Aerosol optical depths and direct radiative perturbations by species and source type

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[1] We have used the Laboratoire de Météorologie Dynamique General Circulation Model (LMDZT GCM) to estimate the relative contributions of different aerosol source types (i.e., fossil fuels, biomass burning, and "natural") and aerosol species to the aerosol optical depth (AOD) and direct aerosol radiative perturbation (DARP) at the top-of-atmosphere. The largest estimated contribution to the global annual average AOD (0.12 at 550 nm) is from natural (58%), followed by fossil fuel (26%), and biomass burning (16%) sources. The global annual mean all-sky DARP in the shortwave (SW) spectrum by sulfate, black carbon (BC), organic matter (OM), dust, and sea salt are -0.62, $+0.55$, -0.33, -0.28, and -0.30 Wm^{-2}, respectively. The all-sky DARP in the longwave spectrum (LW) is not negligible and is a bit less than half of the SW DARP. The net (i.e., SW+LW) DARP distribution is predominantly negative with patches of positive values over the dust source regions, and off the west coasts of Southern Africa and South and North America. For dust aerosols the SW effect is partially offset by LW greenhouse effect. **Citation:** Reddy, M. S., O. Boucher, Y. Balkanski, and M. Schulz (2005), Aerosol optical depths and direct radiative perturbations by species and source type, *Geophys. Res. Lett.*, *32*, L12803, doi:10.1029/2004GL021743.

1. Introduction

[2] Atmospheric aerosols and greenhouse gases significantly alter the Earth's radiation budget. Well-mixed greenhouse gases of anthropogenic origin exert a positive radiative forcing (RF) of 2.4 Wm^{-2} in 2000 compared to pre-industrial times, whereas anthropogenic aerosols are estimated to exert a negative but highly uncertain RF [*Ramaswamy et al.*, 2001]. Rather than RF which strictly speaking applies only to external perturbations, we focus here on the direct aerosol radiative perturbation (DARP) which extends the concept of RF to any aerosol type (i.e., natural and anthropogenic). We only consider the DARP at the top of atmosphere (TOA). Depending upon their chemical nature aerosols can exert a positive (e.g., black carbon (BC)) or negative DARP (e.g., sulfate and sea salt). Organic matter (OM) and mineral dust exert either a positive or a negative DARP depending on particle size, chemical composition, and surface albedo.

[3] In the last decade significant progress has been made in the understanding of the aerosol effects on radiation. A number of global models have been used to study the multi-component aerosol with various degrees of complexity [e.g., *Jacobson*, 2001; *Takemura et al.*, 2002; *Liao et al.*, 2004; *Reddy et al.*, 2005]. Some of these studies are limited to estimating the distributions of aerosol concentrations and aerosol optical depths (AODs), and only a few of them also report DARPs by aerosol species. Except for sulfate [*Haywood et al.*, 1997] and dust [e.g., *Woodward*, 2001; *Liao et al.*, 2004] aerosols, DARPs in the longwave (LW) spectrum are usually not reported.

[4] We have recently presented a global study of the multi-component aerosol in the Laboratoire de Météorologie Dynamique General Circulation Model (LMDZT GCM) [*Reddy et al.*, 2005]. Here we extend this work to estimate the relative contributions of various aerosol sources –fossil fuel, biomass burning, and natural– to AODs and shortwave (SW) and LW DARPs by species, namely sulfate, BC, OM, dust, and sea salt. We also examine the differences in clear-sky versus all-sky DARP.

2. Method

[5] The LMDZT GCM is a grid-point model with a resolution of 3.75° in longitude and 2.5° in latitude with 19 vertical layers. The current version includes sulfate, BC, OM, dust, and sea salt. This model has been thoroughly described and evaluated [*Boucher et al.*, 2002; *Boucher and Pham*, 2002; *Reddy and Boucher*, 2004; *Reddy et al.*, 2005]. The present model setup and emissions are exactly the same as in *Reddy et al.* [2005] and those used for the Aerosol Intercomparison Project (http://nansen.ipsl.jussieu.fr/AEROCOM).

[6] Simulations were carried out by nudging the horizontal model winds to 6 hourly winds from ECMWF analyzes with a relaxation time of 0.1 days. This ensures that the model transport is reasonably constrained by ECMWF meteorology while other dynamical and physical processes are driven by the model parameterizations. Simulations are done using the wind fields for the year 2000 after allowing for two months of spin up.

[7] We have conducted three different experiments. In each experiment we switch on only one aerosol source type

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Table 1. Globally and Annually Averaged AOD at 550 nm (×100) by Aerosol and Source Type

Source →	Fossil Fuels			Biomass			Natural			All sources[a]		
Aerosols ↓	NH	SH	Global	NH	SH	Global	NH	SH	Global	NH	SH	Global
Sulfate	5.08	0.65	2.87	0.13	0.10	0.11	1.17	1.32	1.24	6.38	2.07	4.22
BC	0.24	0.03	0.13	0.28	0.19	0.24	–	–	–	0.52	0.22	0.37
OM	0.34	0.02	0.18	1.81	1.29	1.55	0.40	0.35	0.37	2.55	1.66	2.10
Dust	–	–	–	–	–	–	5.09	0.18	2.63	5.09	0.18	2.63
Sea Salt	–	–	–	–	–	–	1.87	3.62	2.74	1.87	3.62	2.74
(acc. mode)	–	–	–	–	–	–	0.90	1.59	1.24	0.90	1.59	1.24
(coarse mode)	–	–	–	–	–	–	0.97	2.03	1.50	0.97	2.03	1.50
All	5.66	0.70	3.18	2.22	1.58	1.90	8.53	5.47	6.98	16.41	7.75	12.06

[a]The "all sources" columns are obtained as the sum of AODs in the three experiments.

among fossil fuel combustion, biomass burning, and natural sources. Note that some aerosol species stem from all three source types. We include natural organic matter from terpene emissions. However, we miss biomass burning emissions from boreal forest fires. Dust and sea salt are assumed to be natural only. The water associated with sulfate, OM, and sea-salt aerosols contributes to the AOD separately for each aerosol species and each aerosol source. Within each experiment, the DARP is estimated at the TOA in the SW and LW spectrums for each aerosol species separately, by calling the radiation routine with and without the presence of each aerosol type (assuming an external mixture). There is no feedback of the aerosols on the simulated meteorology.

3. Results and Discussion

3.1. Aerosol Optical Depth

[8] The simulated AODs (at 550 nm) were compared to all available AERONET measurements for the period 2000 and 2001. On a monthly mean basis the correlation coefficient was 0.57 (N = 1324) with a bias towards smaller values, especially over biomass burning regions. Moreover 76% of data points fall within a factor of 2 deviation [*Reddy et al.*, 2005]. The largest contribution to the globally and annually averaged AOD is from natural sources (58%) followed by fossil-fuel (26%), and biomass-burning (16%) (Table 1). Within the source from fossil fuel combustion sulfate accounts for 90% of the AOD and concentrates over North America, Europe, and East Asia (Figure 1). As expected, biomass burning is the major source of carbonaceous aerosols. The biomass burning sources are localized in Africa and South America. Natural aerosol sources are dominant all over the oceans and dust source regions which are located over North Africa, West Asia, and Northeast China. Natural aerosols are dominant over both hemispheres, with contributions to the total AOD of 52 and 71% in the NH and SH, respectively. The natural sources are mostly of oceanic origin in the SH with sea salt and DMS, and mostly of continental origin in the NH with mineral dust. Our three different source types have very different latitudinal distributions (not shown). Fossil fuel sources are dominant over the NH and account for 30–55% of AOD in the latitudinal band of 15–60°N. Biomass burning sources account for 30–40% of the total AOD between 30°S and 10°N.

3.2. Direct Aerosol Radiative Perturbation

[9] The DARPs by individual aerosol types are estimated in all three experiments for clear- and all-sky conditions (Table 2 and Figure 2). Assuming an external mixture we also sum up the DARPs by different aerosol types to estimate the total DARP. The clear-sky SW DARP is negative for all aerosol types except for BC. The present estimate is a lower bound for BC because of the assumption of an external mixture. Assuming an internal mixture would at most double this estimate [*Jacobson*, 2001]. Note also the more negative SW DARP by dust compared to previous global modeling studies [*Takemura et al.*, 2002; *Liao et al.*, 2004] due to less absorbing dust optical properties [*Haywood et al.*, 2003; *Reddy et al.*, 2005]. The present global annual mean clear-sky SW DARP by sea salt comes very close to the estimate by *Takemura et al.* [2002] but lower than that by *Liao et al.* [2004].

[10] Natural sources with mostly scattering aerosols play a more important role while biomass burning sources with a significant amount of absorbing aerosols play a less important role. The global annual average clear-sky SW DARP from fossil fuel aerosols is negative (-0.56 Wm^{-2}), with a NH average about 10 times larger than the SH average. The global annual average clear-sky SW DARP from biomass burning sources is close to zero (-0.08 Wm^{-2}), with about an equal negative and positive contribution by OM and BC, respectively. The global annual average clear-sky SW DARP from natural sources is negative at -1.41 Wm^{-2}. The ratio of all-sky to clear-sky SW DARP varies with

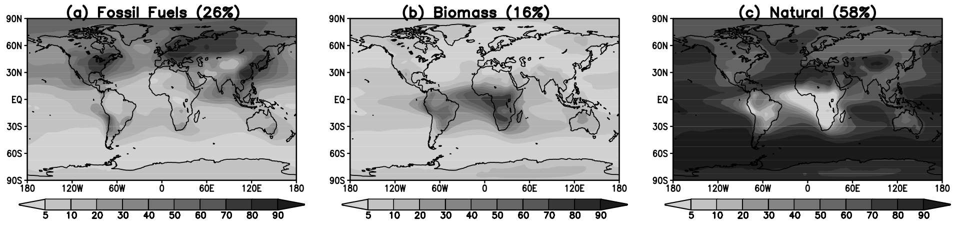

Figure 1. Relative contributions (%) from (a) fossil-fuel, (b) biomass-burning, and (c) natural sources to the annually averaged total AOD (at 550 nm). See color version of this figure in the HTML.

Table 2. Globally and Annually Averaged SW and LW DARP at TOA (Wm^{-2}) by Aerosol and Source Type for Clear- and All-Sky Conditions

Sources →	Fossil Fuels			Biomass			Natural			All sources[a]		
Aerosols ↓	NH	SH	Global	NH	SH	Global	NH	SH	Global	NH	SH	Global
						Shortwave: Clear-sky						
Sulfate	−1.26	−0.13	−0.69	−0.02	−0.02	−0.02	−0.29	−0.29	−0.29	−1.57	−0.44	−1.00
BC	+0.31	+0.03	+0.17	+0.36	+0.23	+0.29	–	–	–	+0.67	+0.26	+0.46
OM	−0.07	−0.00	−0.04	−0.41	−0.30	−0.35	−0.10	−0.08	−0.09	−0.58	−0.38	−0.48
Dust	–	–	–	–	–	–	−0.78	−0.02	−0.40	−0.78	−0.01	−0.40
Sea Salt	–	–	–	–	–	–	−0.39	−0.88	−0.63	−0.39	−0.88	−0.63
All	*−1.02*	*−0.10*	*−0.56*	*−0.07*	*−0.09*	*−0.08*	*−1.56*	*−1.27*	*−1.41*	*−2.66*	*−1.45*	*−2.05*
						Shortwave: All-sky						
Sulfate	−0.78	−0.08	−0.43	−0.02	−0.01	−0.01	−0.18	−0.17	−0.18	−0.98	−0.26	−0.62
BC	+0.34	+0.04	+0.19	+0.40	+0.32	+0.36	–	–	–	+0.73	+0.36	+0.55
OM	−0.04	−0.00	−0.02	−0.32	−0.18	−0.25	−0.06	−0.05	−0.06	−0.42	−0.23	−0.33
Dust	–	–	–	–	–	–	−0.55	−0.01	−0.28	−0.55	−0.01	−0.28
Sea salt	–	–	–	–	–	–	−0.21	−0.40	−0.30	−0.21	−0.40	−0.30
All	*−0.48*	*−0.04*	*−0.26*	*+0.06*	*+0.13*	*+0.10*	*−1.00*	*−0.63*	*−0.82*	*−1.43*	*−0.54*	*−0.98*
						Longwave: Clear-sky						
Sulfate	+0.078	+0.014	+0.046	+0.003	+0.002	+0.002	+0.027	+0.027	+0.027	+0.107	+0.043	+0.075
BC	+0.005	+0.001	+0.003	+0.007	+0.005	+0.006	–	–	–	+0.012	+0.005	+0.009
OM	+0.005	+0.000	+0.003	+0.041	+0.029	+0.035	+0.011	+0.010	+0.010	+0.057	+0.039	+0.048
Dust	–	–	–	–	–	–	+0.368	+0.018	+0.193	+0.368	+0.018	+0.193
Sea salt	–	–	–	–	–	–	+0.178	+0.240	+0.209	+0.178	+0.240	+0.209
All	*+0.088*	*+0.015*	*+0.052*	*+0.051*	*+0.036*	*+0.043*	*+0.584*	*+0.295*	*+0.439*	*+0.722*	*+0.345*	*+0.534*
						Longwave: All-sky						
Sulfate	+0.047	+0.009	+0.028	+0.002	+0.001	+0.002	+0.018	+0.018	+0.018	+0.067	+0.028	+0.048
BC	+0.003	+0.001	+0.002	+0.005	+0.003	+0.004	–	–	–	+0.008	+0.004	+0.006
OM	+0.003	+0.000	+0.002	+0.029	+0.019	+0.024	+0.007	+0.007	+0.007	+0.039	+0.026	+0.033
Dust	–	–	–	–	–	–	+0.262	+0.015	+0.138	+0.262	+0.015	+0.138
Sea salt	–	–	–	–	–	–	+0.106	+0.127	+0.117	+0.106	+0.127	+0.117
All	*+0.053*	*+0.010*	*+0.032*	*+0.036*	*+0.023*	*+0.030*	*+0.393*	*+0.167*	*+0.280*	*+0.482*	*+0.200*	*+0.342*

[a]The "all sources" columns are obtained as the sum of AODs in the three experiments.

aerosol and source types. This ratio is in the range of 42 to 69% for aerosols which are mostly scattering. This spread in values originates from the fact that certain aerosol types (dust, biomass burning) are located predominantly in regions with small cloud cover while others (sea salt, natural, and anthropogenic sulfate) are located predominantly in regions with large cloud cover. The all-sky SW DARP by BC is larger than its clear-sky counterpart (ratio of 120%).

[11] The present estimate of all-sky SW DARP by sulfate lies in the range of IPCC reported values (−0.26 to −0.81 Wm^{-2}) [*Ramaswamy et al.*, 2001]. The all-sky SW DARP by biomass burning sources differs completely from that by fossil fuels in terms of sign and distribution. North and parts of South Africa experiences positive SW DARP of about +2 Wm^{-2} (Figure 2). The positive all-sky SW DARP in biomass burning regions is corroborated by recent estimates made from observed fields of aerosols and clouds off the coast of Namibia [*Keil and Haywood*, 2003] and differs from previous estimates [*Ramaswamy et al.*, 2001]. This probably results from the relative high altitude of emissions in our model, whereby BC may overly cloudy layers. This also results in all-sky SW DARP of +0.55 Wm^{-2} by BC, which is at the upper end of previously reported values for an external mixture. This points out the importance of the representation of the height profile of biomass burning emissions.

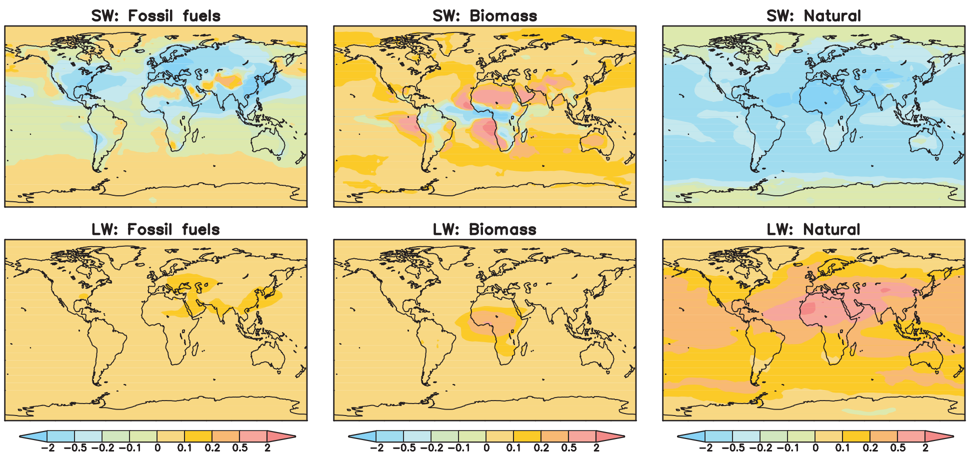

Figure 2. Annually averaged all-sky DARP at TOA (Wm^{-2}) by source type in the (top) SW and (bottom) LW spectrum.

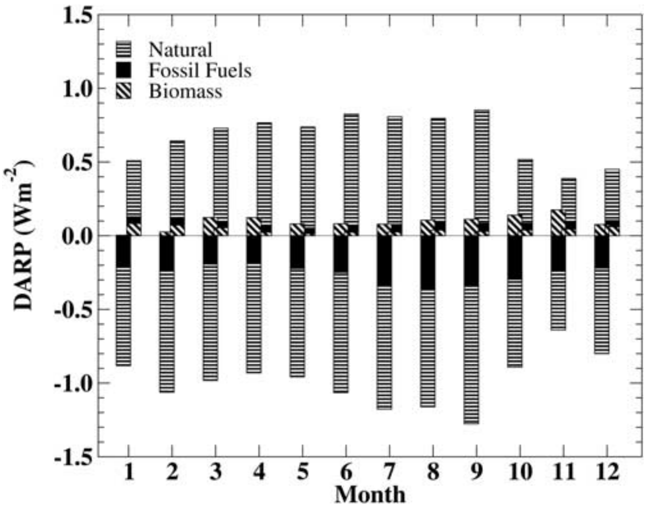

Figure 3. Globally averaged monthly SW (left bar) and LW (right bar) DARP at TOA by source type in all-sky conditions. See color version of this figure in the HTML.

[12] The LW DARP is dominated from natural sources, with the largest contribution in all-sky conditions from dust (40%) followed by sea salt (34%), sulfate (14%), OM (10%), and BC (2%). The LW DARP has values larger than +2 and +0.5 Wm^{-2} over dust and sea salt dominated regions, respectively (Figure 2). The global annual-mean all-sky LW DARP by anthropogenic sulfate (+0.03 Wm^{-2}) is about 3 times larger than that reported by *Haywood et al.* [1997]. Our all-sky LW DARP by dust (+0.14 Wm^{-2}) is at the lower end of reported values (+0.23 to +0.31 Wm^{-2}) [*Woodward*, 2001; *Liao et al.*, 2004]. Our all-sky net (SW + LW) DARP by dust is slightly negative (−0.14 Wm^{-2}) in contrast to previous estimates which reported slightly positive values (+0.07 to +0.14 Wm^{-2}) [*Tegen et al.*, 1996; *Woodward*, 2001; *Liao et al.*, 2004]. The estimated total LW DARP is not negligible and is about one third of SW DARP with an opposite sign.

[13] The global mean seasonal cycle in all-sky SW DARP is mainly controlled by natural sources (Figure 3). During June-September enhanced dust activity, in addition to increased biomass burning, and larger insolation in the NH, augment the SW DARP by 20 to 33% as compared to the annual mean. The all-sky LW DARP shows little seasonality. The all-sky net DARP from fossil fuel and natural sources is negative throughout the year while that from biomass burning sources is always positive.

4. Conclusion

[14] The relative contributions of fossil fuel, biomass burning, and natural sources to AODs have been estimated in the LMDZT GCM. The SW and LW DARPs for individual aerosols (i.e., sulfate, BC, OM, dust, and sea salt) and from different source types have also been estimated. The LW DARP is not negligible and is about one third of the negative SW DARP. As a result it should not be neglected in climate simulations, especially if dust and sea-salt emissions increase during this century as a result of climate change. The net all-sky DARP is predominantly negative with patches of positive values over the dust source regions and/or over bright surfaces. It is also positive off the west coast of South Africa, South and North America. The net global annual mean all-sky DARPs by biomass burning and dust are small but imply a redistribution of the energy in the climate system at the regional scale. In particular the effect of biomass burning aerosols is to heat the atmosphere and to cool the surface. While the present study of DARP by different source types can be useful for evaluating emission control policies, it is now also required to estimate the breakdown by different economic sectors.

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References

Boucher, O., and M. Pham (2002), History of sulfate aerosol radiative forcings, *Geophys. Res. Lett.*, *29*(9), 1308, doi:10.1029/2001GL014048.

Boucher, O., et al. (2002), Simulation of the atmospheric sulfur cycle in the Laboratoire de Météorologie Dynamique general circulation model: Model description, model evaluation, and global and European budgets, *Note Sci. de l'IPSL 23*, 27 pp., Inst. Pierre-Simon Laplace, Paris.

Haywood, J. M., et al. (1997), General circulation model calculations of the direct radiative forcing by anthropogenic sulphate and fossil-fuel soot aerosol, *J. Clim.*, *10*, 1562–1577.

Haywood, J., P. Francis, O. Dubovik, M. Glew, and B. Holben (2003), Comparison of aerosol size distributions, radiative properties, and optical depths determined by aircraft observations and Sun photometers during SAFARI 2000, *J. Geophys. Res.*, *108*(D13), 8471, doi:10.1029/2002JD002250.

Jacobson, M. Z. (2001), Global direct radiative forcing due to multicomponent anthropogenic and natural aerosols, *J. Geophys. Res.*, *106*, 1551–1568.

Keil, A., and J. M. Haywood (2003), Solar radiative forcing by biomass burning aerosol particles during SAFARI 2000: A case study based on measured aerosol and cloud properties, *J. Geophys. Res.*, *108*(D13), 8467, doi:10.1029/2002JD002315.

Liao, H., J. H. Seinfeld, P. J. Adams, and L. J. Mickley (2004), Global radiative forcing of coupled tropospheric ozone and aerosols in a unified general circulation model, *J. Geophys. Res.*, *109*, D16207, doi:10.1029/2003JD004456.

Ramaswamy, V., et al. (2001), Radiative forcing of climate change, in *Climate Change 2001: The Scientific Basis: Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change*, edited by J. T. Houghton et al., pp. 349–416, Cambridge Univ. Press, New York.

Reddy, M. S., and O. Boucher (2004), A study of the global cycle of carbonaceous aerosols in the LMDZT general circulation model, *J. Geophys. Res.*, *109*, D14202, doi:10.1029/2003JD004048.

Reddy, M. S., O. Boucher, N. Bellouin, M. Schulz, Y. Balkanski, J.-L. Dufresne, and M. Pham (2005), Estimates of global multicomponent aerosol optical depth and direct radiative perturbation in the Laboratoire de Météorologie Dynamique general circulation model, *J. Geophys. Res.*, *110*, D10S16, doi:10.1029/2004JD004757.

Takemura, T., et al. (2002), Single-scattering albedo and radiative forcing of various aerosol species with a global three-dimensional model, *J. Clim.*, *15*, 333–352.

Tegen, I., et al. (1996), The influence of mineral aerosols from disturbed soils on climate forcing, *Nature*, *380*, 419–422.

Woodward, S. (2001), Modeling the atmospheric life cycle and radiative impact of mineral dust in the Hadley Centre climate model, *J. Geophys. Res.*, *106*, 18,155–18,166.

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