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

Nezahat Hunter, Rebecca. J. Hooke, Michael P. Higlett, John B. O'Hagan and Richard G.E. Haylock

Public Health England, Chilton, Didcot, Oxfordshire, OX11 0RQ United Kingdom

Correspondence to: Nezahat Hunter (Nezahat.Hunter@phe.gov.uk)

Abstract

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

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

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

1 Introduction

Ultraviolet radiation (UVR) is only a small portion of the radiation we receive from the sun, but has become a topic of increasing concern because of the harmful health effects it can cause. Stratospheric ozone is a naturally-occurring gas that filters the sun's ultraviolet (UV) radiation. It absorbs most of the shorter wavelength UV-B radiation, whereas longer wavelength UV-A radiation mostly passes through the ozone layer and reaches the ground (WMO 2014). However, in the mid-1970s it was discovered that the release of man-made chlorine-containing chemicals could cause stratospheric ozone depletion. In subsequent years temporary ozone holes appeared over the Antarctic and to lesser extent in the Arctic (Farman et al., 1985). It was also observed that stratospheric ozone depletion also extended over populated areas, particular in spring when the ozone layer over Antarctica is dramatically thinned over Australia (Gies et al., 2013). Since the late 1970s, the effects of ozone depletion on UVR have been the subject of a large number of studies published in the literature. These studies have demonstrated that the ozone level decreased up to the mid-1990s which resulted in an increase in the amount of UV radiation reaching the Earth's surface (WHO 2006). Concern was raised that in the long-term ozone depletion would result in significantly increased UVR which in turn may result in increased incidences of skin cancers, particularly melanoma. An increase in UVR can also result in an increase in sunburn, ocular pathologies, premature skin aging and a weakened immune system (UNEP 2010; WHO 2006; AGNIR 2002; Norval et al., 2011, Lucas et al., 2010). However, it is known that exposure to UVR can be beneficial to health by producing vitamin D, which promotes healthy bones and may help in the prevention of certain diseases including heart diseases and cancers (Holick 2007, McKenzie et al., 2009, Young 2009, Epplin & Thomas 2010).

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

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

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

2 Materials and methods

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

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

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

2.2 Total Ozone

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

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

2.3 Cloud cover

The HadISD dataset was created by the Met Office at the Hadley Centre in the UK, which used a sub-set of the station data held in the Integrated Surface Database (ISD) (Dunn et al., 2012 & 2014). The HadISD dataset comprises various selected climate variables, including total cloud cover data that were recorded in various weather stations globally, including in the UK for 1931–2016 (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 of the monthly mean deviation of H_{er}, total ozone or cloud cover data 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 in place of a parametric linear regression analysis, which can be used to correct temporal autocorrelation and to test if the slope of the estimated linear regression line differs significantly from zero (Mann 1945, Kendall 1975, Helsel and Hirsh 1992). If a significant trend was found from the MK test, the rate of change was calculated using the Sen's slope (SS) estimator from nonparametric method (Helsel and Hirsh 1992). If the results of non-parametric analyses were similar to those results obtained by linear regression, the results from non-parametric analyses are not presented.

The relationship between H_{er} and total ozone and cloud cover was also examined using Analysis of Variance (ANOVA) to obtain information about levels of variability within a regression model and to form a basis for tests of significance. The correlation coefficient value (r^2) was calculated to determine a measure of the strength of the relationship between H_{er} and cloud cover and ozone and to quantify how much of the total variation in H_{er} could be explained by ozone or cloud cover. A significance level $p<0.05$ was considered statistically significant.

3 Results

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

Summary statistics from the daily H_{er} are presented in Table 1. Over 25 years H_{er} ranges from 10 J m^{-2} (measured on 9 January 1992, 18 days after winter solstice) to 5655 J m^{-2} (measured on 20 June 2003, at the summer solstice) with a mean of 1303 J m^{-2}.

Figure 1 displays the distribution of the daily H_{er} using boxplots for each season at Chilton. Each box shows the lower 25% quartile Q1, and upper 75% quartile Q3 and central line is the median. The whiskers extended in each direction from the box starts from Q1 to the smallest data point and the upper whisker from the Q3 to the largest data point that is away from the box and measurements falling outside whiskers are possible extreme data values that are outside the typical pattern of the other data points. The observed results show that H_{er} are the highest in the summer months and the lowest in winter months, while during spring and autumn months, H_{er} may change rapidly day to day (Fig.1). The extreme data points were mostly observed in winter and a few in other seasons could be related to natural variability in factors that affect H_{er} including total ozone, cloud cover, aerosols, and climate at this site. In particular, extremely low total ozone periods, which often occur in late winter and early spring, while H_{er} does not exceed normal-ozone summer values. After 2007, in particular in spring, summer and autumn it appears that H_{er} values are well below their expected mean values.

Figure 1: Boxplots of the daily H_{er} data for each season at the Chilton site between 1991 and 2015 (grey solid line represents the mean value for each season)

Figure 2a shows the monthly mean deviation of H_{er} values expressed as percentages. A consistent rise between 1991 and 2003 with a clear peak in 2003 when the H_{er} values were the largest recorded at Chilton over the 25 year period. Thereafter, H_{er} values appeared to decrease. Fig. 2b also shows the mean deviation data in H_{er} for each of the four seasons over the 25 year period. Winter and spring exhibited greater variability in comparison with summer and autumn. During winter months, peaks in H_{er} were observed in various years; however, H_{er} in winter was very low (Table 1) thus the effect on annual exposure is expected to be very small. Among spring months, clear peaks are observed in March 2003 and also in March & April 1997 (Fig. 2b). For summer, H_{er} levels vary less in comparison to other seasons, although summer has the highest H_{er} levels overall and the effect on annual exposure is large. For autumn, peaks are observed in November 2006 and 2007 and the variability in the last few years was stable.

Figure 2: Monthly mean deviation of H_{er} data at Chilton (1991-2015) with trend lines (a) all season combined (b) seasonal.

The regression analyses of H_{er} data indicate that the best fitting single linear trend covering the whole period 1991-2015 has a downward slope but that this slope is not statistically significantly different from a constant value over the period (p=0.27). Two further models were also examined. One is a linear-quadratic function, (LQ), a 2nd degree polynomial, which allows for more gradual variation in the monthly H_{er} across the 25 year period and a second model consists of two linear trends with a node to allow for a single change in linear trend over the 25 year period. Figure 2a also shows the results of fitting these four models to the monthly mean deviation of H_{er} data.

The best fitting model was the last of these which had two linear trends that describe an increasing trend from 1991 to 2003 and a decreasing trend thereafter which defines the year 2003 as the node between two trend lines. The nodal year appeared to be influenced by the particularly high observations in 2003 (Fig. 2a). Thus, in order to avoid bias that might be caused by the highest H_{er} values observed in 2003, the year 2004 was chosen to be the nodal point in preference to 2003.

Based on the results of the initial model fitting to the whole period statistical analyses were also carried out to investigate the long-term variability of H_{er} for two sub-periods (1991-2004 and 2004-2015). Table 2 presents the estimated linear slopes in percentage change per year in H_{er} with 95% confidence intervals (CI) and p-values of the associated significance tests. There is evidence of a statistically significant increasing trend for the first period (1991- 2004) with a mean rate of 1.01% per year (y^{-1}) (95% CI: 0.75%; 1.27%, p<0.001) and a decreasing trend for the second period (2004-2015) with a mean rate of 1.35% y^{-1} (95% CI: 1.98%; 0.72%, p<0.001) based on all the data. While there was evidence of autocorrelation, the results of the non-parametric analyses, which would not be influenced in the same way by the autocorrelation, were similar to those in Table 2 so they are not presented here.

For seasonal trends, the only significantly increasing linear trend was seen in winter from 1991-2004; however, H_{er} level in winter was very low and contributed only a small proportion of the total H_{er} overall. The highest levels of H_{er} observed in summer did not show any significant linear trend for 1991-2004 as the H_{er} levels were steady for this period. The absence of a significant trend in spring for this period might be partly explained by the influence of fairly stable H_{er} levels seen between 1998 and 2002 (Fig.2b). Across the same period in autumn, the trend was found to be approaching statistical significance (p=0.07). For 2004-2015, the estimated trend slope was negative for each season, but the trend was only statistically significant in summer and autumn, (Table 2).

3.2 Total ozone

From the combined Camborne and Reading dataset covering the period 1991-2015 total ozone ranged from a low of 177 DU (measured in January 2006 in Reading) to a high of 524 DU (measured in February 1991 in Camborne) with an overall mean value of 327 DU. The distribution of the daily total ozone values are presented in box plots for the period 1991-2015 for each season (Fig. 3a). The mean value is shown with a grey dashed line and the bold line at 300 DU shows the average amount of total ozone in the atmosphere (http://ozonewatch.gsfc.nasa.gov). Both graphs displayed a large spread of total ozone measurements. The data appear to be varying year to year and there is a much larger spread of total ozone values in winter and spring compared to summer and autumn. The extreme data points are therefore not likely to be erroneous readings. The total ozone values were low in autumn and early winter days with a few exceptional cases in March, April and August. Similarly, the maximum total ozone values were mostly found in late winter and in early spring (Fig. 3b); the solid black line indicates the overall mean value (327 DU) and the grey dashed line represents the baseline ozone level of 220 DU which was not observed over Antarctica prior to 1979 (https://ozonewatch.gsfc.nasa.gov).

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

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

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

The comparable analyses of the seasonal data are also presented in Table 3. The trend for the total ozone data was only statistically significant in winter over the period 1991-2015 (0.43% y^{-1}; 95% CI: 0.19%; 0.67%, $p<0.001$). While there was evidence for autocorrelation in the residuals, Sen's slope trend estimates were found to be very similar to the slope estimates obtained by linear regression. However, there were no statistically significant positive trends identified for any of the seasons either for the first period 1991 to 2004 or for the second period 2004 to 2015 and the 95% confidence interval lower bounds were negative for all seasons. In particular, in summer, a statistical analysis showed that ozone trend was not statistically significant with there being very small when H_{er} was high over any time periods. The significant ozone trends in winter will influence the very low H_{er} at that time of year, but have a little practical influence on overall 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 pattern is not surprising as an increase in cloudiness tends to reduce H_{er} and as a result the RAF become less negative in winter. A test of heterogeneity in RAF values was tested between seasons; there was not statistically significant difference in the RAF values between summer, spring and autumn ($p=0.82$) and also between winter and autumn ($p=0.35$).

Across both sub periods, the inverse correlation between H_{er} and total ozone was statistically significant for all seasons except during winter for the period 1991-2004 (p=0.12). In contrast, for the period 2004-2015, the correlation was stronger in winter (52%) and also in spring (48%) than that in summer (24%) and autumn (17%), although the H_{er} level in winter is very low and the effect of total ozone is negligible. The test heterogeneity in RAF values were also performed for both periods between seasons; there was also no statistically significant differences in RAF values between spring, summer and autumn (p=79) for the period 1991-2004. Although th RAF value in autumn was less negative than that in winter for the period 2004-2015, there was no differences in RAF values between all seasons (p=0.53). The results for both study periods also showed the lowest RAF values were estimated in spring and summer. The daily mean value of H_{er} in summer here is about 14 times larger than in winter.

3.4 Erythema effective radiant exposure (H_{er}) and cloud cover

The long term changes in H_{er} in all weather conditions also differ according to variations in cloud cover. The regression analysis of all the cloud cover data showed a statistically significant downward linear trend with a mean rate of 0.19% y^{-1} (95% CI: -0.34%; -0.04%, p=0.01). When the data for each season was considered separately, a statistically significant downward linear trend was only found in spring (p=0.025) although the trend slope was negative for the other three seasons. The regression analysis of cloud cover for the first period (1991-2004) also showed a statistically significant downward linear trend of 0.68% y^{-1} (95% CI:-1.03; -0.33, p=0.0002), but for 2004-2015 the downward linear trend was small (-0.04% y^{-1}) and not statistically significant (p=0.85). Seasonally, the slope estimates were negative for all four seasons for 1991-2004, but only the trends for winter and spring were statistically significant (-0.93% y^{-1}, p=0.02 and -0.72% y^{-1}, p=0.03 respectively). In contrast, for 2004-2015, there was no evidence of a trend in cloud cover for any season, although the trend estimate was negative for winter and spring, but positive for summer and autumn.

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

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

Table 5 shows the results of the regression analyses of the monthly mean deviation of H_{er} data against cloud cover and the correlation coefficient values to quantify the strength of the relationship by season and for all seasons together at Chilton from 1991 to 2015. A highly statistically significant inverse correlation was found for each season and for all seasons together. For the whole data over the period 1991-2015 the analysis shows a 1% increase in cloud is associated with a decrease of about 1% in H_{er}. This fitted regression line on a scatterplot (Fig.5b) indicates modest correlation between total ozone and H_{er} which was confirmed by an r^2 value of 38% (Fig. 5b) and over 62% of the variation remaining unexplained. Seasonally, the highest correlations were observed in spring (48%) and summer (46%) over the period 1991-2015.

A statistically significant negative correlation was also found for the whole data for the two-sub periods 1991-2004 and 2004-2015 (p<0.001 for both periods) and the results are also presented in Table 5. While the regression slopes were similar for both periods the strength of the correlation was moderate (48%) for the first period, but low for the latter period (27%). For the seasonal data, all regression slope values were negative and statistically significant. The correlation was strongest in spring (66%) and summer (64%) for 1991-2004, but moderate for the same seasons for 2004-2015.

3.5 H_{er}, total ozone and cloud cover

A multiple linear regression analysis was used to investigate how variation in cloud cover and total ozone considered together were associated with changes in H_{er} from 1991 to 2015. The results are presented in Table 6. The estimated slopes for both cloud cover and total ozone were negative and statistically significant for all seasons together. On average H_{er} decreased by 0.82% for each additional 1% increase in cloud cover at constant levels of total ozone. Similarly for every additional 1% increase in total ozone, H_{er} decreased by 1.03% at constant levels of cloud cover. This RAF value is slightly different than the model with no cloud cover effect adjustment, but this value did not differ statistically significant (p=0.50). The correlation coefficient (r^2) was moderate (51%) for all seasons together, of this total variation in H_{er} was explained by these two factors together, with 37% accounted for by cloud cover variation and 14% by total ozone variation.

Across the season specific analyses, the correlation between H_{er} and these two factors was highest in spring (68%) and summer (55%) (Table 6). In both instances cloud cover accounted for the largest variation in H_{er} (47% and 46% respectively), while the variation in H_{er} explained by total ozone was low (21% and 12% respectively) when cloud cover was in the model. This means that 32% and 45% of variation in H_{er} in spring and summer respectively remained unexplained. As expected, the correlation coefficient for winter was the lowest (42%) while for autumn the correlation between H_{er} and these two factors was found to be moderate (51%) with variation in cloud cover accounting for 41% and total ozone explaining only 10% of the total variation in H_{er}. There was also no statistically significant differences in RAF values between all seasons (p>0.50) except between winter and spring (p=0.02) at constant levels of cloud cover.

Table 7 shows the results from the multiple linear regression analysis for two-sub periods (1991-2004 and 2004-2015). For the first period, for every 1% increase in total ozone, H_{er} decreased by 0.79% at constant levels of cloud cover, that is RAF is -0.79. Similarly H_{er} decreased by 0.97% for each additional 1% increase in cloud cover at constant levels of total ozone. For 1991-2004, 55% of the total variation in H_{er} was explained by cloud cover and total ozone together. Cloud cover accounted for 47% of the total while total ozone explained only 8%.

In contrast, for the period 2004-2015, of the 49% of the variation in H_{er} explained by both factors, 33% was explained by total ozone and 16% by cloud cover. The RAF value (-1.25) appeared to be smaller than that RAF for the first sub period, but the test showed that there was no evidence that these values differ (p=0.44). The H_{er} level decreased by 0.65% for each additional 1% increase in cloud cover at constant levels of total ozone.

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

4 Summary and Discussion

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

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

In our previous analysis of the long-term variability of H_{er} between 1991 and 2015 at Chilton the data were divided into two separate time series data due to geophysical phenomena; one from 1991 to 1995 during which the ozone turning point in the mid-1990s because excess volcanic aerosol after the Pinatubo volcanic eruption may cause short-term ozone depletion and as a result enhanced the amount of UVR (WMO 2014) and a second from 1995 to 2015 (Hooke et al., 2017).
In contrast, this current work splits the time series according to statistical analysis; we have shown that the H_{er} data for 1991-2015 (based on a nonlinear model over the full period) were statistically better described by two linear trends; the first a statistically significant increasing linear trend value of 1.01% y^{-1} ($p<0.0001$) for 1991-2004 and the second a statistically significant decreasing trend of 1.35% y^{-1} ($p<0.0001$) from 2004 to 2015. Our finding for the first period is not consistent with our earlier result for the period 1991-1995 where a higher estimate (4.4% y^{-1}) was obtained however the earlier result should be treated with caution due to relatively short time period, 5 years, over which the trend was calculated. Our findings for the second period agree with those of our early study for 1995-2015 but the trend estimate was slightly lower (0.8% y^{-1}).

The finding in this study for the first period (1991-2004) is in good agreement with those from European studies that also reported significant increasing linear trends. At Lindenberg in Germany there was reported an increasing trend of 0.77% y^{-1} during 1996-2003, 0.85% y^{-1} for the period 1999-2004 and 1.4% y^{-1}over the period 1998-2005. The studies at Norrköping in Sweden (with a latitude of 58^{o}) and also at Bilthoven in Holland, both reported an increasing trend during 1996-2004 (1.2% and 0.86% y^{-1}respectively) based on solar zenith angles (SZA) of 60^{o}, but the trend was higher (1.7% y^{-1}) at Bilthoven for the period 1998-2005 when the noon values of the erythemal UV radiation were used (Bais et al., 2007). The study at the Hoher Sonnblick site in Austria (Fitzka et al., 2012) showed a significant upward trend in the erythemally weighted irradiance for the period 1997-2011 with a range from 0.84%±5.2% y^{-1} at 45^o SZA to 1.26%±0.36% y^{-1} at 65^o SZA under all weather conditions. However, a smaller and less significant result was seen at wavelengths of 305 nm (between -0.76%± 1.13% y^{-1} and 0.79%± 0.73% y^{-1}, depending on SZA). The study based at Reading in the UK found a significant increasing linear trend (0.66% per year) for the period from 1993 to 2008 based on the midday values of UV index (Smedley et. al 2012).

The trend in H_{er} in this study over the second period (2004-2015) at Chilton is consistent with values derived for the averaged UV-B data over Canada, Europe and Japan that showed statistically significant evidence of a reduction in UV-B for the period 2007-2011 with the slope estimates ranging from -1.5% to -2% under cloudless conditions (Zerefos et al., 2012). Our findings are also in good agreement with the results of Fountoulakis et al. (2016) at Thessaloniki (latitude of 40^o N) in Greece, where a turning point in the trends of UV irradiance is reported as being in 2006; a statistically significant increasing trend of 0.71% ±0.21% y^{-1} for the period 1994-2006 and a decreasing trend of 0.33% ± 0.32% y^{-1} from 2006 to 2014 but the trend was not statistically significant. It appears that there is a similar behaviour of the trend in the UV irradiance between this UK study and the Greek study, although these countries differ significantly in terms of climate and location. However, a recent study at Uccle in Belgium covering the period 1991-2013 which is similar to that examined in this study found a strongly statistically significant increasing linear trend of 0.7% y^{-1} (De Bock et.al. 2014). In comparison, our results for the period 1991-2015 found a non-significant downward trend.

When the H_{er} data for each season were analysed separately, a statistically significant increasing trend was only found during winter for the first period 1991-2004 despite large inter-month variability and contributes only a small fraction of the annual cumulative H_{er} level in the UK. Much of this significant result might be caused by the excess of low total ozone events observed in winter (detail will be discussed in the next section). However, there was no significant linear trend in H_{er} in either spring or in summer at Chilton. The absence of a significant trend in spring for this period might be due to higher values of total ozone level over the same period.

For the second period 2004-2015, a linear downward trend in H_{er} was observed for all four seasons, but the trend was only statistically significant in summer and autumn. The results of the current study are comparable with those of the Belgian study at the Uccle site from 1991 to 2013, that showed the largest statistically significant increasing trend in H_{er} in spring but a negative trend in winter, albeit not statistically significant (De Bock et. al 2014). In addition, the Austrian study at the Hoher Sonnblick site for the period 1997-2011 also found that the largest and most significant linear trends were during winter and spring.

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

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

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

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

For both sub-periods (1991-2004 and 2004-2015) a statistically significant inverse correlation was observed between H_{er} and total ozone, but the amount of variation in H_{er} explained by that of total ozone was low (18% and 33% for each time period respectively). Our estimates of the size of the trend in ozone are smaller than those reported in the study by Zerefos et al., (2012) based on averaged total ozone and UV-B data over Canada, Europe and Japan. That study found that for the period 2007-2011 the effect of increasing total ozone on UV-B values was about -4% when aerosol optical depth (AOD) was factored into the model. The Belgian study (De Bock et al., 2014) also reported a greater effect of total ozone on the erythemal UV dose (-5%) during the period 1991-2008 when measures of global solar radiation and AOD were taken into account. The Reading study, however, did not find any correlation between the surface UV radiation and total ozone for the period 1993-2008. The authors suggested that the majority of the variability in UV radiation was due to changes in cloud cover and other effects (Smedley et al., 2012).

Examining our data on a season by season basis over the whole period from 1991 to 2015, we found a highly negative slope estimate for each season. However there were some differences in how much of the variation in H_{er} that was explained by total ozone across the seasons. In spring and summer the variability explained was moderate at 41% and 34% respectively but in winter and autumn it was considerably lower at 19% and 21% respectively.

Restricting the data to the first period (1991-2004), we also saw the greatest impact of total ozone on H_{er} in spring (37%) and summer (42%) but for the second period (2004-2015), the impact was bigger in winter (52%) and spring (48%) than that in summer (24%) and autumn (17%). This is mainly due to the strong inverse relationship between H_{er} and ozone that was observed during spring and winter in the period 2008- 2015 (63% and 56% respectively).

4.3 Erythema effective UV radiant exposure (H_{er}) and cloud cover

Cloud cover can have a marked impact on the amount of UVR that reaches the earth's surface. An increase in cloud cover usually results in a reduction of UV radiation below the clouds. Whilst UVR can pass through thin and broken clouds thick clouds tend to reflect, absorb or scatter UV radiation. Puffy, fair-weather clouds deflect rays and can actually increase the UV radiation reaching the earth's surface (Alados-Arboledas et al., 2003). Our analysis of cloud cover variation showed a statistically small significant decreasing trend of -0.68% y^{-1} ($p<0.001$) for the first period (1991-2004) but no significant trend for the second period (2004-2015) although the estimated slope was negative (-0.04% y^{-1}). These findings agree partly with other studies that reported a decrease in cloud cover (Norris and Slingo, 2009; Eastman and Warren, 2013) and also with those that did not find any evidence of a decreasing trend in cloud cover. For example, the studies at the Hoher Sonnblick site in Austria over the period 1997-2011 (Fitzka et al., 2012) and in the study examining data from Europe, Canada and Japan for the period 1995-2011 (Zerefos et al., 2012).

Examining our data on a season by season basis, the only statistically significant trends in cloud reduction was observed in spring and winter during the period 1991-2004. For subsequent years there was no evidence of a trend in observed cloud cover in any season. These observations agree with the findings from the Austria study at the Hoher Sonnblick for 1997-2011 (Fitzka et al., 2012).

The inverse correlation between H_{er} and cloud cover was also found to be strongly statistically significant for both sub-periods. However, while about half the variation in H_{er} was explained by the cloud cover variation in the first period this fell to just over one quarter for the second period.

The same data examined on a season by season basis showed, for the first period, just over half the variation in H_{er} explained by the inverse relationship with cloud cover for spring, summer and autumn but only just over one quarter for winter, although the slope of the relationship did not vary greatly with season. For the second period, the variation in H_{er} explained by cloud cover dropped below 50% for all seasons and in particular the value for autumn (26%) reduced to below that for winter (29%). Although, the Austrian study at the Hoher Sonnblick did not study the correlation between cloud cover and UV measurements, the authors reported that the total cloud reduction of 1.04% y^{-1} was evident for UV measurements at SZA 55^{o} for the period 1997-2011.

4.4 Erythema effective UV radiant exposure (H_{er}), total ozone and cloud cover

Given that we found clear evidence that variation in H_{er} could be partially explained by variation in total ozone and cloud cover separately we considered their combined effects. Over the whole period 1991- 2015 half of the variation in H_{er} could be explained by these two factors with the changes in cloud cover alone accounting for 37% of the variation in H_{er}, while the total ozone variation explained 14%. The unexplained half of the H_{er} variation may be attributed to other factors such as atmospheric aerosol, air pollution or climate. The RAF value was found to be -1.03 at constant levels of cloud cover. Although this RAF value was slightly different than that period between 1991 and 2004 (-0.79) and for 2004-2015 (-1.25) when cloud cover effect in the model, but there was no statistically significant differences in the RAF values (p=0.66). These RAF results are also consistent with the RAF values in the US study in which impact of clouds on the RAF was determined and the RAF ranged from a low of -0.80 to a high of -1.38 (Hall 2017).

The effects of aerosols on surface UV irradiance have also been studied widely in addition to ozone and cloud in Europe (Román et al., 2015, De Bock et al., 2014, Zerefos et al., 2012, Fitzka et al., 2012). Our study, however, did not take account of the effects of aerosols because Chilton is situated in a very rural location in South Oxfordshire in the UK and the levels are generally very stable. The aerosols optical depth (AOD) trend in London was reported in the range of 0-0.004 per year decrease between 2003 and 2015 (Provençal et al., 2017).

Aerosols can affect ground level UV irradiances directly through absorption and scattering solar radiation back out to space, reducing the amount of solar radiation reaching the surface of the Earth. Aerosols can also affect indirectly which are related to modify cloud formation. Atmospheric aerosols originate from both natural sources (such as dust) and from anthropogenic sources – such as air pollution from industry and traffic producing more pollution and atmospheric aerosol, in particular in urban areas. Alpert et al. (2012) reported that aerosols optical depth (AOD) trends declined over the largest cities in Europe during the period (2002-2010) owing to increasing air quality due to environmental regulations (2012). Nevertheless, the influence of the aerosols on UV irradiance has not been fully understood due to their high spatial and temporal variability (WMO 2007).

In this work, the combined effects on a season by season basis the biggest proportion of variability in H_{er} explained by total ozone and cloud cover together was for spring (68%) while the smallest proportion was in winter for period 1991-2015. In each season cloud cover explained far more of the variability than did total ozone, based on the additive linear regression model was used.

The combined effects of total ozone and cloud cover on H_{er} was also assessed using two linear trends with a node at 2004. Over the first period the proportion of H_{er} variability explained by these two factors rose a small amount (4%) but fell slightly for the second period (2%) compared to the proportion when the entire period 1991 to 2015 was considered as a whole.

A major difference was seen in how much of the variability was explained by each factor across the two periods. In the first period cloud cover variation accounted for a lot more of the explained variability compared to ozone (47%:8%) whereas for the latter period the proportions were (16%:33%). This is because for the second period significant correlation between total ozone H_{er} observed in winter and spring during 2008-2015 had a bigger impact on H_{er} than that in cloud cover for the same period.

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

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

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

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

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

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

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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 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:

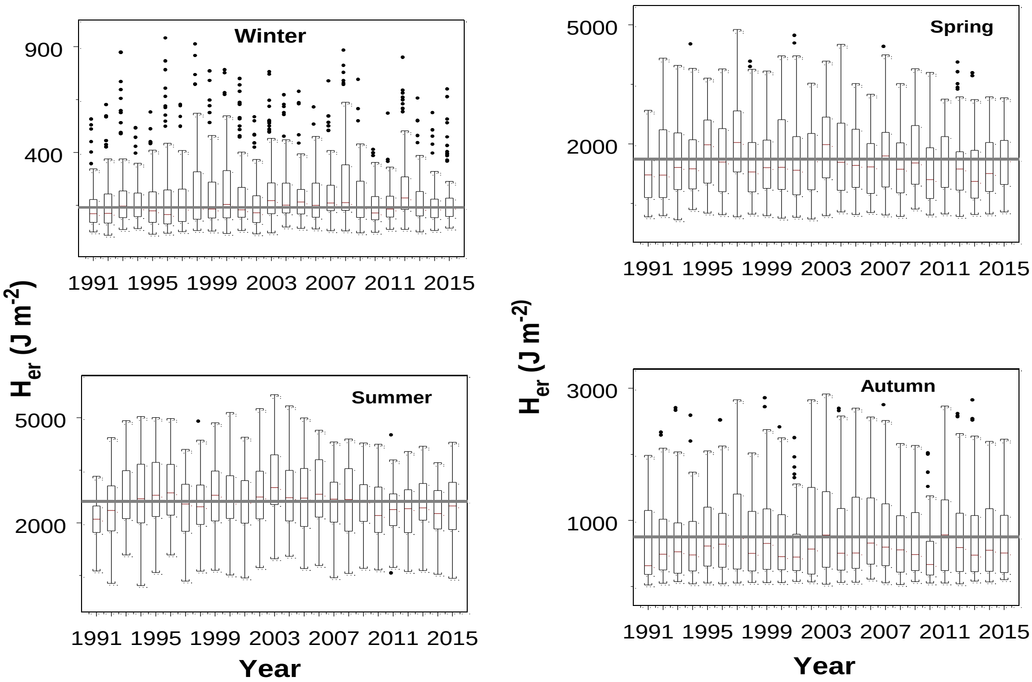
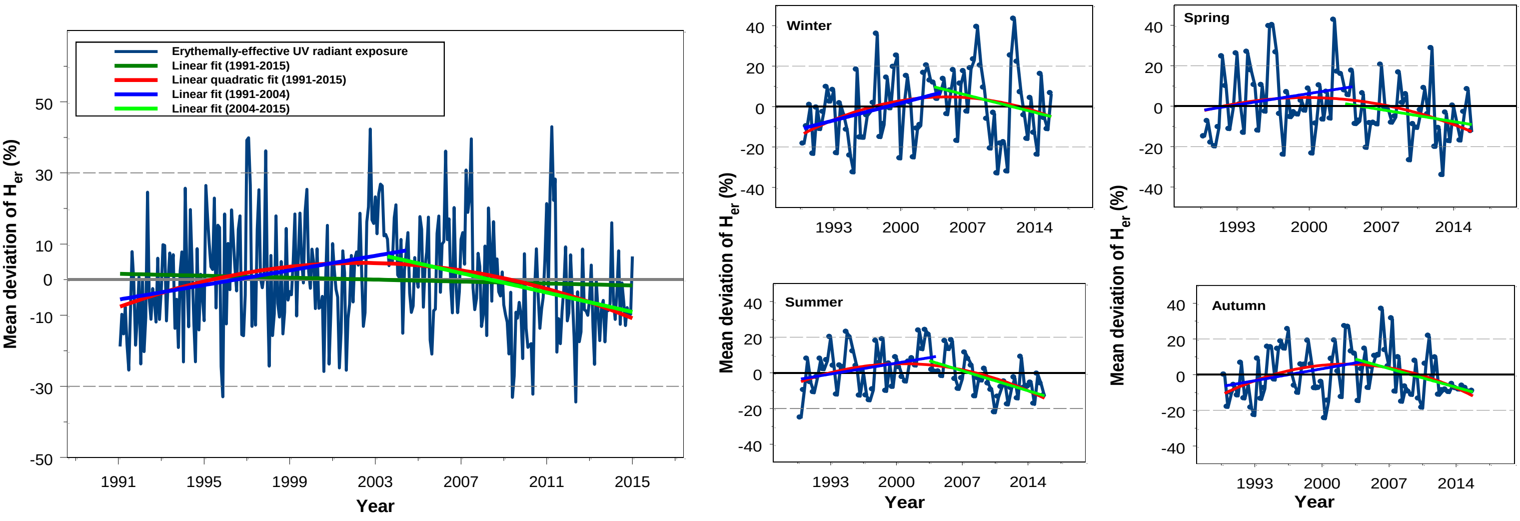

Winter
900
400
1991 1995 1999 2003 2007 2011 2015
Spring
5000
2000
1991 1995 1999 2003 2007 2011 2015
H_{er} (J m^{-2})
Summer
5000
2000
1991 1995 1999 2003 2007 2011 2015
Year
H_{er} (J m^{-2})
Autumn
3000
1000
1991 1995 1999 2003 2007 2011 2015
Year

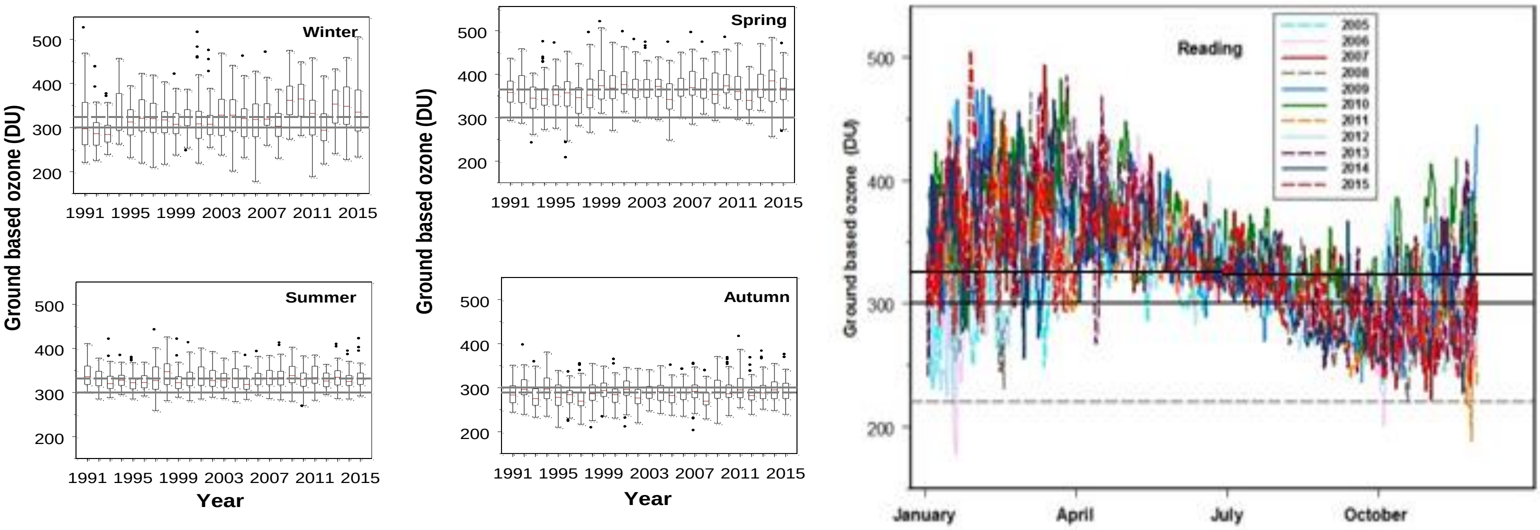
Figure 2:



Erythemally-effective UV radiant exposure
Linear fit (1991-2015)
Linear quadratic fit (1991-2015)
Linear fit (1991-2004)
Linear fit (2004-2015)
Mean deviation of Her (%)
Year
Winter
Spring
Summer
Autumn

(a) (b)

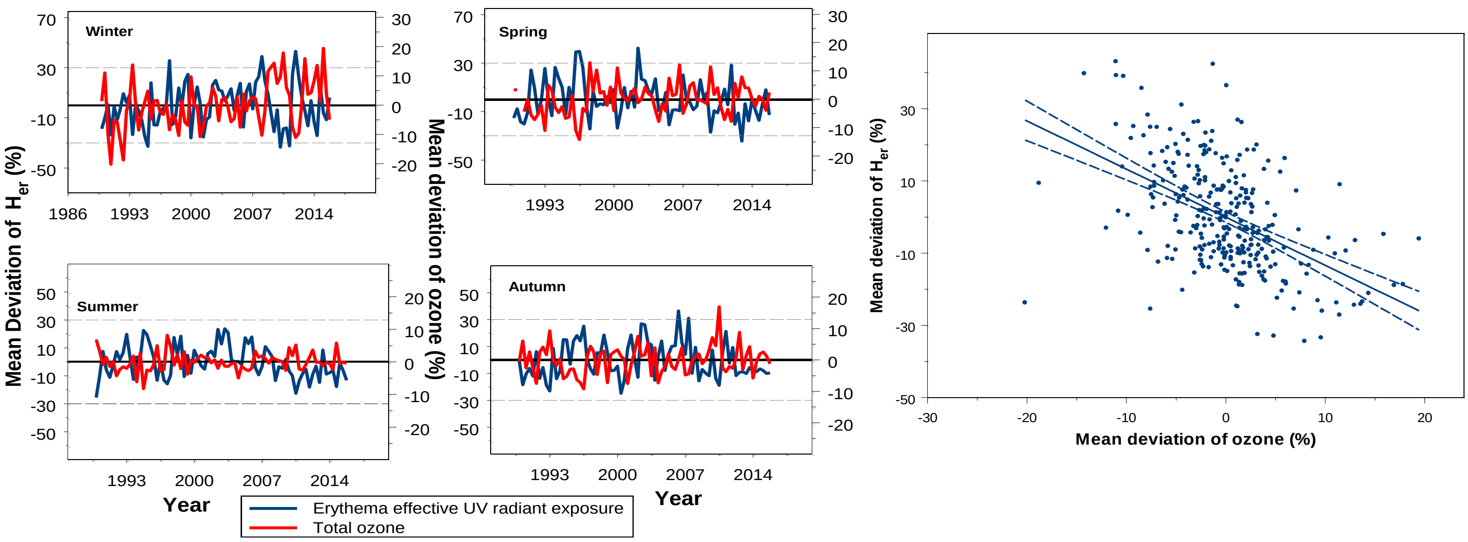
Figure 3:



Winter
Spring
Summer
Autumn
Ground based ozone (DU)
Year
1991 1995 1999 2003 2007 2011 2015
Reading
2005
2006
2007
2008
2009
2010
2011
2012
2013
2014
2015
January
April
July
October

(a) (b)

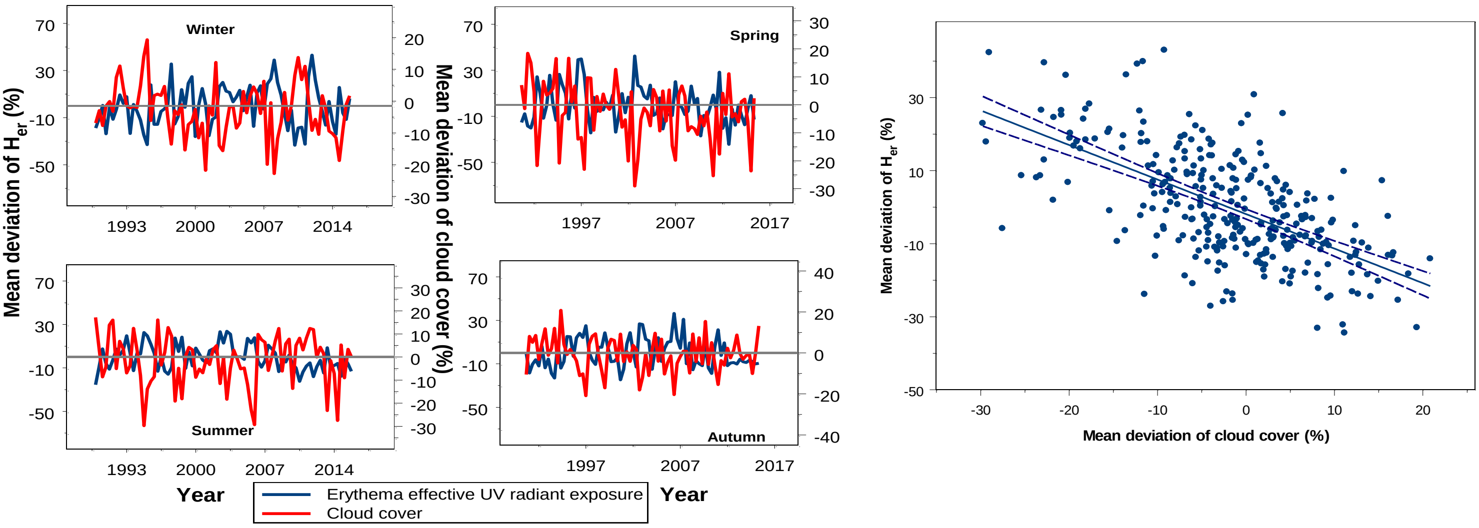
Figure 4:



Winter
Spring
Summer
Autumn
Mean Deviation of H_{er} (%)
Mean deviation of ozone (%)
Year
Erythema effective UV radiant exposure
Total ozone
Mean deviation of H_{er} (%)
Mean deviation of ozone (%)

(a) (b)

Figure 5:


Winter
Spring
Summer
Autumn
Mean deviation of H_er (%)
Mean deviation of cloud cover (%)
Year
Erythema effective UV radiant exposure
Cloud cover
Mean deviation of cloud cover (%)

(a) (b)