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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Global impact of road traffic emissions on tropospheric ozone" *by* S. Matthes et al.

Anonymous Referee #1

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@@@ << General Comments >>

In this manuscript , the authors assess the impact of road traffic emissions on tropospheric ozone with a global chemical model. As the authors state, there have been only limited number of studies on road traffic emissions on ozone. As far as I know, the study by Granier and Brasseur [2003] is the first to investigate the role of road traffic emissions on global ozone. This manuscript similarly gives a quantitative evaluation of road traffic impacts on global tropospheric ozone and can be regarded as an important addition to the previous work [Granier and Brasseur, 2003]. This manuscript also tries to separate the impacts of NOx and NMHCs road traffic emissions. This is a reasonable focus in assessing the impact on ozone, since the NOx/NMHCs ratio is a key

factor of ozone production in source regions and NMHCs can also play an important role in remote ozone production. Furthermore, the authors discuss individual contributions from road traffic emissions in N-America and Europe as well. These points can be regarded as novel aspects of this manuscript and advances from the previous works.

The methodology (experimental setup) to evaluate the impacts of road traffic emissions of NOx and NMHCs appears to be designed reasonably; I am, however, a little concerned about linearity between NOx and NMHCs impacts on ozone which is assumed by the authors. The authors simulate in this study tropospheric ozone using GCM driven meteorology not the observational data like ECMWF. Since GCM can cause significant biases in meteorological variables such as temperature, vertical wind, and PBL regimes, this makes it difficult to compare this study with Granier and Brasseur [2003] which use the ECMWF analyses.

Given still unknown aspects of anthropogenic impacts on tropospheric ozone, this kind of study is useful and informative to understand the current situation of tropospheric ozone, and could also be used for future prospect. The subject of this paper appears to be appropriate to the ACP. However, I would like the authors to consider my questions and revise the manuscript before I recommend the publication of this paper. Details of my comments will be found in the following.

(*) I feel, English usage in this manuscript is generally acceptable, but not so good. Also, several sentences and paragraphs are poorly constructed. I recommend for the authors to have this manuscript scanned by a native English speaker.

@@@ << Specific Comments >>

The manuscript is generally well organized and competent. The methodology is clearly described with the acceptable number of references to earlier works.

Abstract (block 10340): "Improving over previous global modelling studies, which con-

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centrated on road traffic NOx and CO-emissions only,..." Does this mean Granier and Brasseur [2003] ? I think that they included road traffic NMHCs emissions as well as NOx and CO.

"It is revealed that NMHC-emissions from road traffic play a key role..." Please describe what kind of NMHC impact on ozone did the authors find quantitatively. That should be one of the main points of this paper.

"There, during subsidence, PAN acts as a source for NOx, caused by thermal decay. Hence, ozone is produced" –> this does't appear necessary.

"Sensitivity Studies for regional emission show ..." Here, the authors could present the individual impacts of N-American and European emissions which are discussed in Section 4.4.

1.Introduction (block 10341):

line-19: ".., global emissions are still supposed to grow in the future (OECD, 1995)" The authors should include the IPCC-TAR (2001) as a reference here.

line-23: "... E.g., Granier and Brasseur (2003) investigated the impact of NOx and CO emissions from road traffic...." Granier and Brasseur (2003) evaluate the total impact of road traffic emissions on tropo. ozone including NMHCs emissions as well as NOx and CO.

2.Model,emissions,and experimental setup (block 10342):

Please give information on the NOx sources from lightning and soils in your model with global amounts in TgN/yr. And, how do you include the natural (biogenic) emissions of NMHCs; how much did you include them? (TgC/yr). Presumably, these parameters significantly influence the global ozone sensitivity to road traffic emissions in this study.

(block 10343) last paragraph:

The authors consider several types of simulations to isolate the impacts of road traf-

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fic NOx/NMHCs emissions and N-American/European emissions. Especially for their NOx/NMHCs impacts, presumably, they assume a linearity between NOx and NMHCs impacts; i.e., (total O3 change) = (O3 change from NOx) + (O3 change from NMHCs). The authors should mention this point in the text.

The authors use in this study meteorology calculated by a GCM. This causes a difficulty in evaluating NO2 column with the GOME data and in comparing their results with Granier and Brasseur (2003), since there are non-negligible differences (biases) in meteorology between GCMs and actual observation. Also, the authors state that they performed four years simulations for the individual scenarios. Are the meteorological conditions for those four years exactly the same for each of scenarios (CTR90, No_rt, No_NOx, No_NMHC, etc) ? If the meteorology is not identical in all the scenarios, it may be difficult to evaluate quantitatively the pure impacts of individual emission sources.

3.Comparison with observations (block 10344)

The authors compare their simulated tropospheric columns of NO2 with the GOME data as an evaluation of their model. This comparison, however, is not an effective way to evaluate their model simulation. Since there are still great uncertainties in retrieval of the GOME NO2 data, the authors may not be able to draw any definitive/quantitative conclusions from their comparison with the GOME data. The authors should compare their simulated NOx distributions with direct observation like NASA GTEs and surface observation in U.S. and Europe rather than the satellite data. Additionally, in view of the focus of this paper, the authors should evaluate their simulation of tropospheric ozone, PAN, and NMHCs as well as NOx (with sonde/aircraft/surface measurements) to make their main results robust.

line-25: "In Fig.3 additional results from model studies including a chemistry scheme which does not include higher hydrocarbons (ECHAM4.L39(DLR)/CHEM) are given" Results from the ECHAM4.L39/CHEM with a different vertical resolution are not

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needed in this paper. Please remove them to avoid confusion.

(block 10345) line-01: "This corresponds to a stronger reduced vertical mixing in the boundary layer in wintertime" This is also related to a longer lifetime of NOx in wintertime, right?

Fig.3. (block 10361): Do the model results represent NO2 columns at the same local time as the GOME (LTC10:30) ? Is it possible to include comparison of NO2 in European region ?

4.Results:

(block 10347) line1-4 "In the southern hemisphere(SH), ... ,respectively" The meaning of this sentence is not clear. The authors could better present it.

line16- "The road traffic effect to the SH ozone budget in summer appears overproportionally high ..." First, what does "SH ozone budget" mean? isn't it simply "SH ozone"? Anthropogenic emissions (including road traffic) in SH are originally much lower than those in NH. So, even if "road traffic NOx emissions in southern hemisphere amount to only about 3% of the global total", the response shown in Fig.4 is quite natural, not overproportinal.

(block 10348) line2-4 "In source regions, these results are comparable to the findings of Granier and Brasseur (2003), who calculated about the same relative contribution (15%)." Granier and Brasseur (2003) show in fact relative contributions of only 10-15% in source regions which are lower than this study.

line4-8 "However, a remarkable difference occurs in non-source regions where their calculation showed lower relative contributions of 6 to 9%, only. Looking at the impact of individual road traffic emission compounds, the origin of this difference can be attributed to the neglection of NMHC road traffic emissions" Granier and Brasseur (2003) also include NMHC road traffic emissions. So, these sentences do not make any sense. The results of Granier and Brasseur (2003) are just systematically lower

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than those of this study (not only for non-source regions), which is obviously attributable to the difference in model's sensitivity of ozone production"

4.2 Importance of individual emissions for ozone

(block 10349) line9-11 "In January, road traffic NOx emissions even lead to ozone decrease in source regions, caused by a dominating decrease of ozone productivity of NOx with increasing NOx concentrations." This seems to indicate ozone destruction by NOx (titration) rather than explicit ozone productivity, since ozone production in wintertime is not so effective in the mid-high latitudes.

line14-19 "In these strongly confined regions, this results in ozone production and an ozone increase due to road traffic NMHC emission of about 12%. ..." What do "these strongly confined regions" indicate ? and in what regions ozone increase (12%) due to road traffic NMHC is calculated ? Does this paragraph describe NMHCs impacts on ozone for July ?

line20-23 "In January the mechanism remains the same, but acts on a lower level due to slow photochemistry. Northern hemisphere regions in which ozone titration occurs are larger, as well as regions, where NMHC emissions cause an increase in ozone (North America, East Europe)." I think that the NMHC induced ozone changes (Fig.5 right) in January and July are caused by different mechanisms. Ozone increases in N-America and Europe in January seem to result from decrease in ozone destruction by NOx which is coming from lower NOx level associated with PAN formation. To make these points clear, I recommend for the authors to show the changes in ozone production term P(O3) or net production rate P-L(O3) in ppbv/day (or %) for RT, RT_NOx, and RT_NMHC individually in a format similar to Fig5. Such pictures should also demonstrate the NMHC induced ozone production in remote areas associated with long-range transport of PAN, which the authors suggest and discuss in the next section (4.3).

(block 10350) line11-13 "Generally, about 70% of the total ozone increases is cased by NOx emissions from road transport at latitudes, where the main sources are lo-

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cated, and in the free troposphere." This seems to assume a linearity between NOx and NMHCs impacts. Is it really valid? In another words, can the authors reproduce the total ozone changes in Fig.4 (bottom) by summing the individual component shown in Fig.6 ?

line16-17 "Ozone contributions of about 6%, are found in remote regions (e.g., arctic latitudes)." 6% contributions are confined only in the arctic regions (Fig.6). So, "found in remote regions (e.g., arctic latitudes)" should be "found in the arctic regions".

line20- "The mechanism for the long-range impact of NMHC emissions from road transport is additional PAN formation (see Sect. 4.3)." Is it the only mechanism ? How about the direct impact of increased ozone production in source regions ? Even though percentage impacts of NMHC on ozone in source regions appear to be only small, they should have a significant impact on remote ozone levels.

(block 10351) line8- "As will be shown in the next section (Sect. 4.3) the mechanism for long-range impact of NMHC emissions is transport of additionally formed PAN, ..." Again, is it the only factor ?

line12-16 "Our results largely agree with those of Granier and Brasseur (2003), when comparing their total road traffic impact (NOx and CO only) to our .... As Granier and Brasseur (2003) did not account for road traffic NMHC emissions, they underestimated the total impact of road traffic emissions on ozone...." Granier and Brasseur (2003) did account for road traffic NMHC. These sentences do not make any sense. The differences between those two studies should be explained by the different sensitivities of chemical scheme to emission changes, and the different meteorology used in the two models.

line16-19 "The sensitivity experiment for NMHC emissions ..." These two sentences just repeat the same content as already mentioned before. So, the authors should remove the last paragraph "Our results largely ..." (line12-19)

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(block 10352) line1 "in remote regions of more than 6% (Fig.6, middle row)." This should be modified as "in arctic regions of more than 6%"

line12- "Hence, NMHC emissions are responsible for about 90% of PAN enhancement in winter and springtime in arctic latitudes (PN)." Again, the authors appear to assume a linear effect of NOx and NMHCs emissions on PAN. For example, in Fig.7, obviously "NHE-NMHC + NHE-NOx" is not equal to "NHE"(total). The author should make this point clear.

line18-19 "Hence Fig.7 illustrates that road traffic NMHC-emissions are crucial for the formation and long-range transport of PAN which then causes ozone contribution of road traffic in remote regions." To make this suggestion more robust, the authors should show the changes in NOx levels in remote areas due to traffic NMHC emissions (RT_NMHC) just like Fig.7; i.e., dNOx [pptv].

Before Section 4.4; I recommend for the authors to include the information of global budget of tropospheric ozone for individual scenarios (RT, RT_NOx,...): global burden (TgO3), production/destruction (TgO3/yr), surface deposition (TgO3/yr), Stratosphere/Troposphere Exchange (TgO3/yr), and lifetime of O3 (days). Hopefully, the authors could do this by putting an additional table.

4.4 Impact of regional emissions from the USA and Europe

In this section, the authors could include references to the previous intercontinental studies: e.g.,

Wild, O., H. Akimoto, Intercontinental transport of ozone and its precursors in a three-dimensional global CTM, J. Geophys. Res., 106(D21), 27729-27744, 10.1029/2000JD000123, 2001.

Wild O., P. Pochanart, H. Akimoto (2004), Trans-Eurasian transport of ozone and its precursors, J. Geophys. Res., 109, D11302, doi:10.1029/2003JD004501.

5. Summary and conclusions

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(block 10355) line3- "For assessing the climate impact of road traffic emissions NMHCs have to be considered." How about the indirect impacts from NMHCs and CO on OH and CH4 ? These are also keys for assessing the climate impact.

@@@ << Technical Corrections etc. >>

(block 10342) line04: "evaluatex" –> "evaluated"

line18: "model physics are" –> "model physics is"

(block 10344) line19: ", these conditions can differ quite substantially different between individual periods from modelling studies and observations." ??? (need correction)

(block 10346) line28: "(indicated by larger atmospheric regions with contributions above 50% are larger)" ??? (need correction)

(block 10347) line17: "NOx emissions in southern regions" –> "NOx emissions in the southern hemisphere"

(block 10348) line20: "in southern latitudes" –> "in the southern hemisphere"

line24: " relative contributions or more than" –> " relative contributions of more than"

(block 10349) line06: "(left, already discussed in the last sub-section)" –> remove this.

(block 10350) line13: "Zonal gradients are ..." –> "Meridional gradients are ..."

line25: "... in noteworthy" –> "... is noteworthy"

(block 10352) line29: "n source regions" –> "In source regions"

(block 10355) line04: "Regional studies have ..." –> "Our regional studies have ..."

Fig.5 (block 10363) Please change the color scheme; use warm colors (e.g., yellow-red) for positives, and cool colors (blue-green) for negatives.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 10339, 2005.