

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/APCA and 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 usefulness 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 RO<sub>2</sub>. Thus, the formulation already assumes that limiting factors are important. Why is the limiting factor not important between NO<sub>x</sub> 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 exists, some consider NO<sub>x</sub> or VOC only (e.g. Grewe, 2004; Emmons et al., 2012), our method considers NO<sub>x</sub> and VOCs, others use thresholds to judge, whether the chemistry is NO<sub>x</sub> or VOC limited and attribute ozone to NO<sub>x</sub> 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<sub>x</sub> or VOC limit'-threshold are by definition more sensitivity based and not comparable to the contribution estimated by considering NO<sub>x</sub> 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<sub>3</sub> 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  $\Delta_T^{\text{tra}} = O_3 - O_3^{\text{tra}}$  and  $\Delta_T^{\text{shp}} = O_3 - O_3^{\text{shp}}$  were calculated.  $\Delta_T^{\text{tra}}$  and  $\Delta_T^{\text{shp}}$  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  $\Delta_T^{\text{tra}}$ ,  $\Delta_T^{\text{shp}}$  and  $O_3$ . The  $O_3$  RFs of land transport and shipping emissions using the tagging approach are then calculated as follows:

$$\text{RF}_{O_3\text{tra}}^{\text{tagging}} = \text{flux}(O_3) - \text{flux}(\Delta_T^{\text{tra}}), \quad (1)$$

$$\text{RF}_{O_3\text{shp}}^{\text{tagging}} = \text{flux}(O_3) - \text{flux}(\Delta_T^{\text{shp}}), \quad (2)$$

with *flux* 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,  $\Delta O_{3\text{tra}}$  and  $\Delta O_{3\text{shp}}$  are calculated by taking the difference between the unperturbed (*BASE*, see below) and the perturbed simulations (*LTRA95* or *SHIP95*):

$$\Delta O_3 = (O_3^{\text{unperturbed}} - O_3^{\text{perturbed}}) \cdot 20. \quad (3)$$

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,  $\Delta O_{3\text{tra}}$  and  $\Delta O_{3\text{shp}}$  are then treated as described above for  $\Delta_T^{\text{tra}}$  and  $\Delta_T^{\text{shp}}$ . These RFs are called  $\text{RF}_{\Delta O_{3\text{tra}}}^{\text{perturbation}}$

and  $RF_{\Delta O_3shp}^{perturbation}$ , 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  $RF_{O_3tra}^{perturbation}$  and  $RF_{O_3tra}^{tagging}$  (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. Appor-tionment 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<sub>x</sub>, 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<sub>x</sub> 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<sub>x</sub> (NO and NO<sub>2</sub>), 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 NO<sub>y</sub> (NO<sub>x</sub> + NO<sub>z</sub>) is equally available for ozone production. This is problematic because NO<sub>y</sub> photochemical lifetime is much longer than NO<sub>x</sub>. As a result, this Eq 2 will attribute ozone production to NO<sub>x</sub> and NO<sub>z</sub> proportionally. That would lead to ozone being attributed to HNO<sub>3</sub><sup>tag</sup> in the mid to upper troposphere. Unless NO<sub>y</sub> 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 RO<sub>2</sub> 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<sub>3</sub> production rates for each member of the NO<sub>y</sub> family individually, according to their kinetic rate coefficient (e.g. no O<sub>3</sub> is produced in regions, where only HNO<sub>3</sub> 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 misattribution of ozone. Consider a case in which O<sub>3</sub> is locally produced from lightning NO<sub>x</sub> emissions. Using the family approach the tagged NO<sub>y</sub> family locally may consist also of HNO<sub>3</sub> from e.g. anthropogenic emissions. Accordingly, some of the produced O<sub>3</sub> 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 %.

We added a note on this in Sect. 7:

**Grewe (2004) showed for a simple box model that the implementation of the NO<sub>y</sub> family causes an error mainly after the first 12 h after major emission and during this time may lead to an 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<sub>3</sub> 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; Emons 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,  $\Delta O_{3\text{tra}}$  (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 'success' 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<sub>x</sub> 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<sub>x</sub>/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 NO<sub>x</sub> 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<sub>x</sub> 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<sub>x</sub> 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<sub>3</sub> shares in the upper troposphere, where O<sub>3</sub> 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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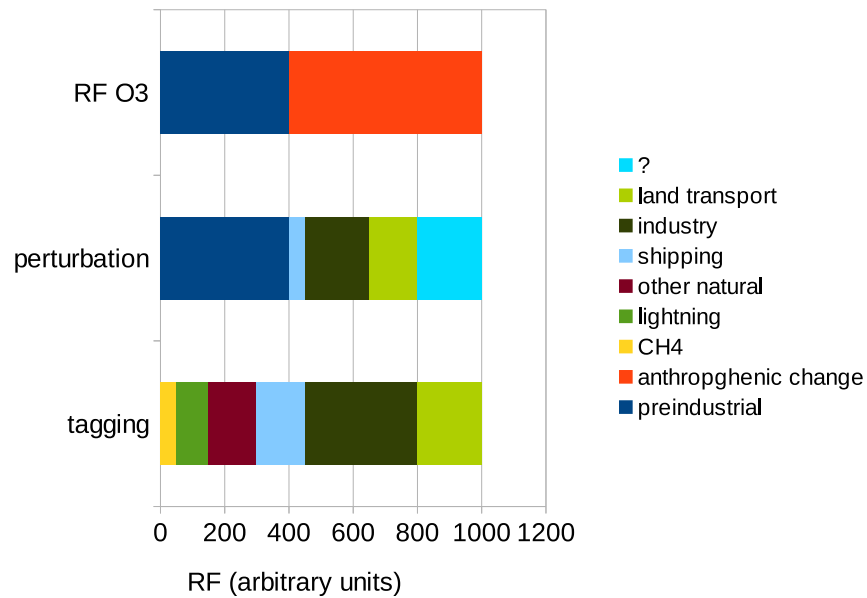


Figure S1: Simplified sketch of three different ways to calculate RFs. 'RF O3' shows the classical way of calculating the anthropogenic RF by calculating the radiative flux of an preindustrial simulation and a simulation with all emissions. 'Perturbation' shows the perturbation approach, here the RF 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.

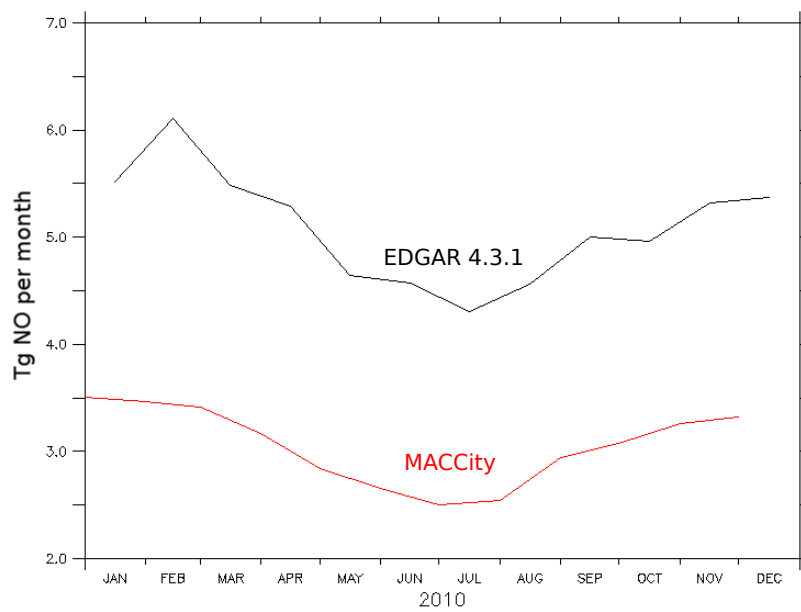


Figure S2: Globally integrated  $\text{NO}_x$  emissions (in Tg (NO) per month) of the anthropogenic non-traffic sector for the MACCity emission inventory (red) and the EDGAR 4.3.1 inventory (black). Shown are values for the year 2010 exemplarily.

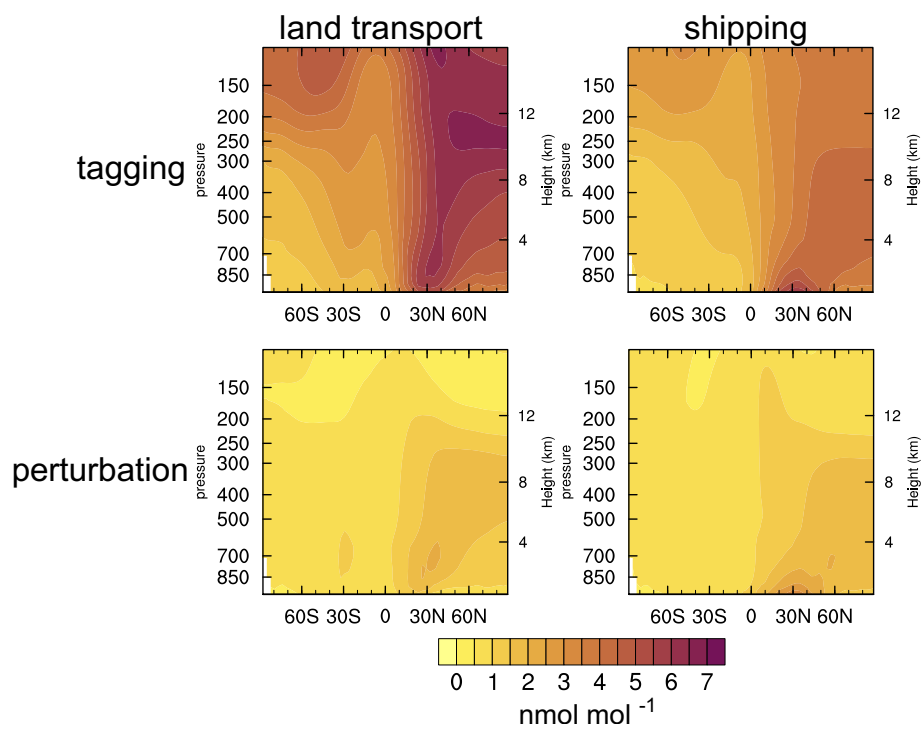


Figure S3: Multi-annual zonal average (2006–2010)  $O_3$  shares as estimated by the perturbation method and the tagging approach. Shown are the contribution and impact of the land transport and shipping emissions to ozone, as estimated by the tagging method and the perturbation approach, respectively.