

Interactive comment on “Source attribution of European surface O₃ using a tagged O₃ mechanism” by Aurelia Lupaşcu and Tim Butler

Anonymous Referee #3

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This is a very nice analysis that provides a lot of useful information and insight regarding the sources of ozone across Europe. The material is appropriate for ACP and I could recommend the paper for publication after it is revised according to my comments below.

General comments: 1) My only concern about the analysis is with regards to the ALPS region. This region combines the high elevations of the Alps (strong influence from long range transport of European and intercontinental origin) with the low elevations of the Po Valley, which is shielded from long-range transport by the Alps and experiences localized and intense air pollution episodes. Given the high variability of source regions and the high variability of local emissions, I don't think that any clear conclusions can be drawn for this region. The authors need to split this region into two parts: 1) the

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Po Valley; and 2) regions above 1500 m elevation. Then we should see that the high elevations have the greatest impact from long-range transport and the low elevations have the strongest impact from local emissions.

2) In general the standard of English is fairly good but it needs a lot of polishing. There are too many instances of grammatical errors or awkward phrasing for me to list individually, but here are a few examples from the first paragraph of the Introduction: “The World Health Organization air quality guideline report that high O₃ concentrations can cause damages to humans and vegetation” “Moreover, it has been shown that tropospheric O₃ also affects radiative forcing and therefore contributing to climate change.” “To maintain a good air quality and understand O₃’s response to climate change, it is important to understand the contribution of different sources of its precursors to the tropospheric O₃ concentration.” Another example is this sentence in the Conclusions, which is difficult to understand: “Thus, we have seen that during the exceedances days, the contribution from local sources sources is ~45 % and 38 % of modeled MDA8 O₃, whilst during nonexceedances values is ~32 % and 2 3% for ALP, respectively GEN.”

Specific Comments:

Page 2, line 5 Here you state: “It has been shown that the background O₃ concentrations have increased during the last several decades due to the increase of overall global anthropogenic emissions of O₃ precursors (HTAP, 2010)” What is meant by background? HTAP uses the term global background to refer to natural ozone that would exist in the absence of anthropogenic emissions. This quantity cannot be measured but must be calculated by global models. Do you really mean to say that the global natural background has increased? Or do you mean that average observed global ozone has increased? According to the extensive review conducted by TOAR-Climate [Gaudel et al., 2018] the current in situ ozone monitoring network is insufficient to quantify ozone changes on the global or hemispheric scale; the available satellite products disagree on the trend, with some showing increases and some showing decreases.

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Line 6 Here you want to demonstrate that ozone air pollution is a current issue, but you only provide one reference that is now 11 years old and only applies to China. Please find additional references that are more current and cover other regions of the world. TOAR has recently published three papers that report present-day ozone using metrics relevant to human health [Fleming et al., 2018], vegetation [Mills et al., 2018] and climate [Gaudel et al., 2018]. These papers also provide up-to-date reviews of the literature concerning the impacts of ozone on humans, vegetation and climate. These papers would be helpful for the Introduction.

In general the standard of English is fairly good but it needs some polishing, for example the following phrases in the first paragraph of the Introduction need more work: “The World Health Organization air quality guideline report that high O₃ concentrations can cause damages to humans and vegetation” “Moreover, it has been shown that tropospheric O₃ also affects radiative forcing and therefore contributing to climate change. “To maintain a good air quality and understand O₃’s response to climate change, it is important to understand the contribution of different sources of its precursors to the tropospheric O₃ concentration.” Another example is this sentence in the Conclusions, which is difficult to understand: “Thus, we have seen that during the exceedances days, the contribution from local sources sources is ~45 % and 38 % of modeled MDA8 O₃, whilst during nonexceedances values is ~32 % and 2 3% for ALP, respectively GEN.”

Page 3, Line 21 Lefohn and Musselman stated that the W126 index “. . . would provide a more appropriate target for air quality management programs designed to reduce emissions from anthropogenic sources contributing to O₃ formation”. I’m not sure why this quote is provided. It seems to imply that W126 is a better metric then AOT40, but there is no agreement among the scientists who study the impacts of ozone on vegetation as to which metric is best. W126 was developed for a few limited crops and trees and is not necessarily applicable to other types of vegetation. For example, Harmens et al. [2018] found that a particular type of wheat is not sensitive to ozone peaks, which means W126 is not the best metric. Mills et al. [2018] give an overview

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of various vegetation metrics and they point out that the flux-based methods are the most accurate, but TOAR did not include these methods because their calculation on the global scale is not yet feasible.

Page 4, Line 4 “Butler et al. (2018) describes in detail” should be “Butler et al. (2018) describe in detail” Because Butler et al. indicates many people, not one.

Page 4, line 18 I don’t understand this phrase: “has been modified to enable to model capacity to be used”

Page 4, Line 19 Should this sentence be a part of the preceding paragraph?

Page 5 line 31 Here and throughout you need to be consistent with regards to the term, ozone concentration. “Ozone concentration” is appropriate when using units of $\mu\text{g m}^{-3}$, but when using units of ppb, the correct term is “ozone mixing ratio”

In many places in the paper the term “NO_x precursors” is used. This seems to be a redundant phrase. Just define NO_x as a precursor gas at the beginning of the paper and then afterwards just use “NO_x” as it will be clear that it is a well-known precursor.

Page 8, line 18 “In all receptor regions, the MDA8 O₃ concentration is dominated by O₃ produced by remote anthropogenic precursors” I think this is overstated because it gives the impression that far more than half of the ozone is from remote anthropogenic precursors. But the range is 30-53%. Please re-phrase this sentence.

Page 8 line 25 This sentence seems out of place and it should appear before the discussion of the stratospheric source.

Page 8 line 29 “Arabic Saudi peninsula” should be Arabian Peninsula

Page 9 line 11 This statement is not quite correct: “Another consequence of enhanced photochemical activity during the summer season is the reduction of stratospheric O₃” It should say: “Another consequence of enhanced photochemical activity during the summer season is that it reduces the relative influence of stratospheric O₃” But in

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addition to changing the relative impact, do the absolute values of stratospheric ozone also diminish in summer, relative to spring?

Page 9 line 31 Is OCN in these studies defined in the same way as in your study? If not then OCN should not be used when referencing the other studies.

Page 12, line 34 I don't understand what is meant by "efficiently complementing". I think you are trying to say that they behave similarly.

Page 13, line 21 Here you say that W126 is more sensitive to local emissions because it does not include a threshold. But, in addition, isn't it also more sensitive to local emissions because this metric places much more weight on the high ozone values, and high ozone is likely to be produced under hot stagnant conditions when local emissions are more important?

References:

Fleming, Z. L., R. M. Doherty et al. (2018), Tropospheric Ozone Assessment Report: Present-day ozone distribution and trends relevant to human health, *Elem Sci Anth*, 6(1):12, DOI: <https://doi.org/10.1525/elementa.273>

Gaudel, A., et al. (2018), Tropospheric Ozone Assessment Report: Present-day distribution and trends of tropospheric ozone relevant to climate and global atmospheric chemistry model evaluation, *Elem Sci Anth*, 6(1):39, DOI: <https://doi.org/10.1525/elementa.291>

Harmens, H., Hayes, F., Mills, G., Sharps, K., Osborne, S. and Pleijel, H., 2018. Wheat yield responses to stomatal uptake of ozone: Peak vs rising background ozone conditions. *Atmospheric Environment*, 173, pp.1-5.

Mills, G, et al. (2018), Tropospheric Ozone Assessment Report: Present-day tropospheric ozone distribution and trends relevant to vegetation. *Elem Sci Anth*, 6(1):47, DOI: <https://doi.org/10.1525/elementa.302>

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