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3-D global simulations of tropospheric CO distributions – results of the GIM/IGAC intercomparison 1997 exercise

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Importance of this Paper: The current generation of 3-D global models used for chemistry and climate global simulations have been compared in the frame of the Tropospheric Ozone (O_3) Global Model Intercomparison Exercise performed in 1997. The objective was to systematically evaluate their capabilities to simulate tropospheric ozone and their precursor gases, including carbon monoxide, and to identify key areas of uncertainty in our understanding of the tropospheric O_3 budget. This exercise was organised by Global Integration Modelling (GIM) Activity of the International Global Atmospheric Chemistry (IGAC) Project.

Abstract

The objective of the Tropospheric Ozone (O_3) Global Model Intercomparison Exercise performed in 1997 was to systematically evaluate the capabilities of the current generation of 3-dimensional global models to simulate tropospheric ozone and their precursor gases, and to identify key areas of uncertainty in our understanding of the tropospheric O_3 budget. This exercise has been organised by Global Integration Modelling (GIM) Activity of the International Global Atmospheric Chemistry (IGAC) Project. The present paper focuses on the capability of the

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models to simulate carbon monoxide, which is an important pollutant in the troposphere. The intercomparison of twelve 3-dimensional global chemistry/transport models shows significant differences between the models although all of them capture the general patterns in the global distribution of CO. © 1999 Elsevier Science Ltd. All rights reserved.

Keywords: 3-dimensional; Global; Models; Intercomparison; Tropospheric chemistry; IGAC; Carbon monoxide; CO; Budget; Methane lifetime

1. Introduction

It is now recognised that the atmospheric concentrations of several chemical and radiatively important trace constituents (gases and particles) are changing in the atmosphere (IPCC, 1994). Human activity is the main reason for these changes, climate changes affecting natural emissions and the radiative balance of the earth being the second forcing agent. Indeed, several trace constituents like aerosols, methane (CH₄), nitrous oxide (N_2O) , nitrogen oxides $(NO + NO_2)$, carbon monoxide (CO), Non-Methane Volatile Compounds (NMVOC) including Non-Methane Hydrocarbons (NMHC), sulphur dioxide (SO₂), dimethylsulphide (DMS) and halocarbons have direct anthropogenic and/or natural emissions to the atmosphere. Others, like ozone (O_3) and secondary aerosols, are produced from chemical reactions initiated by their precursors. In turn, the changing atmospheric concentrations of these trace constituents affect the radiative balance of the earth and may lead to a climate change.

To understand and reliably predict chemical and climate changes in the atmosphere a thorough understanding of the chemical, physical, biological and climatic processes which affect the distributions of trace compounds in the atmosphere, and of the interactions between these processes is required. Only then, the global budgets of trace constituents, their evolution due to natural and anthropogenic forcing and the related feedback mechanisms can be realistically simulated by the mean of numerical models.

The use of *numerical models* is required to take into account the physical processes governing the budget of trace gases in the troposphere and the complex non-linear behaviour of chemical reactions in the troposphere. Such models integrate the most recent information on chemical kinetics of homogeneous and heterogeneous reactions as well as photodissociation coefficients of atmospheric molecules. They also account for interactions between transport, chemistry and the radiative budget in the troposphere, and specifically for: (i) transport of trace constituents like nitrogen species (NO_{ν}) and O_3 from the stratosphere, (ii) emissions of trace gases like O₃ precursors: nitrogen oxides, carbon monoxide, NMVOCs and aerosols occurring mainly in the low troposphere, (iii) dry deposition of various trace gases and particles on the earth's surface, (iv) wet removal of soluble gases and particles, (v) convection, (vi) planetary boundary layer mixing, (vii) synoptic mixing in the troposphere, (viii) cloud microphysics and (ix) homogeneous and heterogeneous chemical reactions.

Recent assessments of the global-scale, radiative effects of greenhouse gases and of aerosols have been performed on the basis of global models which, in most cases, are still under development. The accuracy of such calculations relies strongly on the capability of the models to simulate the global budget of trace gases and aerosols. A major concern for the calculation of the chemically reactive trace gases budget is associated with: (1) the inaccuracies in the transport parameterisation (Jacob et al., 1997), (2) the simplification of the chemistry in the troposphere, required due to the prohibitive number of chemical reactions and species and (3) the uncertainties in the trace gases emissions used in the global models.

Even though current models ignore some important chemical and radiative processes in the atmosphere, it seems crucial for the time being to improve our model estimate of the budgets of O_3 , CH_4 , CO and NO_x . Indeed, O_3 is a greenhouse gas, a pollutant and one of the major oxidising agents in the troposphere. CH_4 is the most chemically important greenhouse gas with direct emissions to

the atmosphere. NO_x , which is also a pollutant, and in particular NO and NO₂, controls O₃ chemical production and destruction in the troposphere. CO is a chemically important gas with primary and secondary emissions in the atmosphere. Depending on the ambient NO_x mixing ratios, CO oxidation by hydroxyl (OH) radicals forming hydrogen peroxy radicals (HO₂) enhances (high NO_x) or depletes (low NO_x) ozone in the troposphere (Crutzen, 1994). CO changes have feedbacks into the abundance of CH₄ through the alteration of OH (Prather, 1996). Thus, although CO itself is not a significant greenhouse gas, changes in its concentration affect ozone and CH₄, both significant greenhouse gases (Daniel and Solomon, 1998).

Effort has already started by WMO/IPCC in 1994 (Stordal et al., 1995; Olson et al., 1997) to evaluate the capability of the chemical models to simulate O_3 , NO_x , HO_x and VOC chemistry. This effort is continued by the Global Integration Modelling (GIM) Activity within the International Global Atmospheric Chemistry (IGAC) Project of IGBP in 1996–1997, and focuses on the capability of the global 3-D chemistry/transport models used for global chemical and climate simulations to calculate the O₃ budget. Ground-based and in situ observations from several regions of the globe are used for validation of the model results. The present paper focuses on the model capability to simulate carbon monoxide since it is a key compound for tropospheric chemistry. Moreover, carbon monoxide has a more simple and better understood chemistry and better documented direct sources than the shorter lived NO_x and VOCs. It is therefore a good tool to evaluate the chemistry and the transport characteristics of photochemical models. Ozone simulations are the topic of another paper in preparation.

2. The GIM/IGAC tropospheric ozone chemistry/ transport 3-D models intercomparison exercise

The objective of the Tropospheric Ozone Global Model Intercomparison Exercise performed in 1997 was to systematically evaluate the capabilities of the current generation of 3-D global models to simulate tropospheric O_3 and other chemical compounds, and to identify key areas of uncertainty in our understanding of the tropospheric O_3 budget. This exercise has been organised by GIM Activity within IGAC Project. The strategy was to *investigate the consistency of the predicted results between models, which use a variety of inputs and mechanisms, to compare model results to the real atmosphere* and identify areas for further research in the close future.

To conduct this exercise, several modelling groups were asked to provide the 'best case' output of their models, and specifically:

- O₃, CO and NO_x (NO + NO₂) monthly mean mixing ratios at surface, 500 and 200 mbar in January and July;
- surface monthly mean concentrations at Barrow, Mace Head, Hohenpeissenberg, Mauna Loa, Barbados Samoa, Cape Grim, Cape Point for O₃; and at Barrow, Mace Head, Mauna Loa, Samoa, Cape Grim for CO;
- monthly varying vertical O₃ profiles at selected ozone sonde stations (Resolute, Hohenpeissenberg, Wallops island, Bermudas, Hilo, Natal, Samoa, Lauder, Syowa, S. Pole);
- seasonal and annual mean terms of O₃ budget up to 300 mbar for the 4 seasons annual mean CH₄ lifetime and mean hemispheric and global burdens.

3. Description of the chemistry transport models

The 12 global 3-D Chemistry Transport Models that participated at this exercise are listed in Table 1 together with the contributing scientists and key references. All these models are global 3-D models. Three of them are climatological models, i.e., they simulate atmospheric transport based on pre-calculated or observed monthly mean winds and temperatures. One of the models is a general circulation model (ECHAM) and the remaining models are chemistry/transport models using precalculated/observed meteorology provided every 4–12 h. This meteorological input is derived from general circulation models or weather prediction models. Models also differ in the parameterisation of the main processes controlling chemical tracer

Table 1

3-D models that participated at the Tropospheric Ozone (O₃) Global Integration Modelling (GIM) intercomparison exercise (3-D models)

IMAGES	JF. Muller, G. Brasseur, C. Granier	Muller and Brasseur, 1995
GFDL	H. Levy	Kasibhatla et al., 1996
HARVARD	Y. Wang, D. Jacob	Wang et al., 1998a,b,c
ECHAM	GJ. Roelofs	Roelofs and Lelieveld, 1995, 1997
TM3	F. Dentener, S. Houweling	Houweling et al., 1998
IMAU3	M. Krol	Krol and Weele, 1997
CTMK	W.M.F. Wauben	Wauben et al., 1997, 1998
MATCH	M.G. Lawrence, P.J. Crutzen	Lawrence, 1996, Lawrence et al., 1995, Lawrence and Crutzen, 1998
MOGUNTIA	N. Poisson, M. Kanakidou	Poisson, 1997, Poisson et al, 1999
MOZART	D.A. Hauglustaine, G.P. Brasseur, S. Walter	Brasseur et al., 1996, 1998
UKMETO	R.G. Derwent, C.E. Johnson, W.J. Collins, D.S.	Collins et al., 1997
	Stevenson	
UIO	T.K. Berntsen, I.S.A. Isaksen	Bernsten and Isaksen, 1997

budgets, i.e. transport by advection, diffusion and convection, chemistry (homogeneous and heterogeneous), wet and dry deposition and emission of trace compounds (including CO) by natural and anthropogenic sources. Detailed model descriptions can be found in the literature (see references in Table 1).

Some of the model characteristics together with the annual global emissions of ozone precursors: NO_x, CO, CH₄ and NMVOC used for the 'best case present day' simulations are summarised in Tables 2 and 3, respectively. Note that the direct annual amount CO emissions adopted in each model can be significantly different: Total primary sources vary from 1040 to 2362 Tg-CO/yr among the various models, as shown in Table 3. These emissions are primarily associated with technological sources, in the northern hemisphere, and with biomass burning, in the tropics. In some models, those not explicitly considering NMVOC chemistry, CO pseudo-primary emissions are used to represent the photochemical source from NMVOC oxidation. However, the major reason for the large range of CO emissions is linked to the uncertainties in the global CO emission estimates deduced from smaller scale observations. The atmospheric dynamics (Table 2a) and the spatial and temporal resolutions of each model are different, implying differences in the propagation of the emissions in each model. Finally, the chemical schemes adopted for NMVOC chemistry representation (Table 2b) and consequently the level and distributions of the hydroxyl (OH) radical and the photochemical production of CO also differ in each model. In addition, one should note that the numerical solvers used to integrate the chemical differential equations are different between models.

One model (GFDL) which participated in the overall intercomparison exercise, uses parameterised chemistry and imposes pre-calculated CO distributions. Therefore the results and the characteristics of this particular model will not be considered in the present paper. All other models provided:

- the calculated CO monthly mean mixing ratios from the best case simulation at the Earth's surface, 500 and 200 mbar in January and July.
- the CO monthly mean concentrations (and standard deviation) at 5 selected surface stations (or the grid points of the models closest to these stations) where observations are available.

4. Results and discussion

The 3-D CO distributions calculated by the 11 models show important differences as demonstrated in Figs. 1–4 for January and Figs. 5–8 for July ((a) at surface and (b) at 500 hPa). Calculated CO distributions reflect CO direct emissions, photochemical production by NMVOC and CH_4

MODEL	IMAU ₃	IMAGES	HAR-	UK-	ECHAM	CTMK	MATCH	MO-	TM3	MOGUNTIA	OIO
Last submission of data on	April 97	April 97	VAKU October 97	METO October 97	March 97	April 97	April 97	ZAK1 Septem- ber 97	March 97	October 97	April 97
(a) Model description GCM/CTM (Y/N) Synoptic Upper boundary Number of	NO NO 100 mb 10	NO NO 50 mb 25	NO YES 10 mb 9	NO YES 100 mb (19) 9	YES YES 10 mb 19	NO YES 5 mb 15	NO YES 2 mb 28	NO YES 5 mb 25	NO YES 10 mb 19	NO NO 100 mb 10	NO YES 10 mb 9
verucati layers Isobaric/sigma Horizontal resolution	Isobaric 10°×10°	Sigma 5°×5°	Sigma 4°×5°	Hybrid 5°×5°	Sigma 3.75° ×3.75°	Sigma 8° × 10°	Sigma 1.9°×1.9°	Hybrid 2.8°× 2.8°	Hybrid 3.75° ×3.75°	Isobaric 10°×10°	Sigma 8°×10°
(b) <i>Chemistry descrip</i> NMVOC chemistry –No of NMVOC ^b Chemical solver	tion in the moc NO ^a 0 QSSA	lels YES 6 QSSA	YES 6 paramete-	YES 11 EBI	NO ^a 0 EBI	NO ^a 0 QSSA	NO ^a 0 QSSA	YES 7 EBI	YES 7 EBI	YES 7 QSSA	YES 6 QSSA
Timestep for chemistry	2 h	6 h ^c	4 h ^d	5 min	30 min	2 h	30 min	20 min	40 min	2 h	30 min
Photolysis rates	Calculated	Tables	Paramete- rised based on 6 stream	2 stream	2 stream	Tables	2 stream	Tables	Tables	Tables	2 stream
Frequency of read- ing or calculating photolysis rates		Ð	4 h	45 min	5 d	15 days	Every 30 min	20 min	15 d	15 d	1 mo
^a CO secondary source ^b Including carbonyls fi ^c At the beginning of α ^d A fourth order integra	is added as a pr which direc ach month of a ation scheme i	pseudo-direct t emissions are simulation tim s used.	source; EBI: E considered in estep is 30 min	ulerian Back the model.	cward, QSSA: t three days.	Quasi Stea	dy-State Assi	Imption; Dt	: timestep.		

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Table 2

Table 3 Emissions of ozone precursors use	d in the CJ	ſMs										
MODEL	IM- AU ₃	IMAG- ES (I)	HAR- VARD	UK- METO	EC- HAM	CTMK	MAT- CH	MO- ZART	TM3	MO- GUN- TIA	OIO	Range
CO direct emissions (Tg-CO/yr) Fuel consumption, industry Biomass burning Vegetation/soil Oceans CO secondary source	1350 540 490 280 40 1063	1283 471 626 166 20 1459	1040 520 520 NO NO 1090	1168 303 725 100 40 1320	1250 450 700 100 840 ^d	1427 383 716 166 162 NC	$\begin{array}{c} 1680^{a}\\ 560^{a}\\ 920^{a}\\ 150^{a}\\ 50\\ 884^{d,a}\end{array}$	1218 382 662 162 13 NC	$ \begin{array}{c} 1110\\ 400\\ 450\\ 75^{\circ}\\ 40\\ 1350\\ \end{array} $	1185 500 515 130 40 1235	2362 ^b 893 714 387 378 NC	1040–2362 840–1459
CO surface loss NO emissions (Tg-N/yr) Fuel consumption, industry	NC 37.1 21 (G)	264 36.85 21	NO 45 21 (G)	270 47.5 21 (G)	NO 37.5 21	NC 47.69 22	233 39 21.3 (G)	NC 39.95 21.4 (G)	320 38.4 22 (G)	330 38 21.3 (G)	NC 42.72 21 (G)	0–330 36.8–47.7
Biomass burning Soil Lightning Aircraft Transport from stratosphere CH4 emissions (Tg-CH4/yr) NMVOC emissions (Tg-C/yr)	6° 4.7 0.6 NC S23 NO ¹	4.9 5.5 5 0.45 NC 1254	12 6 0.5 680 680	9 5 ^h 0.5 0.45 442.4	6° 5.5 8 NO 860 650	5.3° 3.9 0.85 0.64 NO	8.5° 5.5° 0.5 1.0 NO	4.5 6.6 0.45 0.45 NC 741	5° 5.3° 0.6 ¹	6° 5.5 7 0.2 540 520	4.4 12.2 ^g 5.5 0.52 0.52 NC 304 748.8	304–540 442–1254
Anthropogenic Natural	ON N	175 1079	80	58.4 384	330 (CO) (CO)	ON ON	225 ^a (CO) 350 ^a (CO)	125 516	(EG) 400	120 400 +- 150	114 ^m 634.8 ^m	
^a 880 Tg CO/yr anthropogenic bu NMVOC oxidation. ^b Global value form Hough (1991) ^c All biosphere. ^d From CH ₄ oxidation only, for E(^e Distribution taken from Hao et a ^f Yienger and Levy (1995). ^g Biomass burning + soils. ^h Price and Rind (1992); Lawrence ⁱ sum of NO. and HNO.	nings and and geogr CHAM bel 1. (1990). et al. (199	40 Tg CO/ aphical dist ow 300 mb 5).	'yr wildfire ribution as ar.	s; 225 Tg for NO _x ,	CO dry d	oxidation sposition v	of NMV(slocity = 0.	DC from te 03 cm/s ov	chnologica er land.	d sources; 3	50 Tg-CO/y	from natural
¹ Prescribed surface concentrations ^k Prescribed surface concentrations ^k Prescribed surface concentrations ^l Includes only C_2H_6 and C_6H_8 chr (G) GEIA inventory; (EG) EDGA introduced in the form of carbon r ^m Anthropogenic emissions accordi	for CH ₄ av of 1772 pl emistry, als R/GEIA ir nonoxide; ing to EPA	ccording to pbv NH an o direct CF iventory; (I ; natural er	Fung et al d of 1680 ₁ H ₂ O emission) Muller an missions ac	. (1991). ppbv SH. ons 300 Tg id Brasseur cording to	/yr. · (1995), u Isaksen a	pdated; Tu nd Hov (19	rman and] 87).	Edgan; (NC	() not calcı	ılated; (NO)	not used; (CO) emissions

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Fig. 1. CO concentrations (ppbv) as computed by the eleven 3-dimensional global models for the month of January at the surface (1000 mbar).



Fig. 2. CO concentrations (ppbv) as computed by the eleven 3-dimensional global models for the month of January at the surface (continue).



Fig. 3. CO concentrations (ppbv) as computed by the eleven 3-dimensional global models for the month of July at the surface (1000 mbar).



Fig. 4. CO concentrations (ppbv) as computed by the eleven 3-dimensional global models for the month of July at the surface (continue).



Fig. 5. CO concentrations (ppbv) as computed by the eleven 3-dimensional global models for the month of January at 500 mbar.



Fig. 6. CO concentrations (ppbv) as computed by the eleven 3-dimensional global models for the month of January at 500 mbar (continue).



Fig. 7. CO concentrations (ppbv) as computed by the eleven 3-dimensional global models for the month of July at 500 mbar.



Fig. 8. CO concentrations (ppbv) as computed by the eleven 3-dimensional global models for the month of July at 500 mbar (continue).

oxidation and destruction by the OH radical, as well as advective and convective transport away from the source regions and losses to the surface.

4.1. Surface CO distributions

Maximum surface CO concentrations are calculated by all models over technologically developed areas of the world and over biomass burning regions. The calculated maximum CO concentrations depend on the model resolution. The lowresolution models are artificially more diffusive than the higher resolution models by splitting emissions in their relatively large grid boxes. Furthermore, the vertical resolution of the surface layers is also different between models and ranges between 50 and 400 m. The models with the shallowest surface layers generally show the highest concentrations. Convective redistributions of emissions, chemistry and surface loss strongly affect the ultimate surface concentrations.

Comparisons between Figs. 1 and 2 and Figs. 3 and 4 (surface for January and July, respectively) show that lower CO concentrations are calculated in July than in January in the northern hemisphere (NH) due to the strong photochemical sink of CO during NH summertime although the photochemical production of CO is also enhanced during that season. This is not the case in biomass burning areas, where CO concentrations remain high although their maximum values are shifted from the NH tropics in January to the southern hemisphere (SH) tropics in July. Almost all models capture this pattern. However, significant differences exist in the magnitude of CO seasonal cycle and absolute levels as shown in Figs. 1-4. These differences will be further discussed when comparing model results to observations.

Sharper interhemispheric gradients are calculated for NH winter since CO increases in the NH due to technological emissions (about 95% in this hemisphere) and relatively low photochemical destruction under wintertime conditions. The longer lifetime of CO during winter than during summer allows CO to be transported over long distances, so that large concentrations of CO are calculated during winter downwind of the CO source areas. On the contrary, in July, high CO concentrations are more restricted to source regions. These patterns are calculated by all models.

All models capture the general pattern of CO linked to the distribution of its surface sources and photochemical production and destruction of CO. However, the calculated CO levels vary significantly between models due to (i) the annual amount of emissions adopted by each model (Table 3) as discussed above, (ii) the formulation of model dynamics (Table 2a), (iii) the spatial resolution, and (iv) the chemical schemes (Table 2b) resulting in significant differences in the hydroxyl (OH) radical distributions calculated by the models. As a consequence the photochemical production of CO from VOCs and its photochemical sink by reaction with OH radical differ significantly from model to model (Table 3). Differences in the OH levels are reflected (i) on the photochemical source of CO that varies between 840 Tg-CO/yr and 1459 Tg-CO/yr and (ii) on the methane (CH_4) lifetimes due to reaction with OH radical in the troposphere below 300 mbar or the closest model level, calculated by the models to vary between 6.6 and 10 yr (median 7.95 yr). Differences in the calculated CO surface distributions can be partly attributed to the surface losses of CO neglected in several models (see Table 3). Daniel and Solomon (1998) show that accurate assessment of this quantity is important at a climatic time-scale. However, as shown in Table 3, this loss could contribute about 10% to the CO removal in the atmosphere, which is in the range of the uncertainties of CO sources.

4.2. Middle troposphere CO distributions

The calculated CO distributions in the middle and upper troposphere are of particular interest (i) because they affect the OH maximum in the middle tropical troposphere and thus CH_4 levels and (ii) they modify ozone concentrations in the upper troposphere where ozone radiative impact maximises. These CO distributions are even more different between the models than the surface distributions since they strongly depend on the transport of CO in the atmosphere by advection, diffusion and convection. In particular differences in the convection parameterisation result in different patterns in the middle Figs. 5 and 6 and Figs. 7 and 8 and upper tropical troposphere (not shown).

In the middle troposphere at 500 mbar Figs. 5 and 6 and Figs. 7 and 8 the detailed signature of the surface sources of CO is lost since transport zonally mixes CO. The sharp interhemispheric gradient of CO remains obvious in the free troposphere in January. This pattern is almost lost in July in most models both in the boundary layer and the free troposphere, due to the relatively short lifetime of CO in NH summer that prohibits efficient CO transport far from its technological sources. Simultaneously in the SH, the CO mixing ratio is more uniform (wintertime conditions).

A notable feature of most models is that they represent the propagation of the CO biomass burning sources to the free troposphere by deep convection, which is very intense in the tropics. However, this phenomenon is represented very differently by the various models, reflecting the differences in the model parameterisation of the convection.

In general, models calculate an important decrease in CO concentrations between the boundary layer and the middle troposphere, the magnitude of which depends on the season, the geographical location and the model. Thus, the CO concentrations at 500 mbar are 0.2–0.8 times those calculated for the boundary layer with the lowest ratios calculated over continental intensive source areas. The reductions in CO between the surface and the 500 mbar over oceans range from 0.6 to 1.0 (no reduction). At some tropical locations models calculate higher CO concentrations in the middle troposphere than in the boundary layer, reflecting strong vertical convective transport and possibly differences in boundary layer sinks of OH.

4.3. Comparison of model results to observations

The observed monthly averages between different years (Novelli et al., 1992) have been compared to the calculated seasonal variation of surface CO concentrations at five surface stations: Barrow (71°N, 157°W), Mace Head (53°N, 10°W), Mauna Loa (20°N, 155°W), Samoa (14°S, 171°W) and Cape Grim (41°S, 145°E). Table 4 depicts the annual mean calculated and observed CO concentrations at these five stations.

To compare the seasonal trends in the models after removing the differences in annual means between the models shown in Table 4, in Figs. 9 and 10 the seasonal variation of the normalised to the annual mean CO concentrations as calculated by the eleven models and as observed at the five surface stations have been plotted. These normalised concentrations correspond to the ratio of the difference between the monthly mean concentra-

Table 4

Calculated and observed annual mean CO concentrations (in ppbv) at five surface stations

Model	Barrow	Mace Head	Mauna Loa	Samoa	Cape Grim
IMAU3	117.5	114.2	66.0	54.0	59.8
IMAGES	114.4	127.4	76.1	50.3	62.9
HARVARD	121.5	130.8	89.5	58.8	62.2
UKMETO	101.0	118.3	73.9	57.8	66.0
ECHAM	153.6	162.3	92.2	68.1	79.9
СТМК	111.4	107.7	67.3	53.2	58.5
MATCH	149.1	222.8	77.7	59.8	71.5
MOZART	160.2	150.8	78.4	58.1	68.9
TM3	124.6	184.7	79.5	51.0	61.5
MOGUNTIA	135.3	161.8	90.4	54.5	69.2
UIO	131.6	130.2	73.7	60.9	74.1
Observations	148.3 (138.6–148.3)	133.1 (128.8–137.4)	96.3 (89.6–103.0)	62.7 (57.6–67.7)	53.4 (49.9–57.0)



Fig. 9. Seasonal variation of the surface CO concentrations normalised to their annual mean: Comparison between model results and observations at Barrow, Mace Head and Mauna Loa.

tion of CO from its annual mean value divided by the annual mean.

It is notable that the differences between models are larger than the inter-annual variability associated with the observations. Most models tend to underestimate CO levels at Barrow (NH) (Table 4) but capture the seasonal trend within 50% (Fig. 9), which indirectly is an indication for OH levels. At



Fig. 10. Seasonal variation of the surface CO concentrations normalised to their annual mean: Comparison between model results and observations at Samoa and Cape Grim.

Mace Head, an NH coastal station, two models largely overestimate the CO levels whereas the remaining are within 30% of the observed values. More models have difficulties in capturing the observed seasonal trend at this location (Fig. 9). At Mauna Loa station, more representative of the free troposphere, all models underestimate CO levels by 4-30 ppbv (i.e. 6-30%, Table 4) depending on the model and overestimate the CO seasonal amplitude in particular in January (Fig. 9). At Samoa (SH) most models (only one exception) underestimate the CO levels by 3-20% and they do not capture its seasonal cycle. On the contrary, at Cape Grim (SH) they overestimate the CO concentrations by 10-50% as well as the seasonal trend (Fig. 9).

These patterns can be related to (i) the adopted emissions (see Table 3) and emission distributions

in time and space, (ii) to the calculated differences in OH radical concentrations between the NH and the SH which are also reflected in the lifetime of methane (CH₄). As a matter of fact, all models calculate the photochemical CH₄ lifetime in the troposphere up to 300 mbar or the closest model level, to be higher in the SH (median 9.2 yr) than in the NH (median 7.2 yr). This implies a lower concentration of OH in the SH than in the NH. However, the opposite trend in OH levels is expected from the lower CO and CH₄ concentrations in the SH as well as from ¹⁴CO measurements (Warneck, 1988; Brenninkmeijer et al., 1992) which are indirect indications for OH levels. This could indicate that NH industrial emissions might be underestimated in the models, in particular those related to biofuel and waste burning.

The overestimate of CO concentrations at Cape Grim may also result from the fact that the observations depicted in this figure are clean sector data. The model results contain all sectors and include air masses originating the Australian continent. They are therefore expected to be higher than the clean sector observations. Sector selection is also performed at the Mace Head station data, where some models tend to overestimate CO levels and the range of model results largely exceeds that of the observations.

5. Conclusions

The intercomparison of eleven 3-D global chemistry/transport models shows significant differences between the models although all of them capture the general patterns in the observed global distribution of CO. The comparison between model results and observations at selected stations allows the evaluation of the model deviations from the observed annual mean CO concentrations at about $\pm 50\%$, whereas differences between models are more important since models can deviate from their annual mean values by up to 80%.

More accurate emission inventories of CO and other ozone precursors, measurements of the global 3-D distribution of tropospheric CO, and better assessment of the chemical processes determining the oxidizing capacity of the troposphere and especially governing the OH levels could help improving model simulations and reducing related uncertainties and model deviations from reality.

Unfortunately within the present exercise the importance of emission inventories and chemical processes cannot be separated from other contributing factors like transport and deposition differences. In future intercomparison exercises, models should preferably use the same emission inventories as input, thereby ruling out differences between inventories as cause of differences between models.

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