

# On Sulfate Tropospheric Chemistry: An Interactive Global Climate Chemistry Modeling Approach

Sunita Verma\*, H. C. Upadhyaya and O. P. Sharma

\*Centre of Excellence in Climatology, Birla Institute of Technology Mesra, Ranchi, Jharkhand INDIA, [verma.sunita@gmail.com](mailto:verma.sunita@gmail.com)

Centre for Atmospheric Sciences, Indian Institute of Technology Delhi, Hauz-Khas, New Delhi 110016, INDIA.



## Abstract

Atmospheric chemistry plays a crucial role in climate by controlling the abundance and distribution of natural and anthropogenic agents such as greenhouse gases, aerosols, and clouds, which influence incoming and/or outgoing radiation, temperature and precipitation. To accurately represent the sulfur tropospheric chemistry in a global climate chemistry model, a throughput consideration of multi-phase processes is essential i.e. the incorporation of a heterogeneous reaction system of aerosols (both gas phase and aqueous phase, predominating in cloud water), their subsequent transformation, dispersion and the mechanism for their eventual removal from the atmosphere.

In most of the global sulphur models only sulphate mass is estimated as prognostic variable. The number concentrations are inferred assuming a constant sulphate size and size distribution properties. In contrast, in our implementation sulphate number concentrations are also estimated as prognostic variable. Knowledge of the sulphate aerosols number concentration is important to understand the indirect radiative forcing of the aerosols and to refine estimates of direct forcing. The direct and indirect forcings are calculated every time step based on the instantaneous model calculated sulphate mixing-ratios.

## Model Description

### Interactive Chemistry module in LMDZ

Sixteen Prognostic Tracers :

water vapour and liquid water, SO<sub>2</sub>, NO<sub>x</sub>, CO, O<sub>3</sub>, H<sub>2</sub>O<sub>2</sub>, H<sub>2</sub>S, HNO<sub>3</sub>, DMS, DMSO, MSA, Aerosol mass and # conc. in Aitken and accumulation modes

### Important aspect in model:

- Complete Online approach
- Short lived radicals OH-, HO<sub>2</sub> and NO<sub>x</sub> are updated within the model (~CTMs)
- Sulphate aerosol mass and number concentration (prognostic variables).

### Gas Phase Chemistry

$$\frac{dC}{dt} = P - LC \quad C(t+\Delta t) = \frac{P}{L} + \left[ C(t) - \frac{P}{L} \right] \exp(-L\Delta t)$$

$$[H_2SO_4]_{t+\Delta t} = [H_2SO_4]_t + k_{14}[SO_2][OH]\Delta t$$

[H<sub>2</sub>SO<sub>4</sub>] vapour concentration drives the aerosol model and is divided into two modes; a part goes into existing particles growth and a part gives rise to new particles

### Aqueous Phase Chemistry

$$[S(VI)]_{t+\Delta t} = [S(VI)]_t + \frac{d[S(VI)]}{dt}$$

$$[S(IV)]_{t+\Delta t} = [S(IV)]_t - \frac{d[S(IV)]}{dt}$$

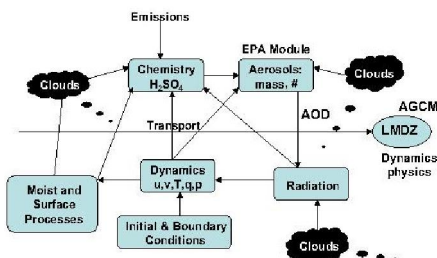
### Aerosol Model: Particle Dynamics

The aerosol module of this model is based on the Community Multiscale Air Quality (CMAQ) model of Binkowski and Shankar (1995), which is also a part of Model-3 of EPA (Binkowski and Roselle, 2003).

In the aerosol model the size distribution of sub-micron particles is represented by two overlapping intervals (modes) assuming a lognormal distribution within each mode.

$$n(\ln D) = \frac{N}{\sqrt{2\pi} \ln \sigma_g} \exp \left[ -0.5 \left( \frac{\ln \frac{D}{D_g}}{\ln \sigma_g} \right)^2 \right]$$

N: particle number concentration within the mode.  
D: particle diameter  
D<sub>g</sub> and σ<sub>g</sub>: geometric mean diameter and geometric standard deviation of the size distribution, respectively.  
The two modes correspond to the nucleation (Aitken) and accumulation modes.



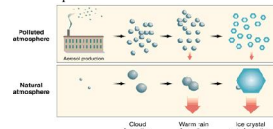
Coupling of chemistry/aerosol processes with dynamics and physics in LMDZ

## Rational

### Why Sulphate Aerosols?

- Important role in global climate and climate change via Direct and Indirect effects.
- Average global mean Radiative Forcing produced by sulphate aerosols is estimated to be - 2 to -3 W/m<sup>2</sup>
- Understanding on formation of sulphate aerosol in the atmosphere is therefore required, which can be achieved by the use of global model that incorporates a chemical model to describe the complex reaction system undergoing in the atmosphere.

The processes which affect clouds. The polluted cloud contains as many as droplets of the half size, twice the surface area, twice the optical depth and higher reflectivity than natural cloud (Toon 2000).



The chemistry and microphysics of aerosols and clouds play an important role in the indirect aerosol-climate effect. The processes that couple aerosol and cloud microphysics are needed for global climate model.

The prediction of cloud droplet number concentration (CDNC) is achieved from the number concentration of sulfate aerosol and cloud properties; hence the model design internally resolves the CDNC to the changes in the cloud water and aerosol parameters. The sulphate aerosol direct radiative forcing is calculated in the radiation code with the difference in shortwave fluxes with aerosol and without aerosol. The computations are done over the entire shortwave spectrum (0.25-4.0μm) and is divided into 24 wavelengths.

Extinction efficiency (α<sub>e</sub>) interaction aerosol and radiations

$$\text{Aerosol Optical Depth (AOD)} \tau = \alpha_e M_s l t$$

l = layer thickness

$$N_d = \max \{ 375 (1 - \exp[-2.5 \times 10^{-9} N_j]), N_{\min} \} \quad N_{\min} = 5 \text{ cm}^{-3}$$

## Numerical Simulations

### Resolution:

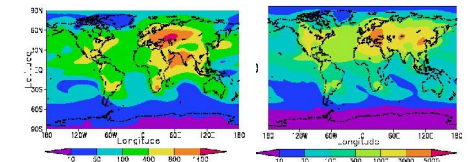
3.75° along longitude, 2.5° along latitude; (96 \* 72) with 19 vertical levels.  
The time step: ~ 30 min physical and chemical parameterization

Initial conditions: 20th December 1998 ECMWF analyses.

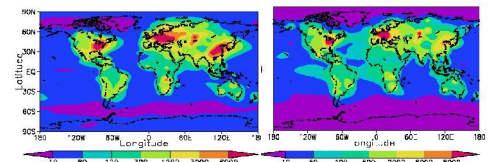
Boundary conditions: SST from ECMWF, other climatological fields

Simulations are carried out for a period of 24 months. The result presented here include the last 12 months

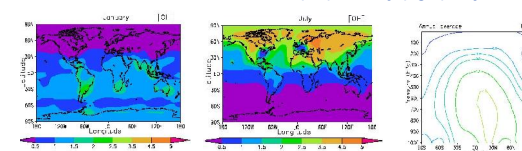
### 1. Distribution of SO<sub>2</sub> (pptv) at the surface in (left) January (right) July.



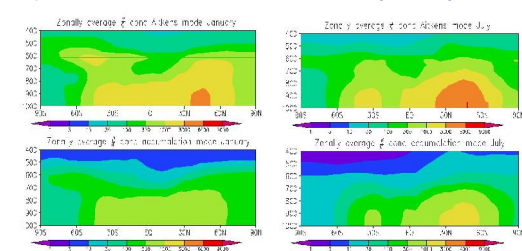
### 2. Distribution of sulphate mixing ratio (pptv) at the surface in (left) January (right) July



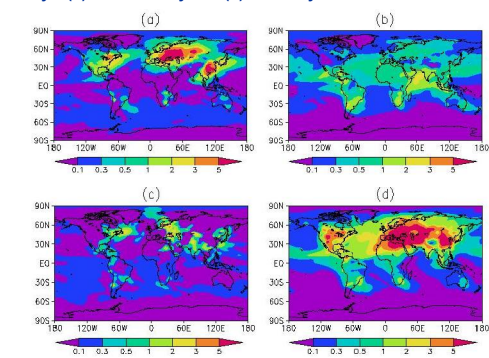
### 3. Distribution of OH at the surface in (left) January (right) July.



### 4. Zonally-averaged profile for Aitken mode and accumulation mode sulphate concentrations for the months of January and July.



### 5. Sulphate mixing ratio (ug/m<sup>3</sup>) in (a) APP January and (c) APP July (b) GPP January and (d) GPP July



## Conclusions

- Sulphate gross production is largely dominated by aqueous phase production (~70%).
- Whereas Gas phase production makes a major contribution to the total sulphate burden.
- Model a useful tool to investigate the climatic effects of anthropogenic sulphate and natural DMS emissions.

### References

- S. Verma, Boucher, O., Reddy, M. S., Upadhyaya, H. C., Le Van, P., Binkowski, F. S., and Sharma, O. P.: Tropospheric distribution of sulphate aerosols mass and number concentration during INDOEX-IFP and its transport over the Indian Ocean: a GCM study, Atmos. Chem. Phys., 12, 6185-6196, doi:10.5194/acp-12-6185-2012, 2012.
- S., Verma, Boucher, O., Reddy, M. S., Upadhyaya, H. C., Levan, P., Binkowski, F., and Sharma, O. P., Modeling and analysis of sulfate aerosol processes in an interactive chemistry GCM, Vol. 112, No. D3, D03207, doi: 10.1029/2005JD006077, J. Geophys. Research, 2007.
- S. Verma, O. Boucher, H.C. Upadhyaya and O. P. Sharma, Sulfate aerosols forcing: An estimate using a three-dimensional interactive chemistry scheme, Atmospheric Environment, Volume 40, Issue 40, 7953-7962, 2006.