VARIATION OF AEROSOL OPTICAL PROPERTIES AND RADIATIVE IMPLICATIONS

Stephen E. Schwartz

Aerosol Metrology for Climate Workshop
Gaithersburg, MD
March 14 – 15, 2011

www.ecd.bnl.gov/steve
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OVERVIEW

Aerosol processes that must be understood

Classes of aerosol properties and processes

Relation to climate change

Aerosol optical properties

Aerosol radiative influences

Some implications for climate change
**DIRECT RADIATIVE FORCING DUE TO ANTHROPOGENIC SULFATE AEROSOL**

\[
\Delta F_R = \frac{1}{2} F_T T^2 (1 - A_c)(1 - R_s)^2 \cdot \bar{\beta} \alpha_{SO_4^{2-}} f(RH) \cdot Q_{SO_2} Y_{SO_4^{2-}} \left( \frac{MW_{SO_4^{2-}}}{MW_S} \right) \theta_{SO_4^{2-}} / A
\]

\( \Delta F_R \) is the area-average shortwave radiative forcing due to the aerosol, W m\(^{-2}\)

\( F_T \) is the solar constant, W m\(^{-2}\)

\( A_c \) is the fractional cloud cover

\( T \) is the fraction of incident light transmitted by the atmosphere above the aerosol

\( R_s \) is the albedo of the underlying surface

\( \bar{\beta} \) is upward fraction of the radiation scattered by the aerosol,

\( \alpha_{SO_4^{2-}} \) is the scattering efficiency of **sulfate and associated cations** at a reference low relative humidity, m\(^2\) (g SO\(_4^{2-}\))\(^{-1}\)

\( f(RH) \) accounts for the relative increase in scattering due to relative humidity

\( Q_{SO_2} \) is the source strength of anthropogenic SO\(_2\) g S yr\(^{-1}\)

\( Y_{SO_4^{2-}} \) is the fractional yield of emitted SO\(_2\) that reacts to produce sulfate aerosol

\( MW \) is the molecular weight

\( \theta_{SO_4^{2-}} \) is the sulfate lifetime in the atmosphere, yr

\( A \) is the area of the geographical region under consideration, m\(^2\)

*Charlson, Schwartz, Hales, Cess, Coakley, Hansen & Hofmann, Science, 1992*
AEROSOL DIRECT SHORTWAVE FORCING

Global Average for Nonabsorbing Aerosol

\[ \Delta F = -\frac{1}{2} F_0 T^2 (1 - A_c)(1 - R)^2 \bar{\beta} \tau \]

\[ \tau = \int \alpha C dz = \int \sigma_{sp} dz \]

- Light Scattering Coefficient
- Mass Concentration
- Mass Scattering Efficiency
- Aerosol Optical Depth
- Mean Upscatter Fraction
- Surface Reflectance
- Cloud Fraction
- Atmospheric Transmittance
- Solar Constant
- Change in Net TOA Flux

Global Average for Absorbing Aerosol

\[ \Delta F = -\frac{1}{2} F_0 T^2 (1 - A_c)(1 - R)^2 \bar{\beta} \tau \omega \left\{ 1 - \frac{2R}{(1 - R)^2} \frac{(1 - \omega)}{\bar{\beta} \omega} \right\} \]

Single Scattering Albedo
AEROSOL PROCESSES THAT MUST BE UNDERSTOOD AND REPRESENTED IN MODELS

- Condensation
- Evaporation
- Surface chemistry
- Coagulation
- Light scattering and absorption $f(RH)$
- Oxidation
- Precursor emissions
- Water uptake
- Primary emissions
- Activation
- Autoconversion
- Subcloud scavenging
- Radiation transfer in clouds
- New particle formation
- Aquous chemistry
- Diffusion
- Evaporation
- Dry deposition

Approach to determine aerosol forcing

Numerical simulation of physical processes

Isomorphism of processes to computer code

Modeling aerosol processes requires understanding these processes, developing and testing their numerical representations, and incorporating these representations in global scale models.
APPROACH TO MODELING AEROSOL RADIATIVE EFFECTS

1. Emissions
2. Transport Chemistry Microphysics Removal Model
3. Size-distributed Composition
4. Optics Model
5. Optical Properties
6. Radiative Effects
7. Radiation Transfer Model
APPROACH TO MODELING AEROSOL RADIATIVE EFFECTS

- Emissions
- Transport Chemistry Microphysics Removal Model
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      - Optical Properties
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    - Radiative Effects
CLASSES OF AEROSOL PROPERTIES AND PROCESSES
CLASSES OF AEROSOL PROPERTIES AND PROCESSES

It is essential to distinguish . . .

Aerosol chemical and microphysical properties.

Aerosol optical properties.

Aerosol radiative influences.

Aerosol radiative forcing.
AEROSOL CHEMICAL AND MICROPHYSICAL PROPERTIES

Size distributed chemical composition: dry, or at reference relative humidity (RH).

Dependence on RH: hygroscopic growth

Liquid vs solid; homogeneous spheres? crystalline vs amorphous . . .

Internal vs external mixture:

Fully internally mixed: All particles have same composition.

Size-dependent internal mixture: Particles of a given size have same composition.

External mixture: Particles of same size may exhibit different composition.
SINGLE-PARTICLE OPTICAL PROPERTIES

Size
Real and imaginary components of index of refraction
Homogeneous, vs spherically symmetric, vs inhomogeneous

*Mie scattering theory* allows calculation of angular dependent scattering for homogeneous or for radially dependent, spherically symmetric refractive index (*e.g.* spherical shells).

*Nonspherical particles*: Approximate methods for spheroids, etc. Numerical methods.

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**AEROSOL OPTICAL PROPERTIES**

*Necessary* aerosol optical properties for calculating radiative influences are:

\[
\{ \text{Scattering coeff} \ \text{Absorption coeff} \} \iff \{ \text{Extinction coeff} \ \text{Single scat albedo} \}
\]

Asymmetry parameter: Average of cosine of scattering angle.

Dependence on *wavelength*.

Dependence on *relative humidity*.

*Also relevant*:

Phase function – Intensity of scattering as function of scattering angle.

Polarization – Spherical vs crystal, pertinent to source.
AEROSOL OPTICAL PROPERTIES

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RADIATIVE INFLUENCES

A change in radiative flux due to of the presence of aerosol.

Requires specification of the radiative situation: solar zenith angle, surface albedo, height of aerosol in atmosphere, cloud-free vs all-sky, . . .

Examples:

Aerosol optical depth: $\Delta$ optical depth due to aerosol.

$\Delta$ Direct normal solar irradiance.

$\Delta$ Diffuse downwelling shortwave irradiance at surface.

$\Delta$ \textit{Absorbed} downwelling shortwave irradiance at surface.

$\Delta$ Upwelling shortwave irradiance at top of atmosphere (TOA).

Might be instantaneous and local; might be temporal and/or spatial average.
RADIATIVE FORCING

A specific difference; a change in radiative flux between two specified conditions.

Examples:

Forcing by the total aerosol: Flux with aerosol minus flux without aerosol.

*Anthropogenic* aerosol forcing: Flux with anthropogenic aerosol minus flux without.

Might be instantaneous, local; might be temporal and/or spatial average; cloud-free or all-sky; might be surface or TOA.
RELATION TO CLIMATE CHANGE OVER THE INDUSTRIAL ERA
GLOBAL ENERGY BALANCE

Global and annual average energy fluxes in watts per square meter

Schwartz, 1996, modified from Ramanathan, 1987
GLOBAL ENERGY BALANCE
Global and annual average energy fluxes in watts per square meter

\[ \frac{1}{4} S_0 \]

\[ \alpha = 31\% \]

\[ 106 \]

\[ \text{Rayleigh} 27 \]

\[ 48 \]

\[ \text{Aerosol} 4 \]

\[ 27 \]

\[ 68 \]

\[ 390 \]

\[ \approx 288K \]

\[ 296 \]

\[ 90 \]

\[ 16 \]

\[ 31 \]

\[ \text{H}_2\text{O, CO}_2, \text{CH}_4 \ldots \]

\[ 69\% = 1 - \alpha \]

\[ \frac{1}{4} S_0 (1-\alpha) = \sigma T^4 \]

Shortwave

Longwave

Schwartz, 1996, modified from Ramanathan, 1987
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Global and annual average energy fluxes in watts per square meter

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\[ \Delta F = +2.6 \text{ W m}^{-2} \]

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GLOBAL ENERGY BALANCE
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\[ 237 \]
\[ 69\% = 1 - \alpha \]
\[ 1/4 S_0 (1-\alpha) = \sigma T^4 \]

Shortwave Longwave

\[ \Delta F = +2.6 \text{ W m}^{-2} \]

H$_2$O, CO$_2$, CH$_4$...

Atmosphere

Schwartz, 1996, modified from Ramanathan, 1987
Negative aerosol forcing substantially offsets GHG forcing. Aerosol forcing is highly uncertain.
Total forcing includes other anthropogenic and natural (solar) forcings. Forcing by tropospheric ozone, ~0.35 W m$^{-2}$, is the greatest of these. Uncertainty in aerosol forcing dominates uncertainty in total forcing.
LIGHT SCATTERING EFFICIENCY

Dependence on particle radius

Ammonium Sulfate, 550 nm

Data of Ouimette and Flagan, 1982
LIGHT SCATTERING CROSS SECTION:
DEPENDENCE ON PARTICLE SIZE, COMPOSITION AND RH

\( \alpha^*, m^2 (g SO_4^{2-})^{-1} \)

moles(SO_4^{2-})/particle

\( (NH_4)_2 SO_4 \)
\( NH_4 HSO_4 \)
\( H_2 SO_4 (97\% \text{ wt\%}) \)

Dry Radius, \( R_0 (\mu m) \)

Nemesure, Wagener & Schwartz, JGR, 95
Larger particles scatter increasingly in the forward direction.
AEROSOL RADIATIVE INFLUENCES
UPSCATTER FRACTION
Dependence on solar zenith angle, particle radius, and refractive index

\[ \mu_0 = \cos(SZA) \]
FORCING PER OPTICAL DEPTH

Global average, cloud-free sky - Scattering aerosol

Forcing per optical depth depends rather strongly on particle size.

Forcing accuracy 0.5 W m\(^{-2}\) requires optical depth accuracy 0.005 - 0.01 (0.01 - 0.02 for 60% cloud cover).
Forcing per optical depth depends rather strongly on particle size – and also rather strongly on aerosol absorption.
Global average sulfate optical thickness is 0.03: **1 W m$^{-2}$ cooling**.

In **continental U. S.** typical aerosol optical thickness is 0.1: **3 W m$^{-2}$ cooling**.
AEROSOL OPTICAL DEPTH AT ARM SGP

Fifteen years of daily average 500 nm AOD in North Central Oklahoma

Green curve is LOWESS (locally weighted scatterplot smoothing) fit.

Michalsky, Denn, Flynn, Hodges, Kiedron, Koontz, Schlemmer, Schwartz, JGR, 2010
AEROSOL COMPOSITION AT ARM SGP

Seven years of *daily average composition* in North Central Oklahoma

Black curve is LOWESS (locally weighted scatterplot smoothing) fit. Note summertime peak of sulfate.
AEROSOL OPTICAL DEPTH AND COMPOSITION AT ARM SGP

Daily average optical depth and composition in North Central Oklahoma

Summertime sulfate peak coincides with maximum optical depth.
AEROSOL COMPOSITION AT ARM SGP

Six days of **30-minute average composition** in North Central Oklahoma

**Preliminary Data**

- First data from new aerosol mass spectrometer installed at SGP.
- Continuous data; for particles with diameter ≤ 0.5 µm.
- Note high nitrate compared to sulfate; substantial organic component.
CLOSURE EXPERIMENTS

“Cubic meter experiments”
Compare measured optical properties with those modeled based on measured aerosol composition and size distribution.
Can be done locally. Require time series of composition, size distribution, and optical properties.

“Column closure experiments”
Compare optical depth, direct beam irradiance, diffuse irradiance; total and/or spectral, at surface or $f(z)$. Require characterization of aerosol optical properties vertically.
LOCAL OPTICAL CLOSURE EXPERIMENT

Aircraft measurements, south of Japan, ACE-Asia, 2001

$d\sigma_{\text{ext}}/d\log D_p$ calculated by Mie theory for measured composition; index of refraction from Bruggeman mixing rule.

Wang et al., JGR 02
LOCAL OPTICAL CLOSURE EXPERIMENT

Aircraft measurements, south of Japan, ACE-Asia, 2001

Calculated extinction coefficient and wavelength dependence agreed with observations within fairly broad uncertainty limits.
MEASURED AND MODELED SCATTERING COEFFICIENT

North Central Oklahoma, May 2003
Submicrometer aerosol, low RH

Modified from Andrews et al. JGR, 2006
MEASURED AND MODELED ASYMMETRY PARAMETER

North Central Oklahoma, May 2003

PCASP, Neph dry        TDMA, 85% RH; Neph, Ambient
AERONET, ambient column

Andrews et al. JGR, 2006
UNCERTAINTY IN AEROSOL DIRECT FORCING

Resulting from typical uncertainty in measurements of input variables

Colored bars denote uncertainties in 24-hr average forcing at equinox resulting from uncertainties in the individual parameters.

Black bar denotes resultant uncertainty in forcing.

Uncertainties are substantial in context of forcings over industrial period.

McCormiskey, Schwartz, Schmid, Guan, Lewis, Ricciuzzi, & Ogren, JGR, 2008
SOME IMPLICATIONS FOR CLIMATE CHANGE
EMPIRICAL DETERMINATION OF EARTH’S CLIMATE SENSITIVITY
Dependence on aerosol forcing

\[ S = \frac{\Delta T_{\text{obs}}}{F_{\text{eff}}} = -\Delta T_{\text{obs}} \]
\[ S = \frac{\Delta T_{\text{obs}}}{F_{\text{tot}} - N} \]
\[ S = \frac{\Delta T_{\text{obs}}}{F_{\text{aer}} + (F_{\text{tot}} - F_{\text{aer}}) - N} \]

\( F_{\text{tot}} - F_{\text{aer}} \) is due mainly to LLGHGs.
Heating rate \( N \) determined from rate of increase of ocean heat content.
Graph is plotted for uncertainty range of aerosol forcing from IPCC AR4.
Sensitivity increases strongly for aerosol forcing > 2 W m\(^{-2}\).
ALLOWABLE FUTURE CO₂ EMISSIONS

Dependence on climate sensitivity and acceptable increase in temperature relative to preindustrial

For $\Delta T_{\text{max}} = 2$ K,

If sensitivity $\Delta T_{2\times}$ is 3 K, no further emissions!
ALLOWABLE FUTURE CO₂ EMISSIONS

Dependence on climate sensitivity and acceptable increase in temperature relative to preindustrial

For $\Delta T_{\text{max}} = 2$ K,

Allowability of future emissions depends on climate sensitivity.
ALLOWABLE FUTURE CO₂ EMISSIONS

Dependence on climate sensitivity and acceptable increase in temperature relative to preindustrial

If $\Delta T_{\text{max}} > 2.1$ K and/or sensitivity $\Delta T_{2\times} < 3$ K, further emissions are allowed without exceeding $\Delta T_{\text{max}}$. 
ALLOWABLE FUTURE CO\textsubscript{2} EMISSIONS

Dependence on climate sensitivity and acceptable increase in temperature relative to preindustrial

If $\Delta T_{\text{max}} > 2.1$ K and/or sensitivity $\Delta T_{2\times} < 3$ K, further emissions are allowed without exceeding $\Delta T_{\text{max}}$.

If $\Delta T_{\text{max}} < 2.1$ K and/or sensitivity $\Delta T_{2\times} > 3$ K, committed temperature increase already exceeds $\Delta T_{\text{max}}$. 

ALLOWABLE FUTURE CO₂ EMISSIONS

Dependence on climate sensitivity and acceptable increase in temperature relative to preindustrial

Allowable future emissions or amount by which present GHGs exceed the allowable threshold depend on climate sensitivity and ΔT_{max}.
ALLOWABLE FUTURE CO₂ EMISSIONS

Dependence on climate sensitivity and acceptable increase in temperature relative to preindustrial

For $\Delta T_{\text{max}} = 2$ K . . .

If sensitivity $\Delta T_{2\times}$ is 3 K, no more emissions.
If sensitivity $\Delta T_{2\times}$ is 2 K, ~30 more years of emissions at present rate.
If sensitivity $\Delta T_{2\times}$ is 4.5 K, threshold is exceeded by ~30 years.

Schwartz, Charlson, Kahn, Ogren & Rodhe, J. Clim. 2010
Aerosols are removed from atmosphere in days to weeks.
Global temperature rapidly increases following removal of aerosol forcing.
Time constant for climate response to step-change in forcing is about 6 years.
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"THE CLIMATE PREDICTION PROBLEM"

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RESEARCH TEAM

"BETTER MODELS"
"THE CLIMATE PREDICTION PROBLEM"

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THE REAL PROBLEM

INADEQUATE KNOWLEDGE OF THE GLOBAL DISTRIBUTION OF AEROSOL PROPERTIES AND PROCESSES!