

## ***Interactive comment on “Attribution of ozone radiative forcing trend to individual NO<sub>x</sub> sources” by K. Dahlmann et al.***

**Anonymous Referee #1**

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### GENERAL COMMENTS

This interesting paper attempts to use a model (E39/C) to attribute changes in ozone radiative forcing over the period 1960–2019 to different NO<sub>x</sub> sources. Whilst I believe the methods employed are scientifically sound and the results will (eventually) be of interest to the atmospheric chemistry community, I cannot recommend publication in its present form. The English is poor in many places, making large sections difficult to comprehend. I have done my best, but this really should have been fixed before submission. It is not my job as a scientific reviewer to fix this.

Scientifically, I raise two major general criticisms that need to be addressed (these are also spelled out in the ‘Specific comments’ section). Firstly, the model employed is a global chemistry-climate model, with stratospheric and tropospheric chemistry. The

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tropospheric chemistry is relatively limited (by comparison to most current tropospheric chemistry models), and includes no treatment of non-methane hydrocarbons. Yet the model is mainly employed here to attribute changes in tropospheric ozone. The limitations of the modelling approach employed need to be clearly stated. It is not at all obvious to me why the sources of stratospheric ozone are presented alongside those arising from tropospheric NO<sub>x</sub> sources. Also, the importance of other ozone precursors, such as methane and CO (never mind NMHCs) is not mentioned.

Secondly, the term radiative forcing is used with a lack of care, which I find surprising given the authors. I am particularly alarmed at the statement in Section 4 (P16144, L2–3) about the ‘total anthropogenic ozone forcing’, and the comparison with the IPCC value for the tropospheric ozone radiative forcing from 1750–2005. It is not at all clear if the values compared are directly comparable as stated, and this needs to be much more carefully described. It has taken the atmospheric chemistry community significant time and effort to clarify how anthropogenic changes in ozone, both tropospheric and stratospheric, contribute to climate change. Publications with statements like this significantly detract from these efforts.

I would like to see the paper eventually published, as the roles of changing efficiencies of ozone production and ozone radiative forcing are interesting topics that have received little attention to date, and I would encourage the authors to sort out the problems with this version of the manuscript so that the community has the opportunity to fully appreciate the significance of their work.

### SPECIFIC COMMENTS

P16132

L23–24: The historical references are mostly about stratospheric ozone, whereas the paper is mostly about tropospheric ozone, which is a bit odd.

L24–25: Perturbations to ozone are thought to represent the third largest radiative forc-

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ing (since pre-industrial times) from a greenhouse gas. It is arguable that indirect human perturbations to water vapour have been more climatically important than ozone changes. The distinction between a radiative forcing GHG and a climatically important GHG is an important one.

P16133

L10: IPCC suggest (see inside front cover of IPCC TAR) that individual chapters are referenced by authors. I'm not sure if this is meant here (and later), or if you did mean to refer to the entire reports.

L13-14: Whilst there is strong evidence that increases in NO<sub>x</sub> emissions have been an important cause of tropospheric ozone increases, there are other important causes, notably increases in CH<sub>4</sub>, CO and NMHCs. Long-term transient modelling studies (e.g., RETRO, Schultz et al., 2007) have typically struggled to simulate observed ozone trends or attribute trends to causes.

L16: 'further ozone increase is expected.' This may be true, but is highly dependent on the future emissions scenario, with some predicting ozone decreases (e.g., see Dentener et al., 2005; Stevenson et al., 2006; Royal Society, 2008).

L18: It is probably also relevant to mention that stratospheric ozone is likely to increase (recover) in future.

P16134

L5: It is not strictly the NO<sub>x</sub> emission altitude that determines the ozone RF, but rather the altitude of the ozone change.

P16135

L4-5: The caveats of using a model with no NMHC chemistry should be clearly stated. Models with no NMHC chemistry are now rare, e.g. all 26 models in the ACCENT inter-comparison (Stevenson et al., 2006) included some NMHC. For example, how does the

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tropospheric ozone budget, burden and lifetime in E39/C compare with the ACCENT models? This is not a plea to discount the E39/C results, as many model features are state-of-the-art (e.g., inclusion of stratospheric chemistry), but just to acknowledge potential deficiencies.

P16136

L13: It is insufficient just to say emissions are 'based on economic scenarios of development of GDP'. More details of the future scenario are needed. Is it documented elsewhere (e.g., IPCC SRES, IIASA etc.), or is it a scenario developed by the authors? What happens to the other ozone precursors (CH<sub>4</sub>, CO)? Are changes in NMHCs considered at all, e.g., as equivalent CO emissions? Future scenarios strongly depend on choices about implementation of air quality legislation, as well as economic growth – is this considered? Some of these factors are mentioned (e.g., catalytic converters in the developed world), but the details should be as clear as possible.

P16137

L1: Similarly for biomass burning emissions – these are said to have 'an almost constant value' – but presumably the seasonal cycle of shifting emissions is represented? Presumably interannual variations (e.g., van der Werf et al., 2003; Schultz et al 2007) in biomass burning (e.g., 1997/8 Indonesian fires) are not included?

L26: It is not only mid-latitude westerlies that transport ozone! Suggest generalise to 'long-range transport'.

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L18-19: I suspect that the higher ozone production efficiency of lightning and aircraft NO<sub>x</sub> is for other reasons in addition to the higher UV at altitude. Possible reasons include a longer NO<sub>x</sub> lifetime and relatively low background NO<sub>x</sub> (compared to the polluted boundary layer).

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L22-25: It isn't clear if the studies referred to are observational or modelling (or both) – the last sentence doesn't make sense.

P16141

L24-25: What is the 'ozone vertical greenhouse efficiencies profile'?

P16143

L1-12: The text describing Figure 8 (including its caption) is not at all clear. I think it is interesting, but I struggled to follow what had been done here.

P16144

L2-3: I am alarmed to see a value for the 'total anthropogenic ozone forcing' of 0.52 W/m<sup>2</sup>, which is then compared to the IPCC estimate of tropospheric ozone forcing (from 1750 to 2005). This needs much more explanation if it is to remain in the paper. Does 'total' mean tropospheric plus stratospheric, and if so, why is it being compared to the IPCC tropospheric value? Is the value for 1750 to 2005, and if not, why is being compared to an IPCC value for this time period? If it is 1750 to 2005, then why is there no description whatsoever of the 1750 model ozone field? Most past studies of this carefully describe what they consider to be 'anthropogenic' – for example it is rather uncertain what biomass burning and soil emissions (which are partly natural and partly anthropogenic) were in 1750.

L8-9: What does 'an agreement in the range of 25%' mean?

L19-21: This sentence does not make sense.

P16145

L20-22: I don't understand this sentence, or much of Section 5.2.

P16146

L1-9: As above, I don't really understand this. Are you saying one method (individual

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components vs summed) is better than the other?

Figure 1: Do Africa and China really share the same NO<sub>x</sub> emissions growth rate?

Figure 2: Would it make more sense to show tropospheric ozone column, rather than total column? It would also be interesting to know what fraction of tropospheric ozone has its source in the stratosphere. 'NO<sub>2</sub>-degradation' in both the figure and the caption should read N<sub>2</sub>O degradation! 'O<sub>2</sub>-Photolyse' should read O<sub>2</sub> photolysis. (Both these mistakes are also in Figure 7)

Figure 4: What do the 2-D and 3-D bars refer to? They aren't described in the caption.

Figure 5: State these are ozone RFs, not total RFs for the named sectors! It should also be clarified that these are RFs due to the occurrence of these NO<sub>x</sub> emissions, as opposed to the commonly used definition of RF (e.g., IPCC) which refers to the change in a component over a time period (typically 1750-2005, or pre-industrial to present-day). Clearly, lightning was still producing NO<sub>x</sub> in pre-industrial times, so it is not a significant component of the pre-industrial to present-day forcing (although it potentially could be, if the emissions have changed, and/or their ozone production efficiency has changed).

Figure 6: As for Figure 5, state these are ozone RF efficiencies.

Figure 7: (See Fig.2). The term 'stratospheric production' is used for N<sub>2</sub>O degradation. Isn't O<sub>2</sub> photolysis and subsequent O<sub>3</sub> formation also stratospheric production?

Figure 8: The caption is particularly misleading. As far as I can tell, these are percentage changes in ozone RF from the 1960s to the 2010s, split by cause. I don't understand how the split is calculated.

Figure 9: State it is ozone RF. Capitalise LW, SW. Similarly for Fig. 10.

#### TECHNICAL CORRECTIONS

There are too many to list – mainly to do with poor English. Here are a few:

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P16133, l4-5: primarily through the reactions  
P16133, l29: enables US  
P16134, l8: levels -> altitudes  
P16134, l12 (and elsewhere): area -> era  
P16134, l20: adding -> the additivity of  
P16134, l25: derivate -> derivative  
P16135, l9: lighting -> lightning  
P16138, l1: resulting FROM  
P16138, l4: primary -> primarily  
P16138, l5: to -> for  
P16138, l6: delete 'rather', and a colon shouldn't (necessarily) be followed by a capital (many places)  
P16138, l12: photolyses -> photolysis (many places)  
P16138, l14: takes -> take  
P16138, l26: just equal -> similar  
P16139, l21 and l22: of -> on  
P16139, l26: at last -> eventually  
P16141, l8: relative -> relatively  
P16142, l3: delete 'may'  
P16142, l22: because OF  
P16142, l2: this -> these

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P16145, l6: of -> off  
P16145, l20: calculated inverse?  
P16146, l7: totalised?  
P16146, l26: relative -> relatively

#### REFERENCES

Dentener et al. (2005) Atmos. Chem. Phys., 5, 1731-1755.  
Royal Society (2008) Ground-level ozone in the 21st century: future trends, impacts and policy implications. (See [royalsociety.org](http://royalsociety.org))  
Schultz et al (2007) RETRO Final Report. (See [retro.enes.org/reports](http://retro.enes.org/reports))  
Stevenson et al. (2006) J. Geophys. Res., 111, D08301, doi:10.1029/2005JD006338.  
van der Werf et al. (2003) Global Change Biol., 9, 547-562

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