Dear editor

Thank you very much for guiding trough the editorial process.

According to the referee comments we thoroughly revised our manuscript.

The most important changes are:

- We added a additional discussion about the perturbation and the tagging approaches in the Introduction.
- We revised Section 2.1 and included a comparison of our tagging method with other methods.
- We added a Section 2.3 containing a brief comparison of the model results with observational data.
- We added an additional figure in Sect. 4.1 to discuss also the ‘inflow’ towards Europe.
- We divided the region ‘Alps’ in two sub-regions (‘Northern Alps’ and ‘Po Valley’) and adjusted Sect. 4.2 accordingly.
- We revised the Discussion completely.

Furthermore, we have carefully checked the language of the revised manuscript and clarified many issues. This leads to a huge amount of (small) changes in different parts of the revised manuscript. Please note, that we performed one last language check after uploading the replies to referee#1 and referee#2. Therefore, some changes of the revised manuscript differ slightly (wr.t. to the language) from our initial replies. We updated these changes in the attached replies to the referees (given in bold).

Attached are the comments to the two referees (original comments in italic, answers in normal fonts, changes in the revised manuscript in bold) together with the revised manuscript. In the revised manuscript all modifications are highlighted (latexdiff).

We are looking forward to your reply,

Mariano Mertens
(on behalf of all co-authors)

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 an indicator for the degree of non-linearity of the chemistry as pointed out by 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 missleading 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 applied 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 11 to 24 %, 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 NO_x and VOC precursors. Land transport emissions contribute mainly to 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 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 NO_x tagging, attribute ozone production under VOC limitation mainly to biogenic sources, while under a 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 NO_x and VOC or HO_x precursors. Similar effects can also be observed when comparing our results to the results of Lupaşcu and Butler (2019), who applied a NO_x 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 NO_x emissions, while we attribute ozone to biogenic 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 %. 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 discrepancy in the results of the different source attribution methods clearly shows that a coordinated comparison between these methods is important. This have 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. Further, they clearly show the importance of fine resolved emission inventories (and models) for source apportionment of short lived chemical species.

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 of the methods. 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 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)

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Dear referee#2
Thank you very much for your detailed 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.

Mertens et al. perform a source attribution study examining the contribution of different emission sectors to air pollution over Europe, with a focus on ozone as a pollutant, a special focus on emissions from the road transport sector, and a regional focus on Europe and Germany. They employ a uniform methodology for "tagging" the emissions of ozone precursors in a system of coupled models, allowing a consistent downscaling to be made from the global scale to the national scale. Furthermore, they compare two simulations performed with different emission inventories, showing the sensitivity of the sectoral contributions to the way in which the emissions from each sector are represented in the emission inventory. This combination of sensitivity and source attribution reveals some interesting information about the behaviour of of tropospheric ozone in the model system used, for example the particularly strong differences in the contribution of land transport emissions to the higher percentiles of the ozone distribution when a spatially more explicit inventory is used.

Reply: We thank referee#2 for this elaborate summary and the detailed review which helped to improve the manuscript considerably.

The manuscript is clearly within the scope of ACP, and the method clearly has a lot of potential to inform international air quality policy. Unfortunately the manuscript in its current form suffers from a number of serious flaws, which must be corrected before it can be accepted for publication.

Reply: We thank you for your overall positive comment. In accordance with your comments (see below for details) and the comments from referee#1 we strongly revised parts of the manuscript. Currently, a final proofreading is performed after which we will upload the revised manuscript. We hope it can then be accepted for publication.

Firstly, the quality of the written English is terrible. The manuscript is littered with grammatical and spelling errors, and written in a generally inaccessible style. I do not feel that it is my job as a reviewer to provide an exhaustive list of these errors. The authors should seek additional help to get the language up to an acceptable standard. I will give one example though: the very title of the manuscript contains a jarring error. The current title basically implies that ozone causes land transport emissions. Clearly this is the other way around. Land transport emissions happen first, and this leads to ozone production. A grammatically correct title could be "Attributing ozone and its precursors to land transport emissions in Europe and Germany".

Reply: We are really sorry for the errors in the first draft of the manuscript. Of course it is not the work of the referees to perform a language editing. We checked the manuscript in detail and corrected many errors. In addition we revised the title according to the suggestion of referee#2.

In the abstract, the authors state that tagging is "required" and that their method is the "only possible" way to examine global to regional scale effects. This language is way too strong and should be toned down before publication. This is especially true given that the authors themselves state on line 28 of page 25 that their results are "consistent" with a perturbation study, and also given the fact that the experiment design doesn't actually make a distinction between land transport emissions in Europe and the rest of the world.

Reply: We think that the scientific community agrees that tagging methods are the only correct way to calculate contributions (for non-linear species). Impacts and contributions can be similar, but they answer completely different questions. Therefore we don't agree that the language of the sentence '[...]the contribution of land transport emissions to tropospheric ozone cannot be calculated or measured directly, instead atmospheric-chemistry models equipped with specific source apportionment methods (called tagging) are required' is too strong. Yet, we rephrased the other sentence to:

We investigate the combined effect of long range transported ozone and ozone which is produced by European emissions by applying the tagging diagnostic simultaneously and consistently on the global and regional scale.

Page 2, lines 26-27: while this is generally true on very small scales (eg. urban areas), the response of ozone to perturbation of precursor emissions in remote regions has been shown to be approximately linear. See for example Wild et al. (2012) and Turnock et al. (2018). Since the authors are also discussing long-range transport, some additional discussion of this here would be relevant.

Reply: We agree with referee#2 that in remote regions (i.e. with low NO_x mixing ratios) the ozone chemistry is (almost) linear w.r.t. NO_x and VOC perturbations. However, with increasing NO_x mixing ratios the chemistry of ozone cannot be considered any more as linear (see for example Fig. 1 in Grewe et al., 2012). Concerning your comment we think it is essential to further discuss the differences of the tagging and the perturbation method to clarify this point. The perturbation approach is based on a Taylor approximation around a base state w.r.t. the chemical regime (called x_0). The goal of this approach is to estimate a sensitivity (e.g. dO_3/dE, where E are the emissions) of the ozone chemistry around x_0 by a Taylor approximation. This sensitivity can be used to estimate a response of ozone on emission changes (as done by Wild et al., 2012). Clearly, this approximation is only valid around x_0, but not for a different base state $\tilde{x}_0$. Further, only for small perturbations non-linear effects can

be neglected (first elements of Taylor series). This means that in regions with large NO_x emissions non-linear effects can be neglected only for very small perturbations (e.g. 5 %) around x_0. As the approximation is only valid around x_0 an extrapolation to larger perturbations leads to larger errors. Therefore, Wild et al., 2012 introduces a non-linearity factor (equation 6 therein) to account for non-linearities for much larger perturbations as the original 20 %. Further they state: 'For emission reductions greater than 60 % this correction remains insufficient, and we do not expect the parameterization to work as well under these conditions.' Clearly, for such large perturbations the Taylor approximation is not valid anymore. This has been discussed in great detail also by Grewe et al., 2010. The tagging approach, however, works in a completely different way. It does not consider the sensitivity of the ozone chemistry to an emission change. Instead, it attributes ozone at any base state w.r.t. the chemistry x_y to the corresponding emissions. Thus, the non-linearities are implicitly taken into account. However, the tagging approach gives no information about the sensitivity of an emission change (e.g. dO_3/dE). In addition, Wild et al. (2012) clearly state:

'The 20 % emission perturbations applied in the HTAP studies were chosen to be small enough to give an approximately linear response while being sufficiently large to provide robust signals in all models. However, the response of O3 to its precursor emissions is known to be non-linear (e.g. Lin et al., 1988), and it is important to characterize where these non-linearities become significant. Scaling a 20 % emission reduction by a factor of five has been shown to underestimate the response to a 100 % reduction (Wu et al., 2009), and while this underestimation is relatively small for VOC emissions, generally less than 10 %, it can exceed a factor of two for NOx emissions (Wu et al., 2009; Grewe et al., 2010), and shows a strong seasonal dependence (Wu et al., 2009). For this reason the sensitivity approach used in the HTAP studies is unsuitable for deriving a full source apportionment for O_3. However, it does not preclude its use in estimating the impact of less severe emission changes.'

This is clearly in line with our argumentation. Yet, to clarify this point in the revised manuscript we changed the paragraph accordingly:
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 an indicator for the degree of non-linearity of the chemistry as pointed out by 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.

Page 4, lines 3-4: Aren't the last two points in this list in fact exactly the same thing?

Reply: We are sorry for the confusion the sentence caused in the original manuscript. Point 3 is dedicated to year to year variability (e.g. years with large biomass burning emissions or summer heatwaves). Point 4 is dedicated to the seasonal variability (e.g. strong biogenic emissions in summer). To clarify this we rephrased the part in the revised manuscript (see our reply to the next point).

Page 4, line 5: I can see how using two different inventories can somewhat account for uncertainties in the emissions, but three years is way to short a period to account for interannual variability. I also do not see how the model uncertainty or the uncertainty in the choice of source apportionment method is accounted for at all in this experiment design. It's fine to mention that there can be a lot of uncertainty, but the authors should not claim to be doing more to address these uncertainties than they actually are.

Reply: Of course three years are not enough to catch the full range of interannual variability. Referee#2 is completely right that we do not account for model and/or methodological uncertainties (e.g. different source apportionment methods). During the writing process of the manuscript we changed the order of the four points, but forgot to change the sentence on p4 l5. We clarified this accordingly. The changed paragraph reads:

Typically, the uncertainties of such source apportionment studies are large. Reasons are:

- **uncertainties in the models (e.g. chemical/physical parametrizations) and trough the choice of source apportionment methods;**

- **uncertainties of the emissions inventories;**
- **seasonal variability of the contributions caused by meteorological conditions and seasonal cycles of emissions (e.g. stronger biogenic emissions and more active photochemistry during summer than winter);**
- **year to year variability of the contributions caused by meteorological conditions or large emissions of specific sources in specific years (for example yearly differences of biomass burning emissions);**

To account for the uncertainties due to different emission inventories we performed simulations with two different anthropogenic emission inventories. To further account for the seasonal variability we investigate the contributions for winter and summer seasons. In addition, we consider always three simulation years to gain insights in the variability of the contribution in different years. The investigation of uncertainties caused by models and/or source apportionment methods is beyond the scope of this study.

Page 5, lines 32-35: These are the only lines in the paper where the authors discuss model evaluation. I understand that the model has been evaluated elsewhere, and the model is basically as good,bad as other models, but I would appreciate some more discussion about how the model performance could be expected to influence the conclusions of the manuscript. Since the authors also want to use their model to examine extreme ozone events (in Section 4.2), there must be at least some analysis of how well the model is capable of representing these events in comparison with observations.

Reply: We added section (Sect. 2.5) including a short evaluation of simulated ozone concentrations in comparison to Airbase data. This Section reads:

A model set-up very similar to the one used for the present study has been evaluated with observational data by Mertens et al. (2016). Generally, the comparison showed a good agreement with observations. The biases are similar to comparable model systems and exhibit a positive ozone bias and negative biases for NO_2 and CO. One important reason for these biases is the too efficient vertical mixing within the COSMO-CLM model. An evaluation of the ozone mixing ratios simulated by *REF* and *EVEU* have already been presented by Mertens et al. (2020), however, mainly focusing on JJA mean values. To investigate the models ability to represent extreme values, we present a brief evaluation of the simulated ozone concentrations in comparison to the Airbase v8 observational dataset (available at, https://www.eea.europa.eu/data-and-maps/data/airbase-the-european-air-quality-database-8, last access 14.2.2020). As the model resolu-

tion of 50 km is too coarse to resolve hot-spots of individual cities we restrict the comparison to those stations which are classified as area types 'suburban' and characterised as 'background'. We focus on JJA 2008 to 2010 and compare the results to overall 350 measurement stations. The measurements are subsampled at the same temporal resolution (3 hourly) as the model data. The comparison with the observational data shows a positive ozone bias of the model, which has been discussed in previous studies (Mertens et al., 2016, 2020). The average root-mean-square-error (RMSE) over all 350 stations is 29.2 µg m^{-3} for *REF*, and 24.3 µg m^{-3} for *EVEU*, respectively. The corresponding mean biases (MBs) are 26.6 % and 20.5 %, respectively (see Table S1). In addition, we calculated also the RMSE and MB for the *REF* simulation considering only measurements and model data at 12 and 15 UTC. For this subsample, both, RMSE and MB decrease considerably. Accordingly, the largest ozone values during daylight are captured very well by the model. As a more detailed comparison between measurements and model result shows, the overestimation of ozone is particularly strong during night. This can partly be attributed to a too unstable boundary layer during night, which is a common difficulty in many models (Travis and Jacob, 2019). In addition, the too strong vertical mixing in the model leads to positive ozone biases at noon and during the night (see also Mertens et al., 2020, 2016). Currently, further investigations are undertaken, about how this bias could be reduced in the future. Besides the too efficient vertical mixing, also too less ozone deposition during night, too low NO or VOC emissions, and successively underestimated ozone depletion during nights could also partly contribute to this bias. For analysing extreme ozone values, we also compare the 95th percentiles of ozone with measurements (see Fig. S1). Overall, the model is able to capture most of the regional variability of the extreme values over Europe. Near the densely populated regions in Benelux, Germany and Italy, however, the model is not able to reproduce the extremes. In these areas the model resolutions (i.e. also for the 12 km domain, which is not shown here) are too coarse to allow for a representation of extreme ozone values in urban areas. As has been shown by prior studies (e.g. Tie et al., 2010) resolutions below 10 km are required to capture high ozone values near cities. Terrenoire et al. (2015) have noted that even with 8 km resolution the performance of the applied CHIMERE model is better at rural than at urban sites. This underestimation can also be quantified using the RMSEs and MBs for the 95th percentile which are listed in Table S1. These results have important implications for the analyses, which are presented in this manuscript. First of all, the too strong vertical mixing in COSMO-CLM/MESSy leads to a positive bias of the contribution of stratospheric ozone at ground-level. Further, also contributions of lightning and aviation at ground-level are likely larger due to this overestimated

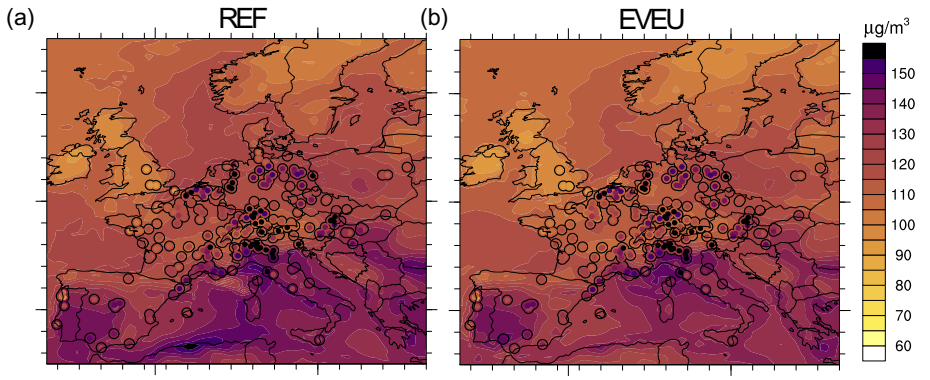

Figure S1: 95th percentile of ozone (in µg/m^3) for the period JJA 2008 to 2010. The background colors show the ozone concentrations as simulated by CM50, the circles represent the location of stations of the Airbase observation data. The inner point represents the measured concentrations, the outer point the concentrations in the respective grid box, where the station is located. All values are based on data every 3 hours.

vertical mixing. This leads to a around 1 percentage point lower contribution of anthropogenic emissions in COSMO-CLM/MESSy compared to EMAC (see Mertens et al., 2020). Due to the coarse model resolution our results are representative for the regional scale, but not for specific urban areas. In these urban areas local emissions and local ozone production/destruction might be more important such that contributions of local sources can be much larger than the values we present. On the regional scale, however, Mertens et al. (2020) showed that the results are quite robust w.r.t. the model resolution (down to 11 km). Because of the stronger ozone bias during night, we further compared the contributions at 12 and 15 UTC with the contributions considering all times of the day. The relative contributions show only small differences, i.e. a slightly larger contribution of anthropogenic emission sources during day (not shown). Therefore, we present always results for all times of the day.

Section 2.1: the authors need to do a lot more here to compare their source apportion- ment method with other methods in the literature. This is especially important, since the authors themselves have stated on Page 4 (line 1) that differences between source apportionment methods are an important source of uncertainty. Kwok et al. (2015) is already mentioned in Section 2.1, and Dunker et al. (2002) is mentioned in the intro- duction. Both of these studies use a regime-dependent attribution methodology, which is actually correctly acknowledged by the authors on page 26 in the Discussion section, but a discussion of how these methodologies differ from the methodology employed by the

Table S1: Root-mean-square error (RMSE, in µ/gm^3) and mean bias (MB, in percent) of the *REF* and *EVEU* simulations compared to Airbase observation data. Given are the scores for the mean values during JJA and DJF, as well as values for the 95th percentile for JJA. For *REF* listed additionally also the scores considering only the values at 12 and 15 UTC.

	RMSE	MB
REF JJA mean	29.2	26.6
REF JJA 12 and 15 UTC	18.7	13.4
EVEU JJA mean	24.3	20.5
REF JJA 95th percentile	26.9	-10.0
EVEU JJA 95th percentile	28.7	-14.2
REF DJF mean	35.1	32.8
EVEU DJF mean	32.8	30.1

authors, and how this could be expected to influence the results of the study is required already in Section 2.1. Similarly, since the authors are also considering the global scale, they should also put their methodology into the context of the existing techniques for source attribution at the global scale. The authors already cite Emmons et al. (2012) elsewhere in the paper, but do not mention this work in Section 2.1, where it would be appropriate to have some discussion of how these methods differ, and how this might influence the results of the study. One very important difference is that Emmons et al. (2012) only consider NOx as a precursor of ozone, while the technique employed by the authors combines the effects of both NOx and VOC precursors. Similarly, the study of Butler et al. (2018) is also missing from the discussion. Butler et al. (2018) account for effects of both NOx and VOC as ozone precursors, but they make some very different design decisions to the technique employed by the authors. The authors must do more to put their method in the context of the previous work, and discuss the relative strengths and weaknesses of the approach they have chosen.

Reply: The method we apply has been discussed in detail by Grewe et al. (2017). Therefore, the intention of this section was only to recap the general idea of the applied tagging method to the reader. It was never indented as a full discussion of our tagging approach compared to other approaches. However, we agree with referee#2 that a short discussion about the different approaches is helpful here, as these differences will also be discussed in the discussion. The newly added part reads:

Some of the categories listed in Table 3 are not directly associated with emission sectors. These categories are stratosphere, CH_4 **and** N_2O**. All ozone which is formed by the photolysis of oxygen, i.e**

$$O_2 + hv \longrightarrow O(^3P) + O(^3P), \quad (1)$$

is labelled as stratospheric ozone. The degradation of N_2O is a source for NO_y (and loss a of ozone) by the reaction:

$$N_2O + O^1D \longrightarrow 2NO. \tag{2}$$

The degradation of CH_4 is considered as source of $NMHC^{CH_4}$. This refers to the reaction:

$$CH_4 + OH \longrightarrow CH_3O_2 + H_2O. \tag{3}$$

As have been discussed recently in detail by Butler et al. (2018) all tagging methods are based on specific assumptions and have specific limitations. The scheme of Grewe et al. (2017), which we apply in the current study, is based on specific assumptions, which differ from other tagging schemes used in regional and global models. One important difference is the question whether ozone formation is attributed to NO_x or VOC precursors. The schemes which are available in the regional models CMAQ (called CMAQ-ISM, Kwok et al., 2015) and CAMx (called CAMx OSAT, Dunker et al., 2002) use threshold conditions to check, whether ozone formation is NO_x or VOC limited. Depending on this the production is attributed to NO_x or VOC precursors only. The scheme of Emmons et al. (2012), applied on the global scale, tags only NO_x and therefore ozone production is only attributed to NO_x precursors. Based on the work of Emmons et al. (2012), Butler et al. (2018) presents a scheme, which attributes ozone formation either to NO_x or VOCs (implying that usually 2 simulations, one with NO_x and one with VOC tagging, are performed). This scheme has also been applied by Lupaşcu and Butler (2019) in a regional model simulation over Europe, using only the NO_x tagging scheme. Compared to these schemes the scheme of Grewe et al. (2017) attributes ozone production always to all associated precursors (i.e. NOx, HO_2 and VOCs) without any threshold conditions. In VOC limited regions, this approach leads to the effect that a NO_x emission reduction of an emission sector reduces the contribution of that sector, and increases the contribution of the other sectors. In contrast, a reduction of VOC emissions decreases the contribution of the respective sector only. The latter is similar to the approaches integrated in CMAQ or CAMx, which attribute ozone production in the case of a VOC limit to VOC precursors only. Compared to a NO_x tagging, our approach leads to lower contributions of NO_x sources, since they compete, not only with other NO_x sources, but also with VOC sources. Because of the family concept, which is necessary to keep the memory consumption and the computational costs low, the tagging method applied in our study can lead to some unphysical artefacts. As an example, Grewe et al. (2017) discuss the production of PAN by NMHCs from CH_4 degradation. Further, due to the combinatorial approach for instance also NMHCs from stratospheric origin can occur in small

amounts, which is also an unphysical artefact. The main reason for this is the definition of the PAN family, which transfers tags from NO_y to NMHCs. Other tagging schemes have specific issues as well. As an example, the scheme of Emmons et al. (2012) does not neglect the O_3-NO_x null cycle, which leads to an overestimation of local sources compared to long range transport sources (see also Kwok et al., 2015). Overall, the impacts of the underlying assumptions on the results are difficult to quantify. Therefore, it is important to study effects of different emission sources with different methods (at best in the same model framework), in order to understand better the strengths and weaknesses of the different approaches and their impact on the source apportionment results.

Also in Section 2.1, the authors could briefly mention how stratospheric ozone is tagged in their approach, since this does not fit into the framework of their Equation 2.

Reply: We added a note about tagging of stratospheric ozone in the revised manuscript (see reply above).

Section 2.2: The authors should make it clear that the tags are applied globally, with no distinction between emissions in Europe and the rest of the world. This is acknowledged later in the manuscript, but the reader would benefit from having this made clear already in this section.

Reply: We added a note in the Section which reads:

In the configuration of the tagging method applied for the present study we use only one global tag for every source category. While this allows to investigate the contributions of all global emissions of a specific emission source to ozone mixing ratios, we are not able to separate contributions from local and long range transport (i.e we cannot separate contributions from, for example, European and Asian land transport emissions to European ozone levels, but we can quantify the contribution of global land transport emissions to European ozone levels).

Page 10, lines 17-24: For some additional context here, it would be nice to know how the proportional contributions of land transport to ambient modelled NOy compare to the proportional contribution of land transport to total NOx in the inventories. Is the contribution as would be expected from simply looking at the emissions, or is it disproportionally higher or lower?

Reply: This is indeed a good question. We calculated the relative share of land transport emissions to all anthropogenic + soil NO_x emissions for June (see Fig. S2, which we add also to the revised Supplement). The contributions of

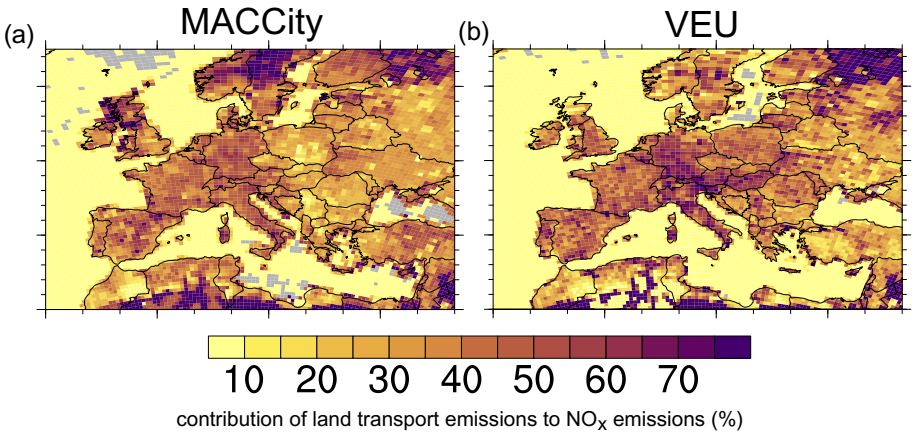

Figure S2: Relative contribution of land transport NO_x emissions to all other emissions (considering soil-NO_x, shipping, anthropogenic, AWB and biomass burning emissions; in %) for July 2009; (a) for the MACCity emission inventory and (b) for the VEU emission inventory.

land transport emissions are in the range of 50 % to 70 %. The contribution is larger in the VEU emission inventory compared to the MACCity emission inventory. The contributions of the emissions are in a similar range as the contribution of land transport emissions to NO_y, however, the regional distribution differs slightly. Near the hot-spots (e.g. Paris) we found smaller relative contributions of land transport emissions to NO_y, while the values are larger in rural areas. We added a note about this in the revised manuscript:

The relative contribution of land transport emissions to ground level NO_y is in the range of 40 % to 70 % in most parts of Europe (see Fig 4). These relative contributions are similar as the share of land transport NO_x emissions to all NO_x emissions (see Fig. S9 in the Supplement), but compared to the share of the emissions the contributions to NO_y are slightly lower near hot-spots, and larger in rural areas.

Section 4.1, page 15: The authors rightly interpret the ozone due to land transport in DJF as coming from long-range transport. I also understand that the limits of the experimental design (one global tag for land transport) make it hard to say anything about long-range transport in JJA, when local photochemistry is more active. But could it be possible to try? For example, could they look at the land transport contribution at the western boundary of the refined grid in JJA, and use this as a rough estimate of the contribution of land transport (and other sectors) in remote regions to baseline ozone in Europe? This could add a lot of value to the study and would be highly relevant for international policymaking.

Reply: This is indeed a good point. As mentioned by referee#2 we cannot di-

rectly estimate the relative importance of 'global land transport emissions' compared to 'European land transport emissions', as we consider only one global tag. To answer this question in detail more tags would be necessary. Based on your suggestion we added a Figure to this Section (see Fig S3) in which we show area averaged contributions for different categories and for different regions. One region we defined here is called inflow and spreads over a large area of the western boundary of the finer domain. We added a paragraph describing this in the manuscript. As you see, we added also a separation between Northern Alps and the Po Basin (see our answer below).

To quantify the contributions of land transport emissions and other emission sources in different regions in more detail, Fig. S3 shows area-averaged relative contributions for JJA and DJF for the *REF* and *EVEU* simulations (absolute contributions are given in Table S1 to Table S8 in the Supplement). The geographical regions were defined according to the definitions of the PRUDENCE project (Christensen et al., 2007), but slightly modified. The region Alps was split up in two separate regions called 'Northern Alps', defined as rectangular box ($46° : 48°$ N and $9° : 13°$ E), and 'Po Valley' ($44° : 46°$ N and $5 : 15°$ E). Note, however, that the region Northern Alps contains parts of Switzerland and Southern Germany, which are still rather flat and subject to large land transport emissions. In addition, we defined a region called 'inflow' ($40° : 60°$ N and $-13° : -11°$ E). This region is used to quantify contributions in the air advected towards Europe. A figure summarizing the definition of all regions is part of the Supplement (Fig. S12). The relative contribution of land transport emissions in the 'inflow' region is about 9 % and very similar in both seasons and for both European emission inventories. During DJF the contributions in all regions are very similar. During JJA the contribution of land transport emissions increases in most regions compared to the 'inflow' (≈ 9 %). In the Po Valley the contribution reaches up to 16 %. Unfortunately, the difference between the contribution in a specific region compared to the contribution in the region 'inflow' cannot be used to calculate O_3^{tra} from European emissions. Such a calculation requires different tags for global and European land transport emissions. The relative contribution of other anthropogenic emissions in the 'inflow' region (≈ 34 %) is also very similar in both seasons. During DJF the contributions in the different regions remain very similar to the contributions in the 'inflow' region. During summer, in contrast, a West-East gradient of the contribution of anthropogenic emissions is present over Europe with a decrease of the contribution of up to ≈ 27 % in Eastern Europe. This decrease is mainly caused by the seasonality of the different emissions (discussed further below). The biogenic emission category shows different relative contributions in the 'inflow' region during DJF (≈ 11 %) compared to JJA (≈ 14 %), which is mainly

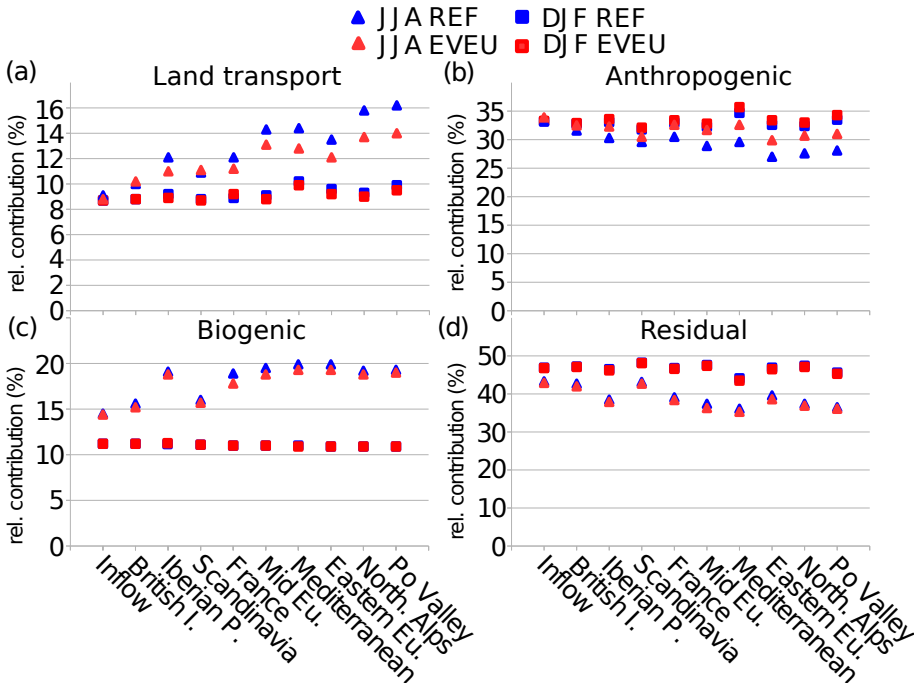

Figure S3: Relative contributions to ground-level ozone (in percent) area averaged in different geographical regions for DJF 2008 to 2010 (triangles) and JJA 2008 to 2010 (squares). Shown are the results of the *REF* (blue) and the *EVEU* simulations (red) for (a) the land transport category, (b) the anthropogenic emissions, (c) the biogenic category, and (d) all other categories. For simplicity the anthropogenic contains the categories anth. non-traffic, aviation and shipping. The residual contains all other categories. The vertical-axis scale differs for (a) to (d).

caused by the strong increase of biogenic emissions during summer compared to winter. In the different regions the relative contributions increase during JJA compared to DJF, and, compared to the 'inflow' up to ≈ 20 %. The contribution of all other tagging categories during DJF is around ≈ 47 % in most regions, and ranges between 41 % and 36 % during JJA.

Page 16, line 3: the seasonal cycle of photochemical activity also plays a role here.

Reply: Indeed. We changed this sentence to:

This seasonal cycle is caused by a complex interplay of the seasonal cycles of different **emission sources, meteorology and photochemical activity.**

Page 16, line 8: is there any way in this study to separate the influence of soil NOx and biogenic VOC? Or are these two different sources inextricably joined together into the "biogenic" sector?

Reply: As they are emitted in the same category there is no possibility to separate them anymore from the simulation results. However, we see where this can deliver important insights, and we are currently revising the tagging method in such a way that these two emissions could be handled separately.

Section 4.2: As mentioned earlier, it would be nice to know how well the model is capable of reproducing the extreme values of ozone as measured. If the model is doing a good job at this, then the results reported here could help to understand these extreme ozone measurements. If the model is not doing well at this, then the results reported here could potentially provide information about systematic model biases, and point the way towards improving the model. As it currently stands, it is not clear at all how these results should be interpreted.

Reply: As discussed above we added a short section with a model evaluation to the revised manuscript. This evaluation shows that the model is able to reproduce the measured 95th percentile of ozone values quite well on the rural scale, but for strong local ozone enhancements the resolution of our model is too coarse (e.g. Tie et al., 2010). The evaluation clearly shows that the model results are not well suited for analysis of the contributions during extreme ozone events on the levels of individual cities. In our analysis, however, we focused on larger geographical regions. We think that on the basis of these larger geographical regions the model results are well suited to investigate the general trends of ozone contributions. Further, our finding that the relative contribution of land transport emissions increase during extreme ozone events compared to the mean conditions is in line of Valverde et al. (2016). They reported a large importance of land transport emissions during high ozone events for Barcelona and Madrid surroundings.

Page 19, line 11: The region "Alps" includes the Po Valley. Does this mean that high mountains are in the same region as a polluted valley? The influences on air quality would be expected to be very different in these regions. High mountains will be more influenced by the free troposphere (and long-range transport), while the valley will be more influenced by local sources. Furthermore, "Alps" and "Po Valley" are used individually in this section and elsewhere in the manuscript. It is not always clear which region is meant. The authors could consider disaggregating this region into two sub- regions for their analysis (which could be quite informative), or at least being clearer about exactly which region they are referring to throughout the text.

Reply: Yes indeed, the region called 'Alps' includes the Po Valley and the Alps. The main intention for this was that we wanted to stick to the geographical

regions defined in the PRUDENCE project. However, we agree that from the point of view of air quality these regions strongly differ. To take this into account, we split the region 'Alps' in two subregions called 'Northern Alps' (defined as 44° : 48° N and 5° : 15° E) and 'Po Valley' (defined as 44° : 46° N and 9° : 13°E) . However, the results for both regions are still very similar. The main reasons are:

- The region Northern Alps contains parts of Southern Germany and also Switzerland, were the mountains are not very high, and much traffic is present.
- Even in the 'higher alps' there are some very important roads with large land transport emissions (e.g. Brenner and Inn valley) which can be clearly seen in the emission inventory. On the 50 km resolution these emissions are mixed over quite large regions.

To better represent the sharp contrast between Alps and the Po Valley a much finer resolution (and fine resolved emission inventories) are necessary, which pose challenging tasks for the future.

Page 19, lines 15-16: the discussion about "uncertainties" in the inventory is very vague here. Could the large range in the contribution of land transport to extreme ozone when using EVEU emissions be related to the higher spatial heterogeneity and existence of more "hot spots" in this inventory compared with REF? There could potentially be some important information here about the need to get the distribution of NOx right in order to capture the high ozone events. A comparison of the REF and EVEU ozone timeseries with some measurements from urban background stations during extreme events could potentially add a lot of value here.

Reply: It is indeed interesting to investigate how different geographical distributions of NO_x emissions could influence the ability of the model to simulate high ozone events. This issue has partly been investigated in previous publications (Tie et al., 2010; Markakis et al., 2015). Compared to these previous studies, the resolution applied here is rather coarse (50 km). The 95th percentiles of ozone for *REF* and *EVEU* are rather similar (see also the newly added evaluation section). When comparing individual stations during specific periods, we noticed that maximum ozone values are not better represented by EVEU compared to REF.
As the 95th percentiles of the ozone values are very similar, we think that the differences of the contributions between the two emission inventories are only caused by the different geographical and sectoral distributions. To clarify this we rephrased and extended the discussion. However, for follow up work we agree that this is still an interesting question and should be further investigated using an improved model set-up at finer resolution. The modified text reads:

The ozone values at the 95th percentile (see Sect. 2.3) and at the other percentiles (see Figs. S1 and S2 in the Supplement), however, are similar for *REF* and *EVEU* (i.e. none of the emission inventories leads to strongly different representation of extreme ozone events in the model). Accordingly, the discussed differences of the relative contributions are not caused by a different representation of the ozone values themselves, but only due to the different geographical and sectoral distributions of the emissions in *REF* and *EVEU*. This demonstrates the large uncertainty, especially for contributions during high ozone events, of the source apportionment analyses which is caused by the uncertainties of emissions inventories (e.g. geographical distribution of emissions, total emissions per sector). These uncertainties must be taken into account in source attribution studies focusing on high ozone events.

Page 23, line 4: the results are not "rather similar", but actually have some important differences, which are subsequently discussed. I think what the authors are trying to say here is that the contribution of land transport is similar in each case, but this is not the meaning which comes across.

Reply: We agree with referee#2 that our original intention of the discussion does not come across. Also the comment from referee#1 shows that this part of the discussion caused confusion. Therefore we revised this part of the discussion completely, taking also into account some more (recently published) work to discuss potential reasons for the differences between the results of the different source apportionment results (taking also into account one of the next comments from referee#2). 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 applied 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 11 to 24 %, 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 NO_x and VOC precursors. Land transport emissions contribute mainly to 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 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 NO_x tagging, attribute ozone production under VOC limitation mainly to biogenic sources, while under a 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 NO_x and VOC or HO_x precursors. Similar effects can also be observed when comparing our results to the results of Lupaşcu and Butler (2019), who applied a NO_x 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 NO_x emissions, while we attribute ozone to biogenic 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 %. 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 discrepancy in the results of the different source attribution methods clearly shows that a coordinated comparison between these methods is important. This have already been suggested by Butler et al. (2018).

Page 25, lines 25-26: This sentence basically conveys no meaning and could be easily deleted with no loss to the manuscript. Alternatively the authors could try to be clearer about what they mean here.

Reply: We rephrased the sentence to:

The result that regions are hot-spots for NO_y from land transport emissions, but not for O_3 from land transport is counter intuitive. The reasons for this is that large amounts of NO_x emissions alone

are not sufficient for large ozone production. This is caused by the non-linearitiy of the ozone chemistry and the strong interdependence of ozone production and meteorological conditions (e.g. Monks et al., 2015).

Page 25, last paragraph: if the previous work only accounts for the contribution of Euro- pean land transport emissions to European ozone, and the current study also includes global emissions, then shouldn't the current study result in a higher contribution than the previous work? The opposite appears to be the case. Can the authors explain this apparent discrepancy?

Reply: We agree. As already discussed above, we tried to clarify this in the new discussion. The differences between the studies, however, are so large that we cannot fully explain the discrepancies, but the discussion hopefully provides some insights. What is really needed to understand the differences between the tagging methods is a detailed inter-comparison of them.

Page 26, line 23: the authors appear to be concluding from the strong influence of the "biogenic" sector that soil NOx emissions are strongly influencing ozone. But couldn't this also be biogenic VOC? How do they separate the influence of these two different sources? A comparison with Butler et al. (2018) could be instructive here, since in that study the separate roles of NOx and VOC as ozone precursors were examined. Comparison of their Figure 3 and Figure 4 indicates that biogenic VOC make a larger contribution to European ozone in summer than biogenic NOx. The authors should discuss this here.

Reply: Of course also biogenic VOCs are very important for the ozone production. As discussed above, we cannot differentiate between ozone produced by biogenic VOCs and soil-NO_x, as we join them together in one category. The soil-NO_x emissions are an important contributor to the NO_x emissions in Europe in summer (see Fig. S4 in the Supplement). Uncertainties of these emissions cause uncertainties of the simulated contributions. However, also the biogenic VOC emissions are uncertain. Therefore, we rephrased this part to clarify that we do not want to say that biogenic VOCs are not important. The discussion of the importance of biogenic VOCs and NO_x with the reference to Butler et al. (2018) was already introduced in the revised discussion above.

Page 28, lines 14-15: the future work proposed by the authors would indeed be ex- tremely interesting from a policymaking perspective. If possible, they should also in- clude as many other sectors as possible. This could help to inform decisions about where emission reductions would be most effective.

Reply: Thanks for this positive comment. Actually, the main focus is on land transport, but we intent to investigate also other categories.

Page 28, lines 28-29: again, it appears that the authors are over-interpreting

their results when they conclude that soil NOx has a strong influence on European ozone levels.

Reply: We guess referee#2 meant lines 18-19. Here we write: 'Here, the focus should not only be on the land transport emissions, but also on other important emissions, including especially biogenic and soil-NO_x emissions, which have large uncertainties and contribute strongly to European ozone levels.' We do not see where we over-interprete the results. However, to make the sentence more clear we add 'VOC' to biogenic:

'Here, the focus should not only be on the land transport emissions, but also on other important emissions, including especially biogenic **VOCs** and soil-NO_x emissions, which **are subject to** large uncertainties and contribute strongly to European ozone levels. '

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

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Attributing ~~land transport emissions to~~ ozone and ~~ozone~~ its precursors to land transport emissions in Europe and Germany

Mariano Mertens[1], Astrid Kerkweg[2,a], Volker Grewe[1,3], Patrick Jöckel[1], and Robert Sausen[1]

[1]Deutsches Zentrum für Luft- und Raumfahrt, Institut für Physik der Atmosphäre, Oberpfaffenhofen, Germany
[2]Institut für Geowissenschaften und Meteorologie, Rheinische Friedrich-Wilhelms-Universität Bonn, Germany
[3]Delft University of Technology, Aerospace Engineering, Section Aircraft Noise and Climate Effects, Delft, the Netherlands
[a]now at: IEK-8, Forschungszentrum Jülich, Jülich, Germany

Correspondence: Mariano Mertens (mariano.mertens@dlr.de)

Abstract. Land transport is an important emission source of nitrogen oxides, carbon monoxide and volatile organic compounds~~, which serves as precursors for tropospheric ozone. Besides the direct negative impact~~ . The emissions of nitrogen oxides ~~, air quality is also affected by these enhanced ozone tropospheric ozoneconcentrations. As ozone is radiativly active , its increase contributes to climate change~~affect air quality directly. Further, all of these emissions serve as precursor for the formation of tropospheric ozone, thus leading to an indirect influence on air quality. In addition, ozone is radiatively active and its increase leads to a positive radiative forcing. Due to the strong non-linearity of the ozone chemistry, the contribution of ~~land transport emissions to tropospheric~~ emission sources to ozone cannot be calculated or measured directly~~, instead~~ . Instead, atmospheric-chemistry models equipped with specific source apportionment methods (~~called tagging~~ e.g. tagging methods) are required. In this study we investigate the ~~contributions~~ contribution of land transport emissions to ozone and ozone precursors using the MECO(n) model system~~, coupling~~ . This model system couples a global and a regional chemistry climate model ~~, which are~~ and is equipped with a tagging diagnostic. ~~For the first time the effects~~ We investigate the combined effect of long range ~~transport and regional effects of regional emissions are investigated. This is only possible by applying a tagging method~~ transported ozone and ozone which is produced by European emissions by applying the tagging diagnostic simultaneously and consistently on the global and regional scale. We performed two ~~three-year simulations~~ simulations each covering three years with different anthropogenic emission inventories for Europe~~by applying our global model with~~ . Therefore, we applied two regional refinements, i.e. ~~a European nest~~ one refinement covering Europe (50 km resolution) ~~in the global model and a German nest~~ and one covering Germany (12 km resolution)~~in the European nest. We find~~ . The diagnosed absolute contributions of land transport emissions to reactive nitrogen (NO_y) near ground-level are in the range of 5 to 10 nmol mol^{-1}~~, corresponding to~~ . This corresponds to relative contributions of 50 to 70 %~~of the ground level values~~. The largest ~~contributions are~~ absolute contributions appear around Paris, Southern England, Moscow, the Po Valley, and Western Germany. ~~Carbon monoxide contributions~~ The absolute contributions to carbon monoxide range from 30 nmol mol^{-1} to more than 75 nmol mol^{-1} near emission ~~hot spots~~ hot-spots such as Paris or Moscow. The ~~contribution of~~ ozone which is attributed to land transport emissions ~~to ozone show~~ shows a strong seasonal cycle ~~which~~ with absolute contributions of 3 nmol mol^{-1} during winter and 5 to 10 nmol mol^{-1} during summer. This corresponds to relative contributions of 8 to 10 % during winter and up to 16 % during summer. ~~Those~~ The largest values during summer are confined to the Po Valley, while the ~~contribution in Western Europa~~

contributions in Western Europe range from 12 to 14 %. ~~The ozone contributions are robust.~~ Only during summer the ozone contributions are slightly influenced by the anthropogenic emission inventory, but these differences are smaller than the range of the seasonal cycle of the contribution to land transport emissions. This cycle is caused by a complex interplay of seasonal cycles of other emissions (e.g. biogenic) and seasonal ~~difference~~ variations of the ozone regimes. ~~This small difference of the ozone contributions due to the emission inventory is remarkable as the precursor concentrations (and) are much more affected by the change.~~ In addition, our results suggest that during events with large ozone values the ~~contribution~~ contributions of land transport ~~emissions~~ and biogenic emissions to ozone increase strongly. Here, the contribution of land transport ~~emission~~ emissions peak up to 28 %. Hence, ~~land transport is~~ our model results suggest that land transport emissions are an important contributor ~~to events of~~ during periods with large ozone values.

1 Introduction

Mobility plays a key role in everyday life, ~~involving~~ which involves the transport of goods and persons. Most of the transport processes rely on vehicles with combustion engines, which emit not only CO_2, but also many gaseous and particulate components, such as nitrogen oxides (NO_x), volatile organic compounds (VOCs), carbon monoxide (CO) or black carbon.

The transport sector with the largest emissions is the land transport sector (involving road traffic, inland navigation and trains). Even ~~tough~~ though the global emissions of many chemical species from the land transport sector have been decreased (e.g. Crippa et al., 2018), the emissions are still very large. For Europe and North America the emissions of NO_x from road traffic have been recently discussed in the public (e.g. Ehlers et al., 2016; Ntziachristos et al., 2016; Degraeuwe et al., 2017; Peitzmeier et al., 2017; Tanaka et al., 2018). NO_x emissions influence the local air quality and lead to exceedances of the nitrogen dioxide (NO_2) thresholds in many cities. Furthermore, NO_x plays an important role for the tropospheric ozone chemistry and serves, together with CO and VOCs, as precursor for the formation of tropospheric ozone (e.g. Crutzen, 1974). Ozone is a strong oxidant and affects air quality (e.g. World Health Organization, 2003; Monks et al., 2015). Large ozone levels impact the vegetation and decrease crop yield rates (e.g. Fowler et al., 2009; Mauzerall et al., 2001; Teixeira et al., 2011). Furthermore, ozone is radiatively active and thus contributes to global warming (e.g. Stevenson et al., 2006; Myhre et al., 2013).

Many studies have been performed ~~investigating~~ which investigated the influence of land transport emissions ~~on~~ to ozone on the global scale (e.g. Granier and Brasseur, 2003; Niemeier et al., 2006; Matthes et al., 2007; Hoor et al., 2009; Dahlmann et al., 2011; Mertens et al., 2018)~~, showing~~ . All of them showed that land transport emissions impact ozone concentrations considerably on the global scale, especially on the Northern hemisphere. As ~~has been~~ outlined by Mertens et al. (2018), these global studies have ~~applied~~ used different methods, ~~rendering a~~ making a direct comparison of the results difficult. Mostly, the so called sensitivity method (or perturbation approach) has been used, ~~comparing results of a reference simulation with the results of a simulation in which changed emissions for~~ in which the results of two different simulation are compared, one simulation with all emissions and one simulation where the emissions of the sector of interest are ~~applied. This method calculates the impact on the concentration resulting from a change of emissions.~~ reduced. In contrast to this, Dahlmann et al. (2011) and Mertens et al. (2018) have used a source apportionment method (by a tagged tracer approach~~) which calculates~~ , called tagging hereafter)

to calculate the contribution of land transport emissions to ozone. ~~This contribution is the part of the concentration from a specific pollutant attributed to the emissions of one specific emission source. Due to the non-linearity of the ozone chemistry the perturbation and the tagging approachgive answers~~ 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 ~~and therefore lead to different results (Wang et al., 2009; Emmons et al., 2012; Grewe et al., 2017; Clappier et al., 2017; Mertens et al., 2018). In teh~~ 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. 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 an indicator for the degree of non-linearity of the chemistry as pointed out by Mertens et al. (2018) in their equation 6. In the following we use the terms 'impact' to indicate results from ~~a sensitivity analysis~~ perturbation approaches and 'contribution' to refer to results of ~~source apportionment studies. Here~~tagging methods. In this study, we are interested in the contribution of land transport emissions to ozone in Europe. Therefore, ~~a source apportionment method is applied~~we chose a tagging method for source apportionment.

The studies discussed above investigated the effect of land transport emissions on the global scale. These results of global models, however, give only very limited information on the contribution of the land transport (or other) emissions to ozone levels on the regional scale, especially as simulated ozone concentrations depend on the model resolution (e.g. Wild and Prather, 2006; Wild, 2007; Tie et al., 2010; Holmes et al., 2014; Markakis et al., 2015). Even ~~tough~~though, land transport is, besides other anthropogenic emissions (e.g. Matthias et al., 2010; Tagaris et al., 2014; Aulinger et al., 2016; Yan et al., 2018) and biogenic emissions (e.g. Simpson, 1995; Solmon et al., 2004; Curci et al., 2009; Sartelet et al., 2012), an important source of ozone precursors in Europe, only few studies ~~have been performed investigating~~ investigated the influence of European land transport emissions on ozone. Reis et al. (2000) have investigated the impact of a projected change of road traffic emissions from 1990 to 2010 on ~~ground level~~ ground-level ozone in Europe, reporting a general decrease of ozone levels due to emission ~~reduction~~reductions. Similarly, Tagaris et al. (2015) have ~~applied the perturbation approach to quantify the~~ quantified the impact of ten different emission ~~scenarios~~ sources on European ozone and PM2.5 levels using the CMAQ model for a specific period (July 2006). Tagaris et al. (2015) have ~~quantified~~ reported an impact of road transport emissions on the maximum 8-hour ozone mixing ratio of 10 % and more in Central Europe. Compared to this, Valverde et al. (2016) have used a source apportionment

method integrated in CMAQ (Kwok et al., 2015) to investigate the contributions of the road traffic ~~emission~~ emissions of Madrid and Barcelona to ozone~~levels on the Iberian peninsula, reporting values~~ . They reported contributions of 11 to 25 % to ozone on the Iberian Peninsula. Similarly, Karamchandani et al. (2017) have applied the source apportionment technique integrated in CAMx (Dunker et al., 2002) to calculate the contribution of eleven source categories on ozone concentrations for one summer and one winter month in 2010, focusing on 16 European cities. Generally, Karamchandani et al. (2017) have reported contributions of 12 to 35 % of the road traffic sector on the ozone levels in different cities. ~~However, in~~ In accordance with other studies Karamchandani et al. (2017) ~~showed~~ have shown that European ozone levels are strongly influenced by long range transport (e.g. Jonson et al., 2018; Pay et al., 2019).

~~So far, all previous~~ Despite the large importance of long range transport, all discussed studies applied the source apportionment method only in ~~a~~ the regional model. In this case the source apportionment method can attribute ozone ~~which stem from lateral or top model boundaries~~ and ozone precursors which is advected towards Europe not to specific emission ~~sources~~source. Instead these ~~contribution are quantifies~~ contributions are quantified as boundary contributions, which are not attributed to emission sources ~~(?)~~(Mertens et al., 2020). Accordingly, all of the previous studies have quantified only the contribution of European land transport emissions on the European ozone levels. ~~This study is therefore dedicated to this gap of knowledge, providing~~ Therefore, this study provides a detailed assessment on the contribution of land transport emissions on ozone and ozone precursors (NO_x, CO) ~~including~~ considering the combined effect of European and global emissions.

To include also the effects of long range transport in regional studies, a global-regional model chain is necessary, which includes a source apportionment method in the global and the regional model. Such a model is the MECO(n) model system (e.g. Kerkweg and Jöckel, 2012a, b; Hofmann et al., 2012; Mertens et al., 2016), which couples the global chemistry climate model EMAC (e.g Jöckel et al., 2010, 2016) at runtime to the regional chemistry model COSMO-CLM/MESSy (Kerkweg and Jöckel, 2012b). ~~Both models are~~ Two regional model refinements are applied, covering Europe and Germany with 50 km and 12 km resolution, respectively. The global model resolution is 300 km. The global and the regional model are equipped with the MESSy interface (Jöckel et al., 2005, 2010) ~~. Due to the MESSy interface the~~ and we apply the same tagging method (Grewe et al., 2017) for source apportionment ~~is used~~ in the global and the regional model. Compared to previous studies, this model system allows ~~for~~ a contribution analysis from the global to the regional scale taking into account the effects of long range transport ~~(?). This is important as long range transport strongly influences European ozone levels.~~ (Mertens et al., 2020).

Typically, the uncertainties of such source apportionment studies are large. ~~Typical reasons for these uncertainties~~ Reasons are:

- uncertainties ~~of the emissions inventories; uncertainties~~ in the models (e.g. chemical/physical ~~parametrization) and differences~~ parametrizations) and trough the choice of source apportionment methods;
- uncertainties of the emissions inventories;
- seasonal variability of the contributions caused by meteorological conditions and seasonal cycles of emissions (e.g. stronger biogenic emissions and more active photochemistry during summer than winter);

– year to year variability of the contributions caused by meteorological conditions or large emissions of specific sources in specific years (for example ~~biomass burning); inter annual variability of the contributions caused by meteorological conditions and seasonal cycles of emissions.~~ yearly differences of biomass burning emissions);

To account for the ~~first three uncertainties we performed two three year long~~ uncertainties due to different emission inventories we performed simulations with two different anthropogenic emission inventories~~for Europe. In~~ . To further account for the seasonal variability we investigate the contributions for winter and summer seasons. In addition, we consider always three simulation years to gain insights in the variability of the contribution in different years. The investigation of uncertainties caused by models and/or source apportionment methods is beyond the scope of this study.

In our analysis we focus on mean and extreme (expressed as 95th percentile) contributions for the multi-year seasonal average values ~~during~~ of winter (December, January, February, hereafter DJF) and summer conditions (June, July, August, hereafter JJA). We focus on results for the European domain~~with 50 kmresolution~~. However, as the model resolution can influence the results, we further investigate ~~results for a~~ also results for the smaller domain covering ~~only Germanywith 12 resolution.~~ Germany.

The manuscript is structured as follows. First, Section 2 contains a brief description of the model system, including an introduction to the applied ~~source apportionment methodas well as more details about the~~ tagging method, a description of the performed model simulations and the applied emission inventories, and a brief comparison of the simulated ozone concentrations with observations. Sections 3 and 4 discuss the contributions of land transport emissions to reactive nitrogen, carbon monoxide and ozone in Europe. Section 5 focuses on the contribution of reactive nitrogen for Germany only based on the finer resolved simulation results. Finally, the ozone budget in Europe and the contribution of land transport emissions to the ozone budget are investigated in Section 6.

2 Description of the model system

In this study the MECO(n) model system is applied (Kerkweg and Jöckel, 2012b; Hofmann et al., 2012; Mertens et al., 2016; Kerkweg et al., 2018). This system couples on-line the global chemistry-climate model EMAC (Jöckel et al., 2006, 2010) with the regional scale chemistry-climate model COSMO-CLM/MESSy (Kerkweg and Jöckel, 2012a). COSMO-CLM (COSMO model in Climate Mode) is the community model of the German regional climate research community jointly further developed by the CLM-Community (Rockel et al., 2008). New boundary conditions (for dynamics, chemistry and contributions) are provided at every time step of the driving model (e.g. EMAC or COSMO-CLM/MESSy) to the finer resolved model instances (COSMO-CLM/MESSy). Accordingly, the MECO(n) model allows for a consistent zooming from the global scale into specific regions of interest.

The simulations analysed in the present study are the same simulations as described in detail by ~~?~~Mertens et al. (2020). Therefore, we present only the most important details of the model set-up. Table 1 lists the ~~MESSy submodels applied in the present study~~used MESSy submodels. The global model EMAC is applied at a resolution of T42L31ECMWF, corresponding to a quadratic Gaussian grid of approx. 2.8° x 2.8° and 31 vertical hybrid pressure levels from the surface up to 10 hPa. The

timestep length is set to 720 seconds. ~~Th archive~~ To achieve a higher resolution we apply two COSMO-CLM/MESSy nesting steps. The first refinement covers Europe with a horizontal resolution of 0.44° and 240 seconds time step length, while the second refinement focuses on Germany with 0.11° horizontal resolution and 120 seconds time step length. Both refinements feature 40 vertical levels from the surface up to 22 km. In the following, the abbreviation CM50 (COSMO(50 km)/MESSy) corresponds to the first refinement (with roughly 50 km resolution) and CM12 (COSMO(12km)/MESSy) corresponds to the second refinement (roughly 12 km resolution). ~~For the calculation of atmospheric chemistry the~~ The MESSy submodel MECCA ~~is applied (Sander et al., 2011)~~ (Sander et al., 2011) is applied in EMAC and COSMO-CLM/MESSy for the calculation of chemical kinetics. The chemical mechanism includes the chemistry of ozone, methane and odd nitrogen. Alkynes and aromatics are not taken into account, but alkenes, and alkanes are considered up to $\mathrm{C_4}$. The Mainz Isoprene Mechanism (MIM1, Pöschl et al., 2000) is applied for the chemistry of isoprene and some non-methane hydrocarbons (NMHCs). The complete namelist set-ups ~~as well as~~ and the mechanisms of MECCA and SCAV (scavenging of traces gases by clouds and precipitation, Tost et al., 2006a, 2010) are part of the supplement.

Anthropogenic~~emissions as well as~~ , biomass burning, agricultural waste burning (AWB) and biogenic emissions are prescribed from external data sources (see Sect. 2.2). Emissions of soil $\mathrm{NO_x}$ are calculated on-line (i.e. during model runtime) following the parametrisation of Yienger and Levy (1995). The same applies for emissions of biogenic VOCs which are calculated following Guenther et al. (1995), and emissions for lightning-$\mathrm{NO_x}$ for which the parametrisation of Price and Rind (1994) is applied.

The simulation period ranges from 07/2007 to 01/2011. The first ~~month~~ months of 2007 are the spin-up phase and the years 2008–2010 are analysed. For reasons of computational costs CM12 has been initialised in May 2008 from CM50 and integrated for the period 05/2008-08/2008 only. Therefore, results of CM12 are analysed only for JJA 2008. To facilitate ~~a~~ an one to one comparison with observations EMAC is 'nudged' by Newtonian relaxation of temperature, divergence, vorticity and the logarithm of surface pressure (Jöckel et al., 2006) towards ERA-Interim (Dee et al., 2011) reanalysis data of the years 2007 to 2010. ~~Sea~~ The sea surface temperature and sea ice coverage are prescribed ~~as boundary conditions for the simulation set-up from this data source. The COSMO/MESSy refinements~~ from ERA-Interim as well. CM50 and CM12 are not nudged, but forced at the lateral and top boundaries against the driving model (e.g. EMAC for CM50 and CM50 for CM12).

One feature of chemistry-climate models is the coupling between chemistry, radiation and atmospheric dynamics, meaning that even small changes in the chemical state of the atmosphere lead to changes in the dynamics (which in turn feed back to the chemistry). This feedback can prevent a quantification of the influence of small emission changes on the atmospheric composition. To overcome this issue Deckert et al. (2011) proposed a so called **q**uasi **c**hemistry **t**ransport **m**odel mode (QCTM mode) for EMAC, which can also be applied in MECO(n) (Mertens et al., 2016). To achieve the decoupling between dynamics and chemistry, climatologies are used within EMAC: (a) for all radiatively active substances ($\mathrm{CO_2}$, $\mathrm{CH_4}$, $\mathrm{N_2O}$, CFC-11 and CFC-12) for the radiation calculations, (b) nitric acid for the stratospheric heterogeneous chemistry (in the submodel MSBM, Multiphase Stratospheric Box Model, ~~(Jöckel et al., 2010)~~Jöckel et al. (2010)) and (c) for OH, $\mathrm{O^1D}$ and Cl for methane oxidation in the stratosphere (submodel CH4). In COSMO-CLM/MESSy only the climatology of nitric acid for the submodel MSBM

is required. The ~~required~~ applied climatologies are monthly mean values from the *RC1SD-base-10a* simulation described by Jöckel et al. (2016).

~~A set-up very similar with the set-up of this study has been evaluated with different observational data by Mertens et al. (2016). Generally, the evaluation exhibited a good agreement with observation. The biases are similar to comparable model systems and exhibit an positive ozone bias and negative biases for and . One important reason for these biases is the to efficient vertical mixing within the COSMO-CLM model.~~

2.1 Tagging method for source attribution

The source apportionment of ozone and ozone precursors is performed using the tagging method described in detail by Grewe et al. (2017), which is based on an accounting system following the relevant reaction pathways and applies the generalised tagging method introduced by Grewe (2013).

For the source apportionment the source terms, e.g. emissions, of the considered chemical species are fully decomposed in N unique categories. The definition of the ten categories considered in the current study are listed in Table 2. The tagging method is a diagnostic method, i.e. the atmospheric chemistry calculations are not influenced by the tagging method. ~~Due to constraints with respect to~~ To minimise the computational resources (e.g. ~~computational~~ computing time and memory ~~) the~~ consumption), the tagging is not performed for the detailed chemistry from MECCA~~is mapped on a family concept for which the tagging is performed~~, but for a simplified family concept. The species of the family concept are ~~given~~ listed in Table 3.

~~All chemical~~ The production and loss rates ~~required for the simplified chemistry, as well as the concentration of all chemical species are~~ and mixing ratios of the chemical species which are required for the tagging method are obtained from the submodel MECCA. ~~Further, loss~~ Loss processes like deposition are treated as bulk process, meaning that the changes of the relevant ~~concentration due to dry-~~ mixing ratios due to dry and wet deposition are ~~memoried~~ memorized and later applied to all tagged species according to their relative contributions.

Due to the full decomposition into N categories, the sum of contributions of all categories for one species equals the total ~~concentration~~ mixing ratio of this species (i.e. the budget is closed):

$$\sum_{\mathrm{tag}=1}^{N} \mathrm{O_3^{tag}} \quad = \quad \mathrm{O_3}. \tag{1}$$

To demonstrate the basic concept of the generalised tagging method we consider the production of $\mathrm{O_3}$ by the reaction of NO with an organic peroxy radical ($\mathrm{RO_2}$) to $\mathrm{NO_2}$ and the organic oxy radical (RO):

$$\mathrm{NO + RO_2 \longrightarrow NO_2 + RO}. \tag{R1}$$

As demonstrated by Grewe et al. (2017) (see Eq. 13 and 14 therein) the tagging method leads to the following fractional apportionment:

$$P_{R1}^{tag} = \frac{1}{2} P_{R1} \left(\frac{NO_y^{tag}}{NO_y} + \frac{NMHC^{tag}}{NMHC} \right). \quad (2)$$

Here, all species marked with ^{tag} represent the quantities tagged for one specific category (e.g. land transport emissions); P_{R1} is the production rate of O_3 by reaction R1, NO_y and NMHC represent the mixing ratios of the tagged family of NO_y and NMHC, respectively. The denominator represents the sum of the mixing ratios over all categories of the respective tagged family/species. Accordingly, the tagging scheme takes into account the specific reaction rates from the full chemistry scheme. Further, the fractional apportionment is inherent to the applied tagging method as due to the ~~combinatorical~~ combinatorial approach, every regarded chemical reaction is decomposed into all possible combinations of reacting tagged species.

~~As discussed by ? the tagging method in~~ Some of the categories listed in Table 2 are not directly associated with emission sectors. These categories are stratosphere, CH_4 and N_2O. All ozone which is formed by the photolysis of oxygen i.e.

$$O_2 + hv \longrightarrow O(^3P) + O(^3P), \quad (R2)$$

is labelled as stratospheric ozone.

The degradation of N_2O is a source for NO_y (and a loss of ozone) by the reaction:

$$N_2O + O^1D \longrightarrow 2NO. \quad (R3)$$

The degradation of CH_4 is considered as source of $NMHC^{CH_4}$. This refers to the reaction:

$$CH_4 + OH \longrightarrow CH_3O_2 + H_2O. \quad (R4)$$

As have been discussed recently in detail by Butler et al. (2018) all tagging methods are based on specific assumptions and have specific limitations. The scheme of Grewe et al. (2017), which we apply in the current study, is based on specific assumptions, which differ from other tagging schemes used in regional and global models. One important difference is the question whether ozone formation is attributed to NO_x or VOC precursors. The schemes which are available in the regional models CMAQ (called CMAQ-ISM, Kwok et al., 2015) and CAMx (called CAMx OSAT, Dunker et al., 2002) use threshold conditions to check, whether ozone formation is NO_x or VOC limited. Depending on this the production is attributed to NO_x or VOC precursors only. The scheme of Emmons et al. (2012), applied on the global scale, tags only NO_x and therefore ozone production is only attributed to NO_x precursors. Based on the work of Emmons et al. (2012), Butler et al. (2018) presents a scheme, which attributes ozone formation either to NO_x or VOCs (implying that usually 2 simulations, one with NO_x and one with VOC tagging, are performed). This scheme has also been applied by Lupascu and Butler (2019) in a regional model simulation over Europe, using only the NO_x tagging scheme. Compared to these schemes the scheme of Grewe et al. (2017) attributes ozone production always to all associated precursors (i.e. NOx, HO_2 and VOCs) without any threshold conditions.

In VOC limited regions, this approach leads to the effect that a NO_x emission reduction of an emission sector reduces the contribution of that sector, and increases the contribution of the other sectors. In contrast, a reduction of VOC emissions decreases the contribution of the respective sector only. The latter is similar to the approaches integrated in CMAQ or CAMx, which attribute ozone production in the case of a VOC limit to VOC precursors only. Compared to a NO_x tagging, our approach leads to lower contributions of NO_x sources, since they compete, not only with other NO_x sources, but also with VOC sources.

Because of the family concept, which is necessary to keep the memory consumption and the computational costs low, the tagging method applied in our study can lead to some unphysical artefacts. As an example, Grewe et al. (2017) discuss the production of PAN by NMHCs from CH_4 degradation. Further, due to the combinatorial approach for instance also NMHCs from stratospheric origin can occur in small amounts, which is also an unphysical artefact. The main reason for this is the definition of the PAN family, which transfers tags from NO_y to NMHCs. Other tagging schemes have specific issues as well. As an example, the scheme of Emmons et al. (2012) does not neglect the O_3-NO_x null cycle, which leads to an overestimation of local sources compared to long range transport sources (see also Kwok et al., 2015). Overall, the impacts of the underlying assumptions on the results are difficult to quantify. Therefore, it is important to study effects of different emission sources with different methods (at best in the same model framework), in order to understand better the strengths and weaknesses of the different approaches and their impact on the source apportionment results.

Besides these general assumptions of the different methods one specific problem occurs when applying ozone source apportionment in regional models; the boundary conditions. Usually, regional studies (e.g. Li et al., 2012; Kwok et al., 2015; Valverde et al. tag ozone from lateral and top boundaries as 'boundary ozone' because no boundary conditions including tagged ozone are available. Recently, Lupaşcu and Butler (2019) have used results from a previous global model simulation including an NO_x tagging as boundary conditions for a regional ozone source apportionment study with WRF-Chem over Europe. As pointed out by Mertens et al. (2020) our approach has no need for results from previous model runs, as in MECO(n) ~~is applied~~ the tagging is performed in all model instances (i.e. in the global model as well as all regional model instances). Thus, consistent ~~lateral and model top boundary conditions can be~~ boundary conditions are provided for the regional model instances ~~. Compared to other source apportionment methods in regional models (e.g. Li et al., 2012; Kwok et al., 2015; Valverde et al., 2016; Pay et al., 2019) our method also attributes emissions outside the domain of the regional model to specific emission sources. Source~~ and source categories containing only contributions from lateral or model top boundaries are not required. In the configuration of the tagging method applied for the present study we use only one global tag for every source category. While this allows to investigate the contributions of all global emissions of a specific emission source to ozone mixing ratios, we are not able to separate contributions from local and long range transport (i.e we cannot separate contributions from, for example, European and Asian land transport emissions to European ozone levels, but we can quantify the contribution of global land transport emissions to European ozone levels).

In the following, we denote absolute ~~contributions~~ contribution of land transport emissions to ozone as O_3^{tra}. Analogously, contributions to the family of NO_y and CO are denoted as NO_y^{tra} and CO^{tra}, respectively (cf. abbreviations in Table 2). These absolute ~~contribution corresponds to this shade~~ contributions correspond to the share of the species total mixing ratio which can

be attributed to emissions of land transport. ~~Similarly,~~ Please note, that the given absolute contributions for ozone are always computed by multiplying the relative contributions to odd oxygen with the ozone mixing ratios. These values are slightly lower as the absolute contributions of odd oxygen. Besides the absolute contributions we investigate relative contributions ~~given~~ which give the percentage of the contribution to the total mixing ratio of the specie.

2.2 ~~Emissions~~ Emission scenarios and numerical experiments

~~Two different emission inventories are used to investigate the uncertainties of these emission inventories~~ To investigate the influence of the uncertainties of anthropogenic emission inventories on the source apportionment results, we perform simulations for two anthropogenic emission inventories. The first emission inventory is the ~~global~~ MACCity inventory (Granier et al., 2011), a global inventory with 0.5 x 0.5° horizontal resolution which corresponds to the RCP 8.5 emission scenario for the analysed time frame (called MAC in the following). The second emission inventory is ~~called VEU . It is a European emission inventory which~~ named VEU and considers only emissions for the European area (0.0625 x 0.0625° horizontal resolution). It has been composed in the DLR project 'Verkehrsentwicklung und Umwelt'. ~~This emission inventory considers only the emission sectors land transport, shipping and anthropogenic non-traffic.~~ For this emission inventory the German land transport emissions were estimated bottom up by means of macroscopic traffic simulations. Based on the travelled kilometres from the traffic simulations the land transport emissions were estimated using emission factors. For the other European countries, as well as for all other emission sectors, a top down approach has been applied. More details about the emission inventory are ~~given~~ provided by Hendricks et al. (2017). Further details about the preprocessing of the emissions is given in Appendix A of Mertens (2017).

Two different simulations ~~were~~ are performed:

- *REF*: The MAC emission inventory is applied in EMAC and all regional refinements (e.g. CM50 and CM12);
- *EVEU*: The MAC emission inventory is applied in EMAC and the VEU emission inventory in the regional refinements.

The VEU emission inventory ~~contains~~ considers only emissions for the sectors land transport, anthropogenic non-traffic (including landing and take-off (LTO) of airplanes) and shipping. Table 4 lists the total emissions of NO_x, CO, VOC, and the ratio of NO_x to VOC for these emission sectors. In general, the total emissions of the land transport sector are quite similar, while the emissions of the sectors anthropogenic non-traffic and shipping ~~sectors~~ are lower in the VEU compared to the MAC emission inventory. Especially the NO_x and VOC emissions are lower by around 30 % and 50 %, respectively. This leads to different NO_x to VOC ratios for the total anthropogenic emissions between both emission inventories. The definition of the emission sectors in VEU is different from the definition in MAC. In the VEU emission inventory LTO emissions are part of the anthropogenic non-traffic sector, but inflight emissions from aircrafts are not considered in VEU. Therefore, the MAC aviation emissions are also applied in the *EVEU* simulation. To avoid a double accounting of the LTO emissions, the aviation emissions in MAC are set to zero in the lowermost level in *EVEU*, leading to a reduction of the aviation emissions of the MAC emission inventory by 0.05 $Tg\ a^{-1}$ (see Table 4). For the emission sectors agricultural waste burning (AWB), biomass burning, lightning and biogenic we apply the same emissions in both simulations (see Table. 5). Total emissions for the global model EMAC, ~~as well as~~ and for CM12 are given in the Supplement (see Section ~~S2~~S4).

Figure 1 displays the geographical distribution of the land transport emissions of NO_x, CO, and VOC applied in the *REF* and *EVEU* simulations and the difference of the emissions between both simulations. Shown are only the emissions of EMAC and CM50, focusing on Europe. The NO_x land transport emissions for CM12 are depicted in the Supplement (Fig. S7). Further, more detailed figures showing the geographical distribution in CM50 are part of the Supplement (Fig. S8). The emissions of CM50 are superimposed onto the emissions applied in EMAC, where the MACCity emissions are applied globally. ~~For the emissions in Europe we not that despite the~~ Despite comparable total emissions ~~the geographical distribution differs~~between the MACCity and the VEU emission inventory over Europe, the geographical distributions differ. Generally, the VEU emission inventory features larger emissions near the hot-spots and lower emissions away from the hot-spots compared to MAC. Further, MAC features larger NO_x emissions especially the Northern part of the British Islands and in Finland. Emissions of CO are especially larger around Estonia in MAC compared to VEU. ~~Especially~~ Particularly, over Germany, the Po Valley and ~~party~~ parts of Eastern Europe VEU features more emissions of NO_x, CO and VOC (see also totals for CM12 in Table S4). Besides the difference between the emissions applied in CM50 (and CM12) it is important to note, that for the *REF* and the *EVEU* simulation the same emissions are applied in EMAC. Therefore, the difference (Fig. 1c) is zero in EMAC.

2.3 Model evaluation

A model set-up very similar to the one used for the present study has been evaluated with observational data by Mertens et al. (2016). Generally, the comparison showed a good agreement with observations. The biases are similar to comparable model systems and exhibit a positive ozone bias and negative biases for NO_2 and CO. One important reason for these biases is the too efficient vertical mixing within the COSMO-CLM model. An evaluation of the ozone mixing ratios simulated by *REF* and *EVEU* have already been presented by Mertens et al. (2020), however, mainly focusing on JJA mean values. To investigate the models ability to represent extreme values, we present a brief evaluation of the simulated ozone concentrations in comparison to the Airbase v8 observational dataset (available at, https://www.eea.europa.eu/data-and-maps/data/airbase-the-european-air-quality-database-8, last access 14.2.2020). As the model resolution of 50 km is too coarse to resolve hot-spots of individual cities we restrict the comparison to those stations which are classified as area types 'suburban' and characterised as 'background'. We focus on JJA 2008 to 2010 and compare the results to overall 350 measurement stations. The measurements are subsampled at the same temporal resolution (3 hourly) as the model data.

The comparison with the observational data shows a positive ozone bias of the model, which has been discussed in previous studies (Mertens et al., 2016, 2020). The average root-mean-square-error (RMSE) over all 350 stations is 29.2 $\mu g\ m^{-3}$ for *REF*, and 24.3 $\mu g\ m^{-3}$ for *EVEU*, respectively. The corresponding mean biases (MBs) are 26.6 % and 20.5 %, respectively (see Table 6). In addition, we calculated also the RMSE and MB for the *REF* simulation considering only measurements and model data at 12 and 15 UTC. For this subsample, both, RMSE and MB decrease considerably. Accordingly, the largest ozone values during daylight are captured very well by the model. As a more detailed comparison between measurements and model result shows, the overestimation of ozone is particularly strong during night. This can partly be attributed to a too unstable boundary layer during night, which is a common difficulty in many models (Travis and Jacob, 2019). In addition, the too strong vertical mixing in the model leads to positive ozone biases at noon and during the night (see also Mertens et al., 2020, 2016).

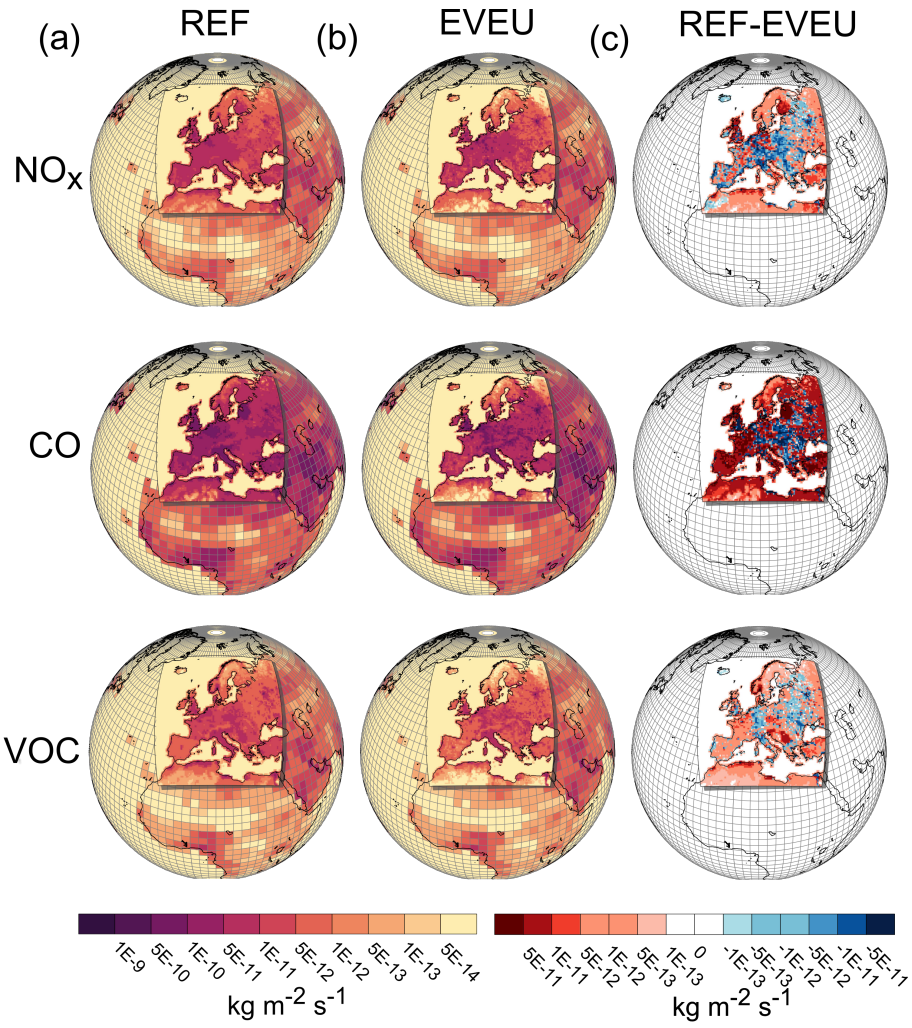

Figure 1. Annually averaged emission fluxes (2008 to 2010) from the land transport sector (in $\mathrm{kg\ m^{-2}\ s^{-1}}$). Shown are the emissions as applied in EMAC (based on the MACCity inventory) and in CM50. The emissions of CM50 are superimposed on the emissions of EMAC. In the region covered by CM50 EMAC also uses the MACCity emissions (not visible). (a) the emissions ~~of the~~ applied in *REF* ~~simulation~~, (b) the emissions ~~of the~~ applied in *EVEU* ~~simulation~~ and (c) the difference of the emissions ~~from~~ between *REF* and *EVEU* ('REF MINUS EVEU'). Shown are the emission fluxes of ~~of~~ NO_x (in $\mathrm{kg\ NO\ m^{-2}\ s^{-1}}$), CO (in $\mathrm{kg\ CO\ m^{-2}\ s^{-1}}$); and VOC (in $\mathrm{kg\ C\ m^{-2}\ s^{-1}}$).

Currently, further investigations are undertaken, about how this bias could be reduced in the future. Besides the too efficient vertical mixing, also too less ozone deposition during night, too low NO or VOC emissions, and successively underestimated ozone depletion during nights could also partly contribute to this bias.

For analysing extreme ozone values, we also compare the 95th percentiles of ozone with measurements (see Fig. 2). Overall, the model is able to capture most of the regional variability of the extreme values over Europe. Near the densely populated regions in Benelux, Germany and Italy, however, the model is not able to reproduce the extremes. In these areas the model

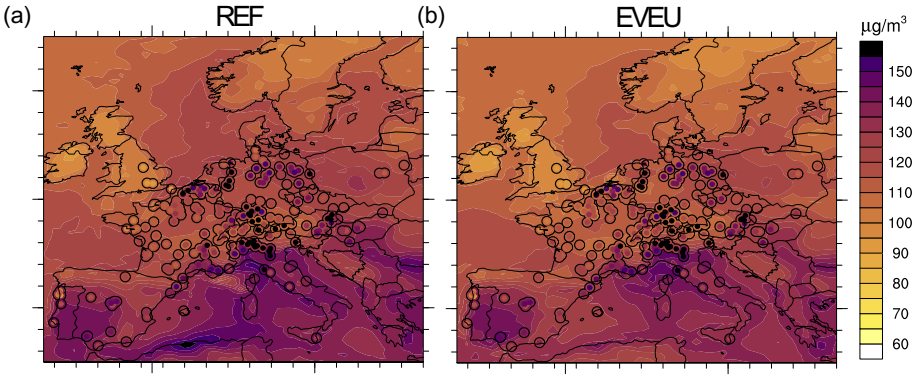

Figure 2. 95th percentile of ozone (in $\mu g/m^3$) for the period JJA 2008 to 2010. The background colours show the ozone concentrations as simulated by CM50, the circles represent the location of stations of the Airbase v8 observation data. The inner point represents the measured concentrations, the outer point the concentrations in the respective grid box, where the station is located. All values are based on data every 3 hours.

resolutions (i.e. also for the 12 km domain, which is not shown here) are too coarse to allow for a representation of extreme ozone values in urban areas. As has been shown by prior studies (e.g. Tie et al., 2010) resolutions below 10 km are required to capture high ozone values near cities. Terrenoire et al. (2015) have noted that even with 8 km resolution the performance of the applied CHIMERE model is better at rural than at urban sites. This underestimation can also be quantified using the RMSEs and MBs for the 95th percentile which are listed in Table 6.

These results have important implications for the analyses, which are presented in this manuscript. First of all, the too strong vertical mixing in COSMO-CLM/MESSy leads to a positive bias of the contribution of stratospheric ozone at ground-level. Further, also contributions of lightning and aviation at ground-level are likely larger due to this overestimated vertical mixing. This leads to a around 1 percentage point lower contribution of anthropogenic emissions in COSMO-CLM/MESSy compared to EMAC (see Mertens et al., 2020). Due to the coarse model resolution our results are representative for the regional scale, but not for specific urban areas. In these urban areas local emissions and local ozone production/destruction might be more important such that contributions of local sources can be much larger than the values we present. On the regional scale, however, Mertens et al. (2020) showed that the results are quite robust w.r.t. the model resolution (down to 11 km).

Because of the stronger ozone bias during night, we further compared the contributions at 12 and 15 UTC with the contributions considering all times of the day. The relative contributions show only small differences, i.e. a slightly larger contribution of anthropogenic emission sources during day (not shown). Therefore, we present always results for all times of the day.

3 Contributions of land transport emissions to ~~ground level~~ ground-level mixing ratios of NO_y and CO in Europe

CO and NO_y are direct pollutants of the land transport sector, with different chemical lifetimes. Please note, ~~that the~~ because of the family concept used by the tagging method we investigate contributions to NO_y and not to NO_x ~~are investigated as the source apportionment method tags the whole family of (without PAN) and not alone~~. Our focus in this section ~~are~~ is on the results on the European scale, results of NO_y for Germany will be discussed in Sect. 5. Figure 3 shows NO_y^{tra} for DJF and JJA, respectively. The largest mixing ~~rations~~ ratios of NO_y^{tra} are simulated near Southern England, the Paris metropolitan region, Western Germany and the Benelux states as well as the Po Valley and the Moscow metropolitan region. In these regions contributions of up to 10 $nmol\ mol^{-1}$ are simulated. In general, larger absolute contributions occur during DJF compared to JJA, but the ~~annual~~ seasonal cycle of the land transport emissions is small in both emission inventories (see supplement Figure S4). Accordingly, the differences of NO_y^{tra} between DJF and JJA are likely not caused by seasonal differences of the emissions, but by larger mixing layer heights ~~as well as~~ and a more effective photochemistry during JJA compared to DJF.

The seasonal change of ~~the~~ NO_y^{tra} is smaller than differences between *REF* and *EVEU*. Near areas with large land transport emissions *EVEU* simulates 3 to 4 $nmol\ mol^{-1}$ larger contributions than *REF*. In most of the hot-spot regions (e.g. Paris and the Po Valley) the differences are even larger and the contributions calculated by *EVEU* are 5 $nmol\ mol^{-1}$ larger ~~as~~ than in *REF*. In some regions the results of both simulations are in total contrast. In *REF* for example, absolute contributions of up to 4 $nmol\ mol^{-1}$ are simulated in Finland, while ~~mixing ratios of~~ *EVEU* simulates absolute contributions below 1 $nmol\ mol^{-1}$ ~~are simulated in *EVEU*~~.

The ~~absolute contributions correspond to relative contributions~~ relative contribution of land transport emissions to ~~ground level~~ ground-level NO_y is in the range of 40 % to 70 % in most parts of Europe (see Fig ~~4).~~ . 4). These relative contributions are similar as the share of land transport NO_x emissions to all NO_x emissions (see Fig. S9 in the Supplement), but compared to the share of the emissions the contributions to NO_y are slightly lower near hot-spots, and larger in rural areas.

During DJF, *REF* simulates the lowest relative contributions of 30 to 50 % over most parts of Europe. During summer the contributions increase up to 60 % with the largest values in Southern Germany, the Po Valley, and ~~southern~~ Southern England. *EVEU* simulates a smaller difference of the contributions between DJF and JJA as *REF*. Further, the maxima are generally slightly larger and contributions of up to 70 % are simulated around the Po Valley and the Paris area. Interestingly, the relative contributions are lower during DJF than during JJA while the absolute contributions are larger during DJF than during JJA. Most likely this is caused by the lower amount of anthropogenic non-traffic NO_x emissions during JJA compared to DJF (see Fig. S4 in the Supplement).

The simulated mixing ratios of CO^{tra} (see Fig. 5) show a similar behaviour as NO_y^{tra}, implying that contributions ~~during~~ in DJF are larger ~~as during~~ than in JJA. This seasonal difference is most likely caused by lower mixing layer heights and increased lifetime of CO during DJF compared to JJA, as OH concentrations are lower in winter compared to summer. Generally, the largest contributions are simulated in southern England, around Paris, Western Germany, the Po Valley and around Moscow. In *EVEU* contributions of up to 75 $nmol\ mol^{-1}$ are simulated around London, Paris, Milan and Moscow, while the results of the *REF* simulation show lower contributions in the Western European regions of mostly 50 to 60 $nmol\ mol^{-1}$. Compared

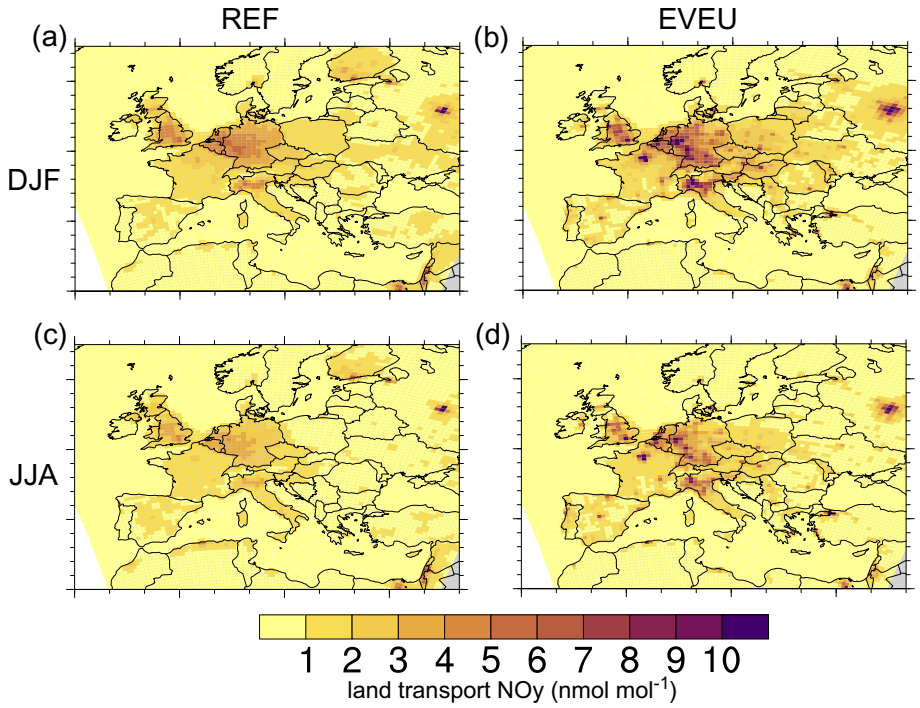

Figure 3. Absolute contribution of land transport emissions to ground-level $\mathrm{NO_y}$ (in $\mathrm{nmol\,mol^{-1}}$) as simulated by CM50. (a) and (b) contributions for the period DJF (2008 to 2010) of the *REF* and *EVEU* simulations, respectively. (c) and (d) contributions for the period JJA (2008 to 2010) of the *REF* and *EVEU* simulations, respectively.

to $\mathrm{NO_y^{tra}}$, however, some hot-spots stand out in the results of the two simulations. *EVEU*, for example, shows larger contributions (40 to 60 $\mathrm{nmol\,mol^{-1}}$) to CO over Hungary or southern Poland. In difference to this, *REF* shows contributions of 30 to 50 $\mathrm{nmol\,mol^{-1}}$ over Estonia. These differences of the contributions are directly caused by differences between the two emission inventories (Fig. 1). Hence, the uncertainties with respect to the CO emissions of land transport in these regions are quite large.

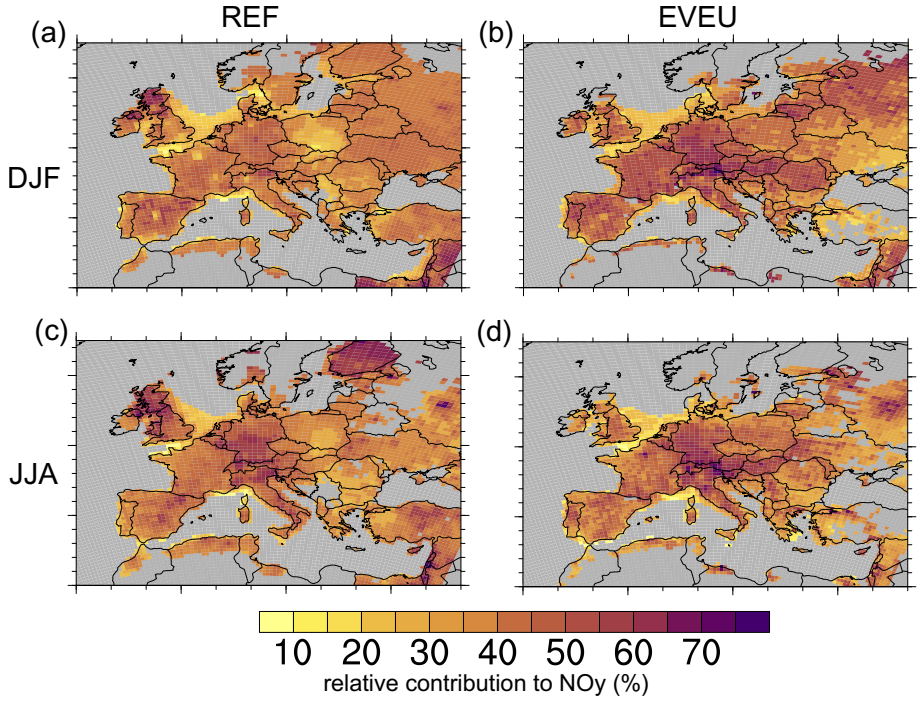

Figure 4. Relative contribution of land transport emissions to ground-level NO_y (in %) as simulated by CM50. (a) and (b) contributions for the period DJF of the *REF* and *EVEU* simulations, respectively. (c) and (d) contributions for the period JJA of the *REF* and *EVEU* simulations, respectively. Grey areas indicate regions where the absolute NO_y mixing ratios are below 0.5 $\mathrm{nmol\ mol^{-1}}$. In these regions no relative contributions are calculated for numerical reasons.

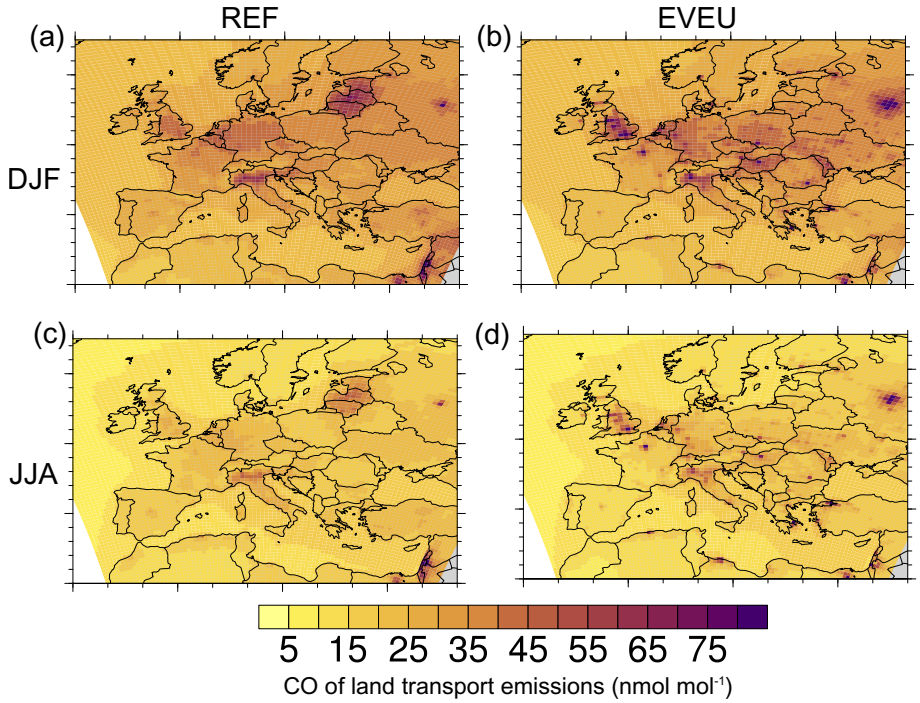

Figure 5. Absolute contribution of land transport emissions to ground-level CO (in $\mathrm{nmol\,mol^{-1}}$) as simulated by CM50. (a) and (b) contributions for the period DJF of the *REF* and *EVEU* simulations, respectively. (c) and (d) contributions for the period JJA of the *REF* and *EVEU* simulations, respectively.

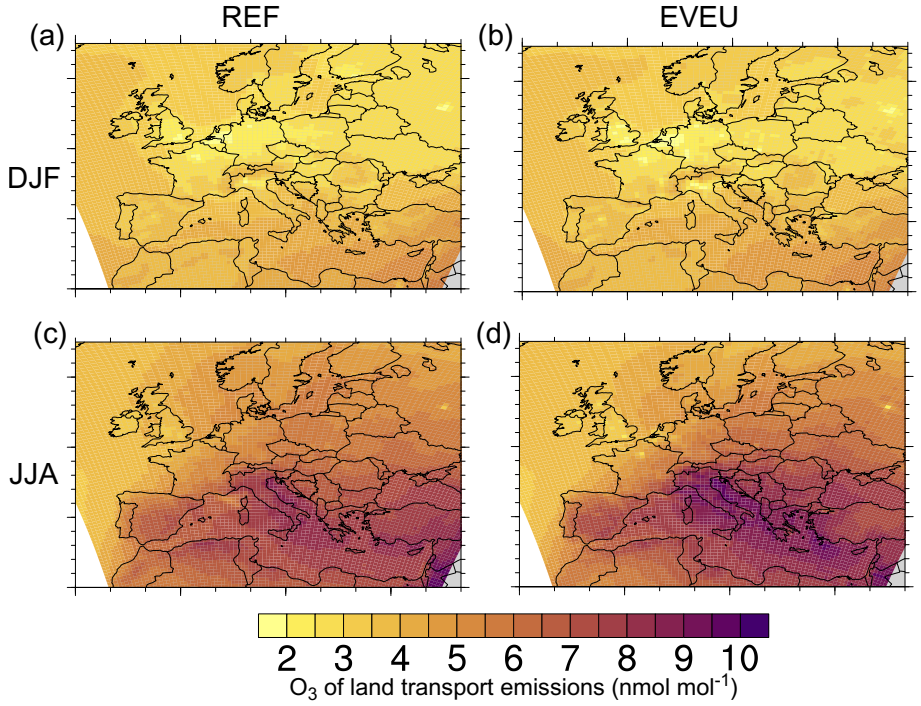

Figure 6. Absolute contribution of land transport emissions to ground-level O_3 (in $\mathrm{nmol\ mol^{-1}}$) as simulated by CM50. (a) and (b) contributions for the period DJF of the *REF* and *EVEU* simulations, respectively. (c) and (d) contributions for the period JJA of the *REF* and *EVEU* simulations, respectively.

4 ~~Contributions~~ Contribution of land transport emissions to ozone in Europe and Germany

In difference to NO_y and CO, ozone is a secondary pollutant~~of the land transport sector. This section quantifies~~. In this section the contribution to ozone is quantified in detail. Besides land transport emissions, however, many other sources contribute to ozone near ~~ground level~~ground-level. Generally, the most important sources which contribute globally to ozone are downward transport from the stratosphere, anthropogenic non-traffic~~emissions~~, shipping, lightning and biogenic emissions ~~(e.g. Lelieveld and Dentener, 2000; Grewe, 2004; Hoor et al., 2009; Dahlmann et al., 2011; Emmons et al., 2012; Grewe et al., 2017)~~(e.g. Table 7 lists the contributions of different emission sources to ozone for Europe averaged for JJA 2008 to 2010 and for the results of *EVEU* and *REF* (see also Fig. S6 for zonally averaged vertical profiles of the contributions)~~Near ground level in Europe the~~. The most important sources for ~~ozone~~ ground-level ozone in Europe are biogenic emissions (≈ 19 %), anthropogenic non-traffic emissions (≈ 16 %), methane degradation (≈ 14 %) and land transport emissions (≈ 12 %). With increasing height the ~~contribution~~ contributions of ground based emission sources ~~decreases, therefore~~ decrease, accordingly the contribution of land transport ~~emission~~ emissions decrease to ≈ 8 % at 600 hPa. At the same time the importance of ozone transported downward from the stratosphere, lightning and aviation increases. At a height of 200 hPa more than 50 % of the ozone is from stratospheric origin. The contribution of land transport emissions drops to around 3 %. Further, the differences between the results of *REF* and *EVEU* decrease with increasing height, indicating the larger importance of long range transport~~which are~~. The latter is equal in both simulations due to identical emissions for the global models and therefore identical boundary conditions for CM50.

4.1 Seasonal average contribution to ground-level ozone

During DJF O_3^{tra} near ground-level simulated by *REF* and *EVEU* (see Fig. 6) ranges between 2 ~~-~~to 4 nmol mol^{-1}. Due to ozone titration the absolute contributions near some hot-spots are lower than these contributions. These absolute contributions correspond to relative contributions of O_3^{tra} ~~to ground level ozone~~ of around 8 % over large parts of Europe (see Fig. 7). Although the European emission inventories differ ~~, the simulated~~ in the simulations, the contributions (absolute ~~as well as~~ and relative) show almost no differences. The emissions of the global model, however, are identical in *REF* and *EVEU* leading to identical contributions at the boundaries of the regional domain. Hence, the contributions during DJF are mainly dominated by long range transport towards Europe which has also been reported by Karamchandani et al. (2017). This is caused by the low ozone production and long lifetime of ozone during winter.

During ~~summer~~ JJA the ozone production increases and local emissions play a larger role. Therefore, O_3^{tra} increases to 5 to 10 nmol mol^{-1}, implying ~~that the relative contribution increases~~ an increase of the contributions to 10 to 16 %. The geographical distribution of the contribution ~~are~~ is similar for both emission inventories, showing increasing absolute and relative contributions from North-West to South-East. The largest relative contributions are simulated around the Po Valley while the largest absolute contributions are shifted downwind ~~of Italy into~~ from Italy to the Adriatic Sea. In these regions the differences between the results of the two simulations are largest, reaching up to 2 nmol mol^{-1} for the absolute and 2 percentage-points for the relative contributions, respectively. The larger differences between the results of *REF* and *EVEU*

during summer compared to winter are mainly caused by the increasing ozone production over Europe during spring and summer. Accordingly, ~~the differences of the emissions between the~~ differences in the emission inventories modify the regional ozone budgets more efficiently.

To quantify the contributions of land transport emissions and other emission sources in different regions in more detail, Fig. 8 shows area-averaged relative contributions for JJA and DJF for the *REF* and *EVEU* simulations (absolute contributions are given in Table S1 to Table S8 in the Supplement). The geographical regions were defined according to the definitions of the PRUDENCE project (Christensen et al., 2007), but slightly modified. The region Alps was split up in two separate regions called 'Northern Alps', defined as rectangular box ($46° : 48°$ N and $9° : 13°$ E), and 'Po Valley' ($44° : 46°$ N and $5 : 15°$ E). Note, however, that the region Northern Alps contains parts of Switzerland and Southern Germany, which are still rather flat and subject to large land transport emissions. In addition, we defined a region called 'inflow' ($40° : 60°$ N and $-13° : -11°$ E). This region is used to quantify contributions in the air advected towards Europe. A figure summarizing the definition of all regions is part of the Supplement (Fig. S12). The relative contribution of land transport emissions in the 'inflow' region is about 9 % and very similar in both seasons and for both European emission inventories. During DJF the contributions in all regions are very similar. During JJA the contribution of land transport emissions increases in most regions compared to the 'inflow' (≈ 9 %). In the Po Valley the contribution reaches up to 16 %. Unfortunately, the difference between the contribution in a specific region compared to the contribution in the region 'inflow' cannot be used to calculate O_3^{tra} from European emissions. Such a calculation requires different tags for global and European land transport emissions. The relative contribution of other anthropogenic emissions in the 'inflow' region (≈ 34 %) is also very similar in both seasons. During DJF the contributions in the different regions remain very similar to the contributions in the 'inflow' region. During summer, in contrast, a West-East gradient of the contribution of anthropogenic emissions is present over Europe with a decrease of the contribution of up to ≈ 27 % in Eastern Europe. This decrease is mainly caused by the seasonality of the different emissions (discussed further below). The biogenic emission category shows different relative contributions in the 'inflow' region during DJF (≈ 11 %) compared to JJA (≈ 14 %), which is mainly caused by the strong increase of biogenic emissions during summer compared to winter. In the different regions the relative contributions increase during JJA compared to DJF, and, compared to the 'inflow' up to ≈ 20 %. The contribution of all other tagging categories during DJF is around ≈ 47 % in most regions, and ranges between 41 % and 36 % during JJA.

As already discussed~~the emission~~ , the emissions of the land transport sector show almost no ~~annual~~ seasonal cycle (Fig S4 in the Supplement), while the absolute and relative contribution of O_3^{tra} shows ~~an annual~~ a seasonal cycle. This ~~annual~~ seasonal cycle is caused by a complex interplay of the ~~annual~~ seasonal cycles of different emission sources~~. The annual~~ , meteorology and photochemical activity. The seasonal cycle of the relative contribution of O_3^{tra} is shown in Fig. 9. The ~~annual~~ seasonal cycle of the absolute contribution is similar to the cycle of the relative contribution, but shows the largest peak during June where the absolute ozone levels are ~~larges (See~~ largest (see Fig. ~~S9~~ S10 in the Supplement). ~~Accordingly, the~~ The contribution peaks between May to July and in October (≈ 13 % averaged over Europe for the column up to 850 hPa) and has a minimum of 9 % during December to March. The decrease of the contribution during the summer ~~month~~ months is mainly caused by the large contribution of biogenic emissions (biogenic VOCs and soil-NO_x) during July and August ~~. Important contributors~~

~~to the lower contributions during winter are the categories stratosphere and industry showing a strong annual~~ and subsequent increasing contributions of O_3^{soi}. The decrease of the contribution during DJF is mainly caused by increasing contributions from the stratosphere and anthropogenic non-traffic emissions. The categories show a strong seasonal cycle with peaks of the contributions during March and May (Fig. S3 in the Supplement). ~~Further, the~~ The indicated standard deviation of the contribution shows that in winter, spring, and autumn the year to year variability (blue shading) is the most important source of uncertainty. Here, differences in regional ~~emission~~ emissions lead only to small differences (orange shading). During summer, however, the differences of the regional emissions strongly contribute to the uncertainties.

The differences between the extreme absolute and relative contributions of O_3^{tra} between *REF* and *EVEU* (expressed as 95th percentile) are larger ~~as~~ than for the mean values. The 95th percentile of the relative contribution of O_3^{tra} to ~~ground level~~ ground-level ozone reaches up to 24 % in the Po Valley using the VEU emission inventory (see Fig. 10). ~~The maxima applying the MAC emission inventory~~ In *REF* the maxima are lower by 4 to 5 percentage points compared to *EVEU*. In contrast to the mean values, the extreme values occur mainly near the regions with the largest land transport emissions, namely over France, Italy and Germany. Over France and Germany extreme values (depending on the applied emission inventory) ~~between~~ in the range of 16 to 18 % occur, while the values in Northern Italy range from 20 to 24 %.

Focussing on Germany, the relative contribution of O_3^{tra} to ~~ground level~~ ground-level ozone is 10 to 15 %. The contribution has a North-West to South-East gradient. One important contributor to this gradient are the strong shipping emissions in the English Channel, North- and Baltic- sea (e.g. Matthias et al., 2010). These emissions lead to larger relative and absolute contributions of shipping emissions in Northern and Western Germany, which decrease towards the South. The absolute contributions are around 2 to 3 $\mathrm{nmol\ mol^{-1}}$ during DJF and 4 to 6 $\mathrm{nmol\ mol^{-1}}$ during JJA (averaged for 2008 to 2010). The largest 95th percentile of the relative contribution of land transport emissions ~~are~~ is simulated in Southern Germany (up to 22 %).

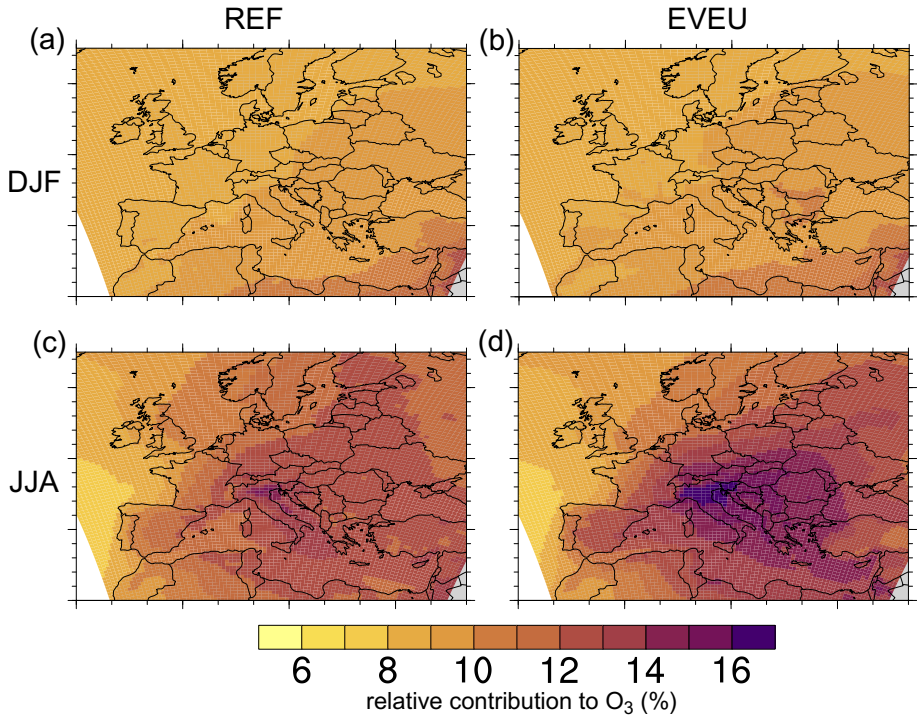

Figure 7. Relative contribution of land transport emissions to ground-level O_3 (in %) as simulated by CM50. (a) and (b) contributions for the period DJF of the *REF* and *EVEU* simulations, respectively. (c) and (d) contributions for the period JJA of the *REF* and *EVEU* simulations, respectively.

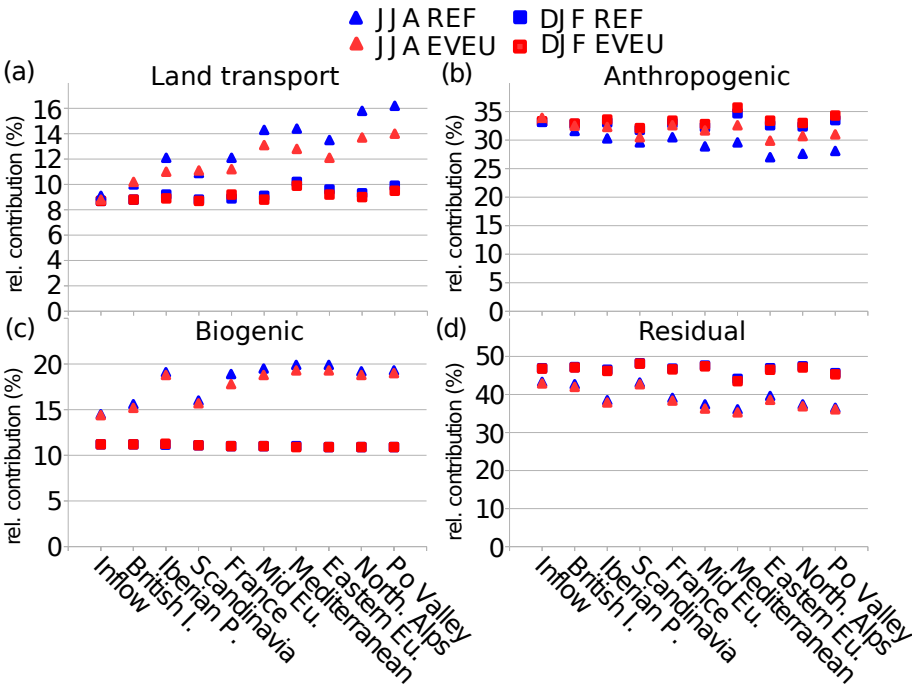

Figure 8. Relative contributions to ground-level ozone (in percent) area averaged in different geographical regions for DJF 2008 to 2010 (triangles) and JJA 2008 to 2010 (squares). Shown are the results of the *REF* (blue) and the *EVEU* simulations (red) for (a) the land transport category, (b) the anthropogenic emissions, (c) the biogenic category, and (d) all other categories. For simplicity the anthropogenic contains the categories anth. non-traffic, aviation and shipping. The residual contains all other categories. The vertical-axis scale differs for (a) to (d).

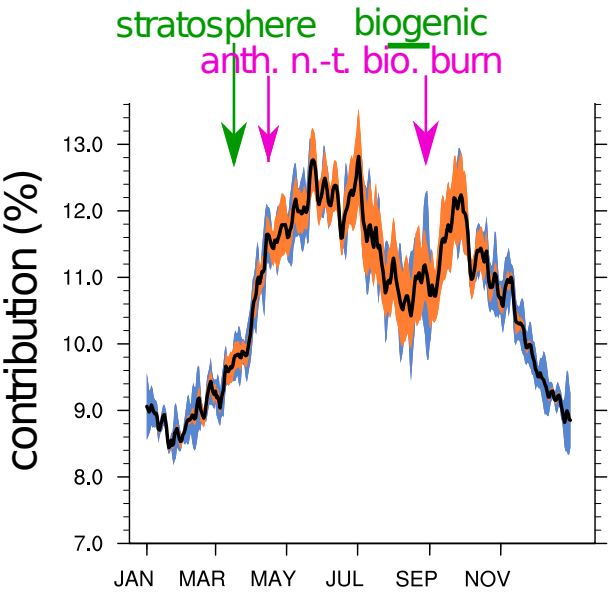

Figure 9. ~~Annual~~ Seasonal cycle of the relative contribution of ~~land transport emissions~~ O_3^{tra} to the ozone column up to 850 hPa (in %). The black line indicates the mean contribution as simulated by CM50, averaged over the years 2008–2010 and the two simulations (*REF*, *EVEU*). The blue shading indicates the standard deviation with respect to time for the years 2008 to 2010 for the *EVEU* simulation. The orange shading indicates the standard deviation with respect to time between the 2008–2010 averaged ~~annual~~ seasonal cycles ~~between~~ of the *REF* and the *EVEU* ~~simulation~~simulations. The coloured arrows indicate the time frames where specific ~~emission~~ categories (stratosphere, anthropogenic non-traffic, biomass burning, and biogenic) have their largest relative contributions. The category biogenic peaks over a wide range, therefore a bar is used instead of the arrow.

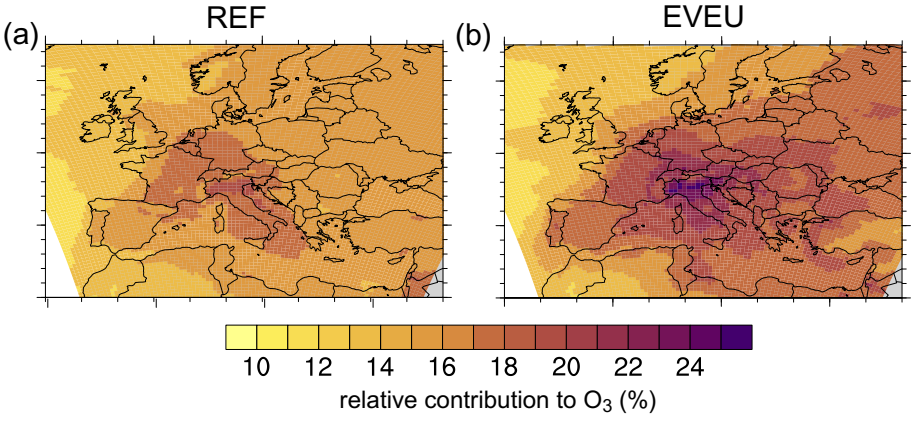

Figure 10. 95th percentile of the relative contribution ~~as simulated by CM50~~ of ~~land transport emissions to ground-level~~ O_3^{tra} (in %) as simulated by CM50 based on 3-hourly model output. (a) and (b) contributions for ~~the period~~ JJA of the *REF* and *EVEU* simulations, respectively.

4.2 Contribution during extreme ozone events

To ~~design mitigation options for periods with~~ better characterise episodes of extreme ozone values, it is important to know which emission sources contribute to and/or drive these extreme ozone values. Therefore, we investigate how land transport emissions contribute to extreme ozone events. As discussed in Sect. 2.3 the contributions we report are representative on the regional scale. For analyses of the local scale the resolution of the model is too coarse.

First, the 99th, 95th and 75th percentile of ~~ozone concentration of~~ the ozone mixing ratios for the period JJA 2008 to 2010 are calculated (based on 3-hourly model output, see Figs. S1 and S2 in the Supplement). Second, the ~~sectors~~ categories contributing to these 99th, 95th and 75th percentile of ozone are analysed. Generally, the contributions to these extreme values have a high spatial variability. To capture ~~these~~ this spatial variability, the contributions are analysed for the whole CM50 domain as well as for specific regional subdomains ~~for which we use the regions defined in the PRUDENCE project (Christensen et al., 2007, , see also Supplement Fig. S10)~~ as introduced above.

The range of contributions in the different ~~subdomains~~ regions is shown in Fig. 11. Generally, the relative ~~contributions~~ contribution of O_3^{tra} (Fig. 11a and b) ~~increase~~ increases for increasing ozone percentiles in most regions. This increase is largest in the regions ~~Alps (including the Po Valley),~~ Po Valley, Northern Alps, Mid Europe, France and the British Islands. The largest contributions of O_3^{tra} occur in the Mediterranean region, ~~the Alps,~~ Northern Alps, Po Valley, Mid Europe and France. Especially in these regions, *EVEU* simulates larger median and maximum relative contributions of O_3^{tra} compared to *REF*. Further, the range of contributions for almost all regions is larger in *EVEU* compared to *REF*. ~~This indicates the large influence~~ The ozone values at the 95th percentile (see Sect. 2.3) and at the other percentiles (see Figs. S1 and S2 in the Supplement), however, are similar for *REF* and *EVEU* (i.e. none of the emission ~~inventory uncertainties on the analysis~~ inventories leads to strongly different representation of extreme ozone events in the model). Accordingly, ~~these uncertainties must kept in mind when designing mitigation option for extreme~~ the discussed differences of the relative contributions are not caused by a different representation of the ozone values themselves, but only due to the different geographical and sectoral distributions of the emissions in *REF* and *EVEU*. This demonstrates the large uncertainty, especially for contributions during high ozone events, of the source apportionment analyses which is caused by the uncertainties of emissions inventories (e.g. geographical distribution of emissions, total emissions per sector). These uncertainties must be taken into account in source attribution studies focusing on high ozone events.

For the 99th percentile of ~~ground level~~ ground-level ozone the median of the relative contributions of O_3^{tra} in the region ~~Alps is around 16~~Po Valley is around 17 % / 22 % (*REF*/*EVEU* simulation), while the 95th percentile is around ~~19~~18 % / ~~24~~25 %. The ~~region with the second~~ contributions in the region Northern Alps are only slightly smaller, as parts of Southern Germany and Switzerland with large land transport emissions are also part of this region. The region with the third largest contributions is Mid Europe (including mainly Germany and the Benelux States). Here, median contributions (at 99th percentile of ozone) of 16 % / 18 % and contributions (at 95th percentile) of 18 % / ~~22~~23 % are simulated. The largest contributions (between 24 and 28 % for the *EVEU* simulation) are mainly simulated in the Po Valley, in South-Western Germany, Western Germany and around Paris. For the lower percentile of ~~ground level ozone the contributions~~ ground-level ozone the contribution of land

transport emissions ~~decrease~~ decreases and reach median contributions of 13 to 16 % and 95th percentiles of 15 to 21 % in the regions Mediterranean, Alps, Mid Europe and France.

The medians of the relative contribution of other anthropogenic emissions (~~including~~ i.e. the emission sectors anthropogenic non-traffic and aviation) range in all regions between 17 ~~%~~ to 25 % (Fig. 11c and d). Hence, the contribution of other anthropogenic emissions is larger ~~as~~ than the contribution of land transport emissions. The increase of the contribution of other anthropogenic emissions with increasing ozone percentiles, however, is lower compared to the ~~contribution of~~ increase of the O_3^{tra}. Accordingly, the relative importance of land transport emissions ~~increase~~ increases with increasing ozone values and hence land transport emissions are an important driver of large ozone values. ~~However, besides the~~ This is in general in line with Valverde et al. (2016) who found that concentration peaks of ozone in Barcelona and Madrid can be explained by ozone attributed to road transport emissions. However, their contributions are in general much larger than the contributions we found (see more details in Sect. 7). Besides the contribution of land transport emissions, however, also the relative contribution of biogenic emissions to ozone increases with increasing ozone levels (Fig. 11e and f). Therefore, also biogenic emissions play an important role during high ozone values.

While the relative ~~contribution~~ contributions to ozone of the shown ~~emission sectors~~ categories increase with increasing ozone levels, the contribution of the shipping emissions and all other ~~emission sectors~~ categories decrease with increasing ozone levels in ~~all most all~~ almost all regions (Fig. S5 in the Supplement). Only in the Mediterranean region *REF* simulates also an small increase of the relative contribution of shipping emissions with increasing ozone levels.

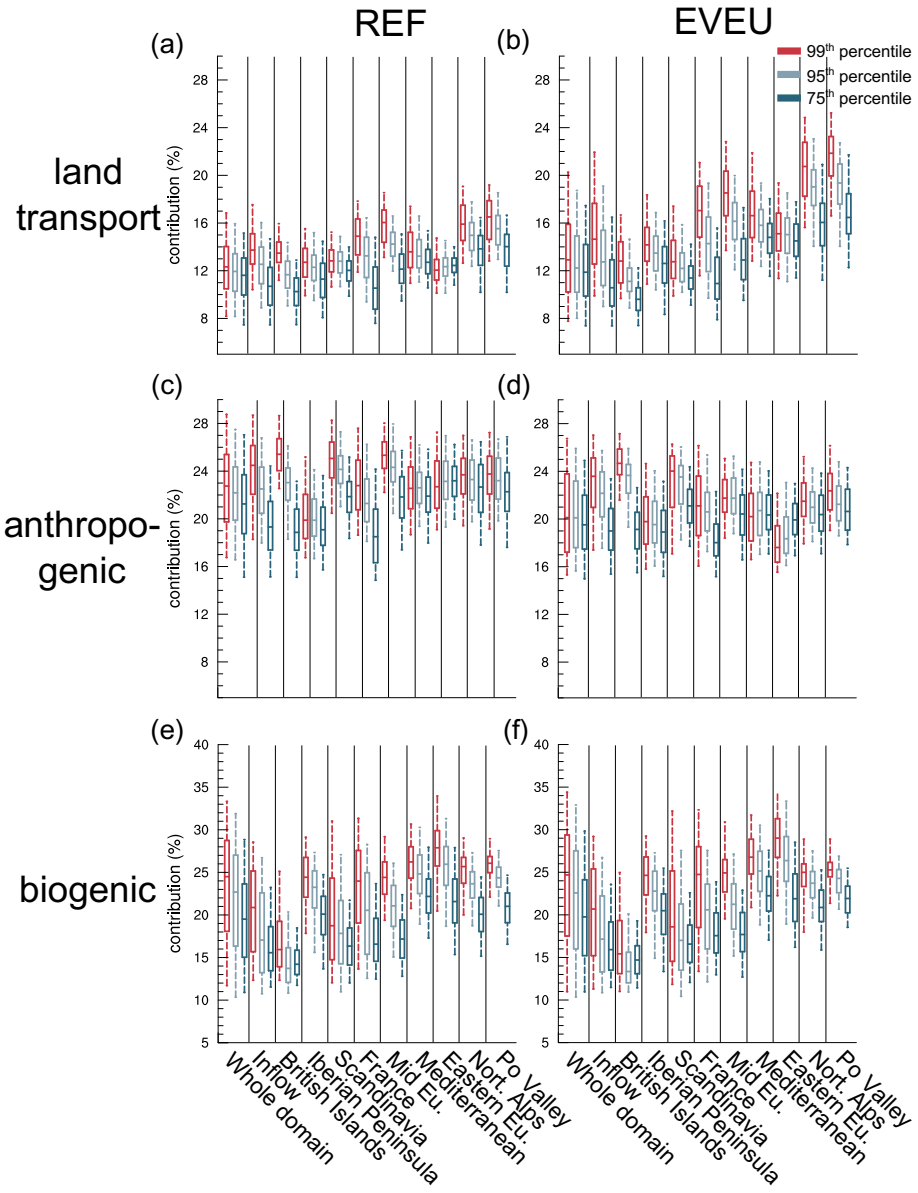

Figure 11. Box-whisker plot showing the ~~contribution~~ contributions of the most important emission sources at the 99th, 95th and 75th percentile of ozone as simulated by CM50. For simplicity only the contributions for land transport~~emissions~~, biogenic ~~emissions~~ and other anthropogenic emissions (anthropogenic non-traffic, and aviation) to ground-level ozone (in %) are shown. Therefore, the contributions do not add up to 100 %. (a) and (b) show the relative ~~contributions~~ contribution of O_3^{tra} ~~at the 99th,95th and 75th percentile of ozone~~; (c) and (d) the relative contribution of anthropogenic emissions (anthropogenic non-traffic and aviation) ~~at the 99th,95th and 75th percentile of ozone~~ ; and (e) and (f) the relative contribution of O_3^{soi} ~~at the 99th,95th and 75th percentile of ozone~~. The lower and upper end of the box ~~indicates~~ indicate the 25th and 75th percentile, the bar the median, and the whiskers the 5th and 95th percentile of the contributions of all gridboxes within the indicated ~~regions~~region. All values are calculated for JJA of the period 2008 to 2010 and are based on 3-hourly model output. The data ~~were~~ are transformed on a regular grid with a resolution of 0.5° x 0.5° to allow for the regional analyses~~on the defined regions~~.

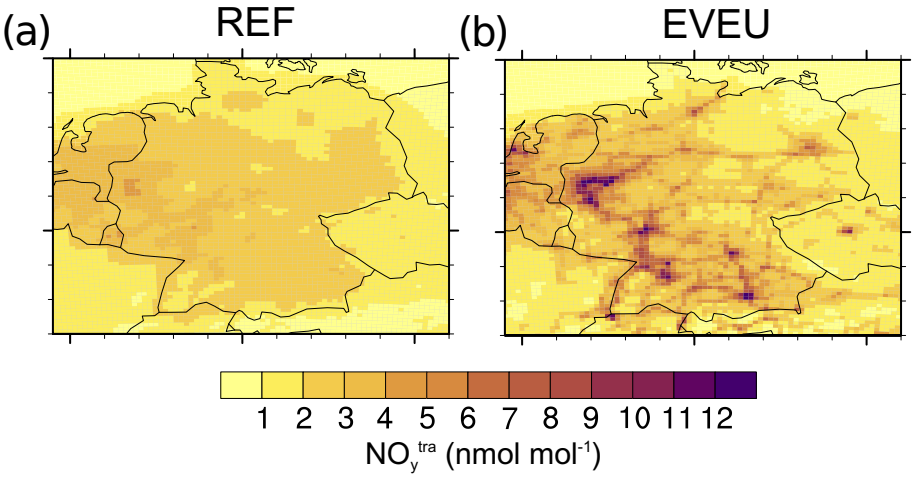

Figure 12. Absolute contribution of NOy^{tra} (in $nmol\ mol^{-1}$) for JJA 2008 of land transport emissions ~~to all reactive nitrogen (, in~~ as simulated by CM12. (a) and (b) show contributions for the period JJA of the *REF* and *EVEU* simulations, respectively.

5 Contribution of land transport emissions to reactive nitrogen in Germany

So far the contributions ~~have been~~ are analysed using the results of the European domain. The resolution of the VEU emission inventory, however, is much finer (roughly 7 km) and therefore the full potential of the emission inventory ~~has not been~~ is revealed. Therefore, this section is dedicated to the results of CM12 focusing on Germany. As shown by ~~? the contributions~~ Mertens et al. (2020) the contribution of land transport emissions to ozone in Germany ~~change~~ changes only slightly, when the model resolution is increased from 50 km to 12 km. The differences caused by the resolution are lower ~~as~~ than the differences between the *REF* and *EVEU* simulation results. Therefore, we focus on the ~~contributions~~ contribution of land transport emissions to NO_y where the results depend stronger on the model resolution. The contribution of O_3^{tra} for Germany is discussed at the end of Sect. 4.1.

Figure 12 shows the absolute contribution of NO_y^{tra} for JJA 2008 as simulated by CM12. As already discussed, the differences between the two emission inventories are rather large. The *REF* simulation ~~results show~~ shows maximum contributions of around 5 $nmol\ mol^{-1}$, while the *EVEU* simulation ~~results show~~ shows contributions of up to 12 $nmol\ mol^{-1}$. These large values occur around the large cities in Bavaria (Munich, Nuremberg) and the large cities in (South-)Western Germany (Stuttgart, Frankfurt, Rhine-Ruhr area). These results indicate the importance of land transport emissions for the mixing ratios of reactive nitrogen levels in German cities. Further, they clearly show the importance of fine resolved emission inventories (and models) for source apportionment of short lived chemical species.

6 Contribution of land transport emissions to the tropospheric ozone budget in Europe

~~To investigate~~ We analyse the contribution of land transport emissions to the ~~European ozone budget~~ ozone budget in Europe by investigating the net ozone production~~() is analysed~~ , which is defined as:

$$P_{O3} = \mathrm{ProdO3} - \mathrm{LossO3}, \tag{3}$$

with production ($\mathrm{ProdO3}$) and loss rates ($\mathrm{LossO3}$) of ozone as diagnosed by the tagging method for the different tagged categories ~~(see Grewe et al., 2017).~~(see Supplement Sect. S5).

Our analysis shows (see Table 8) that the land transport ~~sector is the second important anthropogenic~~ emissions are the second most important anthropogenic emission sector contributing to P_{O3} in Europe. In general, the results obtained with both emission inventories are rather similar, caused by similar ~~total~~ emissions. Integrated up to 850 hPa values of P_{O3} due to land transport emissions of around 13 $\mathrm{Tg(O_3)\ a^{-1}}$ ozone are simulated, while P_{O3} increase to around 23 $\mathrm{Tg(O_3)\ a^{-1}}$ when integrating up to 200 hPa.

The differences of the contributions of O_3^{tra} discussed in Sect. 4 are mainly caused by the differences of the total emissions of the anthropogenic non-traffic sector. The diagnosed net production for the anthropogenic non-traffic category differs by roughly 30 % between *REF* and *EVEU*, while the total net ozone production differs by roughly 15 %, i.e. due to the lower total emissions in the VEU emission inventory compared to the MAC inventory less ozone is produced.

The regions where ozone is predominantly formed by land transport emissions ~~is~~ are displayed in Fig. 13a and Fig. 13b, showing the relative contribution of land transport emissions to P_{O3}. Here, the analysis is restricted to the period May to September where P_{O3} is largest. Additionally, Fig. 13c and Fig. 13d indicate the emission sectors which contribute most to P_{O3} up to 850 hPa in the respective gridbox. Consistent with previous analyses the results show that the relative contribution of land transport emissions to P_{O3} is in general larger in *EVEU* compared to *REF*. The contribution is lowest ~~in~~ over the Atlantic and along the main shipping routes in the Mediterranean Sea. In these regions ozone ~~within the boundary layer~~ up to 850 hPa is mainly formed from shipping emissions (Fig. 13c and d). Generally, the contribution of land transport emissions to P_{O3} is largest over Central Europe, including parts of the Iberian Peninsula, the British Islands and Italy. In these regions the contributions range from 25 % to 35 % in *REF* and 25 % to 40 % in *EVEU*. Further, the regions of large contributions extend much more to the East (including Austria and Hungry) in *EVEU* compared to *REF*. Besides these regions the contributions of land transport emissions to P_{O3} range from 15 % to 20 % in most ~~regions~~areas. However, both simulation results indicate regions especially in Northern Europe, but also in the Mediterranean Sea and Africa with very large contributions (above 35 %). These regions, however, generally show low absolute values of P_{O3}. Therefore, the large ~~contributions~~ contribution of land transport emissions ~~are~~ is not very meaningful. With contributions from 25 % to 40 %, land transport emissions ~~are clearly an important contribution~~ contribute significantly to the ozone production up to 850 hPa. However, in only very few regions (Western Germany, Austria and Northern Italy) and only in *EVEU* land transport emissions are the most important contributor to P_{O3} (Fig. 13).

Outside these regions the results of *REF* and *EVEU* show that biogenic emissions are most important over the Iberian Peninsula~~and over~~ , large parts of Eastern Europe~~as well as over~~ , and Africa. For Central Europe and Northern Europe the

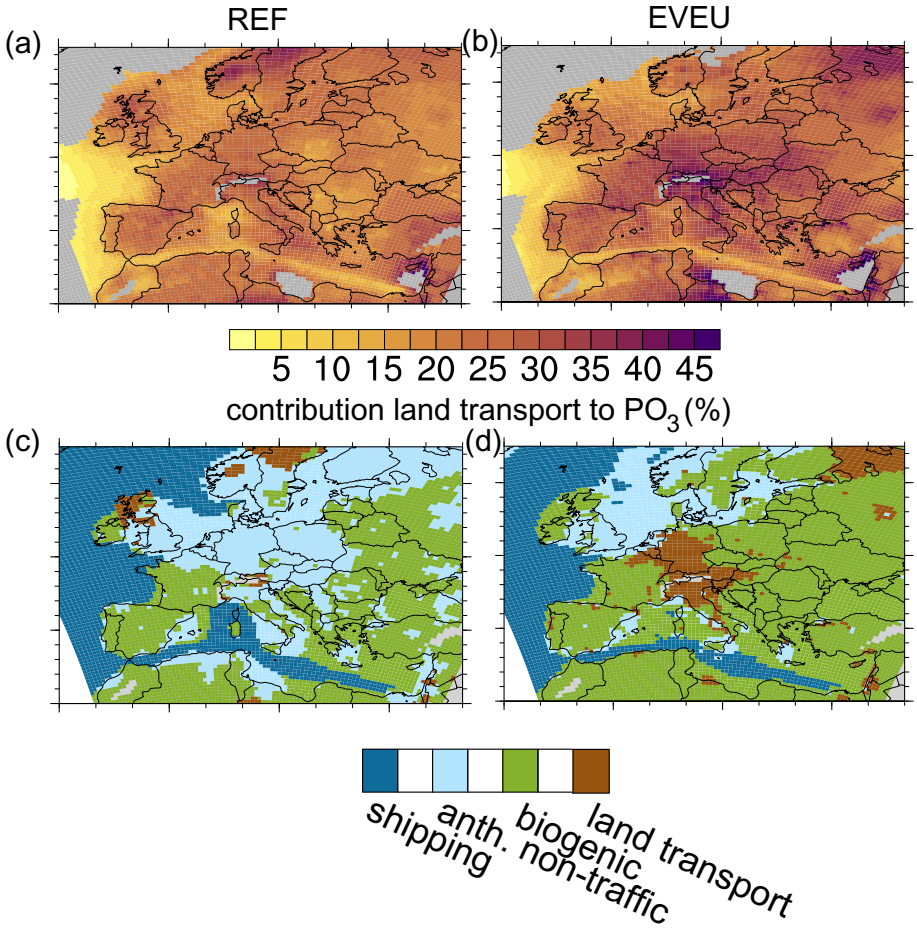

Figure 13. Contribution analysis for P_{O3} integrated from the surface up to 850 hPa. (a) and (b) ~~shows~~ show the relative contribution of land transport emissions to P_{O3} (in %) for the *REF* ~~(a)~~ and the *EVEU* ~~(b)~~ simulation, respectively. (c) and (d) indicate the emission ~~sector~~ sectors which ~~contributes~~ contribute most to P_{O3} up to 850 hPa, for the *REF* and *EVEU* simulation, respectively. Analysed are averaged data for the period May–September 2008 to 2010 as simulated by CM50. Grey areas in (a) and (b) indicate regions where P_{O3} is below $1.5 \cdot 10^{-13}$ mol mol^{-1} s^{-1}. In these regions no relative contributions are calculated for numerical reasons.

REF results indicate that the anthropogenic non-traffic ~~sector~~ category is most important, while *EVEU* indicated biogenic and land transport as being most important. ~~Again, this clearly shows~~ This underlines that the uncertainty of such analysis is strongly influenced by the uncertainties of the anthropogenic and biogenic emission inventories (or parametrizations to calculate these emissions).

7 Discussion

Our analyses ~~demonstrates~~ demonstrate the importance of land transport emissions to European reactive nitrogen (NO_y) ~~concentrations~~mixing ratios. The largest contribution of land transport emissions to NO_y are simulated in Southern England, Benelux, Rhine-Ruhr, Paris and the Po Valley. These regions ~~corresponds~~ correspond well with the regions where ground-level measurements, satellite observations or air-quality simulations report the largest nitrogen dioxide levels (e.g. Curier et al., 2014; Vinken et al., 2014b; Terrenoire et al., 2015; Geddes et al., 2016; European Environment Agency, 2018). ~~Even tough, the absolute contribution (5 to 10) of land transport emissions to~~ While the absolute contributions in these regions strongly depend on the applied emission inventory ~~the relative contributions~~ (5 to 10 $\mathrm{nmol\ mol^{-1}}$), the relative values are 50 % and more. Accordingly, land transport emissions are one of the most important ~~contributor~~ contributors to NO_y in regions with large NO_2 concentrations.

These large amounts of NO_x emissions from land transport clearly contribute to the formation of ozone, but the relative contributions to ozone are lower ~~as~~ than the contributions to NO_y. Here, the mean contributions range between 10 % and 16 % in most regions and even during extreme ozone events the contributions are below ~~25~~30 %. Clearly, land transport emissions are an important contributor to European ozone levels, but they are not the most important contributor to European ozone levels. This is underlined by our analysis of the contribution of land transport emissions to ozone production in Europe, which range between 20 % to 40 % in most areas. The emission sectors which are most important for ozone production in Europe are biogenic emissions ~~as well as~~ and anthropogenic non-traffic emissions. During periods of large ozone values, however, our analyses show that the contribution of land transport emissions to ozone ~~increase~~ increases strongly, while the contribution of anthropogenic non-traffic emissions is only slightly changed. ~~Therefore,~~ This indicates that land transport emissions ~~contribute strongly~~ are an important contributor to large ozone ~~levels~~values.

We find that the regions ~~of the largest contributions~~ with the largest contribution of land transport emissions to ozone are not necessarily identical with the regions ~~of~~ with the largest contributions to reactive nitrogen. The ozone values ~~mainly peak~~ peak mainly in Northern Italy (around the Po Valley) and Southern Germany, which is consistent with the findings of Tagaris et al. (2015). Especially for the Po Valley ~~ground level~~ ground-level measurements show that this is one of the regions in Europe with the largest ~~largest~~ ozone levels (e.g. Martilli et al., 2002; Guerreiro et al., 2014; European Environment Agency, 2018). In ~~the regions of~~ Southern England, around Paris, and the Benelux as well as Rhine-Ruhr regions, where the contribution of land transport emissions to NO_y stands out, the ~~contribution~~ contributions to ozone are not ~~largest.~~ the largest. The result that regions are hot-spots for NO_y from land transport emissions, but not for O_3 from land transport is counter intuitive. The reasons for this is that large amounts of NO_x emissions alone are not sufficient for large ozone production. This is caused by the ~~complexity and non-linearity of ozone chemistry, which depends not only on the amount of ozone precursors but also on the~~ non-linearitiy of the ozone chemistry and the strong interdependence of ozone production and meteorological conditions (e.g. Monks et al., 2015).

~~In general our findings with respect to the contributions of land transport emissions to ozone are in agreement with previous studies either using the perturbation method (Tagaris et al., 2015) or a tagging method (Valverde et al., 2016; Karamchandani et al., 2017).~~

~~In detail, however, previously reported values have been slightly larger. Tagaris et al. (2015) found impacts of up to~~ 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 ~~%in Northern Italy and~~ %. While their largest impacts occur in similar regions as our largest contributions (Southern Germany, ~~and (Valverde et al., 2016) and Karamchandani et al. (2017) report values in the range of 11 to 25 %for most European cities (only for Budapest Karamchandani et al. (2017) reported even larger values of up to 35 %). Compared to these values, however, it is important to note that we investigate the effect of global~~ 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 ~~European ozone values, while the previous studies investigate only effects due to regional emissions and do not attribute contributions of e. g. land transport emissions in North America. Further , in all studies different emissions, different ozone metrics, simulation periods and most importantly different analysis methods are used. Previous publications have appliedthe CMAQ-ISM method (Valverde et al., 2016) or the CAMx OSAT method (Karamchandani et al., 2017). These two methods apply a sensitivity approach to check, if ozone production is~~ ozone over Europe using a tagging method either applied the CAMx model (CAMx OSAT method, Karamcl 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. Depending on this limit ozone production is attributed only~~ 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 11 to 24 %, 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 NO_x and VOC precursors. Land transport emissions contribute mainly to 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 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 NO_x tagging, attribute ozone production under VOC limitation mainly to biogenic sources, while under a 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 NO_x and VOC ~~emissions. The tagging approach of Grewe et al. (2017) used in this study takes competing effects into account and always attributes ozone production to the sectors of all reaction partners (~~or HO_x precursors.

Similar effects can also be observed when comparing our results to the results of Lupaşcu and Butler (2019), who applied a NO_x 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 NO_x emissions, while we attribute ozone to biogenic 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 %. 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 ~~and or~~, soil NO_x and ~~).~~ biogenic VOCs) to fully analyse these differences. The discrepancy in the results of the different source attribution methods clearly shows that a coordinated comparison between these methods is important. This have already been suggested by Butler et al. (2018).

The comparison of the results for the two emission inventories sheds light on the uncertainties associated with such a source apportionment method. The differences of the results for the direct pollutants CO and NO_y are rather large. The mean ozone contributions are much less influenced ~~as~~ than the direct pollutants. Especially during winter and in the middle/upper

troposphere the contributions are mainly dominated by long range transport (e.g. land transport emissions from the rest of the world). In our study, however, we focused ~~on effects only due to European emissions~~only on the uncertainties caused by different emission inventories for Europe. Therefore, we did not investigate the influence of uncertainties from emissions from the rest of the world. Uncertainties of these emissions are likely to influence the contribution from long range transport.

While the mean values of ozone are only slightly influenced, the analysis of extreme values and the analysis of the emission sectors which are most important for regional ozone production differ largely between the different inventories, even ~~tough~~ though the total land transport emissions between the two emission inventories are similar. The differences are mainly caused by the differences of the anthropogenic non-traffic and the shipping emissions between the two emission inventories. Accordingly, the source attribution of land transport emissions is not only influenced by the uncertainties of the land transport emissions~~itself~~, but also by the uncertainties of all other emission sectors. As an example, Kuik et al. (2018) reported an underestimation of road traffic emissions for Berlin of up to 50 %. The impact of such large underestimations on the source attribution results ~~have~~ need to be investigated. Besides ~~the uncertainties of the land transport and other~~ uncertainties of anthropogenic emissions, ~~especially the emissions of the biogenic sector contribute largely to ozone production in Europe. Accordingly, uncertainties in the biogenic emissions contribute to the analysis of this study~~uncertainties of biogenic emissions also contribute to uncertainties of the source apportionment results. In this context especially ~~the soil~~ uncertainties of biogenic VOC emissions and NO_x emissions from soil play an important role~~as~~. As an example, the uncertainties of ~~these emissions~~ soil-NO_x are rather large (Vinken et al., 2014a) and the emissions applied in our model system are at the lower end of current emission estimates. Similar large uncertainties are also reported for biogenic VOC emission inventories (Ashworth et al., 2010; Han et al., 2013; Oderbolz et al., 2013).

Generally, uncertainties caused by the emissions are larger ~~as~~ than the uncertainties, which are caused by the simplifications applied in our source apportionment method, which are in the order of some percent (see also discussion by Mertens et al., 2018). Further, our results indicate a large seasonal variability of the contribution of land transport emissions to ozone. This variability is not only caused by the meteorological conditions but also by the seasonal cycle of other emissions. Accordingly, not only the total emissions of different emission sectors but also their seasonality (and the correct representation of this seasonality) plays an important role.

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 of the methods. 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.

8 Conclusions

In the present study we investigate the contributions of land transport emissions to pollutants in Europe and Germany, focusing on ozone (O_3), carbon monoxide (CO) and reactive nitrogen (NO_y) ~~species~~ by means of simulations with the MECO(n) model system. This model system couples a global chemistry-climate model on-line with a regional chemistry-climate model. To quantify the contributions of land transport emissions to these species we used a tagging method for source apportionment. This tagging method is an accounting system, which completely decomposes the budgets of ozone and ozone precursors into contributions from different emission sources. For the first time such a method is applied consistently in the global as well as the regional models to attribute ozone and ozone precursors to the emissions of land transport~~to ozone and ozone precursors~~. To consider the uncertainties associated with the emission inventories, we performed simulations with two different emission inventories for Europe.

The contribution of land transport emissions to ~~ground level~~ ground-level NO_y depends strongly on the applied emission inventory. In general the contributions range from 5 to 10 $\mathrm{nmol\ mol^{-1}}$ near the European hotspot regions, which are Western and Southern Germany, the Po Valley, Southern England as well as the Paris and Moscow metropolitan region. In most other parts in Central and Southern Europe contributions of around 2 to 3 $\mathrm{nmol\ mol^{-1}}$ are simulated. Generally, absolute contributions during winter are larger ~~as~~ than during summer, but the seasonal differences are smaller ~~as the differences by~~ than the differences between different emission inventories. The absolute contributions correspond to relative contributions of 50 to 70 % to ground-level NO_y, which indicates that land transport emissions are one of the most important sources for NO_y near ~~ground level~~ground-level.

Similar as for NO_y the simulated ~~contributions~~ contribution of land transport ~~emission~~ emissions to CO near ~~ground level~~ ~~depend~~ ground-level depends strongly on the applied emission inventory. Generally, the contributions range ~~between~~ around 30 $\mathrm{nmol\ mol^{-1}}$ during summer in regions which are not directly associated with large land transport emission sources and ~~contributions of~~ more than 75 $\mathrm{nmol\ mol^{-1}}$ near emission ~~hot spots~~ hot-spots such as Paris or Moscow.

The contribution of land transport emissions to ozone, which is a secondary pollutant, shows a geographical distribution which ~~differ~~ differs strongly from the distribution of the primary emissions. The absolute contribution shows a strong North-West South-East gradient with the largest contributions around the Mediterranean Sea. Due to the non-linear behaviour of ozone chemistry and the strong dependency of ozone formation on the meteorology and other precursors as NO_x, regions with large emissions in Western Europe (Benelux, British Islands, Western Germany) show no peak of the contribution of land transport emissions to ozone. ~~This~~ Such a peak is simulated in the Po Valley, where large emissions and favourable conditions for ozone production ~~are present~~prevail. Generally, the contribution has a strong ~~annual~~ seasonal cycle with values of 2 to 3 $\mathrm{nmol\ mol^{-1}}$ during winter and 5 to 10 $\mathrm{nmol\ mol^{-1}}$ during summer. These absolute contributions correspond to relative contributions ~~between~~ in the range of 8 to 16 %. During winter, the results obtained for the two European emission inventories show almost no differences. The contributions are largely determined by long range transport and the ~~inter annual~~ year-to-year variability is the largest source of uncertainty. Of course~~also uncertainties of~~ , also the uncertainties in the emission inventories ~~outside Europe play an important role for the uncertainties~~for emissions outside of Europe can influence the contribution analyses

considerably, but this ~~role has not be~~ has not been investigated in the present study. During summer the differences between the contributions diagnosed using the two emission inventories are larger ~~as the inter annual variabilityand hence~~ than the year-to-year variability. Hence, during summer uncertainties of emission inventories ~~contribute strongly to the uncertainties of the contribution analysis of land transport emissions to ozone. While the emission~~ for Europe influence the contribution analyses considerably.

While the emissions of the land transport sector have almost no seasonal cycle, the contributions have a strong seasonal cycle~~of the contribution of these emissions to ozone show the~~ . This shows the strong influence of seasonal cycles of other emission sources on the ozone production from land transport emissions. Hence, uncertainties ~~in the totals, geographical distributions~~ of total emissions, geographical distribution and the seasonal cycle of other emissions strongly influence the contribution analysis of land transport emissions. Especially during summer ~~one key role are the biogenic emissions~~ biogenic emissions play a key role here. The impact of uncertainties of these emissions ~~need to be studies~~ needs to be studied in more detail. In addition, the impact of the applied source apportionment method needs to be investigated in a coordinated way. Our results suggest, that our methodology, which accounts for NO_x and VOCs at the same time, leads to a partitioning between anthropogenic and biogenic sources partly different from previous studies which account for NO_x or VOCs.

The ~~contributions~~ contribution of land transport emissions to extreme (99th percentile) ozone values is largest in the Po Valley, reaching up to roughly ~~25~~28 %. In other regions of Europe the ~~contributions~~ contribution of land transport emissions to extreme ozone events ~~are~~ is lower and strongly ~~depend~~ depends on the region and the emission inventory. Important is, however, that the contribution of land transport emissions to ozone increase with increasing ozone levels. This indicates that land transport emissions play an important role for high ozone events. Generally, the contribution of land transport emissions ~~contribute to the~~ to ozone production up to 850 hPa ~~to~~ is around 20 and 40 % in most European regions. However, only in very few regions land transport emissions are the most important ~~contributions o~~ contributor to the ozone production. In most ~~region~~ regions anthropogenic non-traffic and biogenic emissions are more important. Our analysis shows that especially also the biogenic emissions are ~~also~~ important during high ozone events~~as the contribution of biogenic emissions increase~~ . Their contribution increases with increasing ozone levels similar to the contribution of land transport emissions. The ~~contributions~~ contribution of anthropogenic non-traffic emissions ~~show~~ shows almost no increase. However, the large differences obtained for the two emission inventories indicate a large uncertainty range of such analysis.

As a next step the ~~performed~~ analysis will be refined using ~~a source apportionment , which differentiate~~ source apportionment categories, which differentiates between contributions of European land transport emissions and land transport emissions from the rest of the world. Such an analysis will help to quantify the importance of European and global land transport emissions ~~,~~ to ozone levels in Europe. Further, more reliable emission estimates are important for follow up studies. Here, the focus should not only be on the land transport emissions, but also on other important emissions, including especially biogenic VOCs and soil-NO_x emissions, which ~~have~~ are subject to large uncertainties and contribute strongly to European ozone levels. To better constrain the uncertainties of the contribution analysis follow up studies are planned (see Sect. 7) in which we will combine observational data of specific aircraft measurement campaigns together with model results including the analysed contributions.

Code and data availability. The Modular Earth Submodel System (MESSy) is continuously further developed and applied by a consortium of institutions. The usage of MESSy and access to the source code is licenced to all affiliates of institutions which are members of the MESSy Consortium. Institutions can become a member of the MESSy Consortium by signing the MESSy Memorandum of Understanding. More information, including on how to become licensee for the required third party software, can be found on the MESSy Consortium Website (http://www.messy-interface.org). The simulations have been performed with a release of MESSy based on version 2.50. All changes are available in the official release (version 2.51). The namelist set-up used for the simulations is part of the electronic supplement. The data used for the figures will be part of the Supplement after the manuscript is accepted for final publication.

Author contributions. MM performed the simulations, analysed the data and drafted the manuscript. AK and PJ developed the model system. VG developed the tagging method. RS drafted the study. All authors contributed to the interpretation of the results and to the text.

Competing interests. The authors declare that they have no competing interests.

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Table 1. Overview of the most important MESSy submodels applied in EMAC and COSMO/MESSy, respectively. Both COSMO/MESSy instances use the same set of submodels. MMD* comprises the MMD2WAY submodel and the MMD library.

Submodel	EMAC	COSMO	short description	references
AEROPT	x		calculation of aerosol optical properties	Dietmüller et al. (2016)
AIRSEA	x	x	exchange of tracers between air and sea	Pozzer et al. (2006)
CH4	x		methane oxidation and feedback to hydrological cycle	
CLOUD	x		cloud parametrisation	Roeckner et al. (2006), Jöckel et al. (2006)
CLOUDOPT	x		cloud optical properties	Dietmüller et al. (2016)
CONVECT	x		convection parametrisation	Tost et al. (2006b)
CVTRANS	x	x	convective tracer transport	Tost et al. (2010)
DDEP	x	x	dry deposition of aerosols and gas phase tracers	Kerkweg et al. (2006a)
EC2COSMO	x		additional ECHAM5 fields for COSMO coupling	Kerkweg and Jöckel (2012b)
GWAVE	x		parametrisation of non-orographic gravity waves	Roeckner et al. (2003)
JVAL	x	x	calculation of photolysis rates	Landgraf and Crutzen (1998), Jöckel et al. (2006)
LNOX	x		NO_x-production by lighting	Tost et al. (2007), Jöckel et al. (2010)
MECCA	x	x	tropospheric and stratospheric gas-phase chemistry	Sander et al. (2011), Jöckel et al. (2010)
MMD*	x	x	coupling of EMAC and COSMO/MESSy (i.e. library and submodel)	Kerkweg and Jöckel (2012b); Kerkweg et al. (2018)
MSBM	x	x	multiphase chemistry of the stratosphere	Jöckel et al. (2010)
OFFEMIS	x	x	prescribed emissions of trace gases and aerosols	Kerkweg et al. (2006b)
ONEMIS	x	x	on-line calculated emissions of trace gases and aerosols	Kerkweg et al. (2006b)
ORBIT	x	x	Earth orbit calculations	Dietmüller et al. (2016)
QBO	x		Newtonian relaxation of the quasi-biennial oscillation (QBO)	Giorgetta and Bengtsson (1999), Jöckel et al. (2006)
RAD	x		radiative transfer calculations	Dietmüller et al. (2016)
SCAV	x	x	wet deposition and scavenging of trace gases and aerosols	Tost et al. (2006a)
SEDI	x	x	sedimentation of aerosols	Kerkweg et al. (2006a)
SORBIT	x	x	sampling along sun synchronous satellite orbits	Jöckel et al. (2010)
SURFACE	x		surface properties	Jöckel et al. (2016)
TAGGING	x	x	source apportionment using a tagging method	Grewe et al. (2017)
TNUDGE	x	x	Newtonian relaxation of tracers	Kerkweg et al. (2006b)
TROPOP	x	x	diagnostic calculation of tropopause height and additional diagnostics	Jöckel et al. (2006)

~~Definition of the chemical families used in the tagging method which diagnoses the source attribution. More details on the species contained in the families are given in the Supplement of Grewe et al. (2017). Tagged species Description Ozone as family of odd oxygen PAN PAN COCO all chemically active nitrogen compounds without PAN in the chemical mechanisms (15) all NMHCs in the chemical mechanisms (42) OH tagged in a steady state approach (see Rieger et al., 2018) tagged in a steady state approach~~

Table 2. Description of the different tagging categories applied in this study following Grewe et al. (2017). Please note that some tagging categories summarise different emission sectors (see description). The last row shows the nomenclature of the tagged tracers exemplary for ozone.

tagging category	description	notation for tagged ozone
land transport	emissions of road traffic, inland navigation, railways (IPCC codes 1A3b_c_e)	O_3^{tra}
anthropogenic non-traffic	sectors energy, solvents, waste, industries, residential, agriculture	O_3^{ind}
ship	emissions from ships (IPCC code 1A3d)	O_3^{shp}
aviation	emissions from aircraft	O_3^{air}
lightning	~~lightning~~ lightning-NO_x emissions	O_3^{lig}
biogenic	on-line calculated isoprene and soil-NO_x emissions, off-line emissions from biogenic sources and agricultural waste burning (IPCC code 4F)	O_3^{soi}
biomass burning	biomass burning emissions	O_3^{bio}
CH_4	degradation of CH_4	O_3^{CH4}
N_2O	degradation of N_2O	O_3^{N2O}
stratosphere	downward transport from the stratosphere	O_3^{str}

Table 3. Definition of the chemical families used in the tagging method. More details on the species contained in the families are given in the Supplement of Grewe et al. (2017).

Tagged species	Description
O_3	Ozone as family of odd oxygen
PAN	PAN
CO	CO
NOy	all chemically active nitrogen compounds without PAN in the chemical mechanisms (15)
NMHC	all NMHCs in the chemical mechanisms (42)
OH	OH tagged in a steady state approach (see Rieger et al., 2018)
HO_2	HO_2 tagged in a steady state approach

Table 4. Average (2008 to 2010) annual total emissions for the CM50 domain of different anthropogenic emission sectors and the total of all emission sectors for NO_x (in $Tg(NO)\ a^{-1}$), CO ($Tg(CO)\ a^{-1}$), VOC ($Tg(C)\ a^{-1}$) and the NO_x to VOC ratio (NO_x/VOC)).

	REF				*EVEU*			
emission sector	NO_x	CO	VOC	NO_x/VOC	NO_x	CO	VOC	NO_x/VOC
land transport	5.2	29	3.1	1.7	5.4	24	3.4	1.6
anthropogenic non traffic	7.3	28	14	0.52	5.1	30	6.5	0.78
shipping	2.4	0.25	0.36	6.5	1.8	0.30	0.096	19
aviation	0.60	-	-	-	0.55	-	-	-
Total	15.5	57.3	17.5	0.88	12.9	54.3	10.0	1.3

Table 5. Average (2008–2010) annual total emissions for the CM50 domain of NO_x (in $Tg(NO)\,a^{-1}$), CO ($Tg(CO)\,a^{-1}$), VOC ($Tg(C)\,a^{-1}$) and the NO_x to VOC ratio (NO_x/VOC). Given are the total emissions of the emission sectors which are identical in *REF* and *EVEU*.

emission sector	NO_x	CO	VOC	NO_x/VOC
biogenic	1.2	4.8	22	0.056
biomass burning	0.26	9.0	0.377	0.73
agricultural waste burning	0.081	2.845	0.0981	0.83
lightning	0.76	-	-	-

Table 6. Root-mean-square error (RMSE, in μ/gm^3) and mean bias (MB, in percent) of the *REF* and *EVEU* simulations compared to Airbase v8 observation data. Given are the scores for the mean values during JJA and DJF, as well as values for 95th percentile for JJA. For *REF* listed additionally also the scores considering only the values at 12 and 15 UTC.

	RMSE (μ/gm^3)	MB (%)
REF JJA mean	29.2	26.6
REF JJA 12 and 15 UTC	18.7	13.4
EVEU JJA mean	24.3	20.5
REF JJA 95th percentile	26.9	-10.0
EVEU JJA 95th percentile	28.7	-14.2
REF DJF mean	35.1	32.8
EVEU DJF mean	32.8	30.1

Table 7. Contribution of different emission ~~source~~ sources area averaged over Europe (defined as rectangular box 10° W to 38° E and 30° N to 70° E) for JJA 2008–2010 at three different altitudes (in %). The values are mean values of the *REF* and *EVEU* simulation, the range indicates the standard deviation between the results of *REF* and *EVEU*.

	ground (%)	600 hPa (%)	200 hPa (%)
stratosphere	7.4 ± 0.1	13.7 ± 0.1	52.0 ± 0.1
CH_4	14.3 ± 0.1	14.7 ± 0.1	8.3 ± 0.1
lightning	8.8 ± 0.2	15.0 ± 0.5	9.0 ± 0.1
aviation	3.7 ± 0.1	5.2 ± 0.1	2.0 ± 0.1
biomass burning	6.1 ± 0.1	4.8 ± 0.1	2.2 ± 0.1
biogenic	18.8 ± 0.3	15.7 ± 0.1	7.5 ± 0.1
shipping	9.2 ± 0.6	4.7 ± 0.1	1.5 ± 0.1
anth. non-traffic	16.4 ± 0.8	13.0 ± 0.2	6.1 ± 0.1
land transport	11.6 ± 0.4	8.3 ± 0.1	3.3 ± 0.1
N_2O	3.6 ± 0.1	5.1 ± 0.0	8.3 ± 0.1

Table 8. Diagnosed net ozone production (P_{O3}) of the ten considered categories (in $Tg\ a^{-1}$) as simulated by CM50. The production rates are integrated over the CM50 domain and up to 850/200 hPa, respectively. The values are averaged for 2008–2010, the ranges indicate one standard deviation with respect to time based on the annual averages ~~from~~ of the individual years~~2008–2010~~.

	P_{O3} integrated up to 850 hpa ($Tg\ a^{-1}$)		P_{O3} integrated up to 200 hpa ($Tg\ a^{-1}$)	
	REF	EVEU	REF	EVEU
land transport	13.2 ± 0.2	13.3 ± 0.3	22.8 ± 0.6	23.4 ± 0.5
anthropogenic non-traffic	22.2 ± 0.5	15.1 ± 0.3	37.8 ± 1.1	26 ± 0.5
shipping	6.7 ± 0.1	5.6 ± 0.1	10.6 ± 0.1	8.8 ± 0.1
aviation	0.3 ± 0.1	0.1 ± 0.1	8.1 ± 0.1	7.9 ± 0.1
biogenic	15.9 ± 0.6	15.3 ± 0.5	28.8 ± 0.7	28.2 ± 0.7
lightning	-0.9 ± 0.1	-1.0 ± 0.1	6.9 ± 0.3	7.0 ± 0.3
biomass burning	2.1 ± 0.2	1.8 ± 0.1	3.8 ± 0.3	3.5 ± 0.3
CH_4 degradation	4.5 ± 0.1	3.6 ± 0.1	12.5 ± 0.4	11.5 ± 0.4
N_2O	-0.2 ± 0.1	-0.3 ± 0.1	1.8 ± 0.1	1.7 ± 0.1
stratosphere	-1.9 ± 0.1	-1.7 ± 0.6	-10.9 ± 0.7	-11 ± 0.7
total	61.8 ± 0.3	51.9 ± 1.0	122.3 ± 2.0	107.4 ± 1.8