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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Attribution of ozone radiative forcing trend to individual NO_x sources" *by* K. Dahlmann et al.

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We are thankful for the helpful comments of referee ♯ 2.

General comments
Referee Comment (RC) Radiative forcing is typically defined as the change in radiative balance due to a perturbation in some atmospheric constituent, solar insolation, or surface quantity. Anthropogenic activity indeed imposes a perturbation on atmospheric constituents, and the authors are on safe ground when they compare the radiation fields of pairs of simulations with and without the anthropogenic emissions of industry, road traffic, ships, and air traffic.
Author Comment (AC) We have explained in more detail that our notion of radiative forcing is consistent with that commonly used in the climate research community,

except for the fact that we calculate ozone radiative forcing with respect to 1960 conditions rather than to the preindustrial state. We think that it is justified to provide also radiative forcings arising from changes in natural emissions, as these are part of interactive feedbacks between natural and anthropogenic components as discussed in the paper. Solar insolation, as included in the IPCC reports, is also a natural radiative balance change and is designated as a radiative forcing accordingly. However, we have now distinguished between radiation balance changes and radiative forcing where necessary. Lightning induced NOx emission change and resulting ozone radiative forcing is, strictly speaking, a feedback rather than a forcing, but may be included among the other forcing with the same justification as the second indirect effect of aerosols (change in cloud radiative forcing due to the impact of aerosol emissions on the hydrological cycle).

(RC) But lightning, biomass burning, soil NO_x emissions, stratospheric N2O degradation, and O2 photolysis are all processes that have been ongoing for millennia. Calculating the total radiative impact of any one of these natural processes, as the authors have done here, tells us little and is not interesting. What would be interesting is whether any of these natural processes have changed (or are expected to change) and to what degree these changes have altered the radiation fields and thus climate. The authors begin to examine the issue of change in natural processes, but they do not go very far in this direction.
(AC) We think it is important to analyse the natural sources like lightning, biomass burning and soils along side those of anthropogenic sources for two reasons: 1) On the one hand side we want to show that the same amount of NO_x emissions has very different effect on ozone as well as radiative forcing. For example lightning produces about 100 molecules ozone per emitted molecule NO_x, while industry only produce 10 molecules ozone per emitted molecule NO_x depending on emission region and altitude. In addition the efficiency to perturb the radiation balance shows strong differences between the different NO_x sources. 2) On the other hand anthropogenic effects are superimposed on a varying natural background which influence each other. Thus for example

soils emission show decreasing RF between 1960 and 2019, although the emissions are constant. We tried to point out this in the introduction.

(RC) In the section 'Additivity', the authors appear to calculate radiative forcing due to ozone by comparing the radiation fields of a pair of simulations, one with all the ozone production processes turned on and one without all these processes turned off. (This is, at least, my understanding). In my view, the authors' approach is akin to comparing a pair of simulations with and without any CO_2 present in the model, an exercise with some pedagogical value but not helpful in understanding changes in current climate. The authors do not convince me that such a calculation for ozone has importance.
(AC) We think that the reviewer misunderstood this section. We rephrased to make it clearer. The purpose of this section is to show to which degree the saturation effect, described in Sec. 5.1 limits the additivity of the RF of a number of different components. Commonly the RF of a specific emission is calculated by the difference between RF with and without this emission or between RF with emission and doubling of this emission. In both cases a relatively high background ozone concentration is assumed, as the ozone produced by all other sources is used as background concentration. Due to the saturation effect discussed in Sec. 5.1 this leads to a lower RF than calculating the RF with no ozone background. In this section we want to quantify how large this effect is for the calculation of the RF of all tropospheric sources. Therefore an additional simulation is performed to analyse the RF resulting from the ozone field of all tropospheric sources. As a result we see that the sum of RF of all tropospheric sources is about 10% lower than the RF of the total ozone field.

(RC) It is not clear what value the examination of stratospheric processes adds to the paper. Is the photolysis rate of O_2 in the stratosphere expected to change? Certainly N_2O abundance in the stratosphere is changing in recent decades, but this issue was not addressed.
(AC) It is important to have a consistent simulation of ozone related processes. Hence stratospheric ozone and the input into the troposphere is included in the model. How-

ever, we agree that those processes are not in the focus of this paper. Therefore we followed the referee comments and set aside these results.

(RC) The paper could be salvaged by (1) looking at the impacts of the changes in natural emissions over time
(AC) It is difficult to study the impact of changes in natural emissions over time as only limited information of NO_x emissions exists. But we analysed the changing impact of natural emissions due to anthropogenic emissions.

(RC) . . . (2) examining trends in the different forcings for significance
(AC) To show the significance of changes in ozone columns we added in Fig. 3a the resulting ozone columns of the different ensemble simulations as grey shaded areas. As the ozone columns are global annual means the differences are quit small and the trends in ozone column are significant. The RF results directly from the changes in ozone and are therefore also significant.

(RC) . . . (3) examining the seasonality of the forcing
(AC) This is an interesting point. Our main foci are the longterm changes of ozone and RF. And we agree that the seasonal effect of ozone and resulting RF are interesting. For this we added the seasonal cycle of NO_x emissions, ozone fields as well as radiative efficiencies of the 90s as a example to the supplement material.

(RC) . . . (4) dropping the discussion of stratospheric ozone forcings
(AC) We dropped the discussion of stratospheric forcings

(RC) Fig. 8, which shows the trends in NO_x emissions, ozone production efficiency, and radiative forcing efficiency for the troposphere over the 1960-2019 time period, would be the focus of this revised paper.
(AC) Fig. 9 (former Fig. 8) is the focus of this paper as it is the summary of Sec. 3. The discussion about absolute ozone production efficiencies and radiative efficiencies are important to understand the different impacts of different NO_x emissions.

(RC) Most references in the paper are outdated, from the 1990s or before.
(AC) We added and discussed some new references (e.g. Mickley et al. (2004), Gauss et al. (2006) and Forster et al. (2007))

(RC) Problems in spelling and grammar appear at the rate of 1-3 per paragraph.
(AC) We excuse for having not checked the language carefully enough for the first draft. Extra effort including an internal review has been made now to improve the readability of the text.

Specific comments.

(RC) Abstract. Here and throughout the paper the author need to be clear about how they are calculating radiative forcing: exactly what perturbations in ozone production have they imposed on their model? For example, the text says, 'Lightning . . . causes the highest specific RF [radiative forcing],' but it is not made clear that this is forcing relative to a case with no lightning.
(AC) We have made it clear that we use the concept of radiative consistently with what is common in the climate research community, but that we deviate from the usual notion in calculating ozone radiative forcing with respect to 1960 conditions rather than to the preindustrial state. We think that it is justified to include changes in natural processes in an inter-comparison of radiative flux changes (i.e. radiative forcings), if a part of interactive feedbacks between natural and anthropogenic components of the net forcing.

(RC) Page 16133. In the introduction the authors need to make clear how their work builds on the work of others. The most recent paper cited here is Stuber et al., 2001. More recent relevant papers, such as Unger et al. [2008] and Fuglesvedt et al. [2008], are not brought up till much later in the paper. Papers examining the relationship between ozone forcing and climate, such as Hansen et al. [2005] and Mickley et al. [2004] are neglected.
(AC) We added and discussed some newer references (e.g. Mickley et al. (2004),

Gauss et al. (2006)) and reformulated the introduction.

(RC) Page 16134-16136. The sources of the all NO_x emissions, past and future, need to be supplied. More details about the natural emissions should be supplied.
(AC) We inserted detailed description about the future scenario: *The growth of anthropogenic NO_x emissions is based on economic scenarios of development of GDP (gross domestic product) according to OECD (1997). [. . .] As mitigation measures are under implementation, we force for the future (2000-2019) the development of road traffic NO_x emissions to follow economic growth, represented by growth of GDP combined with total factor productivity (TFP), reflecting technological improvements. Economic scenarios are based on OECD (1997) providing estimates for individual geographic regions of the globe as described in detail in (Matthes, 2003). [. . .] A detailed analysis of lightning emissions in the employed ensemble simulation can be found in Grewe et al. (2009). Other natural emissions follow Dameris et al., 2005*

(RC) Also, I was surprised that the chemistry model omits non-methane hydrocarbon chemistry. The authors need to provide a quantitative estimate of how this omission affects their results.
(AC) We added a discussion about how the omission of NMHCs affect our results in Sec. 4: *'The net effect of the absence of NMHC effects from the simulations presented in this paper yields potentially an underestimation of the resulting ozone change of about 10 % for the air traffic contribution (Kentarchos and Roelofs , 2002). NMHC chemistry is more important for surface based emission sources than for aircraft emissions. However detailed analysis on sensitivity due to individual NMHC chemistry representations have not been performed. Assessing ozone production due to NO_x emissions from road traffic only, captures around 70% of total ozone impact of road traffic (Matthes et al., 2007).'*

(RC) Page 16137. 'An average annual cycle of each ozone field . . . has been taken.' Are these monthly means? It would have been interesting to look at seasonal effects of

the NO_x emissions, since ozone concentration has a strong seasonal cycle over mid-latitudes.
(AC) We used monthly mean ozone fields for the RF calculation. We were interested in the longterm changes of ozone and RF. But we agree that the seasonal effect of ozone is interesting and added the seasonal cycle of NO_x emissions, ozone fields and ozone production efficiencies to the supplement material.

(RC) Page 16139. 'The reason for a higher ozone production efficiency of lightning and air traffic is the higher amount of UV radiance at higher altitudes.' Actually the reason for the high OPE is greater dilution of NO_x at high altitudes.
(AC) We changed this formulation. *'The reason for a higher ozone production efficiency of lightning and air traffic is the higher amount of UV radiance at higher altitudes, lower background concentration of NO_x and the longer lifetime of ozone.'*

(RC) Figures. Captions for all figures should be stand-alone. Where quantities are calculated (and not observed), that should be noted. Global annual averages should be identified.
(AC) The whole paper is about calculated quantities and we think it would be out of scope to note this in each caption. But we identified global annual mean.

(RC) Figure 4 is not necessary, as it appears to present calculations from another work.
(AC) We prefere to keep Fig. 5 (former Fig. 4) as we think it is necessary for better understanding of the work.

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