Atmos. Chem. Phys. Discuss., 9, C10157–C10166, 2010
www.atmos-chem-phys-discuss.net/9/C10157/2010/

Atmospheric Chemistry and Physics Discussions

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

K. Dahlmann et al.

katrin.dahlmann@dlr.de

Received and published: 27 January 2010

We are thankful for the helpful comments of referee ♯ 1 and followed all his recommendations, except that we still show total ozone columns and not tropospheric columns in Fig. 3 (former Fig. 2) since the differences are minor for anthropogenic sources.

General Comments:
Referee Comment (RC) The English is poor in many places, making large sections difficult to comprehend.
Author Comment (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.

(RC) The model employed is a global chemistry-climate model, with stratospheric and tropospheric chemistry. The 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.
(AC) In Sec. 2 we described the model. We have now introduced a discussion on the advantages and disadvantages of the employed model. *'The effect of NMHC chemistry on ozone concentration changes of e.g. road traffic and air traffic have been described in Matthes (2003) and Kentarchos and Roelofs (2002), respectively. The NMHC chemistry is important in the boundary layer and gets less important in the free troposphere. As we analyse the whole troposphere and stratosphere the NMHC chemistry are not that important and their inclusion would have been too demanding in terms of computational resources.'*

(RC) It is not at all obvious to me why the sources of stratospheric ozone are presented alongside those arising from tropospheric NO_x sources.
(AC) It is important to have a consistent simulation of ozone related processes. Hence stratospheric ozone and the input into the troposphere are included in the model. However, 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) Also, the importance of other ozone precursors, such as methane and CO (never mind NMHCs) is not mentioned.
(AC) We introduced the information about other important ozone precursors and their emission. *'The global increase of tropospheric ozone abundance has mainly occurred as a consequence of rising anthropogenic emissions of various ozone precursor species (NO_x, CO, CH_4, NMHCs). [. . .] Surface concentrations of CO_2, CH_4, N_2O, CO, and 2-D background concentrations of CFCs are taken into account. Carbon monoxide surface concentrations are kept constant over the simulation period, since a clear trend*

over this period is not established Dameris et al. (2005). Methane surface concentrations are included with a trend, based on observations. Fig. 1 shows the calculated implicit emissions of CO and CH_4, which can be deduced from their boundary conditions. [. . .] The absolute values and also the trend in CH4 emissions are comparable to emission estimates [. . .]. Whereas the decrease in CO emissions reflect a decrease in CO and CH4 lifetime due to a simulated OH increase. The change of the CH4 lifetime is comparable to previous studies. [. . .]'

(RC) Secondly, the term radiative forcing is used with a lack of care.
(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. We have now distinguished between radiation balance changes and radiative forcing where necessary.

(RC) 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.
P16144 **(RC)** L2-3: I am alarmed to see a value for the 'total anthropogenic ozone forcing' of 0.52 W/m^2, 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.
(AC) The referee is right. The comparison of both values was not described carefully enough. We rephrased the sentences as followed: *'The IPCC estimate of anthropogenic ozone forcing since pre-industrial times is 0.35 W/m^2 with an uncertainty range of 0.25 to 0.65 W/m^2 (Forster et al., 2007). This RF value includes the effect of all anthropogenic precursor emissions on tropospheric ozone that have occurred between 1750 and 2005 under the assumption of zero anthropogenic emissions and biomass burning sources reduced by 90% for the pre-industrial time (1750). Here we calculate the radiative balance change between contributions from anthropogenic NO_x emissions (industry, ship, road and air traffic) for the decade preceding 2000 and no anthropogenic NO_x emissions, in contrast to our previous definition of RF since 1960. In addition, we consider 90% of the biomass burning emissions and the respective RF to be of anthropogenic origin. Both approaches are comparable although they differ in detail. With our definition we yield a tropospheric ozone radiative balance change of 0.64 W/m^2 for the decade preceding 2000, a value at the upper boundary of the IPCC uncertainty range.'*

SPECIFIC COMMENTS
(RC) P16132 L23-24: The historical references are mostly about stratospheric ozone, whereas the paper is mostly about tropospheric ozone, which is a bit odd.
(AC) We added and discussed some further references about tropospheric ozone (e.g. Mickley et al. (2004) and Gauss et al (2006))

(RC) P16132 L24-25: Perturbations to ozone are thought to represent the third largest radiative forcing (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.

(AC) We changed this sentence into: *'At the beginning of the 21st century the contribution of tropospheric ozone to anthropogenic radiative forcing since pre-industrial times was about 0.35 W/m^2 (Mickley et al., 2004, Gauss et al. ,2006), making it the third most important greenhouse gas perturbed by human activity, next to carbon dioxide (CO_2) and methane (CH_4).'*

(RC) 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.
(AC) We changed the reference accordingly.

(RC) P16133 L13-14: Whilst there is strong evidence that increases in NO_x emissions have been an important cause of tropospheric ozone increases, there are other important causes, notably increases in CH4, 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.
(AC) We introduced the information about other important ozone precursors. The impact of other ozone precursors and their trends is subject to future modelling studies with an updated chemistry.

(RC) P16133 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).
(RC) P16133 L18: It is probably also relevant to mention that stratospheric ozone is likely to increase (recover) in future.
(AC) We have generalised this statement: *It is expected that proceeding emissions in the next decade will lead to further ozone increases in the troposphere, but will depend on development of future technologies (e.g. Dentener, 2005). By contrast the ozone amount in the stratosphere has decreased [. . .] but is expected to recover during the forthcoming decades (Eyring et al, 2007b), which impacts tropospheric ozone (Grewe et al., 2007).*

(RC)P16134 L5: It is not strictly the NOx emission altitude that determines the ozone RF, but rather the altitude of the ozone change.
(AC) We think that this point had been captured correctly already in the draft version.

(RC) 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 intercomparison (Stevenson et al., 2006) included some NMHC. For example, how does the 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.
(AC) We added two references which deal with the effect of higher hydrocarbons on ozone concentrations and included the following text: *'The effect of NMHC chemistry on ozone concentration changes of e.g. road traffic and air traffic are described in Matthes (2003) and Kentarchos and Roelofs (2002), respectively. The NMHC chemistry is important in the boundary layer and gets less important in the free troposphere. As we analyse the whole troposphere and stratosphere the NMHC chemistry are not that important and their inclusion would be very calculation time intensive.'*

(RC) 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? 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.
(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)'

(RC) What happens to the other ozone precursors (CH_4, CO)? Are changes in NMHCs considered at all, e.g., as equivalent CO emissions?
(AC) NMHCs are not considered at all. Emissions of CH_4 and CO are only considered implicit as surface concentrations. We have added a description of this implicit emissions in Sec. 2. *'Surface concentrations of CO_2, CH_4, N_2O, CO, and 2-D background concentrations of CFCs are taken into account. Carbon monoxide surface concentrations are kept constant over the simulation period, since a clear trend over this period is not established Dameris et al. (2005). Methane surface concentrations are included with a trend, based on observations. Fig. 1 shows the calculated implicit emissions of CO and CH_4, which can be deduced from their boundary conditions. [...] The absolute values and also the trend in CH4 emissions are comparable to emission estimates [...]. Whereas the decrease in CO emissions reflect a decrease in CO and CH4 lifetime due to a simulated OH increase. The change of the CH4 lifetime is comparable to previous studies. [...]'*

(RC) 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?
(AC) The referee is right, the seasonal cycle of shifting emissions is represented but interannual variations are not included.
'Natural sources, lightning and soils as well as biomass burning display only a small trend in emissions during the time period considered here. These emissions include a seasonal cycle but with the exception of lightning no interannual variability.'

(RC) It is not only mid-latitude westerlies that transport ozone! Suggest generalise to

'long-range transport'.
(AC) We changed this accordingly.

(RC) P16139 L18-19: I suspect that the higher ozone production efficiency of lightning and aircraft NO_x is for other reasons in addition to the higher UV at altitude. Possible reasons include a longer NO_x lifetime and relatively low background NO_x (compared to the polluted boundary layer).
(AC) We agree. These are also reasons for higher ozone production in higher altitudes.
'The reason for a higher ozone production efficiency of lightning and air traffic are the higher amount of UV radiance at higher altitudes, lower background concentration of NO_x and the longer lifetime of ozone.'

(RC) P16140 L22-25: It isn't clear if the studies referred to are observational or modelling (or both) - the last sentence doesn't make sense.
(AC) We reformulated this sentence and added the information that the studies are observational
'This is consistent with satellite measurements of Uno et al. (2007) and Konovalov et al. (2008), which show increasing NO_x emissions over Eastern Asia and decreasing NO_x emissions over western and Central Europe respectively.'

(RC) P16141 L24-25: What is the 'ozone vertical greenhouse efficiencies profile'?
(AC) was deleted

(RC)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.
(AC) We changed the formulation of this text.
'The time development of ozone RF is the product of changes in NO_x emission, ozone production per NO_x emission and radiative efficiency (RF per ozone change). To analyse the reasons for the changes in RF between 1960s and 2010s displayed in Fig. 6 (former Fig. 5), we present the percentual changes in NO_x emission (E), ozone production per NO_x emission ($\frac{O_3}{E}$) and radiative efficiency ($\frac{RF}{O_3}$) between 1960s and 2010s

in Fig. 9 (former Fig. 8), by splitting up the individual trend terms:

$$\frac{RF^{2010}}{RF^{1960}} = \frac{E^{2010}}{E^{1960}} \times \frac{\frac{O_3^{2010}}{E^{2010}}}{\frac{O_3^{1960}}{E^{1960}}} \times \frac{\frac{RF^{2010}}{O_3^{2010}}}{\frac{RF^{1960}}{O_3^{1960}}}$$

,

(RC) L8-9: What does 'an agreement in the range of 25%' mean?
(AC) We changed the formulation to: *'differences lower than 25%'*

(RC) L19-21: This sentence does not make sense.
(RC) L20-22: I don't understand this sentence, or much of Section 5.2.
(RC) P16146 L1-9: As above, I don't really understand this. Are you saying one method (individual components vs summed) is better than the other?
(AC) We changed the formulation of the whole section.

(RC) Figure 1: Do Africa and China really share the same NO_x emissions growth rate?
(AC) Yes, they have the same emission growth rate. *'Thus for example Africa and China have the same emission growth rates, although China has a much higher GDP growth but also a much higher TFP.'*

(RC) 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.
(AC) Fig.3 (former Fig. 2) is only meant to give an overview of the geographical distribution of the resulting ozone fields. We still want to show total ozone columns and not tropospheric columns since the differences are minor for anthropogenic sources. The question of what fraction of tropospheric ozone has its source in the stratosphere we refere to Grewe et al., 2006.

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

(AC) We deleted this part of the figure. ('typical NO_x range of 2- and 3-D model calculations for the considered location is indicated by the thick solid bars.' (Grooß et al., 1998).)

(RC) 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_x 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_x 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.
(AC) Fig. 6 (former Fig. 5) was changed according to the new definition of RF in this paper. We inserted that this are ozone RFs, not total RFs for the named sectors.

(RC) 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.
(AC) We have reformulated the caption and the description in the text.

Technical correction:
We fully agree.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 16131, 2009.