# Global simulation of aerosol effects on tropospheric photolysis frequencies and photochemistry

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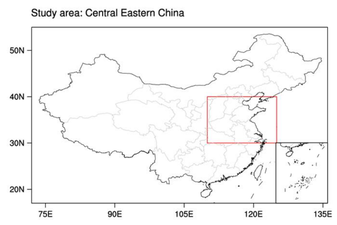
## Introduction

1. Tropospheric ozone ($O_3$) 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, $O_3$ through altering photolysis frequencies using the global chemical transport model GEOS-Chem.
3. 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

| Experiments | Assumptions |
|---|---|
| CTRL | Control run with effects of all aerosols on photolysis frequencies |
| OFF | Without the effect of aerosols on photolysis frequencies |

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 (%).**

## AOD horizontal and vertical distribution

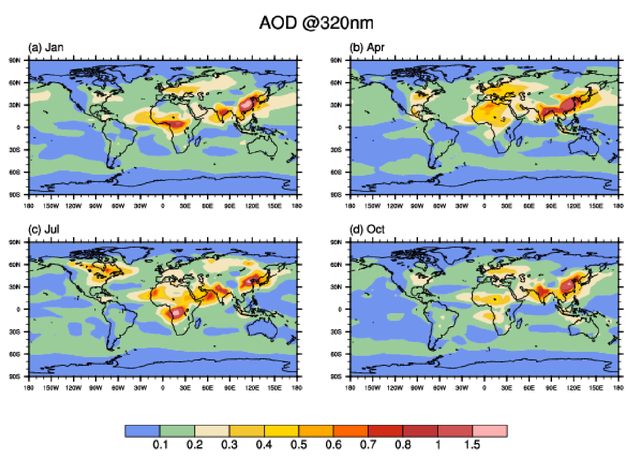



Fig. 1 Simulated global distribution of monthly averaged AOD at 320 nm

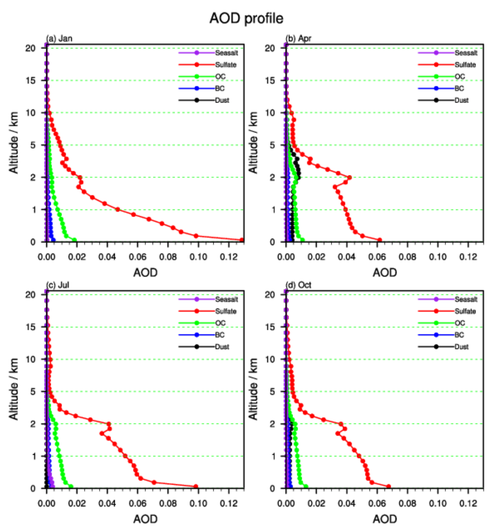



Fig. 2 Simulated vertical distribution of monthly averaged AOD at 320 nm over central eastern China

➢ ***Global distribution of AOD：***

The highest calculated AOD at 320 nm are located in eastern Asia, northern Africa, India and western Europe, which coincides with the higher natural or anthropogenic aerosol concentrations in these regions.

The calculated maximum optical depths in these regions range from 0.6 to 1.5. Global average of AOD is highest in summer, followed by winter, and the lowest in autumn.

➢ ***Vertical distribution of AOD over central eastern China：***

In this region, AOD is composed predominantly of sulfate, followed by organic carbon, mainly scattering aerosols. Optical depths of mineral dust and black carbon (absorbing aerosols) are relatively small.

Among four seasons, sulfate optical depth is maximum during summer, followed by winter. Dust AOD is highest in April, lowest in winter. Carbonaceous AOD is highest in summer. Overall, AOD in central eastern China is maximum during winter, followed by summer.

## References

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## Aerosol-induced impacts on photochemistry

**Vertical profile of the aerosol induced impacts**

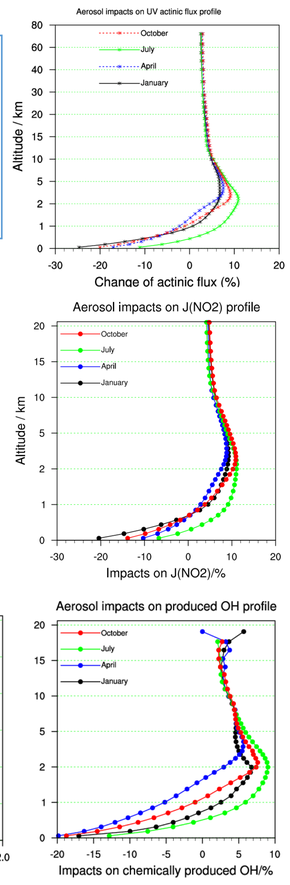



Fig 3. Vertical profile of simulated changes in monthly mean tropospheric photochemistry (%) through aerosol effect on photolysis frequencies

- 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 $O_3$ 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[$NO_2$] and J[$O_3$] 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.

### Aerosol induced impacts on surface-layer photochemistry

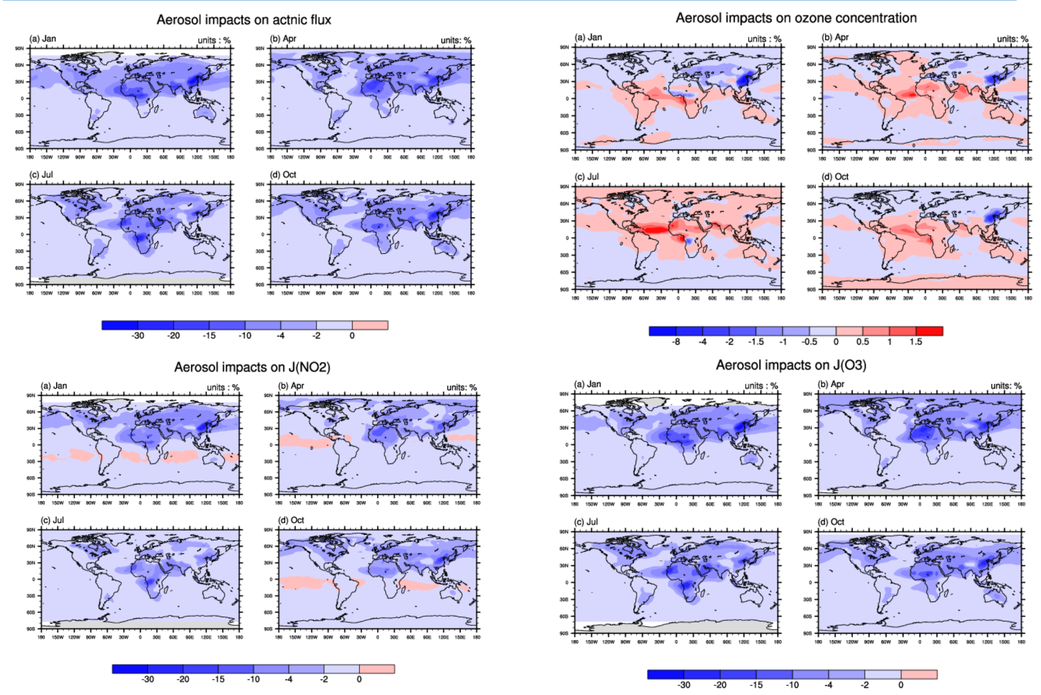



Fig. 4 Simulated changes in monthly mean surface-layer photochemistry (%) through aerosol effect on photolysis rates

The surface-layer J[$O_3$] and J[$NO_2$] are reduced, respectively by 10~30% and 10~40% in regions with high aerosol loading, such as east Asia, India, Europe and north Africa.

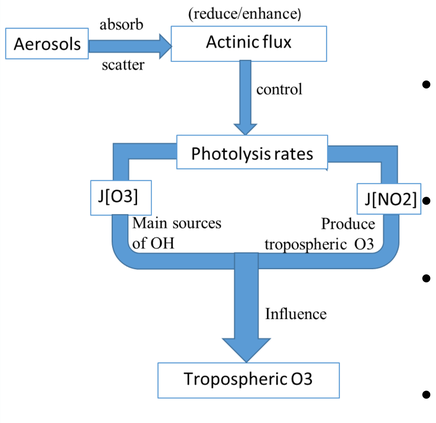
The global monthly mean perturbation of surface $O_3$ by aerosol ranges from -16%~2%.

$O_3$ influencing factor：

➢ Meteorological factors such as solar irradiance and temperature

➢ Precursors, i.e., NOx, NMHC (non-linear)

## 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 $O_3$ 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 $O_3$ by aerosol ranges from -16%~2%. The maximum increase appears in tropical Atlantic ocean during summertime.
- $O_3$ change due to photolysis depends not only on AOD distribution but also on the availability of $O_3$ precursors.

## Acknowledgment:

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