Global simulation of aerosol effects on tropospheric photolysis frequencies and photochemistry

TIAN Rong, MA Xiao-yan
Key Laboratory of Meteorological Disaster, Ministry of Education, Joint International Research Laboratory of Climate and Environment Change, Collaborative Innovation Center on Forecast and Evaluation of Meteorological Disaster, Key Laboratory for Aerosol-Cloud-Precipitation of China Meteorological Administration, Nanjing University of Information Science & Technology, Nanjing 210044, China

Introduction

1. Tropospheric ozone (O3) is an important greenhouse gas in the troposphere and plays a key role in determining the oxidation capacity of the atmosphere. It is well-known that aerosols impact gas-phase atmospheric chemistry by altering photolysis rates and heterogeneous reaction.
2. The focus of this study is to evaluate the impacts of aerosols on distributions and concentrations of tropospheric OH, O3 through altering photolysis frequencies using the global chemical transport model GEOS-Chem. Sensitivity simulations are conducted to quantify the photochemical effects of aerosols including sulfate, nitrate, ammonium, carbonaceous, dust and sea salt.

Methods

• Based upon the 3-D global chemical transport model GEOS-Chem.
• Simulation type: v11-01 NOx-Ox-HC-Aer-Br (aka tropchem)
• Resolution: horizontal resolution 4° × 5° Vertical:46 sigma levels extending up to 0.1 hPa
• Simulation period: January, April, July, October 2013
• Study area: Global and regional (Central Eastern China: 30-40° N, 110-125° E)

Numerical Experiments

<table>
<thead>
<tr>
<th>Experiments</th>
<th>Assumptions</th>
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<tbody>
<tr>
<td>CTRL</td>
<td>Control run with effects of all aerosols on photolysis frequencies</td>
</tr>
<tr>
<td>OFF</td>
<td>Without the effect of aerosols on photolysis frequencies</td>
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Aerosols considered include sulfate, nitrate, ammonium, organic carbon, black carbon, mineral dust and sea salt.

The impact of aerosols on photolysis frequencies can be quantified by CTRL-OFF/ OFF (%).

Aerosol-induced impacts on photochemistry

• Aerosols can reduce actinic flux by scattering or absorbing ultraviolet (UV) radiation, thus reducing photolysis frequencies and OH produced concentrations. As a result, surface-layer O3 concentration is reduced.
• As sulfate and OC are scattering aerosols, they can scatter solar radiation back into the atmosphere, thus increasing the actinic flux above the boundary layer.
• In the summer, with higher sulfate AOD and more backscattering flux above the boundary layer, the increase of actinic flux is maximum (10%). [J(NO2)] and [J(O)] show similar patterns with actinic flux profile.
• Among four seasons, the actinic flux, photolysis frequencies as well as oxidants are significantly reduced at the surface with a maximum reduction in winter, followed by autumn. In summer, the reduction is minimum. The reduction in winter is more significant than other seasons likely due to the larger path length of radiation.

Conclusions

• Aerosols have important impacts on the UV actinic flux, which results in reductions in photolysis frequencies, OH concentrations in the boundary layer.
• Above the boundary layer, scattering aerosols such as sulfate can enhance actinic flux, thus increasing the photolysis frequencies, OH and O3 concentration. The reduction in winter is more significant than other seasons likely due to the larger path length of radiation.
• The global monthly mean perturbation of surface O3 by aerosol ranges from -16%~2%. O3 changing factor:
• Meteorological factors such as solar irradiance and temperature
• Precursors, i.e., NOx, NMHC (non-linear)

References


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