Response of a coupled chemistry-climate model to changes in aerosol emissions: Global impact on the hydrological cycle and the tropospheric burdens of OH, ozone, and NOx

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[1]   In this study, we analyze the response of the coupled chemistry-climate system to changes in aerosol emissions in fully coupled atmospheric chemistry-climate-slab ocean model simulations; only the direct radiative effect of aerosols and their uptake of chemical species are considered in this study. We show that, at the global scale, a decrease in emissions of the considered aerosols (or their precursors) produces a warmer and moister climate. In addition, the tropospheric burdens of OH and ozone increase when aerosol emissions are decreased. The ozone response is a combination of the impact of reduced heterogeneous uptake of N_2O_5 and increased ozone loss in a moister atmosphere. Under reduced aerosol emissions, the tropospheric burden of NO_x ($NO + NO_2$) is strongly reduced by an increase in nitric acid formation but also increased by the reduced N_2O_5 uptake. Finally, we discuss the significant difference found between the combined impact of all aerosols emissions and the sum of their individual contributions. **Citation:** Lamarque, J.-F., J. T. Kiehl, P. G. Hess, W. D. Collins, L. K. Emmons, P. Ginoux, C. Luo, and X. X. Tie (2005), Response of a coupled chemistry-climate model to changes in aerosol emissions: Global impact on the hydrological cycle and the tropospheric burdens of OH, ozone, and NOx, *Geophys. Res. Lett.*, *32*, L16809, doi:10.1029/2005GL023419.

1.   Introduction

[2]   Aerosols affect the radiative balance of the Earth's atmosphere [*Penner et al.*, 2001; *Ramanathan et al.*, 2001; *Menon et al.*, 2002; *Feichter et al.*, 2004; *Wang*, 2004] and its chemical composition [*Tie et al.*, 2001, 2005; *Liao et al.*, 2003; *Martin et al.*, 2003]. The radiative impact is a combination of direct and indirect effects [*Lohmann and Lesins*, 2002; *Ramanathan et al.*, 2005]. The chemistry is directly affected by aerosols through a combination of uptake of chemical species [*Jacob*, 2000] and perturbation of photolysis rates [*Martin et al.*, 2003; *Tie et al.*, 2005].

[3]   Changes in the radiative balance of the atmosphere are likely to affect the hydrological cycle at the regional and global scales [*Ramanathan et al.*, 2001; *Chung and Seinfeld*, 2002]. As the primary source of HO_x (OH +

HO_2) is proportional to the amount of water vapor [*Seinfeld*, 1986], it can be expected that any changes to the hydrological cycle will directly translate into changes in the OH distribution. However, because of the HO_x sink through the HO_2 uptake on aerosols, the OH concentration can also be influenced by changes in aerosol distribution. This influence is of relevance to climate as the tropospheric OH burden modulates the lifetime of methane (and of most chemically reactive species) and therefore its long-term climate forcing. In addition, recent studies have shown that, under the expected warmer and moister climate of the 21st century, the tropospheric ozone loss is likely to be enhanced [*Brasseur et al.*, 1998; *Prather et al.*, 2001, and references therein; *Collins et al.*, 2003].

[4]   In this study, we have performed a set of simulations that highlights the role of aerosols over a wide range of emission scenarios. Under these conditions, we focus on the two most basic ways aerosols can impact a coupled chemistry-climate model: direct radiative forcing and chemical uptake. In particular, we have chosen to simulate the state of the atmosphere when many of the aerosol (or their precursors) emissions are explicitly set to 0. While this is an unrealistic scenario (all aerosol emissions have some natural component to them), it provides an interesting upper limit scenario to the results of a possible decrease in aerosol emissions from their present-day estimates. The analysis in this paper will focus on the global aspects of the problem. To our knowledge, this paper is the first to study the coupled chemistry-climate response to changes in aerosol emissions over a wide range of scenarios.

2.   Model Description

[5]   To perform the simulations, we use the Community Atmosphere Model (CAM3, *Collins et al.* [2005]) coupled to the Model for Ozone and Related Tracers (MOZART) chemistry [*Horowitz et al.*, 2003], including aerosols [*Tie et al.*, 2001, 2005].

[6]   MOZART has a representation of tropospheric chemistry with non-methane hydrocarbons (NMHCs) treated up to isoprene, toluene and monoterpenes. This chemical mechanism is an extension of the mechanism presented by *Horowitz et al.* [2003]; changes include an updated terpene oxidation scheme and a better treatment of anthropogenic NMHCs (J. Orlando and G. Tyndall, personal communication, 2005). The MOZART aerosols have been extended from the work by *Tie et al.* [2001, 2005] to include a representation of ammonium nitrate that is dependent on the amount of sulfate present in the air mass following the

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Table 1. List of Heterogeneous Reactions Considered in This Study[a]

Uptake Reaction	Uptake Coefficient
N2O5	0.1
HO2	0.2
NO2	0.0001
NO3	0.001

[a]The values for the uptake coefficient (γ) are as recommended by *Jacob* [2000].

parameterization of gas/aerosol partitioning by *Metzger et al.* [2002]. In our model, we have included (Table 1) the uptake of N_2O_5, HO_2, NO_2, and NO_3 [*Jacob*, 2000] on aerosols. The effect of aerosols on photolysis rates is not included in this study. Because only the bulk mass is calculated, a lognormal distribution is assumed for all aerosols to calculate the surface area, using different mean radius and geometric standard deviation [*Liao et al.*, 2003].

[7] In summary, CAM3 with interactive chemistry simulates the evolution of the bulk aerosol mass of black or elemental carbon (EC, hydrophobic and hydrophilic), primary organic (POA, hydrophobic and hydrophilic), second organic (SOA, linked to the gas-phase chemistry through the oxidation of atmospheric NMHCs as in the work by *Chung and Seinfeld* [2002]), ammonium and ammonium nitrate (from NH_3 emissions), and sulfate aerosols (from SO_2 and DMS emissions). A description of sea-salt, updated from *Tie et al.* [2005], is also included. Finally, a monthly-varying climatology of dust is used only for radiative calculations.

[8] In this study, as we only consider the direct effect of aerosols, the atmospheric model is coupled to the chemistry solely through the radiative fluxes, taking into account all radiatively active gases and aerosols. The effects of the aerosols on the shortwave fluxes and heating rates are calculated following *Collins et al.* [2002]. In addition, the radiative properties of ammonium nitrate are assumed to be identical to ammonium sulfate [*Liao et al.*, 2003]. In order to capture the full range of climate feedbacks, we use CAM3 in combination with a slab ocean model [*Collins et al.*, 2005]. All simulations started from the end point of an 8-year simulation with the base case emissions and are at least 10 years long, of which the last 9 are analyzed; the overall simulation length was dictated by the need for an equilibrated climate, as measured by the global average surface temperature (see auxiliary material[1]). The horizontal resolution is $2°$(latitude) $\times$ $2.5°$(longitude), with 26 levels ranging from the surface to $\sim$4 hPa.

[9] For this study, we have performed a set of 4 simulations in which the emissions of SO_2, NH_3, EC, and POA are globally scaled by a single factor (0, 0.5, 1, and 1.5); these emissions were selected because they are strongly affected by an anthropogenic contribution, which has dramatically changed over the last century and is likely to change in the future [*Penner et al.*, 2001]. This simple scaling has the advantage of removing the impact that arises from changing the regional distribution of emissions; it is however an approximation to real trends and should be seen as a thought experiment. As an extreme case, we have

[1]Auxiliary material is available at ftp://ftp.agu.org/apend/gl/2005GL023419.

chosen to scale the aerosol emissions to 0. This scenario has the advantage of (1) setting a baseline against which perturbations can be calculated and (2) performing a sensitivity analysis similar to the removal of specific aerosol uptake reactions [e.g., *Dentener et al.*, 1993; *Tie et al.*, 2001; *Liao et al.*, 2003; *Martin et al.*, 2003].

[10] In the base case (scaling = 1), the global integral of the emissions are of 53.4 TgS/year for SO_2 [*Dentener et al.*, 2004], 61.3 TgN/year for NH_3 [*Dentener et al.*, 2004], 19.7 TgC/year for EC and 138.9 TgC/year for POA (combining *Bond et al.* [2004] for the fossil fuel emissions and *van der Werf et al.* [2003] for the biofuel/biomass burning emissions, scaled using the *Andreae and Merlet* [2001] emission factors). The regional distribution of these emissions is shown in the auxiliary material. Only the annual average is shown, although the emissions are available on a monthly basis. In addition we have used the *Granier et al.* [2004] emissions for the ozone precursors. The surface methane concentrations are kept constant at their present-day measured values. The overall distribution of gas-phase chemical species in this study is very similar to MOZART results with the same emissions (not shown). In terms of aerosols, these emissions lead to an annual global atmospheric burden of 1.4 TgC of POA, 0.2 TgC of SOA, 0.2 TgC of EC, 0.6 TgS of SO_4, and 0.3 TgN of NH_4, similar to the range given by *Penner et al.* [2001], *Chung and Seinfeld* [2001, 2005] and *Liao et al.* [2003].

[11] On an annual scale, the distribution of simulated aerosols in the base case leads to an aerosol optical depth (AOD) that is smaller than observed by approximately 20-50% (see auxiliary material, Figure 3), similar to *Ramanathan et al.* [2001] and *Feichter et al.* [2004]. While the AOD maxima over Western Europe and China seem to be well captured, the large aerosol loading areas (identified by large AODs) over Africa, Southeast Asia, North America, and South America are underestimated. Over North America, this AOD underestimate is found even though the simulated surface concentrations (see auxiliary material) are in either good agreement (sulfate) or overestimated (EC and POA). As an annual average, the average global aerosol optical depth is 0.08, on the low end of the estimates given by *Ramanathan et al.* [2001]. Consequently, the radiative forcing of aerosols in this model is likely biased low (there is however a significant error bar on the MODIS observations); therefore, the following analysis is probably only providing a lower bound on the radiative response of the chemistry-climate system to changes in aerosols.

3. Global Impact of Aerosol Emissions

[12] In this section, we first document the role of aerosol emissions in modifying the climate and the atmospheric composition. For the rest of the paper, the term "aerosol emissions" is used to describe the emissions of SO_2, NH_3, EC, and POA.

[13] With increased aerosol emissions, the net solar flux at the surface is reduced; consequently, the latent heat flux and vertically-integrated precipitable water decrease as the aerosol emissions increase (Figure 1a). This latter correlation describes the expected drying of the atmosphere when aerosol emissions are increased [*Ramanathan et al.*, 2001;
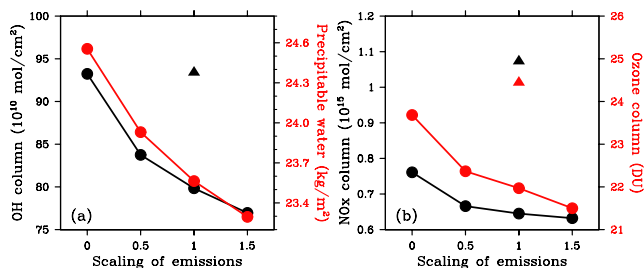
Figure 1. (a) Correlation plot between the scaling of present-day aerosol emissions and total precipitable water (left axis, black) and the tropospheric OH column (from the surface up to 300 hPa, right axis, red). (b) Same as (a) but for the NO_x (left axis, black) and ozone (right axis, red) tropospheric columns. The triangle indicates the simulation in which the chemical uptake by aerosols (see text for details) is removed.

Menon et al., 2002; *Liepert et al.*, 2004]. We also find that the global integral of OH (mass-weighted integral from the surface up to 300 hPa) significantly increases with decreasing aerosol emissions (Figure 1a). This increase (of approximately 15% between the scaling = 1 and the scaling = 0 cases) is due to both the increased amount of water vapor (exemplified here by the total precipitable water) and the decrease in the HO_x sink from aerosol uptake. The OH decrease between the cases scaling = 0 and scaling = 1 is large, considering that changes in global OH from pre-industrial times to present are estimated to be on the order of 9% [see *Lamarque et al.*, 2005, and references therein]. To identify the role of chemistry in the scaling = 0 case, we have also performed a simulation with present-day aerosol emissions but without aerosol uptake; it is identified by the triangle in Figures 1a and 1b. To ensure a meaningful comparison, we have adjusted the scaling = 0 results for the surface area from SOA and sulfate from DMS oxidation. In the case of OH, the effect of turning off the aerosol uptake of N_2O_5 (and to a much lesser extent HO_2) leads to an OH burden that is equivalent to the scaling = 0 case. There is a balance between an increased production and increased loss of OH in the scaling = 0 simulation, with marked regional differences (not shown). The consideration of the impact of aerosols on photolysis rates is likely to further increase the OH change between the scaling = 1 and scaling = 0 cases by approximately an additional 10% [*Martin et al.*, 2003].

[14] The global integral of ozone also shows a strong positive response to decreasing the aerosol emissions. In particular, the difference between the scaling = 0 and scaling = 1 cases amounts to approximately 2 DU (Dobson Units) or 10%. The role of the chemical uptake of aerosols is actually larger, accounting for almost 3 DU (see red triangle in Figure 1b). This means that the removal of aerosol emissions has a feedback that tends to reduce tropospheric ozone. This feedback ($\sim$5%, 1 DU out of 22 DU) is due to the more rapid ozone loss (through the increased $O(^1D) + H_2O$ reaction) when the climate warms up and becomes moister [*Brasseur et al.*, 1998], as it is the case when aerosol emissions are reduced. Indeed, the surface temperature increases by 0.6 K between the scaling = 1 and scaling = 0 cases (see auxiliary material).

[15] Similarly, the global integral of NO_x (which is directly affected by the uptake on aerosols of N_2O_5 and to a lesser extent NO_2) shows an increase (also of approximately 15%) with decreasing aerosol emissions (Figure 1b). As for ozone, we find that this increase is actually larger when only the effect of aerosol uptake is taken into account; this effect is responsible for an almost doubling of tropospheric NO_x burden, similar to *Dentener et al.* [1993]. The reduction from the climate feedback (moving from the simulation identified by the triangle to the scaling = 0 simulation) is in the case of NO_x even larger than in the case of ozone, amounting to $\sim$50%. This is mostly associated to a more rapid conversion of NO_x to nitric acid (through OH + NO_2) in regions of low hydrocarbon/NO_x ratio (K. Murazaki and P. G. Hess, How does global warming contribute to ozone change over the U.S.?, submitted to *Journal of Geophysical Research*, 2005) when aerosol emissions are reduced.

4. Individual Impacts

[16] In addition to the analysis of the response of the chemistry-climate system to the overall scaling of aerosol emissions, we examine here the separate impact of each aerosol emission types. For that purpose, we have performed four additional simulations in which the emissions are scaled by 1.5 one at a time instead of all simultaneously. The goal of this analysis is 1) to identify the relative importance of each aerosol type and 2) to measure the degree of nonlinearity from using separate instead of simultaneous emission changes. With respect to climate (measured here by the integrated precipitable water), SO_2, POA and EC emissions have significant impacts. However, the climate response for EC is of opposite sign than for all the other aerosols; as shown in *Chung and Seinfeld* [2005], the radiative impact of the absorbing EC causes an increase in water vapor consistent with the increase in air temperature. The sum of all separate impacts on the precipitable water (Figure 2a) ends up being approximately 60% of the difference between the scaling = 1.5 and scaling = 1 simulations (the remaining difference is shown in Figure 2 by the dashed
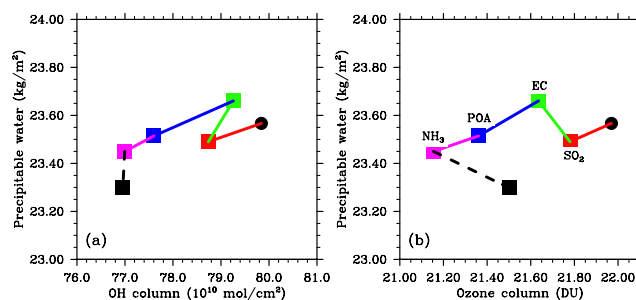
Figure 2. Incremental impact of scaling the aerosol emissions by 1.5 (see text for details) for (a) total precipitable water and the tropospheric OH column and (b) total precipitable water and the tropospheric ozone column. The initial point (base case, scaling = 1) is identified by the black dot. First (red), the impact of increasing SO_2 emissions only (by 1.5) is added. Second (green), EC. Third (blue), POA. Fourth (magenta), NH_3. The simulation in which all emissions were simultaneously increased by 1.5 is identified by the black square.

line). This nonlinear behavior was found in other climate indicators, such as surface temperature or latent heat flux (not shown) and is due to the increased lifetime (from the slower hydrological cycle when all aerosols are included) of the aerosols affected by wet removal; this longer lifetime has the implication that the overall impact of the aerosols is amplified.

[17] With respect to chemistry, while the SO_2, NH_3 and POA emissions act to decrease the OH column, the EC acts to increase it. Also, there is no indication of a significant nonlinearity in the OH response (Figure 2a). On the other hand, the tropospheric ozone column (Figure 2b) is strongly affected by the consideration of separate instead of simultaneous emission changes. As shown in section 3, this is due to a combination of uptake by aerosols and chemistry impacts. In particular, the large nonlinearity seen in the integrated precipitable water translates into an increase in ozone through a decrease in the ozone chemical loss.

5. Discussion and Conclusions

[18] In this study, using a fully coupled chemistry-climate model with aerosols, we have documented changes in the atmospheric climate and chemical state under a variety of aerosol emission scenarios (increase and decrease of present-day conditions). With no other changes imposed (fixed greenhouse gases, such as carbon dioxide and methane, and ozone precursors emissions), we have found a strong response in surface temperature and in the hydrological cycle at the global scale [*Ramanathan et al.*, 2001]. In addition, we have found that, at the global scale, these changes strongly impact the tropospheric OH, ozone and NO_x burdens, with an amplitude that significantly modifies the chemistry through uptake on aerosols. These results indicate that the overall response of the system is critically and nonlinearly dependent on the associated changes in aerosols through their impact on climate and chemical uptake.

[19] Even with the shortcomings of this study (no indirect effect, no impact of aerosols on photolysis rates, and no chemical uptake on dust), we have shown that it is necessary to consider the response of the chemistry-climate system in a fully coupled manner. This coupling is not achieved only through gas-phase chemistry but requires a representation of aerosols (including their mixing state [*Chung and Seinfeld*, 2005]) and their impact on radiation and chemistry.

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