S_g), total ozone column (TOC $_{Q_{O3}}$) and Aerosol Optical Depth (AOD $_{\tau_{aer}}$) (at 320.1nm) is available which allows for an extensive study of the changes in the variables over time. Linear trends were determined for the different monthly anomalies time series. S_{ery} , S_g and Q_{O3} all increase with respectively 7%, 4% and 3% per decade. τ_{aer} shows an insignificant negative trend of -8% per decade. These trends agree with results found in literature for sites with comparable latitudes. A change-point analysis, which determines whether there is a significant change in the mean of the time series, is applied to the monthly anomalies time series of the variables. Only for ~~erythemal UV dose and TOC~~ S_{ery} and Q_{O3} , a significant change point (~~without any known instrumental cause~~) was present in the time series around February 1998 and March 1998 respectively. The change point in ~~TOC~~ Q_{O3} corresponds with results found in literature, where the change in ozone levels (around 1997) is attributed to the recovery of ozone. ~~Linear trends were determined for the different (monthly anomalies) time series. Erythemal UV dose, global solar radiation and TOC all increase with respectively 7%, 4% and 3% per decade. AOD shows an (insignificant) negative trend of -8% per decade. These trends agree with results found in literature for sites with comparable latitudes.~~ A multiple linear regression (MLR) analysis is applied to the data in order to study the influence of ~~global solar radiation, TOC and AOD on the erythemal UV dose~~ S_g , Q_{O3} and τ_{aer} on S_{ery} . Together these parameters are able to explain 94% of the variation in ~~erythemal UV dose~~ S_{ery} . Most of the variation (56%) in ~~erythemal UV dose~~ S_{ery} is explained by ~~global solar radiation~~ S_g . The regression model performs well with a slight tendency to underestimate the measured ~~erythemal UV doses~~ S_{ery} values and with a Mean Absolute Bias Error (MABE) of 18%. However, in winter, negative ~~erythemal UV dose values~~ S_{ery} are modeled. Applying the MLR to the individual seasons solves this issue. The seasonal models have an adjusted R^2 value higher than 0.8 and the correlation between modeled and measured

erythemal UV dose S_{ery} values is higher than 0.9 for each season. The summer model gives the best performance, with an absolute mean error of only 6%. ~~Again, global solar radiation is~~ However, the seasonal regression models do not always represent reality where an increase in S_{ery} is accompanied with an increase in Q_{O3} and a decrease in T_{aer} . In all seasonal models, S_g is the factor that contributes the most to the variation in erythemal UV dose S_{ery} , so there is no doubt about the necessity to include this factor in the regression models. ~~A large part of the influence of AOD is already represented by the global solar radiation parameter. Therefore the~~ The individual contribution of AOD to erythemal UV dose is so low. For T_{aer} to S_{ery} is very low and for this reason, it seems unnecessary to include ~~AOD~~ T_{aer} in the MLR analysis. Including ~~TOC~~ Q_{O3} however, is justified as the adjusted R^2 increases and to increase the adjusted R^2 and to decrease the MABE of the model ~~decreases compared to a model where only global solar radiation is used as explanatory variable.~~

1 Introduction

The discovery of the Antarctic Ozone hole in the mid-1980s triggered an increased scientific interest in the state of stratospheric ozone levels on a global scale (Garane et al. 2006). The ozone depletion not only occurred above the Antarctic, but there is strong evidence that stratospheric ozone also diminished above mid-latitudes (Bartlett and Webb 2000; Kaurola et al. 2000; Smedley et al. 2012). While ozone depletion continued in the 2000s over the polar regions, it has leveled off at mid-latitudes, although ozone amounts still remain lower compared to the amounts in the 1970s (Garane et al. 2006). Stratospheric ozone is expected to recover in response to the ban on ozone depleting substances agreed by the Montréal Protocol in 1987 (WMO 2006; Fitzka et al. 2012). However, it is difficult to predict future changes in ozone as the predictions suffer from uncertainties caused by the general climate change, numerical errors of simulation models and by human behaviour which is not well controllable in several parts of the world. The decline in stratospheric ozone has shifted the focus of the scientific com-

munity and the general public towards the variability of surface UV irradiance (Krzýscin et al. 2011). If all other factors influencing UV irradiance remain stable, reductions in stratospheric ozone would lead to an increase in UV irradiance at the ground, particularly at wavelengths below 320 nm (Garane et al. 2006). Increases of UV irradiance in response to the ozone decline have already been reported for different sites during the 1990s (Garane et al. 2006 and references therein).

The possible increase in UV irradiance raises concern because of its adverse health and environmental effects. Overexposure can lead to the development of skin cancers, cataract, skin aging and the suppression of the immune system (Rieder et al. 2008; Cordero et al. 2009). UV irradiance also has adverse effects on terrestrial plants (Tevini and Teramura 1989; Cordero et al. 2009) and on other elements of the biosphere (Difey 1991). On the other hand, UV radiation does enable the production of vitamin D in the skin, which is positively linked to health effects as it supports bone health and may decrease the risk of several internal cancers (Bernhard 2011 United Nations Environment Programme 2009).

It is important to assess the changes in UV irradiance over prolonged periods of time. Not only do adverse health and environmental effects often relate to long term exposure (from years to a lifetime), also the time scales of the atmospheric processes that are involved (e.g., such as ozone depletion and recovery), are beyond decades (Chubarova 2008; den Outer et al. 2010).

~~In principle, long term trends in UV irradiance can either be inferred~~ Physically, UV trends can only be detected from direct measurements ~~(from ground or space) or reconstructed on Earth. Reconstructed data can be~~ based on proxy data such as ~~total ozone and sunshine duration~~ (Bernhard 2011 the abundance of ozone, solar irradiance, sunshine duration, or regional reflectivity of the earth-atmosphere system measured from the space (Lindfors et al. 2003)). Different sorts of reconstruction models have been used in several studies. They all use various kinds of statistical or model approaches and different meteorological or irradiance datasets (Lindfors et al. 2007 ; Chubarova 2008; Rieder et al. 2010; den Outer et al. 2010; Bais et al. 2011). Techniques are either based on modeling of clear sky UV irradiance or on empirical relationships be-

tween surface UV irradiance and the factors influencing the penetration of UV irradiance through the atmosphere (Kaurola et al. 2000; Trepte and Winkler 2004). In addition to the reconstruction studies, changes in surface UV irradiance have also been studied using ground based measurements at different locations (e.g. den Outer et al. 2000; Sasaki et al. 2002; Bernhard et al. 2006; Fitzka et al. 2012; Zerefos et al. 2012; [Eleftheratos et al. 2014](#)) or even in combination with satellite retrievals (Herman et al. 1996; Matthijsen et al. 2000; Kalliskota et al. 2000; Ziemke et al. 2000; Zerefos et al. 2001; Fioletov et al. 2004; Williams et al. 2004). Some studies combine both models and observations to investigate possible UV irradiance changes (e.g. Kaurola et al. 2000).

Not only stratospheric ozone influences the intensity of UV irradiance reaching the surface of the Earth. Long term changes in solar elevation, tropospheric ozone, clouds, Rayleigh scattering on air molecules, surface albedo, aerosols, absorption by trace gases and changes in the distance between the Sun and the Earth can lead to trends in UV irradiance (~~Bernhard 2011~~ [WMO 2006](#)). Some studies show that increased amounts of aerosols and trace gases from industrial emissions, which absorb UV irradiance in the troposphere, could even compensate for the UV effects caused by the stratospheric ozone decline (Krzýscin et al. 2011, Fitzka et al. 2012). Clouds induce more variability in surface UV irradiance than any other geophysical factor, [besides the solar elevation](#), but their effects depend very much on local conditions (Krzýscin et al. 2011). Surface albedo is determined mostly by snow amount and snow depth (Rieder et al. 2010) and plays a significant role at high altitude and high latitude sites, where UV irradiance can be strongly enhanced due to multiple occurrences of scattering and reflection between snow covered ground and the atmosphere (Fitzka et al. 2012). Several studies have been conducted to quantify the effects of the above mentioned variables on the amount of UV irradiance reaching the ground and many of them have done so by constructing empirical models with UV irradiance (or a related quantity) as a dependent variable (Díaz et al. 2000; Fioletov et al. 2001; de la Casinière et al. 2002; Foyo-Moreno et al. 2007; Antón et al. 2009; De Backer 2009; Huang et al. 2011; Krishna Prasad et al.

2011; El Shazly et al. 2012).

At Uccle, simultaneous measurements of erythemal UV dose, global solar radiation, total ozone column and ~~AOD~~ Aerosol Optical Depth at 320.1 nm are available for a ~~long~~ time period of 23 years (1991-2013). The time series is long enough to allow for reliable determination of significant changes (a minimum of 15 years is required as shown in Weatherhead et al. (1998) and Glandorf et al. (2005)). The availability of the simultaneous time series allows an extensive analysis in which three analysis techniques (~~change-point analysis~~, linear trend analysis, change-point analysis and multiple linear regression analysis) will be combined in order to increase our insights in the relations between the variables. ~~The monthly anomalies time series will be the subject of change-point analysis where the homogeneity of the time series will be investigated.~~ NextFirst, a linear trend analysis will be applied to the monthly anomalies of the time series (both on a daily and seasonal time scale) and the results will be compared with results found in literature. Monthly anomalies are used here 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. The monthly anomalies time series will also be the subject of change-point analysis where the homogeneity of the time series will be investigated. Finally, the multiple linear regression technique (with daily erythemal UV doses as dependent variable and daily values of global solar radiation, total ozone column and ~~AOD~~ Aerosol Optical Depth at 320.1 nm as explanatory variables) will allow us to study the influence of the explanatory variables on the dependent variable on a daily and seasonal basis.

2 Data

In this study, the (all sky) erythemal UV dose, (all sky) global solar radiation, total ozone column and (clear sky) ~~AOD~~ Aerosol Optical Depth at 320.1 nm are investigated over a time period of 23 years (1991-2013). These measurements are performed at Uccle,

Belgium (50°48' N, 4°21' E, 100 m asl), a residential suburb of Brussels located about 100 km from the North Sea shore.

2.1 Daily erythemal UV dose

5 In 1989, the Brewer spectrophotometer instrument#016(~~single monochromator~~), a single monochromator, was equipped with a UV-B monitor (De Backer 2009). This is an optical assembly which enables the Brewer to measure UV-B irradiance using a thin disc of Teflon as a transmitting diffuser (SCI TEC Brewer #016 manual 1988). The Brewer measures the horizontal spectral UV irradiance with a spectral resolution of approximately 0.55 nm, full width at half maximum. The instrument performs UV scans
10 from 290 to 325 nm with 0.5 nm wavelength steps (Fioletov et al. 2002). The erythemal irradiances are calculated using the erythemal action spectrum as determined by the Commission Internationale de l'Eclairage and are integrated to daily erythemal doses (De Backer 2009). For wavelenghts above 325 nm(~~), for which Brewer#016 does not provide data~~), the intensities are extrapolated using a theoretical spectrum weighted by the intensity at 325 nm. This is justified by the fact that, at those wavelenghts, the UV intensity is no longer strongly dependent on ozone and the erythemal weighting function is low. For the calculation of the daily sum, a linear interpolation between the different measurement points is performed. When there is an interruption of 2 hours or more
15 more between the measurements (~~between sunrise and sunset~~) ~~of 2 hours or more~~, the calculated daily sum is rejected. The data (in J/m^2) are available on a regular base since 1991. The instrument is calibrated with 50W lamps on a monthly basis and with 1000W lamps during intercomparisons in 1994, 2003, 2006, 2008, 2010 and 2012. The instrument was also compared with the travelling QUASUME unit in 2004 (Gröbner et al. 2004).
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2.2 Global solar radiation

The global solar radiation is a measure of the rate of total incoming solar energy (both direct and diffuse) on a horizontal plane at the surface of the Earth (Journée and Bertrand 2010). The measurements at Uccle are performed by CM11 pyranometers (Kipp&Zonen; <http://www.kippzonen.com>). For this study, the daily values in J/m^2 (derived from 10 minute and 30 minute data) are used. The data are quality controlled in two steps: first a preliminary fully automatic quality control is performed prior to the systematic manual check of the data (Journée and Bertrand 2010). In May 1996, we switched to a new system and in 2005 half of the instruments were replaced. Corrections to the measurements have been done in 2000, 2001, 2004, 2005, 2007 and 2012. (For the period before 1996, no information is available concerning possible calibrations of the instrument.)

2.3 Total ozone column

Total ozone column values (in DU) are available from Brewer#016 direct sun measurements. The instrument records raw photon counts of the photomultiplier at 5 wavelengths (306.3, 310.1, 313.5, 316.8 and 320.1 nm) using a blocking slit mask, which opens successively one of the five exit slits. The five exit slits are scanned twice within 1.6 seconds and this is repeated 20 times. The whole procedure is repeated five times for a total of about three minutes. The total ozone column is obtained from a combination of measurements at 310.1, 313.5, 316.8 and 320.1 nm, weighted with a predefined set of constants chosen to minimize the influence of SO_2 and linearly varying absorption features from e.g. clouds or aerosols (Gröbner and Meleti 2004). Brewer#016 was calibrated relative to the Dobson instrument in 1984 (De Backer and De Muer 1991) and regularly recalibrated against the travelling standard Brewer instrument #017 in 1994, 2003, 2006, 2008, 2010 and 2012. The stability is also continuously checked against the co-located instruments Dobson#40 (from 1991 until May 2009) and Brewer#178 (since 2001). [Internal lamp tests are performed on a regular basis to](#)

5 check whether the instrument itself is drifting. When instrumental drift is detected, it is corrected for.

2.4 Aerosol Optical Depth

Cheyamol and De Backer (2003) developed a method that enables the retrieval of **AOD** T_{aer} values (at 306.3, 310.1, 313.5, 316.8 and 320.1 nm) using the Direct Sun (DS) measurements of the Brewer instrument. It is also possible to retrieve **AOD** T_{aer} values at 340 nm using Sun Scan (SS) measurements of the Brewer instrument (De Bock et al. 2010). Together with the **AOD**-retrieval method, De Bock et al. (2010) developed a cloud screening procedure to select the clear sky **AOD** T_{aer} values. However, this screening method did not perform well. Hence an improved cloud screening method (described in section 3.1) has been developed and has been applied to **AOD** T_{aer} values retrieved from DS and SS measurements. For this study only the cloud screened **AOD** T_{aer} values at 320.1 nm, retrieved from the DS measurements of the single monochromator Brewer#016, will be used.

3 Method

3.1 Improved **AOD** Aerosol Optical Depth cloud screening method

The initial initial cloud screening algorithm, as described in De Bock et al. 2010, consisted of three steps. First, all **AOD** values larger than 2 were removed. Then, it was verified whether there was a DS observation within five minutes of each individual **AOD** measurement. Finally, the measured irradiances (photon counts) were plotted for days with **AOD** measurement(s) larger than 1.5. If the graph showed clear signs of cloud perturbation, the measurement was removed. The first two steps of this cloud screening were done automatically, whereas the last step had to be done manually. Analysis of the cloud screened data indicated that the performance of this screening technique was not optimal. Therefore it was decided to develop an De Bock et al. 2010, did not perform

well and improvements were needed. The improved cloud screening method ~~–This new cloud screening method~~ makes use of sunshine duration data (from 4 pyrheliometers at Uccle) and is also based on the assumption that the variability of the $AOD_{T_{aer}}$ in the course of one day is either lower than 10% or lower than 0.08 AOD -units(T_{aer} units, which is the maximum uncertainty of the ~~AOD retrieval algorithm~~) T_{aer} retrieval algorithm. Figure 1 gives a schematic overview of the improved cloud screening technique. First it is determined whether the individual $AOD_{T_{aer}}$ measurements were taken within a 10 minute interval of continuous sunshine. The measurements for which this is not the case are removed, after which more than 2 individual measurements per day must remain in order to continue. For each day, we then determine the maximum deviation to the median value. If this value is less than 0.08, we accept all measurements for that day. However, if the maximum deviation exceeds 0.08, the relative standard deviation for that day is calculated. In case this value is less than 10% (which would guarantee a given stability within the diurnal pattern of $AOD_{T_{aer}}$, all the $AOD_{T_{aer}}$ values for that day are accepted. In the other case, the $AOD_{T_{aer}}$ measurement with the largest contribution to the standard deviation is removed (as this measurement is most likely influenced by clouds). The median value will then be recomputed and the previous steps are repeated. Days with 2 or less individual $AOD_{T_{aer}}$ measurements are excluded from the results, since it does not make sense to calculate the deviation to the median and the standard deviation.

The cloud-screened $AODs$ (T_{aer} , both from DS and SS Brewer measurements) were compared to quasi-simultaneous and co-located Cimel level 2.0 quality assured values (with a maximum time difference of 3 minutes). The Cimel sunphotometer, which belongs to BISA (Belgium Institute of Space Aeronomy), is located at approximately 100 m from the Brewer instrument. It is an automatic sun-sky scanning filter radiometer allowing the measurements of the direct solar irradiance at wavelengths 340, 380, 440, 500, 670, 870, 940 and 1020 nm. These solar extinction measurements are used to compute aerosol optical depth at each wavelength except for the 940 nm channel, which is used to retrieve total atmospheric column precipitable water in centimeters.

The instrument is part of the AERONET network (<http://aeronet.gsfc.nasa.gov/>; Holben et al. 2001). The accuracy of the AERONET $AOD_{T_{aer}}$ measurements at 340 nm is 0.02 (Eck et al. 1999). For the period of comparison (2006-2013), the correlation coefficient, slope and intercept of the regression lines have been calculated and the values are presented in Table 1. The results of the comparison show that the cloud screened Brewer $AOD_{T_{aer}}$ values agree very well with the Cimel data.

The advantages of the improved cloud screening method are the removal of the arbitrary maximum level of $AOD_{T_{aer}}$ values and the fact that it runs completely automatic, whereas the old one needed manual verification afterwards). This method has now been applied not only to the $AOD_{T_{aer}}$ retrieval using SS measurements at 340 nm, but also to the method using DS measurements.

3.2 Data analysis methods

Since most statistical analysis tests, such as linear regression and change-point tests, rely on independent and identically distributed time series (e.g. Van Malderen and De Backer 2010 and references therein), most data used in this study are in their anomaly form. 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. Monthly means are only calculated for months with at least 10 individual daily values. For ~~the multiple linear regression analysis, daily values will be used instead of anomaly values.~~

3.2.1 Change-point analysis

~~Change points are times of discontinuity in a time series (Reeves et al. 2007) and can either arise naturally or as a result of errors or changes in instrumentation, recording practices, data transmission, processing, etc. (Lanzante 1996). A change point is said to occur at some point in the sequence if all the values up to and including it share a common statistical distribution and all those after the point share another. The most~~

common change-point problem involves a change in the mean of the time series (Lanzante 1998). There are different tests that can be used to detect a change point in a time series. In this study we use the combination of three tests: the non-parametric Pettitt-Mann-Whitney (PMW) test (based on the ranks of the values in the sequence), the Mann-Whitney-Wilcoxon (MWW) test (a rank-sum test) and the Cumulative Sum Technique (CST). The details of these tests are described in Hoppy and Kiely (1999). The change points discussed further in this study are detected by all three tests (except when mentioned otherwise) and only the change points that exceeded the 90% confidence level were retained. The change points are determined for the monthly anomalies time series of erythemal UV doses, global solar radiation, TOC and AOD at 320.1 nm S_{ery} , S_g and Q_{O3} , 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 τ_{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 τ_{aer} trend calculations outweighs this potential bias. For the multiple linear regression analysis, daily values will be used instead of anomaly values.

3.2.1 Linear trend analysis

Linear trends are calculated for the monthly anomalies of erythemal UV dose, global solar radiation, TOC and AOD S_{ery} , S_g , Q_{O3} and τ_{aer} at 320.1 nm. To determine the significance of the linear trends, the method described in Santer et al. (2000) is used. The least squares linear regression estimate of the trend in $x(t)$, b , minimizes the squared differences between $x(t)$ and the regression line $\hat{x}(t)$

5

$$\hat{x}(t) = a + b(t); t = 1, \dots, n_t \quad (1)$$

Whether a trend in $x(t)$ is significantly different from zero is tested by computing the ratio between the estimated trend (b) and its standard error (s_b)

$$t_b = \frac{b}{s_b} \quad (2)$$

10

Under the assumption that t_b is distributed as Student's t , the calculated t ratio is then compared with a critical t value, t_{crit} , for a stipulated significance level α and $n_t - 2$ degrees of freedom (Santer et al. (2000)).

15

However, if the regression residuals are autocorrelated, the results of the regression analysis will be too liberal and the original approach must be modified. The method proposed in Santer et al. (2000) involves the use of an effective sample size n_e in the computation of the adjusted standard error and calculated t -value, but also in the indexing of the critical t -value. To test for autocorrelation in the residuals of a time series, the Durbin-Watson test is used (Durbin and Watson 1971).

20

The above described linear trend analysis is also applied to the monthly anomalies of the extreme values (minima and maxima) of the variables. The extreme values are calculated by determining the lowest and highest measured value for each month. These trends will be studied together with the relative frequency distribution of the daily mean values. This distribution is determined by using the minimum and maximum values of the entire study period as boundaries and by dividing the range between the boundaries into a certain amount of bins of equal size. The daily values are distributed over the different bins and the relative frequency (in %) is calculated. This will be done for 2 different time periods: 1991-2002 and 2003-2013. Also, the medians for these periods are calculated. In this way, it is possible to investigate whether there is a shift in the frequency distribution of the variables from the first period to the second one. (The results of the analysis of the frequency distribution will only be presented in case they show a significant shift in the data).

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3.2.2 Change-point analysis

Change points are times of discontinuity in a time series (Reeves et al. 2007) and can either arise naturally or as a result of errors or changes in instrumentation, recording practices, data transmission, processing, etc. (Lanzante 1996). A change point is said to occur at some point in the sequence if all the values up to and including it share a common statistical distribution and all those after the point share another. The most common change-point problem involves a change in the mean of the time series (Lanzante 1996). There are different tests that can be used to detect a change point in a time series. In this study we use the combination of three tests: the non-parametric Pettitt-Mann-Whitney (PMW) test (based on the ranks of the values in the sequence), the Mann-Whitney-Wilcoxon (MWW) test (a rank sum test) and the Cumulative Sum Technique (CST). The details of these tests are described in Hoppy and Kiely (1999). The change points discussed further in this study are detected by all three tests (except when mentioned otherwise) and only the change points that exceeded the 90% confidence level were retained. The change points are determined for the monthly anomalies time series of S_{ery} , S_{ga} , QO_3 and τ_{aer} at 320.1 nm. 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.

3.2.3 Multiple linear regression analysis

The goal of a Multiple Linear Regression (MLR) analysis is to determine the values of parameters for a linear function that cause this function to best describe a set of provided observations (Krishna Prasad et al. 2011). In this study, the MLR technique is used to explore whether there is a significant relationship between the erythemal UV dose S_{ery} and three explanatory variables (global solar radiation, TOG and AOD S_{ga}).

Q_{O_3} and τ_{aer}) both on a daily and seasonal scale. We use a linear model where the coefficients are determined with the least squares method:

$$S_{ery} = a \times S_g + b \times Q_{O_3} + c \times \tau_{aer} + d + \epsilon \quad (3)$$

with

- S_{ery} : erythemal UV dose (in J/m²)
- S_g : global solar radiation (in J/m²)
- Q_{O_3} : total ozone column (in DU)
- τ_{aer} : Aerosol Optical Depth at 320.1 nm
- a, b, c: regression coefficients
- d: constant term
- ϵ : error term

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.

The model will be developed based on data from 1991 to 2008. ~~The data~~ 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

Discussion Paper | Discussion Paper | Discussion Paper | Discussion Paper | Discussion Paper

5 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.

The performance of the model and its parameters will be evaluated through different statistical parameters. The adjusted R^2 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. Compared to the R^2 value, the adjusted R^2 value will only increase if a new variable has additional explanatory power. It is possible to test the null hypothesis that a regression coefficient is equal to zero (which would mean that the variable associated with this regression coefficient does not contribute to explaining the variation in UV). This is done by looking at the p-value. If we want to test whether a regression coefficient differs significantly from zero at the 5% level, the p-value should be less than or equal to 0.05. The influence of the variation in the three parameters on the variation of S_{ery} is determined by multiplying the standard deviation of each parameter with its corresponding regression coefficient and dividing this by the average S_{ery} value.

20 The Mean Bias Error (MBE) and the Mean Absolute Bias Error (MABE) are also calculated in order to evaluate the performance of the regression model. The MBE (given in %) provides the mean relative difference between modeled and measured values (Antón et al. 2009):

$$MBE = 100 \times \frac{1}{N} \sum_{i=1}^N \frac{S_{ery_i}^{modeled} - S_{ery_i}^{measured}}{S_{ery_i}^{measured}} \quad (4)$$

5 The MABE (given in %) reports on the absolute value of the individual differences between modeled and measured data (Antón et al. 2009):

$$MABE = 100 \times \frac{1}{N} \sum_{i=1}^N \frac{|S_{ery_i}^{modeled} - S_{ery_i}^{measured}|}{S_{ery_i}^{measured}} \quad (5)$$

4 Results and discussion

4.1 Change-point analysis

4.0.1 Erythemal UV dose

According to the three tests (PMW, MWW and CST) of the change-point analysis, there is a significant shift in the mean of the monthly anomalies of erythemal UV dose which occurs around January 2003. The change point is located suspiciously close to the middle of the time series though. To remove the influence of the presence of one general increasing trend (which would lead to the discovery of a change point in the middle of the time series), the time series was detrended (= original time series – general trend). 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.

4.0.1 Global solar radiation

A significant change point was detected (only by the PMW test) around January 2003 in the time series of global solar radiation. Similar to the erythemal UV dose time series, there is one general trend present, which explains the detection of a change point near the middle of the time series. Thus, it was again decided to look at the detrended time series of global radiation. However, the detected change point around January 2006 (only by the PMW test) was not significant at the 90% significance level.

5 **4.0.1 Total ozone column**

All three tests confirmed the presence of a significant change point around March 1998 in the time series of monthly anomalies of TOC, where the mean before the change point is clearly lower than the one after the change point (Fig. 3). As there is clearly more than one general trend within the entire time series, there is no need for detrending in this case. No ozone calibrations were performed around 1998, so the change point has no known instrumental cause.

4.0.1 AOD at 320.1nm

According to the change point analysis, no significant change was found in the mean of the monthly anomalies of AOD.

15 **4.0.1 Overview and explanations**

The change points in the time series of erythemal UV dose and TOC occur around the same time period (February/March 1998). Since we were able to rule out known instrumental causes for the detected change points in both time series, we can assume that they have some natural/environmental cause and are related to each other. The change point in the TOC time series corresponds with results found in literature. Recent studies have shown that for other stations, the ozone recovery started around 1997 (Steinbrecht et al. 2006 ; Reinsel et al. 2005). Ozone levels seem to follow the change in chlorine concentrations resulting from the regulations of the Montreal Protocol in 1987. When ozone starts to increase, it is expected to have some implications on the UV irradiance as ozone is a strong absorber of UV irradiance in the stratosphere (Wenny et al. 2001). An increase in ozone would normally lead to a decrease in UV irradiance, which is not what was observed at Uccle where the UV irradiance levels continue to increase after 1998. Before 1998, the (insignificant) trends in the time series of TOC and erythemal UV dose are opposite, which is what would be expected.

5 However after 1998, both the (insignificant) TOC and erythemal UV dose trend are positive. So the behavior of TOC can only partly explain the changes observed in the UV irradiance time series and other parameters (such as aerosols and cloudiness) might play an important role.

4.1 Linear trend analysis

4.1.1 Erythemal UV dose

10 A significant positive trend (at the 99% significance level) can be detected in the time series of monthly anomalies of erythemal UV doses S_{ery} (Fig. 42). These values increase with 7% ($\pm 2\%$) per decade. The seasonal trends are presented in Table 2. In spring (March, April and May), summer (June, July and August) and autumn (September, October and November), the erythemal UV dose S_{ery} increases significantly, whereas in winter (December, January and February), the trend is negative. 15 The increase in erythemal UV dose S_{ery} is the largest in spring.

A significant positive trend has been found in the monthly anomalies of both the minimum and maximum values of erythemal UV dose S_{ery} . The minimum values show an increase of +10% ($\pm 4\%$) per decade and the maximum values increased by 7% ($\pm 1\%$) 20 per decade (respectively at the 95 and 99% level). The increase in the median value from $825 J/m^2$ (1991-2002) to $987 J/m^2$ (2003-2013), shows that higher erythemal UV dose S_{ery} values are more frequent in the last period.

4.1.2 Global solar radiation

25 The values of the global solar radiation S_g show an increase of 4% ($\pm 1\%$) per decade at the 99% significance level, which corresponds to an absolute change of $+0.5 (\pm 0.2) W/m^2$ per year for the observed time period (Fig. 42). On a seasonal scale, spring and autumn exhibit a significant positive trend (Table 3). The seasonal trends of global solar radiation S_g , although not significant in summer and winter, have the same sign as the

seasonal ~~erythemal UV dose S_{ery}~~ trends. The trends of ~~global solar radiation S_g~~ are smaller than the ~~UV S_{ery}~~ trends, both on an annual and seasonal scale.

There is a clear difference between the trends of the monthly anomalies of minimum and maximum values of ~~global solar radiation S_g~~ . Both trends are positive, but the increase in the minimum values (+12% ($\pm 5\%$) per decade at 99% significance level) is much larger than the one in the maximum values (+3.2% ($\pm 0.7\%$) per decade at 99% significance level). Study of the median values reveals the presence of an increase from 7880 kJ/m^2 (1991-2002) to 8902 kJ/m^2 (2003-2013). As the global radiation data are all sky data, it is obvious that the minimum values are the ones that are influenced by clouds. If the minimum values increase in time, ~~this could mean that the cloud properties (such as cloud optical depth), i.e. their amount and/or water content, must have~~ changed over the past 23 years. ~~However, this is difficult to prove without direct information or measurements on cloud amount and/or properties.~~

4.1.3 Total ozone column

The monthly anomalies of ~~TOC Q_{O3}~~ show a positive trend of 2.6% ($\pm 0.4\%$) per decade (significant at 99%) (Fig. 42). Significant positive trends occur in spring and summer (Table 4), with the trend in spring being the largest one. As opposed to the seasonal trends of ~~erythemal UV dose and global radiation S_{ery} and S_g~~ , the ones for ~~TOC Q_{O3}~~ are positive for each season. We would expect an increase in ~~TOC Q_{O3}~~ over the past 23 years to be accompanied by a decrease in ~~erythemal UV dose S_{ery}~~ , which is not the case for the Uccle time series. This indicates that other variables might contribute to the change in ~~erythemal UV dose S_{ery}~~ and the contribution of ~~TOC Q_{O3}~~ might be washed out by the influence of these other variables.

Both the minimum and maximum ~~TOC Q_{O3}~~ values increased significantly (99% level) at the same rate (+3.0% ($\pm 0.6\%$) per decade for the minimum values and +3.1% ($\pm 0.6\%$) per decade for the maximum values) over the past 23 years. A clear shift can be seen in the frequency distribution (Fig. 53) of the daily ~~TOC Q_{O3}~~ values. During the second period (2003-2013), higher values are more frequent than during the previous

period (1991-2002), which is supported by the increase in median values from 319.3DU (1991-2002) to 327.9DU (2003-2013). The entire curve of the frequency distribution is shifted, which means that the minimum values of the distribution have also increased between the two decades. After a period with lower $TOG_{Q_{O_3}}$ values in the 1990s, 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.

4.1.4 AOD-Aerosol Optical Depth at 320.1 nm

While the overall trends of ~~erythemal UV dose, global solar radiation and $TOG_{S_{ery}}$, S_g and Q_{O_3}~~ are all positive, the $AOD_{T_{aer}}$ values at 320.1 nm show a negative trend of -8% ($\pm 5\%$) per decade. This trend however is not significant (Fig. 42). The seasonal trends (Table 5) show that the summer and autumn trends are significantly negative, with the largest trend being observed during autumn. Due to a lack of sufficient clear sky data, it was not possible to determine the winter trend for $AOD_{T_{aer}}$.

There are no significant changes in the minimum and maximum $AOD_{T_{aer}}$ values over the 1991-2013 period. From the relative frequency distribution of the daily $AOD_{T_{aer}}$ values (Fig. 64), it can be seen that the frequency of lower ~~AOD-values~~ ($AOD_{T_{aer}}$ values ($T_{aer} < 0.4$)) was higher during the second period (2003-2013). Also the frequency of high ~~AOD-values~~ ($AOD_{T_{aer}}$ values ($T_{aer} > 0.7$)) has decreased towards the second decade. This is in agreement with the overall decrease in $AOD_{T_{aer}}$ over the last 23 years. However, this is not obvious from the median values as they decreased only slightly from 0.38 (1991-2002) to 0.36 (2003-2013).

4.2 Comparison of Uccle trends with other stations

4.2.1 Erythemal UV dose

Long term UV trends for different locations around the world have been the subject of many research articles and it is worth checking the consistency of our results with these studies even though the time periods are never exactly the same as the one studied in this paper (1991-2013). Some trends (observed or modeled/reconstructed) found in literature are presented in Table 6. Looking at these trends, it can be seen that for the stations with comparable latitude to Uccle (45-55°N, ~~stations in blue in Table 6~~), the trends in UV range from -2.1% to +~~8.6~~14.2% per decade. The increase of 7% ($\pm 2\%$) per decade observed at Uccle 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 a more global scale, Zerefos et al. (2012) examined UV irradiance over selected sites in Canada, Europe and Japan between 1990 and 2011. The results (based on observations and modeling for all stations), showed an increase in UV irradiances of 3.7% ($\pm 0.5\%$) and 5.5% ($\pm 0.3\%$) per decade at respectively 305 and 325 nm. For Europe, only the trend at 325 nm (3.4% ($\pm 0.4\%$) per decade) was significant. The COST 726 action (Litynska et al. 2009; www.cost726.org) calculated trend values for European sites and saw a mean positive trend of 4.5% ($\pm 0.5\%$) per decade since 1980 (which was derived from reconstruction models, based on ~~TOG~~

Q₀₃ and measured total solar irradiance).

4.2.2 Global solar radiation

Concerning the global solar radiation, many publications agree on the existence of a solar dimming period between 1970 and 1985 and a subsequent solar brightening period (Norris and Wild 2007; Solomon et al. 2007; Makowski et al. 2009; Stjern et al. 2009; Wild et al. 2009; Sanchez-Lorenzo and Wild 2012). Different studies have calculated the trend in ~~global radiation~~ S_g after 1985. The trend in ~~global radiation~~ S_g from GEBA (Global Energy Balance Archive; http://www.iac.ethz.ch/groups/schaer/research/rad_and_hydro_cycle_global/geba) between 1987 and 2002 is equal to $+1.4 (\pm 3.4) W/m^2$ per decade according to Norris and Wild (2007). Stjern et al. (2009) found a total change in the mean surface solar radiation trend over 11 stations in Northern Europe of $+4.4\%$ between 1983 and 2003. In the fourth assessment report of the IPCC (Solomon et al. 2007), 421 sites were analyzed and between 1992 and 2002, the change of all sky surface solar radiation was equal to $0.66 W/m^2$ per year. Wild et al. (2009) investigated the global solar radiation from 133 stations (from GEBA/World Radiation Data Centre) belonging to different regions in Europe. All series showed an increase over the entire period, with a pronounced upward tendency since 2000. For the Benelux region, the linear change between 1985 and 2005 is equal to $+0.42 W/m^2$ per year, compared to the Pan-European average trend of $+0.33 W/m^2$ per year (or $+0.24 W/m^2$ if the anomaly of the 2003 heat wave is excluded) (Wild et al. 2009). Our trend at Uccle of $+0.5 (\pm 0.2) W/m^2$ per year (or $+4\%$ per decade) agrees within the error bars with the results from Wild et al. (2009), but seems to be somewhat at the high end range.

4.2.3 Total ozone column

Ozone and its trends have been the subject of scientific research since the discovery of ozone depletion. Many studies agree that ozone has decreased since 1980 to the mid 1990s as a consequence of anthropogenic emissions of Ozone Depletion Substances (ODS). This period of decrease is followed by a period of significant increase (Stein-

brecht et al. 2006; Harris et al. 2008; Vigouroux et al. 2008; Krzýscin and Borkowski 2008; Herman 2010; Bais et al. 2011). For the period before the mid 1990s, studies report on decreasing ozone values at Brussels (Bojkov et al. 1995 and Zerefos et al. 1997), Reading (Bartlett and Webb 2000), Lerwick (Smedley et al. 2012), Arosa (Bojkov et al. 1995 and Staehelin et al. 1998), Hohenpeissenberg (Bojkov et al. 1995), Sodankylä (Glandorf et al. 2005) and Thessaloniki (Glandorf et al. 2005) (see Table 7).

5 After the mid-1990s, most studies report on a plateau or a limited increase in ozone. For example, Smedley et al. 2012, found no clear ozone trend in the 1993-2008 period for Reading. Ozone observations from a Brewer instrument at Hoher Sonnblick (by-Fitzka et al. 2012), showed a small but significant increase between 1997 and 2011. Similar behavior was reported for Jungfraujoch in Vigouroux et al. 2008. Our result (a trend of +2.6% per decade) compares well with the trend observed at Hoher Sonnblick, which is the only station with a time period comparable to the one at Uccle). From Fig. 3. From Figs. 2 and 6, it can be seen that a negative trend occurred in the TOC-QO3 values before 1998 and that this trend was followed by an a positive one. However, both trends are not significant at Uccle. It is difficult to unambiguously attribute the ozone trends to changes in ODS because other factors also contribute to ozone variability and trends. These factors are large volcanic eruptions, arctic ozone depletion, long term climate variability, changes in the stratospheric circulation and the eleven year solar cycle (Harris et al. 2008; Vigouroux et al. 2008). According to Rieder et al. 2013, the Equivalent Effective Stratospheric Chlorine and the 11-year solar cycle can be identified as major contributors, but the influence of dynamical features (such as the El Niño Southern Oscillation, North Atlantic Oscillation and Quasi-Biennial Oscillation) on the ozone variability and trends can not be neglected at a regional level.

4.2.4 AOD-Aerosol Optical Depth at 320.1 nm

Trend analysis studies of long time series of AOD-Aerosol Optical Depth are still very scarce at the moment. Some studies however do report on aerosol trends (Table 8). Mishchenko and Geogdzhayev (2007) observed a significant decrease in AOD-Taer

from 1991 to 2005 over much of Europe within the GACP (Global Aerosol Climatology Project; <http://gacp.giss.nasa.gov/>) data. Alpert et al. (2012) studied $AOD_{T_{aer}}$ trends from MODIS (MODerate resolution Imaging Spectroradiometer) and MISR (Multi-angle Imaging SpectroRadiometer) satellite measurements over the 189 largest cities in the world and saw a decrease in $AOD_{T_{aer}}$ over Europe for the 2002-2010 period. The decadal trend observed by de Meij et al. (2012) over Europe between 2000 and 2009 was negative for MODIS (-30%), MISR (-9%) and AERONET (-25%). Zerefos et al. (2012), who investigated the $AOD_{T_{aer}}$ over Europe, Japan and Canada, discovered a general decline in $AOD_{T_{aer}}$ exceeding 10% per year. For Europe specifically, the trend of $AOD_{T_{aer}}$ varied between -16.6% ($\pm 6\%$) per decade when using the GACP dataset and -42.8% ($\pm 5.7\%$) for the MODIS dataset. The ~~(insignificant) trend observed at Uccle~~ (insignificant trend of -8% $\pm 5\%$ per decade) observed at Uccle, lies within the range of trends observed at other European stations. The long term $AOD_{T_{aer}}$ decrease over much of Europe is quite consistent with the supposed reversal from increasing to decreasing anthropogenic sulfur and black carbon emissions owing to the enactment of clean air legislation in many countries (Mishchenko and Geogdzhayev 2007; Chiaccio et al. 2011; Alpert et al. 2012; de Meij et al. 2012; Hsu et al. 2012; Nabat et al. 2013). This change occurred after 1988-1989, the time period when a maximum was reached in the emissions of sulfate aerosols over Europe (Chiaccio et al. (2011)). Many scientists believe that the decadal changes in aerosols have influenced the amount of solar radiation reaching the surface of the Earth and that the decrease in aerosols has played a part in the switch from global dimming to global brightening, which occurred around 1980-1990 (Augustine et al. 2008; Chiaccio et al. 2011). According to Wild et al. (2009), the reduction of aerosols may have played a role during the 1990s but not after 2000. Decreases in cloudiness or cloud albedo may have enabled the continuation of the increase in surface solar radiation over Europe beyond 2000, despite the stabilization of aerosol concentrations.

4.3 Change-point analysis

4.3.1 Erythemal UV dose

According to the three tests (PMW, MWW and CST) of the change-point analysis, there is a significant shift in the mean of the monthly anomalies of S_{ery} which occurs around January 2003. The change point is located suspiciously close to the middle of the time series though. To remove the influence of the presence of one general increasing trend, which would lead to the discovery of a change point in the middle of the time series, the time series was detrended. This is done by subtracting the general trend from the original time series. The change point in the detrended time series is located around February 1998 (Fig. 5). 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.

4.3.2 Global solar radiation

A significant change point was detected (only by the PMW test) around January 2003 in the time series of S_g . Similar to the S_{ery} time series, there is one general positive trend present, which explains the detection of a change point near the middle of the time series. Thus, it was again decided to look at the detrended time series of S_g . However, the detected change point around January 2006 (only by the PMW test) was not significant at the 90% significance level.

4.3.3 Total ozone column

All three tests confirmed the presence of a significant change point around March 1998 in the time series of monthly anomalies of Q_{O_3} , where the mean before the change point is clearly lower than the one after the change point (Fig. 6). As there is clearly more than one general trend within the entire time series, there is no need for

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

4.3.4 Aerosol Optical Depth at 320.1 nm

According to the change-point analysis, no significant change was found in the mean of the monthly anomalies of τ_{aer} .

4.3.5 Overview and explanations

The change points in the time series of S_{ery} and Q_{O3} occur around the same time period (February/March 1998). Since we were able to rule out known instrumental causes for the detected change points in both time series, we can assume that they have some natural/environmental cause and are related to each other.

The change point in the Q_{O3} time series corresponds with results found in literature. Recent studies have shown that for other stations, the ozone recovery started around 1997 (Steinbrecht et al. 2006 ; Reinsel et al. 2005). Ozone levels seem to follow the change in chlorine concentrations resulting from the regulations of the Montréal Protocol in 1987. When ozone starts to increase, it is expected to have some implications on the UV irradiance as ozone is a strong absorber of UV irradiance in the stratosphere (Wenny et al. 2001). An increase in ozone would normally lead to a decrease in UV irradiance, which is not what was observed at Uccle where the UV irradiance levels continue to increase after 1998. Before 1998, the (insignificant) trends in the time series of Q_{O3} and S_{ery} are opposite, which is what would be expected. However after 1998, both the (insignificant) Q_{O3} and S_{ery} trend are positive. So the behavior of Q_{O3} can only partly explain the changes observed in the UV irradiance time series and other parameters, such as aerosols and cloudiness, might play an important role.

4.4 Multiple linear regression analysis

25 Before applying the ~~Multiple~~-multiple linear regression (MLR) technique, it has to be verified that the explanatory variables (~~global solar radiation, TOG and AOD~~ S_g , Q_{O_3} and τ_{aer}) are independent variables. This is done by calculating the correlation coefficients between these parameters. The correlation coefficients between the three variables are low enough (< 0.25) to allow using these variables as independent explanatory variables for the multiple regression analysis. As opposed to the previous analysis methods, the MLR is applied to daily values (~~),~~ instead of monthly anomaly values. For ~~UV and global radiation~~ S_{ery} and S_g , the daily sums are used, whereas for ~~ozone and~~ Q_{O_3} and τ_{aer} , daily mean values are used.

4.4.1 MLR analysis of daily values using ~~OZON~~total ozone column, ~~RAD~~global solar radiation and ~~AOD~~Aerosol Optical Depth

The MLR analysis has been applied to 1246 simultaneous daily values of erythemal UV dose (S_{ery}), global solar radiation (S_g), total ozone (Q_{O_3}) and ~~AOD~~Aerosol Optical Depth (τ_{aer}) between 1991 and 2008. ~~(The amount of regression days was highly limited by the available AOD measurements.)~~ τ_{aer} measurements. The resulting regression equation is:

$$S_{ery} = 690 + 0.000169 \times S_g - 5.01 \times Q_{O_3} + 70.0 \times \tau_{aer} + \epsilon \quad (6)$$

15 (with S_{ery} in J/m^2 ; S_g in J/m^2 and Q_{O_3} in DU).

The adjusted R^2 value of the multiple regression is 0.94, which means that S_g , Q_{O_3} and τ_{aer} together explain 94% of the variation in daily S_{ery} . ~~When looking at the~~ The changes in S_{ery} caused by the variation of each of the three parameters ~~(can be calculated by multiplying the standard deviation of each parameter with its corresponding regression coefficient and dividing this by the average S_{ery} value).~~ From the results, it is clear that S_g ~~(whose variation leads to a change in S_{ery} of 56%)~~ has the biggest influence on S_{ery} , followed by Q_{O_3} (change in S_{ery} of -9%) and τ_{aer} (change in S_{ery} of

1%).

The data from 2009-2013 are used to validate the model (see Fig. 7). The regression equation between the modeled and measured erythemal- S_{ery} values ($f(x) = 0.93x + 113.45$ with x : measured values) and the correlation coefficient (0.96) reveal the good agreement between model and reality. The Mean Bias Error (MBE) of the model is -3%, meaning that the model has a slight tendency to underestimate the measurements, which can be seen in Fig. 7 and 8. The Mean Absolute Bias Error (MABE), which is a useful measure to evaluate the overall performance of the model, equals 18%. This means that the model proposed here, estimates the S_{ery} with a mean error of 18%. Figure 7 and the upper panel of Fig. 8 show that in some cases, negative S_{ery} doses are modeled, which is a sign that the model does not always give realistic results. This is the case only during winter, when the S_g values are much lower than during the other seasons. When moderate to high Q_{O3} values are combined with low S_g values, this leads to negative modeled S_{ery} values according to the regression equation. From Fig. 8 it is also clear that there is a seasonal cycle in the residual values. Therefore, it would be better to perform the multiple regression analysis on a seasonal scale.

4.4.2 Seasonal MLR analysis using total ozone column, global solar radiation and Aerosol Optical Depth

The multiple regression equations for the different seasons are presented below:
Spring:

$$S_{ery} = 1016 + 0.0001542 \times S_g - 5.660 \times Q_{O3} + 92.11 \times \tau_{aer} + \epsilon \quad (7)$$

Summer:

$$S_{ery} = 2010 + 0.0001481 \times S_g - 6.737 \times Q_{O3} - 134.2 \times \tau_{aer} + \epsilon \underline{D} + \epsilon \quad (8)$$

Autumn:

$$S_{ery} = -195 + 0.000143 \times S_g - 1.22 \times Q_{O3} + 120 \times \tau_{aer} + \epsilon \underline{AD} + \epsilon \quad (9)$$

Winter:

$$S_{ery} = 325 + 0.0000750 \times S_g - 1.50 \times Q_{O3} + 101 \times \tau_{aer} + \epsilon \quad (10)$$

For all seasons, more than 80% of the total variation in S_{ery} is explained by the combination of S_g , Q_{O3} and τ_{aer} . This could be concluded from the adjusted R^2 values for each season. What might seem strange is the negative value of the constant term in the regression equation for autumn. However, the p-value for this term is higher than 0.05, which means that this coefficient does not significantly differ from zero at the 95% significance level.

From Fig. 9 and Table 9, it can be concluded that the seasonal models perform well in estimating the measured S_{ery} values. The correlation between the modeled and measured values varies between 0.90 (in winter) and 0.97 (in autumn). The regression equations are shown in both fig. 9 and Table 9. The negative MBE values (except for autumn which has a value close to 0) show that each model has a tendency to underestimate the measured values. The summer model performs best with an absolute mean model error of only 6%. The relative residuals (shown in Fig. 10) are smallest in summer, which again points out that the performance of the summer model in estimating the measured S_{ery} is the best. The spring and autumn models have much higher relative residuals.

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.

Changes in the variation of S_g (Table 10) are the most important and lead to changes in erythemal UV dose S_{ery} between 18% (in summer) and 53% (in autumn). The influence of the variation in Q_{O3} and τ_{aer} is much smaller. Changes in the variation of Q_{O3} always lead to negative changes in S_{ery} (from -2% in summer to -15% in winter), whereas the influence of a change in variation of τ_{aer} varies from a negative value (-1% change in S_{ery}) in summer to positive values in the other seasons, with a maximum of

4% in winter (Table 10). τ_{aer} and S_g have their lowest contribution in summer. Q_{O_3} on the other hand has the lowest contribution in autumn. The influence of Q_{O_3} is highest during winter and spring and this is in accordance with the variation in Q_{O_3} itself which is largest during winter and early spring. For τ_{aer} also, the absolute contribution to the variation in S_{ery} is the highest in winter. As the path length of UV irradiance is higher during winter, aerosols and ozone have more opportunity to influence UV irradiance on its way to the Earth's surface.

5 The influence of τ_{aer} on S_{ery} in the seasonal models is positive (except in summer) which is also the case when the τ_{aer} is used as the only explanatory variable in the models. This does not agree with what was observed in the trend analysis of the monthly anomalies time series, where an increase in erythemal UV dose S_{ery} is accompanied by a decrease in $AOD_{\tau_{aer}}$. It has to be taken into account however, that the negative
10 general $AOD_{\tau_{aer}}$ trend is not significant. Also, this negative trend in $AOD_{\tau_{aer}}$ is too much driven by the high, but sparse values at the beginning of the studied time period. Depending on the circumstances and the physical and optical properties of aerosols, the influence of $AOD_{\tau_{aer}}$ on global and UV irradiance can be either positive or negative. An increase in $AOD_{\tau_{aer}}$ could lead to an increase in global and UV radiation if
15 the increase in $AOD_{\tau_{aer}}$ was caused by an increase in the amount of small scattering aerosol particles. ~~These small particles would enhance the multiple scattering and reflection of UV irradiance, which in turn would increase the UV irradiance observed at the surface of the Earth~~ If there were predominantly particles of size much smaller than the UV wavelengths (i.e. freshly formed particles, Aitken mode particles) and of high single scattering albedo (SSA), the UV radiation could be enhanced by the multiple
20 scattering by these aerosols. However, when the amount of small all particles exceeded a certain (yet, herein in this study not possible to determine) threshold value, extinction would take over and from this point, an increase in $AOD_{\tau_{aer}}$ would lead to a decrease in UV irradiance. Both the composition of aerosols (which determines whether aerosol
25 composition, which determines if a mixture is absorbing or scattering) and the size of the particles rather scattering or absorbing, the aerosol amount, and the aerosol

size distribution determine whether an increase in $AOD_{\tau_{aer}}$ will lead to either an increase or a decrease in UV irradiance. At Uccle, ~~there is not sufficient information on both parameters~~ there is no information on these parameters, hence it is difficult to unambiguously characterize the influence of $AOD_{\tau_{aer}}$ 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 SSA. This will shine a new light on the influence of the aerosols on the UV radiation at Uccle. Antón et al. (2011) already reported that it is hard to determine the effect of aerosols due to their temporal and spatial variability and the difficulties associated with their characterization.

It has already been shown that S_g has the largest influence on S_{ery} , so an important issue that needs to be addressed is whether Q_{O3} and τ_{aer} are actually necessary to capture the variation in S_{ery} . This was investigated by performing the MLR analysis using (1) only S_g , (2) S_g combined with Q_{O3} and (3) S_g combined with τ_{aer} as explanatory variables. The adjusted R^2 value, the MABE and the correlation between modeled and measured S_{ery} values are given in Table 11. From these values, it becomes clear that τ_{aer} only has a minor contribution to the regression model and that to describe the changes in S_{ery} , τ_{aer} might not be needed, except perhaps for spring. ~~It has to be taken into account that τ_{aer} is known to have an influence on S_g , hence it's influence on S_{ery} is already partly represented by the factor S_g . So we could say that τ_{aer} contributes in an indirect way, through S_g , to the variation in S_{ery} . It's direct contribution to S_{ery} only is negligible.~~ For this reason it seems unnecessary to include τ_{aer} in the MLR analysis. Q_{O3} seems to be a more important explanatory variable, as the adjusted R^2 increases for all seasons (~~except summer~~), except summer, and the MABE of the models decreases (~~except in summer~~), when combining S_g and Q_{O3} . The correlation between modeled and measured values does not change much, except in winter (from 0.75 when using only S_g to 0.89 when combining S_g and Q_{O3}). 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 S_{ery} .

5 Conclusions

Of the variables known to influence the UV irradiance that reaches the ground, the variability of global solar radiation, total ozone column and Aerosol Optical Depth (at 320.1 nm) are studied by performing a change-point trend analysis, a trend change point analysis and a multiple linear regression analysis. This is done in order to determine their changes over a 23 year time period (1991-2013) and their possible relation with the observed UV changes at Uccle, Belgium. The erythemal UV dose, TOC and AOD_{S_{ery}, Q_{O3} and T_{aer}} are measured by the Brewer spectrophotometer instruments and the global solar radiation S_g measurements are performed by a CM11 pyranometer.

For TOC and erythemal UV dose, a significant change point (or a significant shift in the mean of the monthly anomalies) was detected around February/March 1998, which has no known instrumental cause. The timing of the change point in ozone corresponds to results found in literature where studies define the change around this time period as the start of ozone recovery, following the regulations of the Montreal Protocol. The trend over the past 23 years was determined for each variable using their monthly anomaly values. An overall positive trend was present in the time series of erythemal UV dose, global solar radiation and TOC S_{ery}, S_g and Q_{O3} of respectively +7% (±2%), +4% (±1%) and +2.6% (±0.4%) per decade. In contrast, the trend of AOD_{T_{aer}}, equal to -8% (±5%) per decade, is (insignificantly) insignificantly negative over the investigated time period. The sign and magnitude of the trends observed at Uccle agree with results found in literature for stations of comparable latitude. The increase in global solar radiation S_g since 1991 could be interpreted as a sign of continuing global brightening over Belgium. The decrease in sulfur and black carbon emissions after 1989, which resulted in enhanced global solar radiation at the Earth's surface, is most probably also the driving mechanism for the decrease in AOD_{T_{aer}}, which in turn could have

an influence by increasing the UV irradiance.

For both erythemal UV dose and global solar radiation S_{ery} and S_g , there is an increase in the frequency of higher values towards the second part of the study period (2003-2013), without the entire frequency distribution shifting. This could be explained by a decrease in cloudiness towards 2003-2013. Several studies report on a decrease in cloud cover over the past decades and a tendency for cumuloform clouds to replace stratiform clouds (Norris and Slingo 2009; Eastman and Warren 2013). This would increase both global solar radiation and UV irradiance S_g and S_{ery} due to enhanced scattering. However, other parameters (such as ozone and aerosols) could also influence the values of erythemal UV dose and global solar radiation S_{ery} and S_g . As opposed to erythemal UV dose and global solar radiation S_{ery} and S_g , a clear shift can be seen in the entire frequency distribution of daily TOG_{QO3} values, with both minimum and maximum values having increased from the 1991-2002 period to the 2003-2013 period, which supports literature findings about an ozone recovery around the end of the 1990s. From the frequency distribution of daily $AOD_{T_{aer}}$ values, it can be derived that between 1991 and 2002, higher $AODs_{T_{aer}}$ values were more frequently present than during the last period (2003-2013), which is in agreement with the overall decrease over the last 23 years.

The seasonal trends of the four variables were also studied and are similar between erythemal UV dose and global solar radiation S_{ery} and S_g , with a positive trend for all seasons except winter. The TOG_{QO3} trend is positive for spring and summer. Normally, we would expect a positive TOG_{QO3} trend to be accompanied with a negative trend in erythemal UV dose S_{ery} . The fact that the observed trends have the same sign, could indicate that the change in UV irradiance is not only influenced by a change in total ozone values. The $AOD_{T_{aer}}$ trend is negative during summer and autumn. The trend in spring is not significant and not enough winter data were present to calculate a winter trend.

For Q_{O_3} and S_{ery} , a significant change point (i.e. a significant shift in the mean of the monthly anomalies) was detected around February/March 1998, which has no known instrumental cause. The timing of the change point in ozone corresponds to results found in literature where studies define the change around this time period as the start of ozone recovery, 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.

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To investigate the influences of global solar radiation, TOG and AOD on the erythemal UV dose S_g , Q_{O_3} and τ_{aer} on S_{ery} , a multiple linear regression was performed using daily values between 1991 and 2008. The three variables together explain 94% of the total variation in the observed erythemal UV dose. Global solar radiation S_g values. S_g has the largest influence on the erythemal UV dose S_{ery} , followed by TOG and AOD Q_{O_3} and τ_{aer} . Data of 2009-2013 were used to validate the model and the MBA and MABE were calculated to evaluate the model performance in terms of overestimation and average error. The MBE value of the model is -3%, which means that the model has a slight tendency to underestimate the measured UV irradiance values. The average error of the model in the estimation of the measurements is equal to 18%. Overall, the model represents reality well, however sometimes (only during winter) negative erythemal UV dose during winter, negative S_{ery} values were modeled. For this reason, seasonal regression models have been developed.

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All seasonal models perform rather well in explaining the variation in UV irradiance (with adjusted R^2 values being larger than 0.8). The negative MBE values show the models' tendencies to underestimate UV irradiance. Again, global solar radiation S_g has the largest influence on erythemal UV dose S_{ery} , followed by TOG and AOD Q_{O_3} and τ_{aer} . The summer regression model performs best, based on the very low MABE values.

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What is seen in reality (i.e. an increase in erythemal UV dose S_{ery} accompanied with an increase in TOG Q_{O_3} and a decrease in AOD τ_{aer}) is not always what is represented

by the models. According to the regression models, ~~TOC and AOD~~ Q_{O3} and T_{aer} respectively always have a negative and positive influence on ~~erythemal UV dose~~ S_{ery} . However, as ~~global solar radiation~~ S_g is obviously the most important factor in explaining the variation in ~~erythemal UV dose~~ S_{ery} , the increase in ~~TOC~~ Q_{O3} (which would be expected to lead to a decrease in ~~erythemal UV dose~~ S_{ery}) and the change in ~~AOD~~ T_{aer} seem to be compensated for by the increase in ~~global radiation~~ S_g .

The question that remains is whether ~~TOC and AOD~~ Q_{O3} and T_{aer} are needed as explanatory variables in the multiple linear regression models. It has been shown that the contribution of ~~AOD~~ T_{aer} to explaining the variation in ~~erythemal UV dose~~ S_{ery} is very small and it can be concluded that this variable is not really needed in the multiple linear regression model. Also its influence is already partly represented by ~~the global radiation parameter~~. ~~Total ozone column~~ S_g , Q_{O3} however, does seem to be a more important factor in capturing the variation in ~~erythemal UV dose~~ S_{ery} 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.

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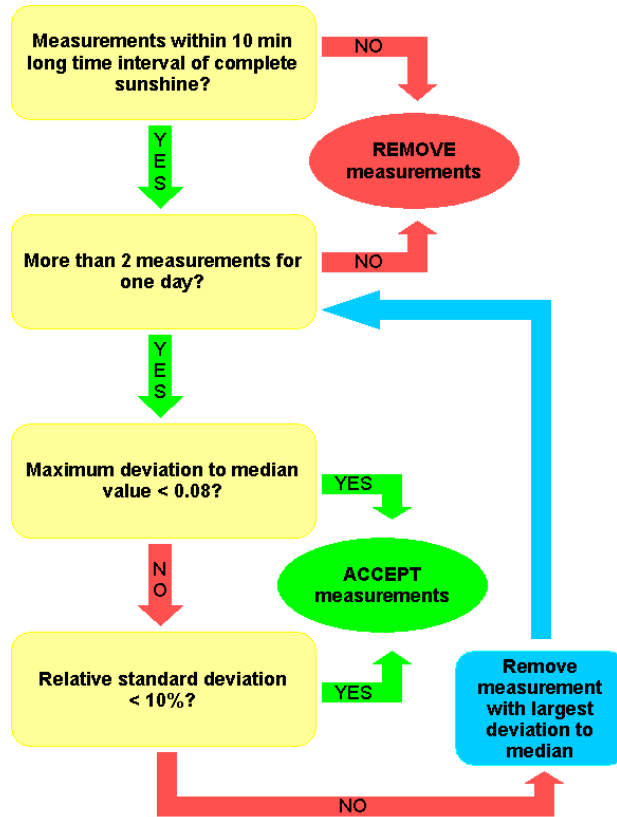


Fig. 1. Improved cloud screening procedure.

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 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 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.

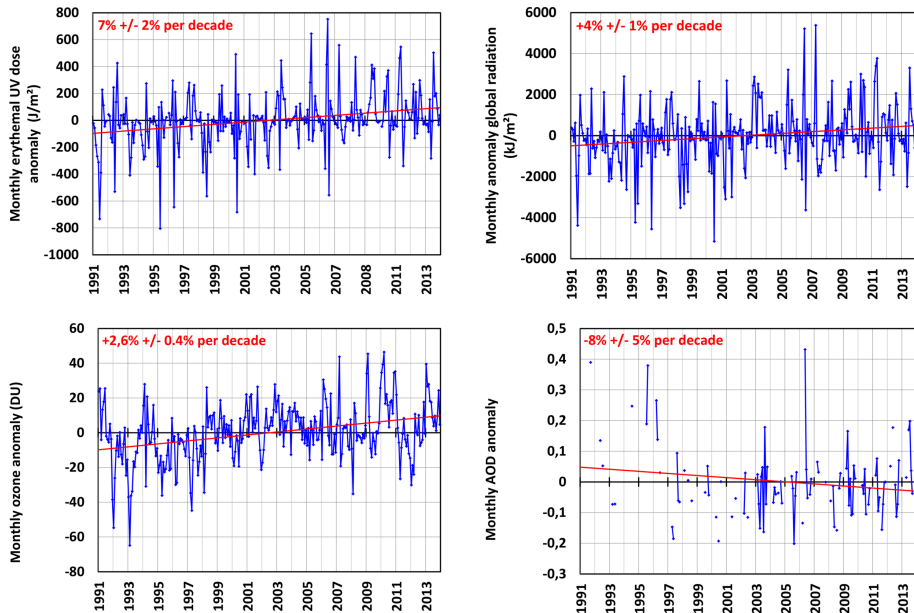


Fig. 2. Trends of monthly anomalies at Uccle for erythemal UV dose (upper left panel), global solar radiation (upper right panel), total ozone column (lower left panel) and [AOD](#) [Aerosol Optical Depth](#) at 320.1 nm (lower right panel) for the time period 1991-2013. The blue lines represent the time series, whereas the red lines represent the trend over the time period.

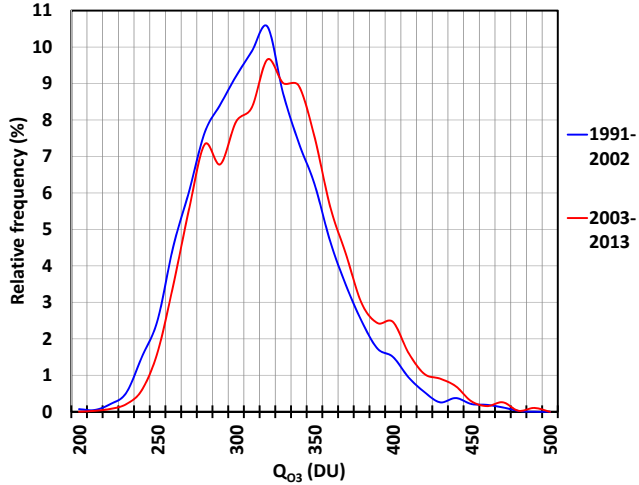


Fig. 3. Relative frequency distribution of daily ~~TOC~~ total ozone column values for the two time periods: 1991-2002 (in blue) and 2003-2013 (in red).

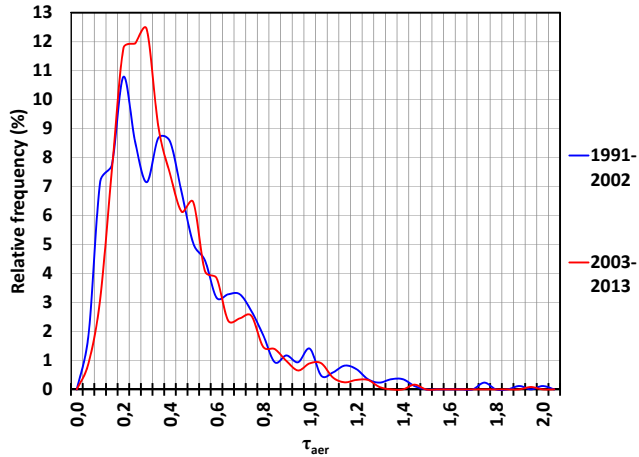


Fig. 4. Relative frequency distribution of daily [AOD-Aerosol Optical Depth](#) values for the two time periods: 1991-2002 (in blue) and 2003-2013 (in red).

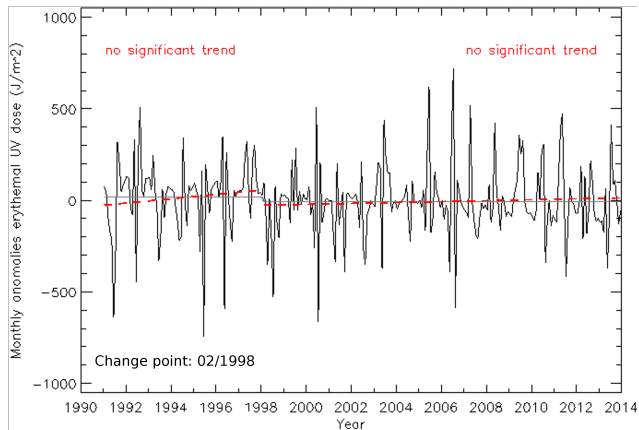


Fig. 5. The black line represents the detrended time series of monthly anomalies of erythemal UV dose (1991-2013). The red (dashed) lines represent the (insignificant) positive trends before and after the detected change point. The grey lines represent the mean before and after the change point.

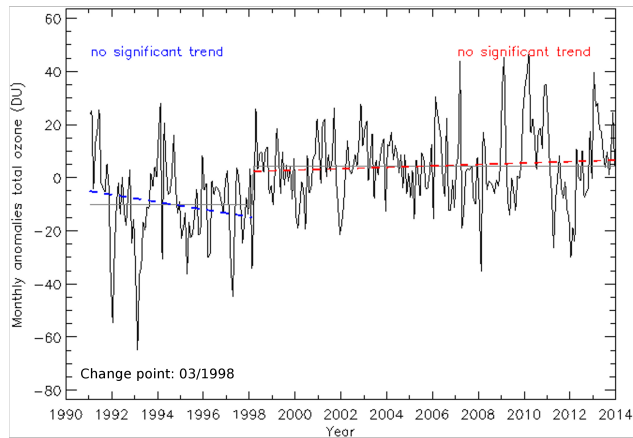


Fig. 6. The black line represents the time series of monthly anomalies of total ozone column (1991-2013). The blue (dashed) line represents the (insignificant) negative trend before the detected change point and the red (dashed) line represents the (insignificant) positive trend after the change point. The grey lines represent the mean before and after the change point.

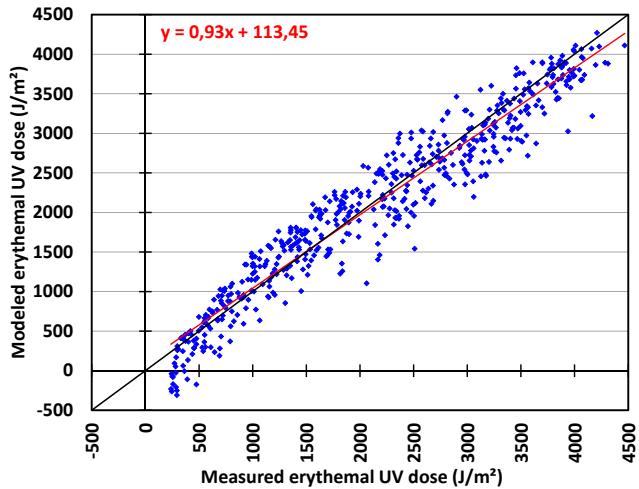


Fig. 7. Scatterplot of the measured and modeled erythemal UV doses at Uccle for the 2009-2013 validation period. The red line represents the regression line of the data ($f(x)=0.93x+113.45$). The black line is the $f(x)=x$ line.

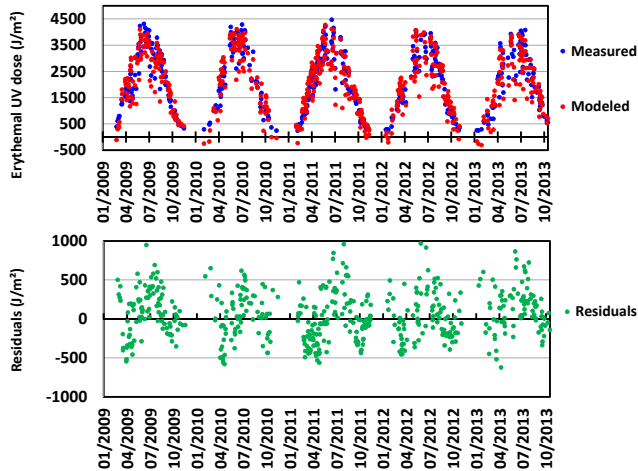


Fig. 8. Validation of the multiple linear regression equation: the upper panel shows the measured (in blue) and modeled (in red) erythemal UV values; the lower panel presents the absolute residuals.

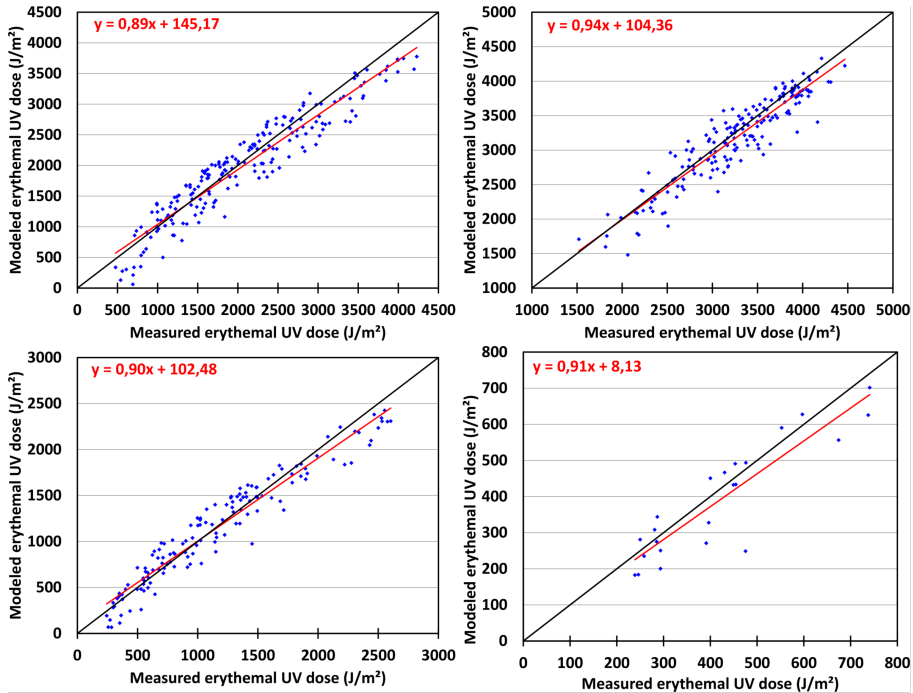


Fig. 9. Scatterplots of the measured and modeled erythemal UV doses at Uccle for the 2009–2013 validation period for spring (upper left panel), summer (upper right panel), autumn (lower left panel) and winter (lower right panel). The red lines represent the regression lines of the data and the black lines are the $f(x)=x$ lines.

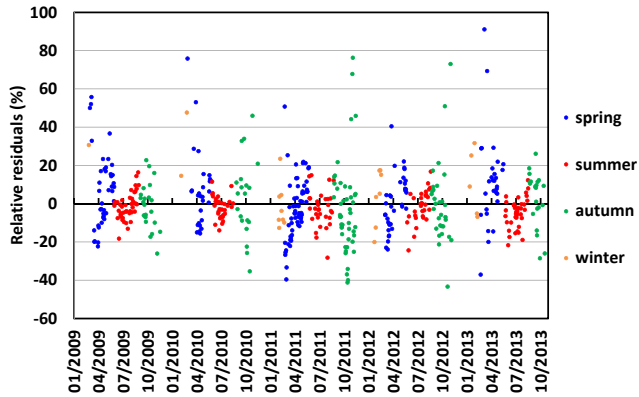


Fig. 10. Relative residuals $(=(\text{measured}-\text{modeled})/\text{measured}\cdot 100)$ of the seasonal multiple regression models. The colors represent the different seasons: blue=spring; red=summer; green=autumn; orange=winter.

Table 1. Comparison of Brewer and Cimel ~~AOD~~ Aerosol Optical Depth values (2006-2013)

		Correlation	Slope	Intercept
DS 320 nm	Brewer#016	0.97	1.004 ± 0.006	-0.067 ± 0.003
DS 320 nm	Brewer#178	0.99	1.007 ± 0.005	0.017 ± 0.002
SS 340 nm	Brewer#178	0.98	0.993 ± 0.007	0.073 ± 0.002

Table 2. Seasonal trends of erythemal UV doses (1991-2013)

Season	Trend per decade	Significance	Significance level
Spring	+9%(±3%)		99%
Summer	+6%(±2%)		99%
Autumn	+7%(±3%)		95%
Winter	-12%(±4%)		99%

Table 3. Seasonal trends of global solar radiation (1991-2013)

Season	Trend per decade	Significance level
Spring	+6%(±3%)	95%
Summer	+2%(±2%)	not significant
Autumn	+6%(±3%)	95%
Winter	-4%(±4%)	not significant

Table 4. Seasonal trends of total ozone column (1991-2013)

Season	Trend per decade	Significance level
Spring	+3%(±1%)	95%
Summer	+1.6%(±0.6%)	95%
Autumn	+1.8%(±0.9%)	not significant
Winter	+3%(±2%)	not significant

Table 5. Seasonal trends of ~~AOD~~ Aerosol Optical Depth at 320.1 nm(1991-2013)

Season	Trend per decade	Significance <u>Significance</u> level
Spring	+2%(±7%)	not significant
Summer	-18%(±8%)	95%
Autumn	-36%(±14%)	95%
Winter	Not enough data	

Table 6. Trends of UV radiation at different stations from (a): Bais et al. (2007), (b): Krzýscin et al. (2011), (c): Smedley et al. (2012), (d): Fitzka et al. (2012), (e): den Outer et al. (2010) and (f): Chubarova (2008).

Station, Country	Latitude/Longitude	Period	Trend/decade	Reference
Measured UV trends				
Sodankylä, Finland	67.42° N / 26.59° E	1990-2004	+2.1% (60° SZA)	(a)
Jokioinen, Finland	60.80° N / 23.49° E	1996-2005	-1.9% (60° SZA)	(a)
Norrköping, Sweden	58.36° N / 16.12° E	1996-2004	+12% (60° SZA)	(a)
Bilthoven, The Netherlands	52.13° N / 5.20° E	1996-2004	+8.6% (60° SZA)	(a)
Belsk, Poland	51.83° N / 20.81° E	1976-2008	+5.6%	(b)
Reading, United Kingdom	51.45° N / 0.98° W	1993-2008	+6.6%	(c)
Hradec Kralove, Czech Rep.	50.21° N / 15.82° E	1994-2005	-2.1% (60° SZA)	(a)
Lindenberg, Germany	47.60° N / 9.89° E	1996-2003	+7.7% (60° SZA)	(a)
Hoher Sonnblick, Austria	47.05° N / 12.96° E	1997-2011	+14.2% (65° SZA)	(d)
Thessaloniki, Greece	40.63° N / 22.95° E	1990-2004	+3.4% (60° SZA)	(a)
Reconstructed or Modeled UV trends				
Sodankylä, Finland	67.42° N / 26.59° E	1980-2006	+3.6%	(e)
Jokioinen, Finland	60.80° N / 23.49° E	1980-2006	+2.8%	(e)
Norrköping, Sweden	58.36° N / 16.12° E	1980-2006	+4.1%	(e)
Moscow, Russia	55.75° N / 37.62° E	1980-2006	+6%	(f)
Bilthoven, The Netherlands	52.13° N / 5.20° E	1980-2006	+2.9%	(e)
Hradec Kralove, Czech Rep.	50.21° N / 15.82° E	1980-2006	+5.2%	(e)
Lindenberg, Germany	47.60° N / 9.89° E	1980-2006	+5.8%	(e)
Thessaloniki, Greece	40.63° N / 22.95° E	1980-2006	+4.4%	(e)

Table 7. Trends of total ozone [column](#) at different stations from (a): Glandorf et al. (2005), (b): Smedley et al. (2012), (c): Bartlett and Webb (2000), (d): Bojkov et al. (1995), (e): Zerefos et al. (1997), (f): Fitzka et al. (2012), (g): Staehelin et al. (1998) and (h): Vigouroux et al. (2008).

Station, Country	Latitude/Longitude	Period	Trend/decade	Reference
Sodankylä, Finland	67.42° N / 26.59° E	1979-1998	-5.7%	(a)
Lerwick, United Kingdom	60.15° N / 1.15° W	1979-1993	-5.8%	(b)
Reading, United Kingdom	51.45° N / 0.98° W	1993-1997	-5.9%	(c)
Brussels, Belgium	50.84° N / 4.36° E	1971-1994	-2.6%	(d)
Brussels, Belgium	idem	1993-1996	-15.0%	(e)
Hradec Kralove, Czech Rep.	50.21° N / 15.82° E	1994-2005	-2.2%	(d)
Hohenpeisenberg, Germany	47.80° N / 11.00° E	1968-1994	-3.5%	(d)
Hoher Sonnblick, Austria	47.05° N / 12.96° E	1997-2011	+1.9%	(f)
Arosa, Switzerland	46.77° N / 9.67° E	1964-1994	-2.7%	(d)
Arosa, Switzerland	idem	1970-1996	-2.3%	(g)
Jungfrauoch, Switzerland	46.55° N / 7.98° E	1995-2004	+4.1%	(h)
Thessaloniki, Greece	40.63° N / 22.95° E	1993-1996	-4.0%	(e)
Thessaloniki, Greece	idem	1990-1998	-4.5%	(a)

Table 8. Absolute and relative trends of [AOD-Aerosol Optical Depth](#) at different stations from (a): Alpert et al. (2012), (b): Nyeki et al. (2012), (c): Fitzka et al. (2012), (d): Kazadzis et al. (2007). MODIS-Terra, MODIS-Aqua and MISR measurements are represented by respectively a '*', '#', and '+' after the station name.

Station, Country	Latitude/Longitude	Period	Trend/decade	Reference
Berlin (*), Germany	52.50° N / 13.40° E	2002-2010	-20.5%	(a)
Berlin (+), Germany	idem	2002-2010	-17.9%	(a)
Berlin (#), Germany	idem	2002-2010	-12.3%	(a)
Warsaw (*), Poland	52.30° N / 21.00° E	2002-2010	-2.4%	(a)
Warsaw (+), Poland	idem	2002-2010	-0.4%	(a)
Warsaw (#), Poland	idem	2002-2010	+12.9%	(a)
Ruhr Area (*), Germany	51.50° N / 7.50° E	2002-2010	-15.7%	(a)
Ruhr Area (+), Germany	idem	2002-2010	-9.3%	(a)
Ruhr Area (#), Germany	idem	2002-2010	-9.3%	(a)
Paris (*), France	48.90° N / 2.40° E	2002-2010	-8.1%	(a)
Paris (+), France	idem	2002-2010	+5.0%	(a)
Paris (#), France	idem	2002-2010	+9.8%	(a)
Hohenpeisenberg, Germany	47.80° N / 11.00° E	1995-2010	-10.6%	(b)
Hoher Sonnblick, Austria	47.05° N / 12.96° E	1997-2011	-5% to -6%	(c)
Barcelona (*), Spain	41.40° N / 2.20° E	2002-2010	-8.8%	(a)
Barcelona (+), Spain	idem	2002-2010	+4.2%	(a)
Barcelona (#), Spain	idem	2002-2010	-2.3%	(a)
Thessaloniki, Greece	40.63° N / 22.95° E	1997-2006	-29.0%	(d)
Madrid (*), Spain	40.40° N / 3.70° W	2002-2010	-18.3%	(a)
Madrid (+), Spain	idem	2002-2010	-10.0%	(a)
Madrid (#), Spain	idem	2002-2010	-7.4%	(a)

Table 9. Performance of the seasonal regression models

	Spring	Summer	Autumn	Winter
Correlation	0.95	0.93	0.97	0.90
Regression equation	$y=0.89x+145.17$	$y=0.94x+104.36$	$y=0.90x+102.48$	$y=0.91x+8.13$
MBE	-4%	-2%	0.06%	-7%
MABE	14%	6%	15%	15%

Table 10. Seasonal changes in UV caused by changes in RAD seasonal influence of the variation of S_g , $OZON_{QO3}$ and $AOD_{T_{aer}}$ on S_{ery}

	Spring	Summer	Autumn	Winter
$AOD_{T_{aer}}$	1%	-1%	2%	4%
$OZON_{QO3}$	-9%	-4%	-2%	-15%
RAD_{S_g}	37%	18%	53%	32%

Table 11. Results of MLR analysis with only RAD_{S_g} , RAD_{S_g} combined with $\text{OZON}_{Q_{O3}}$ and RAD_{S_g} combined with $\text{AOD}_{T_{aer}}$ as explanatory variables

	RAD_{S_g}	$\text{RAD}_{S_g} + \text{OZON}_{Q_{O3}}$	$\text{RAD}_{S_g} + \text{AOD}_{T_{aer}}$
Adjusted R^2			
Spring	0.85	0.90	0.85
Summer	0.81	0.85	0.81
Autumn	0.95	0.95	0.95
Winter	0.65	0.81	0.65
MABE (in %)			
Spring	14.53	14.40	14.33
Summer	6.39	6.21	6.21
Autumn	15.45	15.25	14.89
Winter	22.20	14.25	21.47
Correlation modeled and measured UV values			
Spring	0.93	0.95	0.93
Summer	0.91	0.93	0.91
Autumn	0.96	0.96	0.96
Winter	0.75	0.89	0.76