

Comments to the Authors (ACP-2015-43: *The impact of speciated VOCs on regional ozone increment derived from measurements at the UK EMEP supersites between 1999 and 2012*)

The authors present an in depth look at the regional ozone increment and what is possible when linking various analysis methods and data sources that are not necessarily typically combined. This provides for an interesting and relevant methodology that they present and some interesting insights. While I find the paper quite relevant, there are a number of issues with the presentation/clarity of some aspects of the analysis and process followed given the complexity of some of these linkages. I would recommend that the paper is published, if the comments can be sufficiently addressed.

General comments

There are a variety of concepts and terms used throughout the paper that could do with a better definition or initial explanation, either initially or referenced throughout, to help the reader follow the thread. Some of these are chemical climate, 'positive' and 'negative' O₃ increment and what the (+) and (-) indicate in terms of production vs reduction of ozone, as well as the link to photochemical depletion of the VOCs.

P10: The description on this page (L5-31) is quite involved and complex as to which data is included where, used how, where assumptions are made, etc. It could be quite helpful and provide significant clarify if there were e.g., a flow chart or similar to elucidate the steps/process that is followed.

In many of the sections of this paper, there are a lot of statistics quoted and involved connections made, which are often mixed with a variety of examples for the different months where x might result in one case and y in another. To add clarity to the paper it would be good if these sections/paragraphs, especially in e.g., section 3.3.2 on emission, would include an initial statement of aim at the beginning, so that it is more obvious to the reader why/how the points are important and where the discussion is leading.

Specific comments

P2L21-22: There are a number of factors that affect the utility of gridded emissions for use in measurement and modeling studies, with the aggregated nature of source sectors being just one. This should be acknowledged as it may otherwise seem to be an over simplified assessment of emission inventories.

P3L24: It would be good to include a brief definition of 'chemical climates' here or at the point that it is first addressed beyond the introduction (beginning of methodology section), as most readers will likely not be familiar with the concept and will very likely not look up the referenced material.

P4L12: This sentence doesn't make sense. Please clarify how one allows for the other.

P4L15: Here it is mentioned that you use monthly-averaged diurnal variations but offer no explanation why this averaging period was chosen, and unless I missed it, this choice is also not explained in the methods. A brief justification, here or elsewhere could be beneficial.

P5L13: The sentence makes reference to '...the two shown previously...' but it is not clear what these are. Could the author please clarify?

P6L28-P7L2: While the substitution of Mace Head data to calculate the regional O₃ increment at Auchencorth may be appropriate, but needs to be justified. How do the back trajectories compare? Is such a substitute reasonable?

P7L10: Please give an indication of what 'extensive periods' means in terms of a quantitative range or amount.

P7L10-P8L10: The paragraph is currently quite long and contains different, but related types of information. To make it a bit more clear, I would suggest that the authors insert paragraph breaks at P7L22 and P8L4. In addition, the final part (P8L4-8) summarizes what the inclusion of non-detects means, but does not reiterate why this is important/has been considered. Given the complexity and multiple aspects covered here, it would be good to add information, e.g., at the end of the sentence finishing on P8L8, to say something along the lines of '...indicating that the inclusion of the non-detects as values assumed to be below the LOD was justified.' Otherwise the reader has to go back to the middle p.7 to make that connection. It would just be good to try and connect the dots in a more explicit way in this section.

P8L23: should the sentence say that ethane is the second least reactive? What is meant by second smallest? Also, if it is second smallest, what is the first and why wasn't that used – although I assume that maybe this was methane, in which case that would not make sense, but without this information, it leaves the reader guessing. Also, is there any literature that has done this previously (I'm pretty sure there is) and it would be good to cite. Although I think most analyses have used CO more frequently, but the concept is the same. Might also be worth it to mention lifetime in the reasoning here.

P8L26-27: is this a reasonable assumption to make, that the magnitude of VOC emissions do not differ substantially between day and night? Something as simple as temperature would affect the magnitude of daytime/nighttime emissions of VOCs especially for biogenic VOCs, but also for e.g., evaporative emissions. If this is a reasonable assumption, could you please justify it somehow? Or if I have misinterpreted what is being assumed and refers instead to the emissions being brought in by the trajectories or something else, please clarify this. This statement also seems to be contradicted by the later sentence on L30 that states that night emissions were $\pm 12\%$ compared to day emissions. Or is that sentence meant to communicate that they are the same with a difference of no more than $\pm 12\%$? Please clarify.

P8L30: Does this refer to normal or pocp-weighted emissions?

P9L14-15: The authors mention those factors considered here. Are there other factors that play a large role and are not considered?

P11L14: Why were 2001 and 2011 chosen for Harwell and 2012 for Auchencorth? If all the years looked pretty much the same and the middle year was chosen for all, that would be fine, but it seems a bit random. Please justify the choices. (Or since the other years were clearly also analyzed since they are discussed later on in the section, consider mentioning a justification and/or providing the other plots in SI.)

P12L26-32: What are the standard deviations of the monthly contributions? This would help put the 22% vs 33% difference in perspective. Is this largely within the noise or is this a significant difference.

P12L23-24: The sentence states that six of the VOCs were not measured and gives a % range for their contribution to the monthly total. Could the authors provide a further statement as to whether that is significant enough that that would affect any of the conclusions made earlier in the paragraph? Also, the 2001 6 missing VOCs only pertains to Harwell correct?

Figure 4: Given that in Figure 5 there are also negative contributions from individual VOCs, should there also be negative contributions depicted in Figure 4? Or, if there is a reason they are not shown, can you explain?

Figures 5&6/P13L16-21: Given the presence of positive and negative values, could the authors provide a sentence either in the text or the figure captions to indicate what the positive vs negative contributions indicate, just for ease of reading and clarity. Similarly, L29-31, earlier m+p-xylene is stated to contribute to photochemical depletion (with positive values) and then negative values are discussed. It would be good to explain that these indicate photochemical production.

P13L28-P14L2: Why when earlier summer/July was being discussed, is now the focus on April/(May)?

P14L1-2: Mention that this change/difference will be explored more later. My initial reaction as a reader was one of wanting more explanation since I thought that was where discussion of that point ended.

P15L5-20: It would be worth including some discussion, or at least a mention, on the accuracy of the emission inventories. How do literature sources reporting the source apportionment results of measurement data agree or disagree with the emissions reported in the inventories. Either here, or possibly more appropriately, in the section on uncertainties.

P15L29-P16L9: add an aim/guiding sentence at the beginning of the paragraph. Might also provide clarity if one case e.g, that of April and July is followed through before comparing the conditions for May and June.

Figure 9: Could error bars be added to the plot? How does this compare to other years? Is this fairly consistent or is every year very different?

Figure 9/P16L4-9: Would it be possible to include information on all the aspects that are important to the regional O₃ increment discussed here in one Figure? It might allow for assessing the relationships and connections better, rather than a discussion or listing of percentages in the text.

P16L15: '... the large negative VOC diurnal photochemical depletion...' is a bit confusing. The negative refers only to how this is portrayed in the figures, correct? And the photochemical depletion is what is represented by the fact that it is negative and not positive, correct? In this case I would put the 'negative' in () or somehow indicate that this relates to the representation in the figures. This was similarly an issue with earlier discussion of such information/figures.

P16L10-P17L1: add guiding sentence at the beginning with aim.

P17L1: the previous text discusses VOC speciation, while the following text, starting on the referenced line then discusses emissions and their relationship to countries. I would insert a paragraph break here.

Section 3.3.3: None of this section seems to explicitly address uncertainty (except a brief mention regarding total Gg estimates), as the title would imply, but rather focuses on the mitigation and monitoring part. There are a variety of uncertainties given the back trajectories, the averaging periods, the assumptions associated with POCP values, not to mention the emission inventories. At least some of these should be explicitly addressed.

P18L29-32: The total biogenic VOC emissions estimate in Gg is given, and some information on differing estimates. While the total is mentioned for anthropogenic VOCs, there is no additional information given. Given that the focus is also on mitigation and what can be done, it would be good

to provide a range or at least mention some of the alternative estimates for anthropogenic VOC emissions, since this is also an area of uncertainty.

P19L19-21: The point about disaggregated source sectors is valid, however, I think it also over simplified in that it assumes the emission inventory information to be correct or at least that more highly disaggregated source sector information would be able to be reported in enough detail and at a level of certainty that would allow for such precise identification of mitigation strategies. There is significant uncertainty associated with emission inventories, especially at the more detailed level, and measurements and emission inventories do not always agree. As suggested in a previous point, some literature comparing these two and the importance of sources to VOC emissions should be at least acknowledged, even if it is not discussed in detail.

P19L29-32: constraints in what sense? Of what? Amplified how? Please be more explicit.

Conclusions: An explicit comparison to other methodologies that are similar in some aspect would help communicate what is gained by applying the method presented in this paper. What makes this better than simply considering POCP? Why should people want to use this method?

P20L17-19: Literature exists that also agrees with this point that you are making in these lines about needing further measurement of additional biogenic VOCs, including some of the literature previously referenced, it would be good to include some of these references here.

Minor comments/edits

P3L7-8: it is advised to not only cite reports, but also peer reviewed literature, although the REVIHAAP is in this case quite appropriate.

P7L6-9: For clarity, I would suggest revising the text as follows: 'For 2010-2012, data were available for 27 species at both Harwell and Auchencorth. Concentrations of 6 VOCs at Auchencorth during this period were not above the reported limit of detection (LOD) so their contribution to three regional O₃ increment was not evaluated. For 1999-2001, data were available for 21 VOCs at Harwell only.'

P8L5: '...LOD related non-detects resulted in a small change...'

P8L12: Photochemical has a typo (currently: photochemcial)

P9L9: Remove one of the periods – there are two.

P12L15: '... and maximum monthly total median VOC...' correct?

P13L9: it would be good to include the occasional reminder of 'at Harwell' for the 2001 data, since those years never include any Auchencorth data.

P13L20: '71% of the remaining VOCs' is not clear. Is it that of the total VOCs, 71% were remaining beyond those discussed, or 71% of some other amount of VOCs that is not 100% of all the VOCs measured? If this is also e.g., # of the 27 measured VOCs it might be easier to just state the number of VOCs you are discussing. Similarly, L22 and L23.

P13L20: '...on average 3.4% ± 2.5% to total ...'

P14L16: '...in 2012 are shown in Figure 7.'

P19L7-9: Please specify what the percentage is and ideally then also how much of the total would be accounted for at that point, as it seems that it should be close to 80 or 90%, but it is unclear since I was unsure if the % values refer to %s of the total emissions, or just %s of the VOCs that were not measured.

P19L11: ‘...reductions in individual VOCs ~~from~~ whatever their source(s).’