1 **Relationship between erythema effective UV radiant exposure, total ozone and cloud**
2 **cover in southern England UK: 1991-2015**

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7 **Abstract**

8 The long-term trend over 25 years of solar ultraviolet radiation measurements at Chilton in the
9 south of England (51.6 N^o, 1.3 W^o) has been investigated. Changes in erythema effective UV
10 radiant exposure (H_{er}) have been studied in relation to total ozone and cloud cover by examining
11 variation in the monthly mean deviation data.

12 The statistical analyses of the results showed that the long-term variability of H_{er} can be best
13 characterised in two sub-periods. In the first period between 1991 and 2004, it has been found
14 that H_{er} measurements indicated a small significant increasing linear trend of 1.01% per year
15 (95% CI: 0.75%; 1.27%), while during the second period, between 2004 and 2015, H_{er} showed a
16 small significant decreasing linear trend of 1.35% per year (95% CI: -1.98%; -0.77%). Changes
17 in H_{er} in relation to the combined effect of total ozone and cloud cover in southern England have
18 been investigated. Both cloud cover and total ozone were found to have a highly statistically
19 significant influence on H_{er}. These data also exhibited that between 1991 and 2015, Radiation
20 Amplification Factor (RAF) relates for sunburn of human skin was -1.03 at constant levels of
21 cloud cover that is for every additional 1.0% increase in total ozone, H_{er} decrease by 1.03%.
22 Over the period 1991-2004, cloud cover has explained the largest variation in H_{er} (47%), whilst
23 total ozone has explained only 8% of the changes in H_{er}. For the second period 2004-2015, this
24 pattern is reversed with total ozone having a greater effect on H_{er} variation (33%) than cloud
25 cover (16%). When the data have been examined separately for each season, the largest
26 correlation between H_{er} and total ozone and cloud cover was found during spring for both sub-
27 periods.

28 This study provides robust evidence that both increasing trend for the first period (1991-2004)
29 and decreasing trend for the second period (2004-2015) in H_{er} occur at the same time as
30 increasing total ozone. However, increasing trend in H_{er} over the first period is more strongly
31 associated with the observed reduction in cloud cover, while there is no significant change in
32 cloud cover over the second period that H_{er} is decreasing. All these changes are small and
33 occur within a very variable signal.

34 **1   Introduction**

35 Ultraviolet radiation (UVR) is only a small portion of the radiation we receive from the sun, but
36 has become a topic of increasing concern because of the harmful health effects it can cause.
37 Stratospheric ozone is a naturally-occurring gas that filters the sun's ultraviolet (UV) radiation. It
38 absorbs most of the shorter wavelength UV-B radiation, whereas longer wavelength UV-A
39 radiation mostly passes through the ozone layer and reaches the ground (WMO 2014).
40 However, in the mid-1970s it was discovered that the release of man-made chlorine-containing

41 chemicals could cause stratospheric ozone depletion. In subsequent years temporary ozone
42 holes appeared over the Antarctic and to lesser extent in the Arctic (Farman et al., 1985). It was
43 also observed that stratospheric ozone depletion also extended over populated areas, particular
44 in spring when the ozone layer over Antarctica is dramatically thinned over Australia (Gies et al.,
45 2013). Since the late 1970s, the effects of ozone depletion on UVR have been the subject of a
46 large number of studies published in the literature. These studies have demonstrated that the
47 ozone level decreased up to the mid-1990s which resulted in an increase in the amount of UV
48 radiation reaching the Earth's surface (WHO 2006). Concern was raised that in the long-term
49 ozone depletion would result in significantly increased UVR which in turn may result in increased
50 incidences of skin cancers, particularly melanoma. An increase in UVR can also result in an
51 increase in sunburn, ocular pathologies, premature skin aging and a weakened immune system
52 (UNEP 2010; WHO 2006; AGNIR 2002; Norval et al., 2011, Lucas et al., 2010). However, it is
53 known that exposure to UVR can be beneficial to health by producing vitamin D, which promotes
54 healthy bones and may help in the prevention of certain diseases including heart diseases and
55 cancers (Holick 2007, McKenzie et al., 2009, Young 2009, Epplin & Thomas 2010).

56 The Montreal Protocol came into effect in 1989, banning multiple substances responsible for
57 ozone depletion, and by the mid-2000s it had become apparent that a decline in total ozone had
58 stopped at almost all non-polar latitudes since around 1997 (WMO, 2007). However, the pace of
59 the recovery is affected by changes in temperatures, circulation, and the nitrogen and hydrogen
60 ozone-loss cycles (Waugh et al., 2009). The ozone level has remained relatively unchanged
61 since 2000 with most studies reporting a plateau or a limited increase in total ozone (WMO
62 2014).

63 The most important factor affecting UVR at the earth's surface is the elevation of the sun in the
64 sky - this causes terrestrial UVR to vary with time of day, day of the year and with geographical
65 location (Diffey, 2002). Aside from solar elevation, the most significant factors affecting solar
66 UVR are likely to be stratospheric ozone and cloud cover; UVR may also be affected by a
67 number of other factors including aerosols and air pollutants; many of these factors are also
68 influenced by climate change (Bais et al., 2011). These factors often interact with each other in a
69 complex way and their effect on terrestrial UVR can be hard to quantify.

70 In 1990, due to the widespread concern that the depletion of the ozone layer would cause an
71 increase in population UVR exposure and possible effects on health, the former National
72 Radiological Protection Board (NRPB - now part of Public Health England, PHE) set up
73 monitoring stations at three locations in the UK to measure terrestrial solar UVR (HPA 2012).
74 The Chilton monitoring site is located in a rural area in the south-east of England at
75 approximately 51.6° N, 1.3° W. The analysis of annual erythema effective UV radiant exposure
76 H_{er} over Chilton in 1991-2015 revealed a statistically significant increasing linear trend between
77 1991 and 1995 and a small decreasing linear trend during 1995-2015 (Hooke et al., 2016; Hooke
78 et al., 2017). The analyses described in this paper are complementary to those undertaken by
79 Hooke et al., (2017), which use the same data but with methodological differences that are
80 discussed in the course of the paper. In particular, this work focuses on whether the long-term
81 trend of monthly H_{er} can be linked to changes in ozone depletion and cloud cover, the most
82 significant atmospheric factors that affect terrestrial UVR.

83 **2   Materials and methods**

84

85 **2.1   Erythema effective UV radiant exposure (H_{er})**

86 Details of the methodology for UVR monitoring at Chilton are presented elsewhere (Hooke et al.,
87 2017). A short description of materials and methods is given here, and additional analyses using
88 the same data are pointed out. Erythema effective UV irradiance in the wavelength range 280-
89 400 nm is measured by Robertson-Berger meters (RB-500 and RB-501 since 2004,
90 manufactured by Solar Light Co. Philadelphia, USA). Data from these sensors are sampled to
91 calculate 5 minute mean values that are recorded together with the standard deviation of these
92 readings for each 5 minute period. To convert to H_{er} per day, the erythema effective UVR
93 irradiance data were summed up daily from half an hour before sunrise to half an hour after
94 sunset under all weather conditions (Hooke et al., 2016). The units of H_{er} are defined as the
95 amount of energy (joules) deposited per square meter (J m^{-2}). The first full calendar year of
96 measurements at Chilton began in January 1991. The daily UVR data at this site considered
97 here are the measurements for all available days during the 25 year period from 1st January
98 1991 to 31st December 2015.

100 The broadband detectors measuring erythema effective UV radiation were calibrated annually
101 using a double-grating spectroradiometer. This spectroradiometer was calibrated and traceable
102 to national standards. The daily radiant exposure for 22 clear days during May–October between
103 2003 and 2015 was compared to the daily radiant exposure from the double-grating
104 spectroradiometer and the data from the broadband detectors was found to be within 10% of the
105 spectroradiometer data on all these days (Hooke, 2017).

107 **2.2   Total Ozone**

109 The ground-based instruments, Dobson Spectrophotometers, used to measure daily column
110 ozone were at the UK Meteorological (Met) Office observatories at Camborne in Cornwall (south
111 west of England, Latitude 50.2° N, 5.3° W) for the period 1979-2003. Ozone monitoring was also
112 undertaken at Reading using Brewer spectrophotometers from January 2003 onwards. These
113 instruments measure column ozone, i.e. total ozone, in which includes stratospheric ozone as
114 well as tropospheric ozone in the atmosphere. Total ozone is measured in Dobson Units (DU).

116 These two time series of data from the Camborne and Reading sites can be combined into a
117 single continuous total ozone time series (Smedley et al., 2012). Both sites are located at similar
118 latitude and the Reading site is closer to Chilton (30km to the south-east of Chilton). The
119 combined dataset is considered here as a surrogate for the total ozone data for Chilton over the
120 whole period 1991 to 2015. Data and other information from these sites were obtained from the
121 air quality website (UK-AIR) of the UK Department for Environment Food & Rural Affairs
122 (DEFRA). The details of the instrumentation, the ground-based ozone data and the trend
123 analysis of total ozone from these sites from 1979 to 2008 were published previously (Smedley
124 et al., 2012).

126 **2.3   Cloud cover**

128 The HadISD dataset was created by the Met Office at the Hadley Centre in the UK, which used a
129 sub-set of the station data held in the Integrated Surface Database (ISD) (Dunn et al., 2012 &
130 2014). The HadISD dataset comprises various selected climate variables, including total cloud
131 cover data that were recorded in various weather stations globally, including in the UK for 1931–
132 2016 (Dunn et al., 2016).

Station based cloud cover data in the HadISD dataset are available in various locations for the whole of the UK. The nearest point to the PHE building in Chilton for obtaining cloud cover data is presented here at Benson-Oxfordshire (Latitude 51.6° N, 1.10° W, 15km to the north-east of Chilton) and used as a surrogate value for Chilton. The cloud cover data were calculated hourly from this station's observations of total cloud amount in oktas (1 okta = cloud covering one eighth of the sky = 12.5%). The hourly time series of daily cloud cover values at Benson were obtained from the Centre for Environmental Data Analysis (CEDA) for the period between 1991 and 2015. The daily average cloud amount are used here are based on the recordings at this station from 11am to 2pm GMT.

2.3 Estimating trends

Linear regression analyses were carried out to test whether the estimated slopes in this particular sample of data suggest real long-term trends in the underlying H_{er}, total ozone or cloud cover data in the UK. However, in order to assess the long term trends in H_{er}, total ozone and cloud cover time series seasonal variations have been removed from the monthly mean data. This was done by calculating the overall average H_{er}, total ozone and cloud cover for each month and then subtracting each individual value from their associated average months over the 25 years. For each data set the deviation from averages in percentage of the seasonal corrected monthly mean data was estimated. In contrast, our previously reported analyses were based on annual mean anomaly data from the daily data (Hooke et al. 2017), while the analyses performed here are monthly mean deviation data from the monthly data. Although annual data and monthly means show similar pattern, we have decided using monthly data in order to examine the effects of total ozone and cloudiness changes on the H_{er}.

Longer term variations such as the Quasi-Biennial Oscillation (QBO) and the 11-year solar cycle have not been taken into account. Since the period of the QBO is approximately 2.3 years it affects short term variability rather than long term trends. This fluctuation is small in comparison to the 25 year timescale being analysed in this paper (Harris et al., 2008; Den Outer et al., 2005). The 11-year solar cycle has a longer period and therefore has the potential to impact long term trends, however its effect on erythema effective UV levels is small (den Outer, 2005; Diffey, 2002).

The trend analyses were performed from the regression analysis ofon the corrected monthly mean deviation of H_{er}, total ozone orand cloud cover data sets versus year and t-tests were used to determine whether the slopes of the fitted trend models were significantly different from zero. The shape of trend in the time series was also examined for H_{er}, total ozone and cloud cover by fitting linear and non-linear models to determine whether the observed values generally increase (or decrease) over time. Further analyses were also carried out by examining the changes in H_{er}, total ozone and cloud cover separately for each season (winter, spring, summer and autumn).

The evidence for autocorrelation in the residuals of the regression analysis was also tested using the Durbin-Watson (DW) statistic. It is a well-known method of testing if autocorrelation is a problem undermining the model's inferential suitability (*e.g.,* assessing the confidence in the predicted value of a dependent variable). The test compares the residual for time period *t* with the residual from time period *t-1* and develops a statistic that measures the significance of the correlation between these successive comparisons (Chatfield 1996). If there was evidence for autocorrelation, a non-parametric test (distribution-free), the Mann-Kendall test (MK) was used

183 in place of a parametric linear regression analysis, which can be used to correct temporal
184 autocorrelation and to test if the slope of the estimated linear regression line differs significantly
185 from zero (Mann 1945, Kendall 1975, Helsel and Hirsh 1992). If a significant trend was found
186 from the MK test, the rate of change was calculated using the Sen's slope (SS) estimator from
187 nonparametric method (Helsel and Hirsh 1992). If the results of non-parametric analyses were
188 similar to those results obtained by linear regression, the results from non-parametric analyses
189 are not presented.
190
191 The relationship between H_{er} and total ozone and cloud cover was also examined using Analysis
192 of Variance (ANOVA) to obtain information about levels of variability within a regression model
193 and to form a basis for tests of significance. The correlation coefficient value (r^2) was calculated
194 to determine a measure of the strength of the relationship between H_{er} and cloud cover and
195 ozone and to quantify how much of the total variation in H_{er} could be explained by ozone or cloud
196 cover. A significance level $p < 0.05$ was considered statistically significant.
197
198

3   Results

3.1   Erythema effective UV radiant exposure (H_{er})

201 Summary statistics from the daily H_{er} are presented in Table 1. Over 25 years H_{er} ranges from
202 10 J m^{-2} (measured on 9 January 1992, 18 days after winter solstice) to 5655 J m^{-2} (measured on
203 20 June 2003, at the summer solstice) with a mean of 1303 J m^{-2}.
204
205 Figure 1 displays the distribution of the daily H_{er} using boxplots for each season at Chilton. Each
206 box shows the lower 25% quartile Q1, and upper 75% quartile Q3 and central line is the median.
207 The whiskers extended in each direction from the box starts from Q1 to the smallest data point
208 and the upper whisker from the Q3 to the largest data point that is away from the box and
209 measurements falling outside whiskers are possible extreme data values that are outside the
210 typical pattern of the other data points. The observed results show that H_{er} are the highest in the
211 summer months and the lowest in winter months, while during spring and autumn months, H_{er}
212 may change rapidly day to day (Fig.1). The extreme data points were mostly observed in winter
213 and a few in other seasons could be related to natural variability in factors that affect H_{er}
214 including total ozone, cloud cover, aerosols, and climate at this site. In particular, extremely low
215 total ozone periods, which often occur in late winter and early spring, while H_{er} does not exceed
216 normal-ozone summer values. After 2007, in particular in spring, summer and autumn it appears
217 that H_{er} values are well below their expected mean values.
218
219 **Figure 1**: Boxplots of the daily H_{er} data for each season at the Chilton site between 1991 and
220 2015 (grey solid line represents the mean value for each season)
221
222 Figure 2a shows the corrected monthly mean deviation of H_{er} values expressed as percentages.
223 A consistent rise between 1991 and 2003 with a clear peak in 2003 when the H_{er} values were the
224 largest recorded at Chilton over the 25 year period. Thereafter, H_{er} values appeared to decrease.
225 Fig. 2b also shows the mean deviation data in H_{er} for each of the four seasons over the 25 year
226 period. Winter and spring exhibited greater variability in comparison with summer and autumn.
227 During winter months, peaks in H_{er} were observed in various years; however, H_{er} in winter was
228 very low (Table 1) thus the effect on annual exposure is expected to be very small. Among
229 spring months, clear peaks are observed in March 2003 and also in March & April 1997 (Fig. 2b).

230 For summer, H_{er} levels vary less in comparison to other seasons, although summer has the
231 highest H_{er} levels overall and the effect on annual exposure is large. For autumn, peaks are
232 observed in November 2006 and 2007 and the variability in the last few years was stable.
233
234
235 **Figure 2:** ~~Corrected m~~Monthly mean <u>deviation of</u> H_{er} data at Chilton (1991-2015) with trend lines
236 (a) all season combined (b) seasonal.
237
238 The regression analyses of H_{er} data indicate that the best fitting single linear trend covering the
239 whole period 1991-2015 has a downward slope but that this slope is not statistically significantly
240 different from a constant value over the period (p=0.27). Two further models were also
241 examined. One is a linear-quadratic function, (LQ), a 2nd degree polynomial, which allows for
242 more gradual variation in the monthly H_{er} across the 25 year period and a second model consists
243 of two linear trends with a node to allow for a single change in linear trend over the 25 year
244 period. Figure 2a also shows the results of fitting these four models to the ~~corrected~~ monthly
245 <u>mean deviation of</u> H_{er} data.
246
247 The best fitting model was the last of these which had two linear trends that describe an
248 increasing trend from 1991 to 2003 and a decreasing trend thereafter which defines the year
249 2003 as the node between two trend lines. The nodal year appeared to be influenced by the
250 particularly high observations in 2003 (Fig. 2a). Thus, in order to avoid bias that might be caused
251 by the highest H_{er} values observed in 2003, the year 2004 was chosen to be the nodal point in
252 preference to 2003.
253
254 Based on the results of the initial model fitting to the whole period statistical analyses were also
255 carried out to investigate the long-term variability of H_{er} for two sub-periods (1991-2004 and
256 2004-2015). Table 2 presents the estimated linear slopes in percentage change per year in H_{er}
257 with 95% confidence intervals (CI) and p-values of the associated significance tests. There is
258 evidence of a statistically significant increasing trend for the first period (1991- 2004) with a mean
259 rate of 1.01% per year (y^{-1}) (95% CI: 0.75%; 1.27%, p<0.001) and a decreasing trend for the
260 second period (2004-2015) with a mean rate of 1.35% y^{-1} (95% CI: 1.98%; 0.72%, p<0.001)
261 based on all the data. While there was evidence of autocorrelation, the results of the non-
262 parametric analyses, which would not be influenced in the same way by the autocorrelation, were
263 similar to those in Table 2 so they are not presented here.
264
265 For seasonal trends, the only significantly increasing linear trend was seen in winter from 1991-
266 2004; however, H_{er} level in winter was very low and contributed only a small proportion of the
267 total H_{er} overall. The highest levels of H_{er} observed in summer did not show any significant linear
268 trend for 1991-2004 as the H_{er} levels were steady for this period. The absence of a significant
269 trend in spring for this period might be partly explained by the influence of fairly stable H_{er} levels
270 seen between 1998 and 2002 (Fig.2b). Across the same period in autumn, the trend was found
271 to be approaching statistical significance (p=0.07). For 2004-2015, the estimated trend slope
272 was negative for each season, but the trend was only statistically significant in summer and
273 autumn, (Table 2).
274
275
276 ## 3.2   Total ozone

277 From the combined Camborne and Reading dataset covering the period 1991-2015 total ozone
278 ranged from a low of 177 DU (measured in January 2006 in Reading) to a high of 524 DU

326 (measured in February 1991 in Camborne) with an overall mean value of 327 DU. The
327 distribution of the daily total ozone values are presented in box plots for the period 1991-2015 for
328 each season (Fig. 3a). The mean value is shown with a grey dashed line and the bold line at
329 300 DU shows the average amount of total ozone in the atmosphere
330 (http://ozonewatch.gsfc.nasa.gov). Both graphs displayed a large spread of total ozone
331 measurements. The data appear to be varying year to year and there is a much larger spread of
332 total ozone values in winter and spring compared to summer and autumn. The extreme data
333 points are therefore not likely to be erroneous readings. The total ozone values were low in
334 autumn and early winter days with a few exceptional cases in March, April and August. Similarly,
335 the maximum total ozone values were mostly found in late winter and in early spring (Fig. 3b);
336 the solid black line indicates the overall mean value (327 DU) and the grey dashed line
337 represents the baseline ozone level of 220 DU which was not observed over Antarctica prior to
338 1979 (https://ozonewatch.gsfc.nasa.gov).

339
340
341 **Figure 3**: Daily total ozone values: (a) Box plots for each season for the period (1991-2015) at
342 southern England, (b) Line plots for the period 2005-2015 at Reading.

343 Table 3 presents the estimates of the linear slopes in percentage change per year in total ozone
344 data with 95% confidence intervals (CI). The regression analysis of the trend for the period
345 1991-2015 showed a highly statistically significant increasing linear trend of 0.17% y^{-1} (95% CI:
346 0.09%; 0.25%, p<0.001). The evidence for autocorrelation in the residuals of this regression
347 analysis was tested and the DW test confirmed that the overall level of autocorrelation in the
348 residuals was highly statistically significant (p<0.001). Applying the non-parametric MK test to
349 these data also indicated a strongly statistically significant increasing trend in the total ozone
350 across the full study period and the Sen's slope median trend estimate was 0.13 % y^{-1} (95% CI:
351 0.05%; 0.21%, p<0.001). This slope estimate was smaller than that obtained by the linear
352 regression analysis (Table 3).

353 A model consisting of two trend lines with a node at 2004 was fitted to these data and the results
354 are shown in Table 3. The regression analysis gave slightly different results to those obtained
355 using the non-parametric methodology. The regression analysis found an increasing trend of
356 0.19% y^{-1} in total ozone for the period 1991-2004 which was borderline statistically significant
357 (p=0.06) and a statistically significant (p=0.03) upward trend for 2004-2015 with a value of 0.28%
358 y^{-1} (95% CI: 0.003; 0.53). The non-parametric test also showed that the slope of the trend during
359 1991-2004 was positive (0.16% y^{-1}; 95%CI: -0.02; 0.35), but not statistically significant (p=0.09),
360 while in the latter period the slope trend was positive, 0.22% y^{-1} (95% CI: 0.002%; 0.44%), but
361 this result was of borderline statistical significance (p=0.05).

362 The comparable analyses of the seasonal data are also presented in Table 3. The trend for the
363 total ozone data was only statistically significant in winter over the period 1991-2015 (0.43% y^{-1};
364 95% CI: 0.19%; 0.67%, p<0.001). While there was evidence for autocorrelation in the residuals,
365 Sen's slope trend estimates were found to be very similar to the slope estimates obtained by
366 linear regression. However, there were no statistically significant positive trends identified for
367 any of the seasons either for the first period 1991 to 2004 or for the second period 2004 to 2015
368 and the 95% confidence interval lower bounds were negative for all seasons. In particular, in
369 summer, a statistical analysis showed that ozone trend was not statistically significant with there
370 being very small when H_{er} was high over any time periods. The significant ozone trends in winter
371 will influence the very low H_{er} at that time of year, but have a little practical influence on overall
372 annual dose of H_{er}.

3.3   Erythema effective UV radiant exposure (H_{er}) and total ozone

Further analyses were carried out to examine the relationship between H_{er} and the ground-based ozone. Fig. 4 shows that the relationship between H_{er} and total ozone appears to be an inverse one with H_{er} being high when total ozone is low and vice versa. This is evident for all seasons (Fig.4b). The greater variability in H_{er} observed, in particular in winter and spring, appears to be caused by the greater variability of total ozone for the same seasons (Fig.4a). However, H_{er} effect was negligibly small in winter and might be significant in spring if the total ozone events get low. An inverse relationship was also observed in summer and in autumn, but not to the same extent as that seen in winter and spring. Particular fluctuation in total ozone and H_{er} values was observed after 2007 for each season; higher values of total ozone and lower values of H_{er} were seen and the same pattern was also observed from December 2011 to March 2012. However, the highest H_{er} values recorded at Chilton over the 25 year period were recorded in 2003 and there was not any significant total ozone reduction in the same year. In contrast, higher values of ozone and lower values of H_{er} were seen from November 2009 to January 2011. The recent years from April 2012 to December 2015, the ozone level appeared to be stayed higher, while H_{er} level remained low. In absolute terms, all these observed changes are small and implications for H_{er} in winter are insignificant.

Figure 4: Relationship between monthly mean deviation of H_{er} (1991-2015) and the total ozone data (1991-2015): (a) seasonal, (b) the fitted linear regression line with 95% CI.

Table 4 shows the results of the regression analyses of the monthly mean deviation of H_{er} against the total ozone by season and for all seasons together for the period 1991-2015 and the correlation coefficient estimates (r^2) for each regression model. The inverse correlation between H_{er} and total ozone was found to be strongly statistically significant (p<0.001) for the period 1991-2015 such that a 1.0% increase in total ozone was associated with a 1.3% decrease in H_{er} that is known as the Radiation Amplification factor (RAF) for the erythemal action spectrum for sunburn of human skin and the RAF is about -1.3. The scatterplot between H_{er} and ozone in Figure 4b also shows this fitted regression line in which there is a wide spread of data points around the line indicating a weak correlation which was confirmed by an r^2 value of 25%. However 75% of the variation could not be explained by total ozone alone and that other factors such as cloud cover and aerosols are likely to be important.

The results for the two-sub periods 1991-2004 and 2004-2015 are also presented in Table 4. A statistically significant negative correlation was also found between H_{er} and total ozone (p<0.001 for both periods. The estimated slope was negative for both periods; that is H_{er} was seen to decrease by 1.2% and 1.5% for a 1% increase in total ozone from 1991 to 2004 and for 2004-2015 respectively. These RAF values are slightly different than that RAF for the full study period (1991-2015), but a test of heterogeneity in three RAF values showed that there was no statistically significant difference in them (p=0.68). The corresponding correlation coefficients were found to be weak (18% and 33% respectively) indicating that other factors were also influencing the variation of H_{er} over these periods as mentioned earlier.

For the seasonal data, the inverse correlation between H_{er} and total ozone was also highly statistically significant (p<0.001) for 1991-2015 (Table 4). The highest correlation between H_{er} and total ozone arose in spring (41%) and summer (34%). A 1% increase in total ozone during spring and summer seasons leads to an average of 2.4% and 1.9% decrease in H_{er} respectively. The RAF value in winter was less negative than that value in summer, spring and autumn. This

421 <u>pattern is not surprising as an increase in cloudiness tends to reduce H_{er} and as a result the RAF</u>
422 <u>become less negative in winter. A test of heterogeneity in RAF values was tested between</u>
423 <u>seasons; there was not statistically significant difference in the RAF values between summer,</u>
424 <u>spring and autumn (p=0.82) and also between winter and autumn (p=0.35).</u>
425
426 Across both sub periods, the inverse correlation between H_{er} and total ozone was statistically
427 significant for all seasons except during winter for the period 1991-2004 (p=0.12). In contrast, for
428 the period 2004-2015, the correlation was stronger in winter (52%) and also in spring (48%) than
429 that in summer (24%) and autumn (17%), although the H_{er} level in winter is very low and the
430 effect of total ozone is negligible. <u>The test heterogeneity in RAF values were also performed for</u>
431 <u>both periods between seasons; there was also no statistically significant differences in RAF</u>
432 <u>values between spring, summer and autumn (p=79) for the period 1991-2004. Although th RAF</u>
433 <u>value in autumn was less negative than that in winter for the period 2004-2015, there was no</u>
434 <u>differences in RAF values between all seasons (p=0.53).</u> The results for both study periods also
435 showed the lowest RAF values were estimated in spring and summer. The daily mean value of
436 H_{er} in summer here is about 14 times larger than in winter.
437
438
439 ## 3.4   Erythema effective radiant exposure (H_{er}) and cloud cover
440
441 The long term changes in H_{er} in all weather conditions also differ according to variations in cloud
442 cover. The regression analysis of all the cloud cover data showed a statistically significant
443 downward linear trend with a mean rate of 0.19% y^{-1} (95% CI: -0.34%; -0.04%, p=0.01). When
444 the data for each season was considered separately, a statistically significant downward linear
445 trend was only found in spring (p=0.025) although the trend slope was negative for the other
446 three seasons. The regression analysis of cloud cover for the first period (1991-2004) also
447 showed a statistically significant downward linear trend of 0.68% y^{-1} (95% CI:-1.03; -0.33,
448 p=0.0002), but for 2004-2015 the downward linear trend was small (-0.04% y^{-1}) and not
449 statistically significant (p=0.85). Seasonally, the slope estimates were negative for all four
450 seasons for 1991-2004, but only the trends for winter and spring were statistically significant (-
451 0.93% y^{-1}, p=0.02 and -0.72% y^{-1}, p=0.03 respectively). In contrast, for 2004-2015, there was no
452 evidence of a trend in cloud cover for any season, although the trend estimate was negative for
453 winter and spring, but positive for summer and autumn.

454 Fig.5 shows the relationship between cloud cover and H_{er} for the period 1991-2015. As expected
455 an inverse relationship was observed and peak H_{er} was seen to increase in response to
456 decreasing cloud cover for all seasons.

457
458
459 **Figure 5**: Relationship between the mean deviation of H_{er} (%) and the cloud cover at Chilton
460 (1991-2015): (a) seasonal, (b) correlation plot showing the linear regression line with 95% CI.

461 Table 5 shows the results of the regression analyses of the monthly mean deviation of H_{er} data
462 against cloud cover and the correlation coefficient values to quantify the strength of the
463 relationship by season and for all seasons together at Chilton from 1991 to 2015. A highly
464 statistically significant inverse correlation was found for each season and for all seasons
465 together. For the whole data over the period 1991-2015 the analysis shows a 1% increase in
466 cloud is associated with a decrease of about 1% in H_{er}. This fitted regression line on a scatterplot
467 (Fig.5b) indicates modest correlation between total ozone and H_{er} which was confirmed by an r^2

468 value of 38% (Fig. 5b) and over 62% of the variation remaining unexplained. Seasonally, the
469 highest correlations were observed in spring (48%) and summer (46%) over the period 1991-
470 2015.
471
472 A statistically significant negative correlation was also found for the whole data for the two-sub
473 periods 1991-2004 and 2004-2015 (p<0.001 for both periods) and the results are also presented
474 in Table 5. While the regression slopes were similar for both periods the strength of the
475 correlation was moderate (48%) for the first period, but low for the latter period (27%). For the
476 seasonal data, all regression slope values were negative and statistically significant. The
477 correlation was strongest in spring (66%) and summer (64%) for 1991-2004, but moderate for the
478 same seasons for 2004-2015.
479

3.5 H_{er}, total ozone and cloud cover

481 A multiple linear regression analysis was used to investigate how variation in cloud cover and
482 total ozone considered together were associated with changes in H_{er} from 1991 to 2015. The
483 results are presented in Table 6. The estimated slopes for both cloud cover and total ozone
484 were negative and statistically significant for all seasons together. On average H_{er} decreased by
485 0.82% for each additional 1% increase in cloud cover at constant levels of total ozone. Similarly
486 for every additional 1% increase in total ozone, H_{er} decreased by 1.03% at constant levels of
487 cloud cover. This RAF value is slightly different than the model with no cloud cover effect
488 adjustment, but this value did not differ statistically significant (p=0.50). The correlation
489 coefficient (r^2) was moderate (51%) for all seasons together, of this total variation in H_{er} was
490 explained by these two factors together, with 37% accounted for by cloud cover variation and
491 14% by total ozone variation.
492
493 Across the season specific analyses, the correlation between H_{er} and these two factors was
494 highest in spring (68%) and summer (55%) (Table 6). In both instances cloud cover accounted
495 for the largest variation in H_{er} (47% and 46% respectively), while the variation in H_{er} explained by
496 total ozone was low (21% and 12% respectively) when cloud cover was in the model. This
497 means that 32% and 45% of variation in H_{er} in spring and summer respectively remained
498 unexplained. As expected, the correlation coefficient for winter was the lowest (42%) while for
499 autumn the correlation between H_{er} and these two factors was found to be moderate (51%) with
500 variation in cloud cover accounting for 41% and total ozone explaining only 10% of the total
501 variation in H_{er}. There was also no statistically significant differences in RAF values between all
502 seasons (p>0.50) except between winter and spring (p=0.02) at constant levels of cloud cover.
503
504 Table 7 shows the results from the multiple linear regression analysis for two-sub periods (1991-
505 2004 and 2004-2015). For the first period, for every 1% increase in total ozone, H_{er} decreased
506 by 0.79% at constant levels of cloud cover, that is RAF is -0.79. Similarly H_{er} decreased by
507 0.97% for each additional 1% increase in cloud cover at constant levels of total ozone. For 1991-
508 2004, 55% of the total variation in H_{er} was explained by cloud cover and total ozone together.
509 Cloud cover accounted for 47% of the total while total ozone explained only 8%.
510
511 In contrast, for the period 2004-2015, of the 49% of the variation in H_{er} explained by both factors,
512 33% was explained by total ozone and 16% by cloud cover. The RAF value (-1.25) appeared to
513 be smaller than that RAF for the first sub period, but the test showed that there was no evidence
514 that these values differ (p=0.44). The H_{er} level decreased by 0.65% for each additional 1%
515 increase in cloud cover at constant levels of total ozone.
516

517　The season specific results showed similar negative trends. The highest correlation for the period
518　1991-2004 was observed in spring (82%). Of this total 65% was explained by cloud cover and
519　17% by total ozone. In contrast, the highest correlation value for 2004-2015 was found in winter
520　(67%). Of this total variation cloud cover explained 15% of the variation in H_{er}, while total ozone
521　explained 52%, a much larger contribution. For summer and autumn, cloud cover was found to
522　be the larger influence to the variation in H_{er} (31% and 26%, respectively) in comparison with
523　total ozone (10% and 11%, respectively). The test heterogeneity in RAF values were also
524　performed for both periods between seasons; there was also no statistically significant
525　differences in RAF values between seasons (p>0.10) for both periods (p>0.10).

527　**4   Summary and Discussion**

529　**4.1   Erythema effective UV radiant exposure (H_{er})**

531　This paper reports an analysis of the effect of total ozone and cloud cover on the erythema
532　effective UV radiant exposure (H_{er}) at Chilton between 1991 and 2015. During this period the
533　highest H_{er} levels were observed in 2003. This peak was likely to be due to the exceptionally hot
534　spring and summer with low cloud cover at the site during that year, but not with any significant
535　reduction in total ozone level. It was also the same year that a heat wave affected much of
536　Western Europe including England (Vieno et al., 2010; Beniston 2004). However, hot weather
537　does not necessarily mean high UVR and cold weather does not necessarily mean low UVR
538　(Wong et al 2015). High levels of H_{er} were also reported at two sites, Lindenberg in Germany and
539　at Bilthoven in Holland (den Outer et.al. 2005; WMO et. al 2007) in 2003. These site are at
540　latitudes (49° N, 52° N respectively) which are close to that of Chilton (52° N). Den Outer &
541　colleagues suggested that the high annual erythema effective UV dose received in Holland in
542　2003 was associated with extremely low cloud levels combined with moderately low ozone
543　values. However, no such associations were reported at Uccle in Belgium with a latitude of 51°
544　(De Bock et al., 2014) or at Reading in the UK (Smedley et al., 2012). H_{er} data at Chilton also
545　showed a reversal in trend before and after 2003 with an increasing trend from 1991 to 2003 but
546　a decreasing trend thereafter. In order to avoid bias in the analyses caused by the highest H_{er}
547　values occurring in 2003, the year 2004 was chosen to be the change in point in preference to
548　2003.

550　In our previous analysis of the long-term variability of H_{er} between 1991 and 2015 at Chilton the
551　data were divided into two separate time series data due to geophysical phenomena; one from
552　1991 to 1995 during which the ozone turning point in the mid-1990s because excess volcanic
553　aerosol after the Pinatubo volcanic eruption may cause short-term ozone depletion and as a
554　result enhanced the amount of UVR (WMO 2014) and a second from 1995 to 2015 (Hooke et al.,
555　2017).

557　In contrast, this current work splits the time series according to statistical analysis; we have
558　shown that the H_{er} data for 1991-2015 (based on a nonlinear model over the full period) were
559　statistically better described by two linear trends; the first a statistically significant increasing
560　linear trend value of 1.01% y^{-1} (p<0.0001) for 1991-2004 and the second a statistically significant
561　decreasing trend of 1.35% y^{-1} (p<0.0001) from 2004 to 2015. Our finding for the first period is
562　not consistent with our earlier result for the period 1991-1995 where a higher estimate (4.4% y^{-1})
563　was obtained however the earlier result should be treated with caution due to relatively short time
564　period, 5 years, over which the trend was calculated. Our findings for the second period agree
565　with those of our early study for 1995-2015 but the trend estimate was slightly lower (0.8% y^{-1}).

566
567 The finding in this study for the first period (1991-2004) is in good agreement with those from
568 European studies that also reported significant increasing linear trends. At Lindenberg in
569 Germany there was reported an increasing trend of 0.77% y^{-1} during 1996-2003, 0.85% y^{-1} for the
570 period 1999-2004 and 1.4% y^{-1} over the period 1998-2005. The studies at Norrköping in Sweden
571 (with a latitude of 58°) and also at Bilthoven in Holland, both reported an increasing trend during
572 1996-2004 (1.2% and 0.86% y^{-1} respectively) based on solar zenith angles (SZA) of 60°, but the
573 trend was higher (1.7% y^{-1}) at Bilthoven for the period 1998-2005 when the noon values of the
574 erythemal UV radiation were used (Bais et al., 2007). The study at the Hoher Sonnblick site in
575 Austria (Fitzka et al., 2012) showed a significant upward trend in the erythemally weighted
576 irradiance for the period 1997-2011 with a range from 0.84%±5.2% y^{-1} at 45° SZA to
577 1.26%±0.36% y^{-1} at 65° SZA under all weather conditions. However, a smaller and less
578 significant result was seen at wavelengths of 305 nm (between -0.76%± 1.13% y^{-1} and 0.79%±
579 0.73% y^{-1}, depending on SZA). The study based at Reading in the UK found a significant
580 increasing linear trend (0.66% per year) for the period from 1993 to 2008 based on the midday
581 values of UV index (Smedley et. al 2012).
582
583 The trend in H_{er} in this study over the second period (2004-2015) at Chilton is consistent with
584 values derived for the averaged UV-B data over Canada, Europe and Japan that showed
585 statistically significant evidence of a reduction in UV-B for the period 2007-2011 with the slope
586 estimates ranging from -1.5% to -2% under cloudless conditions (Zerefos et al., 2012). Our
587 findings are also in good agreement with the results of Fountoulakis et al. (2016) at Thessaloniki
588 (latitude of 40° N) in Greece, where a turning point in the trends of UV irradiance is reported as
589 being in 2006; a statistically significant increasing trend of 0.71% ±0.21% y^{-1} for the period 1994-
590 2006 and a decreasing trend of 0.33% ± 0.32% y^{-1} from 2006 to 2014 but the trend was not
591 statistically significant. It appears that there is a similar behaviour of the trend in the UV
592 irradiance between this UK study and the Greek study, although these countries differ
593 significantly in terms of climate and location. However, a recent study at Uccle in Belgium
594 covering the period 1991-2013 which is similar to that examined in this study found a strongly
595 statistically significant increasing linear trend of 0.7% y^{-1} (De Bock et.al. 2014). In comparison,
596 our results for the period 1991-2015 found a non-significant downward trend.
597
598 When the H_{er} data for each season were analysed separately, a statistically significant increasing
599 trend was only found during winter for the first period 1991-2004 despite large inter-month
600 variability and contributes only a small fraction of the annual cumulative H_{er} level in the UK. Much
601 of this significant result might be caused by the excess of low total ozone events observed in
602 winter (detail will be discussed in the next section). However, there was no significant linear trend
603 in H_{er} in either spring or in summer at Chilton. The absence of a significant trend in spring for
604 this period might be due to higher values of total ozone level over the same period.
605
606 For the second period 2004-2015, a linear downward trend in H_{er} was observed for all four
607 seasons, but the trend was only statistically significant in summer and autumn. The results of the
608 current study are comparable with those of the Belgian study at the Uccle site from 1991 to 2013,
609 that showed the largest statistically significant increasing trend in H_{er} in spring but a negative
610 trend in winter, albeit not statistically significant (De Bock et. al 2014). In addition, the Austrian
611 study at the Hoher Sonnblick site for the period 1997-2011 also found that the largest and most
612 significant linear trends were during winter and spring.
613
614 **4.2   Erythema effective UV radiant exposure (H_{er}) and total ozone**
615

616 The significantly increasing trend in total ozone of 0.13% y^{-1} (p<0.001) in the south of England
617 between 1991 and 2015, could be due to natural variability in total ozone. This result is lower but
618 in general good agreement with the significant upward trend reported in European studies: the
619 estimated trend at the Hoher Sonnblick site in Austria during 1997-2011 was 0.19% y^{-1} (Fitzka et
620 al., 2012) and at the Uccle site in Belgian during 1991-2013 the trend was 0.26% y^{-1} (De Bock et.
621 al.,2014). Our finding is also consistent with the result for the period 1995-2011 over Canada,
622 Europe and Japan (Zerefos et al., 2012). The Reading study, however, using a subset of the
623 same total ozone data used here reported a small average increase after 1993, but the trend
624 was not statistically significant from 1993 to 2008 (Smedley et al., 2012). However, the authors
625 noted a small average increase that lies within the range of trends observed at other European
626 stations (Smedley et al., 2012). The analysis of seasonal data found a much larger spread of
627 ozone measurements in winter and spring months compared to those in summer and autumn for
628 the period 1991-2015. The largest and most significant increasing linear trend was found during
629 winter. While there was an upward trend in other seasons it was markedly smaller and not
630 significant. The Reading study did not show any significant trend for any season although an
631 increasing rate in winter was noted for the period 1993-2008. Unlike this study, the Belgian study
632 at the Uccle site only found statistically significant increasing linear trends of ozone in spring and
633 summer for the period 1991-2013 (De Bock et al., 2014). The trend estimates were quite
634 variable between these studies and depend on what period was chosen and therefore comparing
635 these estimates across studies should be treated with caution.

636 We examined whether the long-term behaviour of the measured H_{er} could be explained by total
637 ozone variation. Between 1991 and 2015, while there was a statistically significant inverse
638 relationship between total ozone and H (p<0.001), the total ozone has a weak inverse linear
639 correlation with H_{er} (25%). This is not surprising as the amount of UV radiation reaching the
640 Earth's surface depends not only total ozone but also cloud cover, atmospheric aerosols, air
641 pollution and other climate factors (Calbó et al., 2005). The relationship between changes in H_{er}
642 and total ozone is expressed with the radiation amplification factor (RAF) for sunburn of human
643 skin and this study was found to be the RAF was about -1.3. This value is in good agreement
644 with the reported RAF of -1.1 in the US study (Hall 2017) and the RAF values ranging from -1.3
645 to -1.4 in Spain (Antón et al. 2009).

647 To better compare the variation in H_{er} with that of total ozone two linear trends with a node at
648 2004 were fitted to the ozone data. The trend for total ozone was positive, but not statistically
649 significant (p=0.09) for the period 1991-2004, but there was a strong statistically significant
650 increase of in H_{er} over the same period. In contrast, for the second period 2004-2015, the trend
651 for total ozone showed a borderline statistically significant increasing linear trend (p=0.05) but the
652 H_{er} trend showed a significant decrease over the same period.

654 For both sub-periods (1991-2004 and 2004-2015) a statistically significant inverse correlation
655 was observed between H_{er} and total ozone, but the amount of variation in H_{er} explained by that of
656 total ozone was low (18% and 33% for each time period respectively). Our estimates of the size
657 of the trend in ozone are smaller than those reported in the study by Zerefos et al., (2012) based
658 on averaged total ozone and UV-B data over Canada, Europe and Japan. That study found that
659 for the period 2007-2011 the effect of increasing total ozone on UV-B values was about -4%
660 when aerosol optical depth (AOD) was factored into the model. The Belgian study (De Bock et
661 al., 2014) also reported a greater effect of total ozone on the erythemal UV dose (-5%) during the
662 period 1991-2008 when measures of global solar radiation and AOD were taken into account.
663 The Reading study, however, did not find any correlation between the surface UV radiation and

664 total ozone for the period 1993-2008. The authors suggested that the majority of the variability in
665 UV radiation was due to changes in cloud cover and other effects (Smedley et al., 2012).
666
667 Examining our data on a season by season basis over the whole period from 1991 to 2015, we
668 found a highly negative slope estimate for each season. However there were some differences in
669 how much of the variation in H_{er} that was explained by total ozone across the seasons. In spring
670 and summer the variability explained was moderate at 41% and 34% respectively but in winter
671 and autumn it was considerably lower at 19% and 21% respectively.
672
673 Restricting the data to the first period (1991-2004), we also saw the greatest impact of total
674 ozone on H_{er} in spring (37%) and summer (42%) but for the second period (2004-2015), the
675 impact was bigger in winter (52%) and spring (48%) than that in summer (24%) and autumn
676 (17%). This is mainly due to the strong inverse relationship between H_{er} and ozone that was
677 observed during spring and winter in the period 2008- 2015 (63% and 56% respectively).
678

679 **4.3 Erythema effective UV radiant exposure (H_{er}) and cloud cover**
680
681 Cloud cover can have a marked impact on the amount of UVR that reaches the earth's surface.
682 An increase in cloud cover usually results in a reduction of UV radiation below the clouds. Whilst
683 UVR can pass through thin and broken clouds thick clouds tend to reflect, absorb or scatter UV
684 radiation. Puffy, fair-weather clouds deflect rays and can actually increase the UV radiation
685 reaching the earth's surface (Alados-Arboledas et al., 2003). Our analysis of cloud cover
686 variation showed a statistically small significant decreasing trend of -0.68% y^{-1} (p<0.001) for the
687 first period (1991-2004) but no significant trend for the second period (2004-2015) although the
688 estimated slope was negative (-0.04% y^{-1}). These findings agree partly with other studies that
689 reported a decrease in cloud cover (Norris and Slingo, 2009; Eastman and Warren, 2013) and
690 also with those that did not find any evidence of a decreasing trend in cloud cover. For example,
691 the studies at the Hoher Sonnblick site in Austria over the period 1997-2011 (Fitzka et al., 2012)
692 and in the study examining data from Europe, Canada and Japan for the period 1995-2011
693 (Zerefos et al., 2012).
694
695 Examining our data on a season by season basis, the only statistically significant trends in cloud
696 reduction was observed in spring and winter during the period 1991-2004. For subsequent years
697 there was no evidence of a trend in observed cloud cover in any season. These observations
698 agree with the findings from the Austria study at the Hoher Sonnblick for 1997-2011 (Fitzka et al.,
699 2012).
700
701 The inverse correlation between H_{er} and cloud cover was also found to be strongly statistically
702 significant for both sub-periods. However, while about half the variation in H_{er} was explained by
703 the cloud cover variation in the first period this fell to just over one quarter for the second period.
704
705 The same data examined on a season by season basis showed, for the first period, just over half
706 the variation in H_{er} explained by the inverse relationship with cloud cover for spring, summer and
707 autumn but only just over one quarter for winter, although the slope of the relationship did not
708 vary greatly with season. For the second period, the variation in H_{er} explained by cloud cover
709 dropped below 50% for all seasons and in particular the value for autumn (26%) reduced to
710 below that for winter (29%). Although, the Austrian study at the Hoher Sonnblick did not study
711 the correlation between cloud cover and UV measurements, the authors reported that the total

712 cloud reduction of 1.04% y^{-1} was evident for UV measurements at SZA 55° for the period 1997-
713 2011.
714
715 **4.4   Erythema effective UV radiant exposure (H$_{er}$), total ozone and cloud cover**
716
717 Given that we found clear evidence that variation in H$_{er}$ could be partially explained by variation
718 in total ozone and cloud cover separately we considered their combined effects. Over the whole
719 period 1991- 2015 half of the variation in H$_{er}$ could be explained by these two factors with the
720 changes in cloud cover alone accounting for 37% of the variation in H$_{er}$, while the total ozone
721 variation explained 14%. The unexplained half of the H$_{er}$ variation may be attributed to other
722 factors such as atmospheric aerosol, air pollution or climate. The RAF value was found to be -
723 1.03 at constant levels of cloud cover. Although this RAF value was slightly different than that
724 period between 1991 and 2004 (-0.79) and for 2004-2015 (-1.25) when cloud cover effect in the
725 model, but there was no statistically significant differences in the RAF values (p=0.66). These
726 RAF results are also consistent with the RAF values in the US study in which impact of clouds on
727 the RAF was determined and the RAF ranged from a low of -0.80 to a high of -1.38 (Hall 2017).
728
729 The effects of aerosols on surface UV irradiance have also been studied widely in addition to
730 ozone and cloud in Europe (Román et al., 2015, De Bock et al., 2014, Zerefos et al., 2012, Fitzka
731 et al., 2012). Our study, however, did not take account of the effects of aerosols because Chilton
732 is situated in a very rural location in South Oxfordshire in the UK and the levels are generally
733 very stable. The aerosols optical depth (AOD) trend in London was reported in the range of 0-
734 0.004 per year decrease between 2003 and 2015 (Provençal et al., 2017).
735
736 Aerosols can affect ground level UV irradiances directly through absorption and scattering solar
737 radiation back out to space, reducing the amount of solar radiation reaching the surface of the
738 Earth. Aerosols can also affect indirectly which are related to modify cloud formation.
739 Atmospheric aerosols originate from both natural sources (such as dust) and from anthropogenic
740 sources – such as air pollution from industry and traffic producing more pollution and
741 atmospheric aerosol, in particular in urban areas. Alpert et al. (2012) reported that aerosols
742 optical depth (AOD) trends declined over the largest cities in Europe during the period (2002-
743 2010) owing to increasing air quality due to environmental regulations (2012). Nevertheless, the
744 influence of the aerosols on UV irradiance has not been fully understood due to their high spatial
745 and temporal variability (WMO 2007).
746
747 In this work, the combined effects on a season by season basis the biggest proportion of
748 variability in H$_{er}$ explained by total ozone and cloud cover together was for spring (68%) while the
749 smallest proportion was in winter for period 1991-2015. In each season cloud cover explained far
750 more of the variability than did total ozone, based on the additive linear regression model was
751 used.
752
753 The combined effects of total ozone and cloud cover on H$_{er}$ was also assessed using two linear
754 trends with a node at 2004. Over the first period the proportion of H$_{er}$ variability explained by
755 these two factors rose a small amount (4%) but fell slightly for the second period (2%) compared
756 to the proportion when the entire period 1991 to 2015 was considered as a whole.
757
758 A major difference was seen in how much of the variability was explained by each factor across
759 the two periods. In the first period cloud cover variation accounted for a lot more of the explained
760 variability compared to ozone (47%:8%) whereas for the latter period the proportions were
761 (16%:33%). This is because for the second period significant correlation between total ozone H$_{er}$

762 observed in winter and spring during 2008-2015 had a bigger impact on H_{er} than that in cloud
763 cover for the same period.

764

765 Across the seasons there were marked differences in the proportion of H_{er} variability explained
766 by the two factors. For the period 1991-2004 in spring, the proportion explained rose to 82% but
767 fell to only 31% in winter. However, for the latter period 2004-2015, the variability in H_{er} explained
768 by the two factors was at a maximum in spring and winter (63% and 67% respectively) and at a
769 minimum in summer and autumn (41% and 37% respectively).

770

771 The season specific analysis of these data also showed that the size of the respective
772 contributions that cloud cover and total ozone made to the variation in H_{er} changed between the
773 two periods. For the first period, in spring and summer, cloud cover explained 65% and 63% of
774 the variability respectively compared to the 17% and 7% contributions of total ozone. For the
775 second period in both winter and spring cloud cover explained 15% of the H_{er} variability while
776 total ozone contributed 52% and 48% respectively.

777

778 This study provides robust evidence that both increasing trend for the first period (1991-2004)
779 and decreasing trend for the second period (2004-2015) in H_{er} occur at the same time as
780 increasing total ozone. However, increasing trend in H_{er} over the first period is more strongly
781 associated with the observed reduction in cloud cover, while there is no significant change in
782 cloud cover over the second period that H_{er} is decreasing. All these changes are small and
783 occur within a very variable signal.

784

785 Our findings from the first period partly agrees with those from the Austrian study at Hoher
786 Sonnblick over the period 1997-2011, which reported that the significant increase in H_{er} was
787 attributed to a decrease in cloud cover as well as aerosol optical depth (AOD) rather than a
788 significant increase observed in ozone. However, the authors showed that the changes in the
789 UVR at 305 nm with cloud cover were small and less significant due to the enhanced influence of
790 ozone absorption at the shorter wavelengths (Fitzka et al., 2012). In addition, the Iberian
791 Peninsula study based on data from nine locations in Spain also reported an increase in
792 erythema effective ultraviolet irradiance between 1985 and 2011 and attributed that to changes in
793 AOD and cloud cover rather than total ozone (Román et al., 2015).

794

795 Some of the European monitoring sites have also demonstrated an overall increase in the
796 reconstructed erythema effective UV irradiance observed for the period 1980-2006, two thirds
797 could be attributed to diminishing cloud cover or AOD and only one third to the total ozone
798 reduction (den Outer et al., 2010). The study over Canada, Europe and Japan during 1995-2006
799 also showed that the decline of AOD and significant increase in total ozone were the associated
800 with increased UV-B, although a non-significant trend with cloud cover was found (Zerefos et al.,
801 2012). In contrast, the Belgian study reported individual contribution of insignificant negative
802 trend AOD on erythemal UV dose was very low, while the impact from total ozone was strong
803 (De Bock et al., 2014).

804

805 Our findings from the second period (2004-2015) appear to be partly consistent only with the
806 findings over the period 2007-2011 by Zerefos et al., (2012) in which the authors reported that
807 decreasing trends in the UV-B from 2007 to 2011 were largely driven by increasing total ozone
808 and to a lesser degree of the significant AOD decrease but did not find a statistically significant
809 trend in UV associated with cloud cover. The observed decreasing trend in H_{er} at Chilton for the

810 second period (2004-2015) should be treated with caution because of the lack of other evidence
811 of a decreasing trend in H_{er} data up to 2015 in the literature.
812
813 **Acknowledgements**
814

820

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822
823 **References**
824

825 AGNIR (Advisory Group on Non-ionising Radiation): Health Effects from Ultraviolet Radiation,
826 Report of an Advisory Group on Non-ionising Radiation. Documents of the NRPB 13:1, 2002.
827
828 Alados-Arboledas, L., Alados, I., Foyo-Moreno, I., Olmo, F.J. and Alcántara, A.: The influence of
829 clouds on surface UV erythemal irradiance. Atmospheric Research, 66(4), 273-290, doi:
830 10.1002/joc.1883, 2003.
831
832 Alpert, P., Shvainshtein, O., and Kishcha, P.: AOD trends over megacities based on space
833 monitoring using MODIS and MISR, Am. J. Clim. Change, 12, 117-131,
834 doi:10.4236/ajcc.2012.13010, 2012.
835
836 Antón, M., Serrano, A., Cancillo, M. L. and García, J.A.: Am empirical model to estimate
837 ultraviolet erythemal transmissivity. Ann. Geophys., 27, 1387–398, 2009.
838
839 Bais, A. F., Kazadzis, S., Meleti, C., Kouremeti, N., Kaurola, J., Lakkala, K., Slaper, H., den
840 Outer, P. N., Josefsson, W., Feister, U., and Janouch, M.: Variability in spectral UV radiation at
841 seven European stations, edited by: Gröbner J., One century of UV radiation research.
842 Proceedings of the UV conference, Davos, Switzerland, 1, 27–28, 2007.
843
844 Bais, A. F., Tourpali, K., Kazantzidis, A., Akiyoshi, H., Bekki, S., Braesicke, P., Chipperfield, M.
845 P., Dameris, M., Eyring, V., Garny, H., Iachetti, D., Jöckel, P., Kubin, A., Langematz, U., Mancini,
846 E., Michou, M., Morgenstern, O., Nakamura, T., Newman, P. A., Pitari, G., Plummer, D. A.,
847 Rozanov, E., Shepherd, T. G., Shibata, K., Tian, W., and Yamashita, Y.: Projections of UV
848 radiation changes in the 21st century: impact of ozone recovery and cloud effects, Atmos. Chem.
849 Phys., 11, 7533–7545, doi:10.5194/acp-11-7533-2011, 2011.
850
851 Beniston, M.: The 2003 heat wave in Europe: A shape of things to come? An analysis based on
852 Swiss climatological data and model simulations, Geophys. Res.Lett., 31, L02202,
853 doi:10.1029/2003GL018857, 2004.
854 Calbó,J., Pagès, D., and González, J-B.: Empirical studies of cloud effects on UV radiation: A
855 review. Rev. Geophys., 43, RG2002, doi:10.1029/2004RG000155, 2005. Chatfield C.: *The
856 analysis of time series: an introduction*, Sixth Edition, Chapman and Hall, CRC text in statistical
857 science, 1996.

De Bock, V., De Backer, H., Van Malderen, R., Mangold, A., and Delcloo A.: Relations between erythemal UV dose, global solar radiation, total total ozonecolumn and aerosol optical depth at Uccle, Belgium, Atmos. Chem. Phys.,14, 12251-12270, doi:10.5194/acp-14-12251-2014, 2014.

den Outer, P. N., Slaper, H. and Tax, R.B.: UV radiation in the Netherlands: Assessing long-term variability and trends in relation to ozone and clouds. J. Geophys. Res., 110, D02203, doi:10.1029/2004JD004824, 2005.

den Outer, P. N., Slaper, H., Kaurola, J., Lindfors, A., Kazantzidis, A., Bais, A. F., Feister, U., Junk, J., Janouch, M., and Josefsson, W.: Reconstructing of erythemal ultraviolet radiation levels in Europe for the past 4 decades, J. Geophys. Res., 115, D10102, doi:10.1029/2009JD012827, 2010.

Diffey, B.L.:Sources and measurement of ultraviolet radiation, Methods, 28:4-13, doi:10.1016/S1046-2023(02)00204-9, 2002.

Dunn, R. J. H., Willett, K. M., Thorne, P. W., Woolley, E. V., Durre,I., Dai, A., Parker, D. E., and Vose, R. S.: HadISD: a quality-controlled global synoptic report database for selected variables at long-term stations from 1973–2011, Clim. Past, 8, 1649–1679, doi:10.5194/cp-8-1649-2012, 2012.

Dunn, R. J. H., Willett, K. M., Morice, C. P., and Parker, D. E.: Pairwise homogeneity assessment of HadISD, Clim. Past, 10, 1501–1522, doi:10.5194/cp-10-1501-2014, 2014.

Dunn, R. J. H., Willett, K. M., Morice, C. P., and Parker, D. E.: Expanding HadISD: quality-controlled, sub-daily station data from 1931, Geosci. Instrum. Method. Data Syst., 5, 473–491, doi:10.5194/gi-5-473-2016, 2016

Epplin, J., Thomas, S.A.: Vitamin D: It does a body good. Annals of Long-Term Care 18 (11), 39-45, 2010.

Eastman, R. and Warren, S. G.: A 39-yr survey of cloud changes from land stations worldwide 1971–2009: long-term trends, relation to aerosols and expansion of the tropical belt, Journal of Climate, 26, 1286-1303. Doi:10.1175/JCLI-D-12-00280, 2013.

Farman, J.C., Gardiner, B.G., and, Shanklin, J.D.: Large losses of total ozone in Antarctica reveal seasonal ClO_x/NO_x interaction, Nature, 315, pages 207–210, 1985.

Fitzka, M., Simic, S., and Hadzimustafic, J.: Trends in spectral UV radiation from long-term measurements at Hoher Sonnblick, Austria, Theor. Appl. Climatol., 110, 585–593, doi:10.1007/s00704-012-0684-0, 2012.

Fountoulakis, I., Bais, A.F., Fragkos, K., Meleti, C., Tourpali, K. and Zempila, M.M.: Short- and long-term variability of spectral solar UV irradiance at Thessaloniki, Greece: effects of changes in aerosols, total ozone and clouds. Atmos. Chem. Phys., 16, 2493–2505, doi:10.5194/acp-16-2493, 2016.

Gies, P., Klekociuk, A., Tully, M., Henderson, S., Javorniczky, J., King, K., Lemus-Deschamps, L. & Makin, J.: Low ozone over Southern Australia in August 2011 and its impact on solar ultraviolet radiation levels. Photochemistry and Photobiology, 89, 984–994, doi:10.1111/php.12076, 2013.

Hall E.S.: Comparison of five modelling approaches to quantify and estimate the effect of clouds on the radiation amplification factors (RAF) for solar ultraviolet radiation. Atmosphere, 8, 153, doi.org/10.3390, 2017.

1021 Harris, N.R., Kyrö, E., Staehelin, J., Brunner, D., Andersen, S.B., Godin-Beekmann, S.,
1022 Dhomse, S., Hadjinicolaou, P., Hansen, G., Isaksen, I. and Jrrar, A., 2008. Ozone trends at
1023 northern mid-and high latitudes–a European perspective. In Annales Geophysicae (Vol. 26,
1024 No. 5, pp. 1207-1220).

1026 Helsel, D.R. and R.M. Hirsch.: Statistical methods in water resources. Studies in
1027 Environmental Science 49. New York: Elsevier, 1992.

1029 Holick, M.F.: Vitamin D deficiency, New England Journal of Medicine, 357, 266–281, doi:
1030 10.1056/NEJMra070553, 2007.

1032 Hooke, R.J., Higlett, M.P., Hunter, N., O'Hagan J.B.: Long term variations in erythema effective
1033 solar UV at Chilton, UK, from 1991 to 2015. Photochemical & Photobiological sciences, **16**,
1034 1596-1603 doi: 10.1039/C7PP00053G, 2017.

1036 Hooke, R.J., Higlett, M.P.: Temperature Correction of Historic Erythema Effective Solar Uv Data
1037 Resulting in a Continuous 25-Year Data Set at Chilton, UK, Radiation Protection Dosimetry, 175
1038 (3), 363-367, doi.org/10.1093/rpd/ncw358, 2016.

1041 HPA (Health Protection Agency): Health Effects of Climate Change in the UK 2012. An update of
1042 the Department of Health report 2001/2002. Ed: Vardoulakis S & Heaviside C. Health Protection
1043 Agency (in partnership with the Department of Health), Chilton UK, ISBN 978-0-85951-723-2,
1044 2012.

1046 Lucas, R.: World Health Organisation (WHO) report*:* Solar Ultraviolet Radiation. Environmental
1047 Burden of Disease Series, No. 17, World Health Organization, Geneva, ISBN 978 92 4 159917 7,
1048 2010.

1050 McKenzie, R.L., Liley, J.B., Björn, L.O.: UV radiation: Balancing risks and benefits,
1051 Photochemistry and Photobiology 85(1), 88–98, doi.org/10.1111/j.1751-1097, 2009.

1052 Mann, H.B.: Non-parametric tests against trend, Econometrica 13:163-171, 1945.

1053 Kendall, M.G.: Rank Correlation Methods, 4th edition, Charles Griffin, London, 1975.

1054 Norris, J. R.*,* Slingo, A.: Trends in observed cloudiness and Earth's radiation budget what do we
1055 not know and what do we need to know?*, in* Clouds in the Perturbed Climate System, edited by
1056 J. Heintzenberg *and* R. J. Charlson*,* pp. 17–36*,* MIT Press*,* Cambridge, Mass, 2009.

1058 Norval, M., Lucas, R.M., Cullen, A.P., de Gruijl, F.R., Longstreth, J., Takizawa, Y., van der Leun,
1059 J.C.: The Human Health Effects of Ozone Depletion and Interactions with Climate Change.
1060 Photochemical & Photobiological Sciences 10, 199-225, doi: 10.1039/C0PP90044C, 2011.

1062 Provençal, S., Kishcha, P., da Silva, A.M., Elhacham, E., and Alpert, P.: AOD distributions and
1063 trends of major aerosol species over a selection of the world's most populated cities based on
1064 the 1st Version of NASA's MERRA Aerosol Reanalysis, Urban Clim, 20, 168-191.
1065 doi:10.1016/j.uclim.2017.04.001, 2017.

1067 Román, R., Bilbao, J., de Miguel, A.: Erythemal ultraviolet irradiation trends in the Iberian
1068 Peninsula from 1950 to 2011, Atmospheric Chemistry and Physics, 15**,** 375-391,
1069 doi:10.5194/acp-15-375-2015, 2015.

1071 Smedley, A. R. D., Rimmer, J. S., Moore, D., Toumi, R., and Webb, A. R.: Total ozone and
1072 surface UV trends in the United Kingdom: 1979–2008, Int. J. Climatol., 32, 338–346,
1073 doi:10.1002/joc.2275, 2012.

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1104
1105   UNEP (United Nations Environment Programme): Environmental effects of ozone depletion and
1106   its interactions with climate change: 2010 assessment, 236 pp., UNEP, Nairobi, Kenya,
1107   ISBN:ISBN 92-807-2312-X, 2010.
1108
1109   Waugh, D.W., Oman, L., Kawa, S.R., Stolarski, R.S., Pawson, S., Douglass, A.R., Newman,
1110   P.A., Nielsen, J.E.: Impacts of climate change on stratospheric ozone recovery, Geophysical
1111   Research Letters, 36, doi:10.1029/2008GL036223, 2009.
1112
1113   WHO (World Health Organisation): Solar Ultraviolet Radiation. Global burden of disease from
1114   solar ultraviolet radiation. Environmental Burden of Disease Series, No. 13. World Health
1115   Organization. Geneva, ISBN: 92 4 159440 3, 2006.
1116
1117   WMO: (World Meteorological Organization): Scientific Assessment of Ozone Depletion: 2006,
1118   Global ozone Research and Monitoring Project-Report No. 50, Geneva, Switzerland, 2007.
1119
1120   WMO: (World Meteorological Organization): Scientific Assessment of Ozone Depletion: 2014,
1121   Global ozone Research and Monitoring Project-Report No. 55, Geneva, Switzerland, 2014.
1122
1123   Wong, C.C., Liu, W., Gies, P. and Nixon, R., 2015. Think UV, not heat!. Australasian Journal of
1124   Dermatology, 56(4), pp.275-278.

1125   Young, C.: Solar ultraviolet radiation and skin cancer, Occupational Medicine 59, 82-88, doi:
1126   10.1093/occmed/kqn170, 2009.

1127   Zerefos, C. S., Tourpali, K., Eleftheratos, K., Kazadzis, S., Meleti, C., Feister, U., Koskela, T.,
1128   and Heikkilä, A.: Evidence of a possible turning point in solar UV-B over Canada, Europe and
1129   Japan, Atmos. Chem. Phys., 12, 2469–2477, doi:10.5194/acp-12-2469-2012, 2012.
1130
1131   Vieno M., Dore A., Stevenson D. S., Doherty R., Heal M. R., Reis S., et al.,: Modelling surface
1132   ozone during the 2003 heat-wave in the UK, Atmos. Chem. Phys., 10(16), 7963–7978,
1133   doi.org/10.5194/acp-10-7963-2010, 2010.

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Table 1: Daily H_{er} (J m^{-2}) averaged over the whole period and for each season in Chilton during 1991-2015.

	Min	Mean	Median	Stdev.	Max
Whole data	10	1294	917	1179	5655
Winter (Dec-Feb)	10	188	140	148	933
Spring (Mar-May)	84	1606	1463	943	4880
Summer (June-Aug)	212	2617	2552	944	5655
Autumn (Sep-Nov)	23	746	540	618	2913

Table 2: Estimated trends (in %, y^{-1}) for H$_{er}$ with 95% confidence intervals (CI) at Chilton for two sub-periods: 1991-2004 and 2004-2015.

	Study period			
	1991-2004		2004-2015	
	Estimated trend (95% CI)	p-value	Estimated trend (95% CI)	p-value
Monthly data	1.01 (0.48; 1.54)	<0.001	-1.35 (-1.98; -0.77)	<0.001
Winter (Dec.-Feb.)	1.29 (0.17; 2.41)	0.03	-1.08 (-3.14; 1.02)	0.24
Spring (Mar.-April)	0.84 (-0.40; 2.05)	0.22	-0.88 (-2.10; -0.34)	0.16
Summer (June-Aug.)	0.74 (-0.15; 1.76)	0.09	-1.67 (-2.48; -0.86)	<0.001
Autumn (Sep.-Nov.)	0.98 (-0.04; 2.00)	0.07	-1.56 (-2.68; -0.44)	0.01

Table 3: Estimated trends (in %, y^{-1}) for total ozone with 95% CI at southern England for the ~~corrected~~ monthly mean deviation data and for each season using various study periods.

	Study period					
	1991-2015		1991-2004		2004-2015	
	Estimated trend (95% CI)	p-value	Estimated trend (95% CI)	p-value	Estimated trend (95% CI)	p-value
Whole data	0.17 (0.09; 0.25)	<0.001	0.19 (-0.006; 0.38)	0.06	0.28 (0.03; 0.53)	0.03
Winter	0.43 (0.19; 0.67)	<0.001	0.31 (-0.20; 0.82)	0.24	0.66 (-0.14; 1.59)	0.10
Spring	0.15 (-0.02; 0.32)	0.09	0.22 (-0.16; 0.64)	0.29	0.06 (-0.41; 0.53)	0.80
Summer	0.03 (-0.07; 0.13)	0.52	0.02 (-0.21; 0.25)	0.87	0.13 (-0.09; 0.35)	0.26
Autumn	0.05 (-0.07; 0.23)	0.27	0.05 (-0.30; 0.40)	0.78	0.26 (-0.17; 0.69)	0.24

Table 4: Estimated effect of total ozone on H_{er} with 95% confidence interval based on three study periods (CI).

	1991-2015		1991-2004		2004-2015	
	Estimate (95% CI)	r^2 (%)	Estimate (95% CI)	r^2 (%)	Estimate (95% CI)	r^2 (%)
Whole data	-1.33 (-1.60; -1.06)	25	-1.18 (-1.57;-0.79)	18	-1.50 (-1.85; -1.15)	33
Winter	-0.94 (-1.37; -0.51)	19	-0.54 (-1.23; 0.15)#	6	-1.66 (-2.19; -1.13)	52
Spring	-1.88 (-2.39; -1.37)	41	-1.78 (-2.50; -1.06)	37	-1.87 (-2.54; -1.20)	48
Summer	-2.37 (-3.13; -1.61)	34	-2.47 (-3.35; -1.59)	42	-2.18 (-3.49; -0.87)	24
Autumn	-1.39 (-2.00; -0.78)	21	-1.57 (-2.37; -0.77)	27	-1.19 (-2.07; -0.31)	17

#: $p=0.12$;

Table 5: Estimated effect of cloud cover on H_{er} (%) with 95% CI, based on three study periods.

	1991-2015		1991-2004		2004-2015	
	Estimate (95% CI)	r^2 (%)	Estimate (95% CI)	r^2 (%)	Estimate (95% CI)	r^2 (%)
Whole data	-0.95 (-1.09; -0.81)	38	-1.06 (-1.23;-0.89)	48	-0.82 (-1.04; -0.60)	27
Winter	-1.09 (-1.50; -0.68)	27	-0.96 (-1.47; -0.45)	25	-1.20 (-1.83; -0.57)	29
Spring	-1.05 (-1.30; -0.80)	48	-1.20 (-1.47; -0.93)	66	-0.99 (-1.38; -0.60)	42
Summer	-0.73 (-0.90; -0.54)	46	-0.92 (-1.14; -0.70)	64	-0.53 (-0.78; -0.28)	31
Autumn	-1.05 (-1.34; -0.76)	41	-1.15 (-1.48; -0.82)	53	-0.87 (-1.36; -0.38)	26

Table 6: Estimated effect on H_{er} with 95% CI from the combined effect of both total ozone and cloud cover trend for the period 1991-2015.

	total ozone (95% CI)	cloud cover (95% CI)	r^2 (%)
Whole data	-1.03 (-1.25; -0.81)	-0.82 (-0.94; -0.70)	51
Winter	-0.85 (-1.22; -0.48)	-1.02 (-1.39; -0.65)	42
Spring	-1.41 (-1.81; -1.01)	-0.84 (-1.06; -0.62)	68
Summer	-1.38 (-2.09; -0.67)	-0.56 (-0.76; -0.36)	55
Autumn	-0.98 (-1.49; -0.47)	-0.92 (-1.19; -0.65)	51

Table 7: Estimated effect on H_{er} with 95% CI from the combined effect of total ozone and cloud cover trend for two sub-periods: 1991-2004 and 2004-2015.

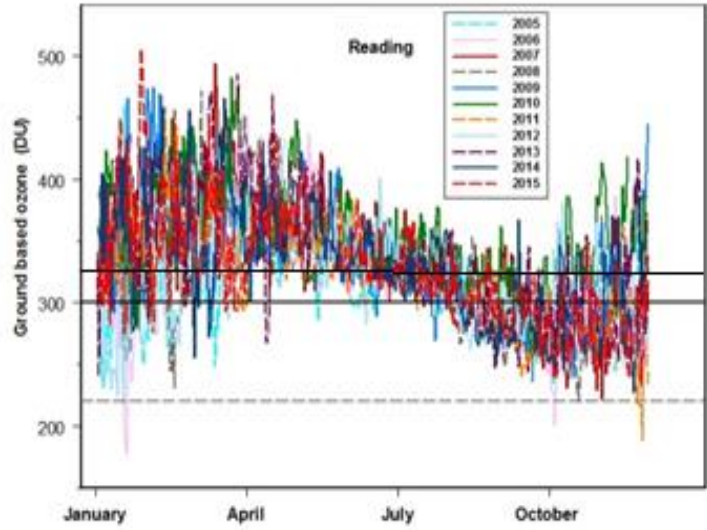|	1991-2004			2004-2015		
	cloud cover (95% CI)	total ozone (95% CI)	r^2 (%)	cloud cover (95% CI)	total ozone (95% CI)	r^2 (%)
Whole data	-0.97(-1.13; -0.81)	-0.79 (-1.26; -0.68)	55	-0.65 (-0.85; -0.45)	-1.25 (-1.56; -0.94)	49
Winter	-0.96 (-1.45; -0.47)	-0.55 (-1.14; 0.04)*	31	-0.89 (-1.34; -0.44)	-1.45 (-1.92; -0.98)	67
Spring	-1.04 (-1.26; -0.82)	-1.26 (-1.67; -0.85)	82	-0.66 (-1.01; -0.31)	-1.36 (-1.99; -0.73)	63
Summer	-0.73 (-0.97; -0.49)	-1.17 (-1.56; -0.78)	70	-0.41 (-0.76; -0.06)	-1.45 (-2.70; -0.20)	41
Autumn	-1.00 (-1.31; -0.69)	-1.03 (-1.64; -0.42)	64	-0.77 (-1.24; -0.30)	-0.97 (-1.75; -0.19)	37

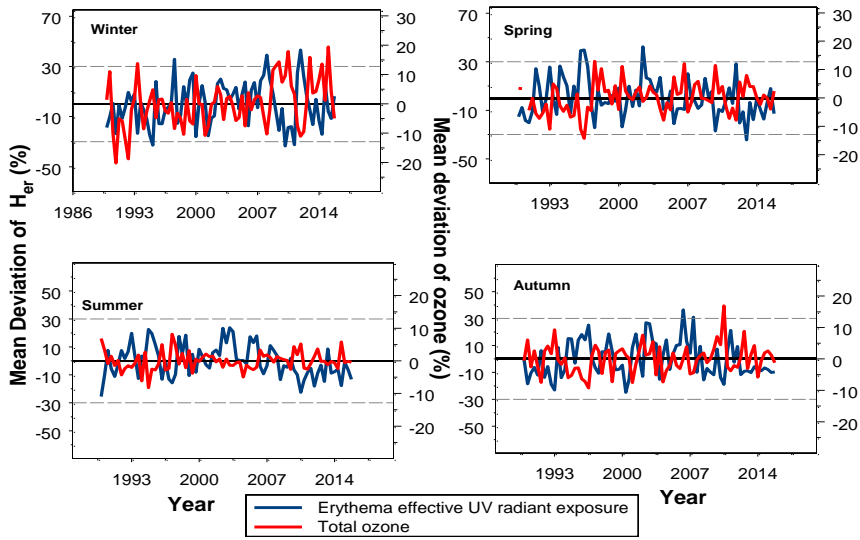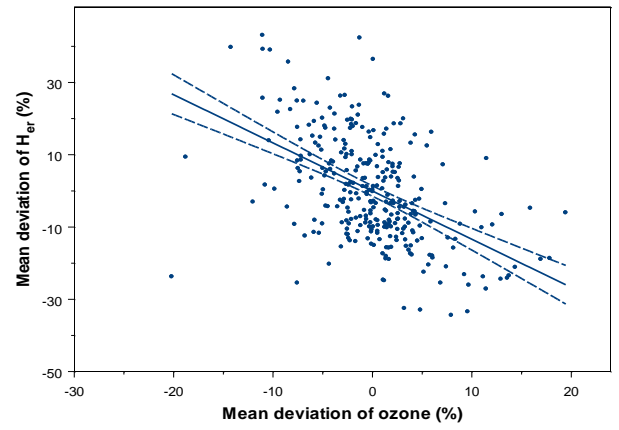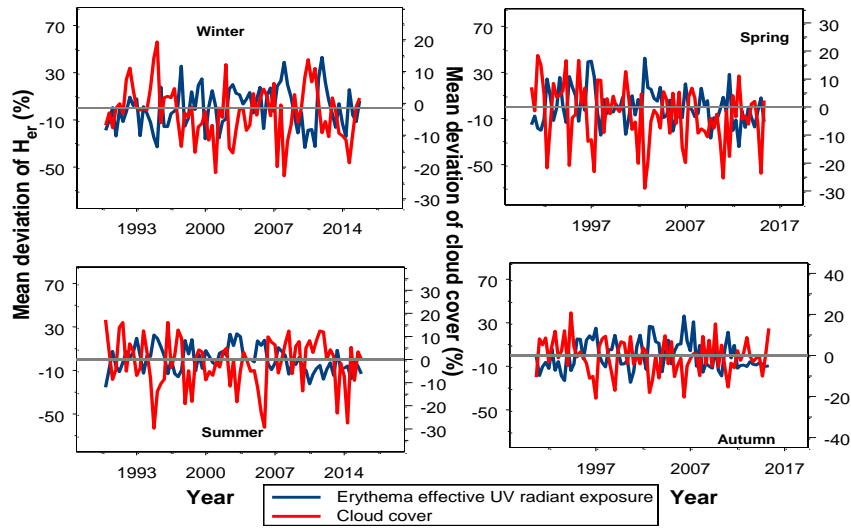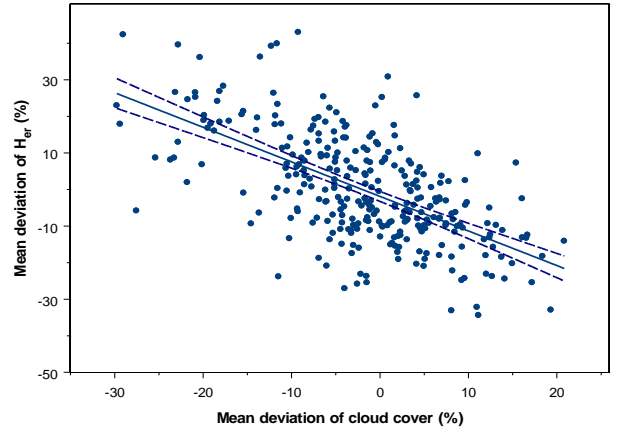*: p-value=0.07

Figure 1:
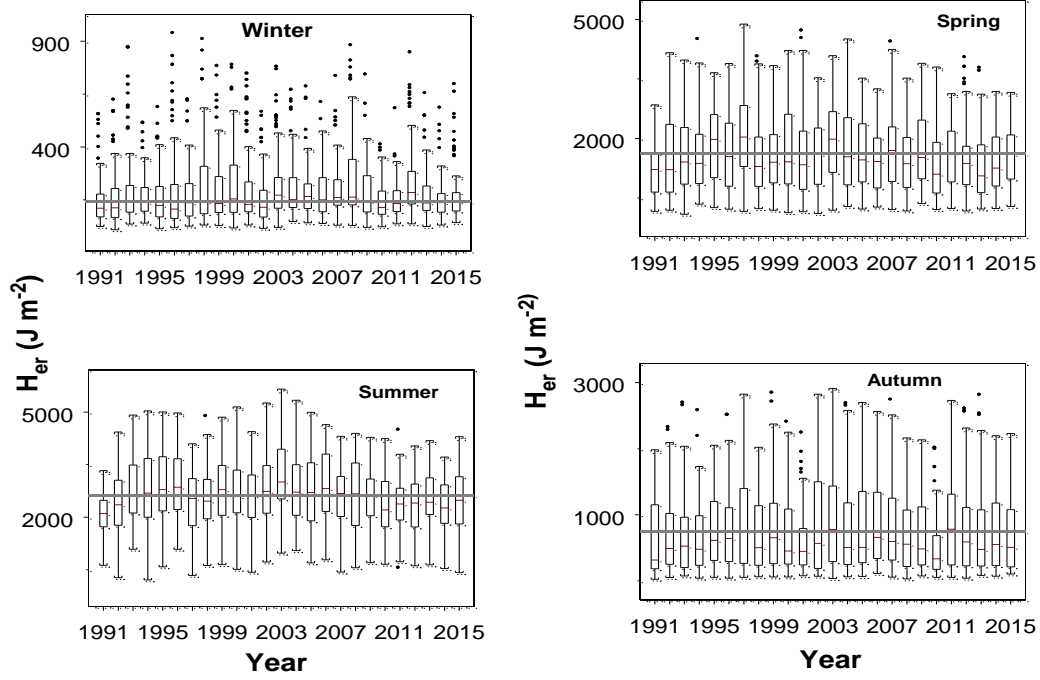
Figure 2:

(a)

(b)

Figure 3:

(a)

(b)

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Figure 4:

(a)

(b)

Figure 5:

(a)

(b)

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