QUANTIFY model evaluation of global chemistry models: carbon monoxide

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ABSTRACT: In the EU Integrated project QUANTIFY, atmospheric chemistry models (ACMs) are one of the major tools to improve the understanding of key processes relevant for the effects of different transportation modes, and their representation in global models. The performance of the ACMs has been tested through comparisons with the ETH model evaluation global database for the upper troposphere and lower stratosphere. Data from measurement campaigns, ozone soundings, and surface data have been processed to support an easy and direct comparison with model output. Since model evaluation focuses on the year 2003, observational data to compare model data with are the SPURT campaign and the commercial aircraft program MOZAIC. The model evaluation indicates a particular problem in the simulation of carbon monoxide. If QUANTIFY emissions inventories are used, models significantly underestimate its tropospheric abundance at northern hemispheric middle latitudes and subtropical latitudes. Potential causes will be discussed.

1 INTRODUCTION

Global atmospheric chemistry models (ACMs), i.e. chemistry transport models (CTMs) and chemistry-climate models (CCMs) have become standard tools to study tropospheric and stratospheric photochemistry and the impact of different emission sources onto the atmospheric composition including scenarios for future emission changes. Studies based on such models were a central element in scientific assessments of the impact of present and future air traffic emissions (Brasseur et al., 1998; Penner et al., 1999; NASA, 1999). In the EU FP6 Integrated Project (IP) QUANTIFY (Quantifying the Climate Effect of Global and European Transport Systems) ACMs are used to improve the understanding of the relative effects of different transportation modes on the atmospheric com-

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position, and their representation in global models. For instance, the impact of present-day traffic emissions on atmospheric ozone and the hydroxyl radical (OH) was evaluated by Hoor et al. (2009). To estimate the reliability of the models and hence of the studies investigating the impact of traffic emissions, it is highly relevant to evaluate how well the models reproduce available observations. A first comprehensive model evaluation of ACMs operated by different groups in Europe was carried out by Brunner et al. (2003; 2005) in the framework of the EU project TRADEOFF. Brunner et al. (2003; 2005) compared model results with trace gas observations from several aircraft campaigns for the period 1995-1998. The present study uses updated versions of the models applied in Brunner et al. (2003; 2005). This paper focuses on the simulation of carbon monoxide (CO), one of the major atmospheric pollutants in densely populated areas, chiefly from exhaust of combustion engines by traffic, but also by incomplete burning of other fuels in industry. In the free troposphere, it has an indirect radiative forcing effect by elevating concentrations of tropospheric ozone through CO oxidation. Model results are compared to data from the commercial aircraft program MOZAIC (Marenco et al., 1998), as well as to aircraft campaign data. The next section summarises the main model characteristics, the boundary conditions used, and the methodology. Results of the model evaluation are shown in Section 3. Conclusions are presented in Section 4.

2 MODELS, DATA, AND METHODOLOGY

Within QUANTIFY model evaluation results from six models were compared with observational data. Four models are CTMs using prescribed operational ECMWF data to simulate meteorological conditions (TM4, p-TOMCAT, OsloCTM2, and MOCAGE) and two are CCMs (LMDzINCA and ECHAM5/MESSy), which were nudged toward operational ECWMF fields.

An overview of the main model characteristics is given in Hoor et al., 2009 (their Table 4), and in Table 1 for MOCAGE and ECHAM5/MESSy. The model setups are described in detail in Hoor et al. (2009) for TM4, p-TOMCAT, OsloCTM2, and LMDzINCA, in Teyssèdre et al. (2007) for MOCAGE, and in Jöckel et al. (2006) for ECHAM5/MESSy.

To force the models toward a realistic atmospheric state, emissions from different source categories were considered in the QUANTIFY numerical simulations. These are described in detail in Hoor et al. (2009). Emissions for the three transport sectors road, shipping, and air traffic were considered. The road traffic emissions inventory was developed within the QUANTIFY project. Except for a sensitivity simulation by OsloCTM2 (which used emissions from the POET project), the emissions used in this study are based on a draft version (Borken and Steller, 2006) (QUANTIFY preliminary, see Table 2 for CO emissions). An overview of CO emissions considered in the QUANTIFY

Table 1: Main characteristics of ECHAM5/MESSY and MOCAGE.

Model	MOCAGE	ECHAM5/MESSy
Operated	CNRM	MPICHEM
Model type	CTM	CCM (nudged)
Meteorology	ECMWF OD	ECMWF OD
Hor. resolution	T21	T42
Levels	60	90
Model top (hPa)	0.07	0.01
Transport scheme	Williamson & Rasch	Lin & Rood
Convection	Bechtold et al. (2001)	Tiedke-Nordeng
Lightning	Climatology	Price and Rind + Grewe
Transp. species	65	82
Total species	82	108
Gas phase reactions	186 + 47	178 + 57
Het. reactions	9	10 (PSC) + 26 (wet-phase)
Stratosph. chemistry	yes	yes
NMHC chemistry	yes	yes
Lightning NO_x (TgN/yr)	5	5

Table 2. CO emissions used in the QUANTIFY model simulations and comparison with TRADEOFF emissions (Brunner et al., 2003) (in Tg CO/yr). (*) Compare number in Hoor et al. (2009), their Table 1.

Species	Emission source	TRADEOFF	QUANTIFY preliminary	QUANTIFY final	OSLO POET
CO					
	Road traffic		73	110	196
	Ships		1.3	1.3	0.1
	Air traffic		1.1	1.1	
	Other anthropogenic		108	108	114
	Domestic burning (DB)		237	237	237
	Biomass burning (BB)	700	508	508	309
	Total anthr. fossil fuel (anthr.+road+ships+air)		183	220	310
	Total anthr. fossil fuel + DB	650	420	457	547
	Vegetation + soil	200	65*	65*	178
	Total	1550	993	1030	1034

simulations is given in Table 2.

Model output was generated and analysed with respect to trace gas observational data using point-by-point output, i.e. at each simulation time step, the instantaneous tracer fields were linearly interpolated to the positions of coinciding observations (Brunner et al., 2003; 2005). This method allows for a very close comparison with observations and fully accounts for the specific meteorological conditions of the measurements. By each modelling group the years 2002 and 2003 were simulated. 2002 was taken as spin-up, the year 2003 provided the base year for comparison with observations and sensitivity simulations (Hoor et al., 2009).

Model results were compared to data from the commercial aircraft program MOZAIC (Marenco et al., 1998), as well as to data from the SPURT (German: SPURenstofftransport in der Tropopausenregion) campaign (Engel et al., 2006). From MOZAIC, the one-minute averages of the CO measurements were evaluated. The 2003 SPURT campaigns took place in February, April, and July 2003 over Europe (Engel et al., 2006; their Fig. 4). Besides CO, ERA40 potential vorticity (PV) interpolated onto SPURT coordinates was used to distinguish between tropospheric and stratospheric air. The SPURT data were time averaged to yield one minute averages.

3 EVALUATION OF MODEL PERFORMANCE

Average model biases $(mean_{model}-mean_{obs})/mean_{obs}*100\%$ and root-mean-square (RMS) differences E of point-to-point model results and measurements are shown in Table 3 for the February 2003 SPURT campaign for the lowermost stratosphere (LMS, PV > 2 PVU) and the upper troposphere (UT, p < 500 hPa and PV < 2 PVU). Additional information on model performance can be summarised in a Taylor diagram (Taylor, 2001; Brunner et al., 2003): the correlation coefficient R, the centred pattern RMS difference E′ between a test vector f (model) and a reference vector r (observations), and the ratio of the standard deviations (σ_f / σ_r) of the two vectors are all indicated by a single point in a two-dimensional plot. For example, in Fig. 1a, the test point by MOCAGE (MO) refers to a correlation coefficient R=0.87, a normalised standard deviation σ_f/σ_r=0.95 (smaller modelled than observed σ), relatively large centred RMS difference (distance between reference and test point, only qualitative statement possible), and a skill score of > 0.9 (parabolic line of constant skill). For more details on the underlying algebra and relationships between statistical quantities see Taylor (2001) and Brunner et al. (2003) for the used definition of the skill score.

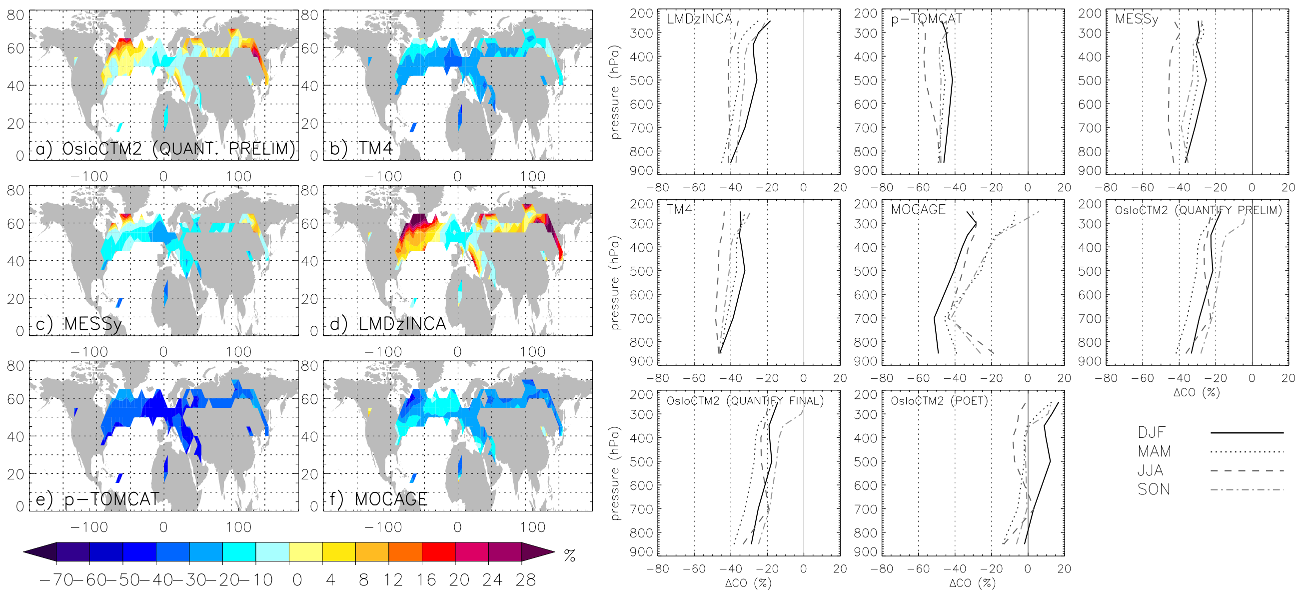
Upper tropospheric CO is underestimated by most models in all campaign months (≈-5% to -50%) except for OsloCTM2 (POET), for which a positive deviation of 10% to ≈35% is found. At higher altitudes in the LMS, negative biases are either significantly reduced or they turn to positive deviations. OsloCTM2, which exhibits positive biases in the UT, shows increased positive deviations from observations in the LMS. It could be suspected that the relatively low CO emissions from road traffic used in the QUANTIFY preliminary simulations (Table 2) might be responsible for the negative bias of most models. However, the negative deviations are not reduced or

Table 3: Mean model biases of CO (in %) for the 2003 SPURT campaigns for the lowermost stratosphere (PV > 2 PVU) (upper part) and the middle to upper troposphere (p < 500 hPa and PV < 2 PVU). Grey shading indicates negative deviation of a model mean from the respective observational value.

Model/Variable	February	April	July
	Lowermost stratosphere (LMS)		
OsloCTM2 (POET)	83±47	61±51	30±52
OsloCTM2	24±29	18±37	3±39
TM4	-13±21	-7±22	-22±27
p-TOMCAT	-27±19	-30±25	-44±23
MOCAGE	-13±26	32±58	-17±24
LMDzINCA	27±39	19±43	-3±38
ECHAM5/MESSy	-1±20	-4±25	-25±25
	Upper troposphere (UT)		
OsloCTM2 (POET)	35±55	37±59	10±54
OsloCTM2	-7±37	0±43	-11±41
TM4	-29±20	-17±30	-34±28
p-TOMCAT	-40±14	-43±23	-52±22
MOCAGE	-26±13	28±61	-19±30
LMDzINCA	-12±37	-5±50	-22±34
ECHAM5/MESSy	-18±27	-10±29	-33±28

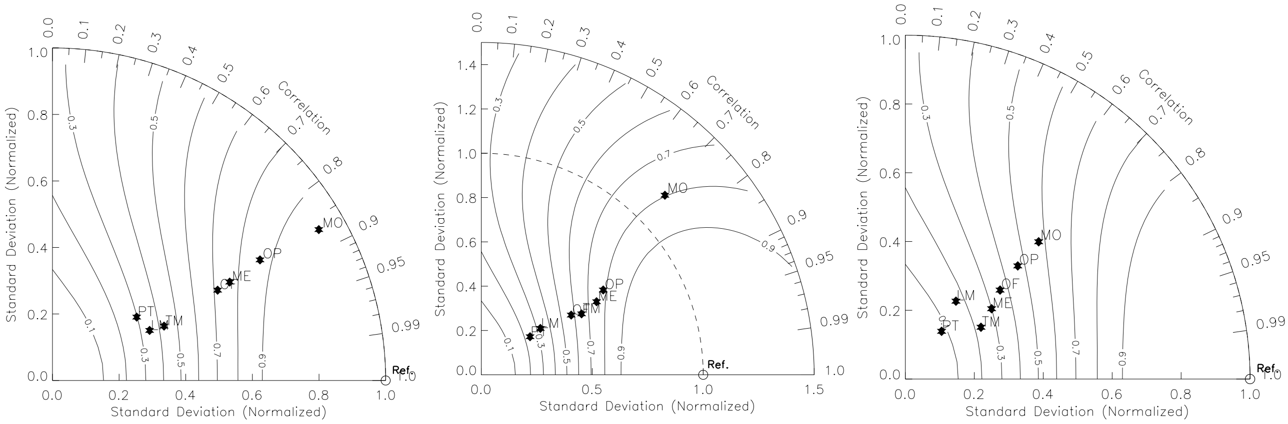
eliminated when using QUANTIFY final road emissions, which are ≈50% higher than the preliminary emissions (Fig. 1b, compare OsloCTM2 simulations PRELIM and FINAL). Hence, the different performance of OsloCTM2 using POET emissions (Table 3, Fig. 1b) can probably not be (fully) explained by the higher road traffic CO emissions. Possibly, emissions of non-methane volatile organic compounds (NMVOCs), which are an additional non-negligible source of CO (IPCC, 2001), may play a role: in the POET emissions inventory these are known to be significantly higher over polluted regions than in other inventories. The altitude dependency of biases is largely reflected by MOZAIC profiles: as presented for Frankfurt, Germany, relative differences show a positive slope with altitude (Fig. 1b). This effect might be connected to an insufficient vertical resolution of the models to resolve the vertical CO gradient across the tropopause.

Using MOZAIC cruise level data, which are mostly representative of the LMS, similar biases as over Europe were identified on the hemispheric scale in all seasons (Fig. 1a for DJF, other seasons not shown). Note that the geographical bias patterns are not homogeneous for most models,



a) b)

Figure 1: Mean model biases for 2003 MOZAIC data (model-MOZAIC) (in %). a) Horizontal distribution from cruise level data at 300 hPa – 170 hPa, DJF 2003, biases only plotted if at least 20 measurements available in 5°x5° grid boxes; b) vertical profiles for Frankfurt, Germany, for DJF (black solid line), MAM (dark grey dotted line), JJA (grey dashed line), and SON (light grey dash-dotted line).



a) b) c)

Figure 2: Taylor diagrams of the comparison between observed and modelled CO for the SPURT campaigns 2003. a) February, b) April, and c) July 2003. Letters denote models: OP (OsloCTM2 with POET emissions), OF (OsloCTM2 with preliminary QUANTIFY emissions), TM (TM4), PT (p-TOMCAT), ME (ECHAM5/MESSy), MO (MOCAGE), and LM (LMDzINCA).

but show maximum negative deviation over Europe and smaller negative or even positive biases over Eastern USA and Siberia. This is due to regional features in the observed distribution, namely a CO maximum over Europe and relatively low mixing ratios over northern America and East Siberia (not shown), which are not fully captured by the models.

CO has a sufficiently large photochemical lifetime of 1-3 months in the troposphere (IPCC, 2001) to be transported on the hemispheric scale (e.g., Stohl et al., 2002). Thus, not surprisingly, the Taylor diagrams reveal high correlation coefficients in winter and spring 2003 ($0.8 \leq R \leq 0.9$) (Fig. 2a and b). In July, only somewhat smaller correlations ($0.5 < R \leq 0.8$) are probably due to the fact that models cannot reproduce small-scale convective events that were encountered during the flights (Hegglin, 2004). However, most models underestimate observed data variability ($\sigma f/\sigma r < 1$), probably also related to inability to reproduce small- or regional-scale features in the observations.

4 CONCLUSIONS

Carbon monoxide is a compound with a rather long lifetime in the troposphere. It is emitted by several emission sources, formed by VOC oxidation and transformed to carbon dioxide by oxidation with OH radicals. Furthermore, vertical and horizontal mixing affects its concentrations. We regard the following processes as most critical to explain the partial disagreement between numerical simulations performed within QUANTIFY and available measurements:

- Tropospheric CO concentrations depend on the applied emissions inventories. While model biases are not affected by either the use of preliminary or final QUANTIFY traffic emissions, the agreement between measurements and model results is improved when using the set of POET CO emissions compared to when using QUANTIFY preliminary or final emissions. However, it remains an open question what the cause(s) for the better model performance of the simulation with POET emission is (are). Additionally, the biomass burning emissions inventory used, which is representative for the year 2000 (specifications see Hoor et al., 2009) may not reflect atmospheric conditions in 2003, as it is known that 2002/2003 biomass burning emissions were anomalously high in the extratropical northern hemisphere (e.g., Yurganov et al., 2005).
- CO can be formed from VOC oxidation. This source is expected to be different from model to model adding additional uncertainty in the comparison between simulations and measurements.
- The sharp vertical gradient in CO concentration across the tropopause is an additional challenge for global simulations. The results indicate that current model resolution may be insufficient to resolve this gradient.

In a further study the information from ozone and nitrogen concentrations will be used to shed more light in the reliability of the numerical simulations performed within QUANTIFY.

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