Dear referee#1,
we thank you very much for your in-dpeth review of our manuscript acp-2019-715. Please find our replies to your comments below. Your original comments are repeated in italics, our replies in normal font, and text passages which we included in the text are in bold. Please note, due to the significant changes in the manuscript so many changes have been added that we don't include all revised texts to the reply but refer to the ranges in our diff document.

The authors apply an ozone tagging method in a global chemistry-climate model to attribute the origin of surface ozone pollution in Europe with focus on the Po Valley and the Benelux.

Answer: Please note that we do not apply a global chemistry-climate model only, but an on-line coupled global-regional chemistry-climate model. This allows us to study air pollution in more detail on the regional scale considering also global impacts (e.g. by long-range transport). The focus of the manuscript is on the results of the nested regional model instances, i.e. on the results with 12 km horizontal resolution.

The work is carefully done but it's not clear to me that there is anything new in the methods or results. I felt that I was reading a technical report rather than a scientific paper, with a tedious deluge of numbers and figures but no real new insight about the origin of ozone. The source attribution for ozone is consistent with what has been documented in many previous papers. The model is not particularly successful at reproducing observations, so it's not clear to me that the source attribution here deserves any more confidence than previous studies. I don't think that this paper is up to the scientific standards of ACP. Maybe I'm missing new scientific insights coming from the paper because they are not properly advertised and/or buried. I couldn't find them in the abstract. I would suggest that the authors submit a much shorter paper focused on what is scientifically new in their results, and including proper citation to the literature.

Answer: Thank you very much for pointing this out. While writing the publication, we indeed might have lost ourselves in some details. Obviously, we have not highlighted the novelty of our work in sufficient detail. Therefore, we would like to highlight our novel approach:

- We apply an on-line nested global/regional CCM to account for finer (12 km) resolution in the target area, but also consider consistently the effect of long range transport.
- The attribution is for NO_x **and** NMHCs **concurrently**.
- With our attribution we distinguish four different source regions **and** 10 sectors.
- The focus of our analyses is on the (land) transport sector.

- Besides JJA mean contributions, we also focus on MDA8 ozone.

New insights / highlight:

- We find that the contributions to ozone from individual sectors, which have large NOx but rather few VOC emissions, are estimated to be lower, if their emissions of NO_x and VOCs are regarded concurrently (in comparison to studies which attribute either only NO_x or only VOCs).

  By design, some of our results differ from previous source attribution studies using a NOx or VOC tagging only. We discuss this in detail in Sect. 7. Given the novel approach, we think our study adds additional information to the topic and not least confirms previous findings with a different methodology.

To highlight the novelty in more detail in the revised manuscript, we changed the introduction, and we sharpened abstract and conclusions. Moreover we moved large parts of the model evaluation to the supplementary material, expanded our analysis to JJA 2017 instead of July 2017, added the region Iberian Peninsula in all analyses and shortened the description of previous analyses. In addition, we added a new Section 6 which analyses the ozone regimes in more detail.

You find the changes in the diff version of the manuscript on the following pages:

- page 1 - page 2, l39; revised abstract
- page 3, l72 - page 6, l162 ; revised introduction
- page 9, l237 - page 14 l375 ; shortened evaluation
- page 17, l436 - page 19 l519 ; sharpened analyses of seasonal mean contributions
- page 24, l520 - p 28 ; sharpened analyses of contributions during episodes of large ozone values
- page 29 - page 30 ; new section 6
- page 34, l726 - page 36, l802; revised conclusion

Specific comments:

The introduction discusses at length the difference between perturbation analysis and attribution by tagging. This is an old story and I don't find it particularly interesting. There's nothing wrong with tagging, it just shouldn't be interpreted as a linear response to a perturbation, and we can leave it at that. It would seem more appropriate for the intro to review past relevant studies on attribution of ozone pollution in Europe - this is lacking.

Answer: We agree that the difference between tagging and perturbation is an "old story", however, from discussions during the review process of previous publications and from discussions on conferences etc. we have the feeling that there is still a lack of understanding of the differences in large parts of the scientific community. Also the second referee asks for results on different mitigation scenarios and why source attribution matters. In that view, we are a bit hesitating to remove the (indeed repeated) discussion. To find a compromise between the comments from the two referees we rephrased the part in the introduction.

You find these changes on page 3, l71 - page 5, l157 of the diff document.

Although the writing is generally fine (albeit tedious), there are a lot of minor grammatical mistakes and typos that could be corrected by a copy editor

Answer: For the revision we checked for typos and grammar, but in any case the manuscript will undergo a final check by the journal.

Page 10, line 12: under- rather than overestimated? There is general ambiguity in referring to frequencies as being under- or over-estimated.

Answer: This sentence has been moved to the Supplement and rephrased there.

Page 10, line 19: surface ozone overestimate is attributed to excessive downward mixing but the model also seems to be too high in the free troposphere based on the aircraft comparisons.

Answer: Yes, indeed, we do have a positive bias of the model results compared to observations. However, we do not see a contradiction here, since an overestimated downward transport will result also in a free tropospheric bias. We revised the overall paragraph:
One main reason for the **positive ground-level** ozone bias is a too strong vertical mixing during the night, **mixing in ozone-rich air from the free troposphere to the boundary layer**. This is is a common problem in many models (Travis and Jacob, 2019). **Moreover, also free-tropospheric ozone is biased high (see disucssuion by e.g. Jöckel et al., 2016).** In consequence, simulated contributions from the stratosphere, from lightning, and from N_2O decomposition to ground level ozone are likely biased high.

Page 20, line 26: I'm surprised that ozone would be produced from ships by ship NMHCs. My understanding is that the ozone production efficiency from ship NOx emissions in models is very high because the chemistry is strongly NOx-limited, unless some specific model parameterization is used to age the NOx faster but that doesn't seem to be used here. I don't think that ship NMHC emissions are needed – there is plenty of CO and methane around for ozone production in the NOx-limited regime. I may be wrong but a reference would be helpful.

Answer: As described in the manuscript, the ozone is not produced by ship NMHCs. The ozone is produced by reactions of ship NO_y with NMHCs from evaporation of gas/oil transported (i.e. leaking) **on board** the ships. Indeed, most ozone is formed by NO_y from the ships with HO_2. Please keep in mind that we perform source attribution of NO_x **and** NMHCs **concurrently**, implying that we would not see this effect with a pure NO_x attribution method. By revising the colour scheme as asked by referee#2, it should now be much clearer that this production of ozone from NMHCs along the ship lines are only a secondary effect.

References

Patrick Jöckel, Holger Tost, Andrea Pozzer, Markus Kunze, Oliver Kirner, Carl Brenninkmeijer, Sabine Brinkop, Duy Cai, Christoph Dyroff, Johannes Eckstein, Franziska Frank, Hella Garny, K. Gottschaldt, Phoebe Graf, Grewe Volker, Astrid Kerkweg, Bastian Kern, Sigrun Matthes, Mariano Mertens, and Andreas Zahn. Earth system chemistry integrated modelling (escimo) with the modular earth submodel system (messy) version 2.51. *Geoscientific Model Development*, 9:1153–1200, 03 2016. doi: 10.5194/gmd-9-1153-2016.

K. R. Travis and D. J. Jacob. Systematic bias in evaluating chemical transport models with maximum daily 8 h average (mda8) surface ozone for air quality applications: a case study with geos-chem v9.02. *Geoscientific Model Development*, 12(8):3641–3648, 2019. doi: 10.5194/gmd-12-3641-2019. URL `https://gmd.copernicus.org/articles/12/3641/2019/`.