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The effect of harmonized emissions on aerosol properties in global models – an AeroCom experiment

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Abstract. The effects of unified aerosol sources on global aerosol fields simulated by different models are examined in this paper. We compare results from two AeroCom experiments, one with different (ExpA) and one with unified emissions, injection heights, and particle sizes at the source (ExpB). Surprisingly, harmonization of aerosol sources has only a small impact on the simulated inter-model diversity of the global aerosol burden, and consequently global optical properties, as the results are largely controlled by model-specific transport, removal, chemistry (leading to the formation of secondary aerosols) and parameterizations of aerosol microphysics (e.g., the split between deposition pathways) and to a lesser extent by the spatial and temporal distributions of the (precursor) emissions.

The burdens of black carbon and especially sea salt become more coherent in ExpB only, because the large ExpA diversities for these two species were caused by a few outliers. The experiment also showed that despite prescribing emission fluxes and size distributions, ambiguities in the implementation in individual models can lead to substantial differences.

These results indicate the need for a better understanding of aerosol life cycles at process level (including spatial dispersal and interaction with meteorological parameters) in order to obtain more reliable results from global aerosol simulations. This is particularly important as such model results

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are used to assess the consequences of specific air pollution abatement strategies.

1 Introduction

One of the largest uncertainties in assessing the human impact on climate is related to the role of aerosol and clouds (IPCC, 2007). The **Aero**sol inter**Com**parison project AeroCom (http://nansen.ipsl.jussieu.fr/AEROCOM) attempts to advance the understanding of global aerosol and its impact on climate by performing a systematic analysis of the results of more than 16 global aerosol models including a comparison with a large number of satellite and surface observations (Guibert et al., 2007[1]; Kinne et al., 2006; Schulz et al., 2006; Textor et al., 2006). In these studies, it was found that significant uncertainty in global modeling of spatial aerosol mass distributions is associated with aerosol processes.

The aerosol mass distributions depend on spatial and temporal distributions of emissions (of aerosols and precursors), on the ambient conditions (e.g., humidity or precipitation) and the transport in the atmosphere as described by the global transport models, as well as on the aerosol microphysical processes (e.g., water uptake or deposition, and chemistry for the production of secondary aerosols) as described by the implemented aerosol module. All these model components are inter-related. AeroCom focuses on the five most important aerosol components: dust (DU), sea salt (SS), sulfate (SO_4), black carbon (BC), and particulate organic matter (POM), and the sum of these components (AER).

In a first set of simulations (AeroCom ExpA, see Textor et al., 2006, referred to as T2006 in the following) each model was run with emission data chosen by the individual participating groups. In these simulations, emission data differed not only because of the use of diverse data sources, but even when referring to the same data source due to different implementation into the models (e.g., regridding, particle size-assumptions). In order to remove the impact of emission diversity on aerosol simulations a sensitivity experiment was performed (AeroCom ExpB) where unified global emission data sets for primary aerosol and aerosol precursors for the year 2000 were prescribed (Dentener et al., 2006).

In this study we compare simulated global mass distributions and underlying processes in AeroCom ExpA and ExpB. In the next two sections we summarize the model-setups and emissions. Then, changes in the diversity of simulated global total aerosol mass distributions are presented and discussed in the context of spatial distributions and residence times of the different aerosol components. New radiative forcing estimates obtained from ExpB and an additional experiment with unified sources for pre-industrial conditions are discussed in Schulz et al. (2006). Supplementary maps and vertical profiles, and many other quantities (tagged FigS in the following) are provided in the electronic supplement to this paper (http://www.atmos-chem-phys.net/7/4489/2007/acp-7-4489-2007-supplement.pdf). Additional information can be found on the AeroCom web site (http://nansen.ipsl.jussieu.fr/AEROCOM/data.html).

2 Model setup

A brief overview of the AeroCom models including a table linking model name abbreviations to the model versions actually used can be found in Table 1, a comprehensive description is given in T2006. The model configurations did not change between ExpA and ExpB, except for three models: In the DLR model, coarse aerosols have been added only in ExpB. Larger changes have been made in KYU, where the interaction between aerosols and clouds has been included for ExpB, and carbonaceous aerosols (BC and POM) are treated externally, unlike the internal treatment in ExpA. In LOA, dry turbulent deposition is only considered in ExpA. In addition, deviations from the recommended AeroCom emissions occurred: In KYU and UIO_GCM, sources of DU and SS remained those of ExpA. For the fine aerosols in KYU, only the emitted aerosol mass flux was matched, but size distributions have not been adapted. In ARQM, emissions have been modified for ExpB, but did not follow the ExpB recommendations. In MATCH, SS sources remained those of ExpA. Due to these deviations, all results of DLR, KYU, LOA, ARQM, as well as UIO_GCM, and the SS and AER results of MATCH are discussed, but not included in the calculation of the model diversities. We also discard the results for all species in UIO_GCM, although the emissions of BC, POM, and the sulfur species are consistent with the AeroCom ExpB recommandations. However, interactions among different aerosol types are taken into account in UIO_GCM, because internal aerosol mixtures are considered. Therefore, the results of all species are influenced by the non-AeroCom emissions of SS and DU. This is, by contrast, not the case in MATCH. Hence, we include GISS, LSCE, MATCH, MOZGN, UIO_CTM, ULAQ and UMI in the statistics, MATCH is excluded from the calculations for SS and AER. The models that are excluded from the statistics are shaded in gray in the figures. Due to our sampling procedure, the statistics on ExpA reported in this paper do not entirely match the results reported in T2006.

The ExpA emissions are discussed in detail by T2006. Models agreed less on the sources of the "natural" aerosol

[1] Guibert, S., Schulz, M., Kinne, S., Textor, C., Balkanski, Y., Bauer, S., Berntsen, T., Berglen, T., Boucher, O., Chin, M., Dentener, F., Diehl, T., Feichter, H., Fillmore, D., Ghan, S., Ginoux, P., Gong, S., Grini, A., Hendricks, J., Horowitz, L., Isaksen, I., Iversen, T., Kloster, S., Koch, D., Kirkevåg, A., Kristjansson, J. E., Krol, M., Lauer, A., Lamarque, J. F., Liu, X., Montanaro, V., Myhre, G., Penner, J., Pitari, G., Reddy, S., Seland, O., Stier, P., Takemura, T., and Tie, X.: Comparison of lidar data with model results from the aerocom intercomparison project, in preparation, 2007.

Table 1. Description of the driver-models and general classification of aerosol modules. Models that follow the AeroCom ExpB recommendations and are included in the calculations of the statistics are marked with $. The following abbreviations are used: accm: accumulation mode; bioburn: biomass burning; coag: coagulation; coam: coarse mode; cond: condensation; dyn: aerosol dynamics apart from sulfur cycle; ext: external; fix: prescribed; hetero: heterogeneous; insol: insoluble; int: internal; mix: mixture; modal M: modal scheme, mass mixing ratio as prognstic variable; modal MN: modal scheme, mass mixing ratio and number conc. as prognostic variables; nucl: nucleation; nuclm: nucleation mode; prog: prognostic; sigma-p: hybrid-sigma p; sol: soluble; supcoam: super coarse mode; thermodyn: thermodynamics.

Model	Global model	Horizont. Resolution (x y)(lon lat)	Vertical Resolution (# of levels) (type)	References for global model	Type of scheme	Number of bins or modes	Aerosol mixing	Aerosol dynamics	References for aerosol module
ARQM	GCM Canadian GCMIII	128×64 2.81°×2.81°	32 sigma-p	Zhang and McFarlane (1995b)	bin	12 all internally mixed	int	nucl, cond, coag thermodyn, cloud processing	Zhang et al. (2001) Gong et al. (2003)
DLR	GCM	96×48	19	Roeckner et al. (1996)	modal MN	A:2, B:3 A: nucm+accm, B:+coam	int	nucl, cond, coag,	Ackermann et al. (1998)
GISS$	GCM modelE	46×72 5°×4°	20 sigma	Schmidt et al. (2006)	bin	13 2 SS, 4 DU, 1 BC, 1 POM, 1 SO_4, 4 DU/SO_4	ext	aging BC POM, hetero DU-SO_4	Koch et al. (1999, 2006); Koch (2001); Bauer and Koch (2005); Cakmur et al. (2006) Koch and Hansen (2005) Miller et al. (2006)
KYU (SPRINT)	GCM CCSR/ NIES/ FRCGC SPRINTARS 5.7b	320×160 1.1°×1.1°	20 sigma	Numaguti et al. (1995); Hasumi and Emori (2004)	bin, modal M for aerosol dyn*	17 bins 10 DU, 4 SS, 1 BC, A; 1 BCPOM, 1 SO_4 (5 modes for aerosol dyn)	B: ext A: partly int for BCPOM	nucl, cond, coag, thermodyn, B: cloud processing	Takemura et al. (2000, 2002, 2005)
LOA	GCM LMDzT 3.3	96×72 3.75°×2.5°	19 sigma	Sadourny and Laval (1984); Hourdin and Armengaud (1999)	bin	16 2 DU, 11 SS, 1 BC (sol+insol), 1 POM (sol+insol), 1 SO_4	ext	aging BC POM	Boucher and Anderson (1995); Boucher et al. (2002); Reddy and Boucher (2004); Guibert et al. (2005)
LSCE$	GCM LMDzT 3.3	96×72 3.75°×2.5°	19 sigma	Sadourny and Laval (1984); Hourdin and Armengaud (1999)	modal MN	5 accm: sol+insol, coam: sol+insol supcoam: sol	ext mix of int modes[2]	aging BC POM	Claquin et al. (1998, 1999); Guelle et al. (1998a, b, 2000) Smith and Harrison (1998) Balkanski et al. (2003) Bauer et al. (2004) Schulz et al. (2006)[2]
MATCH$	CTM MATCH v 4.2	192×94 1.9°×1.9°	28 sigma-p	Zhang and McFarlane (1995a); Rasch et al. (1997) Rasch and Kristjansson (1998)	bin	8 4 DU, 1 SS,1 BC, 1 POM, 1 SO_4	ext	aging BC POM	Barth et al. (2000); Rasch et al. (2000, 2001);
MOZGN$	CTM MOZART v2.5	192×96 1.9°×1.9°	28 sigma-p	Brasseur et al. (1998) Tie et al. (2001, 2005) Horowitz et al. (2003)	bin	12 1 SO_4, 1 POM 1 BC, 5 DU, 4 SS	ext	aging BC POM	Tie et al. (2001, 2005)
UIO_CTM$	CTM OsloCTM2	128×64 2.81°×2.81°	40 sigma	Berglen et al. (2004)	bin	25 8 DU, 8 SS, 4 BC, 4 POM, 1 SO_4	ext except bioburn	aging BC POM	Grini et al. (2002b, 2005) Myhre et al. (2003) Berglen et al. (2004) Berntsen et al. (2006)
UIO_GCM	GCM CCM3.2	128×64 2.81°×2.81°	18 sigma-p	Hack (1994) Kiehl et al. (1998)	modal, M/MN bin for aerosol dyn	12 modes aerosol dyn: 43 bins from 8 int modes DU+SS fix**	4 ext 8 int: mixed +8 fix** from 4 prog	nucl, cond, coag, thermodyn, cloud processing	Iversen and Seland (2002) Kirkevåg and Iversen (2002) Kirkevåg et al. (2005)
ULAQ$	CTM ULAQ	16×19 22.5°×10°	26 log-p	Pitari et al. (2002)	bin	41 7 DU, 9 SS, 5 BC 5 POM, 15 SO_4	ext ext SO_4 microphysics	aging BC POM, Koch, 2001	Pitari et al. (1993, 2002)
UMI	CTM$ IMPACT	144×91 2.5°×2°	30 sigma-p	Schubert (1993); Rotman et al. (2004)	bin	13 3 SO_4, 1 POM, 1 BC, 4 DU, 4 SS	Ext	none	Liu and Penner 2002

∗ KYU describes the size distributions differently for transport and aerosol dynamics. 17 bins are considered for transport. For the aerosol dynamics, a modal approach is employed (one mode per species, sigma fix).

∗∗ UIO_GCM describes the size distributions differently for transport and aerosol dynamics. 12 modes are considered for transport, 4 of them have prescribed size distributions, are transported and not mixed with the other modes (external). The next 4 modes are also transported and only the shape of the distribution is constant (sigma fix). For the aerosol dynamics, these latter 4 modes are internally mixed with 8 prescribed modes, and fitted to 43 bins.

[2] Schulz, M., Balkanski, Y., Textor, C., Guibert, S., Generoso, S., Boucher, O., Breon, F.-M., Hauglustaine, D., and Hourdin, F.: The LMDzT-INCA global aerosol model and its evaluation with surface, lidar and satellite aerosol observations, in preparation, 2007.

components, SS and DU. This is caused by differences in the simulated size spectrum of the emitted particles, by differences in the parameterizations of source strength as a function of wind speed (and soil properties for DU), and by differences in the wind fields themselves. Emissions of the "anthropogenic" species (SO_4, BC, and POM) show better agreement, because of the common use of some few, and usually similar emission inventories. However, these inventories

Table 2. Statistics of models results for DU for the following parameters given in the rows: Emi: emissions, Load: burdens, Wet: wet deposition fluxes, SedDry: total dry deposition fluxes, ResTime: tropospheric residence times, LoadAltF: mass fractions above 5 km height, LoadPolF: mass fractions in polar regions (south of 80° S and north of 80° N), WetofTot: split of removal pathways (mass fraction of wet removal in relation to total removal). The second column gives the unit, and the third the number of models included in the the calculations of the statistics. The next six columns show the means, medians and the model diversities (standard deviations) for experiments A and B.

DUST	unit	#	mean		median		Stdev	
			ExpA	ExpB	ExpA	ExpB	ExpA	ExpB
Emi	Tg/a	7	1640,0	1630,0	1580,0	1670,0	30	4
Load	Tg	7	22,7	21,3	21,3	20,3	21	21
Wet	Tg/a	7	518,0	498,0	516,0	504,0	27	46
SedDry	Tg/a	7	1130,0	1120,0	1040,0	1160,0	50	20
Life	days	7	5,4	4,8	5,1	4,4	26	22
LoadAltF	%	7	14,0	13,4	13,3	13,9	61	61
LoadPolF	%	7	1,7	1,2	1,0	1,0	101	97
WetofTot	%	7	34,9	30,8	36,6	30,3	43	47

Table 3. Statistics of models results for SS, see Table 2 for explanations.

SS	unit	#	mean		median		Stdev	
			ExpA	ExpB	ExpA	ExpB	ExpA	ExpB
Emi	Tg/a	5	8200,0	7720,0	3830,0	7740,0	100	3
Load	Tg	6	7,9	12,7	6,5	12,0	69	31
Wet	Tg/a	5	1320,0	1940,0	1090,0	2220,0	67	45
SedDry	Tg/a	5	6880,0	5780,0	3260,0	5670,0	108	17
Life	days	5	0,5	0,5	0,3	0,6	59	24
LoadAltF	%	6	9,4	9,2	4,5	2,8	111	128
LoadPolF	%	7	4,4	2,9	1,4	1,5	147	127
WetofTot	%	5	21,0	25,3	21,0	28,2	58	45

Table 4. Statistics of models results for SO_4, see Table 2 for explanations. Emi in the first row refers to the sum of emissions of chemical production of SO_4.

SO_4	unit	#	mean		median		Stdev	
			ExpA	ExpB	ExpA	ExpB	ExpA	ExpB
Emi	Tg/a	5	183,0	164,0	205,0	176,0	26	23
Load	Tg	7	2,2	2,1	2,2	2,3	25	25
Wet	Tg/a	6	158,0	142,0	174,0	153,0	27	24
SedDry	Tg/a	6	22,3	20,4	21,5	19,8	31	29
Life	days	6	4,4	4,5	4,5	4,5	19	18
LoadAltF	%	7	32,9	33,3	29,5	30,9	30	28
LoadPolF	%	7	6,7	6,0	7,6	6,9	43	47
WetofTot	%	6	86,9	86,9	87,5	87,0	5	5

have often been improved for certain species or emission types by the individual modelers, and their mix in each of the ExpA models is variable, see references in T2006.

The unified emission data used in ExpB have been recompiled from various recently published inventories, augmented with data generated for the purpose of the AeroCom ExpB as explained in detail by Dentener et al. (2006). The inventory includes fluxes for "natural" emissions of mineral DU, SS, dimethyl sulfide (DMS) from the oceans, sulfur dioxide (SO_2) from volcanoes, sulfate and carbon from

Table 5. Statistics of models results for BC, see Table 2 for explanations.

BC	unit	#	mean		median		Stdev	
			ExpA	ExpB	ExpA	ExpB	ExpA	ExpB
Emi	Tg/a	7	11,6	7,8	11,3	7,8	10	1
Load	Tg	7	0,3	0,2	0,2	0,2	46	26
Wet	Tg/a	6	9,3	6,2	9,4	6,2	17	11
SedDry	Tg/a	6	2,4	1,6	2,6	1,5	33	38
Life	days	7	8,1	7,6	7,2	7,2	41	26
LoadAltF	%	7	21,3	22,8	17,4	17,5	54	53
LoadPolF	%	7	5,1	3,9	6,1	4,3	66	67
WetofTot	%	6	79,0	79,8	78,3	80,0	10	10

Table 6. Statistics of models results for POM, see Table 2 for explanations.

POM	unit	#	mean		median		Stdev	
			ExpA	ExpB	ExpA	ExpB	ExpA	ExpB
Emi	Tg/a	7	95,3	66,6	95,6	66,9	30	1
Load	Tg	7	1,8	1,3	1,7	1,2	15	18
Wet	Tg/a	6	81,8	53,0	79,5	52,1	33	11
SedDry	Tg/a	6	19,9	13,0	19,9	13,9	25	38
Life	days	7	7,4	6,9	7,1	6,7	26	18
LoadAltF	%	7	21,4	22,5	17,5	19,7	59	54
LoadPolF	%	7	3,9	3,3	4,6	3,9	63	65
WetofTot	%	6	79,1	80,3	77,0	78,9	10	10

natural wild-land fires, and POM including secondary organic aerosol. In addition, anthropogenic emissions from biomass burning and fossil fuel burning of SO_2, POM and BC are provided. The prescribed emission fields are generated on a global $1^\circ \times 1^\circ$ spatial resolution, and a temporal resolution ranging from daily to annual. Injection heights for volcanic and wildfire emissions, and size distributions of the primary particulate emissions are prescribed. In this paper we focus on the emissions representative for present-day conditions. The models were nudged to (different) meteorological data sets for the year 2000. Four models without nudging capability (General Circulation Models: ULAQ, UIO_GCM, ARQM, and DLR), provided climatological averages from 5 years of simulation using the same emissions after a spin-up period of one year.

3 Results

3.1 Emissions

A comparison of the emissions in ExpA and ExpB shows that in most models the mass fluxes of "natural" aerosols (coarser sized SS and DU) are larger in the latter experiment (on average by 87% and by 4%, respectively). The emissions of carbonaceous species, BC and POM, are on average by 37% and by 27%, respectively, smaller. The total SO_4 sources decreased by 4%, see below for a discussion. For the model-average relative changes of the source mass fluxes see also Table 8. Model diversities for emissions and the all-model-averages and all-model medians for the annually and globally averaged source fluxes are given in Tables 2–7 and in Figs. S1. Figures S2 show the annual mean zonally averaged emissions.

The implementation of the unified AeroCom sources in ExpB strongly reduced the diversity of global annual emission mass fluxes when compared to ExpA. However, some differences in the emissions remained due to model-specific representations of the particle size distributions (bin schemes or modal schemes, and the number of modes or bins), or simply by inaccurate implementation leading to small deviations from the AeroCom data. In addition, the initial degree of the mixing height, i.e., the impact of the sources on the vertical concentration profile, is governed by the model architecture (e.g., height of model levels and the emission scheme).

The model diversity, i.e., the scatter of the model results, is defined here as the standard deviation of the globally and annually averaged model results, normalized by the all-models-average, for a detailed discussion see T2006. The large emission mass flux diversities in ExpA are sharply reduced in

Table 7. Statistics of models results for AER, see Table 2 for explanations.

AER	unit	#	mean		median		Stdev	
			ExpA	ExpB	ExpA	ExpB	ExpA	ExpB
Emi	Tg/a	5	10100,0	9590,0	5930,0	9680,0	79	3
Load	Tg	6	35,8	36,2	36,7	37,7	18	16
Wet	Tg/a	5	2100,0	2610,0	2010,0	2940,0	50	42
SedDry	Tg/a	5	8000,0	6960,0	5130,0	6870,0	89	17
Life	days	5	1,9	1,3	1,7	1,4	61	16
LoadAltF	%	6	15,3	14,5	12,4	11,2	54	62
LoadPolF	%	7	2,9	2,3	2,0	1,8	94	96
WetofTot	%	5	24,9	27,3	26,3	30,0	43	42

Table 8. Model average relative changes of parameters between ExpA and ExpB expressed as (ExpB-ExpA)/ExpA in [%] for the aerosol species under consideration here (rows). The mean and the median changes are shown in second and third column for emissions, in the fourth and fifth column for loads, in the sixth and seventh column for residence times, and in the eighth and ninth column for the fractions of wet deposition in relation to total deposition, respectively.

	Emi [Tg/a]		Load [Tg]		ResTime [days]		WetofTot [%]	
	Mean	Median	Mean	Median	Mean	Median	Mean	Median
DUST	3,8	3,0	17,5	−4,6	0,0	−9,9	−13,7	−0,9
SS	87,0	102,0	92,8	64,4	53,0	63,5	37,0	47,7
SO_4	−4,1	0,2	9,1	1,1	1,5	5,7	2,1	0,2
BC	−36,5	−31,9	−27,0	−32,8	6,4	4,4	2,6	1,5
POM	−27,0	−30,7	−18,0	−24,6	5,9	8,0	2,4	2,2
AER	46,9	64,9	14,1	3,1	152,0	−22,3	35,5	24,2

ExpB to less than 5%, except for SO_4. The diversity δ of the total SO_4 source in ExpB (δ=23%) is almost as large as in ExpA (δ=26%). SO_4 originates predominantly (about 97% on average in both experiments) from model-specific chemical production as sulfur-containing precursor gases (DMS and SO_2) are oxidized. Direct emission of SO_4 decreased from ExpA to ExpB by 11% and that of the precursor gases SO2 and DMS by 11% and 50%, respectively. 79% (90%) of the secondary SO_4 stems from SO_2, and 21% (10%) from DMS oxidation in ExpA (ExpB). A comparison of the individual processes involved in the sulphur cycle shows, that the diversity in SO_4 sources is due to differences in precursor gas emissions, but differences in the dry deposition of these gases and the chemical production are much more important, see Figs. S3. The individual chemical production pathways in the liquid and gas phase show larger diversities than the total chemical production. This indicates compensation effects. The production of SO_4 from precursor gases leads to the larger diversity of SO_4 when compared to the other aerosol components. Note however, that the statistics of the sulfur cycle is based on only three models, which delivered all quantities involved for both experiments.

3.2 Total mass

The changes in (global annual) masses for individual aerosol components between ExpA and B are generally consistent with those for emissions: models with increased emissions show larger mass and vice versa, for the model-average relative changes of the total masses see also Table 8.

The associated model diversities of the simulated global annual masses are shown in Fig. 1, and in Tables 2–7. In addition, the electronic supplement provides a plot showing the annually masses for the individual models, see Fig. S4. Surprisingly, mass diversity is not considerably smaller in ExpB with harmonized emission mass fluxes. The apparent strong decrease in mass diversities for SS and BC in ExpB results from a few strong outliers in ExpA that are removed in ExpB. These results indicate that diversities for the simulated aerosol mass depend largely on differences of model-specific transports and parameterizations of aerosol interactions with its environment and of microphysical processes, and to a lesser extend on their (precursor) emissions.

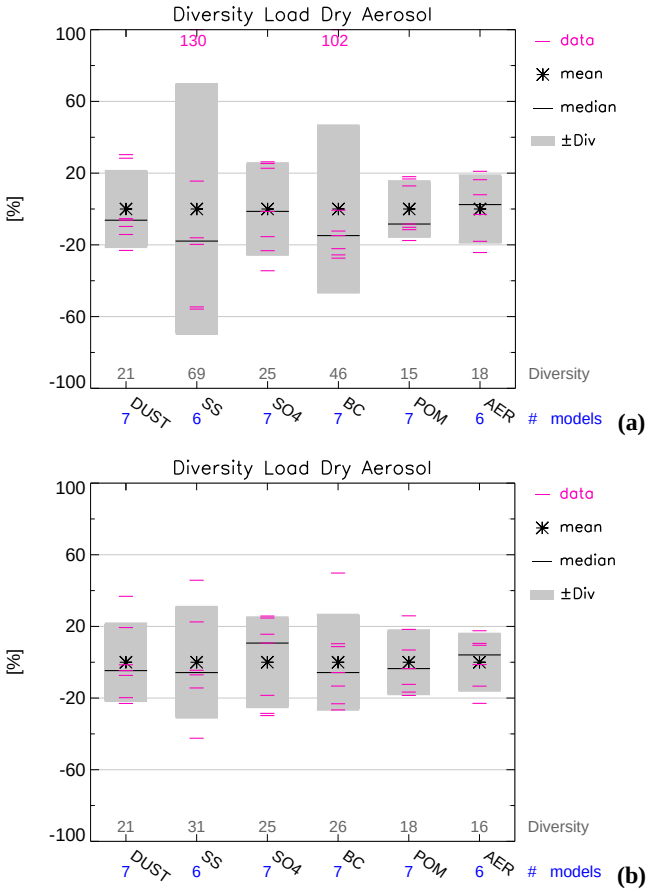

Fig. 1. Model diversities of the global, annual average aerosol burden of the five aerosol species in **(a)** ExpA and **(b)** ExpB. The diversity ranges (±Div = ± standard deviation δ) are indicated by gray boxes, the diversities are given in gray in the lower part of the plot for each species. The individual models' deviations from the all-models-averages are plotted as pink lines ("data"), or as pink numbers if they are outside the scale of the plot. The all-models-averages are indicated by a black star (at 0%) and the medians by a black line (i.e., deviation of the median from the all-models-average). The numbers of models included in the calculation of this statistics are shown in blue below the x-axis.

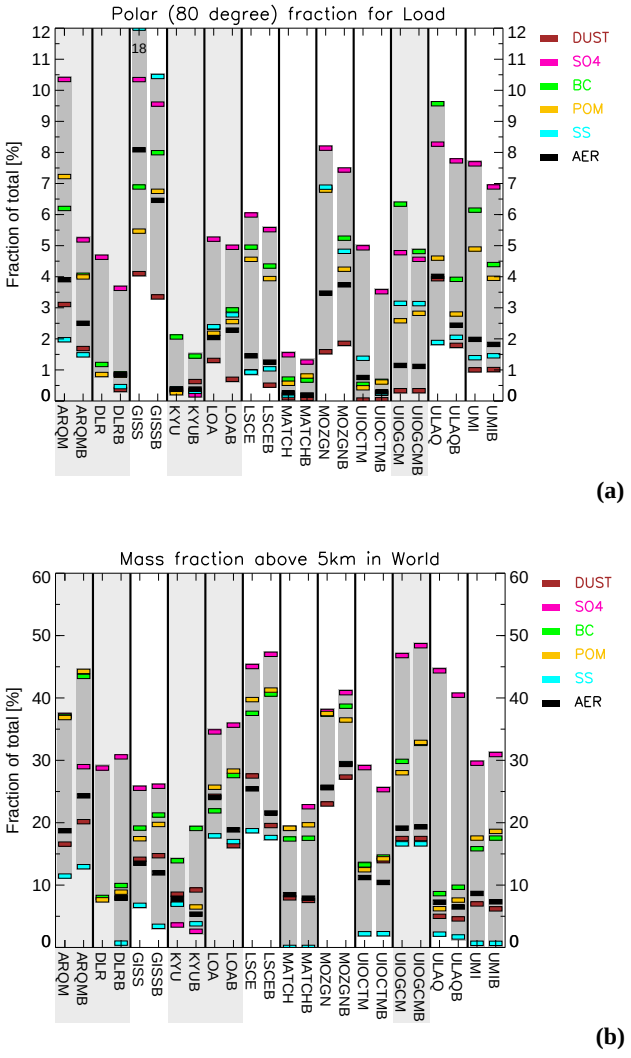

Fig. 2. **(a)** Global, annual average mass fractions in [%] of total mass in polar regions (south of 80° S and north of 80° N) for the AeroCom models. **(b)** Global, annual average mass fractions in [%] of total mass above 5 km altitude for the AeroCom models. The dark gray shadings frame the range for each model. The models that are excluded from the statistics are shaded in light gray in the figures.

3.3 Spatial distributions

Horizontal and vertical dispersal differed considerably among models that participated in AeroCom experiments (T2006), and harmonizing the sources in ExpB does not lead to significantly higher model agreement, see Figs. S5 and S6 electronic supplement for maps. In T2006, we defined two metrics describing the differences of the simulated spatial aerosols pattern. The degree of vertical dispersal was characterized by the mass fractions above 5 km height, where wet scavenging should become increasingly less significant due to the decrease in cloud precipitation efficiency. The degree of horizontal dispersal, i.e., meridional long-range transport, was described by the mass fractions in polar regions (defined as south of 80° S and north of 80° N) because polar regions are far from most aerosol sources and not influenced by differences in local emissions. They thus reflect the long-range transport efficiency of the models.

Meridional and vertical dispersal as simulated in ExpA and ExpB are compared in Fig. 2, the corresponding model diversities are given in Tables 2–7, and in Figs. S7 and S8. The diversities are similar in the two experiments. Hence, harmonization of the spatial and temporal distributions of the aerosol sources did not lead to higher agreement in mass dispersal. This indicates differences in the simulated aerosol transport efficiency itself. The more detailed analysis in

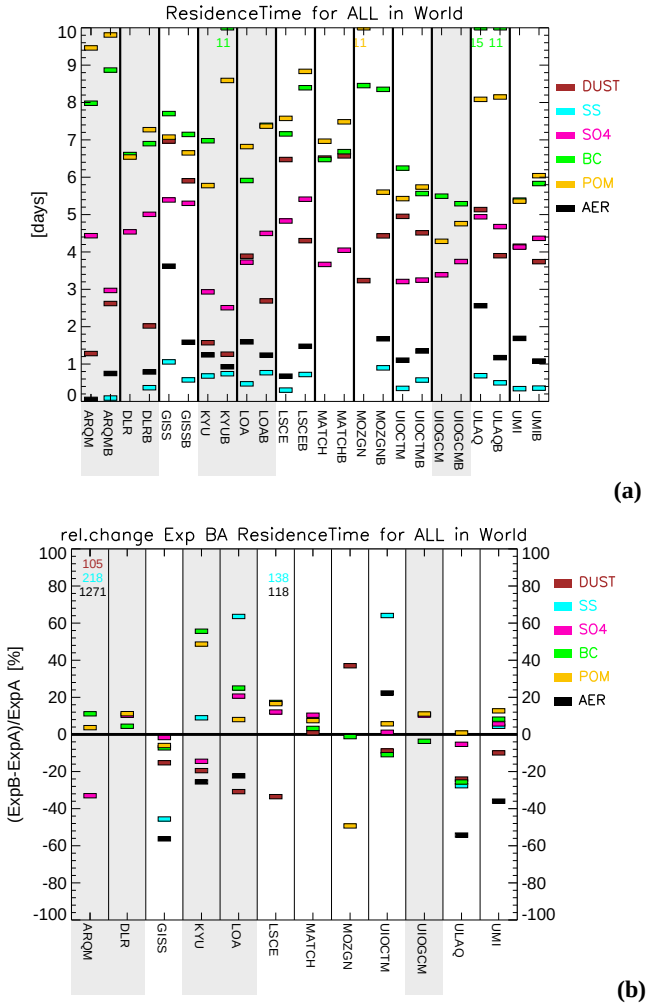

Fig. 3. Tropospheric residence times in ExpA and ExpB in [days], **(b)** Relative changes between ExpA and B expressed as (ExpB-ExpA)/ExpA in [%]. The models that are excluded from the statistics are shaded in light gray in the figures.

Fig. 2 shows, that spatial dispersal is more similar for any pair of simulations performed by an individual model than among all models using the same emissions in ExpB. (For additional layers at different heights (0–1, 1–2.5, 2.5–5, 5–10, >10 km) see the electronic supplement in Fig. S9.) Thus, meridional and vertical dispersals seem to be determined by the model-specific combined effects of transport and the parameterizations of internal aerosol processes.

3.4 Residence times

Another way of looking at model differences is the comparison of the (tropospheric) residence time τ, which is defined as the ratio of global burden and emission. Figures 3 show residence times in ExpA and ExpB and the relative changes between both experiments. The residence time remained unchanged for DU, the relative increases of τ in ExpB relative to ExpA are on average 53% for SS, 2% for SO_4, and 6% for BC and POM, respectively, see also Table 8. The relative increase of τ reached 152% for AER.

The variations of τ between ExpA and ExpB are caused by the changes in spatial and temporal distribution of aerosol sources, that caused changes in the aerosol fields and thus in aerosol removal processes. The modification of the residence times of SO_4 can also be attributed to changes of pre-cursor gas removal and of the conditions, under which its pre-cursor gases are oxidized to SO_4, e.g., the coincidence of clouds and SO_2 (See also the discussion in Sect. 3.1). Model diversities for residence times (Tables 2–7, and Figs. S10) are somewhat smaller in ExpB than in ExpA, especially because some outlier that occured in ExpA have been removed in ExpB.

The residence times depend on the simulated individual removal pathways. We examine these pathways and distinguish between wet and dry deposition, where the latter comprises turbulent deposition and sedimentation (see discussion in T2006). Fine aerosols (SO_4, BC, and POM) are mainly removed by wet deposition (on average about 80–90% by mass in both experiments, see also Tables 2–7). The split between the two removal pathways for fine aerosols is almost exactly the same for most of the models, changes are smaller than 5% (see Fig. 4a for sulfate as an example for the fine fraction), and the diversity among models is similar in both experiments. (The figures for split between the removal path ways for the remaining species and the corresponding model diversities can be found in the electronic supplement Figs. S11 and S12.) Larger changes occur for LOA, KYU, and ARQM, but these models do not entirely fulfill the experiment requirements, see Sect. 2. The split between wet and dry removal is thus not sensitive to a change in emissions and associated assumptions on particles sizes. Since also the meteorological fields are equal in both experiments (at least for the CTMs) and thus the spatial distribution of clouds and precipitation, we can conclude that the changes in residence times for fine aerosols shown in Fig. 3 and Tables 2–7 are due to the changes in the spatial distribution of emissions (and deposition of precursor gases as well as chemical production in the case of SO_4).

Another reason for the variations of τ between the two experiments is the change in particles sizes (especially for the coarse aerosols SS and DU). The split of "fine" (diameter $<1\,\mu$m) and "coarse" (diameter $>1\,\mu$m) mode particles has been discussed in T2006. The mass fractions in the fine mode are 11% (7%), 18% (9%), 93% (93%), 99% (99%), and 99% (98%), for DU, SS, SO_4, BC, and POM in ExpA (ExpB), respectively. Figures S13 show the results in ExpA and ExpB and the corresponding model diversities. The models agree much better on the mass fractions of "anthropogenic" particles in the fine mode (diversities of δ=7% (δ=6%) for SO_4 in ExpA (ExpB), and δ=2% for BC and POM in both experiments) as one would expect. However, the splits are model-specific. Model diversity remains very large for the "natural" aerosols (SS δ=131% (δ=100%), DU δ=67% (δ=94%)

in ExpA (ExpB)). This large disagreement can be explained by the model-specific representations of aerosol size distributions. These lead to differences in the partinioning of the particle sizes recommanded in ExpB on the model size classes, for a more detailed discussion see T2006. In addition, the dependence of the microphysical processes on particle size is not well known, and is differently parameterized in the models.

For the coarser aerosols (SS and DU), dry deposition is with about 65–80% of the removal mass fluxes the dominant process in both experiments (see Tables 2 and 3, and Fig. 4b as an example for SS). Changes between the two experiments exist even for those models which had been shown to have an equal pathway split for fine aerosol. Model diversity of the mass deposited by dry deposition decreased from ExpA to ExpB from 53% to 20% for DU, and from 107% to 19% for SS. These findings indicate the influence of harmonized size distributions in ExpB on the dry removal rates for SS, where at least the spatial distribution of the sources should be similar in both experiments. For DU, the larger model diversity of the sizes in ExpB is not transferred to the residence times. Therefore, in this case the spatial and temporal distribution of the sources seems to play a major role. However, for both coarse aerosol species, the split between the deposition pathways is still rather model-specific and less dependent on the change in the sources.

The interested reader can find additional figures in the electronic supplement showing analyses as those performed in the T2006 paper for both experiments including effective sink rate coefficients in Figs. S14 and S15, and the split between stratiform and convective wet removal in Figs. S16.

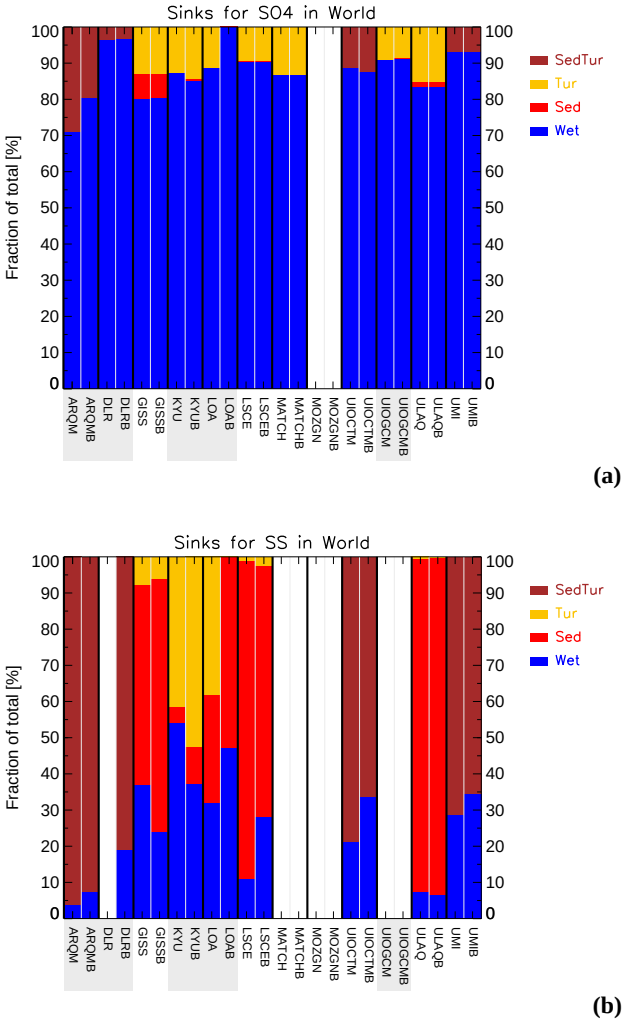

Fig. 4. Contribution of the individual removal processes to the total sink mass flux (annually and globally averaged) for the AeroCom models for **(a)** SO_4 and **(b)** SS in [%]. The color code is given in the legend. Wet refers to wet deposition. If possible we show the individual dry sink rate coefficients (Tur: turbulent deposition, and Sed: sedimentation), otherwise the sum of the two processes (Dry = SedTur) is plotted. The models that are excluded from the statistics are shaded in light gray in the figures.

4 Discussion

The effects of aerosols on climate and air quality are highly uncertain, both in terms of their mechanisms (e.g., importance of secondary organic aerosols, or indirect effects on clouds and precipitation) and in terms of their quantity. The assessment of aerosol effects necessitates a profound knowledge of the aerosol life cycle that can presently not be gained from observations alone. The application of numerical models using high-quality inventories of aerosol precursor gas and primary aerosol emissions and parameterisations of aerosol processes in the atmosphere are required in order to evaluate coherent reduction strategies.

The realistic description of aerosol emissions in modeling is very important. However, despite significant improvements of the inventories in recent years, large uncertainties remain. Crucial information about initial aerosols properties, e.g., related to size distribution or composition, is still missing. In addition, the representation of such properties needs to be improved in the model parameterisations. In our paper, however, we do not examine the quality of emission inventories, but focus on differences among model results, when harmonizing the emission input to models (AeroCom Exp B). The primary intention is to explore, if emission input is the main driver for the simulated model diversity in AeroCom ExpA, where modellers used emission data of their own choice. We can, however, not draw quantitive conclusions on the relative contribution of emission input and other processes to the *uncertainty* of aerosol model simulations, because we only know the model *diversity* from the AeroCom study. It is indeed very likely that the uncertainty of aerosol emissions is even larger than represented by the emission diversity in AeroCom ExpA, because only a few emission inventories with acknowledged high uncertainties are available.

We show that even with (almost) identical emissions, the global species budgets and the simulated relative importance of the contributing processes remains largely unchanged in the individual models. Therefore, the overall model diversity did not considerably decrease in ExpB when compared to that of ExpA. This indicates, that the simulated fate of aerosols is to a large extent model-dependent and controlled by processes other than the diversity in emission input in current global aerosol models. Please note, that the model diversities calculated here are still based on a rather small number of models. Hence, the statistics are not be very robust, and the numbers for the diversities given in this study should be taken as the first estimation of the real diversity of aerosol process modelling on the global scale.

Usually, emission inventories are obtained from bottom-up techniques integrating all available information on the sources. Recently, top-down techniques have been applied in inverse studies using improved satellite information in combination with numerical models in order to infer strength and geographic distribution of the emissions (e.g., Zhang et al., 2005). Model studies have investigated the effect of modified aerosol emissions: Stier et al. (2006) demonstrated non-linear responses of global aerosol fields when modifying aerosol emissions in their simulations considering aerosol component interactions. De Meij et al. (2006) have evaluated the impact of differences in the EMEP and AEROCOM emission inventories on the simulated aerosol concentrations and optical depths in Europe, and demonstrated that seasonal variations in the emissions should be considered. All these studies show important effects when modifying the simulated aerosol emissions that had been neglected so far. Our results indicate that the magnitude of the impact of emission changes on aerosol effects might depend on the individual model configuration. Recent studies have shown that the results from ensemble simulations better represent the observations than the individual models (e.g., McKeen et al., 2005; Pagowski et al., 2005; Vautard et al., 2006). This finding is also true for the AeroCom models. Therefore, we recommend to use an ensemble of models when aiming at a quantitative assessement of the impacts from emission changes, until robust quality measures become available.

This paper shows that differences in aerosol sources alone cannot explain inter-model diversity. The question is, which are the possible causes of the remaining still large model diversity obtained from the simulations in ExpB with unified sources? And which are the critical processes that cause the large differences between models? We have looked at different processes and parameters and tried to group models according to their parameterisations or model architecture. We could, however, not determine any clear relation between the modeling results and the model structure. The processes involved in the aerosol life cycle are strongly inter-related. The importance of a specific process depends on that of all others, and it changes with the ambient conditions and with the aerosol type. For example, wet deposition is the main sink process for highly soluble aerosols like sulfate, but in a region where wet scavenging is less important due to the lack of precipitation, dry deposition becomes the most important process.

More detailed investigations are neccessary that focus on specific regions, seasons, aerosol types and processes. In particular, we expect clarifications from sensitivity studies comparing tendencies of individual processes with constraints imposed on other processes. Tracer experiments are envisaged to examine transport and aerosol dispersal patterns. In addition, we would like to point out, that the model diversity is not only caused by differences in aerosol process modeling but also influenced by the transport (advection and mixing) as well as the meteorological conditions (such as relative humidity, clouds and precipitation) provided by the global host models in which the aerosol modules are implemented. A recent paper by Liu et al. (2007) shows very large differences of the simulated aerosol fields when using different meterological datasets to drive the same aerosol module.

It is a major goal of AeroCom to compare model simulations against measurements. However, existing data mainly refer to surface concentrations or optical properties, because the observations of individual processes is very challenging. Kinne et al. (2006) have shown that although model simulations agree quite well with the observed *total* aerosol optical depth, the contributions from the individual components differ considerably among models. A comparison of the simulated optical depths from ExpA and ExpB in Figs. S17 in the electronic supplement shows, that the composition of the total optical depth is model-specific, and less dependent on aerosol sources, as for the parameters discussed above. Model diversity, see Figs. S17b and c, even slightly increases in ExpB with unified sources for the coarse aerosols (SS and DU), and hence for total aerosol.

5 Conclusions

In this paper, the effects of unified aerosol sources on the simulated aerosol fields have been examined. We compared the results of twelve models for two sets of simulations, one without any constraints on aerosol sources (ExpA), and one where mass fluxes, injection heights and particle sizes of emissions were prescribed (ExpB). Although the aerosol emissions' diversity among models strongly decreased, we realize that it is not straightforward to implement prescribed aerosol (precursor) sources in exactly the same way into different model configurations. Inconsistencies in the actually simulated source fluxes were caused by differences in the model architecture and the representation of the particle size distributions, or simply by inaccurate implementation.

The comparison of the results from ExpA and ExpB shows, that harmonized emissions do not significantly reduce model diversity for the simulated global mass fields. The spatial dispersal and the removal pathways are model-specific

and less depending on the properties of the aerosol sources. This shows that modeled aerosol life cycles depend to a large extent on model-specific differences for transport, removal, chemistry (e.g., formation of sulfate or secondary organics) and parameterizations of aerosol microphysics, and to a lesser extent on the spatial and temporal distributions of the (precursor) emissions. These results indicate the need for a better understanding of aerosol life cycles at process level (including spatial dispersal and interaction with meteorological parameters and processes) in order to obtain more reliable results from global aerosol simulations. This is particularly important as such model results are used to assess the consequences of specific air pollution abatement strategies. We recommend to use an ensemble of model simulation, when assessing the impacts from emission changes, until robust quality measures become available.

The AeroCom initiative aims to better understand which processes are the main contributors to model diversity. The interdependence of the processes involved in the aerosol life cycle complicates this task. Detailed evaluation studies against measurements for different regions and different seasons and looking at specific processes are performed. Efforts are made to establish data test beds on a regional and seasonal basis that are sufficiently accurate to help evaluating specific processes in modeling. Additional simulations dedicated to specific processes are envisaged, where individual parameterizations are tested within at least one global host model.

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