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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.

S. Matthes et al.

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1 General remark:

The two anonymous referees gave a number of constructive comments, which we all tried to include into our revised manuscript.

2 Detailed Response to Referee #1 - 14 November 2005

General comments:

The referee is concerned about assumed linearity between NOx and NMHC impacts on ozone.

Answer: In the work presented, no linearity is assumed. Therefore we think that the referee has the impression that the total impact is calculated by adding the impact of the individual compounds (nitrogen oxides, CO, hydrocarbon). In contrary separate calculations were performed for the calculation of total ozone. For each chemical component (nitrogen oxides, CO, hydrocarbons) an individual scenario simulation was performed which was then compared to the reference simulation. Such a scenario simulations contains all emissions as boundary conditions except those road transport emissions of the considered compound. This approach has been chosen to account for non-linearities. One additionally scenario simulation has been performed where all emitted compounds from road transport were not included. These scenario simulations confirmed the non-linearity of the simulated chemistry, as the summed impact of individual components shows higher ozone changes than the impact of all road transport emissions. As this does not become clear in the current manuscript, we clarified this point in the revised version at the beginning of Section 4.2.

Second, the referee suggests that the major reason for differences of results from this study compared to the study of Granier and Brasseur [2003] could be the different meteorological variables between GCM study and ECMWF analyses.

Answer: We agree that a direct intercomparison between Granier and Brasseur and our work needs particular attention. However we consider both approaches reasonable leading to comparable results for the impact of road transport emissions on ozone. Both approaches have advantages and disadvantages. Using a GCM allows a consistent description of atmospheric processes. Comparing temperatures in our GCM with observational data showed that in July and January deviations of maximum 1K and 2K are found in the lower troposphere, respectively. For other quantities, e.g. vertical wind, and PBL ventilation no global data set is available which could allow a sound

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intercomparison.

Both studies consider multi-annual average results minimizing the influence of inter-annual variability. Granier and Brasseur use a multi-year average meteorology, while we average 4 consecutive years of simulation to calculate changes in ozone concentrations.

We agree, that a comparison has to be done carefully, nevertheless we think that those differences found between Granier and Brasseur and our study originate mainly from the different representation of road transport emissions, and not from different meteorologies used. Granier and Brasseur did not consider NMHC emissions from road traffic in their study. This is based on sensitivity studies which revealed that neglecting NMHC emissions leads to those patterns of ozone impact presented by Granier and Brasseur. Consequently we address this issue in more detail in section 2 and 4 of our manuscript.

Specific comments:

Abstract (block 10340): The referee asks for clarification if Granier and Brasseur [2003] did include road traffic NMHCs emissions in their model calculations. Within their article Granier and Brasseur describe in detail in the Emissions section nitrogen oxide and carbon emission from road traffic. For those two compounds they indicate global emitted amounts and relative contributions to total surface emissions. In the case of NMHCs they do not specify any fraction from road transport. As an inclusion of NMHC emission in a chemistry scheme is not a trivial issue and needs assumptions, a description of the selected procedure would be heavily required if NMHC emissions would have been considered. From this description of emissions methodology we conclude that no NMHC emissions from road traffic were included in detail. Additionally we contacted the authors who confirmed this.

The referee suggests a more quantitative description of the impact of NMHCs. For this reason the following sentence was included in our abstract: In arctic latitudes NMHC

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emissions from road transport are responsible for about 90% of PAN increase from road transport, leading to a contribution to ozone concentration of up to 15%.

Technical change for shortening of the manuscript is followed.

Introduction (block 10341): Referee comments are considered. The additional reference was included, i.e. Special Report on Emission Scenarios.

Referee asks for clarification of emissions considered by Granier and Brasseur. Clarification is provided according to above discussion (see abstract section).

Model, emissions, and experimental setup (block 10342): The referee asks to provide more specific information on NOx emissions from soils and lightning, and also the amounts of biogenic NMHC emissions (in Tg/yr). We included the following sentence: Anthropogenic NOx emissions amount to 27.6 Tg and include 5.0 Tg from biomass burning [Hao et al., 1994] and 27.6 Tg from fossil fuel combustion [Benkovitz et al., 1996]. Natural NOx emissions from soils and lightning account for 5.5 Tg and 5.0 Tg, respectively. NMHC emissions from industry/traffic, biomass burning and vegetation (isoprene) add up to 90 Tg [C], 18 Tg [C] and 400 Tg [C], respectively.

(block 10343) last paragraph: The referee has the impression that the total impact is calculated by adding the impact of the individual compounds (nitrogen oxides, CO, hydrocarbon).

Answer: Above comment is addressed as first general comment in detail, where we clarified that no linearity is assumed.

The referee highlights the difference in meteorology in simulation and observation year with respect to the comparison to the observations. Differences caused by different meteorology are addressed in the next section (block 10344).

Further the referee asks for clarification whether the meteorology in both scenario and control experiments are identical. Answer: Meteorology in both simulations is identical as the calculations are performed without feedback mechanisms from chemical

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changes to the climate system, but driven by constant, identical boundary conditions.

Comparison with observations (block 10344): The referee suggests to further evaluate ozone, PAN, NMHCs and NOx of our modelling system.

Answer: As the system was intensively evaluated within earlier papers, we provide a short overview on respective results. The following paragraph was included: The modelling system used in our study was intensively evaluated within earlier papers, the most prominent being Roelofs and Lelieveld [2000]. They showed that seasonality of surface CO and PAN agrees well with observations. Some differences occur associated with representation of biomass burning emissions (CO) and reduced variability in model simulations (PAN). Additonally they compare ozone station data and sonde data with model values for surface, lower, middle and upper troposphere, showing well reproduced seasonal cycle at all altitudes.

Additionally the referee suggests to use different observational NOx data, e.g. sonde data.

Answer: We use the satellite GOME NOx data as GOME observational data provides global maps of nitrogen oxides. Such global maps allow the analysis of patterns in trace gas distributions, and the analysis of the models ability to capture these general patterns.

line-25: The referee suggests removing results from the ECHAM4.L39/DLR)/CHEM to avoid confusion.

Answer: The inclusion of the earlier model system E39/C helps to understand the impact of NMHC chemistry, which is only included in CBM-IV. The issue of a higher model resolution at the tropopause might be not that important. The seasonal cycle is not influenced by NMHC chemistry. We think this information is helpful to model evaluation. Therefore we kept the figure, but clarified the text. We included: Comparison shows that higher hydrocarbon chemistry in these regions mainly reduces atmospheric NO2

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columns.

(block 10345) line-01: The referee asks whether reduced vertical mixing corresponds to a longer lifetime of NOx in wintertime.

Answer: Yes, this is related to a longer lifetime of NOx in wintertime.

Fig.3 (block 10361): The referee suggests to clarify at what time NO2 columns from the model are extracted.

Answer: The model provides 24 hour averages of NO2. In Lauer et al. [2002] it was shown that for those regions shown in our paper, local time of extracting NO2 columns from the model does not influence the results with respect to the seasonal cycle. However, total amounts may be overestimated by 20% in Europe and 30-50% in Africa [Martin et al., 2002]. Hence, NO2 columns were extracted as monthly means (based on 30 min values). We include the following sentence We used monthly mean NO2 values averaged from half hour values, as Lauer et al. [2002] showed that sampling time does not influence the seasonal cycle.

Fig.3 (block 10361): The referee suggests to show a comparison of NO2 in European Region.

Answer: The European region was excluded in order not to deviate the readers attention from the main scope of the comparison and to keep this aspect short. No principle differences between the results for North America and Europe are found in the comparison between NO2 columns from CBM-IV model results and GOME satellite data.

Results (block 10347) line1-4: Clarification of sentence 'In the southern hemisphere (SH)..' is suggested.

Answer: For clarification we include the following sentence. In the southern hemisphere (SH) relative contributions show an opposite seasonality with higher contributions in summer than in winter, although absolute contributions are lower in summer than in winter. The opposite seasonality in relative contributions is caused by strong

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seasonality of natural emissions, which compensates for the seasonal cycle of NMHC lifetimes and OH concentrations, respectively.

The referee suggests clarification of the sentence on road traffic effect on SH ozone.

Answer: In this sentence it is correct to speak of Southern Hemispheric ozone. The wording overproportional seems to have confused the referee so we rephrase the sentence. Road traffic induced ozone increase in the SH in summer (January) is only a factor of 5 smaller than in the NH, although emissions are lower by a factor of roughly 20.

The referee suggests to further detail the findings from Granier and Brasseur [2003].

Answer: In July, an overall increase in surface ozone due to total road traffic emissions between 8% and 15% in non-source regions in northern extratropics (e.g. North Atlantic, North Pacific) and higher contributions of up to little more than 16% in source regions (e.g. central Europe, USA, Japan) can be found (lower panel). In source regions, these results are comparable to findings of Granier and Brasseur [2003], who calculated about the same relative contribution (10% to 15%). 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 (see Fig. 5, *middle* and *right*), the origin of this difference can be attributed to neglecting of NMHC road traffic emissions. The impact of NOx emissions on ozone is strongly confined within source regions, while the impact of NMHC emissions is visible in remote areas also. These typical different patterns lead us to the conclusion, that main origin of differences between our results and Granier and Brasseur [2003] are attributed to NMHC emissions.

The referee suggests additionally that one reason for differences found is the difference in models sensitivity of ozone production.

Answer: According to an intercomparison of chemical mechanisms [Kuhn et al., 1998]

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our mechanism applied (CBM-IV) was found to lie at the lower range of ozone productivity, which contradicts the hypothesis of a higher sensitivity of our model to ozone production.

(block 10349) line9-11: The referee suggests that less ozone is caused by titration.

Answer: The inclusion of road traffic emissions leads to an increase of both ozone production and ozone loss. In most regions the net-effect on ozone production is positive. However, in Central Europe and East Coast USA large NOx changes result in a negative net ozone production. The total net ozone production remains positive, only the change, caused by the inclusion of road traffic emissions is negative. Therefore, we hesitated to call it titration.

As we compare two model simulations with different nitrogen oxide concentrations the origin of the difference between those two simulations is not due to ozone titration but due to a comparison of ozone values from those two chemical regimes within these model simulations. Hence in our GCM no ozone titration takes place. This is why we do not explain the difference with ozone titration.

(block 10349) line 14-19: Further clarify where strongly confined regions refer to.

Answer: We included the following modified sentence: In these strongly confined areas, road traffic NMHC emission contribute in July about 12% to ozone.

(block 10349) line 20-23: The referee suggests that main origin of differences are different mechanisms in winter than in summer. To make this point clear he suggest to show ozone production plots.

Answer: Indeed this is an interesting question. Unfortunately we can only partly answer it, since we did not save information on individual reactions. In general, the NOx road traffic emissions enhance ozone production and ozone loss both in winter and summer. In source regions this leads to a positive net ozone production in summer and in winter shows a transition from positive values at lower latitudes to negative values at higher

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latitudes, with a transition at around 30-50N. The mechanism between summer and winter is in that sense the same since both production and destruction increases. For the NMHC emissions, we agree with the referee. The mechanism changes. In winter both ozone production and loss is reduced in most areas. In summer, this pattern is seen in a smaller area, since a competing mechanism, caused by the longrange transport of PAN is enhancing both ozone production and loss. The decrease in ozone loss cannot result from ozone reactions with OH or HO2 since those species are increasing. On the other hand NOx is decreasing, which leads therefore to a decreased ozone destruction. However, since the ozone production is decreasing ozone will stabilize at a lower level, which also leads to a lower ozone destruction and one cannot decide which of both mechanisms dominates.

We have included the requested figure and discussed it in section 4.2.

The referee asks whether total ozone changes can be reproduced by summing the individual component shown in Fig. 6.

Answer: Summing up the ozone impact of individual components and comparing it to the total ozone changes, we found that this overestimates tropospheric ozone columns by about 5%, rarely up to 10%. With this knowledge we can estimate the contributions of individual components to the total impact with this assumed linearity and the rescaling by about 5%. We included: This approach has been chosen to account for non-linearities. This non-linearity appears as the summed impact calculated by these individual simulations is i.g. less than 10 percent higher than the simultaneous impact from these components on ozone.

line 16-17: Suggestion followed.

line20 Question is raised whether PAN transport is the only mechanism.

(block 10351) line 8 By looking at the additional figure on net ozone production it becomes clear that ozone production is also changing due to NMHC emissions. So it is

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more adequate to change wording accordingly to: One mechanism for ..

line 12-16 Differences should be explained by chemical scheme and meteorology.

Answer: As discussed above we have to assume that Granier and Brasseur did not account for road traffic NMHC emission. Differences in the chemical scheme are contradicted by finding from Kuhn et al. on the comparable low ozone productivity of CBM-IV. The impact of different meteorology can be minimized by using a kind of climatological mean, which is done in both studies, Granier and Brasseur and our study.

line 16-19 Remove preceding paragraph, as it repeates contents.

Answer: For clarity reasons we decided not to remove the paragraph, but to change the order of the sentence to make the point more clear.

(block 10352) line 1 suggestion followed

line 12- Make point clear of assumed linearity of individual results.

Answer: The comparison of summed impacts of individual components with the total impact of all components together shows that the sum is higher than the total. Nevertheless again for PAN the overestimation lies in the range of less than 10%. For clarification we include: In the following we use results from individual components scenarios for deducing the relative importance of individual species. Hereby it has to be noted, that the sum of individual impacts overestimates the total impact by less than 10%.

line 18-19 Suggestion to show NOx increase in remote regions with a separate figure.

Answer: To limit the length of this paper, we did not include an additional figure.

include the information of global budget of tropospheric ozone for individual scenarios in an additional table.

Answer: Unfortunately budget data was not evaluated from these model simulations.

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Include two more references for long-range transport (O.Wild)

Answer: We included the additional reference on long-range transport from O. Wild and the following paragraph: The results are consistent with the findings of Wild et al. (2001), who studied the intercontinental transport of ozone and its precursors. They analysed the impact of 10 % anthropogenic emission changes from the regions Europe, USA, and Asia, respectively. They found roughly the same impact from emissions from USA and Europe to the ozone budget of the European upper troposphere, which is in agreement with our findings showing a 2% contribution from USA and Europe each. For Asia, Wild et al. (2001) found that one third of the upper troposphere ozone changes arise from European and USA emissions, agreeing with our findings showing a 1.5 % contribution of European and USA road traffic contributions to ozone out of a total of 4%.

The referee suggests that when referring to the climate impact of road traffic also indirect effects from NMHCs and CO have to be considered.

Answer: We agree, but the scope of this paper is impact on tropospheric ozone. The results from climate impact of road traffic is scope of an separate paper.

Technical corrections

Technical corrections were included. Color scheme was not modified, as it was adopted from Granier and Brasseur [2003] in order to allow direct comparison.

3 Detailed Response to Referee #2 - 29 November 2005

Year of emissions: The referee suggests to further emphasize the year of the emissions (1990), and to give an idea on the difference between the 1990 emission and more recent estimates. I agree with the referee that the year of emission is important. We included the year of the emissions in abstract, summary and

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conclusion. As total amounts between 1990 and the suggested EDGAR study do not change considerably we did not include a separate discussion on the issue of changing amounts.

Relative vs. absolute ozone changes: The referee writes that it might be useful to extract the concentration changes and to provide this extra information. He suggests to include an additional figure or replace the existing one.

Answer: I agree with the referee that this is useful information which should be provided. Nevertheless for the reason of the length of the paper, we agreed not to include separate figures but to include additional information on absolute concentration changes in the context for the primary and secondary trace gases. For this reason absolute concentration changes are included in the final version in parenthesis.

Validation of the model: The referee suggests to give a paragraph summing up the findings in an earlier paper on model evaluation. Answer: This issue is reflected by a short summary of evaluation of the model system given in Roeloefs and Lelieveld [2000]. A brief overview is included - see response to anonymous referee No.1 (block 10344).

Plume effects: The referee suggests to mention the impact of the neglection of plume effects in the simulation on the calculated on ozone productivity.

Answer: This is definetely an interesting question. Nevertheless we can not provide an adequate response, as this requires coupling of local, regional and global models. Effects for road traffic emissions are more difficult to isolate. We will address this issue within the IP QUANTIFY and the NoE ACCENT.

Contribution of ozone to episodes: The referee suggest to provide data on air quality issues during episodes. Answer: Unfortunately no statistical information on air quality threshold values has been stored in the model simulations. Reason for this

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is that the horizontal resolution is relatively coarse, making it difficult to reproduce peak values, e.g. measured at stations.

Role of PAN: The referee sees some redundancy in the description of the role of PAN. For this reason we have to reconsider the initial discussions on the role of PAN at several places, and refer mainly to Section 4.2.

Block 10349: The referee sees no relation between ozone titration and NMHC emissions. Answer: I agree with the referee that the statement in the current form, can not be understood. It is much more adequate to talk from the ozone production point of view, where a change in NMHC concentration causes a shift on the ozone production rates. By comparing two different situations of NOx versus NMHC relations, two different ozone production rates result, where the one with the lower NMHCs is at the same time the higher production rate. Refering back to the initial sentence used, this is only equivalent to a change from a situation where additional NOx causes ozone titration resulting in lower ozone values (high ozone precursors) to a situation where additional NOx now causes effective ozone production. For this reason we deleted the fragment - by inhibiting ozone titration.

Block 10343, line 15: The referee suggests that the two different periods cause another significant difference in the comparison. Answer: For clarification we included the following sentence: As year of simulation and observation years are different, a comparison of main pattern can be performed only.

Block 10344, line 10: The referee asks for clarification of the word respective in the context used. The restructured sentence is the following: Deviations of more than 50% are only found in remote regions with column densities of NO2 below $10 \cdot 10^{14} mol.cm^{-2}$ (Fig. 2., *third row*), where small concentrations of tropospheric NO2 in the reference sector influence the determined (low) tropospheric NO2-columns considerably.

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