

## **Response to reviewers**

Atmos. Chem. Phys. Discuss., 15, 1869-1914, 2015: “**Trends and drivers of ozone human health and vegetation impact metrics from UK EMEP supersite measurements (1990-2013)**” by Malley et al.

We thank the reviewers for their time spent reviewing our manuscript and for their recommendations for publication after attention to some revisions. Below we respond to each comment individually and indicate the revisions we have made to the manuscript. Reviewers’ comments are in italics, additional tables and figures are located at the end of the response. Page and line numbers below refer to the original version of the paper published on ACPD.

### **Reviewer #1:**

*This paper presents a detailed analysis of the changes over a 20 year time period in key indices used to assess health and vegetation impacts of ozone at the Harwell EMEP supersite in the south of the UK, including an important evaluation of the role of regional precursor emission control and changes in hemispheric background levels. This is supplemented by a comparison with values of the same indices over more recent years at a second supersite in Scotland. The very detailed analysis of trends in both health and vegetation indices, and by implication impacts, at Harwell clearly demonstrates the different trends in the various indices, and highlights the effect of considering different threshold indices. The paper also very nicely identifies the importance of hemispheric background concentrations and regional ozone production as underlying drivers of change. Both health and vegetation impacts of ozone are of global importance and hence these are important messages of wider importance for assessment of the future benefits of different measures to reduce these ozone impacts. I therefore recommend publication. However, I have a number of comments about the framing and detail of the paper which I suggest the authors consider before final publication.*

#### *Specific comments*

**Reviewer 1 comment:** *The wording of the first para of the abstract implies that data from both sites was important in identifying trends in impact indices and their drivers, but in fact only the time-series from Harwell is long enough to allow analysis of trends. Given that the authors themselves identify (p6, l31) that Harwell is only representative of rural sites within 120km of London, this does place a constraint on the wider relevance of their findings. More discussion of the limitation that this places on wider conclusions about trends and drivers throughout the UK and other parts of NW Europe would be useful.*

Response: We acknowledge that the use of the term ‘trends’ in the first paragraph of the abstract was not sufficiently defined. While long-term temporal trends were only quantified at Harwell, this paper also reports on spatial trends in O<sub>3</sub> health and vegetation impacts across the UK through its comparison between Harwell and Auchencorth. The first sentence of the Abstract has therefore been changed as follows to emphasise that ‘trends’ refers to both spatial and temporal trends:

**Changed text:** “Analyses have been undertaken of the spatial and temporal trends and drivers of the distributions of ground-level O<sub>3</sub> concentrations associated with potential impacts on human health and vegetation using measurements at the two UK EMEP supersites of Harwell and Auchencorth.”

The second and third paragraphs of the Abstract make it clear that the Harwell dataset was used to assess long-term temporal trends, and Auchencorth was used for comparison with Harwell over a shorter (2007-2013 time period).

We have been very clear in defining the geographical representativeness of the sites by reference to previous work that placed these two UK sites in a European context according to measured O<sub>3</sub> variation (Malley et al., 2014). As Harwell is representative of south-east England, the motivation for the spatial comparison between Harwell and Auchencorth (representative of the remainder of the rural UK (Malley et al., 2014)) was to evaluate how the state and drivers of O<sub>3</sub> impacts identified at Harwell compared with the remainder of the UK. While the study period is shorter at Auchencorth, the spatial comparison highlights important policy relevant conclusions about the differences in ozone impact accumulation across the UK. These are emphasised in both the Abstract and Conclusions, which state that, in general, O<sub>3</sub> impacts are determined to a greater extent by hemispheric background concentrations at Auchencorth than at Harwell; and that therefore, across the whole UK, reduction in hemispheric background concentrations is required for substantial improvement. An important exception is during regional O<sub>3</sub> episodes, when elevated POD<sub>Y</sub> is accumulated at Auchencorth compared to Harwell; hence vegetation impacts at Auchencorth would improve to a larger degree than at Harwell from regional O<sub>3</sub> precursor emissions reductions. Again, this contrast between O<sub>3</sub> impacts in south-east England and the remainder of the UK is noted in the abstract and conclusions.

**Reviewer 1 comment:** *The chemical climatology idea is a key conceptual element of the introduction of the paper. However, it subsequently only provides a template for the detailed annexes, and it provides little of the framework for the body of the text and data analysis. Either this element should provide a stronger framework throughout the paper, or it should be omitted, as the key messages of the paper are not really connected to this concept and would be equally valid without it.*

Response: The focus of the Results and Discussion section is on what is learned through the application of the chemical climate approach (impact, state and drivers) on the spatio-temporal changes in the conditions producing O<sub>3</sub> impacts. The presentation and discussion of the material in the Results and Discussion is divided into sub-sections that map onto the chemical climatology framework. The human health (Section 3.1) and vegetation (Section 3.2) sections are further divided into two subsections which consider separately changes in temporal chemical climate phase at Harwell between 1990 and 2013 (Sections 3.1.1 and 3.2.1 for health and vegetation respectively), and spatial changes in chemical climate phase between Harwell and Auchencorth (Sections 3.1.2 and 3.2.2). As the reviewer notes, our Introduction and Methods sections already contain detailed explanation of how the chemical climatology framework was used, including citation to another publication, to which the reader can refer. However, to emphasise more the chemical climatology framework used in the derivation of the results and discussion statistics (but yet retain focus in these sections on the key

findings, as recommended in a further comment below), the following two explanatory sentences have now been added to the start of Sections 3.1 and 3.2.

**Additional text:**

P. 1879 In 23: “This section presents two analyses of the impact, state and drivers of the chemical climatology framework (Figure 2, steps 1-5); specifically, changes in chemical climate phase (Figure 2, Step 6) temporally at Harwell between 1990 and 2013 (Section 3.1.1) and spatially between Auchencorth and Harwell (Section 3.1.2).”

P. 1886 In 24: “This section presents two analyses of the impact, state and drivers of the chemical climatology framework (Figure 2, steps 1-5); specifically, changes in chemical climate phase (Figure 2, Step 6) temporally at Harwell between 1990 and 2013 (Section 3.2.1) and spatially between Auchencorth and Harwell (Section 3.2.2).”

**Reviewer 1 comment:** *The manuscript is long and feels repetitious in places, and it would benefit from a stronger focus on the key findings of the study, rather than describing every piece of data. There are several ways in which this could be achieved; one suggestion from me would be to integrate the different elements implied within Fig 1 for all health and vegetation indices, e.g. from states, through trends, to drivers and phases in turn, with results and discussion integrated in both cases.*

Response: As outlined above, the Results and Discussion is structured to emphasise the investigation of different chemical climate phases within the chemical climatology framework. The collation of health and vegetation impacts (i.e. collation of multiple chemical climates) into a single section would dilute this emphasis on the chemical climatology framework used to derive the statistics. Nevertheless, in recognition of comments on length made by both reviewers, we have undertaken some further editing and restructuring, and deleted some repetitious text, to produce a revised manuscript with better focus on the key findings. The most significant changes are summarised below:

- Section 3.1.3 Comparison between SOMO10/SOMO35 and higher threshold metrics: A new section (3.1.3) has been created to compare SOMO10 and SOMO35 with other commonly used metrics such as the EU target value (60 ppb threshold) and the WHO guideline value (50 ppb). Text describing contributions to SOMO10 and SOMO35 from high O<sub>3</sub> concentrations, and comparison with higher threshold metrics (EU target value (60 ppb) and WHO guideline (50 ppb), has been moved from Sections 3.1.1 and 3.1.2 to Section 3.1.3. The effect is to focus Sections 3.1.1 and 3.1.2 on the description and interpretation of temporal and spatial changes, respectively, in the chemical climate statistics and in the contribution to health impact from regional and hemispheric O<sub>3</sub>. However, important information about the comparability between SOMO10/35 and other commonly used metrics is also retained. Addition of a new Table 1 (in response to a comment below), which contains average POD<sub>Y</sub> across different soil textures, means that statement of these values in the text has been deleted. Specifically, deletion of the following:

P. 1887 In 4: “For crops, there has not been a statistically significant change in PODY between 1990 and 2013. The 1990–2013 average ( $\pm$  SD) PODY for potato was  $2.7 \pm 1.3 \text{ mmol m}^{-2}$ , which corresponds to a 3.4 % reduction in tuber weight. For wheat, the average PODY was  $1.5 \pm 1.1 \text{ mmol m}^{-2}$  (equivalent to a 5.7 % average grain yield reduction).”

P. 1888 In 10 “The average PODY for beech of  $14.7 \pm 3.7 \text{ mmol m}^{-2}$  was four times the critical level (Mills et al., 2011c) and corresponds to a biomass reduction of 16. %. The average PODY for Scots pine was  $27.0 \pm 3.7 \text{ mmol m}^{-2}$ .”

In addressing a comment about repetition of statistics from figures and supplements from Reviewer #2, additional deletions are:

Section 3.1.2:P. 1884, line 2: The first paragraph has been deleted, as it contained no new information which couldn't be found in the figures and supplementary materials; the reader is directed to the additional information contained in the latter at the start of the next paragraph. The information from the last sentence of the paragraph has been incorporated into P. 1885 line 6:

**Original text:** “Elevated SOMO10 and SOMO35 values in 2008 (as also reported by Gauss et al. (2014a) using the EMEP/MSC-W model) resulted from an increased contribution from days with maximum 8-h concentrations above 50 ppb (12% and 36% contributions to SOMO10 and SOMO35 respectively)”

**Amended text:** “Elevated SOMO10 (6% above 2007-2013 average) and SOMO35 (67%) values in 2008 at Auchencorth (as also reported by Gauss et al. (2014a) using the EMEP/MSC-W model) resulted from an increased contribution from days with maximum 8-h concentrations above 50 ppb (12% and 36% contributions to SOMO10 and SOMO35 respectively)”

Section 3.2.2: P. 1891, line 12: The start of the paragraph has been amended to remove repetition of statistics from figures and supplementary materials.

**Original text:** “At Auchencorth, the 2007–2013 average tuber weight reduction for potato predicted by the calculated PODY (Fig. 10a) was  $1.3 \pm 0.5\%$  (Table S4), and for wheat the average yield reduction was  $3.7 \pm 1.5\%$  (Table S6). The LRTAP critical level for impact (Mills et al., 2011c) was only exceeded at this site in 2008 for wheat (5.04% yield reduction). Annual PODY for potato at Auchencorth were consistently lower than at Harwell, while, for wheat, PODY were higher at Auchencorth for 3 of the 7 years.”

**Amended text:** “Annual PODY for potato at Auchencorth (Table S4) were consistently lower than at Harwell, while, for wheat, PODY were higher at Auchencorth (Table S6) for 3 of the 7 years. The LRTAP critical level for impact (Mills et al., 2011) was only exceeded at this site in 2008 for wheat (5.04% yield reduction).”

P. 1893, line 5: The sentence specifying the exact greater proportion of beech and Scots pine PODY accumulated during July and August at Auchencorth has been deleted.

**Reviewer 1 comment:** *I would prefer to see key conclusions that are highlighted in the abstract and conclusions to be clearly supported in the body of the text. For example, 3.2.3 compares POD and AOT40 trends, but the latter are lost in supplementary material. Addition of AOT40 to the time trends in the main figures alongside the POD trends would provide much a clearer demonstration to the reader of this key finding. The authors also make reference to comparison with health guidelines based on 50ppb or 60ppb thresholds, and data to support an inferred difference in trends could usefully be presented.*

Response: We note that Reviewer #2 has commented that the information regarding comparison between SOMO10 and SOMO35 is there, but scattered across multiple sections. An additional section (3.1.3) has been added to more usefully present this comparison, and is partly derived from the relocation of statistics presented in other sections:

### **“3.1.3 Comparison between SOMO10/SOMO35 and higher threshold metrics**

In spite of these spatial differences between the SOMO10 and SOMO35 metrics, both provide a substantially different picture of the extent (proportion of year over which impact metric is accumulated), timing (particular periods when impact metric is accumulated) and severity (magnitude of impact metric) of human health relevant O<sub>3</sub> exposure at Harwell and Auchencorth compared with use of higher threshold metrics such as the WHO air quality guideline (50 ppb) or the EU target value (60 ppb). For example, in 2013, the extent of exceedance of the 60 ppb EU target value across the UK was only 19 days (at least 1 of 81 UK sites exceeding threshold), and the timing of these exceedances was mainly in summer (EEA, 2014b). In contrast, at Harwell in 2013, there were 356 and 130 ADs for SOMO10 and SOMO35 respectively, of which only 27% and 28% was accumulated in summer. In respect of severity, at Harwell in 2010-2013, on average 91% and 66% of SOMO10 and SOMO35 respectively was accumulated on days with maximum 8-h O<sub>3</sub> concentrations below the WHO guideline of 50 ppb, compared to 76% and 38% in 1990-1993 (Table S1). At Auchencorth, an even larger proportion of SOMO10 and SOMO35 were accumulated below 50 ppb, on average 96% and 84% respectively during the 2007-2013 monitoring period (Table S2).

The overall impression from these statistics showing a decline in exposure to concentrations in excess of 35 ppb is that the threat to human health has declined between 1990 and 2013 in south-east England. The comments from the EEA (2014) on the very few episodes in excess of 50 or 60 ppb in 2013 are consistent with this view. However, the recent REVIHAAP (2013) synthesis shows that the lower percentiles of O<sub>3</sub> are also important and it is hard to define a precise threshold below which O<sub>3</sub> is not harmful. Thus the dose of O<sub>3</sub> to humans through respiration may be the more important guide to the potential threat, and as the SOMO10 (and the mean values) have changed little with time, the suggested improvement in air quality from the EEA may be more apparent than

real. An important policy implication of these trends is the degree to which local, regional or global policies are required to decrease the threat to human health from O<sub>3</sub>. In the case of exposures to O<sub>3</sub> in excess of 60 ppb, controls at the European and national scales can be effective, as the measurements demonstrate. However, if the mean or lower percentiles are important, as suggested in recent syntheses, then controls at much larger (hemispheric) scales are required.”

A new Figure 14 has been added showing crop AOT40 between 1990 and 2013 at Harwell, and the Theil-Sen estimate of the trend. Reference to this figure has been added at P. 1894 ln 3, and reference to the POD<sub>Y</sub> figure (8a) for comparison has been added at P1894 ln 5.

**Reviewer 1 comment:** *Soil moisture is an important limiting factor for stomatal ozone flux, but it is unclear to me from the paper how the SWP and PAW was determined for the two sites. No mention is made of direct measurements at the supersites or the met. sites, and so I assume this was done within the DO<sub>3</sub>SE model. However, if the values were modelled, this needs to be explained, and the important assumptions about soil characteristics that need to be made should be stated; were these different for the species to reflect the very different soil types on which they are likely to grow?. In passing, once this variable is included in any index it cannot be stated that a site is regionally representative, as soil characteristics may vary widely over short distances, and indeed are likely to be associated with different vegetation types. This limitation to the use of flux-based indices should be recognised in the paper.*

Response: Soil moisture (SWP and PAW) was modelled, based on the measured meteorological input data. The DO<sub>3</sub>SE model can be run with four different soil textures that have different hydraulic properties that alter the extent of evaporation and hence SWP. The soil textures are sandy loam (soil texture classification = coarse), silt loam (medium coarse), loam (medium) and clay loam (fine). POD<sub>Y</sub> was calculated using the loam (medium) soil texture in the original manuscript but in order to extend the conclusions of the analysis to other soil textures, statistics have now been calculated for all four soil types.

At Harwell, the magnitude of POD<sub>Y</sub> varied between soil textures, but the pattern of accumulation was consistent. At Auchencorth, there was much less variation in POD<sub>Y</sub> with soil texture than at Harwell, indicating soil moisture to have a less limiting influence. This additional analysis shows that the key conclusions from the derivation of these O<sub>3</sub> vegetation chemical climates were consistent across the four soil textures; namely that there was no trend in POD<sub>Y</sub> for any vegetation type between 1990 and 2013 at Harwell and that this was due to non-O<sub>3</sub> factors (e.g. SWP) which reduce stomatal conductance during high O<sub>3</sub> episodes. Text has been changed as follows to emphasise that the conclusions from the paper are applicable across a range of soil textures:

- Table 1: To compare the sensitivity of POD<sub>Y</sub> to soil texture at each site, a new table has been added which shows the average POD<sub>Y</sub> of each vegetation type, for each soil texture, over the study periods at Harwell and Auchencorth. This has also resulted in the deletion of results sentences which duplicate information now found in Table 1. Specific deletions:

P. 1887 ln 5: “The 1990–2013 average ( $\pm$  SD)  $POD_Y$  for potato was  $2.7 \pm 1.3$   $mmol\ m^{-2}$ , which corresponds to a 3.4 % reduction in tuber weight. For wheat, the average  $POD_Y$  was  $1.5 \pm 1.1$   $mmol\ m^{-2}$  (equivalent to a 5.7 % average grain yield reduction).”

P. 1888 ln 10:

**Original text:** “The average  $POD_Y$  for beech of  $14.7 \pm 3.7$   $mmol\ m^{-2}$  was four times the critical level (Mills et al., 2011c) and corresponds to a biomass reduction of 16%. The average  $POD_Y$  for Scots pine was  $27.0 \pm 3.7$   $mmol\ m^{-2}$ . These  $POD_Y$  values are substantially higher than for the crops, due to a lower threshold for exceedance, a longer growing season and other differences in the stomatal conductance response to T, PAR, VPD and SWP.”

**Amended Text:** “The average  $POD_Y$  for beech (Table 1) was four times the critical level (Mills et al., 2011). Beech and Scots pine  $POD_Y$  values were substantially higher than for the crops, due to a lower threshold for exceedance, a longer growing season and other differences in the stomatal conductance response to T, PAR, VPD and SWP.”

**Additional text:**

P. 1874 ln 24: Addition of the following statement in the Methods that soil moisture was calculated from measured meteorological data within the  $DO_3SE$  model, and  $POD_Y$  was calculated for 4 soil textures.

“SWP and PAW were calculated in the  $DO_3SE$  model using the measured meteorological data based on the Penman-Monteith model of evapotranspiration (Bueker et al., 2012). In addition to meteorological conditions, the evaporation of moisture from soil is dependent on the hydraulic properties of the soil texture. Statistics were therefore calculated for four different soil textures, sandy loam (soil texture classification = coarse), silt loam (medium coarse), loam (medium) and clay loam (fine). The properties of these soil textures are detailed in Bueker et al. (2012).”

P. 1886 ln 24: Explanation that the statistics included in the figures and supplementary datasheets are for the loam soil texture.

“The  $POD_Y$  statistics presented in Tables S5-S12, and Figures 8-13 were calculated for the loam (medium) soil texture (Bueker et al., 2012). The representativeness of the conclusions derived from the interpretation of these statistics to other soil textures is discussed in Sections 3.2.1 and 3.2.2.”

P. 1887 ln 4: Introduction of Table 1 in the text.

“The 1990-2013 average  $POD_Y$  values calculated using sandy loam (coarse), silt loam (medium coarse), loam (medium) and clay loam (fine) soil texture properties are shown in Table 1. The ratio between the largest and smallest average  $POD_Y$  due to differences in soil moisture for the different soil textures was 1.57 (wheat), 1.32 (potato), 1.14 (beech) and 1.10 (Scots pine), but the annual pattern of  $POD_Y$  accumulation was consistent across the four soil textures. The statistics in the following sections are those calculated for the loam soil texture, unless otherwise stated, which has intermediate hydraulic properties compared with the three other soil textures.”

- Discussion of the effect of soil moisture in limiting O<sub>3</sub> accumulation during high O<sub>3</sub> episodes has been modified as follows to state that this conclusion is consistent across different soil textures.

SWP during different concentration ranges have been changed to those calculated for clay loam soil texture. Clay loam had the highest SWP, i.e. lowest stomatal conductance limitation due to soil moisture, but during high O<sub>3</sub> concentrations, SWP was sufficiently low to limit stomatal conductance compared to more moderate O<sub>3</sub> concentrations. The limitation was even stronger for the other soil textures.

**Amended paragraph:** P. 1890 ln 1: “Soil water potential (SWP) is a soil texture dependent determinant of potato stomatal conductance in the DO<sub>3</sub>SE model, which decreases when SWP is lower than  $-0.5$  MPa (Bueker et al., 2012; LRTAP Convention, 2010). The 1990-2013 average SWP during hours when O<sub>3</sub> concentrations at Harwell were in the concentration ranges 60-65 ppb, 65-70 ppb and  $>70$  ppb were  $-1.50 \pm 1.32$  MPa,  $-1.14 \pm 0.93$  MPa and  $-1.10 \pm 0.90$  MPa respectively for the clay loam (fine) soil texture. The average SWP during these O<sub>3</sub> concentration ranges were lower, and even more limiting for the other three soil textures. These are substantially lower than the average SWP for the O<sub>3</sub> concentration ranges between 25 and 50 ppb, all of which are above the  $-0.5$  MPa cut-off except 45-50 ppb for sandy loam, silt loam and loam soil textures (average SWP of  $-0.65$ ,  $-0.52$  and  $-0.58$  MPa respectively). Across all soil textures, reduction in the frequency of elevated O<sub>3</sub> concentrations produced during regional photochemical episodes has therefore not reduced POD<sub>Y</sub>, as these elevated O<sub>3</sub> concentrations coincided with other factors (e.g. SWP) which limit stomatal conductance and hence any potential increase in O<sub>3</sub> accumulation resulting from increased O<sub>3</sub> concentrations. Decreasing regional O<sub>3</sub> production resulted in the largest change in concentration bin contributions for potato POD<sub>Y</sub> (Figure 10b). This is due to a later growing season compared with wheat, and a shorter accumulation period and higher maximum stomatal conductance compared with forest trees ( $150$  and  $180$  mmol m<sup>2</sup> s<sup>-1</sup> for beech and Scots pine respectively compared to  $750$  mmol m<sup>2</sup> s<sup>-1</sup> for potato), limiting the O<sub>3</sub> flux during high O<sub>3</sub> episodes.”

- P. 1891 ln 5: The proportion of hours when beech SWP was below the threshold for stomatal conductance limitation is now stated as the range for the different soil textures, and a summary sentence has been added to the end of the paragraph to emphasise consistency across soil textures.

**Original text:** “Between 2010 and 2013, on average 0 and 5 % of hourly SWP values in May and June, respectively, were below this value, compared with 38 and 28 % in July and August, respectively. The effect of SWP on stomatal conductance begins at  $-0.7$  MPa for Scots pine, and therefore has a larger limiting effect. SWP was also found to be one of the most important limiting factors in determining the impact of O<sub>3</sub> on forests across Europe (Emberson et al., 2007).”



**Amended text:** “Between 2010 and 2013, across the four soil textures, on average 0% and 0-9% of hourly SWP values in May and June, respectively, were below this value, compared with 23-51% and 18-31% in July and August, respectively. The effect of SWP on stomatal conductance begins at  $-0.7$  MPa for Scots pine, and therefore has a larger limiting effect. SWP was also found to be one of the most important limiting factors in determining the impact of  $O_3$  on forests across Europe (Emberson et al., 2007). Clay loam had the highest SWP of the four soil textures, and therefore the lowest limitation to stomatal conductance, followed by silt loam, loam and sandy loam. However, the variation in soil moisture between different soil textures due to differences in the extent of evaporation is sufficiently small that the lack of long-term trend in  $POD_Y$  and annual pattern of accumulation is consistent across the soil textures.”

- Insertion of text stating that variation in  $POD_Y$  between soil textures was less at Auchencorth, and the spatial difference between Auchencorth and Harwell were consistent across soil texture.

P. 1891 ln 12 “The 2007-2013 average  $POD_Y$  calculated for the four soil textures is shown in Table 1, and the variation between soil textures is less than at Harwell. The ratio between the largest and smallest average  $POD_Y$  due to differences in soil moisture for the different soil textures was 1.24 (wheat), 1.15 (potato), 1.02 (beech) and 1.02 (Scots pine). The pattern of accumulation, and spatial differences between Harwell and Auchencorth were consistent across soil textures.”

- In the discussion of elevated  $POD_Y$  at Auchencorth during regional photochemical  $O_3$  episodes, “across all soil textures” has been added at P. 1892 ln 5 and ln 13 and P. 1893 ln 12 and 18 to emphasise that the elevated  $POD_Y$  was calculated for all soil textures. To highlight the higher stomatal conductance at Auchencorth compared to Harwell, specific loam soil texture stomatal conductance values are used, and hence “for loam soil texture” has been added to P. 1892 ln 22 to make this clear.
- P. 1893 ln 3: The paragraph has been amended to reflect the range of percentage increases in beech and Scots pine  $POD_Y$  at Auchencorth compared to Harwell across the four soil textures.

**Original text:** “Between 2007 and 2013, Scots pine and beech  $POD_Y$  were on average 31% and 11% higher at Auchencorth compared to Harwell (Fig. 12a). These larger values were due to larger contributions from July and August at Auchencorth (Tables S10 and S12). On average, July and August contributed 7% and 5% more of the annual  $POD_Y$  at Auchencorth for beech (4% and 3% for Scots pine). In these months, higher temperatures at Harwell produced conditions which reduced stomatal conductance. For example, in 2007–2013 at Harwell, SWP was on average 59% higher in July and 82% higher in 10 August than at Auchencorth.”

**Amended text:** “Between 2007 and 2013 across the soil textures, Scots pine and beech  $POD_Y$  were on average 27-37% and 5-19% higher at Auchencorth compared to Harwell (Table 1 and Figure 13a). These larger values were due to

larger contributions from July and August at Auchencorth (Tables S10 and S12). In these months, higher temperatures at Harwell produced conditions which reduced stomatal conductance. For example, in 2007-2013 at Harwell for loam soil texture, SWP was on average 59% higher in July and 82% higher in August than at Auchencorth.”

**Reviewer 1 comment:** *There seem to be a range of different months used for analysis of trends and drivers for vegetation types. In particular, the results for trees should focus on a longer growing season compared to wheat and potato, for which only a limited period of crop development is considered. Furthermore, phenological timings which are under climatic control within DO3SE may also vary between years and show long-term trends, and this needs to be considered.*

Response: A range of different months were used for analysis according to the recommended methods by which the growing seasons were determined in LRTAP Convention (2010). LRTAP Convention (2010) is a mapping manual which sets out the most suitable method to calculate the impact of O<sub>3</sub> on vegetation, and hence is referenced throughout as the source of parameterisation. To make it clearer that this parameterisation extends to the calculation of the growing season, this has now been stated in the methods:

P. 1874 In 4: “POD<sub>Y</sub> values were calculated for two crops (wheat and potato) and two forest trees (beech and Scots pine), accumulated across their respective growing seasons. The length of the growing seasons, and phenological limitation on stomatal conductance throughout the growing season were derived according to methods detailed in LRTAP Convention (2010). The growing seasons for wheat (late April – early August) and potato (late May – early September) were calculated by accumulated temperature and therefore varied inter-annually based on meteorological conditions. For beech, the growing season was calculated using a latitude model (19/04-20/10 at Harwell, 26/04-10/10 at Auchencorth). The Scots pine growing season was the full year.”

More information about the growing season is included in the response to comments by Reviewer #2.

**Reviewer 1 comment:** *The authors claim that the lack of trends in POD<sub>Y</sub> variables is due to changes in nonozone factors, but there is no analysis of trends in the individual factors limiting stomatal conductance, or of conductance itself. On p18, they focus on differences between ozone concentration bins (over what period of the year is not specified) rather than trend analysis, and they also need to consider that SWP/PAW is a cumulative variable and its seasonal development may be linked directly to phenological changes.*

Response: Given that the calculation of stomatal conductance is derived in part by phenology and in part by the meteorological input data, analysis of trends in the non-O<sub>3</sub> factors would require trend analysis in these meteorological variables. The aim of this paper was to assess how O<sub>3</sub> impacts have responded to spatial and temporal changes to their state and drivers, rather than an analysis of climate change over the last 24 years.

In-depth trend analysis of meteorological variables would reduce the focus on the key O<sub>3</sub> findings of the paper, as requested in an earlier comment from this reviewer.

The focus on the POD<sub>Y</sub> accumulated during different O<sub>3</sub> concentration bins (by definition within the growing season of the respective vegetation type) is highly relevant for policy, as it determines a) whether past efforts to control O<sub>3</sub> (and reduce the frequency of high O<sub>3</sub> episodes) has reduced POD<sub>Y</sub>, and b) whether future mitigation strategies would be more effective in reducing vegetation impacts through control of high O<sub>3</sub> episodic concentrations, or more frequent, moderate concentrations. Having established that at Harwell there has been a shift to increasing contribution from moderate concentrations, and reduction in the frequency of elevated O<sub>3</sub> has not reduced POD<sub>Y</sub>, other factors are considered to explain these trends. This structure maintains focus on key findings in relation to past and future O<sub>3</sub> impact mitigation. When considering the limiting effect of soil moisture on stomatal conductance, and hence on POD<sub>Y</sub>, the 1990-2013 median SWP during different O<sub>3</sub> concentration bins is appropriate to demonstrate that high O<sub>3</sub> concentrations coincide with SWP that is substantially more limiting to stomatal conductance across the entirety of the 24 year period. For example (see P. 1890 ln 5 for the analysis to which this relates), between 1990 and 2002 (the first half of the Harwell study period), the average SWP during 60-65 ppb, 65-70 ppb and >70 ppb is lower than the median in 6, 7 and 6 of the 12 years respectively. That approximately half of the years in the first half of the study are below the median suggest that there has not been a trend in SWP of sufficient magnitude to prevent it from generally limiting stomatal conductance during high O<sub>3</sub> concentrations.

### ***Reviewer 1 Technical corrections***

***Reviewer 1 comment:*** Provide the full name the first time that any acronym is mentioned.

Response: The full name of REVIHAAP has been added on first usage.

***Reviewer 1 comment:*** P5, 113. Don't you mean 'during the growing season', not annual?

Response: The text has been amended in line with this recommendation.

***Reviewer 1 comment:*** P5, 116-17. I'm not clear where this comparison of measured and gridded modelling is presented.

Response: Comparison between the POD<sub>Y</sub> calculated in this study and those calculated previously using modelled input data is presented throughout the Results and Discussion. Specifically comparison is found at P. 1887 ln 11, P. 1888 ln 15, P. 1891 ln 10 and P. 1891 ln 17-26.

**Reviewer 1 comment:** P6. L26. *This May-July period is only a relevant comparison for wheat and potato, for the tree species a six-month accumulation period would be more relevant.*

Response: We have been consistent throughout the manuscript, including in the Methods when first described, in stating that the AOT40 calculated for comparison was that which is relevant for comparison with crops, i.e. wheat and potato. Contrasting trends between forest POD<sub>Y</sub> and AOT40 is made through reference on P. 1894 ln 24 to forest-relevant AOT40 calculated previously (Gauss et al., 2014b) across the UK which has decreased in recent years compared to 2000, in contrast to the forest POD<sub>Y</sub> values calculated in this study.

**Reviewer 1 comment:** *The ozone concentration bin diagrams might be clearer and easier for the reader to interpret with a smaller number of bins with cut-offs more clearly related to the key significant trends in the data.*

Response: We agree that a balance is required in the determination of concentration bin size between being small enough to capture trends at different concentration ranges, and being large enough to avoid so many bins that the resulting figure is difficult to interpret. For many of the impacts, there is a contrasting trend at Harwell in the contribution of moderate O<sub>3</sub> concentrations (increasing) and high O<sub>3</sub> concentrations (decreasing), and in between, there are a range of concentrations with no trend in contribution. The issue of aggregating concentrations to a smaller number of bins is that the distinction in these trends becomes blurred, and the point at which trends change is determined with ever decreasing precision. We evaluated that 5 ppb concentration ranges were the optimum bin size to compromise between precisely determining the trends in O<sub>3</sub> concentration distributions associated with impacts, and creating an interpretable figure.

## Reviewer #2:

### *General comments*

*The authors have submitted an interesting manuscript focusing on trends and drivers of ozone concentrations and associated potential impacts on human health (SOMO35 and SOMO10) and vegetation (PODY and AOT40). The study is based on ozone measurements from two UK EMEP supersites, applying a chemical climatology framework. The results indicate that over the period 1990-2013 the relative importance of regional photochemical ozone production have decreased while the importance of hemispheric background concentrations have increased. However, the change in health and vegetation impact metrics differ depending on which metric and threshold is chosen.*

*Ozone is a key air pollutant and it is important to understand how much, and to which direction driving forces will affect ground-level ozone concentrations and their impacts on human health and vegetation. Thus the topic of this manuscript is highly relevant from a scientific and policy-related point of view. The study is comprehensive, well thought through and the manuscript is well written. I therefore suggest publication after minor revisions.*

### *Specific comments*

**Reviewer 2 comment:** *In the introduction and methodology section it is strongly emphasized that the study is based on a chemical climatology approach. When I read the manuscript I feel that this approach is somewhat lost in the result and discussion sections. To be consistent I suggest linking also the second part of the manuscript to the different steps deriving the chemical climate in a more clear way e.g. by more clearly describing different temporal and spatial phases in the chemical climate.*

Response: The same comment was made by Reviewer #1. Therefore please see above for our detailed response and actions on this comment.

**Reviewer 2 comment:** *The manuscript is long and especially the amount of text in the result section is heavy for many readers. Numbers that can be found in figures, previous sections or in the supplementary material is unnecessary to repeat and I suggest reducing e.g. the sections about the spatial differences between the two supersites (section 3.1.2 and 3.2.2). Also I think the manuscript could be improved by integrating results and discussion more.*

Response: Several repetitions of numbers found elsewhere within the manuscript and in figures have been omitted to reduce the length of the Results and Discussion section. As noted in the reviewer comment, these mainly occur in sections 3.1.2 and 3.2.2. The addition of the new Table 1 has also resulted in the deletion of text which now states that data are summarised in the new table. Specific actions and deletions to avoid repetition and reduce the length of the manuscript are detailed above in response to a similar comment by Reviewer #1.

The structure of each individual section is such that an overview of the chemical climate impact, state and drivers statistics is given first, in relation to the change in chemical climate phase being discussed (e.g. temporal at Harwell vs spatial between Auchencorth and Harwell). This structure, coupled with the changes resulting from the previous comment help to emphasise the steps in the chemical climatology framework used to undertake the analysis.

It is when these statistics are considered as a whole (i.e. the chemical climate) that insight is gained in terms of changing relative contribution from hemispheric vs regional processes. We think that the shortening of the results paragraphs as detailed above and in the response to a similar comment by Reviewer #1 helps to focus these paragraphs on communicating the key temporal and spatial trends in the statistics necessary to understand the discussion paragraphs that follow. In addition, text relating to the comparison of SOMO10/35 with the EU target values and WHO air quality guideline values has also been moved to a separate section (see below), and hence presentation of key statistical trends and discussion of their meaning have been moved into closer proximity.

**Reviewer 2 comment:** *Methods: A map with the two supersites would be appreciated. The ones in the supplementary material are very small.*

Response: A map has been added as Figure 2 showing the monitoring sites, and the Met Office stations from which meteorological data was used.

*How come meteorology is not measures at the two supersites? Are the meteorological stations representative for the air quality supersites? Is soil moisture also measured?*

Response: Meteorological measurements are made at the two supersites, but it was appropriate to use the UK Met Office station data which was more readily available and subject to documented quality control procedures ([http://badc.nerc.ac.uk/data/ukmo-midas/ukmo\\_guide.html#2.1](http://badc.nerc.ac.uk/data/ukmo-midas/ukmo_guide.html#2.1)). It is noted that the on-site meteorology is available on request and is planned to be submitted to the Met Office WOW database in the near future.

The location of the Met Office stations are specified such that it is 'representative on a scale required for the station'. The meteorological stations used in this study all measure synoptic scale weather patterns, and therefore are contained within the category of Met Office station with the largest area of representativeness (up to tens of kilometres). The main stations used, Benson and Gogarbank, are 13 km and 14 km from Harwell and Auchencorth respectively, and were therefore considered to be representative of conditions at the supersites. Combining O<sub>3</sub> and meteorological data from different sites to calculate POD<sub>Y</sub> has been done previously at EMEP sites in Sweden (Karlsson et al., 2007).

Harwell: Temperature, wind speed and wind direction are measured but data was only available back to 2005. As the full suite of meteorological parameters required to determine stomatal conductance were also not provided, it was considered more appropriate to use data from nearby stations.

Auchencorth: The full meteorological parameters required to derive stomatal conductance are measured. However Auchencorth is an ombrotrophic bog, and not a site where the vegetation types would be directly grown. Hence it was more appropriate in the calculation of  $POD_Y$  to use meteorological data from the nearby Gogarbank station which has been located to maximise the representativeness of its observations, including areas within south-east Scotland where the vegetation types would be grown.

**Reviewer 2 comment:** P. 1876, line 23: *How can  $POD_Y$  decrease for potato after interpolation of missing data?*

Response: The time period over which potato  $POD_Y$  is accumulated is based on accumulated temperature after plant emergence (outlined in more detail below). When missing values are interpolated, the maximum accumulated temperature (and hence end of the growing season) is reached earlier. Whether an increase or decrease in  $POD_Y$  results from this change in growing season depends on whether the parameters excluded from the growing season or those interpolated produce conditions ( $O_3$  concentration and those affecting stomatal conductance) more favourable for  $POD_Y$  accumulation. For 1993, the excluded time periods had conditions more conducive to  $POD_Y$  accumulation and hence after interpolation  $POD_Y$  decreased by 6%.

The key point of the comparison between interpolated and non-interpolated potato  $POD_Y$  is the magnitude of the difference, not the direction. This example had the largest number of missing values of any year (above the data capture threshold of 75%), and, after interpolation, the change in magnitude of  $POD_Y$  was small. This gives confidence that the  $POD_Y$  values calculated for the other years are not largely biased due to missing values.

**Reviewer 2 comment:** *Results and discussion: I find the comparison between different impact metrics very interesting and highly relevant in a policy context. The comparison between the vegetation impact metrics  $POD_Y$  and AOT40 is discussed in an own section (3.2.3) but for human health metrics the comparison with the WHO air quality guideline (50 ppb) and EU target value (60 ppb) is not as easy to find. Much information is there but spread out in different section. The manuscript would benefit if this information is assembled into one paragraph.*

Response: We agree that the manuscript would benefit if the results relevant to comparison between SOMO10/35 and higher threshold metrics were presented together. Hence a new section, '3.1.3 Comparison between SOMO10/SOMO35 and higher threshold metrics' has been added.

- The final section of 3.1.2, from P. 1885, ln24 has been moved to Section 3.1.3, as have sentences from P. 1880 line 26 and P. 1881 line 8, which show the proportion of SOMO10 and SOMO35 accumulated below the WHO air quality guideline (50 ppb).
- Additional text has been added for the corresponding values at Auchencorth.
- The new section is shown above in response to a comment by Reviewer #1 about the conclusions from comparison with higher threshold metrics being better highlighted in the main text.

**Reviewer 2 comment:** P. 1879, line13-14: *Why are these specific time periods chosen?*

Response: The purpose of the datasheets is to provide a summary of the chemical climate impact, state and drivers statistics. Including individual year statistics for Harwell would result in a datasheet with such a density of statistics that it would be difficult to draw useful information. Hence to reduce the number of statistics, they were averaged into 4-year blocks. The length of the averaging period was chosen as a balance between producing an average representative of the time period whilst avoiding the incorporation of any long-term trends.

**Reviewer 2 comment:** Section 3.2.1: *Has the timing and length of the growth period changed during the 24-year period? Please discuss if/how that could have influenced your results.*

In order for the calculations of  $POD_Y$  in this work to be comparable with those in previous studies, the timing and length of the growing season were all calculated based on the methods outlined in the LRTAP mapping manual (LRTAP Convention, 2010), and summarised individually below. Their application in this work can account for changes in the length and timing of the growing season resulting from long term changes in temperature, but does not account for changes in farming practices (e.g. sowing times for crops).

Wheat:  $POD_Y$  accumulation is based on accumulated temperature, and begins 200 °C.days before mid-anthesis, and ends 700 °C.days after. The wheat phenology function is at maximum between -200 °C.days and 100 °C.days, before decreasing.

Inter-annual variability and long-term changes in temperature are accounted for, and during each year the exact timing and length of the growing season changes. However, any changes in the timing of mid-anthesis, due to changes in sowing dates are not reflected. Changes in sowing dates could both increase and decrease  $POD_Y$ , and the exact change would be determined by the temperature, soil moisture, and radiation conditions during that particular year. For example, we conclude that during summertime  $O_3$  episodes, plant conditions are often such that increased accumulation of  $O_3$  due to higher concentrations is inhibited by reduced stomatal conductance. A trend towards earlier sowing dates would result in the  $POD_Y$  accumulation period occurring earlier in the year, which could increase  $POD_Y$ , due to the lower frequency of conditions which reduce stomatal conductance during the summer (e.g. low soil moisture), but could also decrease  $POD_Y$  due to lower temperatures earlier in the year which reduce stomatal conductance (zero conductance below 12 °C).

Any changes in the length and timing of the growing season will not change the main conclusions from the analysis. There has been a shift towards a larger contribution from hemispheric background concentrations in determining  $POD_Y$  compared to regional episodes, and this has not resulted in a decrease in vegetation impact because during regional  $O_3$  episodes, stomatal conductance is often reduced, and hence additional  $O_3$  accumulation cannot occur.



Potato:  $POD_Y$  accumulation is also based on accumulated temperature, and occurs for 1130 °C.days after plant emergence. It is recommended that day 146 (26<sup>th</sup> May/27<sup>th</sup> in a leap year), is used as the plant emergence date (LRTAP Convention, 2010). The phenology function increased until 330 °C.days before decreasing until the end of the growing season. The conclusions about using this accumulated temperature method for potato are the same as for wheat. Inter-annual variability and long-term changes in temperature are accounted for by changes in the phenology function, but changes in sowing date are not. A change in sow date could similarly result in an increase or decrease in  $POD_Y$ , depending on the particular plant conditions during the year.

Beech:  $POD_Y$  is accumulated across a fixed growing season, which is calculated using a latitude model. Long-term changes in temperature between 1990 and 2014 do not result in an extension or curtailment of the growing season, which would increase or decrease  $POD_Y$  respectively. LRTAP Convention (2010) concluded that the latitude model agreed well with ground observations from studies across Europe covering multiple years up to 2003.

Scots Pine: The growing season for Scots pine is determined by air temperature, and is accumulated throughout the year when temperature is above 0 °C. Changes to the growing season due to long-term changes in temperature are therefore reflected in the calculation of  $POD_Y$ .

Text has been added to the methods to give more information about the difference in growing season between the vegetation types:

P. 1874 ln 4: “ $POD_Y$  values were calculated for two crops (wheat and potato) and two forest trees (beech and Scots pine), accumulated across their respective growing seasons. The length of the growing seasons, and phenological limitation on stomatal conductance throughout the growing season were derived according to methods detailed in LRTAP Convention (2010). The growing seasons for wheat (late April – early August) and potato (late May – early September), were calculated by accumulated temperature and therefore varied inter-annually based on meteorological conditions. For beech, the growing season was calculated using a latitude model (19/04-20/10 at Harwell, 26/04-10/10 at Auchencorth). The Scots pine growing season was the full year.”

***Reviewer 2 Technical corrections:***

***Reviewer 2 comment:*** P. 1871, line 19: write out the REVIHAAP acronym first time mentioned.

Response: The text has been amended in line with this recommendation.

***Reviewer 2 comment:*** P. 1873, line16: space between NOx and

Response: The space was omitted on conversion to the discussion paper format, and is present in the original manuscript.

**Reviewer 2 comment:** P. 1877, line 19-21: difficult sentence, please rephrase. Can the paragraph about the trajectories be written more clearly?

Response: We agree that this section could be better phrased. Text has been reordered to make it clearer earlier in the paragraph how air-mass back trajectories were used to link air-mass history to the state of the chemical climate, i.e. by grouping back trajectories based on their pathway and calculating the proportion of trajectories from each group during SOMO10/35 accumulation and non-accumulation days. The remainder of the paragraph contains specific detail of the clustering method used to achieve this.

**Original text:** “Secondly, the association of the state with air-mass history was investigated using the 2920 4-day HYSPLIT air-mass back trajectories arriving every 3 hours at each site per year (Carslaw and Ropkins, 2013; Draxler and Rolph, 2013; R Core Development Team, 2008). The trajectories were grouped using Ward’s linkage hierarchical cluster analysis, a clustering method that has been shown through simulations to perform effectively (Mangiameli et al., 1996)”

**Amended text:** “Secondly, the association of the state (Step 4) with air-mass history was investigated by grouping back trajectories based on the similarity of their pathway. The proportion of trajectories arriving from each group during SOMO10/35 and POD<sub>Y</sub> ADs and NADs, and over the AOT40 growing season, was then compared. Pre-calculated 4-day HYSPLIT air-mass back trajectories arriving at 3 hour intervals (2920 trajectories per year) were used (Carslaw and Ropkins, 2013; Draxler and Rolph, 2013; R Core Development Team, 2008), and grouped using Ward’s linkage hierarchical cluster analysis which has been shown through simulations to perform effectively (Mangiameli et al., 1996)”

The final sentence of this paragraph (P. 1878, line 11) has been removed as it is now repetition of the first sentences of the amended text.

**Reviewer 2 comment:** P. 1878, line 24: . . .ADs and NADs for SOMO10/35 and POD<sub>Y</sub>?

Response: Text has been amended to make clear that daily emissions estimates were compared on SOMO10/35 and POD<sub>Y</sub> exceedances days.

**Original text:** “The 96 hourly emissions estimates for each trajectory were summed, and averaged across the 8 trajectories arriving each day, producing a daily average trajectory NO<sub>x</sub> emissions estimate which was compared on ADs and NADs.”

**Amended text:** “The 96 hourly emissions estimates for each trajectory were summed, and averaged across the 8 trajectories arriving each day, producing a daily average trajectory NO<sub>x</sub> emissions estimate which was compared on SOMO10/35 and POD<sub>Y</sub> ADs and NADs.”

**Reviewer 2 comment:** *P. 1884, line 11: . . .representative of much of the rural west and north of the UK. . .*

Response: Text has been amended in line with the recommendation.

## References used in response to reviewers:

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### Additional Tables and Figures:

Table 1: Average  $\pm$  SD wheat, potato, beech and Scots pin  $\text{POD}_Y$  calculated for 4 different soil textures (see Bueker et al. (2012) for a description of their hydraulic properties) over the monitoring periods at Harwell and Auchencorth.

<b>Harwell 1990-2013 average</b>	<b>Sandy loam (coarse)</b>	<b>Silt loam (medium coarse)</b>	<b>Loam (medium)</b>	<b>Clay loam (fine)</b>
<b>Wheat <math>\text{POD}_Y</math> (<math>\text{mmol m}^{-2}</math>) (grain yield % reduction)</b>	1.21 $\pm$ 1.07 (4.61%)	1.75 $\pm$ 1.19 (6.64%)	1.51 $\pm$ 1.14 (5.72%)	1.90 $\pm$ 1.19 (7.22%)
<b>Potato <math>\text{POD}_Y</math> (<math>\text{mmol m}^{-2}</math>) (tuber yield % reduction)</b>	2.35 $\pm$ 1.27 (3.03%)	3.10 $\pm$ 1.42 (3.99%)	2.64 $\pm$ 1.32 (3.40%)	3.10 $\pm$ 1.46 (4.00%)
<b>Beech <math>\text{POD}_Y</math> (<math>\text{mmol m}^{-2}</math>) (biomass % reduction)</b>	14.0 $\pm$ 3.7 (15.4%)	16.0 $\pm$ 3.5 (17.6%)	14.7 $\pm$ 3.7 (16.2%)	16.1 $\pm$ 3.4 (17.7%)
<b>Pine <math>\text{POD}_Y</math> (<math>\text{mmol m}^{-2}</math>)</b>	26.2 $\pm$ 5.5	28.7 $\pm$ 5.3	27.0 $\pm$ 5.6	28.8 $\pm$ 5.3
<b>Auchencorth 2007-2013 average</b>				
<b>Wheat <math>\text{POD}_Y</math> (<math>\text{mmol m}^{-2}</math>) (grain yield % reduction)</b>	0.85 $\pm$ 0.45 (3.23%)	1.01 $\pm$ 0.38 (3.86%)	0.96 $\pm$ 0.39 (3.65%)	1.05 $\pm$ 0.37 (3.99%)
<b>Potato <math>\text{POD}_Y</math> (<math>\text{mmol m}^{-2}</math>) (tuber yield % reduction)</b>	0.95 $\pm$ 0.41 (1.22%)	1.08 $\pm$ 0.46 (1.39%)	0.99 $\pm$ 0.41 (1.28%)	1.09 $\pm$ 0.47 (1.40%)
<b>Beech <math>\text{POD}_Y</math> (<math>\text{mmol m}^{-2}</math>) (biomass % reduction)</b>	16.6 $\pm$ 1.6 (18.3%)	16.9 $\pm$ 1.2 (18.6%)	16.7 $\pm$ 1.5 (18.4%)	16.9 $\pm$ 1.2 (18.6%)
<b>Pine <math>\text{POD}_Y</math> (<math>\text{mmol m}^{-2}</math>)</b>	35.9 $\pm$ 3.6	36.5 $\pm$ 2.7	36.2 $\pm$ 3.3	36.6 $\pm$ 2.7

Figure 1: Map of the United Kingdom and Ireland showing the location of the two UK EMEP supersites (purple circles) at Auchencorth and Harwell, as well as the location of the UK Met Office stations from which meteorological data was used (green circles).

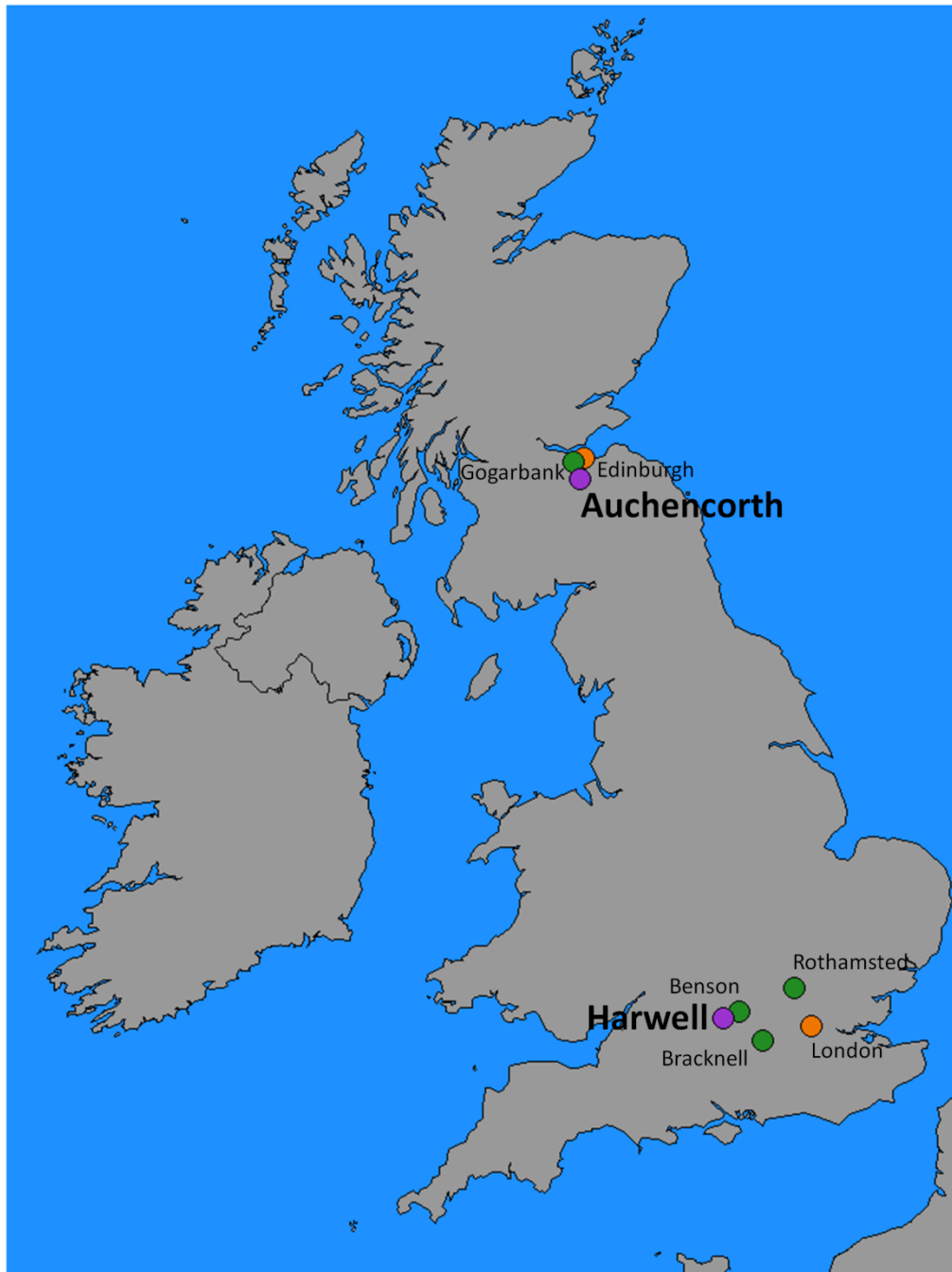


Figure 14: Crop-relevant AOT40 (calculated between May and July) at Harwell for the period 1990 to 2013. The Theil-Sen trend estimate of median trend (shown in red) is  $-3.6\% \text{ y}^{-1}$  ( $p = 0.001$ ).

