Atmospheric Chemistry and Physics

Global impact of road traffic emissions on tropospheric ozone

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Abstract. Road traffic is one of the major anthropogenic emission sectors for NO_x, CO and NMHCs (non-methane hydrocarbons). We applied ECHAM4/CBM, a general circulation model coupled to a chemistry module, which includes higher hydrocarbons, to investigate the global impact of 1990 road traffic emissions on the atmosphere. Improving over previous global modelling studies, which concentrated on road traffic NO_x and CO emissions only, we assess the impact of NMHC emissions from road traffic. It is revealed that NMHC emissions from road traffic play a key role for the impact on ozone. They are responsible for (indirect) long-range transport of NO_x from road traffic via the formation of PAN, which is not found in a simulation without NMHC emissions from road traffic. Long-range transport of NMHC-induced PAN impacts on the ozone distribution in Northern Hemisphere regions far away from the sources, especially in arctic and remote maritime regions. In July total road traffic emissions (NO_x, CO and NMHCs) contribute to the zonally averaged ozone distribution by more than 12% near the surface in the Northern Hemisphere midlatitudes and arctic latitudes. In January road traffic emissions contribute near the surface in northern and southern extratropics more than 8%. Sensitivity studies for regional emission show that effective transport of road traffic emissions occurs mainly in the free troposphere. In tropical latitudes of America up to an altitude of 200 hPa, global road traffic emissions contribute about 8% to the ozone concentration. In arctic latitudes NMHC emissions from road transport are responsible for about 90% of PAN increase from road transport, leading to a contribution to ozone concentrations of up to 15%.

1 Introduction

Ozone plays an important role in the troposphere due to its impact on the oxidizing capacity of the atmosphere, on air quality, and its contribution to the greenhouse effect. In the atmosphere NO_x, CO and NMHCs act as ozone precursors, by forming radicals which finally contribute to ozone formation (Crutzen et al., 1999). CO and NMHCs are oxidized by OH while nitric oxide determines oxidation pathways possibly leading to ozone production. The relevant NO_2 related reactions are:

$$NO + O_3 \rightarrow NO_2 + O_2 \tag{R1}$$

$$NO_2 + O_2 \rightarrow O_3 + NO \tag{R2}$$

$$NO + HO_2 \rightarrow NO_2 + OH \tag{R3}$$

$$O_3 + HO_2 \rightarrow 2O_2 + OH \tag{R4}$$

Reaction (R2) is a net reaction of the photolysis of NO_2 and subsequent ozone formation. Reactions (R1) and (R2) describe a photostationary equilibrium of NO/NO_2 and O_3 without net ozone production. Only if ozone is substituted in NO_2-formation (Reaction R3) a net ozone production takes place. These conditions apply above a threshold in the NO mass mixing ratio (typically above 10–30 ppt) when HO_2 reacts preferably with NO (R3) and not with O_3 (Reaction R4). As HO_2 is formed during oxidation of CO and NMHCs (Crutzen et al., 1999; Atkinson, 1990), their emissions are influencing significantly the above described photochemistry.

Road traffic is one of the main emitters of these ozone precursors, formed by the combustion of fossil fuels inside internal combustion engines (gasoline and diesel). Although since the nineties reductions in road traffic specific emissions were achieved in certain regions due to the introduction of catalytic converters resulting in decreasing emission indices, global emissions are still supposed to grow in the future (OECD, 1995),(IPCC, 2000). Reasons for this are both,

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Table 1. Global annual anthropogenic emissions of nitrogen oxides (NO_x), carbon monoxide (CO) and non-methane hydrocarbons (NMHCs), for references see text. Anthropogenic total, fossil fuel combustion and herein included emissions due to road traffic are given (o/w = of which).

Emissions	NO_x Tg [N]	CO Tg [CO]	NMHCs Tg [C]
Anthropogenic	27.6	678.4	108.2
Fossil fuel	22.6	478.4	35.4
o/w road traffic	8.8	236.9	26.6

Fig. 1. NO_x emission $[10^{-11}\,\mathrm{g\,cm^{-2}\,s^{-1}}]$ from road traffic in 1990.

traffic growth rates, e.g. in developing regions, and fuel intense vehicles counteracting mitigation strategies. In contrast to other transport modes, e.g. shipping and aviation, only few global modelling studies on the impact of road traffic exist. E.g., Granier and Brasseur (2003) investigated the impact of NO_x and CO emissions from road traffic and estimated relative contribution of such emissions to ozone concentrations near the surface in the Northern Hemisphere of between 12% and 15% in industrialized regions and about 9% in remote regions. In our study we also consider the additional impact of NMHC emissions from road traffic. The related impact on climate is dealt within a forthcoming paper (Matthes et al., 2007[1]).

The applied global atmosphere-chemistry model, emission data and experimental setup are described in Sect. 2. In Sect. 3 simulated NO_2 columns are evaluated against satellite data, and extending results of existing model evaluation (Roelofs and Lelieveld, 2000; Houweling et al., 1998). In Sect. 4, results of our modelling study are presented and discussed, with respect to the effect of total road traffic emissions, individual contributions from NO_x, CO, and NMHCs emissions, and with respect to regional road traffic emissions. Section 5 gives a summary and concludes this study.

2 Model, emissions, and experimental setup

In order to study the impact of road traffic emissions on the atmospheric composition we use the global circulation model ECHAM4 (Roeckner et al., 1996) coupled to the chemistry module CBM-IV (Gery et al., 1989), using a parameterized stratospheric chemistry. The coupled model system (ECHAM4/CBM) has been developed and adapted for global modelling (Roelofs and Lelieveld, 2000). ECHAM4 is a spectral general circulation model which solves the primitive equations; for the present study it is used in T30 horizontal resolution with 19 layers vertical resolution (model top layer centered at 10 hPa), model physics is calculated

on the associated Gaussian grid of about $3.75° \times 3.75°$ geographical longitude vs. latitude. Water vapour, cloud water and the 35 tracers included in the chemistry model are transported by a semi-Lagrangian advection scheme. CBM-IV is a carbon-bond-mechanism based on structural lumping with explicit representation of e.g. alkanes, alkenes, isoprene, aceton, formaldehyde. Aromatic compounds are neglected in the scheme used here, but sensitivity experiments performed with our chemistry model give a lower estimate of the contribution of aromatic compounds to atmospheric ozone in the order of 3% in strongly confined regions.

Emissions are implemented as flux boundary conditions into the model system. The year 1990 was chosen as reference year, since complete emission data and atmospheric observations were available. Global values of anthropogenic emissions (NO_x, CO, and NMHCs) are given in Table 1. Anthropogenic NO_x emissions amount to 27.6 Tg and include 5.0 Tg from biomass burning (Hao and Liu, 1994) and 22.6 Tg from fossil fuel combustion (Benkovitz et al., 1996). Natural NO_x 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. Road traffic emissions (Matthes, 2003; Matthes and Sausen, 2000) of nitrogen oxide (included within fossil fuel combustion) were calculated from the fossil fuel related emissions (Benkovitz et al., 1996) by extracting the fraction of road traffic given by Olivier et al. (1996). The geographical distribution of NO_x emissions from road traffic is shown in Fig. 1. Carbon monoxide emissions from fossil fuel combustion are taken from Olivier et al. (1996), but have been enhanced by 15% to yield the global amount given in OECD (1995). Emissions from biomass burning are adopted from Hao and Liu (1994). The NMHC emissions from road traffic were adapted according to Houweling et al. (1998): individual emitted NMHC species (Olivier et al., 1996) are

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[1]Matthes, S., Stuber, N., Grewe, V., and Ponater, M.: Radiative forcing of road transport emissions, in preparation, 2007.

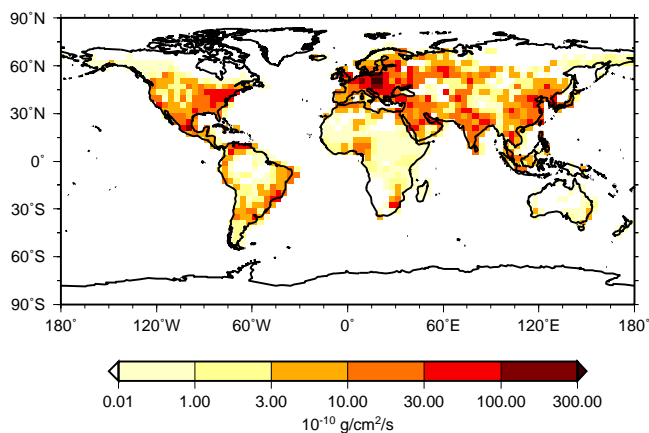lumped and thereby included in the carbon-bond-scheme of the chemistry module according to Gery et al. (1989). With this procedure around 80% of total mass of NMHC emissions according to Olivier et al. (1996) can be integrated into the model.

To determine the impact of emissions from road traffic, a number of individual simulations were performed (Table 2): a control run (CTR90) and six scenario simulations. A scenario consists of identical conditions except those emissions which are to be investigated are excluded in the model system. Three scenarios for individual species emitted by road traffic (No_NOx, No_NMHC, No_CO), one excluding all road traffic emissions (No_rt), and two scenarios without all road traffic emissions in a specific region (No_rtusa, No_rteur) were calculated. The impact of the individual road traffic emission component is then calculated as the difference between the respective scenario and CTR90 on a monthly mean basis (four years averaging period after two years spin-up time). This approach has been widely used. However it differs from the methodology applied by Granier and Brasseur (2003). They use an averaged meteorology whereas we average the chemical impact and hence allow more non-linearities, e.g. a larger variability in transport affecting chemistry.

3 Comparison with observations

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. The representation of the atmospheric NO_x distribution is of crucial importance for the simulation of the impact of road traffic emission. Satellite data of tropospheric NO_2 have been made available only recently (Richter and Burrows, 2002) from the GOME measurements. We compare these with simulated NO_2 columns based on monthly means of four consecutive years, following Lauer et al. (2002), who applied the so-called tropospheric excess method (TEM) on model data. We used monthly mean NO_2 columns averaged from half hour values, as Lauer et al. (2002) showed that sampling time does not influence the seasonal cycle. However, total amounts may be overestimated by 20% in Europe and 30–50% in Africa (Martin et al., 2002). As year of simulation and observation years are different a comparison of main patterns can be performed only.

Figure 2 gives a global picture of the model's capability and indicates that the model system is able to capture the main pattern of the atmospheric NO_x distribution. Nega-

Table 2. Overview of model simulations performed; rt = road transport,w/o = without.

Name	Description
	Base runs
CTR90	emissions from all sources
No_rt	w/o NO_x, NMHCs, CO from road traffic
	Effect of individual emission
No_NOx	w/o NO_x from road traffic
No_NMHC	w/o NMHC from road traffic
No_CO	w/o CO from road traffic
	Regional effects
No_rtusa	w/o emissions from road traffic in USA
No_rteur	w/o emissions from road traffic in Europe

tive values indicate that the reference sector in this latitude is not free of tropospheric contributions to the total NO_2 column (see also discussion on application of TEM below). A conclusion on the quantitative model performance is hard to draw, first, because of uncertainties in the observational data caused by clouds and a low sensitivity in surface-near NO_2, which may dominate the total tropospheric column, and, second, because of different meteorology between observational data and modelled data. The tropospheric NO_2 columns are influenced by vertical mixing within the boundary layer, which is determined by the respective meteorological conditions. As a general circulation model calculates its own meteorological state and does not use measured meteorological data as input, these conditions can differ quite substantially between different individual periods from modelling studies and observations. As observational data in winter is sparse, this can become prominent then. A vertically limited boundary layer can promote an enrichment of NO_x in this layer, causing high trospheric NO_2 columns.

Comparing the amplitude of the seasonal cycle of the tropospheric NO_2 column densities in industrialized regions (eastern USA; similar for Europe, not shown) indicates an agreement within 20% (Fig. 3). Taking into account a 20% overestimation due to different sampling times (see above) agreement might be even better. In Fig. 3 additional results from model studies including a chemistry scheme which does not include higher hydrocarbons (ECHAM4.L39(DLR)/CHEM) are given (Lauer et al., 2002). Comparison shows that higher hydrocarbon chemistry in these regions mainly reduces atmospheric NO_2 columns. In industrialized regions of the Northern Hemisphere deviations between modelled and observed values are higher in winter than in summer. This corresponds to a stronger reduced vertical mixing in the boundary layer in wintertime. On the other hand, the observations have a small sensitivity in the surface layer of the atmosphere due to physical reasons. This can serve as an explanation why
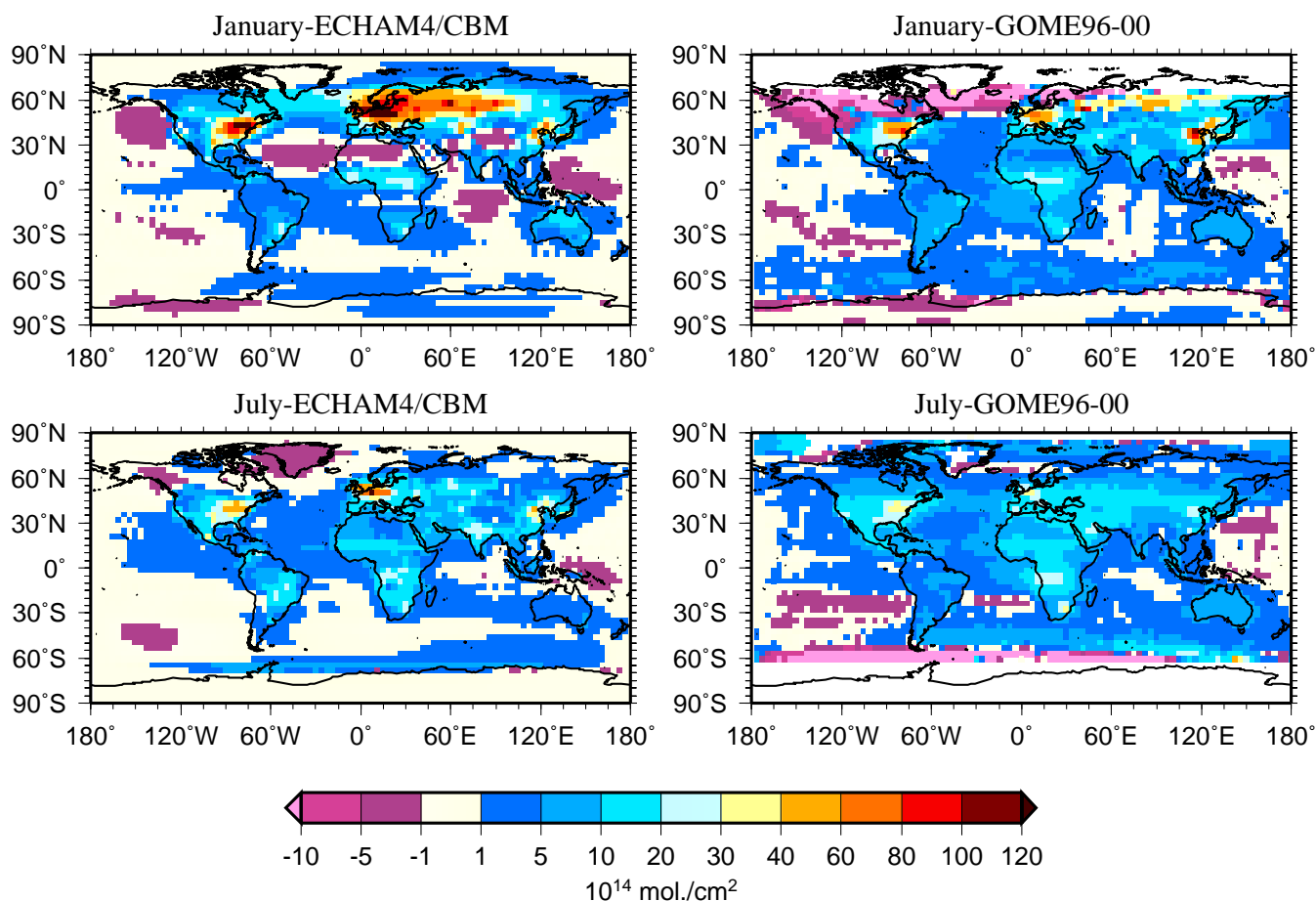
Fig. 2. Tropospheric NO_2 columns [10^{14} mol. cm^{-2}] derived from GOME satellite data (right) (Richter and Burrows, 2002) and ECHAM4/CBM model simulations (left) for January (top) and July (bottom).

modelled data are higher than observed values. Additionally, clouds in wintertime prevent observations with the satellite instrument, which causes a sparse data base for northern hemispheric winter (December and January) and can cause a systematic bias of measurements. Deviations between observed and modelled NO_2 columns amount to values between 15% and 30%. Deviations of more than 50% are only found in remote regions with column densities of NO_2 below 10×10^{14} mol. cm^{-2} (Fig. 3, third row), where small concentrations of tropospheric NO_2 in the reference sector influence the determined small tropospheric NO_2-columns considerably. Therefore within these model regions, TEM can not be applied reliably in order to deduce NO_2 tropospheric columns.

The preceeding comparison shows that the ECHAM4/CBM is able to reproduce the general pattern of the global NO_2 column density distribution calculated with the TEM within regions where the reference sector does show no tropospheric influence ("clean air"). However absolute maximum values in industrialized areas (high NO_x-regions) show a tendency of higher values in the model

than in observations. Due to non-linear photochemistry this would cause a lower estimate of ozone productivity of NO_x emissions.

4   Results

4.1   Impact of total road traffic emissions

The changes in zonal monthly mean concentrations (CO, NO_x, alkanes, ozone) due to total road traffic emissions are presented in Fig. 4 for January and July (difference between the simulations No_rt and CTR90). Road traffic contributes to atmospheric carbon monoxide concentrations in the free troposphere of the summertime northern extratropics by more than 12% (Fig. 4, first row) or 10 ppb (not shown). In January, contributions of more than 24% (20 ppb) are reached in the Northern Hemisphere due to a longer chemical lifetime (less photolysis in winter). In the southern extratropics relative contributions in the free troposphere are lower than in the northern extratropics due to smaller road traffic emissions in southern latitudes. Relative contributions
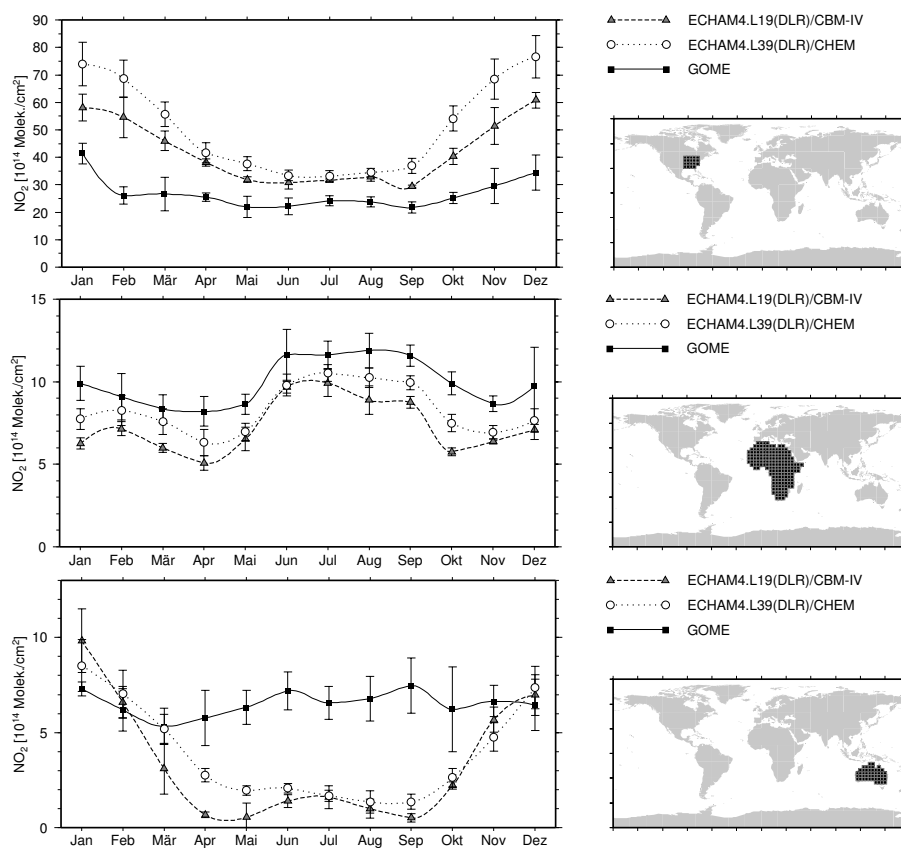
Fig. 3. Regional tropospheric NO_2-columns derived from GOME satellite data (Richter and Burrows, 2002) and ECHAM4 model simulations (in 10^{14} mol. cm^{-2}). The respective geographic region for averaging is indicated in black: eastern USA, Africa and Australia.

amount to less than 8% (5 ppb) in January (summer) and to less than 12% (10 ppb) in July (winter). The highest values occur in source regions, with maximum contributions to atmospheric CO in January in the Northern Hemisphere near the surface (up to 32% or 100 ppb).

Road traffic emissions contribute to atmospheric NO_x (Fig. 4, 2nd row) by a similar order of magnitude as to CO, ranging from 8% to 24% in the extratropics (1 ppt to 200 ppt), depending on altitude and season. However, the impact of NO_x emissions compared to CO is much more confined to the vicinity of the source regions as the atmospheric lifetime of NO_x is about one order of magnitude shorter than the lifetime of CO. In midlatitudes near the surface, contributions up to 32% are calculated (10 ppt NH, 200 ppt SH), whereas in the free troposphere values around 12% are found (1–10 ppt). Beside these high contributions in midlatitudes, relative contributions of more than 50% appear in arctic regions (2 ppt). These relative contributions correspond to absolute contributions in arctic regions of about 2 ppt NO_x. This effect is particularly pronounced in winter (January). As will be shown in the sensitivity studies in the next section these high contributions are caused by NMHC road traffic emissions leading to enhanced formation of PAN. PAN is then transported

throughout the hemisphere and decomposes in subsidence regions (Sect. 4.3). This represents an indirect transport of NO_x from road traffic.

The impact of road traffic emissions on atmospheric concentration of alkanes (Fig. 4, 3rd row) reaches relative contributions of more than 50% (up to 5 ppt January, 2 ppt July). Alkanes are selected as a representative for a NMHC with comparatively long chemical lifetime. These high relative contributions show that road traffic represents one of the major emitters of alkanes, for which anthropogenic sources dominate. In the Northern Hemisphere, again relative contributions are higher in winter than in summer (indicated by larger atmospheric regions with contributions above 50%), due to longer chemical lifetimes in winter. In the Southern Hemisphere (SH) relative contributions show an opposite seasonality with higher contributions in summer than in winter (Fig. 4), although absolute contributions are lower in summer than in winter (Matthes, 2003). The opposite seasonality in relative contributions is caused by strong seasonality of natural emissions, which compensates for the seasonal cycle of NMHC lifetimes and OH concentrations, respectively.

Characteristic differences in the pattern of the relative contributions of road traffic emissions to the distribution of
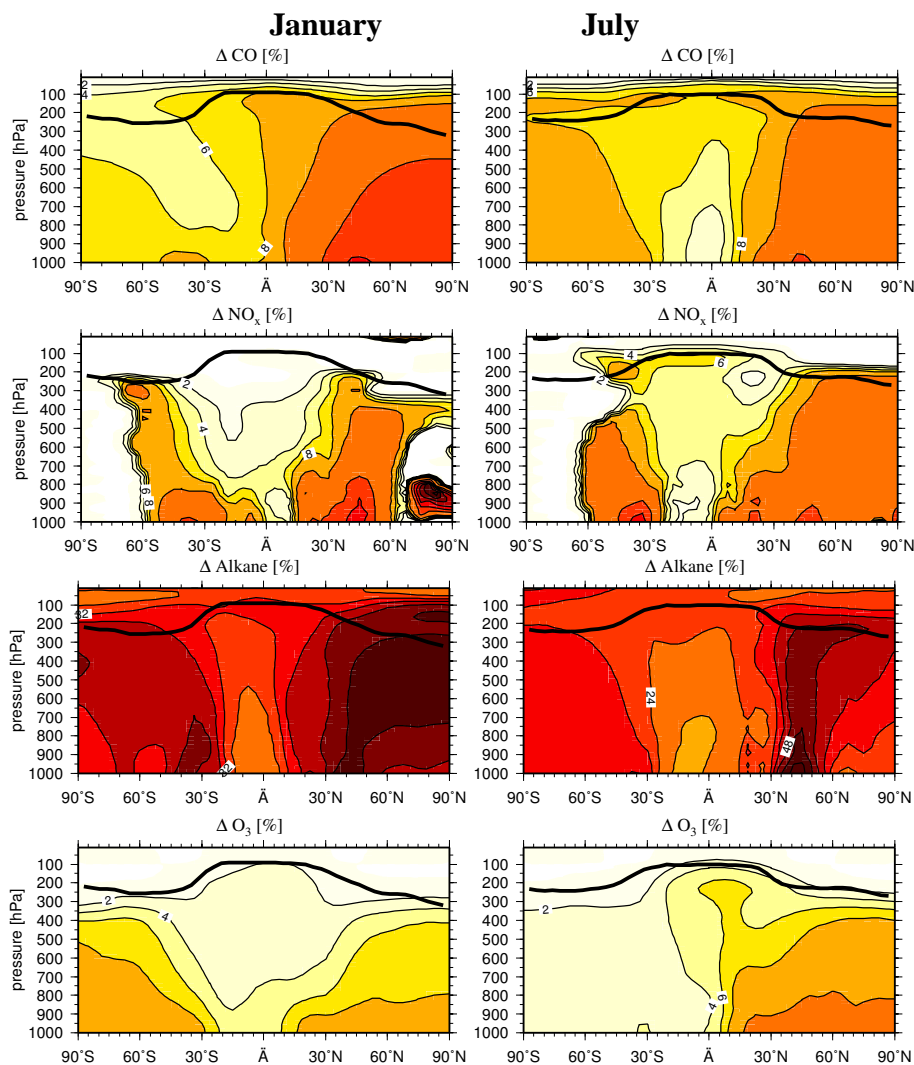
Fig. 4. Relative contribution of road traffic emissions to atmospheric CO, NO_x, NMHCs and O_3 zonally averaged distributions (in %). Isolines are 2, 4, 6, 8, 12, 24, 32, 48, 56%.

atmospheric ozone (Fig. 4, bottom) can be found between January and July. Its seasonality shows a pattern opposite to the primary species (see above), with maximum ozone contributions occurring in summer and minimum values in winter. While ozone formation is increased by an enhanced abundance of its precursors, not only the concentration of precursors is of importance, but also the photochemical activity which has its maximum in summer. In the summer hemispheres, relative contributions to the atmospheric burden of ozone of more than 12% (NH, up to 5 ppb) and 8% (SH, up to 1 ppb) from road traffic induced ozone occur. In winter hemispheres relative contributions are lower by a factor between 1.5 and 2 in the NH and of about 3 in the SH.

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. Reason for this comparatively high relative ozone contribution in the

Southern Hemisphere is the low background concentrations of trace gases, especially nitrogen oxides. As productivity of ozone production decreases with increasing NO_x concentrations (Liu et al., 1980, 1987) NO_x emissions from road traffic are much more effective when emitted in the Southern Hemisphere. In the Northern Hemisphere ozone production takes place at high NO_x concentrations, often in the saturation regime. Thus a further NO_x increase only leads to a weak increase of the production rate or even to a decrease.

For comparison of our results to those presented by Granier and Brasseur (2003), the horizontal distribution of relative contributions to surface ozone is shown (Fig. 5, left). In July, an overall increase in surface ozone due to total road traffic emissions between 8% and 15% (2–5 ppt) in non-source regions in northern extratropics (e.g. North Atlantic, North Pacific) and higher contributions of up to little more than 16% (10 ppt) in source regions (e.g. central Europe,
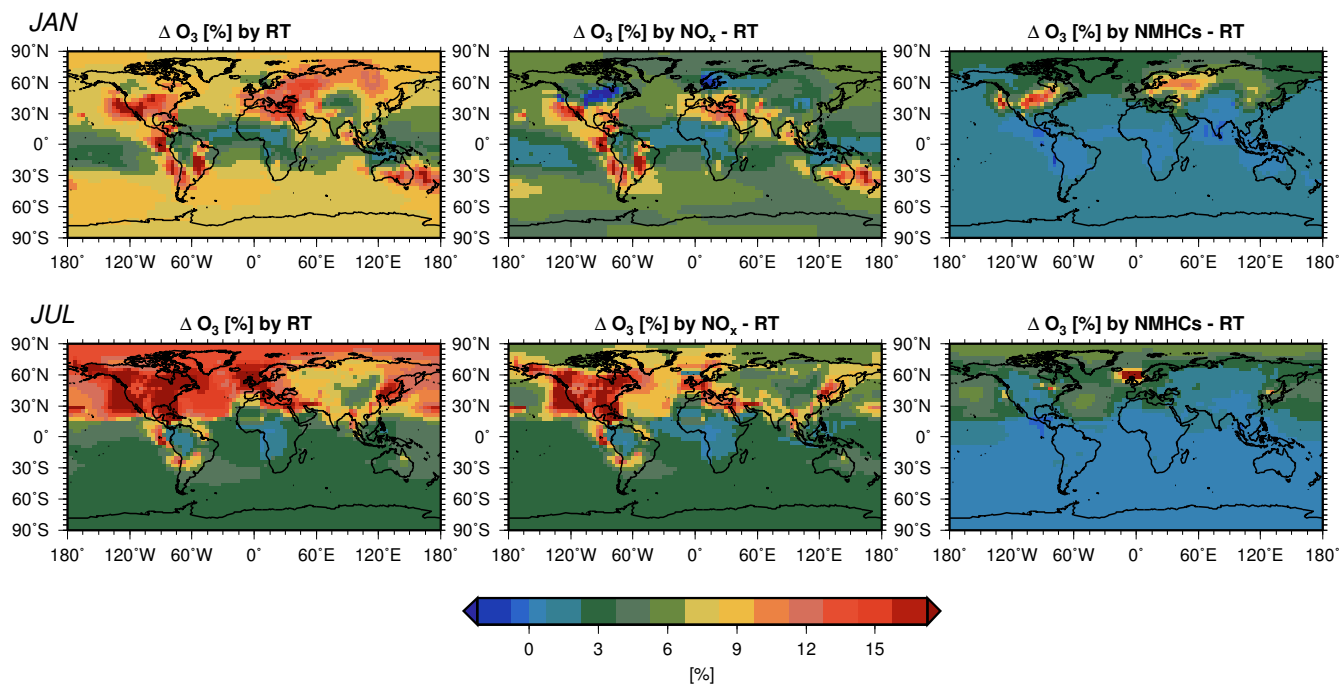
Fig. 5. Relative contributions [%] of total and individual road traffic emissions to the surface ozone distribution in January (upper row) and July (lower row); total emissions (left), NO_x only (middle) and NMHC only (right).

USA, Japan) can be found (lower panel). In source regions, these results are comparable to the 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 NO_x 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 from road traffic which they did not consider. In the Southern Hemisphere, relative contributions remain lower than on the Northern Hemisphere, with values of more than 8% being calculated only in July in continental source regions and more than 4% contribution only in certain outflow regions (e.g. Pacific ocean, east coast of South America, tropical pacific), which is comparable to the findings of Granier and Brasseur (2003).

As Granier and Brasseur (2003) give only values for July, no direct comparison is possible for January. By comparing the impact of total road traffic emissions with the impact of NO_x emission only (Fig. 5), the significant role of NMHCs becomes again obvious. In the Northern Hemisphere in January, the relative contribution of total road traffic emissions

remain much lower in source regions compared to July with only more than 8%, sometimes even less due to weaker photochemistry (as mentioned before). Relative contributions of more than 16% are only found in a few locations, e.g. southern USA, Arabian Peninsular. In January however, the relative contributions in southern latitudes are higher due to enhanced photochemistry (summer) and low background concentrations (see discussion above). There, relative contributions of more than 16% are found in industrialized centres (e.g. South America, New Zealand). Over large parts of southern hemispheric extratropic oceans relative contributions of more than 8% are simulated. This emphasises the considerable long-range impact of road traffic emissions on ozone in remote regions.

4.2 Importance of individual emissions for ozone

To determine the impact of individual components from road traffic emissions we will discuss now separate sensitivity experiments for NO_x, CO and NMHC emissions only (Table 2). 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.

In Fig. 5, the individual contributions of both NO_x (middle) and NMHCs emissions (right) together with the total impact of road traffic to surface ozone are shown. Maximum contributions to atmospheric ozone from road traffic
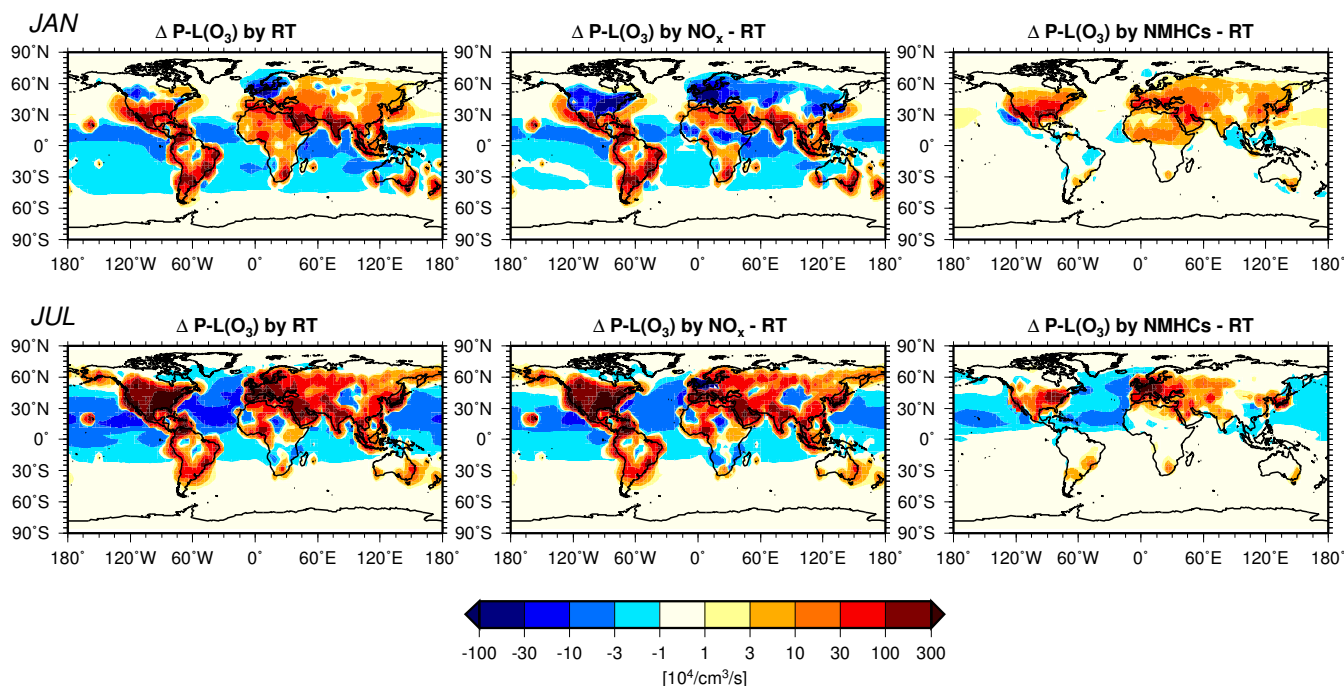
Fig. 6. Contributions [%] of individual road traffic emissions – NO_x (left), NMHCs (middle)) and CO (right) to distribution of net ozone production.

NO_x emissions are found in summer in source regions and downwind regions (about 15%), especially over North America and Europe. In January, road traffic NO_x emissions even lead to ozone decrease in strongly confined areas of source regions, caused by a dominating decrease of ozone productivity of NO_x with increasing NO_x concentrations.

NMHCs (like CO) act as a source for the hydroperoxyl radical (R5)

$$RH + OH \rightarrow R + HO_2 \qquad (R5)$$

thereby influencing Reactions (R3) and (R4). In these strongly confined areas, road traffic NMHC emission contribute in July about 12% to ozone. In other Northern Hemisphere regions, especially in higher latitudes, an ozone increase by around 4% is found, with higher contributions (up to 6%) in remote areas (e.g. oceans, Arctic) than in sources regions (approximately 2%), other than the hot spots (e.g. parts of Europe and USA).

To better understand the ozone changes Fig. 6 shows changes in net ozone production rate. NO_x road traffic emissions enhance ozone production and ozone loss in winter and summer. In source regions this leads to a positive net ozone production in summer. In winter a transition is found at around 30–50 N which ranges from positive values at lower latitudes to negative values at higher latitudes. In winter NMHC emissions lead to a reduced production and a reduced loss of ozone resulting in an increased net ozone production (Fig. 6). In summer, this pattern is seen in a smaller area, since a competing mechanism, caused by the long-range transport of PAN is enhancing both ozone production and loss. This decrease in ozone loss results either from lower ozone concentrations or lower NO_x concentrations, since OH and HO_2 are increasing.

The impact of CO emissions from road traffic on atmospheric ozone is not shown because it is about a factor 10 smaller than that of NO_x emissions in both January and July.

Figure 7 displays the zonal mean ozone change due to individual emission components from road traffic, in order to quantify the different role of road traffic NO_x and NMHC emissions in the free troposphere (for totals see Fig. 4, bottom). The effect of both, total emissions and NO_x emissions, is strongest in latitudes where the main source regions of road traffic emissions are located (15° N–60° N). In July, NO_x emissions in northern hemispheric source regions contribute by more than 10% to the zonally averaged near surface ozone. At 800 hPa and below, a relative contribution of more than 8 % can be attributed to road transport NO_x emissions in these latitudes. More regional considerations show (see Sect. 4.4), the atmospheric distributions in these altitudes are predominantly influenced by regional emissions. This is consistent with the short atmospheric lifetime of NO_x of only several days. At higher altitudes up to 300 hPa, relative contributions of global road traffic NO_x emissions of more than 4% can be noticed in July. Generally, about 70% of the total ozone increases is caused by NO_x emissions from road transport at latitudes, where the main sources are

located, and in the free troposphere. Meridional gradients are distinct for NO_x related ozone increase especially in July, reflecting fast photochemistry and related short atmospheric lifetime.

In July, NMHC emissions contribute considerably less than NO_x emissions to ozone in those source latitudes (15° N–60° N) with only about 3% (Fig. 7, middle row). Ozone contributions, of about 6 %, are found in arctic regions. In these remote regions the effect due to NMHC emissions has about the same strength as due to NO_x emissions (Fig. 7, upper row), which is consistent with above discussed contributions to surface ozone. One mechanism for the long-range impact of NMHC emissions from road transport is additional PAN-formation (see Sect. 4.3). In January, road traffic NMHC emissions contribute strongest to zonally averaged ozone in source latitudes with more than 4% relative contribution, by inhibiting ozone titration, and more than 2% in northern hemisperic extratropics, but outside the main source regions. Again the strong contribution in arctic latitudes is noteworthy.

As mentioned above, the impact of CO emissions from road traffic on atmospheric ozone is about a factor of 10 smaller than that of NO_x emissions in both January and July. Relative contributions of 1% and 2% can be found in latitudes north of 30° N below 400 hPa, elsewhere it is even less.

To sum up these results it can be noted that the total impact of road traffic emissions (NO_x, CO, NMHCs) is dominated by the impact of NO_x emissions in source regions and while it is substantially influenced by the impact of NMHC (and CO) emissions in non-source regions. This importance of NMHC emissions (and partly CO) became obvious from the comparison between the impact due to exclusive consideration of NO_x and the impact by consideration of all three emission species together (NO_x, NMHCs, CO). Particularly, relative contributions of road traffic emissions in remote regions increase, e.g. in arctic regions in July, from about 8% to about 15%. As will be shown in the next sub-section (4.3) the mechanism for long-range impact of NMHC emissions is transport of additionally formed PAN, which acts as a temporal reservoir for NO_x and allows an indirect long-range transport of NO_x from source regions.

When comparing our separate sensitivity experiments for NO_x and CO emissions only (No_NOx, No_CO) with total road traffic impact (NO_x and CO only) given by Granier and Brasseur (2003), respective results for impact on ozone largely agree. But as Granier and Brasseur (2003) did not account for road traffic NMHC emissions, they underestimated the total impact of road traffic emissions on ozone. The sensitivity experiment for NMHC emissions (No_NMHC) revealed that in remote areas NMHCs act as an additonal source for road traffic induced ozone. In these areas ozone is formed due to long-range transport of PAN from NMHC road traffic emissions.
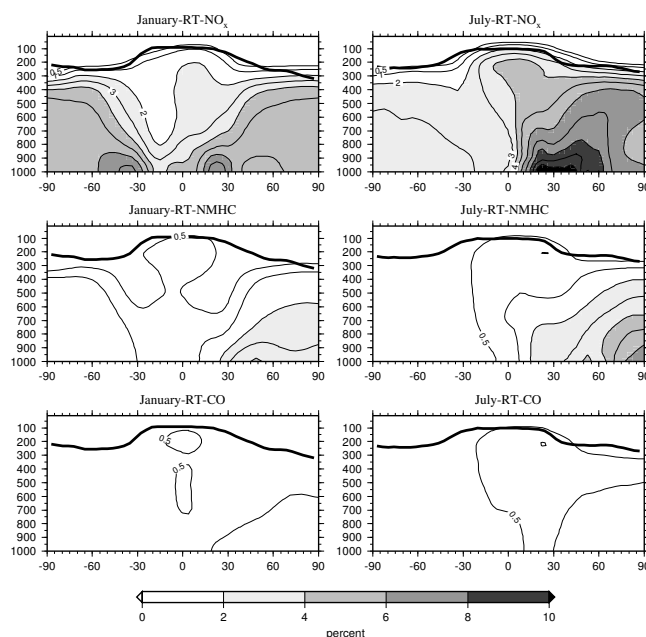

Fig. 7. Contributions [%] of individual road traffic emissions – NO_x (upper), NMHCs (middle)) and CO (lower) to atmospheric distribution of ozone. Isolines are equidistant with 1% difference, plus one additional line for 0.5%.

4.3 Role of road traffic NMHC emissions in PAN formation and long-range transport into Arctic latitudes

We have mentioned several times the importance of PAN as reservoir species for transferring NO_x emissions to regions far away from the sources. NMHC emissions from road traffic form PAN together with atmospheric NO_x in source regions. PAN is thermally stable at low temperatures and can be transported over long distances to remote regions. Subsidence causes an adiabatic heating, causing PAN to decay thermally and leads to additional NO_x. In low-NO_x regions this NO_x leads to ozone production and as a consequence causes high relative contributions of road traffic emissions to zonally averaged ozone concentrations in arctic regions of more than 6% (Fig. 7, middle row).

As one example for remote regions, the regionally averaged concentration changes of PAN due to road traffic emissions are shown in Fig. 8 in both northern hemispheric mid-latitudes and arctic regions. A strong PAN enhancement is indicated due to all road traffic emissions (solid lines), a slightly weaker due to NMHC emissions only (dashed lines) and clearly weaker due to NO_x emissions only (dotted lines). In spring, in arctic (PN) and mid-latitudes (NHE), total road traffic emissions are responsible for a PAN enhancement of 75 ppt (solid line, regional average north of 68° below 550 hPa), whereas NMHC emissions alone cause in arctic latitudes an increase of about 65 ppt (dotted line). On the other hand, NO_x emissions alone cause a PAN increase of
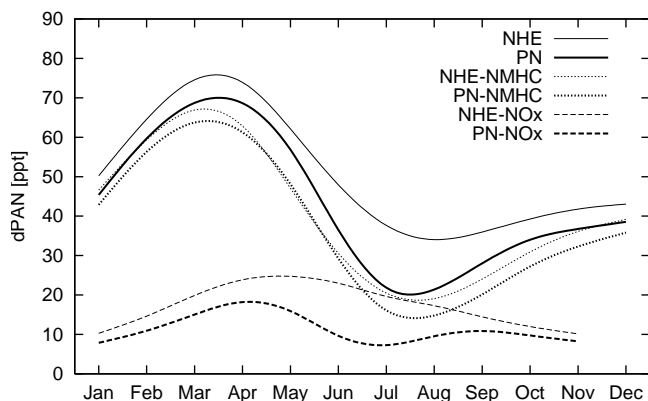
Fig. 8. Individual contributions of road traffic emissions to PAN in two regions: (1) mid-latitudes near the surface (NHE; 30° N–60° N, below 750 hPa) and (2) arctic regions (PN; 68° N–90° N, below 550 hPa); total (solid), NMHCs (dotted), NO_x (dashed)).
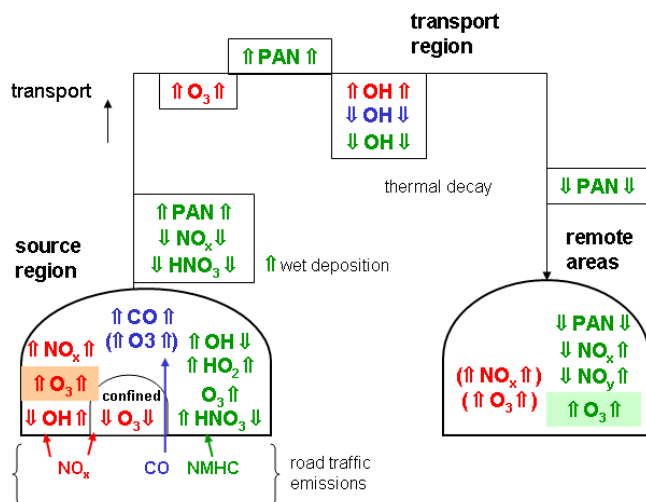

Fig. 9. Scheme illustrating the impact of individual components of road traffic emissions on atmospheric composition: NO_x (red), CO (blue), NMHCs (green). An arrow up indicates increase, down a decrease; the arrows on the left of the specie describe the change in winter, those on the right the change in summer. Weak processes are in parenthesis, major processes are highlighted with a coloured background.

only about 20 ppt (dashed line). In the following we use these results from individual components scenarios for deducing an estimate on 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%. NMHC emissions are responsible for about 90% of PAN enhancement in winter and springtime in arctic latitudes (PN). Due to higher atmospheric temperatures in spring and summer, the absolute size of this effects decreases from April on. In summer absolute values only amount to about 20 ppt contribution due to gaseous road traffic emissions (NO_x, NMHCs and CO). Nevertheless NMHCs still represent the major contributor with about 50%. Hence Fig. 8 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.

The synthesis of the role for the impact of all three individual emission components is illustrated in Fig. 9 for the three different regions: source regions (e.g. Europe), transport regions (free troposphere) and remote regions (e.g. Atlantic ocean, Arctic). The scheme includes the impact on the Hydroxylradical. In source regions, NO_x emissions from road traffic (red colored) are mainly important for ozone production depending on season. This causes a decrease in winter (January) and an increase in summer (July) of the concentrations of the atmospheric hydroxyl radical. In strongly confined regions (high-NO_x) increasing HO_x can strongly reduce ozone productivity and overcompensate the NO_x increase due to road traffic emissions, causing a decrease of ozone. In source regions, generally NMHC emissions convert additional OH into HO_2 during their oxidation (R5). Radicals formed during the oxidation process produce more HO_x than they consume within their initial reaction. The enhanced HO_x increases ozone productivity of nitrogen oxides. In winter HO_x increases so strongly in source regions of the

Northern Hemisphere that – in spite of a relative decrease of HO_x – OH increases. The increased OH concentrations form an enhanced sink for NO_x and more HNO_3 is produced. In transport regions PAN concentrations are enhanced and NO_x are reduced due to NMHC emissions from road traffic. On the other hand, HNO_3 concentrations are reduced due to wet deposition in transported air masses. During subsidence of air masses PAN decomposes thermally and the PAN concentration decreases. In remote regions the above mentioned impact of NMHCs makes these species responsible for a major ozone increase.

Figure 5 showed that ozone destruction due to road traffic NO_x emissions occurs in the industrialized areas in Central Europe and North America. Since NMHC road traffic emissions cause long-range transport of NO_x (see above) the geographic origin of emissions is of primary interest. Therefore, we performed studies of regional road traffic emissions (NO_x, NMHCs, CO): USA only or Europe only (No_rtusa and No_rteur in Table 2), which will now be discussed.

4.4 Impact of regional emissions from the USA and Europe

The results of the USA and Europe related impact simulations are shown in Fig. 10. The meriodionally averaged, relative contributions to atmospheric ozone (Fig. 10, upper row) indicate that long-range transport occurs eastward. In the free troposphere (above 900 hPa) over the USA in mid-latitudes, global road traffic contributes by about 8% (6 ppb), whereas emissions from the USA itself contribute more than 4% (3 ppb). In the free troposphere over the Atlantic, 4%–8%
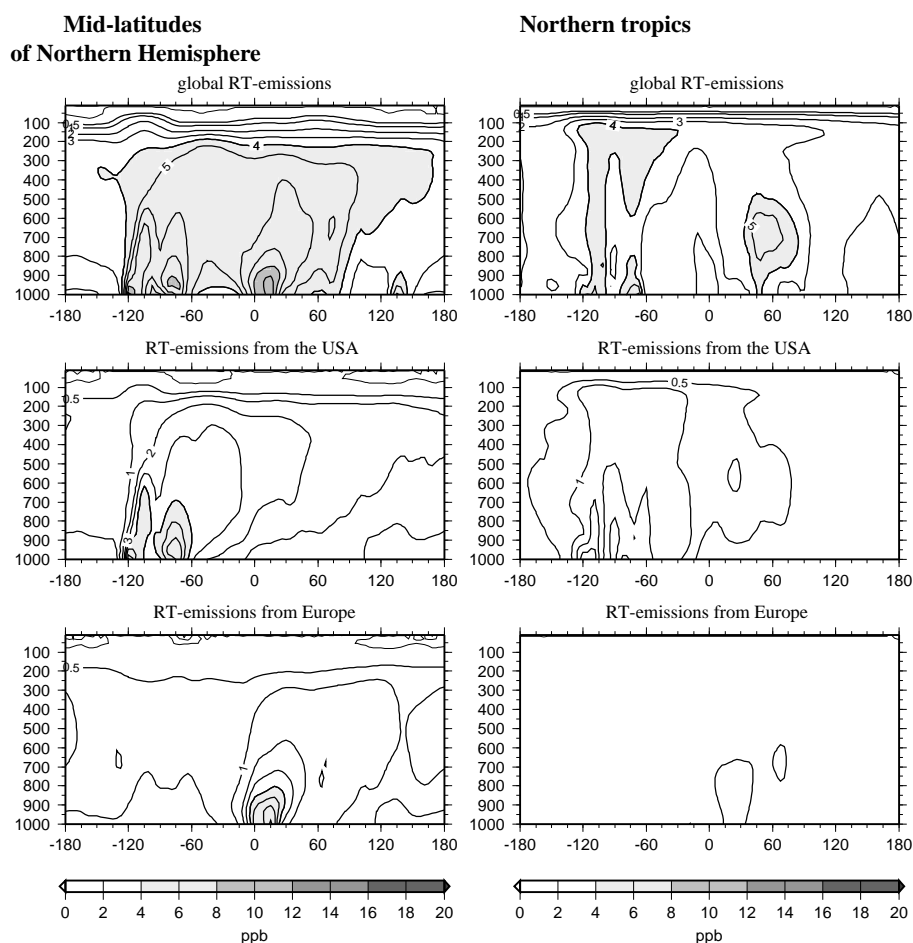
Fig. 10. Contributions [ppbv] of road traffic (RT) emissions to meridionally averaged ozone: global emissions (upper), emissions from the USA (middle) and emissions from Europe (lower) in July. Mid-latitudes denote meridionally averaging over 30° N–60° N and northern tropics over Equator – 30° N; isolines distance 1 ppb, plus one additional at .5 ppb.

(2–4 ppb) of the ozone concentration has its origin in road traffic emissions in USA and about 2% (1 ppb) originates from Europe, out of a road traffic contribution from global road traffic emissions of 8% to 12% (4–5 ppb). In the free troposphere over Europe in midlatitudes, emissions from the USA still contribute by more than 3% (2 ppb), where the impact of global road traffic emissions amounts to 8% (5 ppb) (700 hPa–300 hPa; 30° E). and road traffic emissions from Europe contribute by up to 4% (2 ppb). Further east (30° E– 130° E) in the free troposphere relative contributions from Europe and the USA have about the same size of 2% (1 ppb) here; global emissions contribute more (about 8% or 5 ppb). Over Asia, where global road traffic emissions contribute up to 8% (4 ppb) to mid-latitude ozone abundance a contribution of about 2% (1 ppb) due to road traffic emissions in the USA can be found (Fig. 10, middle row) whereas on the other hand, emissions from Europe contribute there about 2% (0.5 ppb). Results show that Europe and USA road traffic emissions can represent up to 50% of the global road traffic

contribution to the ozone distribution in mid-latitudes in the free troposphere. Surface ozone concentrations are only significantly affected by long-range transport in remote areas.

In northern tropics (Fig. 10, right panels), global road traffic emissions contribute more than 8% (4 ppb) above the USA/Central America to atmospheric ozone. Road traffic in the USA represents with around 2% (1 ppb) only about a quarter of these total ozone contributions (up to 200 hPa in tropical latitudes). European emissions from road traffic show a maximum relative contribution of 1% (0.5 ppb) around 30° E (Arabian Peninsular), clearly illustrating the minor importance of European road traffic emissions for the tropics. The sensitivity studies show the strong zonal transport of road traffic impact on ozone, and the much weaker meridional transport. The results are consistent with the findings of Wild and Akimoto (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 more than 2% contribution from USA and Europe each. For Asia, Wild et al. found that one third of the upper troposphere ozone changes arise from European and USA emissions, agreeing with our findings showing a 1.5 ppb contribution of European and USA road traffic contributions to ozone out of a total of 4 ppb.

5   Summary and conclusions

Our results indicate that in July 1990 road traffic emissions (NO_x, CO, and NMHCs) contribute to the zonally averaged tropospheric ozone concentration by more than 12% in Northern Hemisphere midlatitudes and arctic latitudes. In January, road traffic contributes near the surface both in northern and southern extratropics more than 8%. Relative contributions near the surface to northern hemispheric midlatitudes ozone amount to more than 12% in remote regions. The simulations with ECHAM4/CBM show that the exclusive consideration of NO_x for assessing the impact of road traffic emissions neglects an important impact due to NMHC emissions. This holds in particular for the consequences of long range transport of emissions for atmospheric ozone in remote regions which is underestimated by about 30% when only considering NO_x and CO emissions. For assessing the climate impact of road traffic emissions NMHCs have to be considered. Our regional studies have emphasised the regional and long-range contributions to ozone enhancement in the free troposphere, showing that global emission distributions have to be well known even for regional ozone studies in the free troposphere. Future increase in southern hemispheric regions, e.g. due to economic development in these regions will lead to comparative high ozone contributions to atmospheric ozone.

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References

Atkinson, R.: Gas-phase tropospheric chemistry of organic compounds: a review, Atmos. Environ., 24A, 1–41, 1990.

Benkovitz, C., Scholtz, M., and Pacyna, J.: Global gridded inventories of anthropogenic emissions of sulfur and nitrogen, J. Geophys. Res., 101, 29 239–29 253, 1996.

Crutzen, P., Lawrence, M., and Pöschl, U.: On the background photochemistry of tropospheric ozone, Tellus, 51A-B, 123–146, 1999.

Gery, M., Whitten, G., Killus, J., and Dodge, M.: A photochemical kinetics mechanism for urban and regional scale modelling, Geophys. Res. Lett., 94, 12 925–12 956, 1989.

Granier, C. and Brasseur, G.: The impact of road traffic on global tropospheric ozone, Geophys. Res. Lett., 30, 1086, doi:10.1029/2002GL015972, 2003.

Hao, W. and Liu, M.: Spatial and temporal distribution of tropical biomass burning, Global Biogeochem. Cycles, 8, 495–503, 1994.

Houweling, S., Dentener, F., and Lelieveld, J.: The impact of nonmethane hydrocarbon compounds on the tropospheric photochemistry, J. Geophys. Res., 103, 10 673–10 696, 1998.

IPCC: Special Report Emission Scenarios, Tech. rep., Intergovernmental Panel on Climate Change, Cambridge University Press, New York, NY, USA, 2000.

Lauer, A., Dameris, M., Richter, A., and Burrows, J.: Tropospheric NO_2 columns: a comparison between model and retrieved data from GOME measurements, Atmos. Chem. Phys., 2, 67–78, 2002,
http://www.atmos-chem-phys.net/2/67/2002/.

Liu, S., McFarland, M., Kley, D., Mahlman, J., and Levy II, H.: On the origin of tropospheric ozone, J. Geophys. Res., 85, 15 879–15 888, 1980.

Liu, S., Trainer, M., Fehsenfeld, F., Parrish, D., Williams, E., Fahey, D., Hübler, G., and Murphy, P.: Ozone production in the rural troposphere and the implication for regional and global ozone distribution, J. Geophys. Res., 92, 4191–4207, 1987.

Martin, R., Chance, K., Jacob, D., Kurosu, T., Spur, R., Bucsela, E., Gleason, J., Palmer, P., Bey, I., Fiore, A., Li, Q., Yantosca, R., and Koelemeijer, R.: An improved retrieval of tropospheric nitrogen dioxide from GOME, J. Geophys. Res., 107(D20), 4437, doi:10.1029/2001JD001027, 2002.

Matthes, S.: Globale Auswirkung des Straßenverkehrs auf die chemische Zusammensetzung der Atmosphäre, Ph.D. thesis, Ludwig-Maximilians Universität München, 2003.

Matthes, S. and Sausen, R.: Abschätzung der Emissionen in den EU-15 Staaten im Zeitraum 1980 bis 2015, Tech. rep., German Space Center (DLR), Oberpfaffenhofen, Germany, 2000.

OECD: Motor vehicle pollution, Tech. rep., OECD Organisation for economic co-operation and development, Paris, France, 1995.

Olivier, J., Bouwman, A., van der Maas, C., Berdowski, J., Veldt, C., Bloos, J., Visschedijk, A., Zandveld, P., and Haverlag, J.: Description of EDGAR 2.0: A set of global emission inventories of greenhouse gases and ozone depleting substances for all anthropogenic and most natural sources on a per country basis on a $1° \times 1°$ grid, Tech. rep., RIVM, Bilthoven, Netherlands, 1996.

Richter, A. and Burrows, J.: Retrieval of Tropospheric NO_2 from GOME Measurements, Adv. Space Res., 29, 1673–1683, 2002.

Roeckner, E., Arpe, K., Bengtsson, L., Christoph, M., Claussen, M., Dümenil, L., Esch, M., Giorgetta, M., Schlese, U., and Schulzweida, U.: The Atmospheric General Circulation Model ECHAM-4: Model Description and Simulation of Present-day Climate, Tech. rep., Max-Planck-Institute for Meteorology, Hamburg, Germany, 1996.

Roelofs, G.-J. and Lelieveld, J.: A three dimensional chemistry-general circulation model: Influence of higher hydrocarbon chemistry, J. Geophys. Res., 105, 22 697–22 712, 2000.

Wild, O. and Akimoto, H.: Intercontinental transport of ozone and its precursors in a three-dimensional global CTM, J. Geophys. Res., 106, 27 729–27 744, 2001.