

Lecture 39. Global change due to anthropogenic aerosols: sulfates.

Objectives:

1. Natural and anthropogenic sulfates.
2. Climatic effect of tropospheric sulfate aerosols.

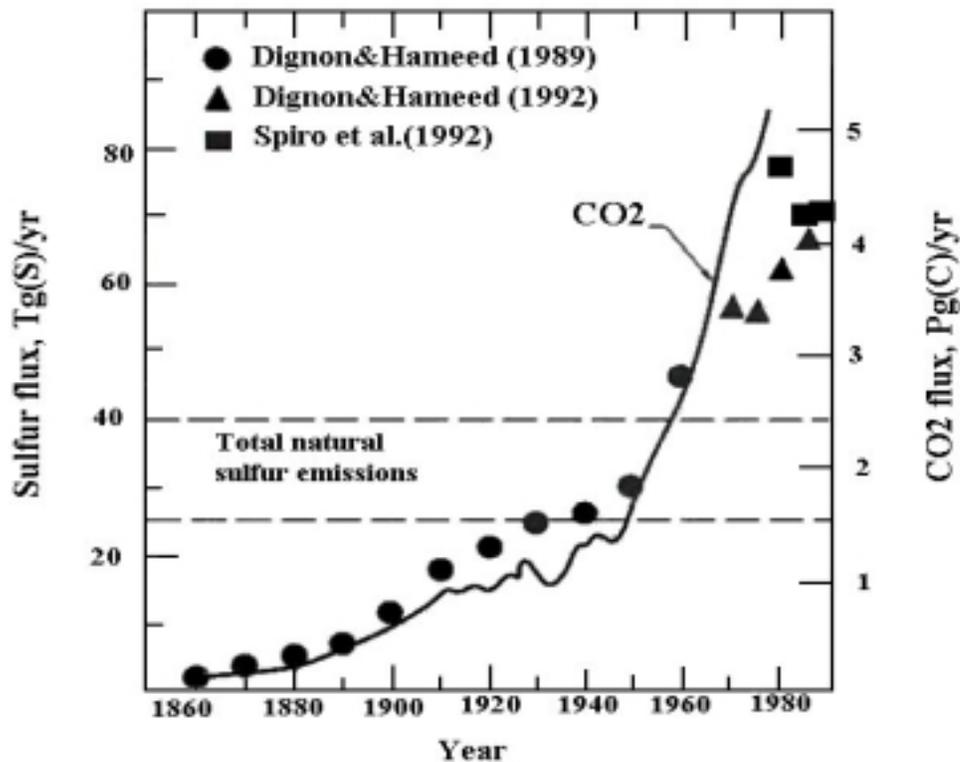
Readings: Seinfeld and Pandis (1998): p. 1156-1160

1. Natural and anthropogenic sulfates.

1991 Charlson et al. estimated direct aerosol forcing by sulfate aerosols.

Authors employed a global mean climate model (box model) and global anthropogenic emissions of SO₂.

Figure 39.1 Global anthropogenic SO₂ and CO₂ emissions since 1860 (IPCC 1995).



- Global sulfur emissions to the atmosphere have natural and anthropogenic sources.

Table 39.1 Present estimates of global natural sulfur emission.

| Source | Emissions (Tg S yr ⁻¹) | Species emitted |
|------------------|------------------------------------|---|
| Volcanic | 10 (+/- 5) | SO ₂ , H ₂ S, COS |
| Soils and plants | 1-4 | H ₂ S, DMS, COS |
| Biomass burning | 2-3 | SO ₂ , COS |
| Oceanic | 36 (+/- 20) | DMS, H ₂ S, COS, CS ₂ |

| Species | Emissions (Tg S yr ⁻¹) |
|--|------------------------------------|
| DMS (CH₃SCH₃) dimethyl sulfide | 35 (+/- 20) |
| SO₂ | 12 (+/- 6) |
| H₂S (hydrogen sulfide) | 3 (+/- 2) |
| CS₂ (carbon disulfide) | 1 (+/- 1) |
| COS (carbonyl sulfide) | 1 (+/- 1) |

NOTE: Dimethyl sulfide (DMS) is the dominant natural sulfur compound emitted from the world's ocean (of about 50%-80% of total natural sulfur emission). However, the uncertainty factor in the DMS estimate is of 2 to 3 and it remains an unresolved issue.

Major problems: complex pattern of DMS emission and poor understood DMS chemistry.

DMS oxidation chemistry starts with oxidation via reactions with OH and NO₃, followed by multi-steps mechanism, which is not completely understood. Therefore, the simplified DMS chemistry is used in the models.

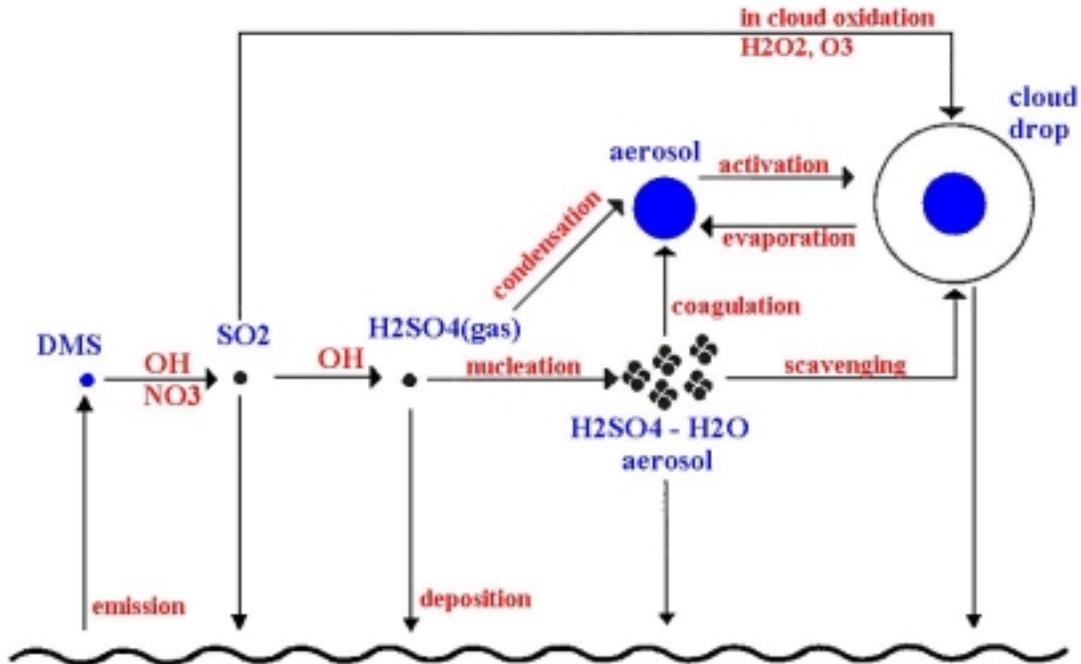
Simplified DMS chemistry:



where MSA is methane sulfonic acid $\text{CH}_3\text{SO}_2\text{OH}$, and α is the parameter.

- Because of the photochemical source of OH, DMS removal by OH occurs during daytime, leading to a pronounced diurnal cycle in DMS concentration.
- Once SO_2 formed it can be further converted to H_2SO_4 and sulfate aerosol.

Figure 39.2 Simplified chemical and physical processes of sulfate aerosol formation in the marine boundary layer.



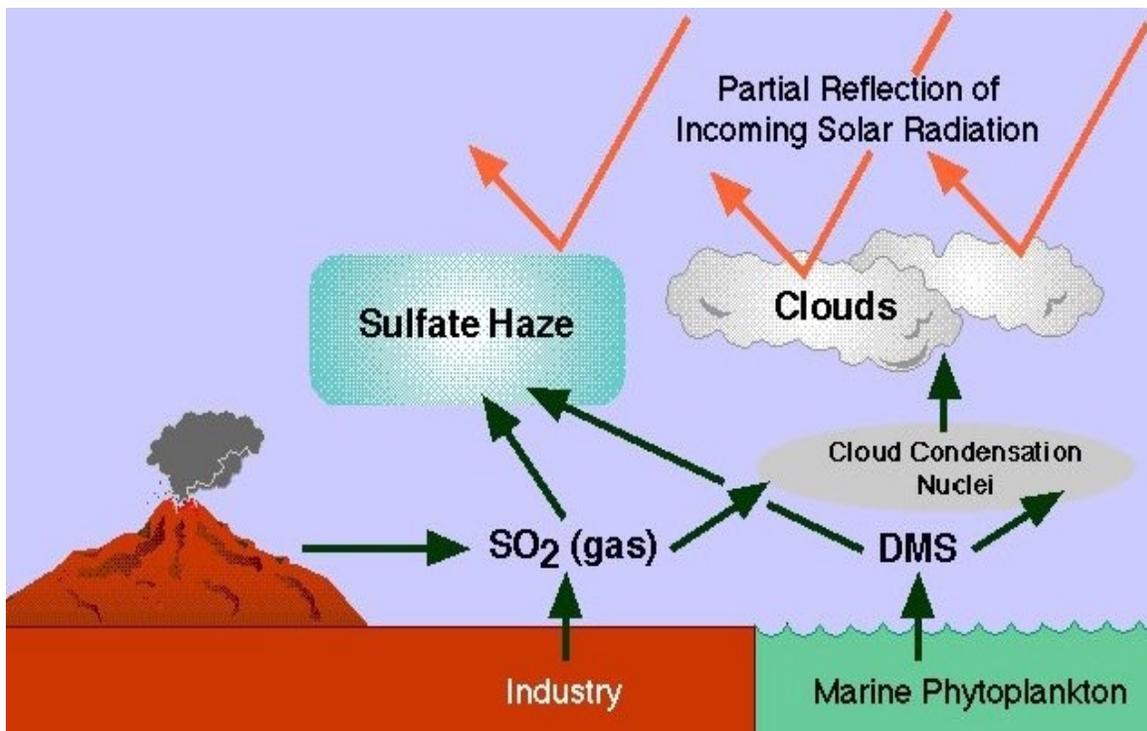
NOTE: compare the formation of sulfates in marine conditions to the formation of sulfates from industrial SO₂ pollution (Lecture 23).

2. Climatic effect of tropospheric sulfate aerosols.

In general, the mechanisms by which atmospheric aerosols influence radiative forcing of climate can be distinguished into three categories:

- **Direct forcing.** Aerosols can scatter and/or absorb solar radiation, and therefore alter atmospheric temperature.
- **Indirect (cloud) forcing.** Aerosols can alter cloud particle size distribution and cloud particle concentration and, hence, cloud albedo and the atmospheric temperature.
- **Indirect effects of aerosols on heterogeneous atmospheric chemistry.** Aerosol particles can serve as the site of atmospheric chemical reactions thereby increasing the rates of these reactions.

Figure 39.3 Climatic effects of tropospheric sulfate aerosols.



- **Anthropogenic sulfates** produce negative radiative forcing, leading to global cooling. Recent estimates of global mean forcing by sulfates are in the range from -0.3 to -1.3 W/m^2 . Major uncertainties: spatial and temporal distribution of sulfates and their optical properties.
- **Direct radiative forcing due to anthropogenic sulfates** is characterized by complex spatial distribution (in contrast to the forcing due to greenhouse gases).

Figure 39.4 Annual mean direct radiative forcing by anthropogenic sulfates (Kiehl et al., 1997)

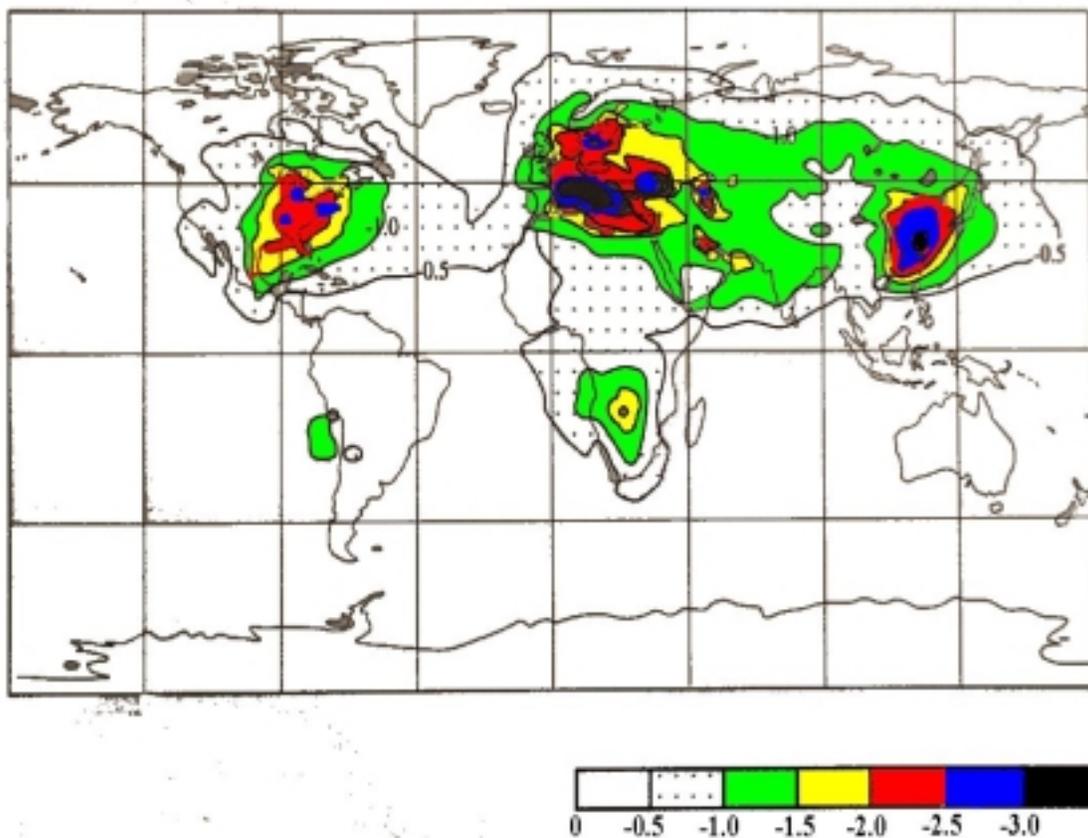
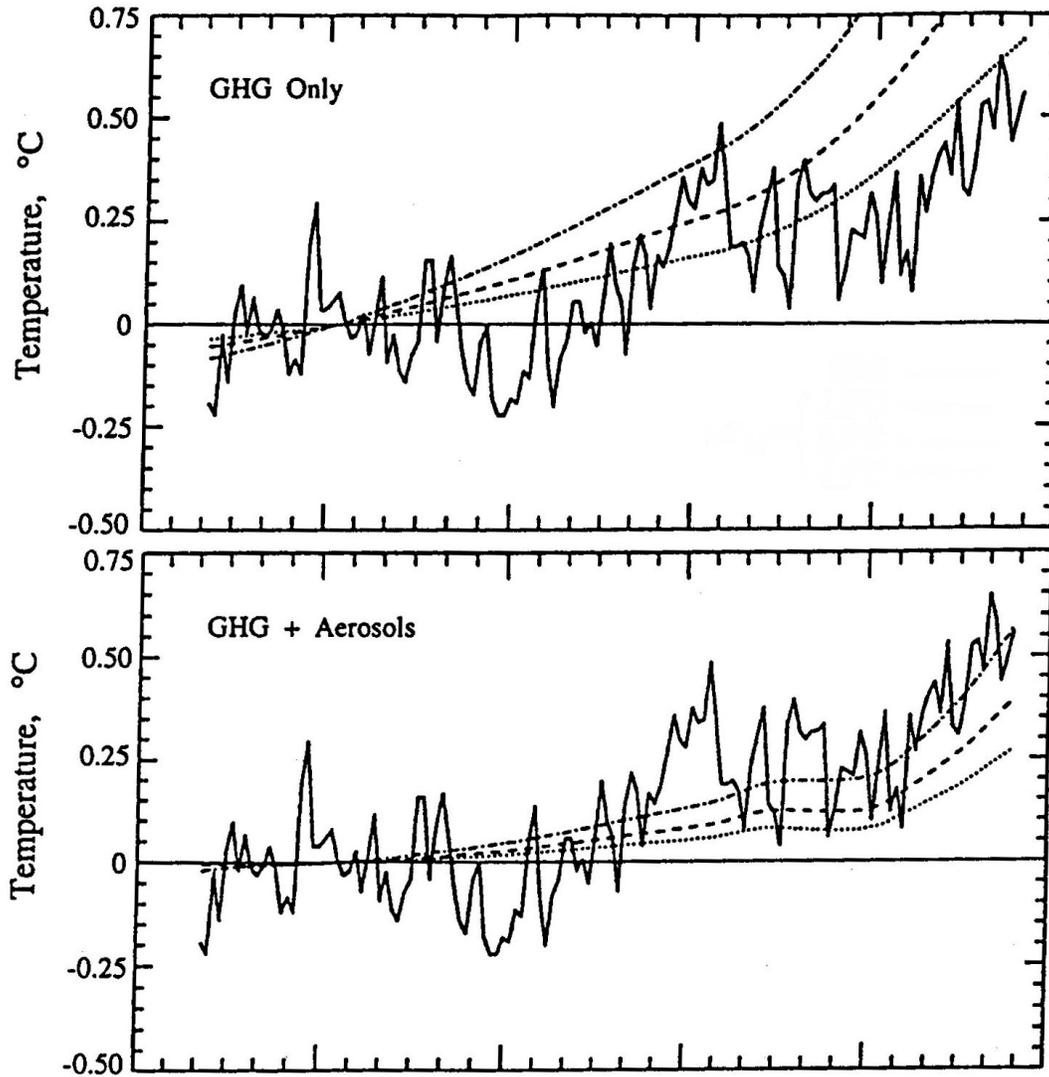


Figure 39.5 Comparison of observed (solid line) and modeled global mean temperatures (dashed lines), assuming greenhouse gases only (top panel), and greenhouse gases plus sulfates (bottom panel). Modeled temperatures were predicted by the model assuming CO₂-doubling scenario for three values of climate sensitivity (IPCC 1995).



- Accounting for aerosol forcing produces a better correspondence of observed and GCM modeled temperature records than is greenhouse gases (GHG) alone are considered.