## Response to Reviewer #1

We would like to thank the reviewer for the helpful comments on the manuscript. It helped to improve our manuscript. Please find our comments in italic.

This publication describes the development of a parameterized source-receptor model at global scale to have a tool for fast assessment of the atmospheric impacts of (changes in) road transport emissions. The paper makes the impression that it was written a few years ago, as some relevant recent references were not included. Overall, it was not clear to me, why this model would be specific to road-transport emissions, as the modeling principle could be relevant for other source types as well. While the ability to describe source contribution and source emission sensitivity was one of the 'sales' arguments for the paper, the element of describing how the 'contribution' element is tested, and how it is used for assessment was not well developed in the paper. The paper is perhaps also a bit too much overselling it's uniqueness. While it is good to have several modelling tools that can make rapid impact assessments, there are now several assessment tools in the literature, including the TM5-FAST model, but also e.g. Wild et al. 2012, and Butler et al. (2018) https://gmd.copernicus.org/articles/11/2825/2018/

Thank you for your comments. As also suggested by reviewer 2, we thoroughly restructured the method section. The description of the tagging method and the introduction of the term contribution are now at the beginning of the method section. This makes the usefulness of the terms contribution and impact clearer to reader. To our knowledge, so far no other response model assesses the contribution (determined by a tagging method) and the impact of road traffic emissions together.

As suggested, we added the references to our manuscript. For example, the study Wild et al. (2012) is compared to TransClim in the conclusions. The study Butler et al. (2018) describes another tool for source attribution of tropospheric ozone. As we use the tagging scheme introduced by Grewe et al. (2010), the paper of Butler et al. (2018) does not directly relate to the response model TransClim. Thus, we did not include it in the assessment of the response models in the conclusion. Furthermore, Van Dingenen et al. (2018) provides indeed a similar tool as TransClim, however, with a different approach and with different impact metrics. As discussed in the conclusion, the approach used in TransClim is not necessarily bound to road traffic emissions. It can be easily extended on other traffic modes or other emission sources. For now, the goal of TransClim is to assess the climate effect of mitigation strategies for road traffic.

I recommend to publish the paper with major revisions, after considering the comments above and the detailed remarks below.

Detailed remarks:

2 and particulate matter- along with NO2 one of the most important pollutants- a bit strange to leave it out in this list, even your study is not focussing on it. *As suggested, we included "particulate matter" in the list.* 

5 it may be worth to mention here the characteristic of the model that make it specific for transport. In general such models do not need to be specific for transport, but could be applied to any sector.

The approach of TransClim is indeed very flexible. It is easy to extend the LUTs to include other transport modes or even other emission sectors. This is discussed in the conclusion.

4 I understand that you define impact on two dimensions: response and contribution. This needs to be clearer highlighted in the abstract. The combined approach to contribution and responses (I think) is what makes Transclim and this analysis special. (?)

We added more explanation to the abstract.

24 and probably more than this, including methane (for certain engine types) and HCFCs/HFCs from airconditioers.

*Yes, this is an incomplete list of the pollutants emitted by road traffic. We adapted the sentence.* 

25 here you correctly include PM (but not in abstract). And PM is influencing ozone through heterogeneous reactions.

Thank you, we added particulate matter in the abstract.

- 35 Most studies? Are there studies that don't and why? In a few cases, road traffic emissions can also decrease winter-time ozone in the troposphere. We added an example to the text.
- 37 What did Reis and Tagaris find? Sentence is is now without purpose. Thank you, we gave more information on the study Tagaris et al. (2015).

39 Explain better what the 0.8 % refers to. Global ERF; German ERF, 0.8 of world traffic? I can imagine many possibilities...The 5 mK is a global number, or Germany?

Hendricks et al. (2018) determines the contribution of German road traffic emissions to the total anthropogenic stratosphere-adjusted radiative forcing and the global mean surface temperature change. We adapted the sentences accordingly.

41 to 52. This paragraph, which is essentially the overall method is not terribly well elaborated. I propose you first indicate that you want a method and tool that can both assess sensitivity to emission changes, and current contribution and call that impact. I don't really see why an emission perturbation is solely defining impact. Later you define 'total effect' an unnecessary and ambiguous term. Clear language may help the understanding. L. 43- I only partly agree that perturbation method do not take non-linear relations into account- for instance a perturbation on a preindustrial situation would give a very different result from the perturbation of present-day. I propose: "depending on the chemical regime, large errors may occur when extrapolating emission perturbation relationships to larger perturbations." L44 'it quantifies only impact of emissions': apart from what is impact, I think it is important that you talk here about emission perturbations (sensitivities).

This paragraph gives an overview of the methods commonly used and introduces the perturbation and tagging method. Thank you for pointing at the necessity to clearly define the terms used. Very much appreciated! We rephrased the paragraph to better explain the terms impact, contribution and effect. Correct, the perturbation approach is a linearization of the current chemical state. And we agree that for larger perturbations errors may occur. However, this is not the point we made. In non-linear systems the perturbation/sensitivity approach and contribution method must give different results, which are significantly even for small perturbations (e.g. Grewe et al. 2010).

57 unclear until this point what you do with methane- from a variety of perspectives. Although direct emissions may not be terribly high, indirect emissions may be more significant, e.g. from oil and gas production. A large part of the fuel is used for transport. Changes in emissions of NOx, VOCs, and CO will affect the lifetime of CH4 on timescales of up to ca. 20 years. There are methods how to include the effect of lifetime changes on CH4 itself (an important effect) and O3. Here or before it should be already be mentioned if/how this is included. TransClim computes the contribution of various emissions and precursors to the methane destruction and by that the contribution to methane lifetime as well as changes caused by OH changes. Methane as an ozone precursor is not yet regarded. Direct emissions of methane from road traffic are small and thus neglected. We added this information to the introduction.

73 The graph is not extremely informative, as it doesn't provide much insight in 1) the scale of emission perturbation (grid, region, world?), the type of perturbation (annual, monthly, all components together, or separate, size of perturbation), time scale of effects, equilibrium or transient.... Is this figure needed?

We deleted the figure and rephrased the paragraph. It gives an overview of how TransClim works and thus guides the reader through the method section.

89 This section is in part not requirements but rather a description of assumptions. Rename?

We have formulated requirements which are important for the performance of TransClim. We actually tested various methods in Rieger (2018) and the algorithm which performs the best is presented in this manuscript. Here we only refer to the results in terms of conclusions. We have reformulated the text to clarify this.

## 93 where is CH4?

This study focuses on the effect of road traffic emissions on ozone. The effect on methane is a side aspect which is evaluated for German road traffic emissions in table 5. The requirements are similarly applicable to methane.

97 As explained before, most models consider non-linearity. The point is that the nonlinear response to changes should be computed within I a certain margin of accuracy.

Most response models which are available in this research field do not consider the non-linear behaviour of the tropospheric ozone chemistry (see conclusion). They assume a linear relationship between emission change and ozone change. Consequently, it is important that TransClim regards this non-linearity. As you mentioned, it is further important that the deviations between the results computed by TransClim and EMAC remain low. This condition is covered by requirement (6). Please see also our comment above.

98 Explain why you think the choice of these big continental scale regions is appropriate for the problems that need to be quantified.

The emission regions are chosen in such a way that the climate effect of road traffic emissions from different part of the world can be evaluated. The source-receptor relations differ for different regions. This set of emission regions is not fix. If there is a desire to refine the emission regions over a particular region, then new emission variation simulations need to be performed with EMAC and integrated to the LUTs of TransClim. On purpose, we build up the algorithm of TransClim very flexible, so the integration of new emission regions is very easy (see also section 2.4.2 and conclusion). We added further explanation to section 2.4.2. Besides, there is a large difference in local air quality responses and climate impact responses. These emission regions are not applicable for air quality assessments.

103 Again here: calculate is one thing, but with which accuracy. Is this ERF or RF. In either case to what extent is this state of the art and method?

We consider the stratosphere-adjusted radiative forcing here. The stratosphereadjusted radiative fluxes are calculated by the global chemistry-climate model EMAC which is a state-of-the-art model. The question how accurate can the algorithm of TransClim determine the stratosphere-adjusted radiative forcing is evaluated later in the manuscript in section 3 "Model evaluation". 105 Background refers to a hypothetical situation without (transport?) emissions. Is this what is meant. Or do you rather mean that the large ozone trajectory according to socioeconomic and technological assumptiosn as used in the climate community should be considered. Also note that the RCps are now superseded by SSPs (with some consequences for emission trajectories). This is not a major issue for the concept, but this could be mentioned somewhere.

As suggested by reviewer 2, we omitted this point here to focus on the points which TransClim is able to do. We added the consideration of the background ozone concentrations as future improvements of TransClim in the conclusion section.

107 this is about specifying efficiency and accuracy.

Yes, this is correct. But the response model has to be efficient and provide low errors; otherwise it does not provide an additional benefit. Thus, efficiency and accuracy is a requirement for the algorithm.

111 (Figure 2). Please clarify if the red dots are representing what has be done in terms of perturbations. In this case it may be a bit problematic that not more perturbation lower than 1 have been implemented, as in several world regions this may be the overall trajectory that emissions are going already, and will even more so go in future. It is also not very clear how the point 6 is taken into account (changing baseline ozone).

The red dots are just a sketch of the emission scaling and do not represent the actual emission variation simulations. These can be found in table 2 now. We indicated this in the text. As mentioned in the conclusion, the requirement 6 could not be regarded by this algorithm and will be investigated in future studies.

131 what is a standard computer? What do you calculate for an emission scenario, each year, every 10 years?

Here, a standard computer describes a work station, in contrast to a high performance computing system. We noted this in the text as well. The runtimes are given for an arbitrary emission scenario in one emission region. The runtimes are not depended on the emission scenario, but on the number of performed interpolations which depend on the number of considered emission regions.

138-146 in an earlier part of the text it should already be explained what problems need to be solved, and why these large regions are appropriate for this.

The current set of emission regions is not fix. If needed, additional emission variation simulations can be performed with EMAC for smaller emission regions. We added application examples for TransClim in the introduction.

156-169 Again some further rationale for this model set-up should be provided. Nowadays (2021) 2.8x2.8 doesn't look very state-of-the art (e.g. look at the Van Dingenen paper (2018), that use a 1x1 resolution. Is the high vertical resolution needed in view of the course horizontal resolution? Why 'free running' (I assume you mean not constrained by (re-) analysed data?)- where there could be clear advantages of putting some constraints- e.g using prescribed SST or nudging. Is the explanation in I. 176?

In terms of resolution, the model we used has a simulation resolution compared to CMIP models from ACCMIP (Lamarque et al. Geosci. Model Dev., 6, 179–206, 2013, www.geosci-model-dev.net/6/179/2013/) as well as for the new runs in ACCMIP, which are currently published. EC-Earth3-AerChem (van Noje et al. 2021, https://gmd.copernicus.org/articles/14/5637/2021/) has a 2° by 3° resolution for chemical tracer transport.

Yes, free running is referring to a climate simulation, where the atmospheric circulation is calculated by the primitive equations and not prescribed by atmospheric winds etc. This is now explained in the text.

176 I have no idea what a QCTM mode is- abbreviations need to be duly explained We repeated the explanation of the abbreviation here again and further referred to the paragraph above where this abbreviation was explained.

173-184. Overall this makes a sound impression, it would help the reader to explain why this is important, and what kind of 'improvements' are found compared to more conventional 'off-line' calculation of radiative forcing.

We reformulated the whole paragraph.

185 MaccCity (if I remember well based on EDGAR3) is pretty old by now- and goes up to2000 (?). I understand that the development of this paper has been taken a while, but there are now inventories like EDGAR5, CEDs that have updated emissions with more recent years.

MACCity provides emission data for anthropogenic and biomass burning for the period 1990 - 2010. It is based on the data set ACCMIP and RCP8.5. In this study, we provide a data set for the LUTs for the year 2010. However, the approach used in TransClim is very flexible. So it is easy to extend the LUT with more emission variation simulations of more recent years.

195 see similar information in Van Dingenen. Thank you for this hint. We added the reference to the manuscript.

198 Can you specific how many simulations are available, and also how the change baseline according to RCP was considered? As a first dimension?

For each emission region, 21 emission variation simulations have been performed. We added this information and an additional table (table 2) to the manuscript. The current setup of TransClim is not able to consider changes of ozone background concentrations. This will be regarded in future studies, as discussed in the conclusions.

247 This synthetic emission is instructive, intuitively I would say that one can still expect problem in Northern Europe (as well as western Europe, Germany) where 'titration' effects can mess up the analysis.

As the evaluation of this test case shows, TransClim reproduces the results obtained by EMAC very well. In the source region Europe, the errors remain below 0.3%. In particular, over Northern Europe, TransClim underestimates the EMAC results at most. But still, the errors are so small that they do not hamper the applicability of TransClim. While titration is an important aspect for air quality assessments, we focus on the climate impact here.

250 What results are you talking about here? SARF globally for one year?

As suggested by reviewer 2, we deleted figure 6. It showed the tropospheric  $O_3$  and  $O_3^{tra}$  columns in Dobson units obtained by TransClim and the relative errors towards EMAC. Now, Figure 5 shows the relative errors between TransClim and EMAC results and figure A1 in the appendix show the absolute values of the tropospheric  $O_3$  column, the tropospheric mean of OH and radiative flux of  $O_3$  at top of the atmosphere as well as the corresponding contributions.

254 are you really discussing O3 or the O3 RF?

The former text was related to  $O_3$  and its contributions of road traffic emission to ozone  $O_3^{tra}$ . We rewrote the whole paragraph.

279 underestimation of what? I am not sure that 7 % deviation is 'very low', this could link to the specification section earlier

We rephrased the whole section.

285 Clarify whether these are transient simulations, equilibrium or something else? VEU1 simulations are equilibrium chemistry simulations for 2030 and the climate response is calculated transient based on the transient changes in emissions and the results of the equilibrium simulations.

296 I think it is a bit confusing to the readers to call it a German emission scenario and consequently apply it to all of Europe. Can the authors expand on the 'robust' signal issue? Is this an apparent drawback of having a model unconstrained by analysed meteorology?

Although the noise of the chemical perturbation is significantly reduced by the QCTM mode of EMAC, it may be still challenging to quantify the climate effect of a small signal resulting from a change in German road traffic emissions by the perturbation method. To avoid this problem, Hendricks et al. (2018) enhanced the signal and perturbed the European transport emissions. The climate response of German transport emissions is determined by downscaling the European response with the ratio of German to European NO<sub>x</sub> emissions. Hendricks et al. (2018) assess this scaling procedure as follows: "Estimating the German effect by this scaling procedure requires the assumption that the radiative forcing per emitted amount of pollutant is similar for the European and the German emissions. Since pollutants released over Europe usually experience vigorous mixing, uncertainties due to this assumption are probably small." We adapted the text.

299 In view of the previous remarks: perhaps for this paper it is not very necessary to highlight the Germany case- it sounds a bit like a 'patch' to me.

Agreed, the procedure might be seen as a 'patch'. The method that we apply here (TransClim) reduces the signal-to-noise ratio. And hence, TransClim is capable of addressing the effects of German emissions, only. Although the global impact is minor, it is still of interest to compare different climate mitigation options. Therefore, we would like to keep this passage. We refer to the differences between European and German emission effects in e.g. table 5. In addition this information is necessary to understand the reply wrt. the referee's comment for line 393.

300 Finally CH4. But what is done with this information? German road traffic emissions influence not only the tropospheric ozone but also the lifetime of methane. Both are important greenhouse gases. We added this intention to the text.

- 322 3 years. Clarify if you mean 3 years from a transient simulation, or what? VEU1 Simulations are equilibrium chemistry simulations for the year 2030. The resulting climate response is determined transient (see in detail comment above).
- 393 'only' is normative language. 24 % seems high. How is it comparing gto the specs? *TransClim reproduces the ozone radiative forcing of European road traffic obtained by EMAC very well. Here, the deviation is only 4 %. But for German road traffic, the deviation is significantly larger (24%). However, this can be taken as a benchmark, because this deviation is caused by the different methods how the climate effect of German road traffic emissions is determined: TransClim determines the climate effect by comparing a simulation with German road traffic emissions to a simulation without German road traffic emissions. In VEU1, the climate effect of German road traffic emissions is determined by downscaling the European climate effect (see also above).*

346-350 Indeed interesting, but unfortunately without explanation. *We added a possible explanation.* 

384 standard computer?

A standard computer describes a work station. This information is already added in section 2.5.

431 Probably a more authorative publication is Van Dingenen et al. (2018), which also extensively describes methodology, error analysis against a range of issues (deviation from linearity, deviation from 'additionality', using a wide range of high/and low end scenarios, and comparison with other literature estimates of similar scenarios. Although the Van Dingenen paper does not give a detailed regional analysis of ozone columns and RF, the analysis shows e.g. for 2030 deviations for summer surface ozone in the order of 4- 9 % for most regions under a high emission scenario and 8-13 % for a low scenario (with an outliers of around 20 %). However, this includes effects of CH4, and by far more regions that in the current study. Interestingly an comparison with a range of publications including results of AR5 showed that FASST was well within a range of other scenario results. Based on the analysis in this paper, I can not support the statement that deviations are 'far less' than from FASST- given the much more limited scope of the evaluation.

Thank you. We added the study of Van Dingenen et al. (2018) to our manuscript. Based on this study and the evaluation given in our manuscript, a direct comparison of the errors is difficult because different metrics are given. As also suggested by reviewer 2, we added more evaluation simulations to our manuscript. These show that TransClim performs well and the deviations from the full global model EMAC remain generally below 10%. We adapted the paragraph.

438 TM5-FASST does include ozone precursors (including CH4)- so it is not clear what is meant with this sentence.

To our knowledge, TM5-FASST does not compute the contributions of a specific emission sector to the ozone concentration using a tagging method. We modified the text accordingly.

440 The study by O. Wild et al. (2012) on the HTAP included a parameterisation of non - linearities of ozone. https://doi.org/10.5194/acp-12-2037-2012, 2012.

Thank you for pointing out this study. We added a paragraph about it to the conclusions.