



Response of a coupled chemistry-climate model to changes in aerosol emissions



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Abstract. In this study, we analyze the response of a coupled chemistry-climate (the NCAR Community Atmosphere Model, CAM3) system to changes in aerosol emissions in fully coupled atmospheric chemistry-climate-slab ocean model simulations. Using this model 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. **At the global scale, a decrease in emissions of the considered aerosols (or their precursor : SO₂, NH₃, primary organic and soot) produces a warmer and moister climate. Without most aerosols, the globally-averaged surface temperature is approximately 0.5°C warmer.** In addition, the tropospheric burdens of OH and ozone significantly increase when aerosol emissions are decreased. These chemical responses are a combination of the impact of reduced heterogeneous uptake and impact (such as increased ozone loss) of a moister atmosphere.

Model description and setup

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].

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 Chung and Seinfeld [2002]), ammonium and ammonium nitrate (from NH₃ emissions), and sulfate aerosols (from SO₂ 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.

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 horizontal resolution is 2°(latitude)×2.5°(longitude), with 26 levels ranging from the surface to ~4 hPa.

References

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Effects of aerosols on atmosphere

Aerosols affect

➤ the **radiative balance** of the Earth's atmosphere by a combination of direct and indirect effects
➤ its **chemical composition**; uptake of chemical species [Jacob, 2000] and perturbation of photolysis rates [Martin et al., 2003; Tie et al., 2005].

Depending on their chemical composition, shape and size, aerosols will absorb or scatter light

- **scatter**: the radiative energy is sent back to space and therefore lost from the system. Aerosols exert a net cooling of the atmosphere
- **absorb**: the radiative energy acts to heat the absorbing layer in the atmosphere

both imply a **loss of energy** at the surface

↳ **Direct effect of aerosols**

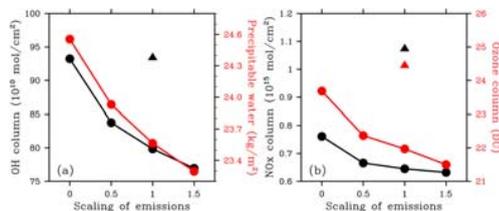
The indirect effects are

- **Semi-direct effect** : absorption can lead to strong heating, which can drop the relative humidity and thus evaporate clouds
- **First indirect effect** : aerosols increase the number of cloud drops, which enhance the reflection back to space by clouds (Twomey effect). This is not considered in this study

Impact on hydrological cycle and chemical composition

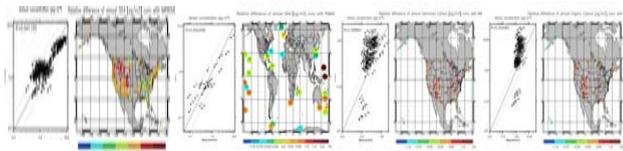
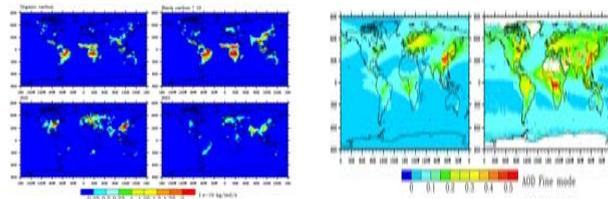
With **increasing** aerosol emissions, we find that, at the global and annual scale,

- the **net solar flux** at the surface is **reduced**
- the **latent heat flux** and vertically-integrated precipitable water **decrease** (see below, left panel, red line)
- the global integral of **OH** (mass-weighted integral from the surface up to 300 hPa) significantly **decreases** (below, left panel, lack line), due to both the decreased amount of water vapor and the increase in the HO_x sink from aerosol uptake (this latter effect is shown by the triangle)
- the global integral of **ozone** (right panel, red line) also **decreases**, due to the increased O(1D)+H₂O reaction in a warmer and moister climate and the increase in chemical uptake
- the global integral of **NO_x** (=NO+NO₂) **decreases** (right panel, black line), due to slower conversion of NO_x to nitric acid (through OH+NO₂) in regions of low hydrocarbon/NO_x ratio [Murazaki and Hess, 2005]



Analysis of the simulated aerosols and aerosol optical depth

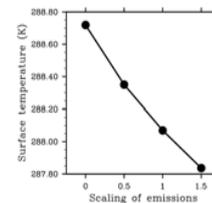
On an annual scale, the distribution of simulated aerosols in the base case (using the emissions shown below, top left) leads to an aerosol optical depth (AOD) that is smaller than observed by approximately 20-50% (Figure below, top right), similar to Ramanathan et al. [2001] and Feichter et al. [2004]. 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. Comparison with observations indicates little bias with for sulfate over the US but an overestimate of organic and black carbon.



Impact on surface temperature

With **increasing** aerosol emissions, we find that the global annual average **temperature decreases**; when all aerosol emissions are set to 0, the surface temperature increases by approximately 0.5K (see figure to the right). This is a direct consequence of the reduction of the net solar flux at the surface with increasing aerosols.

Under pre-industrial conditions (with much smaller aerosol emissions than the present-day levels), the surface temperature becomes therefore significantly warmer.



Conclusions

With no other changes imposed (fixed greenhouse gases, such as carbon dioxide and methane, and ozone precursor emissions), we have found a **strong response to changes in aerosol emission in surface temperature, hydrological cycle and chemical composition 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.