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Contrasts in the effects on climate of anthropogenic sulfate aerosols between the 20th and the 21st century

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[1] In this study, we examine the time evolution of the relative contribution of sulfate aerosols and greenhouse gases to anthropogenic climate change. We use the new IPSL-CM4 coupled climate model for which the first indirect effect of sulfate aerosols has been calibrated using POLDER satellite data. For the recent historical period the sulfate aerosols play a key role on the temperature increase with a cooling effect of 0.5 K, to be compared to the 1.4 K warming due to greenhouse gas increase. In contrast, the projected temperature change for the 21st century is remarkably independent of the effects of anthropogenic sulfate aerosols for the SRES-A2 scenario. Those results are interpreted comparing the different radiative forcings, and can be extended to other scenarios. We also highlight that the first indirect effect of aerosol strongly depends on the land surface model by changing the cloud cover. **Citation:** Dufresne, J.-L., J. Quaas, O. Boucher, S. Denvil, and L. Fairhead (2005), Contrasts in the effects on climate of anthropogenic sulfate aerosols between the 20th and the 21st century, *Geophys. Res. Lett.*, *32*, L21703, doi:10.1029/2005GL023619.

1. Introduction

[2] Sulfate aerosols are currently the second most important anthropogenic forcing for the climate after the greenhouse gases (GHG) [*Ramaswamy et al.*, 2001]. They reflect solar radiation (direct effect) and they modify cloud radiative properties (indirect effects). They have a strong cooling effect that has partly canceled the climate warming due to GHG increase during the 20th century. Nevertheless the residence time of sulfate aerosol in the atmosphere is a few days whereas the residence time of GHG such as CO_2 is a few decades. Moreover, the indirect effect of sulfate aerosols is a very non-linear function of aerosol burden [e.g., *Boucher and Pham*, 2002]. Therefore the relative importance of sulfate aerosols and GHG on climate change may vary with time. Here we explore this question by performing simulations with a climate model, some of which differ only by their sulfate aerosol concentrations.

2. Model Description

[3] The climate model used for the simulations is the newly developed IPSL-CM4 model [*Marti et al.*, 2005] that has been used to perform climate change simulations for the IPCC fourth assessment report. Its components are LMDZ-4 (F. Hourdin et al., The LMDZ4 general circulation model: Climate performance and sensitivity to parametrized physics with emphasis on tropical convection, submitted to *Climate Dynamics*, 2005, hereinafter referred to as Hourdin et al., submitted manuscript, 2005) for the atmosphere, ORCA [*Madec et al.*, 1998] for the ocean, ORCHIDEE [*Krinner et al.*, 2005] for the land surface, LIM [*Fichefet and Maqueda*, 1997] for the sea-ice and OASIS [*Valcke et al.*, 2004] for the coupling.

[4] The direct and indirect effect of sulfate aerosols are parameterized as by *Boucher and Pham* [2002] (hereinafter referred to as BP02), *Pham et al.* [2005] (hereinafter referred to as PBH05) and *Quaas et al.* [2004b]. Only the effect of aerosol concentration on cloud droplet radius not affecting cloud liquid water content (called first indirect effect [*Twomey*, 1974]) is considered. For this effect, a couple of observational-based constraints exist and can be used to evaluate model parameterization [e.g., *Lohmann and Lesins*, 2002]. For the effect of cloud droplet radius change on precipitation and cloud lifetime (second aerosol indirect effect [*Albrecht*, 1989]), however, no such constraints exist yet, so that it is so far very difficult to validate such parameterizations at global scale [e.g., *Rotstayn and Liu*, 2005].

[5] A key point of our parameterization is the relationship between the cloud droplet number concentration N_d (in droplets per cm^{-3}) and the aerosol mass concentration m_a (in μgm^{-3}). We make use of the empirical parameterization of *Boucher and Lohmann* [1995]:

$$N_d = 10^{a_0 + a_1 log(m_a)}. \quad (1)$$

To avoid unrealistic droplet number concentrations, N_d is restricted to be within [20, 1000] droplets per cm^{-3}. In previous versions of LMDZ, the original values of the empirical constants $a_0 = 2.21$ and $a_1 = 0.41$ (formula "D" of *Boucher and Lohmann* [1995]) were used and *Quaas et al.* [2004a] have evaluated the direct and first indirect effect of sulfate aerosols using the POLDER satellite data [*Buriez et al.*, 1997; *Deuzé et al.*, 1999]. *Quaas et al.* [2004a] found a decrease of cloud top droplet effective radius as aerosol index increases in both model results and POLDER observations, but the decrease was too strong in the model. Moreover, the mean droplet radius was too small in the model. Using the same diagnostics, we adjust the two constants a_0 and a_1 in order to fit better the observations

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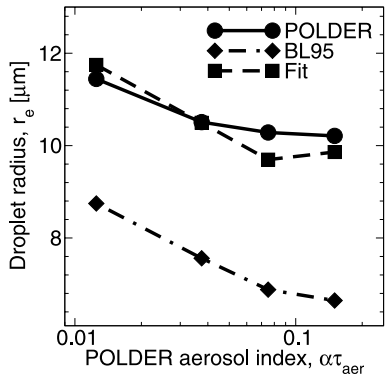

Figure 1. Relationship between cloud droplet effective radius (μm) and aerosol index for POLDER observations (line) and for the LMDZ model with the original [*Boucher and Lohmann*, 1995] "D" formula (dash-dot) and with the adjusted formula (dash).

(Figure 1) and we obtain $a_0 = 1.7$ and $a_1 = 0.2$. It should be noted, that this estimate value of a_1 could further be reduced when taking into account not only sulfate, but also other potential cloud condensation nuclei like sea salt or carbonaceous aerosols [*Quaas and Boucher*, 2005].

3. Climate Response for the 20th and 21st Century

[6] The runs presented here have been performed after a spin-up run of 330 years and in parallel to a 500 years control run (see auxiliary material[1] for details). Two runs have been performed over the period 1860–2100. In the first one, both the greenhouse gases and the sulfate aerosol concentrations vary with time. In the second run, the greenhouse gas concentration varies with time whereas the sulfate aerosol concentration is held fixed to its pre-industrial value (see auxiliary material for details). For the period 2000–2100 the IPCC SRES-A2 scenario has been adopted. We make use of the monthly mean 3D distributions of sulfate aerosols pre-computed by BP02 for the pre-industrial period and for the 20th century, and by PBH05 for the 21st century. Natural forcing are not present in the forcings as considering the volcanic eruptions for the 20th century but not for the 21st century would have lead to an artificial bias.

[7] For the period from 1860 to 2000, the model predicts an increase of the global mean 2m air temperature (T2m) that reaches 1.4 K when only the GHG increase is considered, and that is reduced to 0.9 K when sulfate aerosols increase is also considered (Figure 2; see also Table S1). This latter temperature increase is 0.2 K higher than the observed value, and this difference may be explained by various reasons: natural forcings (such as volcanic aerosols or change in solar activity) being neglected, a too high climate sensitivity or a too low aerosol first indirect effect, absence of a second aerosol indirect effect,...

[8] For the 21st century and for the SRES-A2 scenario, the model gives a temperature increase of 3.5 K in both cases whether anthropogenic sulfate aerosols are considered or not (Figure 2). This figure highlights that the sulfate aerosol change has an important impact on the temperature increase during the 20th century, but a negligible impact (compared to the impact of GHG increase) during the 21st century for this scenario.

[9] We will now consider directly the anomaly of the surface air temperature due to sulfate aerosol change over the whole period 1860–2100. The anomaly of T2m increases (in absolute value) from 1860 to around 2020, then remains almost constant within the range of natural variability (Figure 3 (top)). This cooling is mainly located over the Northern hemisphere continents where the sulfate aerosol sources are the most important and it ranges from −1 to −2 K (Figure 3 (bottom)). The cooling slowly spreads out over the whole globe and becomes almost uniform when the aerosol concentration decreases in the late 21st century.

4. Radiative Forcing

[10] We compare here the aerosol radiative forcings (see auxiliary material) with previously published results, in particular those obtained by BP02 and PBH05 with previous versions of the LMDZ model. In our simulations, the radiative forcing change of sulfate aerosol in 1995 relative to 1860 are −0.72, −0.50 and −0.22 W.m^{-2} for the total, direct and first indirect effect, respectively (Table 1). The direct effect is close, but somewhat higher in absolute value (0.1 W.m^{-2}), than the value reported by BP02 and is within the range proposed by *Ramaswamy et al.* [2001]. In contrast, the indirect effect in our simulation is four times lower than the value reported by BP02 (−0.2 W.m^{-2} instead of −0.8 W.m^{-2}), and is in the lower part of the range proposed by *Ramaswamy et al.* [2001] (0 to

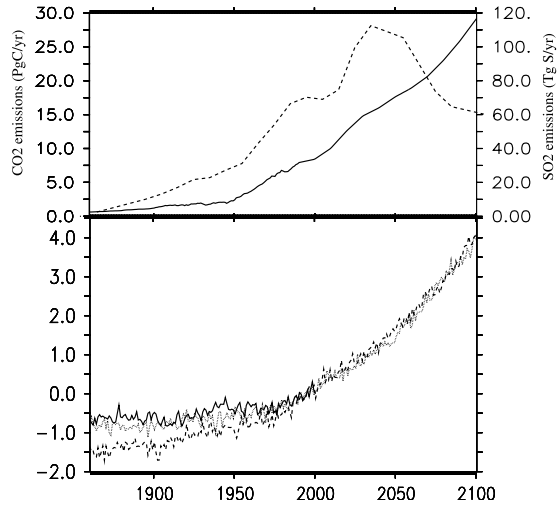

Figure 2. (top) Time evolution of the CO_2 (solid, left axis) and the sulfate emissions (dashed, right axis). (bottom) Time evolution of the 2m air surface temperature anomaly (in K) for observations (solid), for the run with time evolving sulfate aerosols (dot), for the run with pre-industrial sulfate aerosols (dash). The anomalies are computed relative to the 1990–2000 mean. For the 21st century, the SRES-A2 scenario is used.

[1]Auxiliary material is available at ftp://ftp.agu.org/apend/gl/2005GL023619.

Table 1. Radiative Forcing (W.m^{-2}) Relative to 1860 of the Well Mixed Greenhouse Gases (GHG) and as Provided by *Ramaswamy et al.* [2001], and of the Sulfate Aerosols Through Their Direct (ADE), First Indirect (AIE) and Total Effects as Computed by the IPSL-CM4 Model and as Previously Published by BP02 and PBH05

			Sulfate Aerosols					
			IPSL-CM4			BP02 and PBH05		
Year	Scenario	GHG	ADE	AIE	Total	ADE	AIE	Total
1945	-	0.65	−0.25	−0.12	−0.37	−0.13	−0.43	−0.56
1995	-	2.20	−0.50	−0.22	−0.72	−0.41	−0.83	−1.24
2045	A2	4.63	−0.97	−0.27	−1.24	−0.76	−1.08	−1.84
	A1B	4.57	−0.67	−0.21	−0.87	−0.51	−0.88	−1.39
	B1	3.91	−0.50	−0.22	−0.92	−0.61	−0.95	−1.56
2095	A2	7.85	−0.56	−0.15	−0.72	−0.41	−0.83	−1.24
	A1B	6.21	−0.24	−0.09	−0.33	−0.18	−0.58	−0.76
	B1	4.49	−0.22	−0.09	−0.31	−0.16	−0.53	−0.69

−1.5 W.m^{-2}). Sensitivity runs allow us to identify the two main sources of this important difference with BP02: (i) the adjustment of the parameters of equation (1) and (ii) the replacement of the simple bucket model used as the land surface model in the previous version of LMDZ by the more complex ORCHIDEE model. The bucket model leads to a larger cloud cover over continents in the mid latitudes, that increases the impact of the first indirect effect of sulfate aerosols. These two modifications almost equally contribute to the difference with BP02.

[11] The radiative forcing changes over three 50 years periods are displayed (Figure 4) for our simulations and as computed by BP02 and PBH05. This gives us an example of radiative forcing changes with two models that have a very different aerosol indirect effect. For the period 1945–1995, the ratio between the GHG forcing change and the sulfate aerosol forcing change is about −30% (−50% for BP02). During the first half of the 21st century, the GHG radiative forcing increases faster than during the second half of the 20th century, for all scenarios. In contrast, the aerosol forcing remains constant or decreases in absolute value over the same period. Moreover the difference between our results and BP02 and PBH05 decreases. During the second half of the 21st century, sulfate aerosols play an even more different role. The change of their radiative forcing is positive for all the three scenarios. The absolute value of the forcing decreases during this period owing to a decrease of sulfate emissions. This forcing being negative, the sulfate emission decrease leads to a positive radiative forcing change.

5. Summary and Conclusion

[12] The adjustment of the first indirect effect in the LMDZ model to POLDER data leads to a reduction of the sulfate aerosol indirect effect radiative forcing by a factor of 2 compared to a simulation using the original parameters proposed by *Boucher and Lohmann* [1995]. Compared to a previous version of LMDZ, the indirect effect has been reduced by another factor of 2 by changing the land surface model that leads to a decrease of evaporation and of the cloud cover over continents (Hourdin et al., submitted manuscript, 2005).

[13] For the 20th century, the model produces a temperature increase that is consistent with observations when sulfate aerosols are considered, and a temperature increase that is much higher when they are neglected. On the contrary, the model produces the same temperature increase during the 21st century and under the SRES-A2 scenario if sulfate aerosols are considered or not. This feature can be explained by considering the radiative forcing change during the 21st century and is not expected to be strongly modified if one considers a stronger indirect effect with a higher radiative forcing such as the one computed by PBH05.

[14] The impact of anthropogenic sulfur emissions on climate is very different during the three 50 year periods considered here (1945–1995, 1995–2045, 2045–2095). During the first period (1945–1995), their cooling effect attenuates the temperature increase due to the GHG increase. During the second period (1995–2045), their effect becomes small compared to the GHG increase and finally during the third period (2045–2095) the decrease of sulfate

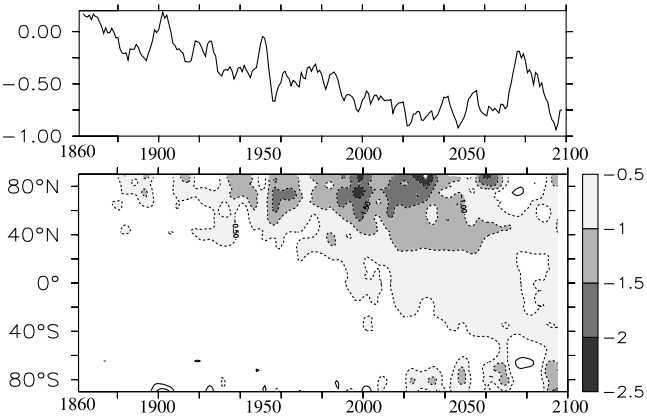

Figure 3. Time evolution of the 2m air temperature anomaly (K) due to sulfate aerosols increase (i.e., difference between the simulation with and without anthropogenic sulfate aerosols). (top) Global mean anomaly. (bottom) Zonal mean anomaly.

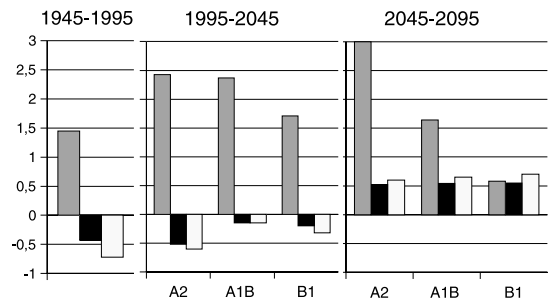

Figure 4. Radiative forcing (grey: GHG; dark: sulfate aerosols computed by the IPSL model; white: sulfate aerosols computed by BP02 or PBH05) change over three 50 year periods: 1945–1995, 1995–2045 and 2045–2095. The changes are computed as the difference between the two ten-year mean centered on each period limit. For the 21st century, the changes are computed for the SRES A2, A1B and B1 scenarios.

concentration leads to a positive change of the radiative forcing. For the SRES-B1 scenario, which has low GHG emissions, one can therefore anticipate that the temperature increase due to sulfate aerosol reduction for the period (2045–2095) will be significant compared to the temperature increase due to GHG increase over the same period.

[15] The decrease of the relative importance of sulfate aerosol effects compared to the GHG increase effect has two main reasons. The first and the most important one is simply the decrease in sulfate emissions. Indeed the forcing efficiency of the direct effect (in W(g sulfate)$^{-1}$) is almost constant in our runs, as previously found by PBH05. The second reason is a decrease of the forcing efficiency of the first indirect effect, which decreases by a factor of two during the 21th century, mainly because of a shift in regional patterns of emissions and a saturation in the indirect effect [PBH05]. In our case, this phenomenon plays a small role as the first indirect effect is small compared to the direct effect.

[16] The results presented here are based on a semi-empirical model of the first indirect model and, given the range of possible radiative forcing and possible climate response to forcing, we believe that these results are robust. Nevertheless, one should remember that the interactions between aerosol and climate are far from being understood, that there is a large variety of aerosols whereas we consider only sulfate aerosols, and that important observations such as the global dimming and its recent slowing down are currently not explained and may be partly due to aerosols.

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