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**Traffic emissions:  
impact on ozone and  
OH**

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# The impact of traffic emissions on atmospheric ozone and OH: results from QUANTIFY

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Received: 9 July 2008 – Accepted: 1 September 2008 – Published: 21 October 2008

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Published by Copernicus Publications on behalf of the European Geosciences Union.

## Abstract

To estimate the impact of emissions by road, aircraft and ship traffic on ozone and OH of the present-day atmosphere seven different atmospheric chemistry models simulated the atmospheric composition of the year 2003. Based on newly developed global emission inventories for road, maritime and aircraft emission data sets each model performed a series of five simulations: A base scenario using the full set of emissions, three sensitivity studies with each individual sector of transport reduced by 5% and one simulation with all traffic related emissions reduced by 5%. The approach minimizes non-linearities in atmospheric chemical effects and are later scaled to 100%.

The global annual mean impact of ship emissions on ozone in the boundary layer leads to an increase of ozone of 1.2%, followed by road (0.87%) and aircraft emissions (0.3%). In the upper troposphere between 200–300 hPa both road and ship traffic affect ozone by 1.1%, whereas aircraft emissions contribute 0.9%. However, the sensitivity of ozone formation per NO<sub>x</sub> molecule emitted is highest for aircraft exhausts.

The local maximum effect of the summed traffic emissions on the ozone column predicted by the models is 4.0 DU and occurs over the northern subtropical Atlantic. The impact of traffic emissions on total ozone in the Southern Hemisphere is approximately half of the northern hemispheric perturbation.

Below 800 hPa both ozone and OH respond most sensitively to ship emissions in the marine boundary layer over the Atlantic, where the effect can exceed 10% (zonal mean) which is 80% of the total traffic induced ozone perturbation. In the Southern Hemisphere ship emissions contribute relatively strongly to the total ozone perturbation by 60%–80% throughout the year (equivalent to 1–1.5 ppbv).

Road emissions have the strongest impact on ozone in the continental boundary layer and the free troposphere in summer. They also affect the upper troposphere particularly during northern summer associated with strong convection in mid latitudes. Ozone perturbations due to road traffic show the strongest seasonal cycle in the northern troposphere, and can even change sign in the continental boundary layer during winter.

ACPD

8, 18219–18266, 2008

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The OH concentration in the boundary layer is most strongly affected by ship emissions, which has a significant influence on the lifetime of many trace gases including methane. Methane lifetime changes due to ship emissions amount to 4.1%, followed by road (1.6%) and air traffic (1.0%).

## 1 Introduction

The rise in energy consumption by the growing human population and the increasing mobility are associated with emissions of air pollutants in particular by road and air traffic as well as international shipping. These emissions are expected to increase in future, affecting air quality and climate (Kahn Ribeiro et al., 2007) and vice versa (Hedegaard et al., 2008). The impact of air traffic emissions has been subject of various investigations (e.g. Hidalgo and Crutzen, 1977; Schumann, 1997; Brasseur et al., 1996; Schumann et al., 2000) also assessing projections for the future (e.g. Sovde et al., 2007; Grewe et al., 2007). For the present day atmosphere these studies indicated an increase of ozone of 3–6% due to aircraft emissions in the region of the North Atlantic flight corridor. More recent studies calculated an overall maximum effect of 5% for the year 2000 in the northern tropopause region (Grewe et al., 2002). Depending on season the values typically range between 3 ppbv and 7.7 ppbv in January and September, respectively (Gauss et al., 2006). The radiative forcing due to the additional O<sub>3</sub> from air traffic is estimated to be of the order of 20 mW/m<sup>2</sup> (Sausen et al., 2005).

Relatively few studies have dealt with the impact of road traffic (Granier and Brasseur, 2003; Niemeier et al., 2006; Matthes et al., 2007), and ship emissions (Lawrence and Crutzen, 1999; Corbett and Koehler, 2003; Eyring et al., 2005; Dalsoren and Isaksen, 2006; Endresen et al., 2007; Eyring et al., 2007). Endresen et al. (2003) reported peak ozone perturbations of 12 ppbv for the marine boundary layer during northern summer over the northern Atlantic and Pacific regions. Eyring et al. (2007) used a multi-model approach based on EDGAR emissions and reported somewhat lower values of

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5–6 ppbv for the North Atlantic. They also calculated a maximum column perturbation of 1 DU for the tropospheric ozone column associated with radiative forcings of 9.8 mW/m<sup>2</sup>. For road emissions Matthes et al. (2007) maximum contributions to surface ozone peaking at 12% in northern midlatitudes during July. Similar values of 10% are reported by Niemeier et al. (2006) for current conditions.

Besides the effects of pollutants on ozone a potential change of the OH concentration is of importance in particular for regional air quality and the self-cleaning capacity of the atmosphere (Lelieveld et al., 2002). Changes of methane loss rates due to anthropogenic emissions are reported to be on the order of 0.5%/yr of which 1/3 (0.16%) is due to an increase of OH from anthropogenic CO, NO<sub>x</sub> and non-methane hydrocarbons (NMHCs) (Dalsoren and Isaksen, 2006). In addition, the regional OH distribution can differ substantially due to the short lifetime of OH and NO<sub>x</sub> in particular in the lower troposphere and the different response of the HO<sub>x</sub>-NO<sub>x</sub>-O<sub>3</sub>-system to NO<sub>x</sub> perturbations (Lelieveld et al., 2002, 2004). Although generally the presence of carbon compounds such as CH<sub>4</sub>, NMHCs, and CO act as a sink for OH, the latter can be efficiently recycled under high NO<sub>x</sub> conditions. The reaction of NO with HO<sub>2</sub> produces O<sub>3</sub> and recycles OH making the system less sensitive to perturbations. In pristine regions with low NO<sub>x</sub> and high OH concentrations conditions are favourable for OH-formation following ozone production from NO<sub>x</sub>-perturbations. Since emissions from the three transport sectors are emitted into rather different environments their impact on O<sub>3</sub> and OH may differ strongly.

Thus, no study has consistently investigated the combined effects of these three modes of transport and directly compared their influences on the current composition of the atmosphere. The EU-project QUANTIFY (Quantifying the Climate Impact of Global and European Transport Systems) is the first attempt to provide an integrated view on the effects of traffic on various aspects of the atmosphere. This study focuses on the global impact of traffic exhaust emissions on the current chemical state of the atmosphere. The climatological implications and future projections are subject of followup studies.

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## 2 Emissions and simulation setup

Emissions for the three transport sectors road, shipping and air traffic were replaced in the EDGAR data base with respect to the individual emission classes and recalculated separately for QUANTIFY. Emissions from road transportation were developed bottom-up: Vehicle mileage and fleet average emission factors were estimated for the year 2000. The inventory differentiates between five vehicle categories and four fuel types and covers 12 world regions and 172 countries. It is adjusted for the year 2000 to the national road fuel consumption (Borken et al., 2007, and references therein) or national statistics or own calculations. National emissions are allocated to a  $1^\circ \times 1^\circ$ -grid essentially according to population density with emissions from motorised two- and three-wheelers biased towards agglomerations and emissions from heavy duty vehicles biased towards rural areas. The emissions used in this study are based on a draft version (Borken and Steller, 2006). The final inventory includes improved emission factors. With fuel consumption and CO<sub>2</sub> emissions only 3% higher, the final global emissions of NO<sub>x</sub>, NMHC and CO are higher by 33%, 48% and 51%, respectively. The integrated annual emissions which were used in this study for selected species are given in Table 1.

The emissions for ship traffic were reconstructed for QUANTIFY based on fuel- and activity-based estimates by Endresen et al. (2007). They report a fuel consumption being about 80 Mt lower than in Eyring et al. (2005) or Corbett and Koehler (2003). They attribute the difference to the different assumptions for the operation at sea. For further details see Endresen et al. (2007).

Aircraft emissions are based on the AERO2K dataset (Eyers et al., 2004) including the emissions from military aviation. The emissions for each transport sector are given in Table 1.

Non-traffic emissions used in the present modelling exercise are based on the latest release of the EDGAR32FT2000 emission inventory (van Aardenne et al., 2005; Olivier et al., 2005) including emissions of greenhouse gases and ozone precursors for the

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year 2000 with the exception of methane which was prescribed as a surface boundary condition.

For biomass burning monthly means for the year 2000 were used based on GFED estimates with multi-year (1997–2002) averaged activity data using Andreae and Merlet (2001) and Andreae (2004, personal communication) for  $\text{NO}_x$  emission factors (BB-AVG-AM). Lightning  $\text{NO}_x$  was specified at 5 TgN/year, representing the current best estimate (Schumann and Huntrieser, 2007).

NMHC's were subdivided into individual organic and partly oxidized species. The partitioning of the NMHC's was performed according to von Kuhlmann et al. (2003a) and is shown in Table 2 for biomass burning and fossil fuel related emissions. The mass of NMHC's given in kg(NMHC)/yr was converted to kg(C)/year using a ratio of 161/210 for molecules(C)/molecules(NMHC). The specific ratios were then applied to calculate the individual NMHC partitioning.

Biogenic emissions for isoprene and NO emissions from soil were also included based on online calculations with the ECHAM5/MESSy model (Ganzeveld et al., 2006; Kerweg et al., 2006) and provided as offline fields to all partners.

To compare the emissions provided by QUANTIFY with other projects and data bases some recently used inventories are also given in Table 1. Notably the road traffic emissions are lower than in other inventories. The final road traffic emissions by Borcken et al. (2007) are higher by 33% for  $\text{NO}_x$  and about 50% for CO emissions than those used in this study. Nonetheless, low total CO emissions are a result of lowered average emission factors accounting for effective use of catalytic converters in light duty vehicles.

The resulting source strengths of  $\text{NO}_x$  from the different sectors, which were used in QUANTIFY are shown in Fig. 1. Globally, road  $\text{NO}_x$  emissions are dominated by the eastern US and western Europe as well as India and eastern China, the latter with an increase rate of 6%/year (Ohara et al., 2007) only between 2000 and 2003. Besides the continental coast lines on the Northern Hemisphere and around the major shipping routes ship traffic over the northern central Atlantic between 25°–55° N is a significant

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source of NO<sub>x</sub> over a large area. A second NO<sub>x</sub> source from ship emissions covering a large area is traffic along the east coast of Asia. Besides the continental eastern US and western Europe the maximum emissions from air traffic also occur over the North Atlantic, though further north than the shipping maxima. In the Southern Hemisphere

NO<sub>x</sub> emissions are largely dominated by non-traffic sources and biomass burning.

The simulation period covers the years 2002 and 2003 with 2002 as spin-up. Each participating model calculated the chemical state of the atmosphere for present day conditions using all emissions as described above. The perturbation simulations were performed by reducing the emission of each individual transport sector by 5% (see Table 3). This relatively small reduction was applied to avoid nonlinear responses of the chemical system which would occur by setting the respective emissions to zero. To check for linearity a final simulation was carried out with all transport sectors simultaneously reduced by 5%. Post processing confirmed the linearity of the small scale perturbation approach allowing to integrate the effects of the individual transport sectors in this setup.

In the following all perturbations are scaled to 100 % unless explicitly mentioned otherwise.

### 3 Participating models

Six models were applied to estimate the effect of traffic emissions on the current atmospheric chemical composition. Five of them simulated a two years period and included higher order chemistry schemes. One model (E39/C) was used in a different mode to provide information on the interannual variability of the traffic impact using a ten year transient simulation. Four models are CTMs using prescribed operational ECMWF data to simulate the meteorological conditions (TM4, p-TOMCAT, OsloCTM2 and UCI).

The other two models are coupled CCMs (chemistry climate models). LMDzINCA was nudged to the operational ECMWF fields whereas E39/C was operated as a climate model. Some general properties of the models are listed in Table 4. Except for E39/C

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all models included explicit NMHC chemistry. The TM4 uses a Carbon Bond Mechanism reaction scheme (CBM4) not including acetone chemistry. Three of the models do not include stratospheric chemistry reactions (TM4, p-TOMCAT, LMDzINCA). The number of species ranged from 42(TM4) to 125(LMDzINCA). LMDzINCA and TM4 used their own biogenic and oceanic emissions, respectively (see below).

### 3.1 TM4

The KNMI chemistry transport model TM4 (van Noije et al., 2006a,b) is driven by ECMWF analysed meteorology and contains a chemistry scheme derived from the Carbon Bond Mechanism reaction scheme (CBM4) (Houweling et al., 1998). It was run at a horizontal resolution of  $2 \times 3$  degrees with 34 model levels from the surface up to 10 hPa.

The lightning parameterisation (Meijer et al., 2001) uses convective precipitation from ECMWF to describe the horizontal distribution of lightning and normalised profiles calculated by Pickering et al. (1998) to distribute lightning produced  $\text{NO}_x$  vertically between cloud base and cloud top.

Vertical emission profiles were also adopted as included in the respective emission files. In addition to the QUANTIFY emissions oceanic emissions were taken from the POET emission inventory, ammonia emissions from EDGAR 2.0, and volcanic  $\text{SO}_2$  and DMS (Dimethylsulfide) emissions from the standard model configuration of TM4.

### 3.2 LMDzINCA

The CNRS-LSCE model, LMDz-INCA had a resolution of  $3.75^\circ$  in longitude and  $2.5^\circ$  in latitude with 19 levels extending from the surface up to about 3 hPa and is driven by ECMWF operational data (Hauglustaine et al., 2004).

The NMHC setup of the LMDz-INCA model was used. It considers detailed tropospheric chemistry with a comprehensive representation of the photochemistry of non-methane hydrocarbons (NMHC) and volatile organic compounds (VOC) from biogenic,

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anthropogenic, and biomass burning sources.

Most anthropogenic emissions were taken from the EDGAR3.2FT2000 database. The effective injection height of biomass burning emissions into the atmosphere was taken into account with the emission heights calculated in the RETRO (REanalysis of the TROpospheric chemical composition over the past 40 years) project. The lightning source was determined interactively in LMDz-INCA with a modified Price and Rind (1992) parameterization, and its total was prescribed at  $\approx 2$  Tg[N]/year. Biogenic sources were calculated with the vegetation model ORCHIDEE. Oceanic emissions were taken from Folberth et al. (2005).

### 3.3 OsloCTM2

The Oslo CTM2 model is a 3-D chemical transport model driven by ECMWF meteorological data and extending from the ground to 10 hPa in 40 vertical layers. The horizontal resolution for this study was Gaussian T42 ( $2.8^\circ \times 2.8^\circ$ ). The model was spun up for several years with emissions from the POET and RETRO projects, then for additional 6 months with the emissions provided for the QUANTIFY project. One restart file was archived for March 2002. From this file the five different scenarios were started, which were run for 22 months.

### 3.4 p-TOMCAT

The model used during the first part of the QUANTIFY project is the global offline chemistry transport model p-TOMCAT. It is an updated version (see O'Connor et al. (2005)) of a model previously used for a range of tropospheric chemistry studies (Savage et al., 2004; Law et al., 2000, 1998). Convective transport was based on the mass flux parameterization of Tiedtke (1989). The parametrizations includes descriptions of deep and shallow convection with convective updrafts and largescale subsidence, as well as turbulent and organized entrainment and detrainment. The model contains a nonlocal vertical diffusion scheme based on the parameterization of Holtslag and Boville (1993).

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In this study p-TOMCAT was run with a  $5.7^\circ \times 5.7^\circ$  horizontal resolution and 31 vertical levels from the surface to 10 hPa. The offline meteorological fields used are from the operational analyses of the European Medium Range Weather Forecast model. The chemical mechanism includes the reactions of methane, ethane and propane plus their oxidation products and of sulphur species, it includes 96 bimolecular, 16 termolecular, 27 photolysis reactions and 1 heterogeneous reaction on sulphuric acid aerosol. The model chemistry uses the atmospheric chemistry integration package ASAD (Carver et al., 1997) and is integrated with the IMPACT scheme of Carver and Scott (2000). The ozone and nitrogen oxide concentrations at the top model level are constrained to zonal mean values calculated by the Cambridge 2-D model (Law and Nisbet, 1996). The chemical rate coefficients used by p-TOMCAT have been recently updated to those in the IUPAC Summary of March 2005. The model parameterizations of wet and dry deposition are described in Giannakopoulos et al. (1999).

### 3.5 UCI

The UCI CTM is a 3-D eulerian chemistry-transport model driven by meteorological data from the ECMWF IFS version cycle 29r2 at T42L40 (see OsloCTM2). Tropospheric chemistry is handled by ASAD software package (Carver et al., 1997), containing 38 species (28 transported) and 112 reactions (Wild et al., 2003), while in the stratosphere, it employs the stratospheric linear ozone scheme (Linoz) and conducts linear calculations of (P-L) (Hsu, 2004). Photolysis rates are calculated by the Fast-JX package (Bian and Prather, 2002). Transport scheme uses the second order moments (Prather, 1986). Lightning is parameterized with the method of Price and Rind (1992). Convection is simulated, following the ECMWF Tiedtke convection diagnostics.

### 3.6 E39/C

Impacts by road and ship traffic were provided by DLR based on simulations with E39/C. The perturbation fields are derived as monthly mean values from a transient

simulation from 1990 to 1999 (Dameris et al., 2005; Grewe, 2007). The meteorology is calculated by the climate model ECHAM4.L39(DLR) and therefore does not represent an individual year. Nevertheless, it represents the late 20 century climate and is therefore to some extent comparable to the other simulations of the year 2003. In particular the emissions which were used by the other participating CTMs are different. The impacts are derived using tagging methods (Grewe, 2004) and hence differ from the 5% change approach used by other modelling groups in QUANTIFY. However, both methods try to identify the individual contributions from sectors. Changes in the contributions are detected comparably by both approaches (Grewe, 2004), i.e. the standard deviation based on interannual variability is similar for both approaches.

## 4 Traffic induced ozone changes

### 4.1 Total ozone perturbation

The integrated effect of the emission reduction (ALL-case) by 5% is shown in Fig. 2 for January and July, respectively. The column ozone distribution change was integrated from the surface up to 50 hPa. The results from E39/C are not included in the calculation of the mean fields since the results of E39/C were obtained with a different setup and a different method.

The mean total ozone perturbations in Fig. 2 exhibit strong hemispheric differences of the traffic emissions with almost zero effect in the Southern Hemisphere, but maximum effects of about 4 DU during Northern Hemisphere summer (3 DU during winter). All models simulate a similar location of the strongest ozone perturbation extending from the northern subtropical Atlantic to central Europe.

The maximum ozone perturbation is strongest pronounced during northern summer. Interestingly its southern hemispheric seasonal cycle – despite being weak – is in phase with the Northern Hemisphere with a maximum effect of less than 2 DU. This is most likely due to interhemispheric transport and mixing of ship and road emissions

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occurring during northern summer, which exceed the effects of emissions on the Southern Hemisphere (see also Fig. 5).

Note furthermore that the effects of traffic emissions over the Pacific and Indian Ocean downwind of the densely populated coastal areas and sources of pollution, are not as strong as over the central Atlantic Ocean. A comparison with the  $\text{NO}_x$  emissions (Fig. 1) indicates that over the northern hemispheric Atlantic in particular ship traffic and also aircraft emit large amounts of  $\text{NO}_x$  and that these high emissions occur over a relatively large area. Over the eastern US and western Europe the sum of all three emission categories reach a maximum. Their combination is responsible for the relatively strong perturbations in these regions.

To assess the robustness of the perturbation signal two tests are performed. First the model to model differences in terms of the associated standard deviation is assessed to estimate the impact of model uncertainties. Second, the impact of the chosen meteorology (year 2003) is tested by comparing to the interannual variability of the transport signal derived from the transient E39/C simulation, which includes natural variability of the troposphere and the stratosphere due to variations of ozone influx, transport patterns (e.g. induced by El Nino), and others (Grewe, 2007). The inter-model standard deviations (one- $\sigma$ ) are shown in Fig. 2 (middle). Overall the models calculate very similar patterns with a relative standard deviation mostly below 15% over large regions of the globe where the effect of the perturbation is strongest. In the Southern Hemisphere, the relative standard deviations are in general somewhat higher (between 20–30%), since the absolute column perturbations are not as large as in the Northern Hemisphere. Largest deviations occur over the tropical central pacific, where concentrations of ozone are still relatively low. Thus, small perturbations may lead to relatively large variations between the models. Note however, that calculated ozone perturbations in particular over Europe and the central Atlantic are relatively robust indicating a significant impact of traffic emissions in these regions.

To assess the interannual variability of the signal we compared the transport induced ozone changes to a ten year transient simulation of E39/C. As can be seen from Fig. 2

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(bottom) the traffic induced perturbation signal simulated by E39/C over the ten years period is on the order of 0.8 DU and 1.2 DU in January and July, respectively. Largest variations occur in coastal regions, where synoptic variability leads to advection of either relatively clean maritime air or air from polluted urban areas. Although the perturbation signal from E39/C is largest compared to the other models, the ensemble mean response of the models is still larger than the interannual variability of the transport induced ozone changes according to E39/C. Thus, focusing on a particular year is justified within a 10% uncertainty for ozone since this is the climatological variability simulated consistently within one model.

## 4.2 Zonal mean ozone perturbation

The zonal mean ozone perturbation for the integrated traffic emissions (Fig. 3) shows the largest effect in the northern subtropics/lower-middle latitudes. In the Northern Hemisphere boundary layer the mean ozone perturbation peaks at 5.5 ppbv in mid latitudes at 40–50° N during summer decreasing to less than 3 ppbv during winter. The perturbation mixing ratio peaks in the upper troposphere/lower stratosphere of the northern extratropics during summer, largely due to aircraft emissions as will be seen later (Fig. 4).

The relative perturbation in the upper troposphere is of the order of 4–6% during January and July. However, as indicated in Fig. 3 the strongest relative effect can be as large as 16% in the Northern Hemisphere boundary layer during summer. During winter the perturbation peaks at 10% and is located in the tropical boundary layer.

For the Southern Hemisphere the models calculate the strongest absolute effect on the ozone mixing ratio also for the upper troposphere, although the changes relative to the unperturbed case maximize in the marine boundary layer. In the Southern Hemisphere, both, absolute and relative changes are about 50% lower than in the Northern Hemisphere. The effect on ozone in the southern boundary layer is only about 2 ppbv during January and July.

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As illustrated in Fig. 3 largest uncertainties between the models are found near the tropopause and during summer when convection plays an important role in vertical transport in particular in the northern extratropics. Large emissions by traffic take place in that latitude belt and thus have the highest probability to be redistributed from the surface to higher altitudes via convection during the summer months. The associated variations between the individual models in that region range from 3.5 ppbv (LMDzINCA) to 6 ppbv (p-TOMCAT) at 250 hPa during July resulting in a standard deviation of 2 ppbv. In the extratropics of the Southern Hemisphere the effect of summer convection is not as pronounced due to the hemispheric differences of the emissions.

Interestingly, the interannual variability of the zonal mean ozone perturbations of around 0.5 ppbv ( $\approx 5\%$ ) for both January and July, is much smaller than the variability among the models ( $\approx 20\%$ ). This indicates that year-to-year changes in meteorology lead to changes in the horizontal pattern of ozone perturbations (and e.g. convection), but that the effect of vertical transport and mixing differs less, although horizontally displaced.

### 4.3 Effects by transport modes

The effects of the respective emissions from road, ship and aircraft on ozone are shown in Figs. 4–5 for January and July, respectively. The results indicate that the road emissions have the strongest effect on ozone in the summer boundary layer over the eastern US and central Europe extending over the Mediterranean to the Arabian Peninsula (Fig. 4). During winter the average effect of road traffic on ozone in the highly industrialized regions of the extratropics almost vanishes or is even inverted. Often under stable boundary layer conditions the emissions accumulate, on average to more than 2 ppbv over the industrialized centers over the eastern US, Europe and parts of East Asia (not shown). Under these conditions additional  $\text{NO}_x$  from (road) traffic locally leads to the titration of ozone. In addition the ozone production efficiency can be increased by re-

duced  $\text{NO}_x$  emissions under high  $\text{NO}_x$  conditions leading to a higher ozone productivity per emitted  $\text{NO}_x$ .

Matthes et al. (2007) and Niemeier et al. (2006) find larger relative ozone perturbations due to road traffic of about 10% for some regions in the Northern Hemisphere boundary layer performing similar model simulations. Part of the deviations to our results can be explained by the use of different emissions, which are based on the EDGAR 1990 fuel estimates in Matthes et al. (2007) and are about 25% higher for  $\text{NO}_x$  from road than in this study. The inverted sensitivity of ozone to road emissions during winter is also reported by Niemeier et al. (2006) with the maximum effect over Europe exceeding 25% at the surface. The latter effect is simulated by all five models in our study, which contribute to the average shown in Fig. 4. Quantitatively, the results of Niemeier et al. (2006) also exceed our calculations by a factor of 2–3 at the surface.

Ship emissions have a significant impact throughout the year also with a maximum in July over the central Atlantic/western European region. In the Northern Hemisphere their effect dominates the boundary layer perturbation during January and July. Over the northern Pacific region mainly the emission from ships contributes significantly to the ozone perturbation, whereas the North Atlantic region additionally is affected by road emissions, which are transported efficiently from the highly polluted Eastern US to the Atlantic.

Not too surprising, in the northern UTLS region the effect of aircraft emissions dominates the ozone perturbation (Fig. 5) being approximately half as large during January compared to July associated with the enhanced level of photochemistry. The ozone perturbation from road traffic during summer is even higher than the perturbation due to aircraft emissions in winter, highlighting the role of road traffic for the chemical state of the UTLS in summer (see also Fig. 6). Ship emissions, despite of the high importance in the July boundary layer (Fig. 4) do not show a similar effect in the UTLS, indicating that convective and large scale transport from the marine boundary layer do not have the same impact as continental convection for road traffic.

This is also evident from Fig. 6, which shows the temporal evolution of the ozone per-

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turbations for the different modes of transportation. Comparing ship and road induced ozone perturbations ship emissions have a larger impact on boundary layer ozone than road traffic over the whole year. In the upper troposphere and lower stratosphere the ozone perturbation from road traffic peaks in summer over in the northern mid latitudes exceeding the effect on the boundary layer. The effect of ship emissions does not exhibit this strong seasonal signal in the upper troposphere although their effect on surface ozone shows a similar zonal and temporal distribution as for road. Globally the effect of ship emissions on ozone in the boundary layer larger than those from road emissions, whereas both are almost equal in the upper troposphere (Table 5). Notably in the upper troposphere aircraft emissions do not dominate the annual mean global effect between 200–300 hPa. Their influence is rather concentrated to latitudes  $>40^{\circ}$  N (see. Fig. 5) whereas road and ship traffic affect the UT globally.

These findings are also evident in the vertical zonal mean cross sections (Fig. 5), highlighting the importance of surface emissions from ship and road traffic for the tropical upper troposphere. It also shows that the impact of road traffic is of larger importance for the extratropical troposphere in the northern summer hemisphere. In the tropics ship emissions are the strongest contributor to the ozone perturbation in the tropical transition layer (TTL), being of greater importance than aircraft emissions there. As evident from Fig. 5 the impact of road and ship emissions seems to influence the whole TTL region from  $20^{\circ}$  S– $20^{\circ}$  N in particular during northern summer. During January, when the Inter Tropical Convergence Zone (ITCZ) is located in the Southern Hemisphere, where the emissions are much lower, the TTL is only weakly affected. Note also that for the upper troposphere in the Southern Hemisphere the effect of ship emissions is among all modes of transport the strongest contributor in January as well as in July (comp. also Fig. 6).

#### 4.4 Relative importance of traffic sectors

In addition to the perturbation of  $O_3$  the contribution of the different transport sectors relative to the total effect from traffic is of interest. As shown in Fig. 7 the relative im-

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5 pact of road and ship emissions differ substantially although both are released at the surface. In the region between the surface and 900 hPa the relative effect of ship emissions clearly dominates the total perturbations by all traffic emissions in each summer hemisphere. In the Southern Hemisphere during January the perturbations of ship emissions can almost contribute up to 85% of the total ozone perturbation, since ship emissions are the major source of pollutants south of 40° S. In the northern extratropics the effects from ships are most important in the boundary layer below 950 hPa, where the mean perturbation relative to the background can be as high as 16% (see also Fig. 7). As evident from Fig. 7 the contribution of ship emissions is particularly important in the subtropical monsoon regions and the tropics, where convection leads to upward transport of the emissions and to a relative contribution of 50% up to the tropical tropopause.

The relative contribution of road emissions peaks in the free troposphere in both summer hemispheres with a pronounced seasonal cycle in the northern extratropics. The maximum effect on ozone occurs in July, when convection at these latitudes is strongest. Thus during that time of the year pollutants from road traffic have a much higher probability to be transported to tropopause altitudes compared to winter. This is also evident from Fig. 5 where the ozone perturbations in July below the tropopause are almost equal to the effect from aircraft.

20 Note that ship emissions do only weakly affect the free and upper troposphere of the northern extratropics during July. Since ships largely emit over the open oceans where convection is not as strong as over the continents, they have a much lower potential to reach the upper troposphere of the extratropics compared to road emissions. Therefore, in the winter hemispheres the effects of road and ship traffic on ozone show similar patterns (Fig. 5). As indicated by Fig. 7 the relative contribution of road traffic on the free and upper extratropical troposphere is about 15% larger than that of ships. Directly at the tropopause and in the lower stratosphere the contribution of aircraft dominates in the Northern Hemisphere, whereas in the Southern Hemisphere upper troposphere its effect on ozone is the smallest of the three types of traffic.

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To estimate the efficiency of the ozone perturbation from each transport sector we normalized the ozone perturbation of each sector to the respective number of  $\text{NO}_x$  molecules assuming a global average lifetime of 22 days for ozone (Stevenson et al., 2006). As evident from Table 6 road and ship emissions have similar efficiencies to perturb ozone. However, the  $\text{NO}_x$  emissions from air traffic are more than twice as efficient in producing ozone. Since these emissions take place in the UTLS region the lifetime of the reservoir species  $\text{HNO}_3$  and PAN are much longer than at the surface. Therefore each  $\text{NO}_x$  molecule can be recycled more often to produce ozone before being removed via precipitation scavenging and dry deposition of  $\text{HNO}_3$ .

Note that despite some individual differences, all models indicate the largest efficiency to perturb ozone for  $\text{NO}_x$  emissions from aircraft and the weakest efficiency for road traffic.

## 5 OH

### 5.1 Global OH

The effects of different transport systems on tropospheric OH concentrations are shown in Fig. 8 for January and July, respectively. Interestingly, ship emissions have the largest impact on the global OH budget in the boundary layer. During northern summer the effect of  $\text{NO}_x$  from maritime traffic on OH can be as large as  $5 \cdot 10^5$  molecules/ $\text{cm}^3$  at  $40^\circ$  N equivalent to an increase of up to 15%. During winter the effect of ship emissions is about a factor of two lower, thereby reaching a maximum in the latitude band from  $40^\circ$  S to  $30^\circ$  N. Since the OH concentrations are globally highest in these regions, the increase by about  $5 \cdot 10^5$  molecules/ $\text{cm}^3$  is of similar magnitude as the annual mean concentration in temperate and high latitudes, and is therefore highly significant on a global scale. Also in winter the boundary layer OH perturbation is still of the order of 10%. As can be seen from Fig. 8 ship emissions are dominating the annual zonal mean OH perturbation in the boundary layer throughout the year.

The effect of road emissions on the OH distribution shows a pronounced seasonal cycle in particular in the Northern Hemisphere. The maximum effect occurs during northern summer in the continental boundary layer between 800–900 hPa reaching  $1 \cdot 10^5$  molecules/cm<sup>3</sup> or about 3.5% (zonal annual mean). Since road emissions are largely emitted at some elevation over the continents the zonal mean effect on OH at 1000 hPa is less pronounced compared to 900 hPa. However, in particular during northern summer road transport seems to be most important for the lower troposphere in the extratropics up to 500 hPa.

The horizontal distribution (Fig. 9) reveals that the effect on a regional basis can be much stronger than  $1 \cdot 10^5$  molecules/cm<sup>3</sup>. The largest impact is found in the industrialized regions of the Eastern US, central Europe and East Asia reaching  $5 \cdot 10^5$  molecules/cm<sup>3</sup>. In these regions OH has the highest potential to be recycled via the reaction of NO with HO<sub>2</sub>, which is produced after the initial reaction of OH on the carbon containing reactive species (e.g. CO, CH<sub>4</sub>, NMHCs). The additional NO<sub>2</sub> from this reaction in turn leads to the enhanced formation of ozone and OH production via O<sup>1</sup>(D). During winter the emissions of CO and NMHCs in these regions act as a direct sink, since solar radiation is reduced.

Comparing the impact of road and ship traffic, both surface sources of air pollution, the effects on OH are very different, although the annual amount of emitted NO<sub>x</sub> is similar (see Table 1). The zonal mean perturbation of road peaks at about 4% during northern summer and 2% in the Southern Hemisphere summer. The regional effect during July can exceed 10% in the continental boundary layer for road traffic. As evident from Fig. 8 ship emissions have a larger effect even in the zonal mean. The large differences of both means of transportation on OH can be related to the different regions and pollution conditions. As will be seen later, the sensitivity of OH to traffic emissions is largest in the still relatively pristine regions over the (sub-)tropical oceans. Background concentrations of ozone, NO<sub>x</sub> and other NMHCs are low, and the solar irradiation and water vapor available for OH formation are high. The recycling potential of OH by NO<sub>x</sub> is lower than over the continents and is efficiently enhanced by ship emissions. The

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reason is that at already enhanced  $\text{NO}_x$  levels the reaction of OH with  $\text{NO}_2$  becomes a significant sink of hydroxyl radicals, which is not the case in the  $\text{NO}_x$ -poor marine boundary layer. On the other hand these low  $\text{NO}_x$  concentrations accompanied with high OH make the system more sensitive for  $\text{NO}_x$  perturbations than in polluted areas (Lelieveld et al., 2002). Furthermore, road emissions occur at higher altitudes and latitudes where average water vapor concentrations and solar irradiation are lower and in regions where other anthropogenic sources of pollution cause enhanced  $\text{NO}_x$  levels. The combination of these factors leads to a lower sensitivity of OH to perturbations from road traffic than for ship emissions.

The regional perturbation pattern (Fig. 9) illustrates that locally the effects of road and ship emissions on OH are similar during northern summer. The strongest effect is simulated for the ship emissions over the northern subtropical Atlantic in the same region where also ozone exhibits the largest perturbation (see Fig. 4). Note furthermore that in particular during northern summer the effect on OH at high northern latitudes over Europe is as high as over the eastern subtropical Pacific. Thus the increasing ship traffic at high northern latitudes as a response to the diminishing ice coverage due to the relatively rapid climate change (Lemke et al., 2007, IPCC) might have strong effects on the oxidation capacity and therefore on the composition of the atmosphere in high latitudes.

Similar to ozone, aircraft emissions have the largest effect on OH in the upper troposphere of the northern extratropics during summer. Despite the fact that the absolute perturbation is about a factor of 2 lower during winter, the relative perturbation exceeds 12%. The larger relative change during northern winter is due to the strong seasonal variation of the background latitudinal OH distribution. During winter the extratropical zonal mean OH at 300 hPa is less than  $1 \cdot 10^5$  molecules/cm<sup>3</sup>, whereas in summer the mean OH in the same region is reaching  $2.5 \cdot 10^5$  molecules/cm<sup>3</sup>.

Note that aircraft emissions have the maximum impact on OH over the subtropical Atlantic south of the main North Atlantic flight corridor. Larger humidity values and the smaller solar zenith angles at lower latitudes during that time of the year are more

favourable for OH production than further north. Notice that ship emissions also significantly contribute to the OH perturbation in the middle to upper troposphere of the subtropics up to 200 hPa caused by convective uplift of NO<sub>x</sub> from maritime transport in the tropical and subtropical latitudes.

## 5.2 Methane lifetime

As seen in the previous section ship emissions have the largest effect on OH leading to the largest reduction of methane lifetime (Table 7) among the three major modes of transport. Road emissions are only half as efficient in perturbing OH on a global scale, and the effects of air traffic are the smallest. The changes by 4.12% (ensemble mean) due to ship emissions exceed the 1.56% reported by Eyring et al. (2007), although their emissions of 3.1 TgN/yr are not very much lower compared to the 4.4 TgN/yr in this study (see Table 1). However, the distribution of the ship emissions in Eyring et al. (2007) and Stevenson et al. (2006) is based on the EDGAR3.2 dataset, where the emissions are mainly concentrated on the major shipping routes. The ship emissions used in QUANTIFY have been generated by merging the daily fields of COADS and AMVER data (Automated Mutual-assistance Vessel Rescue system) (Endresen et al., 2003) and are distributed over a larger area (comp. Fig. 1 and Eyring et al. (2007), their Fig. 1). This explains the stronger effect on CH<sub>4</sub> lifetime, since a larger fraction of NO<sub>x</sub> is emitted over the tropical oceans over a widespread area. Recall that conditions for OH production are favourable at low latitudes and in addition the local CH<sub>4</sub> lifetime by OH oxidation is shortest also due to the high temperatures.

Both factors are also the reason for the smaller effects of road and aircraft emissions on OH and on the methane lifetime, since they occur in higher latitudes and altitudes where annual mean OH concentrations and temperatures are lower.

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### 5.3 Sensitivity of OH production

From Fig. 9 it is evident that ship emissions strongly affect the still relatively clean regions mostly in the tropics, but also in the extratropics. In contrast, road emissions are largely released into an environment already affected by emissions from other sources both of natural (soils, lightning) and anthropogenic origin. The different effects on OH from road and ship traffic in the boundary layer are presented in Fig. 10 showing the  $\Delta\text{NO}_x$  and  $\Delta\text{OH}$  perturbations for road and ship emissions, respectively. Coloured data points are indicative for low  $\text{NO}_x$  conditions ( $<20$  pptv) in different latitude belts. The correlations show that the sensitivity of OH to  $\text{NO}_x$  perturbations is much higher for ship traffic than for road emissions even under low- $\text{NO}_x$  conditions. As expected the  $\Delta\text{OH}/\Delta\text{NO}_x$ -ratio is highest for ship emissions in the relatively pristine maritime conditions in the tropics (red). The impact of road emissions is insignificant in these regions under low- $\text{NO}_x$  conditions. In particular in the southern extratropics (green dots) there is a high sensitivity to  $\text{NO}_x$  perturbations from ships.

In relatively  $\text{NO}_x$  enriched conditions, under which most of the road emissions occur, the effect of ship  $\text{NO}_x$  on OH is also higher than that of road traffic. The OH response for ship emissions is an almost linear function at the lower  $\Delta\text{NO}_x$  levels, whereas the efficiency of OH production from road is weaker and levels off at lower  $\Delta\text{NO}_x$  perturbations. OH perturbations from road  $\text{NO}_x$  emissions maximize at  $4 \cdot 10^5$  molecules/cm<sup>3</sup>, while OH perturbations from ship exceed  $5 \cdot 10^5$  molecules/cm<sup>3</sup> and show no saturation tendency. This can be attributed to the higher level of NMHCs and  $\text{NO}_2$  over the continents acting as a sink for OH at high pollution levels.

## 6 Conclusions

In the frame of QUANTIFY the combined and relative effects of road, ship and aircraft emissions on the composition of the current atmosphere has been investigated. Six models simulated the effects of traffic on the current state of the atmosphere using a

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small (5%) perturbation approach. The results especially highlight the global impact of ship emissions on the chemical state of the marine boundary layer. Directly at the surface their impact can exceed 10% relative to the ozone background contributing 60–80% to the total perturbation by traffic. They strongly contribute to the tropospheric ozone column perturbation of 4 DU over the Atlantic Ocean.

For the global OH budget the effect of ship emissions is most important accounting for a perturbation up to  $5 \cdot 10^5$  molecules/cm<sup>3</sup> equivalent to about 15% relative to the base case. Since emissions of NO<sub>x</sub> largely occur in the pristine regions in the (sub-)tropics they efficiently lead to OH-formation and to additional ozone production. The consequent reduction of the lifetime of methane is about 4% for ship traffic while road and aircraft lead to 1.8% and 1% changes, respectively.

Road emissions in the Northern Hemisphere affect ozone in particular during summer, when the low solar zenith angles in the extratropics enhance photochemistry. Continental convection leads to vertical redistribution of the road emissions resulting in a significant contribution to the ozone perturbation in the northern extratropical UTLS of 3.5 ppbv being about 30% of the aircraft effect on ozone at 250–300 hPa. The efficiency of ozone production per NO<sub>x</sub> molecule emitted is highest for aircraft emissions (22 O<sub>3</sub>-molecules/molecule NO<sub>x</sub>), whereas the efficiencies for ozone from ship or road NO<sub>x</sub> are around 10 and 6 O<sub>3</sub>-molecules/molecule NO<sub>x</sub>, respectively. The high efficiency of air traffic on ozone perturbations arises from the relatively long lifetime of NO<sub>x</sub> and its reservoir species PAN and HNO<sub>3</sub> at the altitudes where the emissions occur.

*Acknowledgements.* The QUANTIFY project is funded by the European Union within the 6th research framework programme under contract 003893.



This Open Access Publication is  
financed by the Max Planck Society.

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**Table 1.** Emissions from different sources provided by QUANTIFY for the year 2000 in TgN NO<sub>x</sub> and TgC CO and NMHC, respectively (for NMHC's the conversion of 161/210 was used). Soil and biogenic isoprene emissions were taken from Ganzeveld et al. (2006), biogenic emissions of hydrocarbons and CO according to von Kuhlmann et al. (2003a,b).

source	QUANTIFY <sup>a,b,c</sup>			Accent <sup>d</sup>			Edgar <sup>e,f</sup>			Retro <sup>g</sup>	
	NO <sub>x</sub>	CO	NMHC	NO <sub>x</sub>	CO	NMHC	NO <sub>x</sub>	CO	NMHC	NO <sub>x</sub>	CO
Road	6.85	31.3	7.7	–	–	–	8.67	79.8	25.9	9.7 <sup>h</sup>	91.6 <sup>h</sup>
Ship	4.39	0.6	0.34	–	–	–	6.3	0.55	1.3	3.6	0.47
Air	0.67	–	–	0.79	–	–	0.71	0.04	0.42	0.73	–
tot. traffic	11.91	31.9	8.04	16.07	83.36	36.6	17.6 <sup>i</sup>	86.6 <sup>i</sup>	29.84 <sup>i</sup>	–	–
Non-traffic	27.1 <sup>e</sup>	341.3 <sup>e</sup>	100.3 <sup>e</sup>	21.9	447.4	76.13	27.1	341.3	100.3	19.3	275.6
biogenic, soil	6.89 <sup>j</sup>	48.2	340.5	5	100	772	–	–	–	9.4	77.3

<sup>a</sup> Road emissions: Borken and Steller (2006); <sup>b</sup> Ship emissions: Endresen et al. (2007);

<sup>c</sup> Aircraft emissions: Eyers et al. (2004); <sup>d</sup> Emission inventory developed within ACCENT (Atmospheric Climate Change: The European Network of Excellence) and GEIA (Global Emissions Inventory Activity); <sup>e</sup> van Aardenne et al. (2005); Olivier et al. (2005); <sup>f</sup> Ship emissions from Eyring et al. (2005); <sup>g</sup> Reanalysis of the tropospheric composition over the past 40 years, data taken from report on emissions data sets and methodologies for estimating emissions, [ftp://ftp.retro.enes.org/pub/documents/reports/D1-6\\_final.pdf](ftp://ftp.retro.enes.org/pub/documents/reports/D1-6_final.pdf); <sup>h</sup> Total land based traffic; <sup>i</sup> Including non-road land transport; <sup>j</sup> Ganzeveld et al. (2006); Kerkweg et al. (2006).

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**Table 2.** Partitioning of NMHC's used for the emissions in QUANTIFY (von Kuhlmann et al., 2003a,b).

species	NMHC specifications	
	biomass burning	fossil fuel
C <sub>2</sub> H <sub>6</sub>	0.1014350	0.05974
C <sub>3</sub> H <sub>8</sub>	0.0322390	0.09460
C <sub>4</sub> H <sub>10</sub>	0.0416749	0.7154
C <sub>2</sub> H <sub>4</sub>	0.1904856	0.03715
C <sub>3</sub> H <sub>6</sub>	0.0849224	0.01570
CH <sub>3</sub> OH	0.1075290	0.01436
CH <sub>3</sub> CHO	0.0483586	0.
CH <sub>3</sub> COOH	0.1136230	0.
CH <sub>3</sub> COCH <sub>3</sub>	0.0501278	0.02375
HCHO	0.0609397	0.004787
HCOOH	0.0404954	0.
MEK	0.1281699	0.03447

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**Table 3.** Model simulations performed for QUANTIFY.

experiment	emission		
	road	ship	aircraft
Base	100%	100%	100%
Road	–5%	100%	100%
Ship	100%	–5%	100%
Air	100%	100%	–5%
All	–5%	–5%	–5%

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**Table 4.** Participating models.

Model	TM4	p-TOMCAT	OsloCTM2	LMDzINCA	UCI-CTM	E39/C
Operated	KNMI	UCAM-DCHEM	UiO	LSCE	UCI	DLR
Model Type	CTM	CTM	CTM	CCM (nudg)	CTM	CCM
Meteorology	ECMWF OD	ECMWF OD	ECMWF OD	ECMWF OD	ECMWF OD	–
Hor. Resolution	2° × 3°	T21	T42	3.75° × 2.5°	T42	T30
Levels	34	31	40	19	37	39
Model Top(hPa)	10	10	10	3	2	10
Transport Scheme	Russel-Lerner	Prather	Prather	van Leer	Prather	Williamsen and Rasch
Convection	Tiedke	Tiedke	Tiedke	Tiedke	Tiedke	Tiedke
Lightning	Meijer et al. (2001)	Price and Rind	Price and Rind	Price and Rind, modified	Price and Rind	Grewe et al. (2001)
Transp. Species	26	35	76	66	28	12
Total Species	42	51	98	96	38	37
Gas phase reactions	68+16	112+27	163+47	291+51	90+22	107
Het. reactions	2	1	7	4	0	8
Strat. Chem.	no	no	yes	no	LINOZ	yes
NMHC Chemistry	yes, CBM4	yes	yes	yes	yes	no
References	van Noije et al., 2006a; 2006b	O'Connor et al. (2005)	Gauss et al. (2003) Isaksen et al. (2005)	Hauglustaine et al. (2004), Folberth et al. (2005)	Wild et al. (2003), Hsu et al. (2005)	Grewe (2007), Damiris et al. (2005)

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**Table 5.** Globally averaged ozone perturbations by transport mode for the lower and upper troposphere in ppbv. Also given is the percentage relative to the unperturbed case.

	1000–800 hPa			300–200 hPa		
	Mean	$\sigma$	%	Mean	$\sigma$	%
Road	−0.87	0.22	2.89	−1.17	0.30	0.95
Ship	−1.20	0.26	3.99	−1.12	0.21	0.91
Air	−0.30	0.07	1.0	−0.89	0.46	0.72
All	−2.37	0.44	7.9	−3.20	0.57	2.61

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**Table 6.** Global annual mean ozone production in molecule of ozone per molecule of emitted  $\text{NO}_x$ . For the calculation a global mean lifetime of ozone of 22 days (according to Stevenson et al., 2006) was assumed.

	TM4	OSLO	LMDz	UCI	TOMCAT	Mean	$\sigma$
Road	5.2	7.2	5.6	5.8	4.3	5.6	0.9
Ship	7.9	10.8	7.6	9.4	9.4	9.0	1.2
Air	22.0	20.3	24.1	23.1	46.3	27.2	9.7

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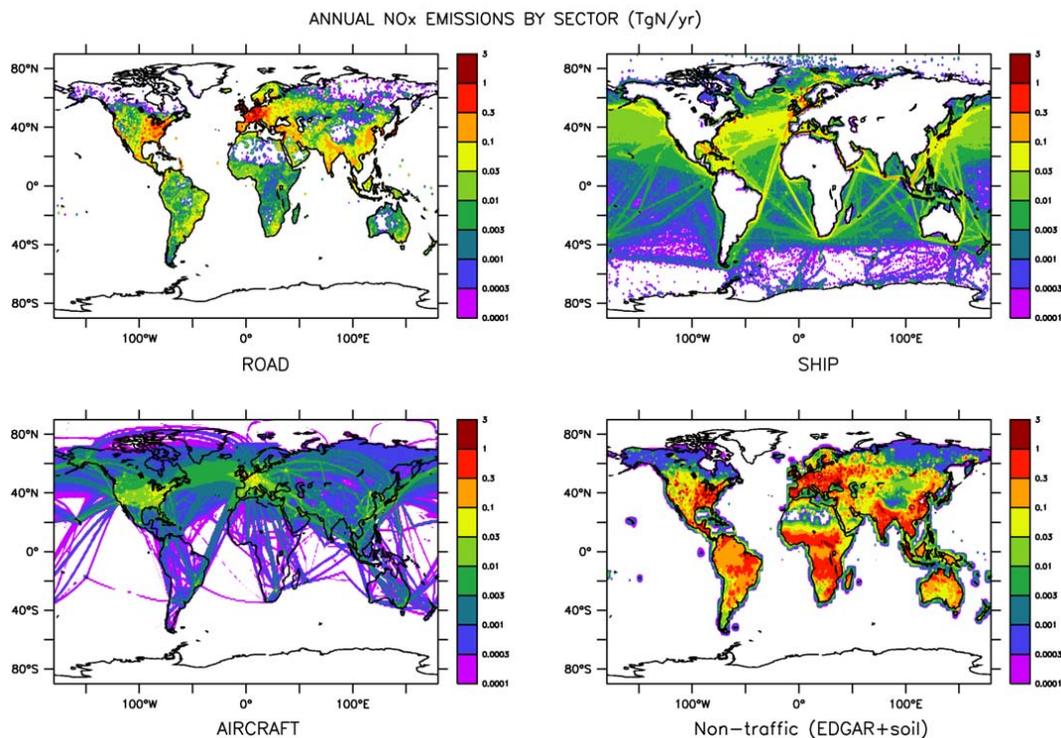
**Table 7.** Methane lifetime in years for the BASE case and relative changes due to a decrease of traffic emissions scaled to 100%. Top boundary for the integration was 50 hPa. Note that no feedback factor is included.

	TM4	OSLO	LMDz	UCI	TOMCAT	Mean	$\sigma$
Base (yr)	7.23	8.49	10.23	7.45	11.43	8.97	1.63
Road (%)	1.86	1.36	1.91	1.28	1.63	1.61	0.25
Ship (%)	3.25	3.63	4.12	3.51	6.10	4.12	1.02
Air (%)	0.81	0.63	1.06	0.93	1.79	1.04	0.40

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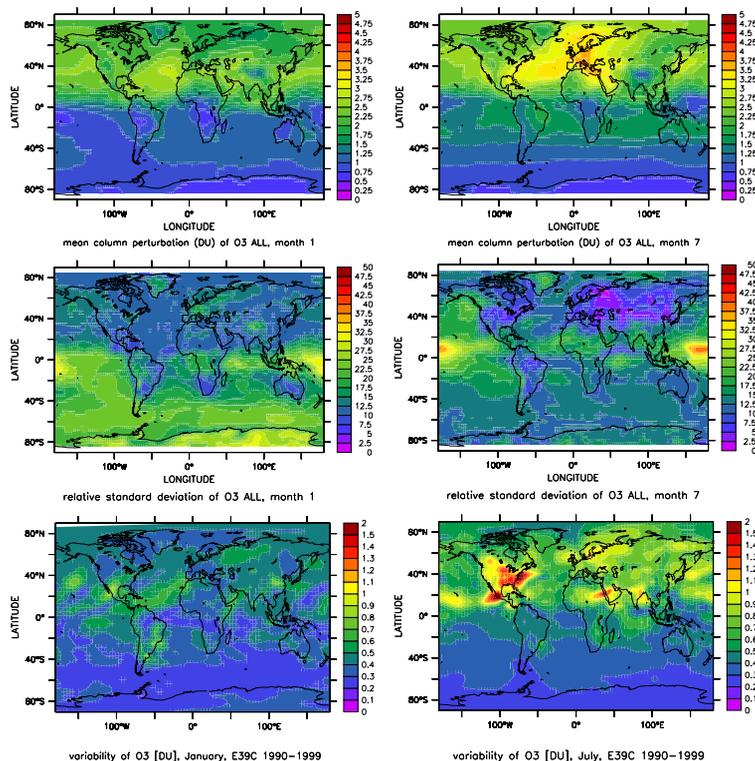


**Fig. 1.** NO<sub>x</sub> emissions (TgN/m<sup>2</sup>/year) used in QUANTIFY by sector.

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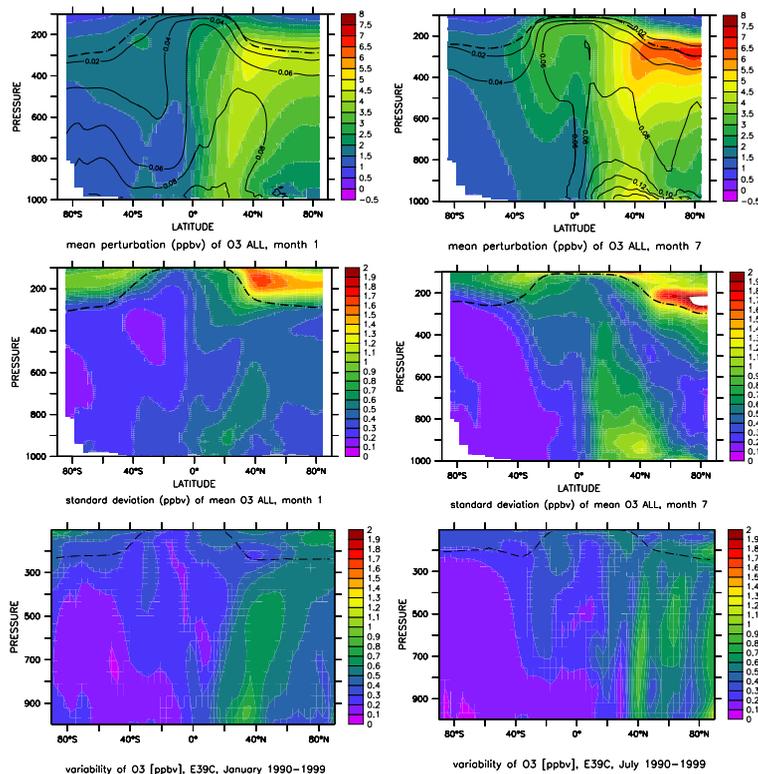


**Fig. 2.** Mean column ozone perturbations (scaled to 100%) for the integrated emissions from all types of transport obtained from TM4, LMDzINCA, OSLO CTM2, UCI and p-TOMCAT (top row) for January (left) and July(right). The corresponding relative standard deviations (relative to the ensemble mean perturbation) are displayed in the middle row. The lower two panels show the interannual variation of the detrended transportation induced ozone perturbation based on E39/C.

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**Fig. 3.** As Fig. 2, but for the zonal mean in ppbv. Solid contours show the perturbation relative to the mean unperturbed simulation and the dashed line indicates the tropopause.

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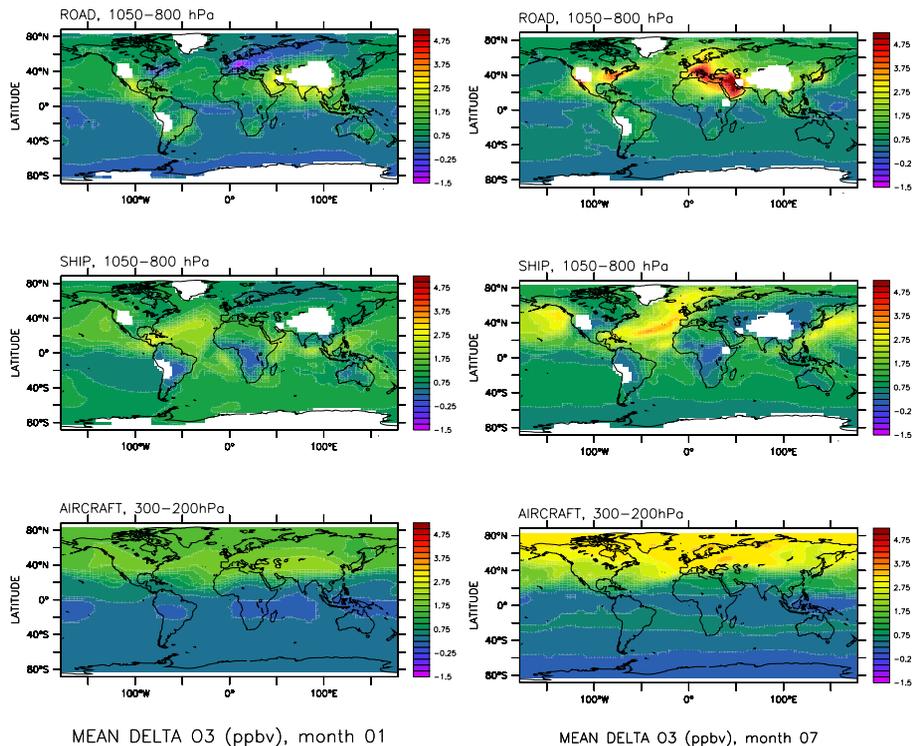
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**Fig. 4.** Mean perturbations of ozone (ppbv) in the PBL (surface-800 hPa) during January (left column) and July (right) for the different modes of transportation. The values for aircraft are shown for 300–200 hPa.

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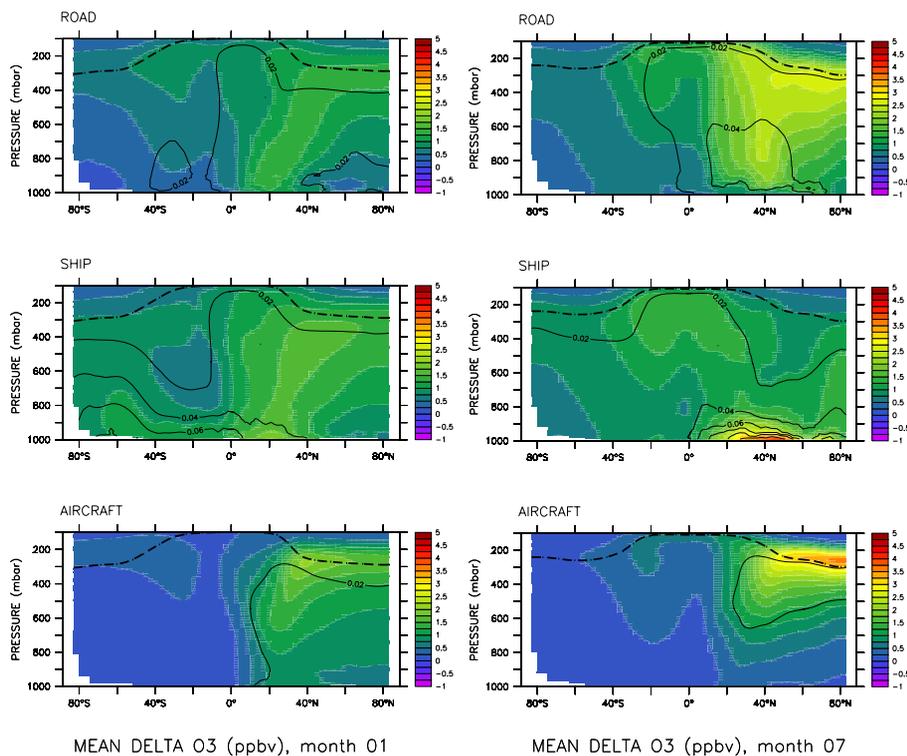
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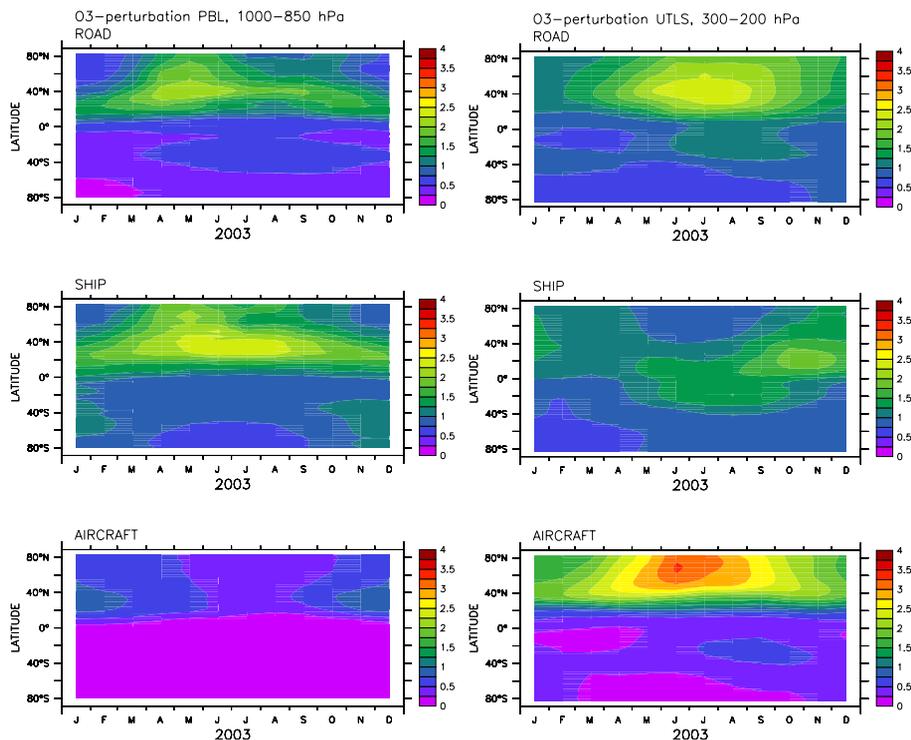


**Fig. 5.** Same as Fig. 4, but for the zonal mean ozone perturbation. Solid contours show the change relative to the base case simulation, dashed line indicates the tropopause.

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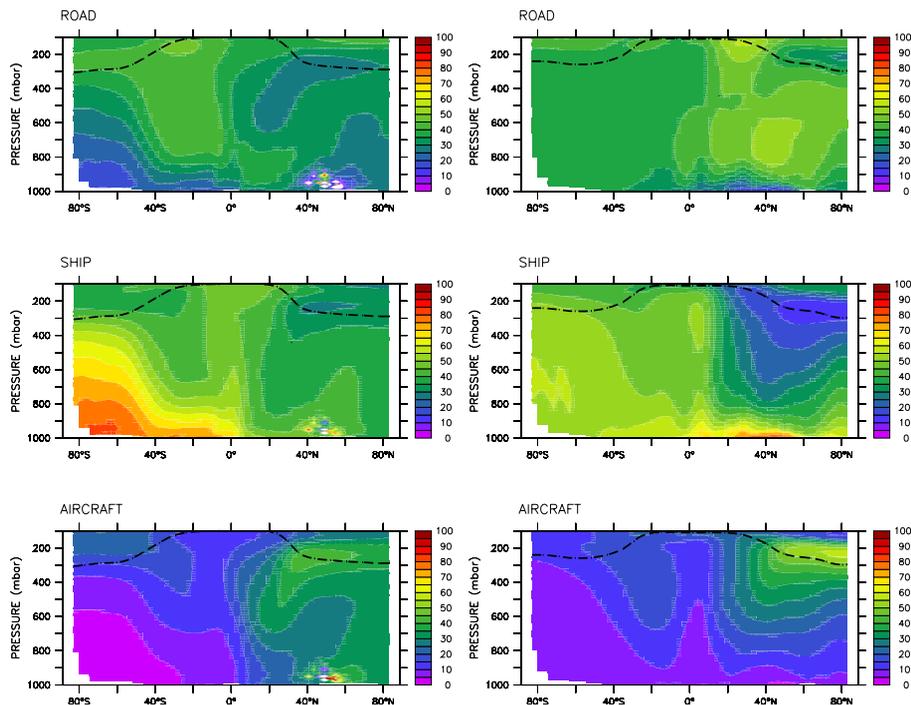


**Fig. 6.** Temporal evolution of the ozone perturbation in ppbv for the boundary layer (left column) and the UTLS region (right).

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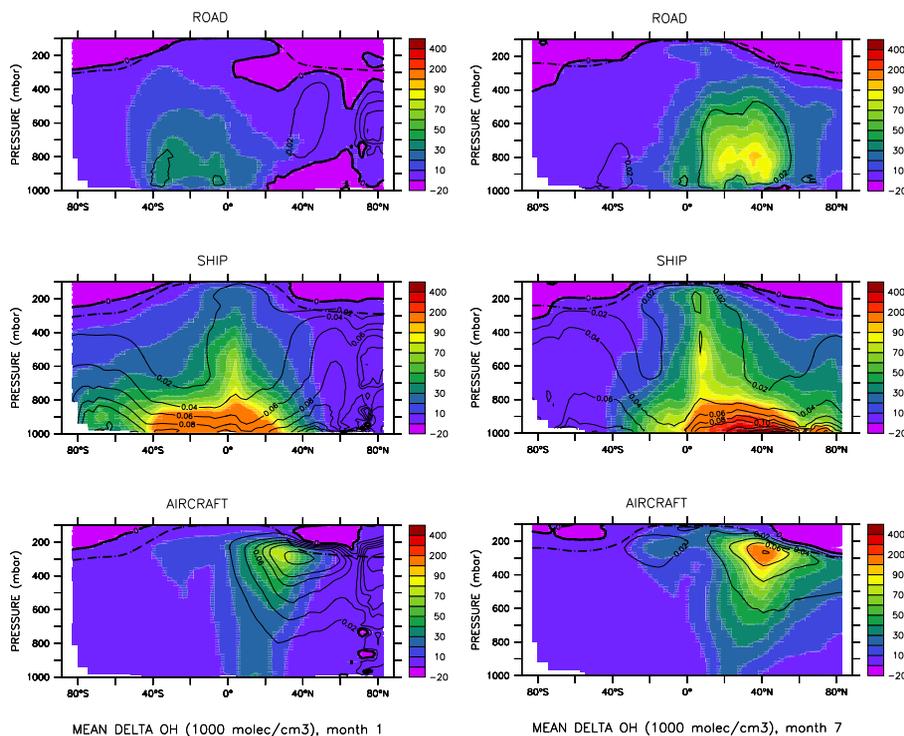


**Fig. 7.** Relative contribution of each transport sector to the total ozone changes due to the sum of emissions in all transport sectors.

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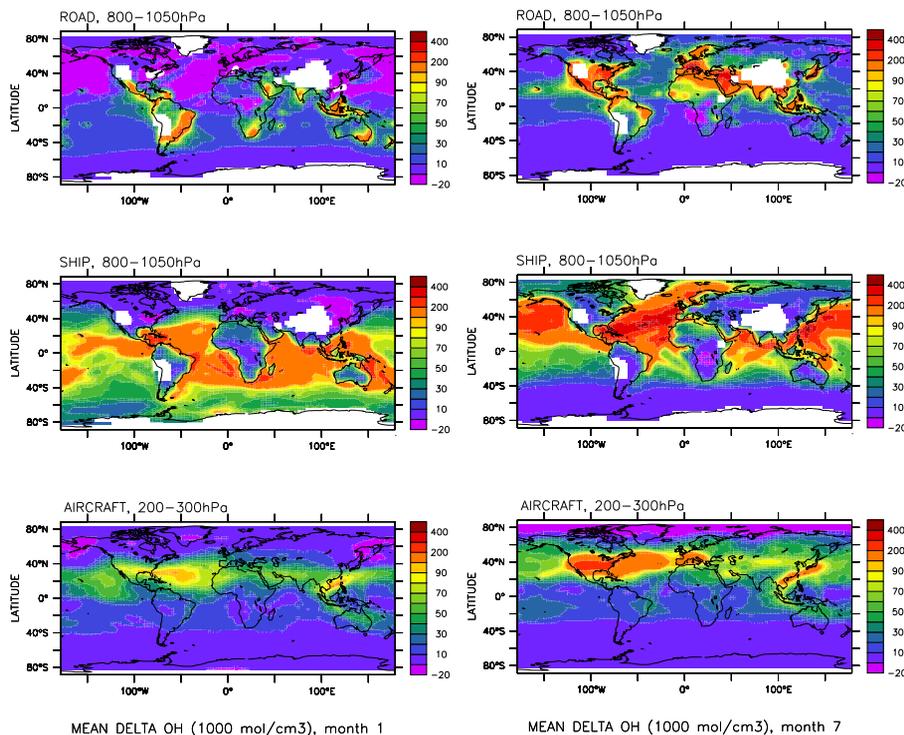


**Fig. 8.** Zonal mean OH perturbation for January (left) and July for road (top), ship (middle) and aircraft (bottom) emissions, respectively (shading). Contours show the perturbation relative to the base case simulation.

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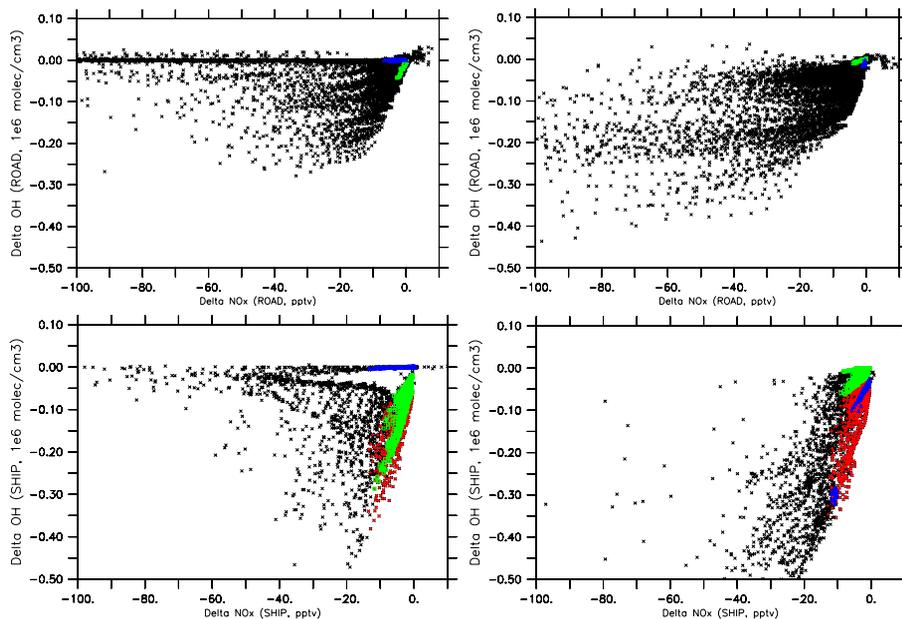


**Fig. 9.** Mean OH perturbation in the boundary layer during January (left) and July (right) between 800–1000 hPa for road – (top) and ship emissions (middle). The effect of aircraft emissions is displayed for 250–300 hPa (bottom).

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**Fig. 10.** Correlations between  $\Delta\text{OH}$  and  $\Delta\text{NO}_x$  for road traffic (upper row) and ship traffic (lower row) for January (left) and July (right). All coloured points are for low  $\text{NO}_x$  conditions coded by latitudes: (sub-)tropics (red), Northern Hemisphere (blue  $>30^\circ\text{N}$ ) and Southern Hemisphere (green,  $<30^\circ\text{S}$ ). The effect of ship emissions is shown at 950 hPa over the oceans and of road emissions over land up to 850 hPa.

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