Atmos. Chem. Phys. Discuss., 7, 1699–1723, 2007
www.atmos-chem-phys-discuss.net/7/1699/2007/

Atmospheric Chemistry and Physics Discussions

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

C. Textor[1,19], M. Schulz[1], S. Guibert[1], S. Kinne[2], Y. Balkanski[1], S. Bauer[3], T. Berntsen[4], T. Berglen[4], O. Boucher[5,18], M. Chin[16], F. Dentener[6], T. Diehl[17], J. Feichter[2], D. Fillmore[7,1], P. Ginoux[9], S. Gong[10], A. Grini[4], J. Hendricks[11], L. Horowitz[9], P. Huang[10], I. S. A. Isaksen[4], T. Iversen[4], S. Kloster[2,6], D. Koch[3], A. Kirkevåg[4], J. E. Kristjansson[4], M. Krol[6,12], A. Lauer[11], J. F. Lamarque[7], X. Liu[13,8], V. Montanaro[14], G. Myhre[4], J. E. Penner[13], G. Pitari[14], S. Reddy[5,9], Ø. Seland[4], P. Stier[2,20], T. Takemura[15], and X. Tie[7]

[1]Laboratoire des Sciences du Climat et de l'Environnement, Gif-sur-Yvette, France
[2]Max-Planck-Institut für Meteorologie, Hamburg, Germany
[3]Columbia University, GISS, New York, USA
[4]University of Oslo, Department of Geosciences, Oslo, Norway
[5]Laboratoire d'Optique Atmosphérique, Université des Sciences et Technologies de Lille, CNRS, Villeneuve d'Ascq, France
[6]European Commision, Joint Research Centre, Institute for Environment and Sustainability, Climate Change Unit, Italy
[7]NCAR, Boulder, Colorado, USA
[8]Battelle, Pacific Northwest National Laboratory, Richland, USA

[9]NOAA, Geophysical Fluid Dynamics Laboratory, Princeton, New Jersey, USA
[10]ARQM Meteorological Service Canda, Toronto, Canada
[11]DLR-Institut für Physik der Atmosphäre, Oberpfaffenhofen, Germany
[12]Institute for Marine and Atmospheric Research Utrecht (IMAU) Utrecht University, The Netherlands
[13]University of Michigan, Ann Arbor, MI, USA
[14]Universita degli Studi L'Aquila, Italy
[15]Kyushu University, Fukuoka, Japan
[16]NASA Goddard Space Flight Center, Greenbelt, MD, USA
[17]Goddard Earth Sciences and Technology Center, University of Marylan Baltimore County, Baltimore, Maryland, USA
[18]Hadley Centre, Met Office, Exeter, United Kingdom
[19]Service d'Aéronomie, CNRS/UPMC/IPSL, Paris, France
[20]Department of Environmental Science and Engineering, California Institute of Technology, Pasadena, USA

Received: 18 December 2006 – Accepted: 19 January – Published: 2 February 2007

Correspondence to: C. Textor (christiane.textor@aero.jussieu.fr)

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 diversity for aerosol burden, and consequently 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 on 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 diversity for these two species was caused by few outliers. The experiment also indicated 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 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, 2001). The Aerosol inter Comparison project AeroCom (http://nansen.ipsl.jussieu.fr/AEROCOM) attempts to advance the understanding of the 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 distribution 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 secondary aerosols) as described by the implemented aerosol module. All these model components are inter-related, since aerosol mass is conserved. AeroCom focuses on five most important aerosol components: dust (DU), sea salt (SS), sulfate (SO4), 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)) each model was run with emission data for each aerosol component 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, 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 (Dentener et al., 2006) were prescribed.

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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 are provided on the AeroCom web site (http://nansen.ipsl.jussieu.fr/AEROCOM/data.html).

2 Model setup

A comprehensive description of the AeroCom models including a table linking model name abbreviations to the model versions actually used can be found in Textor et al. (2006). The model configurations did not change between ExpA and ExpB, except for three models: in the DLR model, coarse aerosols have been implemented 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. In KYU_B, 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, and ARQM, SS results of MATCH and UIO-GCM, and DU results of UIO_GCM are discussed, but not included in the calculation of the model diversities shown in Fig. 1 and Tables 1–6. Due to this sampling procedure, the statistics on ExpA reported in this paper does not entirely match the results reported in Textor et al. (2006).

The ExpA emissions are discussed in detail by Textor et al. (2006). Models agree less on the sources of the "natural" aerosol 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 (SO4, BC, and POM) show better agreement, because of the common use of some few, and usually similar emission inventories. However, these inventories 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 Textor et al. (2006).

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 natural wild-land fires, and particulate organic carbon including secondary organic aerosol. In addition, anthropogenic emissions from biomass burning and fossil fuel burning of SO_2, particulate organic matter and black carbon 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, which could only operate in a climatological mode (ULAQ, UIO_GCM, ARQM, and DLR), provided averages from 5 years of simulation.

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 (on average by 77% and by 9%, respectively). The emissions of carbonaceous emissions, BC and POM, are on average by 33% and by 23%, respectively, smaller. The total SO4 sources decreased by 8%, see below for a discussion. For the model-average relative changes of the source mass fluxes see also Table 7.

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. In addition, the initial degree of the mixing height is governed by the model architecture (e.g., height of model levels and the emission scheme). Some further discrepancies were caused by the use of intermediate versions of the AeroCom emission data.

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 Textor et al. (2006). Model diversities along with all-model-averages and all-model medians for the annually and globally averaged source fluxes are given in Tables 1–6. The large emission mass-flux diversities in ExpA are sharply reduced in ExpB to less than 5%, except for SO4. The diversity of the total SO4 source in ExpB (21%) is almost as large as in ExpA (25%). SO4 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 SO4 decreased by 11% and that of the precursor gases SO2 and DMS by 11% and 50%, respectively, in ExpB. 79% (90%) of the secondary SO4 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 SO4 sources is due to differences in precursor gas emissions, but differences in the dry deposition of these gases and the chemical production are as important. This leads to the larger diversity of SO4 when compared to the other aerosol components. Note however, that the statistics of the sulfur cycle is based on only four 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. However, this is not true, when diversity in size assumptions comes into play, as for DU and SS. For the model-average relative changes of the total masses see also Table 7.

The associated model diversities of the simulated global annual masses are shown in Fig. 1, and in Tables 1–6. 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 aerosol interactions with its environment and microphysical processes, and to a lesser extend on their (precursor) emissions.

3.3 Spatial distributions

Horizontal and vertical dispersal differs considerably among models that participated in AeroCom experiments (Textor et al., 2006). Meridional dispersal (represented here as mass fractions in polar regions) and vertical dispersal (as mass fractions above 5 km altitude) simulated in ExpA and ExpB are compared in Fig. 2. Model diversities for polar and upper troposphere mass fractions (Tables 1–6) are similar in two experiments,

and the more detailed analysis in Fig. 2 shows, that the aerosol dispersal did not significantly change between the two experiments. 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. Thus, meridional and vertical dispersals seem to be determined by the model-specific combined effects of transport and the parameterization of internal aerosol processes.

3.4 Residence times

Another way of looking at model differences is the (tropospheric) residence time τ, which is defined as the ratio of burden and emissions. Figs. 3 show residence times in ExpA and ExpB and the relative changes between both experiments. τ is on average by 8% smaller for DU and by 27% larger for SS in ExpB relative to ExpA. The residence times of SO4 and BC changed by +5% and by –3%, respectively, and remained unchanged for POM, see also Table 7. The variations of τ between the two experiments are caused by the changes in particles sizes (leading to the larger changes for the coarse aerosols SS and DU), and by the changes in spatial and temporal distribution of aerosol sources, spatial distributions, and their removal processes. The modification of the residence times of SO4 can also be attributed to changes of pre-cursor gas removal and of the conditions, under which its pre-cursor gases are oxidized to SO4, e.g., the coincidence of clouds and SO_2 (See also the discussion in Sect. 3.1). Model diversities for residence times (Tables 1–6), are smaller in ExpB than in ExpA, which is consistent with efforts to harmonize emissions 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 Textor et al., 2006). Fine aerosols (SO4, BC, and POM) are mainly removed by wet deposition (on average about 80–90% by mass in both experiments, see also Tables 1–6). 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. Larger changes occur for LOA, KYU, and ARQM since 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 the also the meteorological fields are equal in both experiments 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 1–6 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 SO4).

For the coarse aerosols (SS and DU), dry deposition is with about 70–80% of the removal mass fluxes the dominant process in both experiments (see Tables 1 and 2 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 50% to 20% for DU, and from 108% to 17% for SS. These findings indicate the influence of harmonized size distributions in ExpB on the dry removal rates. However, the split between the deposition pathways is still rather model-specific and less dependent on the change in the sources.

4 Conclusions

The important effects of aerosols on climate change and air quality in combination with the large uncertainty of the magnitude of these effects necessitates profound knowledge of the aerosol life cycle. The application of numerical models using high-quality inventories of aerosol precursor gas and primary aerosol emissions are required in order to evaluate coherent reduction strategies. However, current emission inventories are associated with large uncertainties. Usually they 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).

Recent model studies have investigated the effect of changing 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. 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. Our results indicate that the findings from such studies depend to a large extent on the individual model configuration. Therefore, we recommend to use an ensemble of models when assessing the impacts from emission changes, until robust quality measures become available.

In this paper, the effects of unified aerosol sources on the simulated aerosol fields has 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 diversity of aerosol sources among models strongly decreased, we realize that is it 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, intermediate versions of the emissions data sets, 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 dispersals and removal pathways are model-specific and less depending on the properties of the aerosol sources. This indicates 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) 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.

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, but we expect clarifications from sensitivity studies comparing tendencies of individual processes with constrains 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 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 host model. Therefore additional studies dedicated to specific processes are necessary, where several parameterizations for a specific process are tested within at least one global host model.

As it is a major goal of AeroCom to compare model simulations against measurements. Detailed evaluation studies against measurement 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.

Acknowledgements. This work was supported by the European Projects PHOENICS (Particles of Human Origin Extinguishing “natural” solar radiation In Climate Systems) and CREATE (Construction, use and delivery of an European aerosol database), and the French space agency CNES (Centre National des Etudes Spatiales). The authors would like to thank the Laboratoire des Sciences du Climat et de l’Environnement, Gif-sur-Yvette, France, and the Max-Planck-Institut für Meteorologie, Hamburg, Germany. The work of O. Boucher forms part of the Climate Prediction Programme of the UK Department for the Environment, Food and Rural Affairs (DEFRA) under contract PECD 7/12/37.

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Table 1. Statistics of models results for DU: emissions, burdens, mass fractions above 5 km height, mass fractions in polar regions (south of 80 S and north of 80 N), tropospheric residence times, split of removal pathways (mass fraction of wet removal in relation to total removal). Shown are the means, medians and the model diversities for all species in AeroCom 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
ResTime	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	%	8	1,5	1,1	1,0	0,8	109	102
WetofTot	%	7	34,9	30,8	36,6	30,3	43	47

Table 2. Statistics of models results for SS: emissions, burdens, mass fractions above 5 km height, mass fractions in polar regions (south of 80 S and north of 80 N), tropospheric residence times, split of removal pathways (mass fraction of wet removal in relation to total removal). Shown are the means, medians and the model diversities for all species in AeroCom experiments A and B.

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
ResTime	days	5	0,5	0,6	0,3	0,6	59	31
LoadAltF	%	6	9,4	9,2	4,5	2,8	111	128
LoadPolF	%	8	4,3	2,9	1,6	1,8	142	117
WetofTot	%	5	21,0	25,3	21,0	28,2	58	45

Table 3. Statistics of models results for SO4: emissions, burdens, mass fractions above 5 km height, mass fractions in polar regions (south of 80 S and north of 80 N), tropospheric residence times, split of removal pathways (mass fraction of wet removal in relation to total removal). Shown are the means, medians and the model diversities for all species in AeroCom experiments A and B.

SO4	unit	#	mean		median		Stdev	
			ExpA	ExpB	ExpA	ExpB	ExpA	ExpB
Emi	Tg/a	6	179,0	162,0	198,0	171,0	25	21
Load	Tg	8	2,1	2,0	2,0	2,0	27	26
Wet	Tg/a	7	156,0	141,0	171,0	149,0	25	23
SedDry	Tg/a	7	21,3	19,4	20,5	19,2	32	31
ResTime	days	7	4,2	4,4	4,1	4,4	20	18
LoadAltF	%	8	34,6	35,2	33,7	35,7	30	29
LoadPolF	%	8	6,4	5,8	6,8	6,2	43	46
WetofTot	%	7	87,5	87,5	88,5	87,4	5	5

Table 4. Statistics of models results for BC: emissions, burdens, mass fractions above 5 km height, mass fractions in polar regions (south of 80 S and north of 80 N), tropospheric residence times, split of removal pathways (mass fraction of wet removal in relation to total removal). Shown are the means, medians and the model diversities for all species in AeroCom experiments A and B.

BC	unit	#	mean		median		Stdev	
			ExpA	ExpB	ExpA	ExpB	ExpA	ExpB
Emi	Tg/a	8	11,7	7,8	11,3	7,8	9	1
Load	Tg	8	0,3	0,2	0,2	0,1	46	28
Wet	Tg/a	7	9,3	6,2	9,3	6,0	16	10
SedDry	Tg/a	7	2,5	1,6	2,6	1,8	31	34
ResTime	days	8	7,8	7,3	6,8	6,9	41	28
LoadAltF	%	8	22,4	24,1	18,3	19,4	50	49
LoadPolF	%	8	5,3	4,0	6,2	4,4	60	60
WetofTot	%	7	78,4	79,3	75,4	76,9	9	9

Table 5. Statistics of models results for POM: emissions, burdens, mass fractions above 5 km height, mass fractions in polar regions (south of 80 S and north of 80 N), tropospheric residence times, split of removal pathways (mass fraction of wet removal in relation to total removal). Shown are the means, medians and the model diversities for all species in AeroCom experiments A and B.

POM	unit	#	mean		median		Stdev	
			ExpA	ExpB	ExpA	ExpB	ExpA	ExpB
Emi	Tg/a	8	93,6	66,6	88,7	66,9	28	1
Load	Tg	8	1,7	1,2	1,6	1,2	23	20
Wet	Tg/a	7	79,2	53,0	74,5	52,7	32	10
SedDry	Tg/a	7	19,7	13,1	18,7	13,9	23	34
ResTime	days	8	7,0	6,7	7,0	6,4	30	21
LoadAltF	%	8	22,3	23,8	18,3	19,7	53	49
LoadPolF	%	8	3,7	3,2	4,6	3,4	62	61
WetofTot	%	7	78,8	80,1	77,4	79,1	9	9

Table 6. Statistics of models results for AER: emissions, burdens, mass fractions above 5 km height, mass fractions in polar regions (south of 80 S and north of 80 N), tropospheric residence times, split of removal pathways (mass fraction of wet removal in relation to total removal). Shown are the means, medians and the model diversities for all species in AeroCom experiments A and B.

AER	unit	#	mean		median		Stdev	
			ExpA	ExpB	ExpA	ExpB	ExpA	ExpB
Emi	Tg/a	5	10 100,0	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
ResTime	days	5	1,9	1,4	1,7	1,4	61	17
LoadAltF	%	6	15,3	14,5	12,4	11,2	54	62
LoadPolF	%	8	2,7	2,2	1,7	1,5	96	97
WetofTot	%	5	24,9	27,3	26,3	30,0	43	42

Table 7. Model average relative changes of parameters between ExpA and ExpB expressed as (ExpB-ExpA)/ExpA in [%] for emissions, load, residence time, and fraction of wet deposition in relation to total deposition.

	Emi [Tg/a]		Load [Tg]		ResTime [days]		WetofTot [%]	
	Mean	Median	Mean	Median	Mean	Median	Mean	Median
DUST	8.6	3.0	–1.7	–3.7	–7.7	–9.9	–11.3	–0.9
SS	76.6	95.7	88.2	64.4	26.6	4.4	37.5	20.0
SO4	–8.3	–7.4	–4.2	0.2	4.7	5.7	–0.0	0.1
BC	–33.2	–30.8	–34.9	–32.8	–2.5	–1.2	1.2	0.7
POM	–23.1	–18.5	–26.7	–29.3	–0.1	7.4	1.6	2.2
AER	37.8	60.9	8.1	3.1	–1.1	–35.9	18.4	5.9

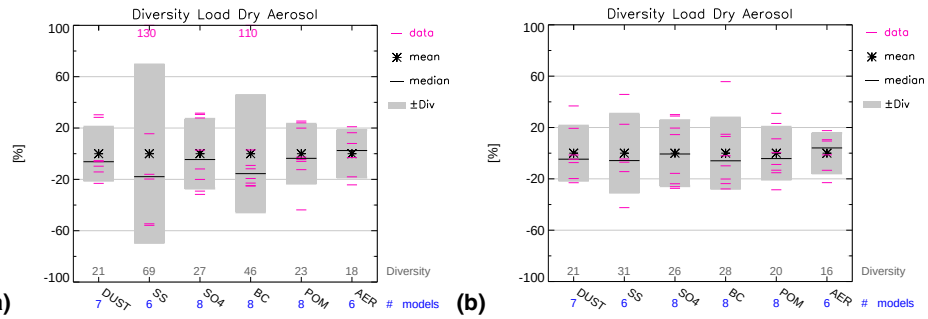

Fig. 1. Model diversities of the global, annual average aerosol burden of the five aerosol species in **(a)** ExpA and **(b)** ExpB. The diversities are indicated by gray boxes ("div"=normalized standard deviation). The individual models' deviations from the all-models-averages are plotted as pink lines ('data"), or as 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.

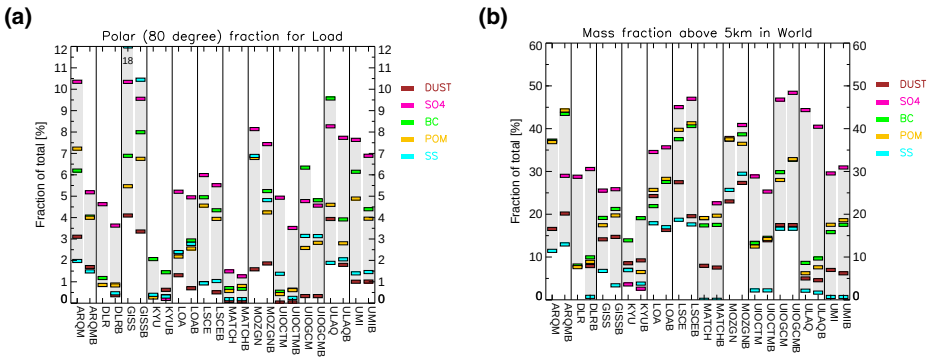

Fig. 2. **(a)** Global, annual average mass fractions in [%] of total mass in polar regions (south of 80 S and north of 80 N) for the AeroCom models. **(b)** Global, annual average mass fractions in [%] of total mass above 5 km altitude for the AeroCom models. The gray shadings frame the range for each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

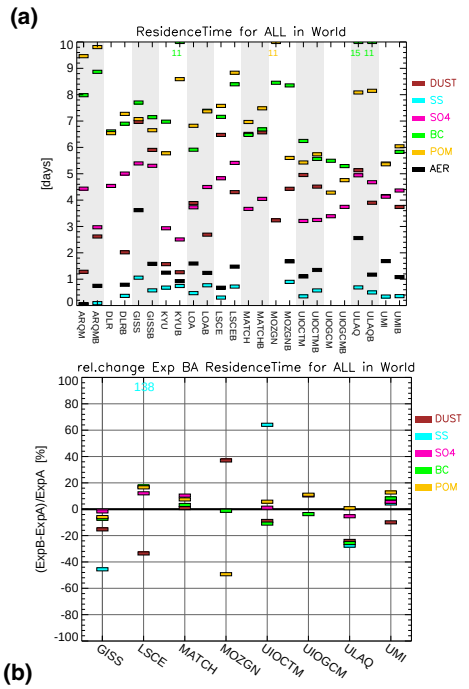

Fig. 3. Tropospheric residence times in ExpA and ExpB in [days], **(b)** Relative changes between ExpA and B expressed as (ExpB-ExpA)/ExpA in [%].

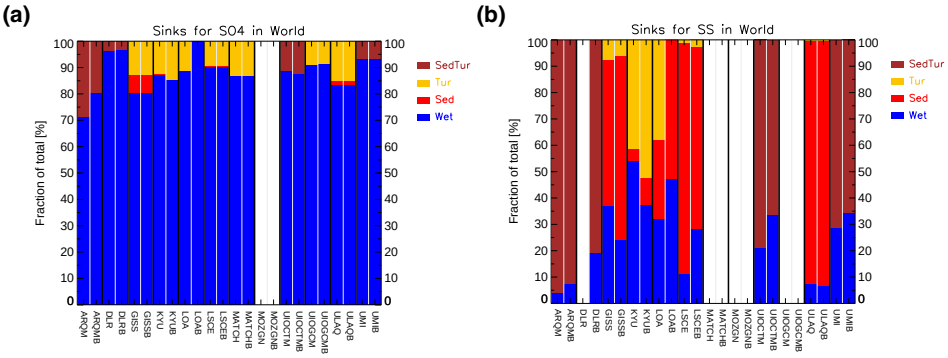

Fig. 4. Contribution of the individual removal processes to the total sink mass flux (annually and globally averaged) for the AeroCom models for **(a)** SO4 and **(b)** SS. The color code is given in the legend. Wet refers to wet deposition. If possible we show the individual dry sink rate coefficients (Tur: turbulent deposition, and Sed: sedimentation), otherwise the sum of the two processes (Dry=SedTur) is plotted.