

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, reproducibility 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_{O_3}), respectively, for a particular region. The m denotes the slope, which corresponds to an approximation of the derivative $dP_{\text{O}_3}/d\text{NO}_x$ in the unperturbed simulation, which is calculated by the difference in ozone production and NO_x mixing ratios in the unperturbed and perturbed simulation. $x_0 = \text{NO}_x^u$ is the NO_x mean mixing ratio in the unperturbed simulation and $b = P_{\text{O}_3}^u - dP_{\text{O}_3}/d\text{NO}_x \text{NO}_x^u$, where $P_{\text{O}_3}^u$ is the mean ozone production in the unperturbed simulation.

Based on the linearised ozone production ($P_{\text{O}_3}^{\text{lin}}$) calculated by the tangent approach, we define a saturation indicator Γ , which helps to analyse the ozone sensitivity further:

$$\Gamma = \frac{\text{y - axis intercept}}{\text{y - value of unperturbed simulation}} = \frac{P_{\text{O}_3}^{\text{lin}}(\text{NO}_x = 0)}{P_{\text{O}_3}^{\text{lin}}(\text{NO}_x = \text{unperturbed})}. \quad (1)$$

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 ($P_{\text{O}_3}^{\text{lin}}(\text{NO}_x = 0) = P_{\text{O}_3}^{\text{lin}}(\text{NO}_x = \text{unperturbed})$). 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 ($dP_{\text{O}_3}/d\text{NO}_x$), $\Gamma = 1$ corresponds to $dP_{\text{O}_3}/d\text{NO}_x = 0$, while $\Gamma > 1$ corresponds to $dP_{\text{O}_3}/d\text{NO}_x < 0$ and vice versa.

5. It is unclear why gamma is defined in terms of the intercept instead of the slope ($d\text{O}_3/d\text{NO}_x$). The intercept will be leveraged by the amount of the NO_x emissions. That is, the impact of the slope will be amplified when the NO_x emissions are large by changing the intercept to a greater extent than if the NO_x emissions are small.

Reply: Of course Γ could also be defined in terms of the slope (dPO_3/dNO_x). 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 NO_x is shown, with the well-known result that in regions of high NO_x 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

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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 therefore 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 intercomparison, 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 $RF_{O3tra}^{tagging}$ and $RF_{\Delta O3tra}^{perturbation}$ (which we define in Sect. 2):

We obtain a global net RF for land transport of $RF_{O3tra}^{tagging} = 92 \text{ mW m}^{-2}$. The shortwave RF is 32 mW m^{-2} and the longwave RF is 61 mW m^{-2} . The estimated RF of ship traffic is $RF_{O3shp}^{tagging} = 62 \text{ mW m}^{-2}$ 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 O₃ changes calculated for different perturbed emission sources (e.g. land transport and aviation) is not necessarily the total O₃ change if all emissions are reduced at the same time (e.g. Wang et al., 2009; Grewe et al., 2010).

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