Dear referee#1
Thank you very much for your review of our manuscript acp-2019-715. Please find our replies to your comments below. In the following, referee comments are given in italics, our replies in normal font, and text passages which we included in the text are in bold.

This publication presents an analysis of the role of transport emissions on different pollutants by using a tagging source apportionment approach. The uncertainties related to the use of different emission inventories are also assessed. The paper is well structured although the English should be reviewed. In this respect, I listed some possible improvements (see the minor comment section) but the whole text would need to be revised. Although I find this work of interest, I listed below some major concerns I have regarding the methodology proposed by the Authors and would appreciate some additional information in the text regarding these points before I could recommend publication.

We thank referee#1 for this overall positive comment and all other comments which helped to improve the manuscript. For the revised manuscript we checked the English language and clarified several issues (see below for our specific comments). Currently, we perform a final proofreading before uploading the revised manuscript.

1. As noted by the Authors in their introduction, sensitivity analysis and tagging approach are two approaches that are used to answer different questions. Sensitivity deliver impacts whereas tagging delivers contributions. While it is rather clear that impacts can be used to inform on the potential effects of emission reductions on air quality levels, it is rather unclear how the contributions estimated for the Authors can be used in practice. In one of their earlier work, the Authors mentioned the possibility of using contributions in complement to the impacts to inform on the potential of emission reductions that go beyond the threshold covered by the perturbation or sensitivity method. But this possibility is not mentioned in this work. On the contrary, confusion is introduced in some sections in which the Author seem to indicate that contributions can be used to support air quality strategies, e.g. in Section 4.2 (first three lines).

Reply: Contribution analyses provide no direct information about potential benefits from emission reductions (see also Thunis et al., 2019). As discussed by Mertens et al. (2018), which was mentioned by referee#1, the combination of the sensitivity approach with the tagging approach can help to better understand the changes in atmospheric composition by specific emission reductions. The goal of this manuscript, however, is not to investigate potential mitigation options. The goal is to quantify the contribution of the (current state of) land transport emissions to ozone and ozone precursors. The tagging method is well suited to answer this question. Such a quantification of the current status is, to our understanding, the first step in understanding the influence of different emission sources on the atmospheric composition, but can of course not replace additional sensitivity simulations. We clarified this in the revised manuscript.

The new section in the Introduction reads:
In contrast to this, Dahlmann et al. (2011) and Mertens et al. (2018) have used a source apportionment method (by a tagged tracer approach, called tagging hereafter) to calculate the contribution of land transport emissions to ozone. The perturbation approach is based on a Taylor approximation to estimate the sensitivity of ozone (or other chemical species) at a base state (w.r.t the chemical regime) to an emission change. The tagging approach, however, attributes all emissions at any base state (w.r.t. the chemical regime) to the corresponding tagged emissions, but gives no information about the sensitivity of ozone to an emission change (see also, Grewe et al., 2010). For a chemical specie that is controlled by linear processes, the perturbation and the tagging approaches lead to identical results, however, the ozone chemistry is strongly non-linear. Therefore, only for small perturbations around the base state (w.r.t. the chemical regime) the response of ozone on a small emission change can be considered as almost linear, but the perturbation approach does not allow for a complete ozone source apportionment (e.g. Wild et al., 2012). As an example, Emmons et al. (2012) have reported that tagged ozone is 2–4 times larger than the contribution calculated by the perturbation approach. As has been outlined in numerous publications, this difference is due to different questions these methods answer. The perturbation approach investigates the impact of an emission change on the mixing ratios of ozone and is therefore well suited to evaluate for example mitigation options. The tagging approach quantifies the contribution of specific emission sources onto the ozone budget for a given state of the atmosphere (Wang et al., 2009; Emmons et al., 2012; Grewe et al., 2017; Clappier et al., 2017; Mertens et al., 2018). These contributions do, however, not necessarily change linearly with potential changes in emissions. The difference between the results from the perturbation and tagging approaches can actually be used as in indicator for the degree of non-linearity of the chemistry as pointed out Mertens et al. (2018) in their equation 6. In the following we use the terms 'impact' to indicate results from perturbation approaches and 'contribution' to refer to results of tagging methods. In this study, we are interested in the contribution of land transport emissions to ozone in Europe. Therefore, we chose a tagging method for source apportionment.

In addition we clarified this at several parts of the manuscript (see 'diff' version). Especially the first part of Section 4.2 reads now: "**To improve the understanding of extreme ozone events, ...**"

2. *Along the same lines, the Authors mention that these findings based on*

tagging are in line with other studies using perturbation methods (see l27 in the discussion section). How can these conclusions be reached when it is clearly mentioned in the introduction that perturbation methods and tagging are expected deliver different results. These two statements contradict each other, unless O3 may be considered as a linear species, in which case both methods would indeed converge to the same conclusions

Reply: We agree with referee#1 that this part of the conclusions was miss-leading in the original manuscript and your comment is very much in line with the comment from referee#2. We rewrote large parts of the conclusions including a more detailed comparison of the results from different tagging methods for Europe. This comparison helped to understand the results of the different tagging approaches in more detail. The new part of the discussion reads:

A detailed comparison of our results with previous studies is complicated: First, we apply one global tag for the land transport sector and do not differentiate between local produced ozone and long range transported ozone. In comparison to our approach similar regional studies usually attribute ozone only to the emissions within the regional domain and attribute long-range transported ozone to the boundary conditions. Second, the tagging methods applied in various studies differ. Third, the applied emission inventories differ, so do ozone metrics and simulated periods. Tagaris et al. (2015), who calculated the impact of different emission sectors on ozone using a 100 % perturbation of the respective emission sectors reported an impact of European road transport emissions of 7 % on average for the maximum 8 hr ozone values in July 2006. In most regions impacts above 10 % have been reported, with maximum local impacts (Southern Germany, Northern Italy) of above 20%. While their largest impacts occur in similar regions as our largest contributions (Southern Germany, Northern Italy), our mean contributions are larger than their impacts, but the maximum contributions are lower than their maximum impacts. Further, around London and in parts of Northern England their impacts (see Fig. 3 therein) are around 2 to 4 %, while our contributions are in the range of 8 to 10 %. Hence, impact and contribution differ largely in these regions. This is in line with previous work, stating that the contributions to ozone are more robust, i.e. less dependent on the background, as the perturbations or impacts (Grewe et al., 2012, 2019). All the studies that we are aware of and which reported contributions of land transport emissions to ozone over Europe using a tagging method either use the CAMx model (CAMx OSAT method, Karamchandani et al., 2017) or the CMAQ model (CMAQ-ISM method, Valverde et al., 2016; Pay et al., 2019). As discussed, these two methods apply a sensitivity approach to check, whether ozone production is NO_x **or** VOC **limited. These previous studies considered only European**

emissions, while we consider the combined effect of European emissions and long range transport. Therefore, one would expect that our contribution analysis shows larger contributions as previous studies. However, our contributions in general are lower compared to previously reported values. As an example, Karamchandani et al. (2017) reported contributions around larger European cities in the range of 24 to 11 %, in Budapest even up to 35 %. Valverde et al. (2016) reported contributions of road transport emissions from Madrid and Barcelona of up to 24 % and 8 %, respectively. Similarly, Pay et al. (2019) diagnosed contributions of road transport emissions on ozone of 9 % over the Mediterranean Sea and up to 18 % over the Iberian Peninsula, however for a specific summer episode only (July 2012). To discuss potential reasons why our contributions are lower compared to previous estimates, we analysed our results for July 2010, to compare these contributions directly with the findings of Karamchandani et al. (2017). As an example, Karamchandani et al. (2017) reported contributions of 17 % around Berlin, while our contributions are in the range of 12–14 %. Further they diagnosed contributions from the biogenic sector of around 11 % around Berlin, while we find contributions of the biogenic sector of around 18 %. Generally, the contributions reported by Karamchandani et al. (2017) seem to be much more variable over Europe compared to our results. A reason for this might be the different treatment in the apportionment of $\mathrm{NO_x}$ and VOC precursors. Land transport emissions contribute mainly to $\mathrm{NO_x}$ emissions, while biogenic emissions are an important source of VOCs. As shown by Butler et al. (2018), anthropogenic emissions contribute most to ozone over Europe, if a $\mathrm{NO_x}$ tagging is applied, while biogenic emissions are the most important contributor, when a VOC tagging is applied (Figs. 3 and 4 therein). Accordingly, those approaches which use a threshold to perform either a VOC or $\mathrm{NO_x}$ tagging, attribute ozone production under VOC limitation mainly to biogenic sources, while under a $\mathrm{NO_x}$ limitation ozone is attributed mainly to anthropogenic sources (including land transport emissions). Most likely this leads to a much stronger variability between anthropogenic and biogenic contributions compared to our approach, where ozone is always attributed to $\mathrm{NO_x}$ and VOC or $\mathrm{HO_x}$ precursors. Similar effects can also be observed when comparing our results to the results of Lupaşcu and Butler (2019), who applied a $\mathrm{NO_x}$-only tagging for the period April to September 2010 and considered regional as well as global sources similar to our approach. They reported contributions of biogenic emissions in Europe for the period July - September between 5 and 13 % over Europe. Our results show contributions of biogenic emissions which are much larger (15 to 26 % for the same period). In there approach, ozone is only attributed to biogenic $\mathrm{NO_x}$ emissions, while we attribute ozone to biogenic $\mathrm{NO_x}$ and VOC emissions. Further, our estimated stratospheric contribution to

ground-level ozone is also larger than the contributions reported by Lupaşcu and Butler (2019). In this case, our results indicate contributions for July to September in the range of 5 to 10 % compared to their 2 to 4 %. Similarly, for lightning-NO_x our model shows larger contributions (6–12 %) compared to the 3–6 % diagnosed by Lupaşcu and Butler (2019). These differences of the contributions for the stratospheric and the lightning category can partly be attributed to the more efficient vertical mixing in COSMO-CLM. Mertens et al. (2020) reported a maximum difference of the contributions from the stratosphere and lightning to ozone between EMAC and COSMO-CLM/MESSy of 30 % at maximum. As the difference between our results and the results of Lupaşcu and Butler (2019) are much larger as these 30 %, the difference can most likely not entirely be attributed to differences in vertical mixing. Rather, the differences can probably be explained by the different contributions of the biogenic category (due to different tagging methods) and by the different contributions of lightning and stratospheric sources. However, the different studies provide not enough insights about the applied emissions (e.g. for lightning-NO_x, soil NO_x and biogenic VOCs) to fully analyse these differences. The discrepancies between the results of the different source apportionment methods show clearly the importance of a coordinated comparison of different source apportionment approaches, as has already been suggested by Butler et al. (2018).

3. In some sections, many numbers are given to characterize the various contributions, e.g. Section 5. A few additional lines to detail the implication these results may have would be useful.

Reply: We thank referee#1 for this suggestion. Generally, our manuscript is composed in such a way that the first sections present only our findings while we discuss the implications in the following sections. Concerning Section 5 we added the following sentence:

These results indicate the importance of land transport emissions for the mixing ratios of reactive nitrogen levels in German cities.

Most of these comments address spelling errors or unclear grammatical sentences. But I would strongly suggest the Authors to review the whole text regarding the English writing.

Reply: Thank you very much for the corrections. We added all of them. For the revised manuscript we checked the manuscript carefully and will perform a final proofreading before uploading it. We are very sorry for the large amount of spelling errors in the original manuscript.

1. In many sentences 'as' is used in place of 'than' (e.g. p19 l27; p22 l6; p26

l25. . .)
Reply: We checked the whole manuscript and fixed it (hopefully) everywhere.

2. P1 l28: teh - the
Reply: Fixed !

3. P3 l20: quantifies - quantified Reply: Fixed !

4. P4 l29: Th - the
Reply: Changed to 'to'

5. P5 l34: an - a
Reply: Fixed

6. P5 l35: to - too
Reply: Fixed

7. P8 l9: not - note
Reply: Fixed

8. P8 l8: party - part
Reply: Changed to parts

9. P19 l16: kept - be kept
Reply: Fixed

10. P19 l 25: the text within parentheses is unclear
Reply: Changed to **(i.e. the emission sectors anthropogenic non-traffic and aviation)**

11. P19 l29: increase - increases
Reply: Fixed

12. P19 l34: all most - almost
Reply: Fixed

13. P22 l14 & 15: sentence is unclear
Reply: We changed the sentence to: **We analyse the contribution of land transport emissions to the ozone budget in Europe by investigating the net ozone production, which is defined as:**

14. P23 l3: second - second most
Reply: Fixed

15. P23 l11: is displayed - are displayed

Reply: Fixed

16. Discussion section: could the Authors add a few words to explain how all these contribution numbers can be validated? Can we use contributions to know which inventory might be closer to the truth?
Reply: This is indeed a good point. What can be done is an evaluation of model results and diagnosed contributions with measurements for specific periods to check, if processes are implemented correctly (or if they are missing). Examples could be periods with large influence of stratospheric ozone (where models should show large stratospheric contributions) or measurements in city plumes, for which models should show a large contribution of ozone from anthropogenic categories. We added a short discussion on this in the revised manuscript. A crucial point is also the differences in the tagging methods, which need to be investigated in more detail to understand strengths and weaknesses of the different approaches better. In our opinion, contributions alone do not help to discuss individual emission inventories. At the end all information (measured and simulated ozone mixing ratios, and contributions) can help to estimate if emission inventories are in a plausible range. However, in our opinion they cannot help to judge, if an emission inventory is right. The additional part in the discussion reads:

Challenging remains also the question on how to evaluate these source apportionment results. Clearly, a comparison of different source apportionment methods would help in revealing individual strengths and weaknesses. In addition, we plan to include source apportionment results in the process of model evaluation (and suggest similar to other modelling groups). By comparing measurements and model results for specific episodes or for specific regions (e.g. in plumes of cities, in regions with strong lightning activity or events of stratospheric intrusions) it can be investigated, if the diagnosed contributions are in a plausible range. Further, the influence of model biases on the analysed contributions can be estimated. A direct evaluation of these contributions, however, is not possible.

17. P25 l3: corresponds - correspond
Reply: Fixed

18. P25 l7: depend - depends
Reply: Fixed

19. P25 l8: contributor - contributors
Reply: Fixed

20. P25 l17: increase - increases
Reply: Fixed

21. P25 l19: regions of - regions with
Reply: Fixed

22. P25 l25: not largest - not the largest
Reply: Fixed

23. P27 l11: by different - between different
Reply: Fixed

24. P27: l28 to 30: please check the use of the word 'uncertainty' which is used many times in a couple of sentences
Reply: We rephrased the sentences to:
Of course, also the uncertainties in the emission inventories for emissions outside of Europe can also influence the contribution analyses considerably, but this has not been investigated in the present study. During summer the differences between the contributions diagnosed using the two emission inventories are larger than the year-to-year variability. Hence, during summer uncertainties of emission inventories for Europe influence the contribution analyses considerably.

25. P28 l2: studies - studied
Reply: Fixed

26. P28 l9: o - ?
Reply: Fixed

27. P28 l9 region - regions
Reply: Fixed

28. P28 l11 increase - increases
Reply: Fixed

29. P28 l19-20: can the Author develop a little bit more on how they plan to use observation data to validate the contributions? I believe this is a key point and one of the major benefits of the tagging approach.
Reply: For the revised manuscript we added some more details about this in the discussion section (see our reply above). In the conclusion section we added a reference to the discussion section.

We are looking forward to your reply,
Mariano Mertens
(on behalf of all co-authors)

References

Butler, T., Lupascu, A., Coates, J., and Zhu, S.: TOAST 1.0: Tropospheric Ozone Attribution of Sources with Tagging for CESM 1.2.2, Geosci. Model Dev., 11, 2825–2840, https://doi.org/10.5194/gmd-11-2825-2018, URL https://www.geosci-model-dev.net/11/2825/2018/, 2018.

Clappier, A., Belis, C. A., Pernigotti, D., and Thunis, P.: Source apportionment and sensitivity analysis: two methodologies with two different purposes, Geosci. Model Dev., 10, 4245–4256, https://doi.org/10.5194/gmd-10-4245-2017, URL https://www.geosci-model-dev.net/10/4245/2017/, 2017.

Dahlmann, K., Grewe, V., Ponater, M., and Matthes, S.: Quantifying the contributions of individual NOx sources to the trend in ozone radiative forcing, Atmos. Environ., 45, 2860–2868, https://doi.org/http://dx.doi.org/10.1016/j.atmosenv.2011.02.071, URL http://www.sciencedirect.com/science/article/pii/S1352231011002366, 2011.

Emmons, L. K., Hess, P. G., Lamarque, J.-F., and Pfister, G. G.: Tagged ozone mechanism for MOZART-4, CAM-chem and other chemical transport models, Geosci. Model Dev., 5, 1531–1542, https://doi.org/10.5194/gmd-5-1531-2012, URL http://www.geosci-model-dev.net/5/1531/2012/, 2012.

Grewe, V., Tsati, E., and Hoor, P.: On the attribution of contributions of atmospheric trace gases to emissions in atmospheric model applications, Geosci. Model Dev., 3, 487–499, https://doi.org/10.5194/gmd-3-487-2010, URL http://www.geosci-model-dev.net/3/487/2010/, 2010.

Grewe, V., Dahlmann, K., Matthes, S., and Steinbrecht, W.: Attributing ozone to NOx emissions: Implications for climate mitigation measures, Atmos. Environ., 59, 102–107, https://doi.org/10.1016/j.atmosenv.2012.05.002, URL http://www.sciencedirect.com/science/article/pii/S1352231012004335, 2012.

Grewe, V., Tsati, E., Mertens, M., Frömming, C., and Jöckel, P.: Contribution of emissions to concentrations: the TAGGING 1.0 submodel based on the Modular Earth Submodel System (MESSy 2.52), Geosci. Model Dev., 10, 2615–2633, https://doi.org/10.5194/gmd-10-2615-2017, URL https://www.geosci-model-dev.net/10/2615/2017/, 2017.

Grewe, V., Matthes, S., and Dahlmann, K.: The contribution of aviation NOx emissions to climate change: are we ignoring methodological flaws?, Environmental Research Letters, 14, 121 003, https://doi.org/10.1088/1748-9326/ab5dd7, 2019.

Karamchandani, P., Long, Y., Pirovano, G., Balzarini, A., and Yarwood, G.: Source-sector contributions to European ozone and fine PM in 2010 using AQMEII modeling data, Atmos. Chem.

Phys., 17, 5643–5664, https://doi.org/10.5194/acp-17-5643-2017, URL https://www.atmos-chem-phys.net/17/5643/2017/, 2017.

Lupaşcu, A. and Butler, T.: Source attribution of European surface O_3 using a tagged O_3 mechanism, Atmos. Chem. Phys., 19, 14 535–14 558, https://doi.org/10.5194/acp-19-14535-2019, URL https://www.atmos-chem-phys.net/19/14535/2019/, 2019.

Mertens, M., Grewe, V., Rieger, V. S., and Jöckel, P.: Revisiting the contribution of land transport and shipping emissions to tropospheric ozone, Atmos. Chem. Phys., 18, 5567–5588, https://doi.org/10.5194/acp-18-5567-2018, URL https://www.atmos-chem-phys.net/18/5567/2018/, 2018.

Mertens, M., Kerkweg, A., Grewe, V., Jöckel, P., and Sausen, R.: Are contributions of emissions to ozone a matter of scale?, Geosci. Model Dev., 13, 363–383, https://doi.org/10.5194/gmd-13-363-2020, URL https://www.geosci-model-dev.net/13/363/2020/, 2020.

Pay, M. T., Gangoiti, G., Guevara, M., Napelenok, S., Querol, X., Jorba, O., and Pérez García-Pando, C.: Ozone source apportionment during peak summer events over southwestern Europe, Atmos. Chem. Phys., 19, 5467–5494, https://doi.org/10.5194/acp-19-5467-2019, URL https://www.atmos-chem-phys.net/19/5467/2019/, 2019.

Tagaris, E., Sotiropoulou, R. E. P., Gounaris, N., Andronopoulos, S., and Vlachogiannis, D.: Effect of the Standard Nomenclature for Air Pollution (SNAP) Categories on Air Quality over Europe, Atmosphere, 6, 1119, https://doi.org/10.3390/atmos6081119, URL http://www.mdpi.com/2073-4433/6/8/1119, 2015.

Thunis, P., Clappier, A., Tarrason, L., Cuvelier, C., Monteiro, A., Pisoni, E., Wesseling, J., Belis, C., Pirovano, G., Janssen, S., Guerreiro, C., and Peduzzi, E.: Source apportionment to support air quality planning: Strengths and weaknesses of existing approaches, Environment International, 130, 104 825, https://doi.org/https://doi.org/10.1016/j.envint.2019.05.019, URL http://www.sciencedirect.com/science/article/pii/S0160412019301539, 2019.

Valverde, V., Pay, M. T., and Baldasano, J. M.: Ozone attributed to Madrid and Barcelona on-road transport emissions: Characterization of plume dynamics over the Iberian Peninsula, Science of The Total Environment, 543, Part A, 670 – 682, https://doi.org/http://dx.doi.org/10.1016/j.scitotenv.2015.11.070, URL http://www.sciencedirect.com/science/article/pii/S0048969715310500, 2016.

Wang, Z. S., Chien, C.-J., and Tonnesen, G. S.: Development of a tagged species source apportionment algorithm to characterize three-dimensional transport and transformation of precursors and secondary pollutants, J. Geophys. Res. Atmos., 114, n/a–n/a, https://doi.org/10.1029/2008JD010846, URL `http://dx.doi.org/10.1029/2008JD010846`, d21206, 2009.

Wild, O., Fiore, A. M., Shindell, D. T., Doherty, R. M., Collins, W. J., Dentener, F. J., Schultz, M. G., Gong, S., MacKenzie, I. A., Zeng, G., Hess, P., Duncan, B. N., Bergmann, D. J., Szopa, S., Jonson, J. E., Keating, T. J., and Zuber, A.: Modelling future changes in surface ozone: a parameterized approach, Atmos. Chem. Phys., 12, 2037–2054, https://doi.org/10.5194/acp-12-2037-2012, URL `https://www.atmos-chem-phys.net/12/2037/2012/`, 2012.