Dear editor,

thank you very much for guiding the editorial process.

According to the referee comments we thoroughly revised our manuscript. In particular, we revised the Introduction, Section 4 and the Conclusion to meet the comments of the referees. To describe the method of our radiative forcing calculations in more detail, we further changed the structure of Section 2 and 6 and added more details concerning the method (new Sect. 2.3).
Further, we checked the manuscript for a consistent wording.

In general, we think that the referees comments are answered in detail. In some cases, however, we think that there were misunderstandings. In these cases we tried to be more precise in our manuscript.

Attached are the comments to the two referees (original comments in italic, answers in normal fonts, changes in the 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)

We thank referee#1 for many useful comments which helped to improve the manuscript. In the following, referee comments are given in italics, our reply's in normal font, and text passages which we included in the text, in bold.

Overview: This paper estimates contributions to ozone using a tagging methodology. They focus on land transportation and shipping, which are important sectors. They compare their results to comparable studies from the past and attempt to distinguish between perturbation and "contributions." The methods are generally clear and the results are well presented. There are several points of interpretation and extension of this work to conclusions that go beyond what the work supports. The main problem in this paper is cooption of terms that this reviewer believes are inappropriate. Much of this is framing, but has important implications that need to be better fleshed out.
Reply: We thank referee#1 for these positive comments. We modified the text accordingly and described the terms we use in more detail or changed parts which might be misleading. Please see below for more detailed responses.

The field has historically estimated "contribution" in many ways including perturbation, source apportionment tagging (e.g., CAMx OSAT/APCA and CMAQ ISAM), renormalized sensitivities (e.g., DDM or adjoint). Yet this paper argues that "only tagging estimates the contribution of emissions." Note that many tagging techniques (OSAT/APCAvand ISAM) have sensitivity-based metrics to account for relative importance (e.g., Sillman-ratio threshold). One goal of the relative importance approaches is to make a "contribution" that is meaningfully consistent with sensitivity because of its useful- ness to policy makers. These relative importance factors are omitted in the technique applied in this paper. Why is this combinatorial tagging the only approach that can estimate "contribution"? If combinatorial tagging is somehow more appropriate, then why not include all reactants? The ad absurdum argument would then say that a large fraction of all ozone is simply natural due to molecular oxygen required for the formation of RO2. Thus, the formulation already assumes that limiting factors are important. Why is the limiting factor not important between NOx and VOC in "contribution"?
Reply: We agree with referee#1 that in the past the term 'contribution' has been used for the results of different methods. However, in the last years this difference between 'impact' (sensitivity, e.g. perturbation or DDM) and 'contribution' (source apportionment, e.g. tagging) has been discussed in several publications from both, the chemistry-climate, and air quality communities (e.g. Grewe et al., 2010; Clappier et al., 2017). Of course, large differences between various source apportionment methods exits, some consider NO_x or VOC only (e.g. Grewe, 2004; Emmons et al., 2012), our method considers NO_x and VOCs, others use thresholds to judge, whether the chemistry is NO_x or VOC limited and attribute ozone to NO_x or VOC emission sources (e.g. Dunker et al., 2002; Kwok et al., 2015).
The calculated contribution of course heavily depends on the applied source apportionment methods. We don't want to judge on any of these approaches

being right or wrong. However, the contributions calculated using a 'NO_x or VOC limit'-threshold are by definition more sensitivity based and not comparable to the contribution estimated by considering NO_x and VOC only, or together.
Our goal was not to say that only the combinatorial tagging can be used to calculate contributions. But the general difference between these source apportionment methods, which usually have closed budgets, and the sensitivity methods is important to us. We revised large parts of the Introduction (see also reply to referee#2) to make clear that we separate between impact/contribution and sensitivity/source apportionment.
The most important change with respect to this comment is:
With respect to the influence of different emission sources on ozone itself, typically two different questions are of interest (e.g. Wang et al., 2009; Grewe et al., 2010; Clappier et al., 2017):

- **How sensitive does ozone respond to changes of a specific emission source (sensitivity study)?**
- **How large is the contribution of different emission sources to ozone (source apportionment)?**

Sensitivity studies are important to investigate the influence of an emission change on, for instance, ozone. Often, the so called perturbation approach has been applied, in which the results of two (or more) simulations are compared: one reference simulation with all emissions and a sensitivity simulation with perturbed emissions. Source apportionment, in contrast, is important to attribute different emission sources to climate impact (such as radiative forcing) or extreme ozone events. Source apportionment studies often use tagged tracers in order to estimate contributions of different emission sources, for instance, to ozone. In this tagging approach, additional diagnostic species are introduced, which follow the reaction pathways of the emissions from different sources (e.g. Lelieveld and Dentener, 2000; Dunker et al., 2002; Grewe, 2004; Gromov et al., 2010; Butler et al., 2011; Grewe et al., 2012; Emmons et al., 2012; Kwok et al., 2015). Other methods exist for both type of studies, which we neglect here for simplicity (see e.g. Clappier et al., 2017).
In a linear system, both approaches, perturbation and tagging, lead to the same answer (e.g. Grewe et al., 2010; Clappier et al., 2017). The O_3 chemistry, however, is highly non-linear. Therefore, both approaches lead to different results, not because of uncertainties in the method, but because they give answers to different questions. Here, we use the following wording to discriminate between these two types of questions and methods, knowing that other authors may use them differently: The impact of a source is calculated by the sensitivity method (here the perturbation approach), while the contribution is

calculated using the source apportionment method (here tagging approach, e.g., Wang et al., 2009; Grewe et al., 2010; Clappier et al., 2017). Accordingly, the impact indicates the effect of an emissions change, while the contribution enables an attribution of ozone (and associated radiative forcing) to specific emissions sources.

The IPCC AR5 WG1 Chapter 8 defined radiative forcing as "an instantaneous change in net (down minus up) radiative flux (shortwave plus longwave; in W m^{-2}) due to an imposed change." AR5s definition is generally consistent with previous definitions (e.g., Seinfeld and Pandis 2006; Jacob 1999). Contribution as defined as the combinatorial tagging is not consistent with an imposed change. First, there is no imposed change. In fact, removing those emissions (tra or shp) would not impose a change of similar magnitude. Thus, the idea that transport or shipping contributes to RF proportionally to combinatorial tagging is conceptually flawed.

Reply: We are not sure, if we understand this comment correctly. From what we understand, referee#1 is arguing that only with the perturbation approach (e.g. by removing the traffic emissions) a radiative forcing (RF) could be calculated. If so, this is an important point and the referee's comment indicates that we need to clarify our RF calculations in more detail to show that it is actually largely in agreement with the IPCC RF definition. To clarify this, we start with the IPCC definition of the tropospheric ozone RF, which is the RF for the ozone change between 1850 and a current situation. We are here interested in attributing this RF to individual source of ozone, such as land transport emissions. For this, we need to know the ozone attributable to the respective emission source. If we add up all RFs for different emission sources based on ozone fields calculated by the perturbation approach, the sum of the RFs calculated for different emission sources is drastically lower than the total tropospheric ozone RF (e.g. Grewe et al., 2012). Hence, the use of the perturbation approach is not in line with the IPCC definition to attribute different emission sources to ozone (see also the simplified sketch in Fig. S1 which is also part of the revised Supplement).

In contrast, the idea of the tagging approach is to attribute the RF of O_3 proportional to the share of O_3 corresponding to the individual emission sources (as performed in a previous study by Dahlmann et al., 2011). The benefit of using the contribution of an emission source (in contrast to using the impact of the emission source) is that for the contribution the sum of the individual radiative forcings is equal to the total RF, i.e. $\sum_i^n RF^i \approx RF$ with RF^i being the radiative forcings of the individual emission source i of n total emission sources. This does not hold for the perturbation approach (Grewe et al., 2012).

To add more details of our approach, we moved the description of the RF calculations from Sect. 6 to Sect. 2 and added further explanations. In addition, we added some details concerning the assumptions used in this method in the Supplement. The description of our RF method in Sect. 2.3 is now:

The radiative forcing (RF) of ozone is defined as the net flux change caused by a change (e.g. between two time periods like pre-industrial and present day, Myhre et al., 2013). Here, we are interested in the

contribution of land transport and shipping to this RF. Due to the non-linearities in the ozone chemistry (see also Sect. 4), we estimate the contribution of the land transport/shipping emissions to ozone and then calculate the RF of these O_3 shares individually. This approach is consistent with the IPCC RF definition, since the sum of all individual RF contributions approximately equals the total RF (for a detailed example see Dahlmann et al., 2011).
Thus, to calculate the O_3 RFs of land traffic and shipping emissions, additional simulations were performed applying the stratospheric adjusted radiative forcing concept (e.g. Hansen et al., 1997; Stuber et al., 2001; Dietmüller et al., 2016). For this, monthly mean fields of the simulation ***RC1SD-base-10a*** are used as input data, of the radiation scheme, except for O_3, which stem from the ***BASE*** simulation. Calculations of the RF based on the results of the tagging approach in accordance with Dahlmann et al. (2011) were performed as follows:

1. Based on the results of the ***BASE*** simulation, monthly mean values of Δ_T^{tra}=O_3 - O_3^{tra} and Δ_T^{shp}=O_3 - O_3^{shp} were calculated. Δ_T^{tra} and Δ_T^{shp} corresponds to the share of O_3 excluding O_3 from land transport and shipping emissions, respectively.

2. Multiple radiation calculations (Dietmüller et al., 2016) were performed, calculating the radiative flux of Δ_T^{tra}, Δ_T^{shp} and O_3. The O_3 RFs of land transport and shipping emissions using the tagging approach are then calculated as follows:

$$\mathrm{RF}_{\mathrm{O3tra}}^{\mathrm{tagging}} = rflux(\mathrm{O_3}) - rflux(\Delta_T^{\mathrm{tra}}), \tag{1}$$

$$\mathrm{RF}_{\mathrm{O3shp}}^{\mathrm{tagging}} = rflux(\mathrm{O_3}) - rflux(\Delta_T^{\mathrm{shp}}), \tag{2}$$

   with $rflux$ being the radiative fluxes calculated for the respective quantity. Accordingly, the calculated RFs measure the flux change caused by the ozone share of land transport and shipping emissions, respectively.

Calculating the RFs based on the results of the perturbation approach is similar to (e.g. Myhre et al., 2011). First, $\Delta O_{3\mathrm{tra}}$ and $\Delta O_{3\mathrm{shp}}$ are calculated by taking the difference between the unperturbed (***BASE***, see below) and the perturbed simulations (***LTRA95*** or ***SHIP95***):

$$\Delta \mathrm{O_3} = (\mathrm{O_3}^{\mathbf{unperturbed}} - \mathrm{O_3}^{\mathbf{perturbed}}) \cdot 20. \tag{3}$$

As we consider 5 % perturbations these differences are scaled by a factor of 20 to yield a 100 % perturbation. To calculate the RFs using the perturbation approach, $\Delta O_{3\mathrm{tra}}$ and $\Delta O_{3\mathrm{shp}}$ are than treated as described above for Δ_T^{tra} and Δ_T^{shp}. These RFs are called $\mathrm{RF}_{\Delta \mathrm{O3tra}}^{\mathrm{perturbation}}$

and $\mathrm{RF}^{\mathrm{perturbation}}_{\Delta\mathrm{O3shp}}$, respectively. Accordingly, the method to calculate the RFs of the O_3 shares analysed by the perturbation and the tagging approach are the same. The differences between $\mathrm{RF}^{\mathrm{perturbation}}_{\mathrm{O3tra}}$ and $\mathrm{RF}^{\mathrm{tagging}}_{\mathrm{O3tra}}$ (and the same for shipping) arise only due to differences of the the differently calculated O_3 shares.
The benefit of using the contribution of an emission source (in contrast to using the impact of the emission source) is that for the contribution the sum of the individual radiative forcings is equal to the total RF, i.e. $\sum_i^n RF^i \approx RF$ with RF^i being the radiative forcings of the individual categories i of n total categories. This hold for the perturbation approach (Dahlmann et al., 2011; Grewe et al., 2012). However, the calculations of the RF is still subject to some specific assumptions, which we discuss in detail in the Supplement.

The authors assert that this technique is useful in understanding changes in emissions (particularly section 4.1). The current state of practice uses an emission reduction matrix to explore sensitivities at multiple emissions reductions (20, 40, 60, 80%) of both NOx and VOC. How is tagging this technique more useful than the iterative NOx/VOC matrix?
Reply: We think that there is a misunderstanding. In the conclusion (last sentences) we clearly state:

'To investigate mitigation options, the tagging method cannot replace sensitivity studies and vice versa. However, we clearly demonstrated that a combination of both methods strengthen the investigation of mitigation options and should be the method of choice.'

As demonstrated in Sect. 4.1 we prefer to apply the tagging method in all sensitivity simulations performed at different emission reduction levels. This is important, because in a non-linear system the success of a particular mitigation option (e.g. reducing road traffic emissions by 10 %) strongly depends on the history of previous emission reductions. For instance in this case the sensitivity method measures the success of all mitigation options, while the additionally applied tagging method provides a more in depth understanding. The additional tagging method helps in attributing the remaining ozone to different sources and demonstrates that, for instance, emissions from industry contribute more to ozone after land transport emissions are reduced, because the ozone production efficiency of the industry emissions increase.
As discussed in the answer to referee#2 we rephrased Sect. 4.1 (page 13–14 of the revised manuscript) to make this more clear. In Addition, we changed the sentence above to: **To investigate mitigation options, the tagging method cannot replace sensitivity (i.e. perturbation) studies and vice versa.**

Finally, I have concern about the methodology as described in Eq 2. Apportionment based on fraction of NOy and NMHC concerns me. See Page,Line

comments.
Reply: Please see below for a detailed answer.

Much of this critique is specific to the interpretation and assertions of unique value. The methods and results are internally consistent. I am skeptical of the species family approach as described. The biggest issue is that the article attempts to fully own the term "contribution", applies combinatorial tagging to RF in an odd way that needs to be clearly distinguished from traditional RF, and implies regulatory value that is likely already met. Most of these comments can be addressed by revising the interpretation.
Reply: As described in detail (above and below) we changed parts of the manuscript to clarify the differentiation between impact and contribution.

1,3: recommend "complementary" because the dynamics of "competition"
Reply: Both, VOC and NO_x, are precursors of ozone and both species are attributed to ozone in our approach, as well as in the approaches by (e.g. Dunker et al., 2002; Kwok et al., 2015). In this sense NO_x and VOC compete for the production.
Since the wording seems to confuse, we have rephrased the sentence:
...but also their non-linear interaction in producing ozone.

1,5-7: The regions are not clear in the abstract. Consider adding "ocean" to each region to be consistent with text and clarify.
Reply: Thanks! Added in the abstract and the conclusion!

1,20: This is a narrow definition of the word contribution and I have seen no argument that combinatorial tagging is the only way to define contribution.
Reply: As discussed above, we not to intend to restrict tagging only to our combinatorial approach, but to all tagged tracer approaches. We added 'source apportionment' in the first paragraph of the abstract to make this more clear. In the Introduction we also added some more details (see above):
We quantify the contribution of land transport and shipping emissions to tropospheric ozone for the first time with a chemistry-climate model including an advanced tagging method (also known as source apportionment), which considers not only the emissions of NO_x **(**NO **and** NO_2**),** CO **or volatile organic compounds (**VOC**) separately, but also their non-linear interaction in producing ozone.**

2,15: It is not important to know "contribution" as defined by combinatorial tagging to define mitigation strategies. In fact, knowing sensitivity is fundamentally more important to mitigation since the mitigation intends to impose a change.
Reply: Indeed sensitivities are important to measure mitigation options, but it is also important to know which emission source contributes most to the ozone budget, in order to investigate, which emission sectors are worth to mitigate.

We rephrased the introduction to make this more clear (see above).

3,4: F should be f
Reply: Thanks! Changed!

4,23-5,2: If implemented as discussed, this approach assumes two things that are fundamentally at odds with our understanding of atmospheric chemistry. First, it assumes that all NOy (NOx + NOz) is equally available for ozone production. This is problematic because NOy photochemical lifetime is much longer than NOx. As a result, this Eq 2 will attribute ozone production to NOx and NOz proportionally. That would lead to ozone being attributed to HNO3^{t}ag *in the mid to upper troposphere. Unless NOy is being defined differently than the field convention, this is troubling. Second, and less concerning, NMHC are not all equally reactive nor do they have equal RO2 yields. Assuming concentration fractions are proportional to combinatorial contribution is not consistent with the chemical mechanism.*
Reply: As discussed in Sect. 7, we are aware of the simplifications of the family approach. These simplifications are necessary in order to have a reasonable balance between complexity of the model and the demand regarding the computational resources (see discussion by Grewe et al., 2017). However, it is important to keep in mind that our tagging method relies on the diagnosed production and loss rates from the chemical solver (MECCA, Sander et al., 2011). MECCA calculates the O_3 production rates for each member of the NO_y family individually, according to their kinetic rate coefficient (e.g. no O_3 is produced in regions, where only HNO_3 is present, see also our chemical mechanism in the Supplement). The family concept in the tagging method, however, can under certain circumstances indeed lead to a missatribution of ozone. Consider a case in which O_3 is locally produced from lightning NO_x emissions. Using the family approach the tagged NO_y family locally may consists also of HNO_3 from e.g. anthropogenic emissions. Accordingly, some of the produced O_3 would be attributed to anthropogenic emissions instead of the lightning emissions. This effect has been investigated by Grewe (2004), who concludes that this effect is important mainly during the first 12 h after a major emission and during this time may lead to an error caused by the family concept of up to 10 %.
Wee added a note on this in Sect. 7:
Grewe (2004) showed for a simple box model that the implementation of the NO_y family causes an error mainly after the first 12 h after major emission and during this time may lead to and error caused by the family concept of up to 10 %.

5,23: Februar[y]
Reply: Thanks! Fixed!

6,12: Is the seasonality of non-traffic reasonable and expected?
Reply: Yes. The sectors 'Energy' and 'Residential' are important contributors to the non-traffic emissions, especially during winter, e.g. due to heating. For a

comparison, Fig S2 shows the monthly total anthropogenic non-traffic emissions of the MACCity (used in our study) and the EDGAR emission inventory.

6,24: Why is July most comparable? What did those studies look at?
Reply: In all other studies O_3 impacts for July conditions are presented. Therefore, we report our values also for July conditions. We changed the sentence to make this more clear:
Please note that we list our values in Table 3 for July conditions only, to be comparable to other studies, since they also reported values for July conditions.

7,3: Reword or edit grammar
The sentence "However, compared to other 5 % studies our results show, especially for NA, slightly larger values. This might be caused by a different geographical distribution and larger CO and NMHC emissions in our applied emission inventory. " was changed to:
However, in general our simulation results show larger values compared to these previous findings. These differences are noticeable especially for the NA region. The differences might be caused by a different geographical distribution of the emissions, as well by larger CO and NMHC emissions in the emission inventory we applied.

7,8: This assumes that contribution == tagging, which the authors need to further consider.
Reply: As discussed above the differentiation between perturbation (impact) and tagging (contribution) is well known and discussed in more detail in the references provided in this sentence. We rephrased this sentence and add a new reference (Clappier et al., 2017) to make this more clear:
The comparison of our results using the 5 % perturbation approach with the results using the tagging approach clearly confirms the known differences between estimates of the impact (perturbation) and contribution (tagging) (e.g. Wang et al., 2009; Grewe et al., 2010; Emmons et al., 2012; Grewe et al., 2012, 2017; Clappier et al., 2017).

9,1-4: Are these ratios of partial column or average ratios?
Reply: We always consider partial columns up to 850 hPa in DU in Sect. 4. We rephrased the paragraph slightly to make this more clear. In addition we added a proper unit to Fig.4. The sentence is now:
To investigate this effect in more detail, $\Delta O_{3\mathrm{tra}}$ (see Eq. 3) is analysed further. Here, we consider not only ground-level values, but partial ozone columns integrated from the surface up to 850 hPa **(called 850PC, in** DU**).**

10,2: consider replacing "almost" with "closest to".
Reply: Thanks! Changed!

10,7-12: Are not mitigation strategies more aligned with sensitivities?
Reply: Of course, the success of a mitigation strategy is measured for instance by the reduction of ozone. This can be assessed with the perturbation approach. However, the perturbation approach does not give any information about changes of the ozone production efficiency from one sector, if other emissions are changed. This can be achieved with the tagging approach. Therefore, we propose to combine both methods (see next answer).

10,20-31: See discussion of sensitivity matrix, which is the current approach for developing mitigation.
Reply: As noted above we do not propose to replace sensitivity studies with tagging simulations, because tagging cannot replace perturbation to investigate the successes of a mitigation strategy. We propose to combine both methods, because the success of a mitigation measure depends on the sensitivity. Therefore, the success of one individual emission reduction strongly depends on the history of all previous emission reductions. The perturbation approach provides the general 'sucess' with respect to changes in ozone, while the results of the tagging approach allow an in-depth understanding of the results, an attribution of ozone to emission sources, and show how the production efficiency of other emission sources increase, if for instance road traffic emissions are decreased.
We largely rephrased Sect. 4.1 (see page 13–14 of the revised manuscript) making this point more clear:

The tagging approach does not give any information about the sensitivity of the ozone chemistry with respect to a change of emissions.
....
A combination of tagging and perturbation is a powerful tool for putting additional pressure on unmitigated emission sources, because, even if the absolute ozone levels do not change, their shares in high ozone values (or radiative forcing) increase.
11,20: "[global] land transport." This section is tricky because the production may come from upwind sources. Try to be more explicit.
Reply: This is indeed a very good point. To make this more clear we added an additional sentence at the beginning of the paragraph:
Please note, in our tagging method we distinguish only between different emission sources, but not between emission regions. Therefore, the budgets analysed for distinct geographical regions might not be solely influenced by regional emissions, but also by upwind sources.

13,24: Be more specific than 'some'.
Reply: We changed the sentence accordingly:
Recent updates of the tagging scheme with respect to differences of the HO_x family show an influence of 1–3 percentage-points on the relative contribution of land transport and shipping emissions (Rieger et al., 2017).

13,25: trough − > through?
Reply: Yes! Tanks!

13,25: is the author referring to engineering simplifications in the CTM?
Reply: Yes. We rephrased the sentence:
Therefore, we conclude that the error through the simplifications of the tagging method is estimated to be smaller than the errors arising from approximations applied in the global chemistry-climate-models itself (physics and chemistry parameterisations, e.g. 20 % given by Eyring et al., 2007).

13,28-29: CAMx OSAT/APCA[camx.com] and CMAQ ISAM [doi: 10.5194/gmd-8-99-2015] are a couple of examples of similar complexity to this scheme.
Reply: We are well aware of these approaches, which are mainly used in regional air quality modell (and, to our knowledge are not used in global chemistry-climate models). However, as discussed, these approaches are based on thresholds of the NO_x/VOC sensitivity, well chosen for the intended purpose. But they are not comparable to our approach, which accounts for the competing effects between all species. Approaches by Emmons et al. (2012) or Butler et al. (2011) are also available on the global scale, but consider either only NOx or VOC only. We rephrased this sentence:
Other available tagging schemes, however, are based on kinetic approaches (Gromov et al., 2010), consider either only NO_x or VOC (e.g. Emmons et al., 2012; Butler et al., 2011), or are based on thresholds depending on whether the ozone chemistry is NO_x or VOC limited (e.g. Dunker et al., 2002; Kwok et al., 2015). The differences between the assumptions and the scales on which they are applied render a detailed comparison impossible.

14,22-24: One interpretation is that the radiative forcing in this paper is an overestimate due to the lack of realism in the tagging compared to an actual imposed response.
Reply: We do not agree with Referee#1 on this point. The larger RFs using the tagging approach compared to the perturbation approach are due to larger ozone shares. As discussed above, the methodology of calculating the RFs is the same between tagging and perturbation. However, to make this more clear we add zonal averages of the contribution and the impact of both emission sources to the Supplement and to this reply (see Fig. S3). Further, we stressed this point in more detail in Sect. 6 and in the conclusion. In Sect.6 the following note were added:
However, the RF obtained by the tagging approach is much larger than the RF obtained by the perturbation approach. In particular, the peak at around 20°N is more enhanced for the tagging approach. This is mainly caused by the larger O_3 shares in the upper troposphere, where O_3 is most radiative active, as estimated by the tagging compared to the perturbation approach (see Supplement for a figure

showing the individual shares).
Further the following note were added to the conclusion:
While our estimates of the contribution of land transport and shipping emissions to tropospheric ozone are similar compared to previous studies using a 100 % perturbation, our estimates of the radiative forcing are larger by a factor of 2–3 compared to previous estimates using the perturbation method. As discussed in detail, this large difference compared to previous values is largely attributable to differences in the methodology, leading to different estimates of the ozone shares attributable to land transport and shipping emissions, respectively.

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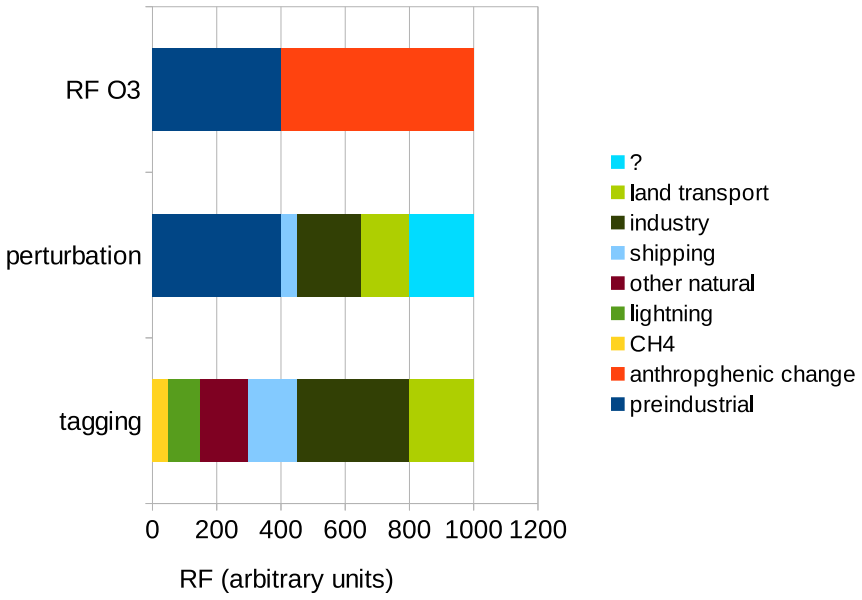

Figure S1: Simplified sketch of three different ways to calculate RFs. 'RF O3' shows the classical way of calculating the anthropogenic RF by calculating the radiative flux of an preindustrial simulation and a simulation with all emissions. 'Perturbation' shows the perturbation approach, here the RF of different emission sources is estimated by perturbation simulations turning specific emissions off. This approach, however, leads to a part of ozone which can not be attributed to one sector (marked with ?). This is mainly caused by changes of the ozone production efficiency. The 'tagging' method estimates a radiative forcing for every specific category. Accordingly, a complete attribution of the RF to specific emission sources is possible.

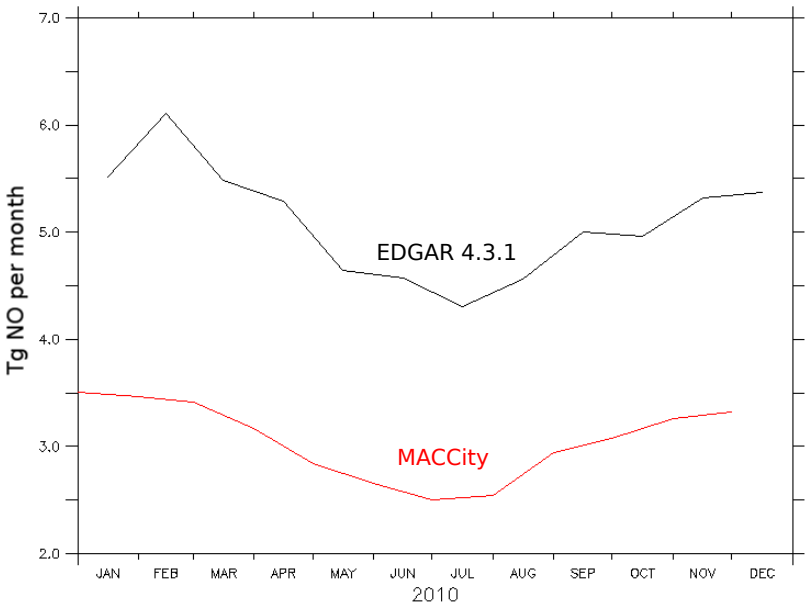

Figure S2: Globally integrated NO_x emissions (in Tg (NO) per month) of the anthropogenic non-traffic sector for the MACCIty emission inventory (red) and the EDGAR 4.3.1 inventory (black). Shown are values for the year 2010 exemplarily.

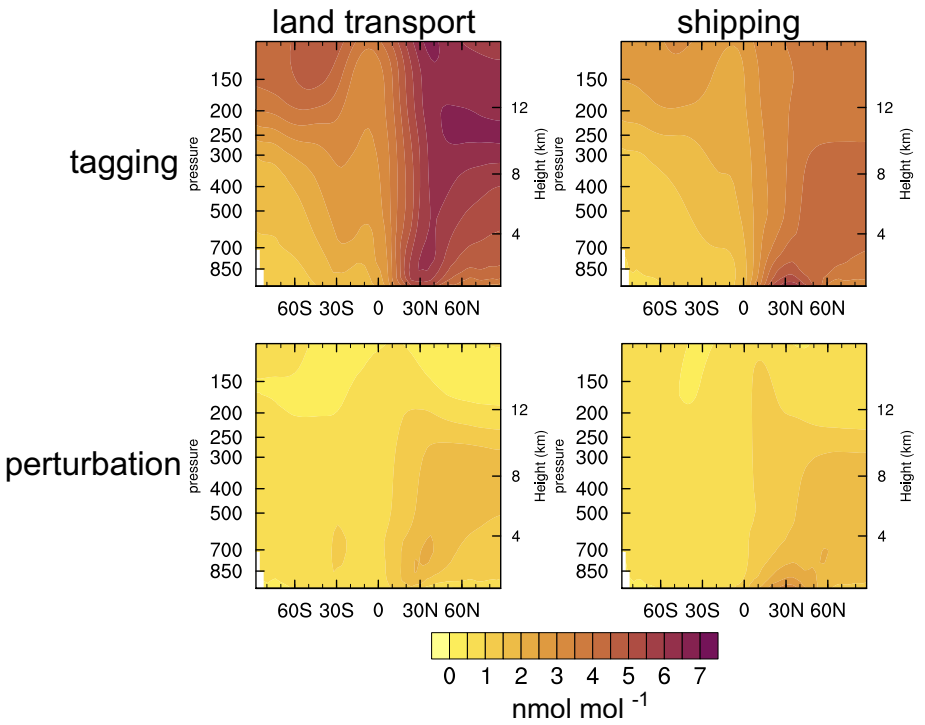

Figure S3: Multi-annual zonal average (2006–2010) O_3 shares as estimated by the perturbation method and the tagging approach. Shown are the contribution and impact of the land transport and shipping emissions to ozone, as estimated by the tagging method and the perturbation approach, respectively.

We thank referee#2 for many useful comments, which helped to improve the manuscript. In the following, referee comments are given in italics, our reply's in normal font, and text passages which we included in the text, in bold.

This paper offers a nice overview of the impact of shipping emissions on ozone through the use of two methodologies: the tagging methodology and the perturbation methodology. The paper is well written and extremely thorough with a clear comparison to previous studies.

Reply: We thank referee#2 for this very positive and encouraging comments.

1. The authors state two goals in this study (p3, l4-6) in determining ozone from shipping emissions: to review previous studies and to give the results of the tagging method. The results from the authors use of the tagging methodology nicely complements estimates from the contribution method. I think the paper works as a review paper. However, as written, I question whether the paper stands very well on its own as a new piece of research. There does not seem to be enough new. Part of the author's justification for this paper is that no one has investigated the ozone contributions from transportation using the tagging approach. Just because something has not been done does not mean it is scientifically interesting or worth pursuing. There are probably other emission sectors that have not been investigated using the tagging approach. It doesn't seem that there should be a new paper written for each of these sectors. I think the authors need to better justify their study than simply state it has not been done. Why do we need another paper on the emission contributions from transportation emissions given the uncertainty? Specifically, what new insights does the tagging approach give? (This needs to be better clarified, see below). What do we learn about the tagging approach here that we didn't know before?

Reply: We thank Referee#2 for acknowledging the review character of our manuscript. Of course, the general difference between tagging and perturbation is well known and has been discussed in many studies (which we cite in the manuscript), especially for simplified models. Of course, it might not be worth to study the difference between impact and contribution for each sector in detail. However, land transport and shipping emissions are very important anthropogenic emission sectors and are therefore subject to mitigation. Further, we want to highlight the following points:

- We here confirm previous results (using different methods) with a new method. This is very important, in particular this shows that those results are robust. Moreover, reproduciblability with different methods is an important aspects in science.
- To our knowledge, we applied for the first time the tagging and the perturbation approach simultaneously and consistently for land transport and shipping emissions,

- including a consistent way of calculating the radiative forcing (RF), thus allowing for a detailed comparison of the results.
- Further, we consider for the first time in a chemistry-climate-model the interactions between NO_x and VOC. Our results indicate that the RFs calculated by Dahlmann et al. (2011) and Grewe et al. (2012) using a NO_x only tagging are likely too large. Accordingly, we present new best estimates of the ozone RF, which are between previous estimates using the perturbation and the NO_x only tagging.
- In addition, the tagging method allows us to present detailed results with respect of the influence of the land transport and shipping emissions on the tropospheric ozone budget.

To stress these aspects more, we revised the Introduction, Section 4/6 and the Conclusion. Please find the detailed differences in the 'diff version' of the revised manuscript.

2. Why does the present study use a 5% perturbation? The results are sensitive to this perturbation. Some justification is needed. It would be helpful for comparison purposes if the authors also gave their results for a 100% perturbation in their tables. To what extent does the discrepancy with the tagging method come from the assumed 5% reduction? It appears a 100% emission reduction gives similar results to the tagging method. Reporting on a 100% emission perturbation would also help compare with other studies.
Reply: As discussed in previous studies, the small perturbation approach minimises the impact of non-linearity. A 100% perturbation is considered as not being realistic (e.g. Hoor et al., 2009; Grewe et al., 2010; Koffi et al., 2010).
In the revised manuscript we added a note on this in Sect. 4:
The 5 % perturbation was chosen as previous studies showed that this small perturbation sufficiently minimises the impact of the non-linearity of the chemistry on the results (e.g. Hoor et al., 2009; Grewe et al., 2010; Koffi et al., 2010).

3. Equation (3): Is a factor of 20 missing?
Reply: In the first version of our manuscript we focused on differences between the 5 % perturbations. Accordingly, no factor was missing. In the revised manuscript we revised this part of the manuscript (see below) and made the factor of 20 more clear.

4. The definition of gamma needs to be clarified in more detail in the text. After looking in detail at the figure and reading the text the meaning of gamma became clear, but it should not have been this difficult. Please clarify the definition of gamma in the text explicitly stating what the y intercept is and stating that y is the average net ozone production rate in a particular region.
Reply: We rephrased the section about Γ considering also your next point to make the definition of Γ more clear. Our definition of Γ is also used in science

of economics. There, elasticity (η) is defined as $\eta = 1 - \Gamma$. In economics η measures the change of an economic variable, if another variable is changed.
The changed paragraph is now:
Based on the results of the *REF* and *LTRA95* simulations, the ozone sensitivity is calculated with the tangent approach in accordance with Grewe et al. (2010) by solving a linear equation ($y = m \cdot (x - x_0) + b$)**. Here,** x **and** y **are the average** NO_x **mixing ratio and the net** O_3 **production** (P_{O3})**, respectively, for a particular region. The** m **denotes the slope, which corresponds to an approximation of the derivative** dP_{O3}/dNO_x **in the unperturbed simulation, which is calculated by the difference in ozone production and** NO_x **mixing ratios in the unperturbed and perturbed simulation.** x_0=NO_x^u **is the** NO_x **mean mixing ratio in the unperturbed simulation and b =** P_{O3}^u-dP_{O3}/dNO_x NO_x^u**, where** P_{O3}^u **is the mean ozone production in the unperturbed simulation.**
Based on the linearised ozone production (P_{O3}^{lin}**) calculated by the tangent approach, we define a saturation indicator** Γ**, which helps to analyse the ozone sensitivity further:**

$$\Gamma = \frac{\text{y} - \text{axis intercept}}{\text{y} - \text{value of unperturbed simulation}} = \frac{P_{O3}^{lin}(NO_x = 0)}{P_{O3}^{lin}(NO_x = \text{unperturbed})}. \quad (1)$$

This value is a quantitative indicator of the chemical regime, showing how much an emission change of one specific sector is compensated by increased ozone productivity of other sectors. $\Gamma = 1$ **indicates a saturated behaviour of the ozone production i.e. the ozone production does not change, if emissions are changed (**$P_{O3}^{lin}(NO_x = 0) = P_{O3}^{lin}(NO_x = \text{unperturbed})$**). Accordingly, there is no ozone reduction because the change of the emissions is entirely compensated by the increase of the ozone production efficiency of other emissions.** $\Gamma > 1$ **indicates an overcompensating effect, i.e., reduced** NO_x **emissions lead to an increase of the ozone production (corresponding to the** VOC**-limited regime). Finally,** $\Gamma = 0$ **indicates a linear response of the system (with a y-intercept at zero). Accordingly, the ozone change introduced by an emission change is not compensated by an increase of the ozone production efficiency. For** $\Gamma = 0.5$ **the ozone change is half compensated by a change in the ozone production efficiency. In terms of the estimated derivative (**dP_{O3}/dNO_x**),** $\Gamma = 1$ **corresponds to** $dP_{O3}/dNO_x = 0$**, while** $\Gamma > 1$ **corresponds to** $dP_{O3}/dNO_x < 0$ **and vice versa.**

5. It is unclear why gamma is defined in terms of the intercept instead of the slope (dO3/dNOx). The intercept will be leveraged by the amount of the NOx emissions. That is, the impact of the slope the will be amplified when the NOx emissions are large by changing the intercept to a greater extent than if the NOx emissions are small.

Reply: Of course Γ could also be defined in terms of the slope (dPO3/dNOx). However, we use Γ as indicator to check whether the ozone production increases, decreases or stays the same with changed emissions. Exactly the same were possible using the slope. To make this more clear we added a comparison between the slope and Γ for the different regimes (see above).

6. Figure 5 clearly demonstrates that the perturbation approach gives different estimates under different conditions. However, it does not show how the tagging approach differs. Some more work is needed here to better understand how these two approaches give different answers depending on ambient conditions and transportation emissions. From line 9 onwards (on page 9) the well-known dependence of ozone production on NOx is shown, with the well-known result that in regions of high NOx a decrease in emissions has little impact on the ozone concentration. There is not much new here. The text and figures don't explicitly show that the tagging approach gives a different answer than the perturbation approach. And isn't the discrepancy between the two methods well known. What is new?

Reply: New is the quantification of the competing effects by combining tagging with the perturbation method and the calculation of the Γ value. Of course, the response of the ozone chemistry to NO_x emissions, as well as the difference between impact and contribution, are well known. We clearly state this in our text and refer to previous publications. It shows that the basic chemical response is in line with previous studies, forming the base for a better understanding and quantification of the underlying processes. We revised the Section 4 (including 4.1 see below) in large parts to quantify the difference between tagging and perturbation in more detail. Please see page 11–14 of the revised manuscript for the changed sections:

As discussed in the previous section and by previous studies (e.g. Wang et al., 2009; Grewe et al., 2010) the perturbation approach

.....

even if the absolute ozone levels do not change, their shares in high ozone values (or radiative forcing) increase.

7. The authors state: 'Combining the tagging and the perturbation approach is there- fore the best way to measure the success of a mitigation strategy.' (p10, l19-20). The authors argue that the perturbation approach gives different answers depending on the current state. I suppose the tagging approach gives the same ozone reduction regardless of the mitigation pathway. This should be clearly stated. Nevertheless, it is unclear how one would use the tagging method to decide on mitigation issues. Perhaps a concrete example would be helpful here? This is important because it would provide a needed justification of the tagging approach. It is crucial that the paper clearly gets this across. It seems to me the tagging approach is useful in assigning blame: for example, if you want to apportion blame for an ozone pollution outbreak or for the radiative forcing due to ozone. It is not clear to me how one would use the tagging method practically in assessing mitigation options.

Reply: We are very thankful to referee#2 for this comment. Of course it is very important to us to get the benefit of combining tagging and perturbation across. Obviously in the first manuscript this point was not stressed enough. Therefore, we revised Subsection 4.1 in large parts (.

8. The loss rate of ozone is very dependent on how it is calculated (page 11). How are the losses calculated in the present study? Are they calculated in the same manner in the comparison studies?
Reply: We considered the following loss rates (cf. equation 14 in Grewe et al. (2017)):

- reactions of O_3 with OH and HO_2,
- effective loss reactions of O_3 with NO_y species,
- reactions of O_3 with NMHCs, and
- reactions mainly of $O^1(D)$ with different species (e.g. $O^1(D) + H_2O$) leading to an effective O_3 loss.

We added our detailed chemical mechanisms which indicates the reactions, which are considered for effective loss and production of O_3 to the Supplement. We added a note on this in our description of the tagging method:
The chemical mechanism including all diagnosed production and loss rates for the tagging method are part of the Supplement. The analysed production and loss rates in Sect. 5 are calculated in accordance with Eq. 13 and 14 of Grewe et al. (2017).

Indeed the values presented by Young et al. (2013), which we use for comparison, are results of a multi-model intercomparison. As stated by Young et al. (2013) not all models, which participated in the intercomparision, calculate ozone loss in a comparable manner (exact details, however, are not given). We added a note on this in the revised manuscript:
Further, it is important to note that loss rates are not calculated consistently in all models presented by Young et al. (2013).

9. P12, l 16: 'We obtain. . ..' Using which method?
Reply: To make this more clear we differentiate in the revised manuscript between $RF^{\text{tagging}}_{\text{O3tra}}$ and $RF^{\text{perturbation}}_{\Delta\text{O3tra}}$ (which we define in Sect. 2):
We obtain a global net RF for land transport of $RF^{\text{tagging}}_{\text{O3tra}} = 92$ mW m^{-2}**. The shortwave RF is 32** mW m^{-2} **and the longwave RF is** 61 mW m^{-2}**. The estimated RF of ship traffic is** $RF^{\text{tagging}}_{\text{O3shp}} = 62$ mW m^{-2} **and smaller than the land transport RF.**

10. The section on uncertainties should also discuss the uncertainties in the perturbation method. In particular this method is sensitive to the perturbation assumed.

Reply: Thanks. This is indeed a good point. We added:
However, also the perturbation approach faces an important limitation. The calculated impact largely depends on the magnitude of the chosen perturbation and the impacts are only valid for this specific perturbation (e.g. Hoor et al., 2009). In addition, the perturbation approach has a fundamental problem, namely a non-closed budget. This means that the sum of O_3 changes calculated for different perturbed emission sources (e.g. land transport and aviation) is not necessarily the total O_3 change if all emissions are reduced at the same time (e.g. Wang et al., 2009; Grewe et al., 2010).

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Revisiting the contribution of land transport and shipping emissions to tropospheric ozone

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Abstract. We quantify the contribution of land transport and shipping emissions to tropospheric ozone for the first time with a chemistry-climate model including an advanced tagging method (also known as source apportionment), which considers not only the emissions of ~~(~~nitrogen oxides (NO_x, NO and NO_2), carbon monoxide CO or ~~non-methane hydrocarbons () separatelybut the competing effects of all relevant ozoneprecursors~~volatile organic compounds (VOC) separately, but also their non-linear interaction in producing ozone. For summer conditions a contribution of land transport emissions to ground level ozone of up to 18 % in North America and South Europe is estimated, which corresponds to 12 $\mathrm{nmol\ mol^{-1}}$ and 10 $\mathrm{nmol\ mol^{-1}}$, respectively. The simulation results indicate a contribution of shipping emissions to ground level ozone during summer in the order of up to 30 % in the Northern Pacific Ocean (up to 12 $\mathrm{nmol\ mol^{-1}}$) and 20 % in the Northern Atlantic Ocean (12 $\mathrm{nmol\ mol^{-1}}$). With respect to the contribution to the tropospheric ozone burden, we quantified values of 8 % and 6 % for the land transport and shipping emissions, respectively. Overall, the emissions from land transport contribute around 20 % to the net ozone production near the source regions, while shipping emissions contribute up to 52 % to the net ozone production in the Northern Pacific Ocean. To put these estimates in the context of literature values, we review previous studies. Most of them used the perturbation approach, in which the results ~~from~~ for two simulations, one with all emissions and one with changed emissions ~~for~~ of the source of interest, are compared. For a better comparability with these studies, we also performed additional perturbation simulations, which allow a consistent comparision of results using the perturbation and the tagging approach. The comparison shows that the results strongly depend on the chosen methodology (tagging or perturbation ~~method~~approach) and on the strength of the perturbation. A more in-depth analysis for the land transport emissions reveals that the two approaches give different results particularly in regions with large emissions (up to a factor of four for Europe). ~~With respect to the contribution of land transport and ship traffic emissions to the tropospheric ozone burden we quantified values of 8 and 6 for the land transport and shipping emissions, respectively. Overall, the emissions from land transport contribute to around 20 of the net ozone production near the source regions, while shipping emissions contribute up to 52 to the net ozone production in the Northern Pacific.~~ Our estimates of the ~~radiative ozone~~ ozone radiative forcing due to emissions of land transport and shipping emissions are, based on the tagging method, 92 $\mathrm{mW\ m^{-2}}$ and 62 $\mathrm{mW\ m^{-2}}$, respectively. ~~Again these results are larger by a factor of 2–3 compared to previous studies~~ Compared to our best estimates, previously reported values using the perturbation approach ~~, but largely agree with previous studies which investigated the difference between the~~

~~tagging and the perturbation method~~are almost a factor of 2 lower, while previous estimates using a NO_x only tagging are almost a factor of 2 larger. Overall our results highlight the importance of ~~differing~~ differentiating between the perturbation and the tagging approach, as they answer two different questions. ~~We~~ In line with previous studies, we argue that only the tagging approach (or source apportionment approaches in general) can estimate the contribution of emissions, ~~while only the perturbation approachinvestigates~~ which is important to attribute emission sources to climate change and/or extreme ozone events. The perturbation approach, however, is important to investigate the effect of an emission change. To effectively asses mitigation options both approaches should be combined. This combination allows to track changes in the ozone production efficiency of emissions from sources which are not mitigated and shows how the ozone share caused by these unmitigated emission sources subsequently increases.

1 Introduction

Ozone in the troposphere has several well known effects: it contributes to global warming due to its radiative properties (e.g. Stevenson et al., 2006; Myhre et al., 2013), and large concentrations of ozone are harmful to humans and to plants (e.g. World Health Organization, 2003; Fowler et al., 2009). In addition, ozone is an important source for the OH radical, which controls the cleansing capacity of the troposphere (e.g. the lifetime of methane, Naik et al., 2013). Due to these different effects ozone is a central species of atmospheric chemistry (Monks et al., 2015).

Two important sources of ozone exist in the troposphere – the downward transport from the stratosphere and the in-situ production from ~~precursors~~ precursor emissions (e.g. Lelieveld and Dentener, 2000; Grewe, 2004). The most important precursors of ozone are carbon monoxide (CO), methane (CH_4), ~~non-methane hydrocarbons (~~volatile organic compounds (VOC) and nitrogen oxides (NO_x=NO+NO_2, e.g. Haagen-Smit, 1952; Crutzen, 1974; Monks, 2005). These precursors have anthropogenic as well as natural sources. Important natural sources of VOCs are biogenic emissions (e.g. Guenther et al., 1995), while NO_x is emitted by lightning (e.g. Schumann and Huntrieser, 2007) and soil (e.g. Yienger and Levy, 1995; Vinken et al., 2014). ~~anthropogenic~~ Anthropogenic sources of ozone precursors, on the other hand, include emissions from industry, land transport ~~(containing the sectors road traffic, inland navigation and railways, e.g. Uherek et al., 2010)~~ (containing the sources road traffic, inland na
shipping (e.g. Eyring et al., 2010). With respect to the influence of different emission sources on ozone itself, typically two different questions are of interest (e.g. Wang et al., 2009; Grewe et al., 2010; Clappier et al., 2017) :

~~To define mitigation strategies or to calculate climate impacts, it is important to know which emission source contributes to what extent to the ozone concentration. Because of~~

- How sensitive does ozone respond to changes of a specific emission source (sensitivity study)?
- How large is the ~~non-linearity of the chemistry it is not possible to calculate the amount of produced ozone directly from the amount of the emissions. Instead, the~~ contribution of different ~~sources needs to be estimated by means of simulations with advanced models, either chemistry transport models or chemistry climate models. However, only the latter allow to directly quantify the impacts of the chemical species on the climate.~~ emission sources to ozone (source apportionment)?

~~Many of such investigations have been performed in the past to estimate the contribution of road traffic (but not the total land transport eff~~
~~shipping (e.g. Lawrence and Crutzen, 1999; Eyring et al., 2007; Hoor et al., 2009; Koffi et al., 2010; Holmes et al., 2014) emissions~~
~~to tropospheric ozone onthe global scale. Typically two different approaches are used in such studies – the perturbation and the~~
~~tagging approach . Most of previous studies used the perturbation approach~~ Sensitivity studies are important to investigate the influence of an emission change on, for instance, ozone. Often, the so called perturbation approach has been applied, in which the results ~~from a reference simulation including all emission sources with the results of a perturbed simulation with changed emissionsof a specific source are compared. In the tagging approach~~of two (or more) simulations are compared: one reference simulation with all emissions and a sensitivity simulation with perturbed emissions. Source apportionment, in contrast, is important to attribute different emission sources to climate impact (such as radiative forcing) or extreme ozone events. Source apportionment studies often use tagged tracers in order to estimate contributions of different emission sources, for instance, to ozone. In this tagging approach, additional diagnostic species are introduced which ~~follows~~ follow the reaction pathways of the emissions from different sources ~~(e.g. Lelieveld and Dentener, 2000; Grewe, 2004; Emmons et al., 2010; Butler et al., 2011; Grewe et al.,~~ ~~Assuming~~ (e.g. Lelieveld and Dentener, 2000; Dunker et al., 2002; Grewe, 2004; Gromov et al., 2010; Butler et al., 2011; Grewe et al., 20 Other methods exist for both type of studies (e.g. sensitivity and source apportionment), which we neglect here for simplicity (see e.g. Clappier et al., 2017) .

In a linear system~~both approaches lead to identical answers (e.g. Grewe et al., 2010) . In case of a non-linear system (as the~~ , both approaches, perturbation and tagging, lead to the same result (e.g. Grewe et al., 2010; Clappier et al., 2017) . The O_3 chemistry~~) both methods~~ , however, is highly non-linear. Therefore, both approaches lead to different results~~as they answer~~ , not because of uncertainties in the method, but because they give answers to different questions. ~~The perturbation approach addresses the question: 'What is~~ Here, we use the following wording to discriminate between these two types of questions and methods, knowing that other authors may use them differently: The impact of a source is calculated by the sensitivity method (here the perturbation approach), while the contribution is calculated using a source apportionment method (here tagging approach, e.g., Wang et al., 2009; Grewe et al., 2010; Clappier et al., 2017) . Accordingly, the impact indicates the effect of an emissions change, while the contribution enables an attribution of ozone (and associated radiative forcing) to specific emissions sources.

In the past, many studies have been performed to estimate the ~~change in ozone, if the emissions of the source of interest are changed?' (e.g. Wu et al., 2009; Grewe et al., 2010) . Accordingly the perturbation approach calculates the~~ impact of ~~e. g. land transport emissions on tropospheric ozone~~road traffic emissions (but not the total land transport effect, e.g., Granier and Brasseur, 2003; N the global scale. However, only few studies exist estimating the contribution of road traffic emissions on ozone: Dahlmann et al. (2011) and Grewe et al. (2012) used a tagging approach considering only NO_x. Further, these studies focussed mainly on globally averaged tropospheric ozone columns and associated radiative forcings without regional quantifications of the contribution. Similar, for the shipping sector previous studies focused on the calculation of the impact (e.g. Lawrence and Crutzen, 1999; Eyring et al., 2007; Hoor et Only Dahlmann et al. (2011) reported results of the O_3 due to shipping emissions using a NO_x-only tagging approach.

~~To investigate the contribution(also called source attribution) the so-called tagging approachis much better suited. It has been shown that the contributions estimated by the tagging approach are larger~~

It is well known that the impact is usually smaller compared to the ~~impacts calculated using the perturbation approach (e.g. Grewe et al., 2012; Emmons et al., 2012; Grewe et al., 2017) . However, so far no study investigated the contribution of land transport~~ contribution (e.g. Grewe et al., 2012; Emmons et al., 2012; Grewe et al., 2017) . Furthermore, impacts are usually not additive. This means that the ozone changes (impacts) which are calculated for different emission sources by perturbing one of the emission source is not the same as perturbing all of the emission sources at the same time. This does not only hold for the ozone concentration but also for the associated ozone radiative forcing. As land traffic and shipping emissions ~~in detail using a tagging approach. In addition~~are important sources of ozone precursors, it is very important to calculate not only their impact on ozone, but also the contribution of these emissions to ozone in detail. Further, our approach tags for the first time not only NO_x and VOC individually, but both ozone precursors concurrently (Grewe et al., 2017).

~~The~~ Therefore, the goal of the present study is twofold: ~~First~~ first we review estimates of the contribution ~~/~~and impact of land transport ~~/road traffic and ship traffic~~ and shipping emissions on tropospheric ozone and the resulting radiative forcing. Second, we present new results ~~using a detailed tagging method, which has so far not been used to investigate~~ analysing the contribution of land transport and shipping emissions ~~. This includes a comparison of the tagging and the perturbation method. Furtherwe provide detailed information about the influence of the emissions from~~ in detail using a tagging approach. These new results quantify for the first time the contributions of the considered emissions on (ground-level) ozone in detail. Further, we also report results using a perturbation approach in a consistent manner to bridge the gap between previous studies and our new results. This allows a detailed comparison of the impact and contribution, as well as the associated ozone radiative forcings of land transport and shipping ~~on the tropospheric ozone budget~~emissions between the perturbation approach, NO_x-tagging and NO_x-&VOC tagging.

The paper is organised as follows: In Sect. 2 we give an overview of the used model system and describe the applied set-up. In Sect. 3 we analyse ~~the~~ our simulation results with respect to ~~the contribution~~ contribution versus impact of land transport and shipping emissions to ground level ozone including a detailed overview and discussion of the results from previous studies. In Sect. 4 we compare our results using the perturbation and the tagging approach in more detail. Section 5 gives more detailed insights into the tropospheric ozone budget. ~~Finally, Sect. 6 analyses the~~ The contribution of the land transport and shipping emissions to radiative forcing due to ozone ~~.~~ is analysed in Sect. 6, while Sect. 7 gives a discussion about the uncertainties associated with the tagging and the perturbation approaches, respectively.

2 Model description and set-up

2.1 Model description

We applied the ECHAM/MESSy Atmospheric Chemistry (EMAC) chemistry-climate model (Jöckel et al., 2006, 2010, 2016) equipped with the TAGGING technique described by Grewe et al. (2017). EMAC uses the second version of the Modular Earth Submodel System (MESSy2) to link multi-institutional computer codes. The core atmospheric model is the 5th generation European Centre Hamburg general circulation model (ECHAM5 Roeckner et al., 2006). For the present study we applied EMAC (ECHAM5 version 5.3.02, MESSy version 2.52) in the T42L90MA-resolution, i.e. with a spherical truncation of T42

(corresponding to a quadratic Gaussian grid of approx. 2.8 by 2.8 degrees in latitude and longitude) with 90 vertical hybrid pressure levels up to 0.01 hPa. The simulation set-up is almost identical to the one of the simulation *RC1SD-base-10a* described in detail by Jöckel et al. (2016) alongside with an evaluation of the resulting model simulation. Therefore, we describe only the most important details and differences. A comparison with the results of the simulation presented here and the *RC1SD-base-10a* is part of the Supplement of the present manuscript.

The chosen simulation period covers the years 2004 to 2010. The years 2004–2005 serve as spin-up, while the years 2006–2010 are analysed. Initial conditions for the trace gas distribution were taken from the *RC1SD-base-10a* simulation (Jöckel et al., 2016). Lightning NO_x is parameterised after Grewe et al. (2002) with global total emissions of ≈ 4.5 $Tg(N)$ a^{-1}. Emissions of NO_x from soil and biogenic C_5H_8 emissions were calculated using the MESSy submodel ONEMIS (Kerkweg et al., 2006), using parameterisations based on Yienger and Levy (1995) for soil-NO_x and Guenther et al. (1995) for biogenic C_5H_8. The applied gas phase mechanism in MECCA (Sander et al., 2011) incorporates the chemistry of ozone, methane and odd nitrogen. Alkanes and ~~Alkenes~~ alkenes are considered up to C4, while the oxidation of C_5H_8 and some non-methane hydrocarbons (NMHCs) are described with the Mainz Isopren Mechanism version 1 (von Kuhlmann et al., 2004). Further, heterogeneous reactions in the stratosphere (submodel MSBM, Jöckel et al., 2010) as well as aqueous phase chemistry and scavanging (SCAV, Tost et al., 2006) are included. Emissions of methane (CH_4) are not considered explicitly. Instead pseudo-emissions are calculated using the submodel TNUDGE (Kerkweg et al., 2006) . TNUDGE relaxes mixing ratios in the lowest model layer towards observations using Newtonian relaxation (see also Jöckel et al., 2016) .

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. Also the ~~the~~ sea surface temperature and sea ice coverage are prescribed as transient time-series from ERA-Interim too. To allow for identical meteorological conditions in sensitivity experiments with changed emissions, the quasi chemistry transport model mode (QCTM-mode, Deckert et al., 2011) of EMAC was used. In this mode, climatologies of the radiative active trace gases are prescribed for the calculation of the radiation. Further, climatologies are used for processes which couple the chemistry and the hydrological cycle. The applied climatologies are monthly average values taken from the *RC1SD-base-10a* simulation.

2.2 Tagging method for source attribution

The tagging is performed using the MESSy TAGGING submodul described in detail by Grewe et al. (2017). This tagging method is an accounting system following the relevant reaction pathways and applies the generalized tagging method introduced by Grewe (2013). This method diagnoses the contributions of different categories to the regarded species without influencing the full chemistry. A prerequisite for this method is a complete decomposition of the source terms, e.g. emissions, of the regarded species in N unique categories. As a consequence of the complete decomposition, the sum of the contributions of all tagged categories of one specie equals the total concentration of this specie (i.e. the budget is closed):

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

As an example of this method consider the production of O_3 by the reaction of NO with an organic peroxy radical (RO_2) to NO_2 and the organic oxy radical (RO):

$$NO + RO_2 \longrightarrow NO_2 + RO. \quad \text{(R1)}$$

For this reaction the tagging approach leads to the following fractional apportionment (c.f. Eq. 13 and 14 in Grewe et al., 2017, for a detailed example):

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

In this case the variables marked with ^{tag} represent the tagged production rate of O_3 by reaction R1 (P_{R1}) as well as the tagged families of NO_y and NMHC (details given below) of one individual category (e.g. land transport). Accordingly the fractional apportionment is inherent to the method based on a combinatorial approach, which decomposes every regarded reaction into all possible combinations of reacting tagged species. This takes into account the specific reaction rate constant from the full chemistry scheme (implicitly by the production and loss rates from the chemistry solver). The chemical mechanism including all diagnosed production and loss rates for the tagging method are part of the Supplement. The analysed production and loss rates in Sect. 5 are calculated in accordance with Eq. 13 and 14 of Grewe et al. (2017) .

The applied method considers ten categories (detailed definition is given in Table 1). To minimize the needed amount of memory and computational performance, not every individual specie is tagged. Instead a family concept is chosen. The following families are taking into account: O_3, NO_y, PAN, NMHC and CO. Additionally, OH and HO_2 are tagged by a steady state approach. In the following, we denote absolute contributions of land transport and shipping emissions to ozone diagnosed with the tagging method as O_3^{tra} and O_3^{shp}, respectively.

~~As anthropogenic emissions~~

2.3 Radiative forcing

The radiative forcing (RF) of ozone is defined as the difference of the net radiative fluxes caused by a change (e.g. between two time periods Here, we are interested in the contribution of land transport and shipping to this RF. Due to the non-linearities in the ozone chemistry (see also Sect. 4), we estimate the contribution of the land transport/shipping emissions to ozone and then calculate the RF of these O_3 shares individually. This approach is consistent with the IPCC RF definition, since the sum of all individual RF contributions approximately equals the total RF (for a detailed example see Dahlmann et al., 2011) .

Thus, to calculate the O_3 RFs of land traffic and shipping emissions, additional simulations were performed applying the stratospheric adjusted radiative forcing concept (e.g. Hansen et al., 1997; Stuber et al., 2001; Dietmüller et al., 2016) . For this, monthly mean fields of the simulation *RC1SD-base-10a* are used as input data, of the radiation scheme, except for O_3, which stem from the *BASE* simulation. Calculations of the RF based on the results of the tagging approach in accordance with Dahlmann et al. (2011) were performed as follows:

1. Based on the results of the *BASE* simulation, monthly mean values of Δ_T^{tra}=$\mathrm{O_3}$ - $\mathrm{O_3^{tra}}$ and Δ_T^{shp}=$\mathrm{O_3}$ - $\mathrm{O_3^{shp}}$ were calculated. Δ_T^{tra} and Δ_T^{shp} corresponds to the share of $\mathrm{O_3}$ excluding $\mathrm{O_3}$ from land transport and shipping emissions, respectively.

2. Multiple radiation calculations (Dietmüller et al., 2016) were performed, calculating the radiative flux of Δ_T^{tra}, Δ_T^{shp} and $\mathrm{O_3}$. The $\mathrm{O_3}$ RFs of land transport and shipping emissions using the tagging approach are then calculated as follows:

$$\mathrm{RF}_{\mathrm{O3tra}}^{\mathrm{tagging}} = rflux(\mathrm{O_3}) - rflux(\Delta_T^{\mathrm{tra}}), \tag{3}$$

$$\mathrm{RF}_{\mathrm{O3shp}}^{\mathrm{tagging}} = rflux(\mathrm{O_3}) - rflux(\Delta_T^{\mathrm{shp}}), \tag{4}$$

with $rflux$ being the net radiative fluxes calculated for the respective quantity. Accordingly, the calculated RFs measure the flux change caused by the ozone share of land transport and shipping emissions, respectively.

Calculating the RFs based on the results of the perturbation approach is similar to (e.g. Myhre et al., 2011) . First, $\Delta\mathrm{O_{3tra}}$ and $\Delta\mathrm{O_{3shp}}$ are calculated by taking the difference between the unperturbed (*BASE*, see below) and the perturbed simulations (*LTRA95* or *SHIP95*):

$$\Delta\mathrm{O_3} = (\mathrm{O_3}^{\mathrm{unperturbed}} - \mathrm{O_3}^{\mathrm{perturbed}}) \cdot 20. \tag{5}$$

As we consider 5 % perturbations (e.g. the emissions of land transport and shipping are decreased by 5 %, see Sect. 2.4) these differences are scaled by a factor of 20 to yield a 100 % perturbation. To calculate the RFs using the perturbation approach, $\Delta\mathrm{O_{3tra}}$ and $\Delta\mathrm{O_{3shp}}$ are than treated as described above for Δ_T^{tra} and Δ_T^{shp}. These RFs are called $\mathrm{RF}_{\Delta\mathrm{O3tra}}^{\mathrm{perturbation}}$ and $\mathrm{RF}_{\Delta\mathrm{O3shp}}^{\mathrm{perturbation}}$, respectively. Accordingly, the method to calculate the RFs of the $\mathrm{O_3}$ shares analysed by the perturbation and the tagging approach are the same. The differences between $\mathrm{RF}_{\mathrm{O3tra}}^{\mathrm{perturbation}}$ and $\mathrm{RF}_{\mathrm{O3tra}}^{\mathrm{tagging}}$ (and the same for shipping) arise only due to differences of the the differently calculated $\mathrm{O_3}$ shares.

The benefit of using the contribution of an emission source (in contrast to using the impact of the emission source) is that for the contribution the sum of the individual radiative forcings is equal to the total RF, i.e. $\sum_i^n RF^i \approx RF$ with RF^i being the radiative forcings of the individual categories i of n total categories. This hold for the perturbation approach (Dahlmann et al., 2011; Grewe et al., 2012) . However, the calculations of the RF is still subject to some specific assumptions, which we discuss in detail in the Supplement.

In general, we consider only the direct RF due to changes of the $\mathrm{O_3}$ concentration. We calculate no RF due to changes of the methane concentration caused by anthropogenic emissions. These changes would lead to a negative RF due to decreased methane concentrations. Especially for shipping emissions the negative RF due to methane can be larger compared to the positive ozone forcing (e.g. Myhre et al., 2011) .

2.4 Simulation set-up

As anthropogenic emissions inventory we chose the MACCity emission inventory (Granier et al., 2011), which follows the RCP8.5 scenario (Riahi et al., 2007, 2011) for the analysed period. The monthly varying anthropogenic emissions are represented on a grid with 0.5° x 0.5° spatial resolution. The geographical distribution of the land transport (containing road traffic, inland navigation and railways) and the shipping sector are shown in Fig. 1. Additionally, the total emissions of CO, NO_x and NMHCs of the most important emission sectors are given in Table 2.

~~We conducted three different simulations~~ Three different simulations were conducted: one with all emissions (*BASE*), one with a 5 % decrease of the land transport emissions of NO_x, CO and VOCs (*LTRA95*), and one with a 5 % decrease of the shipping emissions of NO_x, CO and VOCs (*SHIP95*). The 5 % perturbation was chosen as previous studies showed that this small perturbation sufficiently minimises the impact of the non-linearity of the chemistry on the results (e.g. Hoor et al., 2009; Grewe et al., 2010; Koffi et al., 2010) .

All three simulations were equipped with the full tagging diagnostics. To quantify the contribution of the emission sources the tagging results of the *BASE* simulation are used. The simulations with a decrease of the land transport and shipping emissions were performed to allow for a direct comparison between the tagging and the perturbation method. The additional tagging diagnostics in the perturbed simulations allow for a more detailed investigations in the change of the ozone production (see Sect. 4).

In the present study we focus on the source regions of land transport and shipping emissions. Therefore we use the same geographical regions as defined by Righi et al. (2013) to investigate the contribution these emissions. The regions are Europe (EU), North America (NA) and Southeast Asia (SEA) for land transport, and North Atlantic Ocean (NAO), Indian Ocean (IO) and North Pacific Ocean (NPO) for the shipping emissions.

3 Contribution to ground level ozone

First, we analyse the absolute amount of O_3 produced by land transport (tra) and ship (shp) exhaust as analysed with the ~~Tagging method. We denote absolute contributions diagnosed with the tagging method as and , respectively~~tagging approach. Additionally we indicate also the relative contribution of O_3^{tra} and O_3^{shp} to near ground level O_3. For all quantities multi-annual, seasonal average values for ~~December–Februar~~ December–February (DJF) as well as June–August (JJA) for the years 2006–2010 (for DJF starting with December 2005) were computed.

3.1 Land transport

Figure 2a and Fig. 2b show the seasonal average values of O_3^{tra} for DJF and JJA . The maximum absolute contribution for each hemisphere are simulated during local summer conditions when the photochemistry is most effective. Most geographical locations of these maxima correspond to the regions with the largest land transport emissions. The largest absolute contributions of 8–14 $nmol\ mol^{-1}$ are simulated during JJA on the Northern Hemisphere in North America (8–12 $nmol\ mol^{-1}$), Southern

Europe (8–10 $\mathrm{nmol\ mol^{-1}}$), the Arabian Pensinsula (12–14 $\mathrm{nmol\ mol^{-1}}$), India (8–10 $\mathrm{nmol\ mol^{-1}}$) and Southeast Asia (6–10 $\mathrm{nmol\ mol^{-1}}$). In Asia the largest values are simulated around the Korean Peninsula rather than in China. This lower contribution of land transport emissions in China compared to Europe or North America is mainly caused by a much larger fraction of other anthropogenic emissions (e.g. industry and households) compared to land transport emissions (e.g. Righi et al., 2013). Accordingly much more O_3 is produced in China by other anthropogenic emissions compared to land transport. The local maxima (4–6 $\mathrm{nmol\ mol^{-1}}$) on the Southern Hemisphere are simulated during DJF, when the photochemistry is most active. These maxima are located in South America and South Africa. Corresponding the regions with the largest land transport emissions on the Southern hemisphere (cf. Fig. 1).

The relative contribution of O_3^{tra} to near ground level O_3 is depicted in Fig. 2c and Fig. 2d. Values of 14–16 % are simulated during DJF around the source regions on the Southern Hemisphere, but the absolute values on the Southern Hemisphere are lower compared to the Northern Hemisphere. The simulated relative contributions on the Northern Hemisphere during DJF is around 10 %. Only around the Arabian Peninsula values of 14–16 % are found. During JJA, these maxima increase to 14–18 % over North America and 12–16 % for the other hotspot regions on the Northern Hemisphere. One important reason for the change of the contribution from DJF to JJA (on the Northern Hemisphere) is the strong seasonal cycle of the anthropogenic non-traffic sector in our applied emission inventory, showing large emissions during winter and lower emissions during summer. This leads to larger contributions of the anthropogenic non-traffic category during DJF compared to JJA.

To review estimates of the impact ~~/~~and contribution of previous studies and to compare the new results with previous values, Table 3 summarises the amount of emissions as well as reported impacts/contributions of road traffic emissions from previous studies. So far, only the effects of road traffic emissions alone and not the total effect of land transport emissions have been investigated. With respect to the ozone precursors road traffic emissions are the largest contributor to the land transport sector. The contributions of inland navigation and railways are smaller than the uncertainties of the road traffic emissions. Therefore we argue that our results of the land transport sector can be compared with previous studies considering only road traffic emissions (cf. also the amount of applied emissions in different studies in Table 3). In general, we are focussing on global studies only. Regional effects of road traffic emissions have been investigated too (e.g. Reis et al., 2000; Tagaris et al., 2015; Hendricks et al., 2017), but because of the coarse resolution of global models a quantitative comparison between findings of regional studies with these global studies is not straightforward and probably not meaningful. Please note that we ~~provide~~ list our values in Table 3 ~~the values of the present study only for July to allow for a better comparability~~for July conditions only, to be comparable to other studies, since they also reported values for July conditions. In addition the impact of the land transport emissions were calculated by with the results of the unperturbed and perturbed simulation (*BASE* minus *LTRA95*) which is scaled by 20 to estimate a 100 % perturbation. Figures showing the contribution/impact for the results of the present study are part of the Supplement.

Previously, the impact of road traffic emissions on ozone concentration has been investigated mainly using 100 % and 5 % perturbation approaches. Most previous studies applied similar amounts of road traffic emissions as the present study used for land transport emissions (9–10 $\mathrm{Tg\ a^{-1}}$). The fraction of NO_x emissions from road traffic compared to all emissions was largest

in the studies of Granier and Brasseur (2003), Niemeier et al. (2006) and Matthes et al. (2007). These studies also applied the largest CO and VOC emissions, while the individual fractions vary across the studies.

In general, the results of all considered studies can be separated into three groups: (1) The largest values are reported by the present study (using the tagging ~~method~~approach) as well as by Niemeier et al. (2006). (2) Slightly lower values are given by Granier and Brasseur (2003) and Matthes et al. (2007), while (3) Hoor et al. (2009) and Koffi et al. (2010) report the lowest impact. These studies, however, differ not only in the emission inventories and models used, but also in the methods. The lowest values are in general reported by studies using the 5 % perturbation (scaled to 100 %), which is confirmed by our results using the same method. However, ~~compared to other 5 studies our results show, especially for NA, slightly larger values. This~~ in general our simulation results show larger values compared to these previous findings. These differences are noticeable especially for the NA region. The differences might be caused by a different geographical distribution ~~and larger and emissions in our applied emission inventory~~ of the emissions, as well by larger CO and NMHC emissions in the emission inventory we applied. Further, differences in the atmospheric composition as simulated by the different models can influence the production rates of ozone, which might contribute to the differences of the simulated impacts.

The comparison of our results using the 5 % perturbation approach ~~and~~ with the results using the tagging approach clearly confirms the known ~~underestimation of the contribution by the perturbationapproach (e.g. Wu et al., 2009; Grewe et al., 2010; Emmons et a~~ between estimates of the impact (perturbation) and contribution (tagging, e.g. Wang et al., 2009; Grewe et al., 2010; Emmons et al., 2012; C Depending on the region, we find a difference of up to a factor of 4. The reason for this difference is investigated in more detail in Sect. 4.

Granier and Brasseur (2003), Niemeier et al. (2006) and Matthes et al. (2007), however, also used a perturbation approach, but report values, which are more similar to our estimate using the tagging method. This is likely caused by the larger emissions applied in these studies compared to all other studies. Accordingly, the contribution of the road traffic emissions is underestimated by the perturbation method, but the larger emissions (and fraction) of the road traffic category lead to results, which are similar as estimated by the tagging method with smaller emissions. Of course also other factors, like differences between the models, chemical mechanisms, geographical distribution, and different seasonal cycles of the emissions can contribute to differences between the studies. The influence of these factors, however, is difficult to reveal.

3.2 Ship traffic

The absolute contribution of O_3^{shp} are shown in Fig. 3a and Fig. 3b. Similar to the shipping emissions (cf. Fig. 1), O_3^{shp} shows a strong North-South gradient. The maximum values in the Northern Hemisphere are located between 20°–30° N during DJF ($\approx$ 6 nmol mol^{-1}). These maxima move northwards during summer and increase in magnitude (10–12 nmol mol^{-1}). This shift is caused by the increase in the photochemical activity in the Northern hemisphere during summer. Most shipping emissions are located north of 30° N (see Fig. 1). With increasing ozone production during spring and summer more O_3^{shp} near the regions with the largest emissions are formed, compared to the regions of 20–30° N.

The largest values of the relative contribution of O_3^{shp} during DJF are around 14 % and are co-located with the regions of the largest values of O_3^{shp} (Fig. 3c). The maxima of the contribution increase during JJA to around 30 % in the Northwestern

Pacific, while the values in the Northeastern Pacific are around 18–22 %. In the Northern Atlantic maximum contributions of 20 % are simulated (Fig. 3d).

Table 4 summarises emissions and results of previous studies. In general most studies used similar global NO_x shipping emissions of around 4 $Tg(N)\ a^{-1}$. The largest impact/contribution of shipping emissions is limited to distinct areas within the investigated geographical regions. Therefore the range of the given contributions/impacts within the geographical regions is large. The displacement between the regions of emissions and largest ozone production is well known (e.g. Endresen et al., 2003; Eyring et al., 2007) and mainly caused by complex interplay between NO_x emissions, transport of precursors and ozone production.

Similar as discussed for the impact/contribution of land transport emissions, there is a large discrepancy between the results using the 100 % and the 5 % perturbation method. The studies using the 100 % method report impacts in the Atlantic and the Pacific in the range of 4–11 $nmol\ mol^{-1}$ (corresponding to 12–40 %). In general the previous studies report larger impacts in the Pacific compared to the Atlantic. Only Eyring et al. (2007) reported a larger perturbation in the Northern Atlantic compared to the Pacific, which can most likely be attributed to differences in the emission inventories, as Eyring et al. (2007) applied lower emissions in the Northern Pacific compared to the Northern Atlantic.

Hoor et al. (2009) and Koffi et al. (2010) report absolute impacts (5 % perturbation) in the range of 2–6 $nmol\ mol^{-1}$. Our model results using a 5 % perturbation suggest somewhat larger impacts of around 2–8 $nmol\ mol^{-1}$ (10–22 %) in the Atlantic and Pacific. Most likely this difference can be attributed to different shipping emissions applied.

The absolute contributions diagnosed using the tagging approach are larger and in the range of 3–11 $nmol\ mol^{-1}$ (relative contribution: 10–33 %) in the Atlantic and Pacific. These contributions are at the lower end of the contributions reported by the studies using the 100 % approach. Compared to these studies, however, we applied the largest shipping emissions. Accordingly, a larger contribution compared to other studies can be expected. As the used models and emission inventories in all studies are very different we can only speculate about possible reasons.

One reason for this discrepancy might be the resolution of the model simulations. In previous studies a variety of resolutions were used (especially in the multi model approaches by Eyring et al. (2007) and Hoor et al. (2009). Our horizontal resolution of $\approx 2.8°$ is at the finer end of most of these resolutions (only Dalsøren et al. (2009) used $\approx 1.875°$). A coarse resolution leads to a strong dilution of the shipping emissions. This effect can lead to an overestimation of the O_3 production (e.g. Wild and Prather, 2006). Our results are also influenced by this problem too, because a resolution of T42 dilutes the emissions over large areas. A model with finer resolution, effective emissions, or a plume model (e.g. Franke et al., 2008; Holmes et al., 2014) diagnoses likely smaller contributions. Another important contributor to the differences is the geographical distribution of ship emissions. If the ship tracks are too narrow, the ozone production might be suppressed (see discussion by Eyring et al., 2007). Further, differences in the seasonal cycles of emissions con contribute to the differences.

4 Comparing perturbation and tagging approach

As discussed in the previous section and by previous studies ~~(e.g. Wu et al., 2009; Grewe et al., 2010)~~ (e.g. Wang et al., 2009; Grewe et al., perturbation approach, which is often used for source attribution, and the tagging approach lead to different results.

To investigate this effect in more detail, ~~the differences between the results of the *BASE* and *LTRA95* simulations with respect to are calculated~~ $\Delta O_{3\mathrm{tra}}$ (see Eq. 5) is analysed further. Here, we consider not only ground-level values, but partial ozone columns integrated from the surface up to 850 hPa (called 850PC, in DU).

$$\Delta O_3 = {O_3}^{\mathrm{unperturbed}} - {O_3}^{\mathrm{perturbed}},$$

~~where ${O_3}^{\mathrm{perturbed}}$ and ${O_3}^{\mathrm{unperturbed}}$ are the tropospheric columns from the simulation with 5 reduced emissions and the original simulation, respectively.Similar $\Delta O_3^{\mathrm{tra}}$, the difference of the tagged ozone due land transport emissions, is calculated. In a next step we calculate the ratios $\Delta O_3^{\mathrm{tra}}/\Delta O_3$. This ratio indicates by how much the results using the tagging approachdiffer from the results of the~~ To quantify the difference between the perturbation and the tagging approach in more detail, Fig. 4a shows the 850PC of $\Delta O_{3\mathrm{tra}}$. Figure 4b shows the 850PC of (O_3^{tra}) for the *BASE* simulation. A qualitative comparison already indicates a relative large difference between the impact (as estimated by the perturbation approach~~.~~

~~The corresponding values of the partial columns up to 850 are shown in Fig.~~ , Fig. 4a) and the contribution (by the tagging approach, Fig. ~~??. In general the ratio is largest in the Northern Hemisphere, where most land transport sources are located. In most regions~~4b). Figure 4c shows the relative difference between both quantities, indicating a difference between 40–80 %. The lowest differences are found on the Southern Hemisphere, while the difference is largest near the hotspot regions (North America, Europe and South-East Asia). Here, the impact is up to a factor of four lower compared to the contribution (not shown). A large relative difference is also indicated in some regions near the equator. In these regions, however, the absolute difference is low. The only region where a difference below 20 % is simulated is in parts of South America. This difference between the impact and the contribution is not confined to the lower troposphere, but is present throughout the troposphere (additional figures showing zonal averaged impact and contributions are part of the ~~ratio is around 1.5–2. Over South Americathe ratio is one, so the tagging and the perturbation method show the same results, but in this region land transport emissions are rather small. Especially near the hotspot regions (Europe, Southeast Asia) ratios of up to 4 are simulated. Accordingly, the perturbation approach largely underestimates the contribution of land transport emissions to ozone in the regions of large land transport emissions~~Supplement).

To ~~understand the reason for the different ratios in different regionsin more detail~~further investigate why the difference between impact and contribution largely change between the regions, the dependency between NO_x ~~emissions~~ mixing ratios (caused by changes of the emissions) and the net O_3 production of the results for the year 2010 is analysed. Figure. 5 ~~a~~ shows this dependency for the whole globe (black) and some chosen areas (coloured dots). Generally the ~~the~~ well known dependency (e.g. Seinfeld and Pandis, 2006) between O_3 production and NO_x concentrations can be observed. In pristine regions a net loss of O_3 is, present (first regime). With increasing NO_x ~~emissions~~ mixing ratios the net O_3 production increases strongly ~~. This second regime is usually~~ (called NO_x ~~limited~~-limited regime). The production of O_3 ~~decrease~~ decreases again with even

larger NO_x values. In this third regime, however, the production of O_3 can be increased if the NMHC emissions are increased (called NMHC-limited regime). Every dot represents a different grid box of the model with different meteorological conditions and background mixing ratios of CO, NMHC etc. Therefore, the dependency between the NO_x mixing ratio and the net O_3 production differs for every grid box and is not given by one single function (which is the case for boxmodel calculations with prescribed conditions).

In different regions of the world the O_3 production takes place in different chemical regimes, depending on the amount of NO_x emissions. Therefore, the coloured dots highlight the individual relationship between NO_x mixing ratio and production of O_3 for four different regions.

Depending on the ~~regime of the production and the strength of the perturbation in the individual~~ chemical regime in the different regions, the ~~production responds differently on emissions in the different regions (e.g., Dahlmann et al., 2011) . To illustrate this in more detail, the dots in Fig. 5b show the average dependency between~~ ozone chemistry responds differently to the perturbation applied in the perturbation approach (e.g., Dahlmann et al., 2011) .

Based on the results of the *REF* and *LTRA95* simulations, the ozone sensitivity is calculated with the tangent approach in accordance with Grewe et al. (2010) by solving a linear equation ($y = m \cdot (x - x_0) + b$, see Supplement for additional Figures). Here, x and y are the average NO_x mixing ratio and the net O_3 production ~~for the different geographical regions. Additionally, we calculated the estimated derivative of the ozone sensitivity based on the perturbed and the unperturbed simulation (see Grewe et al., 2010, for a in depth discussion with idealised examples as well as Fig.5c) .~~ (P_{O3}), respectively, for a particular region. The m denotes the slope, which corresponds to an approximation of the derivative dP_{O3}/dNO_x in the unperturbed simulation, which is calculated by the difference in ozone production and NO_x mixing ratios in the unperturbed and perturbed simulation. x_0=NO_x^u is the NO_x mean mixing ratio in the unperturbed simulation and b = P_{O3}^u-dP_{O3}/dNO_x NO_x^u, where P_{O3}^u is the mean ozone production in the unperturbed simulation.

Based on the ~~estimated derivatives~~ linearised ozone production (P_{O3}^{lin}) calculated by the tangent approach, we define a saturation indicator Γ~~can be calculated, which is defined as:~~

$$\Gamma = \frac{\text{y} - \text{axis intercept}}{\text{y} - \text{value of unperturbed simulation}},$$

, which helps to analyse the ozone sensitivity further:

~~with y being the net production rate (cf. Fig. 5).~~

$$\Gamma = \frac{\text{y} - \text{axis intercept}}{\text{y} - \text{value of unperturbed simulation}} = \frac{P_{O3}^{lin}(NO_x = 0)}{P_{O3}^{lin}(NO_x = \text{unperturbed})}. \quad (6)$$

Accordingly, Γ compares the production rate of ozone of the base case with unperturbed emissions ($NO_x = \text{unperturbed}$) with the approximated production rate of ozone, if NO_x emissions are set to zero ($NO_x = 0$), assuming a linear ozone chemistry. This value is a quantitative ~~measure of how the ozone production in the different region responds to a change of the emission strength. $\Gamma = 0$ indicates a linear response of the system (with an y-intercept at zero)~~indicator of the chemical regime, showing how much an emission change of one specific sector is compensated by increased ozone productivity of other

sectors. $\Gamma = 1$ indicates a saturated behaviour of the ozone production i.e. the ozone production does not change, if emissions are changed ($P^{lin}_{O3}(NO_x = 0) = P^{lin}_{O3}(NO_x = unperturbed)$). Accordingly, there is no ozone reduction because the change of the emissions is entirely compensated by an increasing ozone production efficiency of other emissions. ~~This point corresponds to the threshold between the - and - limited regime.~~ $\Gamma > 1$ indicates an ~~overcompensation~~ overcompensating effect, i.e., reduced NO_x emissions lead to an increase of the ozone production (corresponding to the VOC-limited regime).

~~Accordingly (see Fig. 5b)~~Finally, $\Gamma = 0$ indicates a linear response of the system (with a y-intercept at zero). Accordingly, the ozone change introduced by an emission change is not compensated by an increase of the ozone production efficiency. For $\Gamma = 0.5$ the ozone change is half compensated by a change in the ozone production efficiency. In terms of the estimated derivative (dP_{O3}/dNO_x), ~~the response of the net ozone production on the emissions perturbation in North Africa ($\Gamma = 0.3$)and South America ($\Gamma = 0.4$)is almost linear. In South-East-Asia ($\Gamma = 0.6$) the ozone production response is between the linear and saturated behaviour, while over Europe the ozone production is almost saturated ($\Gamma = 0.9$).~~ $\Gamma = 1$ corresponds to $dP_{O3}/dNO_x = 0$, while $\Gamma > 1$ corresponds to $dP_{O3}/dNO_x < 0$ and vice versa.

Table 5 lists the Γ values of the four different regions together with a brief interpretation of these values (additional information and figures concerning Γ are part of the Supplement). In general, only the regions North Africa and South America show a response of the O_3 chemistry, which is close to linear ($\Gamma = 0.2 - 0.3$). As known (e.g. Wang et al., 2009; Grewe et al., 2010; Clappie for this linear case the perturbation and the tagging approach lead to the same results (e.g. the contribution can be estimated using a perturbation approach). In all other regions the contribution is largely underestimated by the perturbation approach.

~~This shows that a reduction of land transport emissions in Europe would only slightly alter the ozone budget, because the efficiency of ozone production from other emission sources increases, if land transport emissions are decreased (Grewe et al., 2012) .~~

~~This example clearly shows~~ This underlines the importance of ~~the discrimination of the tagging and the perturbationapproach. Clearly~~discriminating between tagging and perturbation. Clearly, both approaches answer different, but equally important questions. The perturbation approach answers the question on the impact of an emission ~~reduction~~change. This approach is important to estimate effects due to mitigation measures (e.g. Williams et al., 2014). The tagging approach in contrast, disentangles the ozone budget into the contributions of the individual emission ~~sectors~~ sources and is important to investigate e.g. the contribution of radiative forcing of individual emission sources ~~. However, even~~ (see Sect. 6) or to quantify contribution of different emission sources to extreme ozone events. However, the tagging approach can not be used to quantify the impact of an emission change, while the perturbation approach should not be used to quantify the contribution. As demonstrated, in regions where ozone responses more linearly to emission changes, both approaches differ slightly, but in regions where large emissions occur (e.g., Europe, South-East Asia) the perturbation approach largely underestimates the contributions and should not be used for source apportionment. However, if mitigation options are investigated the ~~Tagging~~ tagging approach should be combined with the perturbation approach (see next subsection).

4.1 Combining Tagging and Perturbation approach in mitigation studies

The ~~response of the atmospheric composition on~~ tagging approach does not give any information about the sensitivity of the ozone chemistry with respect to a change of emissions~~can not be obtained from only one simulation using the tagging approach. This requires an additional simulation with changed emissions (perturbation approach), but the perturbed and the unperturbed simulation should be both equipped with a tagging diagnostic.~~ . Accordingly, the success of an emission reduction, e.g. measured in terms of reduced ozone concentration, is evaluated using the perturbation approach. Wang et al. (2009) proposed to first use a tagging simulation estimating these sources, which contribute largest to ozone and therefore have the largest mitigation potential. However, we propose to equip all simulations, the unperturbed reference simulation and all simulations with changed emissions, with the tagging approach.

In this case ~~, the tagging diagnostics allows to quantify the contribution of the emissions from the sector of interest. Usually,~~ the ~~reduction of~~ results of the perturbed simulations quantify the changes in ozone due to mitigation options. The tagging results provide additional information, which are important to quantify the accountability of different emission sources to the ozone concentration or the associated radiative forcing. These additional information are important, because the ~~contribution is much larger than the reduction of ozone itself, because the efficiency of the ozone productivity from the other sectors can be altered, even if the emissions themselves are unchanged. Combining the tagging and the perturbation approach is therefore the best way to measure the success of a mitigation strategy. If only the perturbation approach is used to evaluate a mitigation strategy, the~~ success of one specific mitigation option largely depends on the history of previous mitigations (Grewe et al., 2012). ~~This problem is sketched in~~

To present the benefits of combining both methods in more detail, Fig. 6 sketches an idalised example of four different mitigation options. For each of the idealised mitigation options we assume a decrease of the emissions of one specific emission source by 10 arbitrary units. Mitigation option 1 reduces the land transport emissions, mitigation option 2 the shipping emissions and mitigation option 3 the emissions from industry. ~~If the resulting ozone concentration is considered~~

With respect to the ozone concentration (Fig. 6a) only mitigation option 3 ~~seems to be successful~~ is successful in largely reducing the ozone concentration. Having only the results with respect to the ozone concentration in mind one could attribute the ozone change completely to the emissions change of the industry sector. From this point of view there would be no benefit to reduce land transport or shipping emissions. ~~Due to mitigation option 1, however, the contribution of land transport emissions~~

However, if all simulations are additionally equipped with a tagging method the contribution of the different emission sources to the ozone concentration ~~decreases, but the ozone production efficiency of all other emissions increases. Mitigation~~ is anlysed (Fig. 6b). For each of the considered cases both, the ozone concentration, and the contribution of the different emission sources to this ozone concentration differ. This additional contribution analysis shows that even if due to mitigation option 1 the overall ozone concentration increases, the contribution of the road traffic emissions is lowered. At the same time the contribution of all other emission sources, which are not changed, increase, because the ozone production efficiency increase. However, if every emission source is made responsible for their individual contributions to ozone levels (for air quality mitigation purpose) or their individual contributions to ozone radiative forcing (for climate mitigation purpose), an obvious benefit exists for a specific

emission source to reduce it's emissions even if overall O_3 levels are only slightly reduced. These additional information are only available using the tagging approach.

This gets even more clear, if mitigation option 2 ~~decreases the contribution of shipping emissions (which are reduced in this case), while the overall ozone concentration does not change.~~ is considered in which the shipping emissions are reduced. The overall ozone concentration remains unchanged, as the ozone chemistry is in a saturated regime ($\Gamma = 1$). The contribution of the shipping emissions, however, decrease strongly, while the contribution of emissions from industry and household increase. Accordingly, the emission sources household and industry are more responsible for the ozone values and/or ozone radiative forcing, while the emission sources road traffic and shipping are less responsible. This puts pressure onto these emission sources to reduce emissions of ozone precursors.

In mitigation option 3 the emissions of the industry sector are reduced. In this case, the response of the ozone concentration to emission changes is close to linear ($\Gamma \approx 0$) and the ozone concentration is reduced strongly. This emission reduction causes a reduction of the ozone production efficiency, leading not only to a reduction of the contribution of the industry emissions, but also to a further reduction of the contribution of all other sources.

The large effect of the ozone concentration for option 3 is only the effect of all previous mitigation options. In contrast, if the emissions from industry instead of the land transport emissions are reduced in mitigation option 1, this mitigation would almost have no effect on the ozone concentration. ~~This demonstrates the importance of combining perturbation and tagging to evaluate mitigation options.~~ Clearly, the effect of one specific mitigation option strongly depends on the history of previous mitigation options. A combination of tagging and perturbation is a powerful tool for putting additional pressure on unmitigated emission sources, because, even if the absolute ozone levels do not change, their shares in high ozone values (or radiative forcing) increase.

5 Analysis of the ozone budget

For more details about the influence of emissions of land transport and ship traffic on the ozone burden, we analysed the burden as well as production and loss rates of O_3, O_3^{tra} and O_3^{shp}, respectively. These analyses were performed globally, as well as for the distinct geographical regions defined in Sect. 2. Please note, in our tagging method we distinguish only between different emission sources, but not between emission regions. Therefore, the budgets analysed for distinct geographical regions might not be solely influenced by regional emissions, but also by upwind sources.

The global total tropospheric burden of O_3 averaged for 2006–2010 is 318 Tg, which is in the range of 337 ± 23 Tg presented by Young et al. (2013) as a results of a multi-model intercomparision, but please note that we used a fixed value of 200 hPa for the tropopause. Of these 318 Tg, globally 24 Tg are produced by land transport emissions, while 18 Tg are produced by emissions from shipping. The relative contribution of the burden of O_3^{tra} to the total ozone is thus around 8 % globally and 10 % in the regions Europe, North America and Southeastern Asia. The relative contribution of the burden of O_3^{shp} is around 6 % globally and 8 % near the important source regions. The difference between the rather large contribution of the shipping emissions near ground level (cf. Sect. 3) and the much smaller contribution for the whole troposphere is mainly caused by

the confinement of the contribution of shipping emissions to the lowermost troposphere (e.g. Eyring et al., 2007; Hoor et al., 2009).

To better understand the effect of land transport and shipping emissions on the atmospheric composition, we analysed the production and loss rates of O_3 from land transport and shipping emissions globally and for the individual regions, respectively. The corresponding numbers are shown in Figs. 7 and 8. Globally integrated production rates of 5274 $\mathrm{Tg\ a^{-1}}$ (averaged 2006–2010) are simulated, while the loss rate is 3972 $\mathrm{Tg\ a^{-1}}$, leading to a net production of O_3 of 1301 $\mathrm{Tg\ a^{-1}}$. Similar values of 5110 ± 606 $\mathrm{Tg\ a^{-1}}$ for the production are reported by Young et al. (2013). The values of the loss are lower than reported by Young et al. (2013), but still within the spread of the different models (4668 ± 727 $\mathrm{Tg\ a^{-1}}$, again note different definition of the tropopause). Further, it is important to note that loss rates are not calculated consistently in all models presented by Young et al. (2013) .

Globally a net production of 165 $\mathrm{Tg\ a^{-1}}$ from the land transport ~~category~~ emissions is simulated, corresponding to a contribution of 13 % to the total net O_3 production. The contribution of the land transport category to the total net O_3 production near the source regions is 19 % over Europe (24 $\mathrm{Tg\ a^{-1}}$), 21 % over North America (39 $\mathrm{Tg\ a^{-1}}$) and 17 % over Southeast Asia (51 $\mathrm{Tg\ a^{-1}}$).

A global net O_3 production of emissions from shipping of 129 $\mathrm{Tg\ a^{-1}}$ is simulated, corresponding to a contribution of 10 % to the total net O_3 production. Regionally, the importance of the shipping ~~category~~ emissions to the net O_3 production is much larger. Here contributions of 34 % over the Northern Atlantic (26 $\mathrm{Tg\ a^{-1}}$), 19 % over the Indian Ocean (17 $\mathrm{Tg\ a^{-1}}$) and 52 % over the Northern Pacific (36 $\mathrm{Tg\ a^{-1}}$) are simulated. The larger relative contributions near the source regions compared to the land transport category are mainly caused by less or almost no emissions of other sources in the shipping region. Especially over land, other important sources, such as anthropogenic non traffic and NO_x emissions from soil, decrease the relative importance of the land transport emissions. However, even near the source regions emissions of land transport contribute to around 20 % to the net O_3 production in these regions.

6 Radiative Forcing

~~We performed additional simulations to calculate the stratosphere adjusted radiative forcing (RF, e.g. Hansen et al., 1997; Stuber et al., 200 land transport and ship traffic contributions to . In these simulations only the dynamical processes and the radiation calculation are considered.~~

~~The monthly mean fields of the simulation *RC1SD-base-10a* are used as input data for the radiation scheme. To determine the contribution of the land transport and the ship emissions of the tagging results, the monthly means of 'O_3minus ' and 'minus ' were fed into additional radiative calculation calls (Dietmüller et al., 2016) . Finally, we calculated the stratospheric adjusted radiative forcing of and by subtracting the fluxes of 'minus ' (analogue for) from the fluxes of (see also Dahlmann et al., 2011) . The approach to calculate the RF by the results of the perturbation approach is similar to e.g. Myhre et al. (2011) . We first calculate Δbetween the unperturbed and the perturbed simulation and multiply this difference with a factor of 20. This scaled~~

~~difference is then treated exactly as and to calculate the RF. In general we consider only the direct RF due to changes of the concentration. We calculate no RF due to changes of the methane concentration caused by the anthropogenic emissions. These changes would lead to a negative RF due to decreased methane concentrations. Especially for shipping emissions the negative RF due to methane can be larger compared to the positive ozone forcing (e.g. Myhre et al., 2011) .~~

We obtain a global net RF for land transport of ~~92~~ $\mathrm{RF}^{\mathrm{tagging}}_{\mathrm{O3tra}} = 92\ \mathrm{mW\ m^{-2}}$. The shortwave RF is 32 $\mathrm{mW\ m^{-2}}$ and the longwave RF is ~~61 . The~~ 61 $\mathrm{mW\ m^{-2}}$. The estimated RF of ship traffic is ~~62~~ $\mathrm{RF}^{\mathrm{tagging}}_{\mathrm{O3shp}} = 62\ \mathrm{mW\ m^{-2}}$ and smaller than the land transport RF. The shortwave RF of ship emissions is 22 $\mathrm{mW\ m^{-2}}$ and the longwave is 40 $\mathrm{mW\ m^{-2}}$. To review estimates of the RF of land transport and shipping emissions and to compare our results with previous estimates, Table 8 compares our results with previous studies. As noted ~~above~~ in Sect. 2.3 only the RF of $\mathrm{O_3}$ is shown, RF of ~~changed~~ changes due to $\mathrm{CH_4}$ are not considered.

Most studies have estimated a lower RF of land transport/road traffic emissions of around 30 $\mathrm{mW\ m^{-2}}$~~. These studies use~~ , using the perturbation approach. ~~Only~~ The review of Uherek et al. (2010) gives a range for the RF due to road traffic emissions of $50 - (54 \pm 11)\ \mathrm{mW\ m^{-2}}$. Compared to these values Dahlmann et al. (2011) give larger estimates of around 170 $\mathrm{mW\ m^{-2}}$ ~~. They, however, used~~ using a $\mathrm{NO_x}$ only tagging approach and larger global land transport $\mathrm{NO_x}$ emissions of roughly 13 $\mathrm{Tg(N)\ a^{-1}}$~~and a tagging method which considers only~~. Comparing the RF per $\mathrm{Tg(N)\ a^{-1}}$ Dahlmann et al. (2011) reported values of around 14 $\mathrm{mW\ m^{-2}\ Tg^{-1}(N)\ a}$, while our estimates are around 10 $\mathrm{mW\ m^{-2}\ Tg^{-1}(N)\ a}$.

Also for the RF due to shipping emissions previous estimates using the perturbation ~~method~~ approach (around 20–30 $\mathrm{mW\ m^{-2}}$) are lower compared to our findings of around 60 $\mathrm{mW\ m^{-2}}$. Only the tagging study by Dahlmann et al. (2011) report values which are more similar to our estimates (49 $\mathrm{mW\ m^{-2}}$), but this study used lower ship emissions of around 4 $\mathrm{Tg(N)\ a^{-1}}$ while we applied roughly 6 $\mathrm{Tg(N)\ a^{-1}}$. Accordingly, our results suggest ~~an~~ a RF of 10 $\mathrm{mW\ m^{-2}\ Tg^{-1}(N)\ a}$, while Dahlmann et al. (2011) reported values of around 12 $\mathrm{mW\ m^{-2}\ Tg^{-1}(N)\ a}$. Obviously, the $\mathrm{NO_x}$ only tagging used by Dahlmann et al. (2011) leads in general to a larger RF per Tg(N) compared to our $\mathrm{NO_x}$– & VOC-tagging.

For a more detailed comparison we also calculated the RF due to land transport and shipping using the 5 % perturbation ~~method~~approach. By this ~~method we estimate a global net RF of around 24~~ approach we estimate $\mathrm{RF}^{\mathrm{perturbation}}_{\mathrm{\Delta O3tra}} = 24\ \mathrm{mW\ m^{-2}}$ (scaled to 100 %) for land transport emissions and ~~around 22~~ $\mathrm{RF}^{\mathrm{perturbation}}_{\mathrm{\Delta O3shp}} = 22\ \mathrm{mW\ m^{-2}}$ (scaled to 100 %) for shipping emissions. Both values are at the lower end of previous estimates of the RF using the perturbation approach. Remarkable, however, is the difference of a factor of three to four between our results using the perturbation and the tagging approach, despite identical model~~and emissions. Accordingly, especially for calculations of radiative forcings, it is very important to differentiate between the tagging and~~ , emissions, and a consistent calculation of the RF for the impact and the contribution of emissions.

These results have important implications with respect to current estimates of the RF due to land transport (and shipping) emissions. Previous best estimates of an RF of $50 - (54 \pm 11)\ \mathrm{mW\ m^{-2}}$ by Uherek et al. (2010) are too low, because these estimates are based on the perturbation approach~~and the different scientific questions they answer.~~ . Previous studies using a $\mathrm{NO_x}$-only tagging (Dahlmann et al., 2011; Grewe et al., 2012) reported larger values of up to 170 $\mathrm{mW\ m^{-2}}$, because the $\mathrm{NO_x}$-only tagging does not consider competing effects of $\mathrm{NO_x}$ and VOCs. Accordingly, our best estimate (92 $\mathrm{mW\ m^{-2}}$) of the RF due to land transport emissions lies between both previous estimates. Compared to this Uherek et al. (2010) gives an

estimate of 171 $\mathrm{mW\ m^{-2}}$ of the combined land transport $\mathrm{CO_2}$ forcing, while Righi et al. (2015) reports a RF of land transport aerosol in the order of -81 to $-12\ \mathrm{mW\ m^{-2}}$.

The zonal averages of the shortwave, longwave and net radiative forcing for land transport and ship traffic are shown in Fig. 9. Solid (dashed) lines indicate the RF due to the tagging (perturbation) ~~method~~approach. The overall behaviour of RFs deduced by tagging and perturbation ~~method~~ approach compare very well. However, the RF obtained by the tagging ~~method~~ approach is much larger than the RF obtained by the perturbation ~~method~~approach. In particular, the peak at around 20°N is more enhanced for the tagging ~~method~~approach. This is mainly caused by the ~~fact that the tagging method leads to larger values~~ larger $\mathrm{O_3}$ shares in the upper troposphere, where $\mathrm{O_3}$ is most radiative active, as estimated by the tagging compared to the perturbation approach ~~. Especially in this area is most radiative active.~~ (see Supplement for a figure showing the individual shares). In all cases, the longwave radiative forcing with ≈ 65 % dominates over the shortwave radiative forcing with ≈ 35 %. The overall shape of the net forcing corresponds to the tropospheric $\mathrm{O_3^{tra}}$ and $\mathrm{O_3^{shp}}$ column (not shown). In general, the RFs of land transport and ship traffic are largest in the Northern Hemisphere, where most emissions occur. The overall behaviour of the RF zonal means compares quite well with that reported by Myhre et al. (2011), however, we simulate larger absolute values as discussed above.

Figure 10 shows the vertical profile of land transport and ship traffic radiative forcing for the tagging and perturbation ~~method~~approach. Tagging and perturbation ~~method~~ approach show the same behaviour. However, the tagging ~~method~~ approach has larger values. Most flux changes are simulated in the lower/middle troposphere (300–1000 hPa). Here, the shortwave RF is negative. In contrast, the longwave forcing is positive throughout the whole atmosphere. The vertical profiles correspond to the fraction of $\mathrm{O_3^{tra}}$ (respectively $\mathrm{O_3^{shp}}$) to $\mathrm{O_3}$: the fraction increases with height until it peaks at 850 hPa. In this regime, the largest flux changes occur as well. Above, it continuously decreases with height, so do the flux changes.

7 Uncertainties

The general limitations of the tagging diagnostics applied in this study have been discussed by Grewe et al. (2017), therefore we here discuss only the most important details. The mathematical method itself is accurate, but the implementation into the model requires some simplifications ~~like~~ such as the introduction of chemical families. ~~Recent~~ Grewe (2004) showed for a simple box model that the implementation of the $\mathrm{NO_y}$ family causes an error mainly after the first 12 h after major emission and during this time may lead to and error caused by the family concept of up to 10 %. Further, recent updates of the tagging scheme with respect to differences of the $\mathrm{HO_x}$ family show an influence of ~~some percent on the values of (Rieger et al 2017, in prep.). Therefore~~the error trough these simplifications 1–3 percentage-points on the relative contribution of land transport and shipping emissions to ozone (Rieger et al., 2017) .

Therefore, we conclude that the error through the simplifications of the tagging method is estimated to be smaller than the errors ~~due to model simplifications and assumptions (physics and or chemistry, e.g. 20 % given by Eyring et al., 2007)~~ arising from approximations applied in the global chemistry-climate-models itself (physics and chemistry parameterisations, e.g. 20 % given by Ey

For the future it would be very interesting to compare results from different tagging methods in more detail to have more quan-

titative information about the influence of the simplifications chosen by different methods. ~~To our knowledge~~Other available tagging schemes, however, ~~no other tagging schemes with similar complexity as the scheme applied by us exist so far~~are based on kinetic approaches (Gromov et al., 2010) , consider either only NO_x or VOC (e.g. Emmons et al., 2012; Butler et al., 2011) , or are based on thresholds depending on whether the ozone chemistry is NO_x or VOC limited (e.g. Dunker et al., 2002; Kwok et al., 2015) . The differences between the assumptions and the scales on which they are applied render a detailed comparison impossible.

However, also the perturbation approach faces an important limitation. The calculated impact largely depends on the magnitude of the chosen perturbation and the impacts are only valid for this specific perturbation (e.g. Hoor et al., 2009) . In addition, the perturbation approach has a fundamental problem, namely a non-closed budget. This means that the sum of O_3 changes calculated for different perturbed emission sources (e.g. land transport and aviation) is not necessarily the total O_3 change if all emissions are reduced at the same time (e.g. Wang et al., 2009; Grewe et al., 2010) .

Clearly, the largest source of uncertainties are the emission inventories. Especially for source attribution not only the uncertainties of the emissions source of interest are important, but also the uncertainties of all other emissions sources. As an example, the emissions of NO_x from soil are poorly constrained (e.g. Vinken et al., 2014). This is in particular problematic as part of the soil-NO_x emissions take place in similar regions as the land transport emissions. Therefore NO_x from both emissions sources influences the ozone production concurrently.

Also with respect to the RF calculation our approach uses some assumptions (for the tagging and the perturbation results, respectively) which we discuss in detail in Sect. 2.3 and the Supplement. Compared to calculations of the ozone radiative forcing by comparing two simulation results applying conditions for for present day and preindustrial times we estimate a difference of of 10–30 % (for details see Supplement). This difference are smaller as the factor 2–3 between the results of the tagging and the perturbation approach.

8 Summary and Conclusion

We estimate the contribution of land transport and shipping emissions to tropospheric ozone for the first time with an advanced tagging method which considers not only NO_x, but also CO and ~~.~~

VOC. Our results indicate a maximum contribution of land transport emissions during summer of up to 18 % to ground level ozone in North America and 16 % in Southern Europe, which corresponds to up to 12 $\mathrm{nmol\ mol^{-1}}$ in North America and 10 $\mathrm{nmol\ mol^{-1}}$ in Europe.

The largest contribution of shipping emissions to ground level ozone was simulated in the Northern Pacific Ocean and the Northern Atlantic Ocean. During summer, contributions of up to 30 % were simulated in the Northwestern Pacific Ocean, corresponding to up to 12 $\mathrm{nmol\ mol^{-1}}$. In the Northern Atlantic Ocean contributions of up to 20 % during summer were calculated (up to 12 $\mathrm{nmol\ mol^{-1}}$). The comparison with previous estimates clearly show that the results strongly depend on the chosen method. Perturbation studies using a 5 % approach usually show the lowest contribution (scaled to 100 %) in the ~~regions considered~~ considered regions, while most 100 % perturbations, as well as the tagging approach show the largest contributions.

Overall, emissions of land transport and ship traffic contribute by 8 % and 6 %, respectively, to the tropospheric ozone burden. Land transport emissions contribute by around 20 % to the tropospheric ozone production near the source regions. The contribution of shipping emissions to the net ozone production near the source regions is with values of up to 52 % in the Northern Pacific even larger as the contribution of land transport emissions to the net production.

~~We~~ Using the tagging method we estimate a global average radiative forcing due to ozone caused by land transport emissions of 92 $\mathrm{mW\ m^{-2}}$ and 62 $\mathrm{mW\ m^{-2}}$ caused by ~~to ship~~ shipping emissions. In general, radiative forcings are largest on the Northern Hemisphere and peak at around 30° N. While our estimates of the contribution of land transport ~~/~~and shipping emissions to tropospheric ozone are similar compared to previous studies using a 100 % perturbation, our estimates of the radiative forcing are larger by a factor of 2–3 compared to previous estimates using the perturbation method. As discussed in detail, this large difference compared to previous values is largely attributable to differences in the methodology, leading to different estimates of the ozone shares attributable to land transport and shipping emissions, respectively. Previous estimates of the ozone RF due to land transport emissions using a $\mathrm{NO_x}$-only tagging method, however, are too large as they do not consider the competing effects of $\mathrm{NO_x}$ and VOCs. Accordingly, 92 and 62 $\mathrm{mW\ m^{-2}}$ are the current best estimates of the ozone RF due to land transport and shipping emissions, as estimated using a source apportionment method.

Our results clearly ~~show that a differentiation between results using the tagging and the perturbation method is very important.This holds especially for investigations of the radiative forcing~~indicate that it is important to differentiate between sensitivity methods (i.e. perturbation), which estimate the impact, and the source apportionment methods (i.e. tagging) which estimate the contribution of emissions, because both approaches give answers to different questions. The perturbation approach measures the effect of an emission ~~reduction (or increase)~~change, while only the tagging approach yields contributions of individual emission sources to ozone concentration. This difference is very important when interpreting the results, in particular when investigating the radiative forcing of individual emission categories. To investigate mitigation options, the tagging method cannot replace sensitivity (i.e. perturbation) studies and vice versa. However, we ~~clearly demonstrated that a combination of both methods strengthen the investigation of mitigation options and should be the methodof choice~~demonstrated that even if mitigation options are investigated, the sensitivity simulations should be equipped with a tagging method. The tagging approach provides very valuable additional information about the changes of the contributions to ozone due to the mitigation option, which puts additional pressure on unmitigated sources.

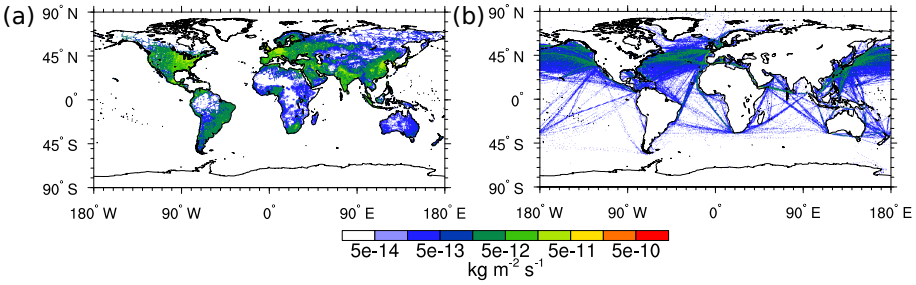

Figure 1. Average (2006–2010) emissions flux of NO_x (in $kg(N)\ m^{-2}\ s^{-1}$) emissions from **(a)** land transport and **(b)** shipping.

Table 1. Description of the different categories as used by the TAGGING submodel.

tagging categories	description
land transport	emissions of road traffic, inland navigation, railways (IPCC code 1A3b_c_e)
anthropogenic non-traffic	sectors Energy, Solvents, Waste, Industries, Residential, Agriculture
ship	emissions from ships (IPCC code 1A3d)
aviation	emissions from aircraft
lightning	lightning NO_x emissions
biogenic	on-line calculated isoprene and soil-NO_x emissions, off-line emissions from biogenic sources and agricultural waste burning (IPCC code 4F)
biomass burning	biomass burning emissions
CH_4	degradation of CH_4
N_2O	degradation of N_2O
stratosphere	downward transport from the stratosphere

Table 2. Average (2006–2010) annual total emission of CO (in $\mathrm{Tg(CO)\ a^{-1}}$), $\mathrm{NO_x}$ (in $\mathrm{Tg(N)\ a^{-1}}$) and NMHC (in amount of carbon) of the most important emission categories. The category 'other' contains the emissions of the sectors biomass burning, agricultural waste burning as well as other biogenic emissions.

	CO ($\mathrm{Tg(CO)\ a^{-1}}$)	NMHC ($\mathrm{Tg(C)\ a^{-1}}$)	$\mathrm{NO_x}$ ($\mathrm{Tg(N)\ a^{-1}}$)
land transport	152	17	10
shipping	1	2	6
anthropogenic non-traffic	411	73	17
soil NOx			6
lightning NOx			5
biogenic $\mathrm{C_5H_8}$		493	
other	416	15	5

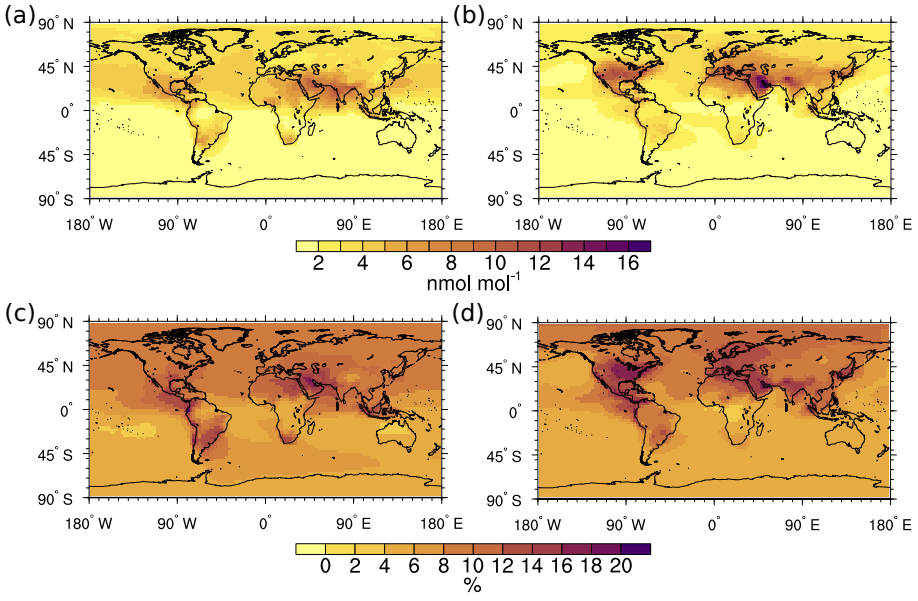

Figure 2. Seasonal average values of the absolute and relative contribution of $\mathrm{O_3^{tra}}$ to near ground level $\mathrm{O_3}$. The upper row give the absolute values (in $\mathrm{nmol\ mol^{-1}}$) for winter (DJF, **(a)**) and summer (JJA, **(b)**), respectively. The lower row shows the DJF **(c)** and JJA **(d)** values of the contribution (in %).

Table 3. Summary of previous global model studies investigating the contribution/impact of land transport/road traffic emissions to ozone. Method denotes the percentage of the emissions reductions (perturbation). The other columns list the amount of land transport/road traffic emissions as well as the fraction (f) compared to the emissions used in the studies for NO_x (in $Tg(N)\ a^{-1}$), CO (in $Tg(CO)\ a^{-1}$) and NMHC ($Tg(C)\ a^{-1}$). The four rows from the right list the contribution of the land transport/road traffic categories as estimated by these studies in mixing ratios and/or percent. Where possible, we show the estimated contribution for the geographical regions defined in Sect. 2 as well as zonal average values. All contributions are given to near ground level ozone and for July conditions. The table is ordered by the year of publication. A '-' indicates missing information.

study	method	NO_x	fNO_x	CO	fCO	NMHC	fNMHC	NA	EU	SEA	ZM
								$nmol\ mol^{-1}$	$nmol\ mol^{-1}$	$nmol\ mol^{-1}$	$nmol\ mol^{-1}$
		$Tg\ a^{-1}$	%	$Tg\ a^{-1}$	%	$Tg\ a^{-1}$	%	%	%	%	%
GB03	100%	10	24	207	14	-	-	-	-	-	-
								11–15	9–15	5–12	-
NM06	100%	9	30[a]	196	36[a]	36	27[a]	5–20	5–15	5–10	-
								10–50	-5–25	5–50	-
NM06	100%	9	30[a]	196	36[a]	36	27[a]	zonal mean			-
											up to 10
M07	100%	9	24	237	-	27	5	-	-	-	-
								13–16	9–16	3–16	-
M07	100%	9	24	237	-	27	5	zonal mean			up to 5
											up to 12
H09	5 %[b]	7	15	31	7	8	2	2–5[c]	2–6[c]	1–4[c]	-
								-	-	-	-
K10	5 %[b]	9	18	110	11	11	1	2–5	-1–5	1–3	-
								-	-	-	-
K10	100 %	9	18	110	11	11	1	zonal mean ground level			-
											up to 7
this study	tagging	10	20	152	16	17	3	3–14	3–13	2–11	
								6–19	8–18	5–16	
this study	tagging	10	20	152	16	17	3	zonal mean mid latitudes NH			3–7
											9–11
this study	5 %[b]	10	20	152	16	17	3	1–9	-1–6	-1–5	-
								1–12	-3–9	-2–12	-
this study	5 %[b]	10	20	152	16	17	3	zonal mean mid latitudes NH			2–4
											1–2

[a] Fraction only compared to all anthropogenic emissions. [b] Given values scaled to 100 %. [c] Given for average values from 800 hPa to the surface.
Abbreviations are: GB03 (Granier and Brasseur, 2003), N06 (Niemeier et al., 2006), M07 (Matthes et al., 2007), H09 (Hoor et al., 2009), K10 (Koffi et al., 2010).

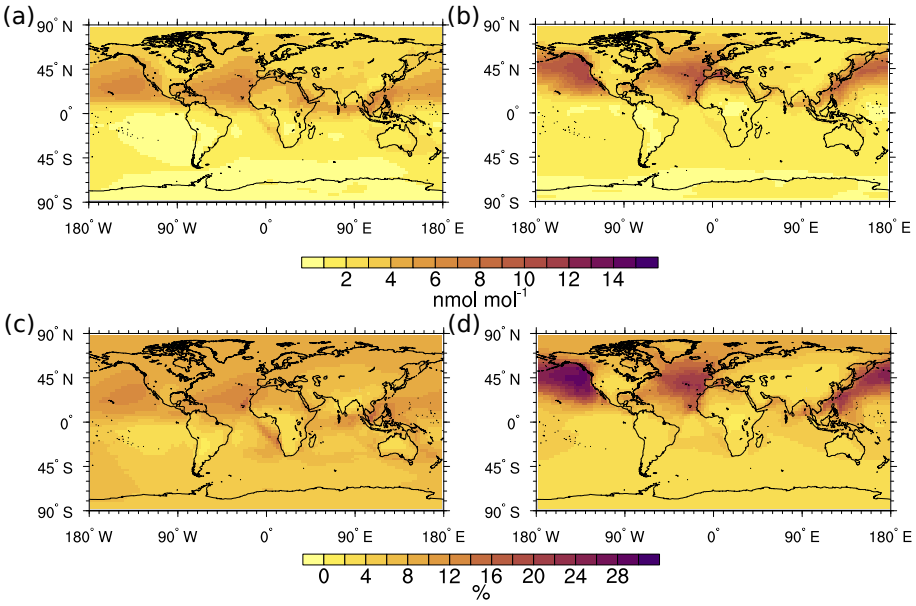

Figure 3. Seasonal average values of the absolute and relative contribution of O_3^{shp} to near ground level O_3. The upper row give the absolute values (in $\mathrm{nmol\ mol^{-1}}$ for DJF **(a)** and JJA **(b)**, respectively. The lower row shows the DJF **(c)** and JJA **(d)** values of the contribution (in $\%$).

Table 4. Summary of previous global model studies investigating the contribution/impact of shipping emissions to ozone. Method denotes the percentage of the emissions reductions (perturbation). The other columns list the amount of shipping emissions as well as the fraction (f) compared to all emissions used in the studies for NO_x (in $Tg(N)\ a^{-1}$). The four rows from the right list the contribution of the shipping category as estimated by these studies in mixing ratios (upper row) and/or percent (lower row). Where possible, we show the estimated contribution for the geographical regions defined in Sect. 2 as well as zonal average values. For the geographical regions we give only the values larger than the background values. All contributions are given to near ground level ozone and for July conditions. The table is ordered by the year of publication. A '-' indicates missing information.

study	method	NO_x	fNO_x	Atlantic	Pacific	India	Zonal Mean
				$nmol\ mol^{-1}$	$nmol\ mol^{-1}$	$nmol\ mol^{-1}$	$nmol\ mol^{-1}$
		$Tg\ a^{-1}$	%	%	%	%	%
ED03	100%	4	8	4–12	4–11	3–4	-
				-	-	-	-
E07	100%	3	11[a]	2–12	1–4	1–4	-
				12–36	12–24	12–18	-
E07	100%	3	11[a]	zonal mean mid latitude NH			1–1.5
							-
H09	5%[c]	4	10	2–4	2–3	1–2	-
				-	-	-	-
D09	100 %	5	-	-	-	-	-
				14–33	14–40	9–12	-
K10	5%[c]	4	8	2–5	3–6	1–2	-
				-	-	-	-
K10	5%[c]	4	8	zonal mean			up to 1.5
							-
K10	100%	4	8	up to 8	up to 9	-	-
				-	-	-	-
K10	100%	4	8	zonal mean			up to 3
							-
this study	tagging	6	12	3–9	4–11	2–5	-
				10–24	10–33	9–15	-
this study	tagging	6	12	zonal mean mid latitudes NH			3–6
							10–15
this study	5 %[c]	6	12	2–8	2–7	1–4	-
				10–18	11–22	4–10	-
this study	5 %[c]	6	12	zonal mean mid latitudes NH			2–4
							5–8

[a] No information available. [b] Fraction only compared to all anthropogenic emissions. [c] Given values scaled to 100 %. [d] Given for average values from 800 hPa to the surface. Abbreviations are: ED03 (Endresen et al., 2003), E07 (Eyring et al., 2007), H09 (Hoor et al., 2009),D09 (Dalsøren et al., 2009), K10 (Koffi et al., 2010).

Table 5. Comparison of Γ values (definition see text) between the four considered regions and interpretation of these values.

	Γ	Interpretation
Europe	0.9	90 % of the O_3 reduction due to land transport emissions are compensated by increased ozone production. Ozone contribution and impact differ largely.
Southeast Asia	0.6	10 % reduction of land transport emissions will lead to a 4 % reduction in ozone due to increased ozone productivity. Ozone contribution and impact differs largely.
North Africa	0.4	Only small compensation effects; ozone contribution and impact differ only slightly.
South America	0.3	Land transport emission reduction almost scales with ozone reduction. Impact and contribution are almost equal.

Table 6. Burden of O_3 and O_3^{tra} integrated up to 200 hPa (in Tg). Average values for the period 2006–2010.

	O_3 (Tg)	O_3^{tra} (Tg)	contribution O_3^{tra} (%)
Global	318	24	8
Europe	15	2	10
North America	21	2	10
Southeast Asia	25	2	9

Table 7. Burden of O_3 (total) and O_3^{shp} (shipping) integrated up to 200 hPa (in Tg). Average values for the period 2006–2010.

	O_3 (Tg)	O_3^{shp} (Tg)	contribution O_3^{shp} (%)
Global	318	18	6
North Atlantic Ocean	24	2	8
Indian Ocean	27	1	5
North Pacific Ocean	32	2	8

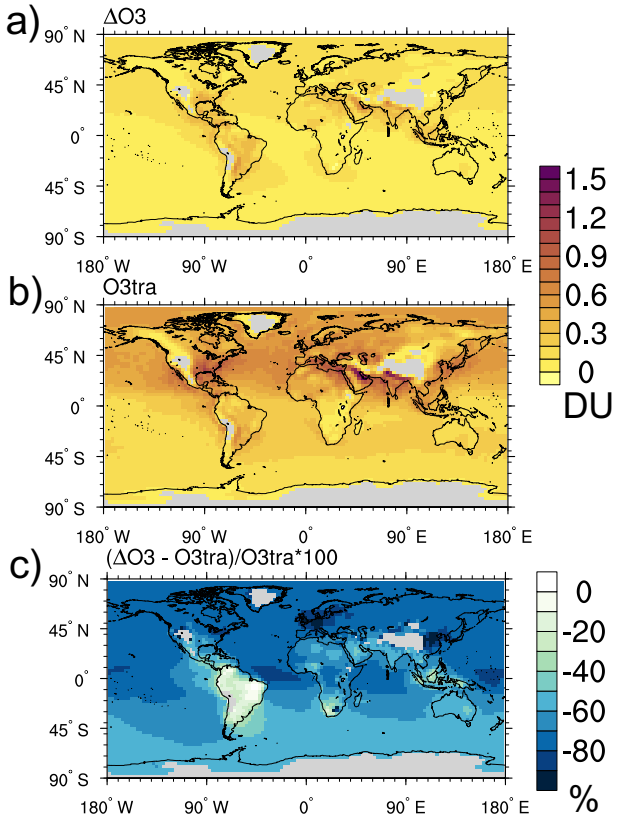

Figure 4. Multi-annual averages (2006–2010) of **(a)** ΔO_3 (impact), **(b)** ~~ΔO_3^{tra}~~ O_3^{tra} (contribution, both in DU) of the *REF* simulation and **(c)** ~~$\Delta O_3^{tra} / \Delta O_3$~~ the relative difference between the impact and the contribution of land transport emissions (in %). ~~The differences~~ All values are calculated for the partial columns from the surface of up to 850 hPa (850PC).

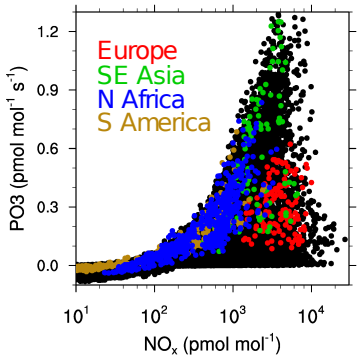

Figure 5. Dependency between NO_x mixing ratios and net O_3 production. ~~(a) Gridbox values:~~ The black dots represent monthly mean values at ground level for the year 2010 of every individual grid box. The individual colours indicate monthly average values during May–August (Northern Hemisphere) and November–February (Southern Hemisphere) for individual regions (defined as rectangular areas~~, see Appendix I~~). ~~(b) Regional values: The single dots represent year 2010 averages for the four regions shown in (a). In addition the tangents were calculated for every region by comparing the perturbed and the unperturbed simulations. The black rectangle highlights the region shown in (c). The x-axes of (c) and (b) are linear, while (a) uses a logarithmic x-axis.~~

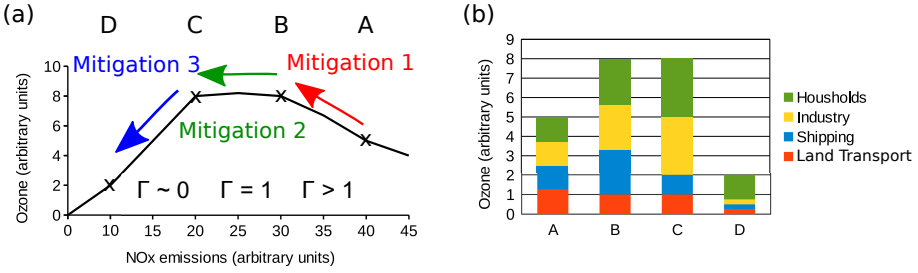

Figure 6. Idealised example explaining the difference of the perturbation and the tagging approach for the evaluation of mitigation increases. (a) shows the dependency between NO_x emissions and ozone (both in arbitrary units). Three different mitigation options are indicated by the colored arrows. In addition, the approximate value of Γ (see text for definition) is given. (b) shows the contribution of the ozone concentration at the four marked points in (a). In this example it is assumed that only four emission categories exist, emitting the same amount of emissions at point A.

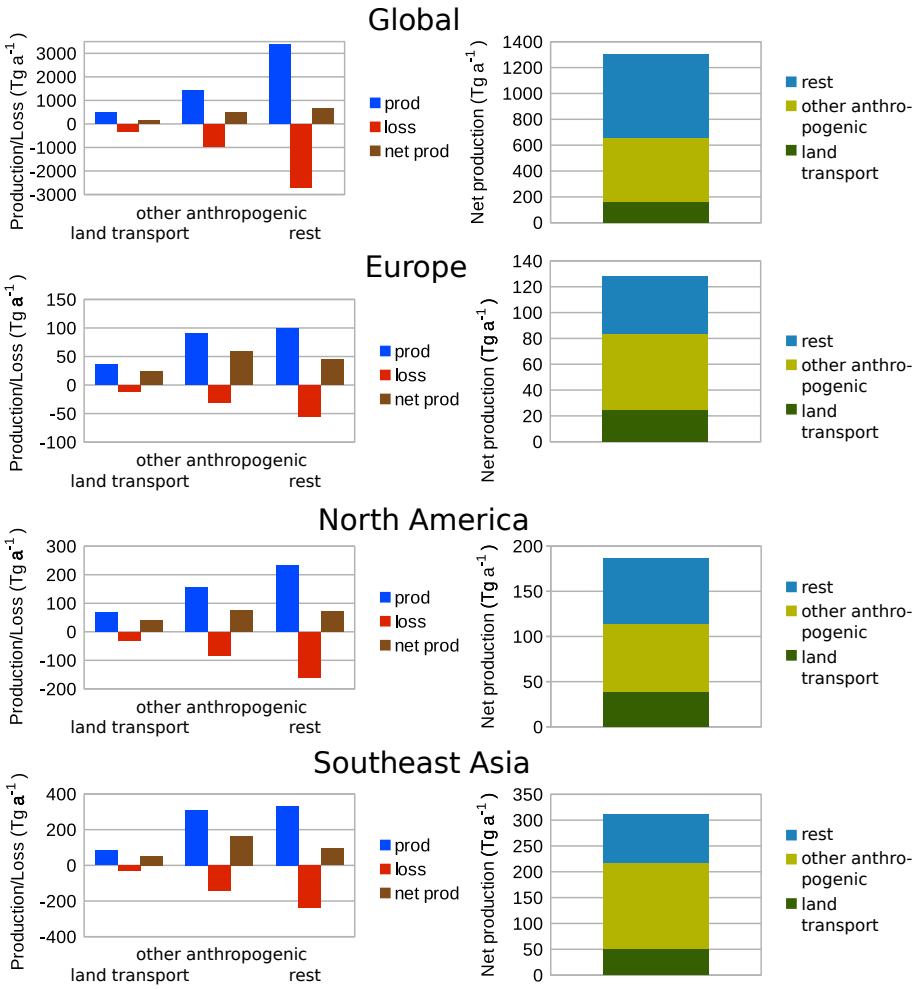

Figure 7. Production and loss rates of O_3 from different sectors (integrated up to 200 hPa and averaged for 2006–2010). The left side shows the individual production and loss rates as well as the net O_3 production, while the right side shows only the net production of the different sectors. For simplicity only land transport, other anthropogenic (shipping, anthropogenic non-traffic and aviation) and rest (all other tagging categories) are shown.

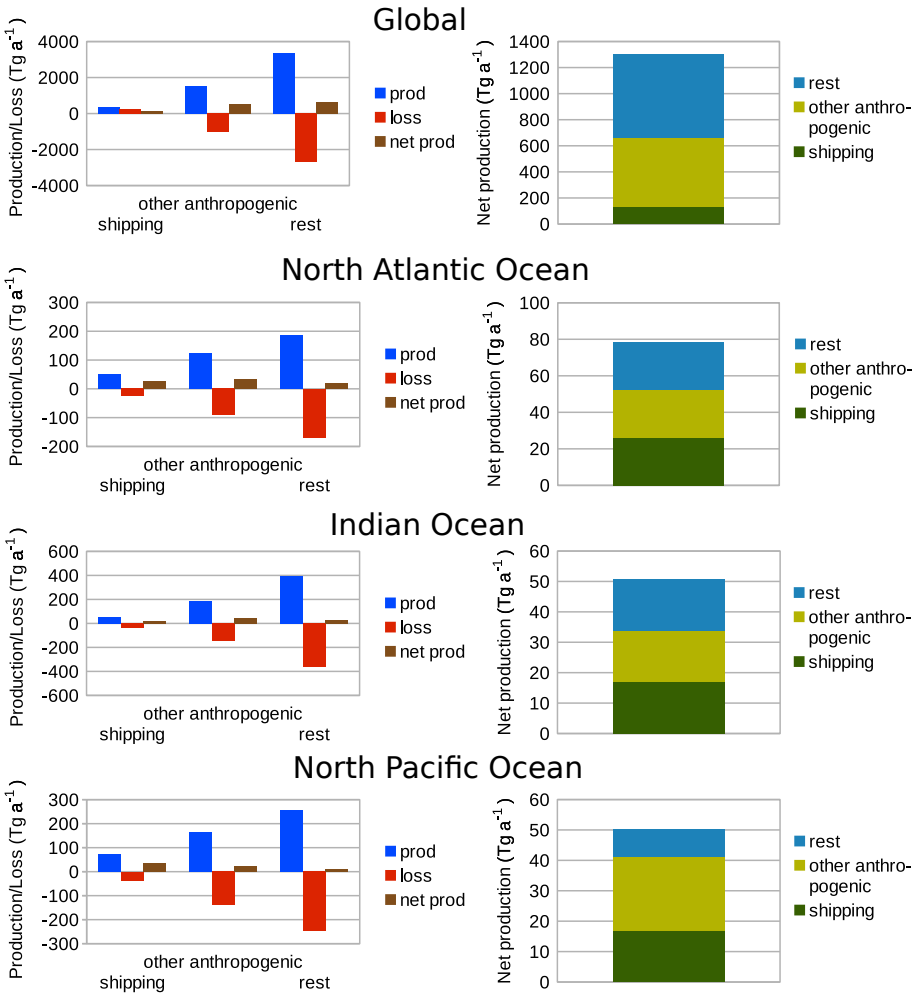

Figure 8. Production and loss rates of O_3 from different sectors (integrated up to 200 hPa and averaged for 2006–2010). The left side shows the individual production and loss rates as well as the net O_3 production, while the right side shows only the net production of the different sectors. For simplicity only shipping, other anthropogenic (land transport, anthropogenic non-traffic and aviation) and rest (all other tagging categories) are shown.

Table 8. Global estimates of the annually averaged radiative forcing due to O_3 caused by emissions of land transport/road traffic (global RF road) and ship emissions (global RF shp). Please note that individual studies use different methods for the calculation of the radiative forcing e.g. some studies give instantaneous values, while other studies stratospheric adjusted values (see last row).

Study	method	global RF road ($mW\ m^{-2}$)	global RF shp ($mW\ m^{-2}$)	RF type
Endresen et al. (2003)	100 %	-	29	scaling of tropospheric ozone column change
Niemeier et al. (2006)	100 %	30 / 50 (January / July)	-	instantaneous at TP[e]
Eyring et al. (2007)	100 %	-	10 ± 2	instantaneous at TP[e] decreased by 22 %
Fuglestvedt et al. (2008)	100 %	54 ± 11	32 ± 9	stratospheric adjusted
Hoor et al. (2009)	5 %	28[a]	28[a]	-
~~Dahlmann et al. (2011)~~ [c] ~~tagging~~ review Uherek et al. (2010)		$50 - (54 \pm 11)$	-	-
Dahlmann et al. (2011)	NO_x-tagging	170[c]	49[c]	fixed dynamical heating
Dahlmann et al. (2011) [c]	100 %	31[c]	-	fixed dynamical heating
Myhre et al. (2011)	5 %	31[a]	24[a]	
Grewe et al. (2012)	~~tagging~~[c] NO_x-tagging	132[c]	-	fixed dynamical heating
Grewe et al. (2012)	100 % [c]	24[c]	-	fixed dynamical heating
Holmes et al. (2014)	5 %	-	27[d]	-
this study	~~tagging~~ NO_x/VOC-tagging	92	62	stratospheric adjusted
this study	5 %	24[a]	22[a]	stratospheric adjusted

[a] Scaled to 100 %. [b] For year 2000 conditions. [c] For year 1990 conditions. [d] Calculated by scaling the RF value of the 'instant dilution' case for a change of 1 $Tg\ a^{-1}$ with the total amount of used emissions by Holmes et al. (2014). [e] Tropopause

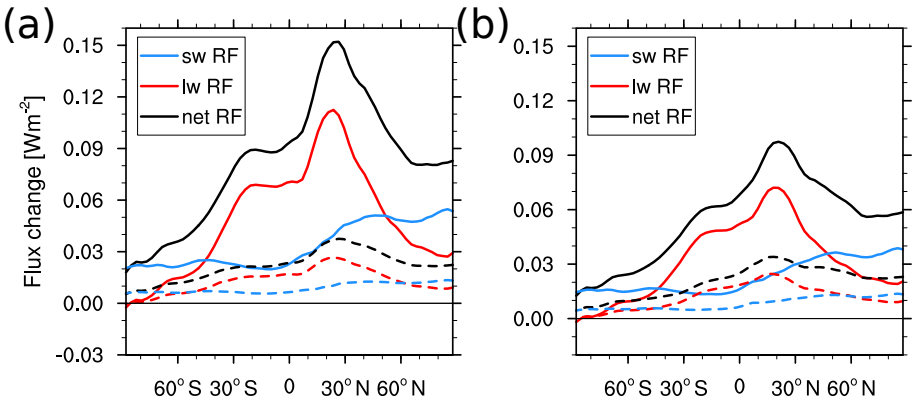

Figure 9. Zonal mean of shortwave, longwave and net radiative O_3 forcing of (a) land transport and (b) ship traffic. The continuous lines give the results of the tagging method, the dashed lines of the perturbation method.

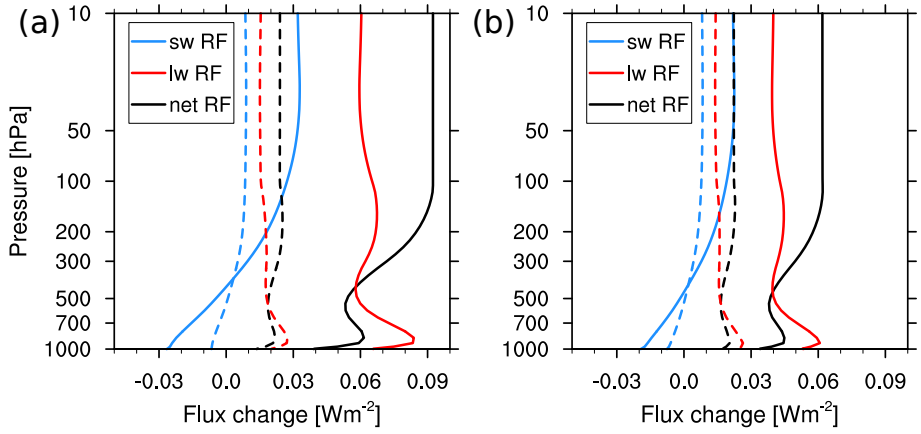

Figure 10. Vertical profile of globally averaged shortwave, longwave and net radiative O_3 forcing of (a) land transport and (b) ship traffic. The continuous lines give the results of the tagging method, the dashed lines of the perturbation method.

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