
Anonymous Referee #1

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General comments

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 consider-
ing 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

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.

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.

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.
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.

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 DO3SE 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.

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.

The authors claim that the lack of trends in PODy variables is due to changes in non-ozone 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 phonological changes.

Technical corrections

Provide the full name the first time that any acronym is mentioned.

P5, l13. Don’t you mean ‘during the growing season’, not annual?

P5, l16-17. I’m not clear where this comparison of measured and gridded modelling is presented.

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.

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.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 1869, 2015.