UNDERSTANDING THE INFLUENCES OF ATMOSPHERIC AEROSOLS ON CLIMATE AND CLIMATE CHANGE AND REPRESENTING THEM IN MODELS: A TALL ORDER

Stephen E. Schwartz

Challenges in Characterizing Small Particles
Exploring Particles from the Nano- to Microscales
Washington DC
October 25-26, 2010
www.ecd.bnl.gov/steve
OVERVIEW

Aerosol influences on climate and climate change

Earth’s energy balance and perturbations

Climate sensitivity – definition, importance, past and current estimates

Estimates of aerosol forcing and total forcing

Implications of current uncertainty in climate sensitivity

Determining climate sensitivity with models

Empirical determination of climate sensitivity

Determining aerosol forcing

Concluding remarks
Radiative Forcing by Tropospheric Aerosol

Partial Reflection and Absorption of Incoming Solar Radiation

Aerosol Haze

Clouds

Organics
Dust
SO₂
Soot
Sea salt
Organics
DMS

Land Use Changes
Industrial Emissions
Biomass Burning
Ocean
AEROSOL IN MEXICO CITY BASIN
AEROSOL IN MEXICO CITY BASIN

Light scattering by aerosols decreases absorption of solar radiation.
Fire plumes from southern Mexico transported north into Gulf of Mexico.
Aerosols from ship emissions enhance reflectivity of marine stratus.
GLOBAL ENERGY BALANCE
Global and annual average energy fluxes in watts per square meter

\[
1/4 S_0 \approx 254K
\]

\[
\alpha = 31\%
\]

\[
1/4 S_0 (1 - \alpha) = \sigma T^4
\]

Schwartz, 1996, modified from Ramanathan, 1987
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\(\alpha = 31\%\)

Stefan-Boltzmann radiation law

Schwartz, 1996, modified from Ramanathan, 1987
Greenhouse gas forcing is considered accurately known.
Gases are uniformly distributed; radiation transfer is well understood.
GLOBAL ENERGY BALANCE
Global and annual average energy fluxes in watts per square meter

\[ \frac{1}{4} S_0 \approx 254K \]

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

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

Schwartz, 1996, modified from Ramanathan, 1987
HOW MUCH WARMING IS EXPECTED?

Equilibrium change in global mean surface temperature = Climate sensitivity × Forcing

\[ \Delta T = S \times F \]

S is *equilibrium* sensitivity. Units: K/(W m\(^{-2}\))

Sensitivity is commonly expressed as “CO\(_2\) doubling temperature”

\[ \Delta T_{2\times} \equiv S \times F_{2\times} \]

where \( F_{2\times} \) is the “CO\(_2\) doubling forcing” ca. 3.7 W m\(^{-2}\).
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HOW MUCH WARMING IS EXPECTED?

For increases in CO₂, CH₄, N₂O, and CFCs over the industrial period

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

*Expected* temperature increase:

\[ \Delta T_{\text{exp}} = \frac{F}{F_{2\times}} \times \Delta T_{2\times} = \frac{2.6}{3.7} \times 3 \text{ K} = 2.1 \text{ K} \]

*Observed* temperature increase:

\[ \Delta T_{\text{obs}} = 0.8 \text{ K} \]
THE WARMING DISCREPANCY

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How can we account for this *warming discrepancy*?
Why Hasn’t Earth Warmed as Much as Expected?

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Brookhaven National Laboratory, Upton, New York

Robert J. Charlson
University of Washington, Seattle, Washington

Ralph A. Kahn
NASA Goddard Space Flight Center, Greenbelt, Maryland

John A. Ogren
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Henning Rodhe
Department of Meteorology, Stockholm University, Stockholm, Sweden
From Forcing by Long-lived Greenhouse Gases

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WHY HASN’T EARTH WARMED AS MUCH AS EXPECTED... FROM FORCING BY LONG-LIVED GREENHOUSE GASES?

- Uncertainty in greenhouse gas forcing.
- Countervailing natural cooling over the industrial period.
- Lag in reaching thermal equilibrium.
- Countervailing cooling forcing by aerosols.
- Climate sensitivity lower than current estimates.
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GLOBAL ENERGY BALANCE
Global and annual average energy fluxes in watts per square meter

\[ \Delta F = -1.2 \text{ W m}^{-2} \]
\[ \alpha = 31\% \]

\[ 106 \]
\[ 48 \]
\[ 27 \]
\[ 169 \]
\[ 296 \]
\[ 90 \]
\[ 16 \]

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

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

\[ 237 \approx 254K \]

\[ 343 \]

\[ 390 \approx 288K \]

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

\[ 68 \]

\[ \text{Latent heat} \]
\[ \text{Sensible heat} \]

\[ \text{Atmosphere} \]

Schwartz, 1996, modified from Ramanathan, 1987
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.
MONTHLY AVERAGE AEROSOL JUNE 1997
Polder radiometer on Adeos satellite

Optical Thickness $\tau$
$\lambda = 865$ nm

Ångström Exponent $\alpha$
$\alpha = -\frac{d \ln \tau}{d \ln \lambda}$

Small particles are from gas-to-particle conversion.
Cloud albedo is calculated for observed data and for average effective radius for each day. Forcing is calculated for indicated conditions relative to October 26.

<table>
<thead>
<tr>
<th>Date, 2000</th>
<th>Effective radius $r_e$, $\mu$m</th>
<th>Optical Depth</th>
<th>Net flux at TOA, W m$^{-2}$</th>
<th>Forcing relative to 10/26, W m$^{-2}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>10/26</td>
<td>10.2</td>
<td>15.1</td>
<td>293</td>
<td>—</td>
</tr>
<tr>
<td>10/21</td>
<td>7.8</td>
<td>20.8</td>
<td>266</td>
<td>27</td>
</tr>
<tr>
<td>02/18</td>
<td>5.8</td>
<td>28.3</td>
<td>240</td>
<td>53</td>
</tr>
</tbody>
</table>

*Kim, Schwartz, Miller, and Min, JGR, 2003*
CLIMATE FORCINGS OVER THE INDUSTRIAL PERIOD

Extracted from IPCC AR4 (2007)

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.
WHY HASN’T EARTH WARMED AS MUCH AS EXPECTED. . .

FROM FORCING BY LONG-LIVED GREENHOUSE GASES?

• Uncertainty in greenhouse gas forcing.
• Countervailing natural cooling over the industrial period.
• Lag in reaching thermal equilibrium.
• Countervailing cooling forcing by aerosols.
• Climate sensitivity lower than current estimates.
IMPLICATIONS

ALLOWABLE FUTURE CO$_2$ EMISSIONS

How much fossil carbon can be burned and emitted into the atmosphere (as CO$_2$) without exceeding a given threshold for “dangerous anthropogenic interference” with the climate system?

Answer depends on target threshold and climate sensitivity.

Premise of the calculation:

Forcings by LLGHG’s only; *no aerosol forcing.*

Result expressed as equivalent CO$_2$.
ALLOWABLE FUTURE CO₂ EMISSIONS

Dependence on $\Delta T_{2\times}$ for indicated values of maximum allowable temperature increase above preindustrial

For $\Delta T_{\text{max}} = 2$ K and IPCC AR4 best estimate $\Delta T_{2\times} = 3$ K, allowable future emissions are essentially zero.

For $\Delta T_{\text{max}} = 2$ K and IPCC “likely” range of $\Delta T_{2\times}$, 2 to 4.5 K allowable future emissions range from ~35 years at present emission rate to negative by like amount.
APPROACHES TO DETERMINING CLIMATE SENSITIVITY

Climate models

Evaluate by performance on current climate
Evaluate by performance over instrumental record

Empirical

Sensitivity = Time constant/Heat Capacity
Paleo: \( \frac{\Delta Temperature}{\Delta Flux} \), paleo to present
Instrumental record \( \frac{\Delta Temperature}{\text{(Forcing} - \text{Flux)}} \)
Satellite measmt.: \( [\frac{d(\text{Forcing} - \text{Flux})}{dT}\text{emperature}]^{-1} \)
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Simulations that incorporate anthropogenic forcings, including increasing greenhouse gas concentrations and the effects of aerosols, and that also incorporate natural external forcings provide a \textit{consistent explanation of the observed temperature record.}
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These simulations used models with different climate sensitivities, rates of ocean heat uptake and magnitudes and types of forcings.

IPCC AR4, 2007
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**How can this be?**

IPCC AR4, 2007
CORRELATION OF AEROSOL FORCING, TOTAL FORCING, AND SENSITIVITY IN CLIMATE MODELS

Nine coupled ocean-atmosphere models; two energy balance models

\[
S = \frac{\Delta T}{F}
\]

\[
F = \Delta T S^{-1}
\]

Total forcing is linearly correlated with inverse sensitivities of the models. Climate models with lower sensitivity (higher inverse sensitivity) employed a greater total forcing.

Slope (0.8 K) is approximately equal to observed temperature change. Models accurately reproduce known temperature change.

Modified from Kiehl, GRL, 2007
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Greater total forcing is due to smaller (less negative) aerosol forcing.

Modified from Kiehl, GRL, 2007
APPROACHES TO DETERMINING CLIMATE SENSITIVITY

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Evaluate by performance on current climate
Evaluate by performance over instrumental record

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Paleo: $\Delta Temperature/\Delta Flux$, paleo to present
Instrumental record $\Delta Temperature/(Forcing – Flux)$
Satellite measmt.: $[d(Forcing – Flux)/dTemperature]^{-1}$
From known temperature change, forcing, and heating rate

\[ F_{\text{eff}} = F - H \]

\[ \Delta T = S F_{\text{eff}} \]

\[ F_{\text{eff}} = \Delta T S^{-1} \]

Known effective forcing intersecting with known temperature increase permits empirical determination of inverse sensitivity.
EMPIRICAL DETERMINATION OF CLIMATE SENSITIVITY

Effect of uncertainty in forcing

\[ F_{\text{eff}} = F - H \]
\[ \Delta T = SF_{\text{eff}} \]
\[ F_{\text{eff}} = \Delta TS^{-1} \]

Present uncertainty in aerosol forcing precludes precise determination of climate sensitivity from temperature increase over industrial period.
CLIMATE MODEL DETERMINATION OF CLIMATE SENSITIVITY

Effect of uncertainty in forcing

\[ F_{\text{eff}} = F - H \]
\[ \Delta T = S F_{\text{eff}} \]
\[ F_{\text{eff}} = \Delta TS^{-1} \]

Uncertainty in aerosol forcing allows climate models with widely differing sensitivities to reproduce temperature increase over industrial period.
Climate sensitivity and aerosol forcing are *intrinsically coupled*, in climate models and in empirical determination of sensitivity.

Confident determination of climate sensitivity requires *great reduction in uncertainty in aerosol forcing* over the industrial period.
THE PATH FORWARD
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Determine aerosol forcing with high accuracy.
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Determine aerosol forcing with high accuracy.

Multiple approaches are required:

- **Laboratory studies** of aerosol processes.
- **Field measurements** of aerosol processes and properties: emissions, new particle formation, evolution, size distributed composition, optical properties, CCN properties, removal processes . . .

Represent aerosol processes in *chemical transport models*.

Evaluate models by *comparison with observations*.

*Satellite measurements* for spatial coverage.

Calculate forcings in *chemical transport models and GCMs*. 
AEROSOL PROCESSES THAT MUST BE UNDERSTOOD AND REPRESENTED IN MODELS

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

Modeling aerosol processes requires understanding these processes, developing and testing their numerical representations, and incorporating these representations in global scale models.
Surface measurements: AERONET network.
Satellite measurements: composite from multiple instruments/platforms.
Are the models getting the “right” answer for the wrong reason?
Are the models getting the “right” answer because the answer is known?
Are the satellites getting the “right” answer because the answer is known?
AEROSOL OPTICAL DEPTH IN 17 MODELS (AEROCOM)

Comparison also with surface and satellite observations

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SEA SALT AEROSOL MASS EMISSIONS

Annual average in AEROCOM models; $10^{12}$ kg yr$^{-1}$

Range of global annual mean is a factor of 50.

Textor et al., ACP 2006; courtesy Michael Schulz

http://dataipsl.ipsl.jussieu.fr/cgi-bin/AEROCOM/aerocom/aerocom_work_annualrs.pl
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ORGANIC CONTRIBUTIONS TO TROPOSPHERIC AEROSOL

Mass-spec determination of primary vs secondary organics

New analytical techniques permit identification of formation mechanisms.

Evolution of Organic Aerosols in the Atmosphere
AEROSOL INTERACTIONS WITH MAJOR ATMOSPHERIC CYCLES

Aerosol processes are central to research in the Department of Energy Atmospheric System Research Program.
SECONDARY AEROSOL PRODUCTION
Eight aircraft flights above and downwind of Mexico City, March 2006

Parcel photochemical age measured using $-\log(\text{NO}_x/\text{NO}_y)$ as clock.
Aerosol normalized to CO above background to account for dilution.

*Fivefold increase* in organic aerosol.
Measured increase in organic aerosol exceeds modeled based on laboratory experiments and measured volatile organic carbon *tenfold.*
AEROSOL TRANSPORT AND EVOLUTION
Mexico City, March 22, 2006

FLEXPART age and back trajectories

Component ID by scanning transmission electron microscopy

Number fraction and number size distribution

Moffet, Tivanski, Hopkins, Desyaterik, Fast, Barnard, Laskin, Gilles; PNNL, LBL
NEW MEASUREMENTS YIELD NEW INSIGHTS

Atmospheric Nucleation Event: 09/16/10

- \( N_{1-6 \text{ nm}} \sim 100X N_{10-500 \text{ nm}} \)
- Highly dynamic \( \rightarrow \) need size/time dep. meas.

See poster of Chongai Kuang
NEW MEASUREMENTS YIELD NEW INSIGHTS

Atmospheric Nucleation Event: 09/16/10

- conventional measurements [GROWTH]
- new sub 6 nm measurements [NUCLEATION]

- \( N_{1-6 \text{ nm}} \approx 100X N_{10-500 \text{ nm}} \)
- Highly dynamic \( \rightarrow \) need size/time dep. meas.

See poster of Chongai Kuang
Critical supersaturation for cloud drop formation increases with decreasing particle size.
Critical supersaturation in maritime air is consistent with seasalt composition.
Continental influence (off California coast) increases critical supersaturation for a given particle size.

Change in supersaturation shows dependence on composition.
Particles above cloud layer showed greater increase in supersaturation than particles below cloud.
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Composition measured with PILS (particle into liquid sampler) showed high organic fraction in above cloud aerosol.
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Measurements with aerosol mass spectrometer showed organic material in CCN size range.
LABORATORY STUDIES

Aerosol flow reactor in Finlayson-Pitts lab, UC Irvine
PRODUCT CHARACTERIZATION IN NO₃ – α-PINENE REACTION

**IR spectroscopy**

![IR spectroscopy chart](image1)

**HPLC**

![HPLC chart](image2)

**Aerosol mass spectrometry**

![Aerosol mass spectrometry chart](image3)

**High-resolution AMS**

![High-resolution AMS chart](image4)

**Single Particle Laser Ablation Time-of-flight mass Spec (SPLAT)**

![SPLAT chart](image5)

Finlayson-Pitts, UC Irvine
AEROSOL PARTICLE NUMBER CONCENTRATION

Average particle number concentrations North America, July 2004

Aitken mode particles \((D \leq 100 \text{ nm})\)

Accumulation mode particles \((D \geq 100 \text{ nm})\)

Strong dependence on new particle formation mechanism

Strong dependence on size of primary emissions

Accurate representation of number concentrations and aerosol indirect effects requires improved knowledge of \textit{new particle formation rate} and \textit{size distributed emissions}.

\textit{Chang, Schwartz, McGraw & Lewis, JGR, 2009}
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*Measurement based determination of aerosol forcings.*
DIRECT DETERMINATION OF AEROSOL FORCINGS AT ARM SITES

Measurements 24-7-365

Drone
Net SW and LW at TOA

3-D Characterization of Aerosol and Cloud Properties

Characterization of 3-D Cloud Properties by Radars, Tomography

~50 km

Scanning Cloud Radars

AMF

ARM Central Facility

Radiometers

Sunphotometers
CONCLUDING OBSERVATIONS

• Radiative forcing by incremental greenhouse gases *already in the atmosphere* could potentially lead to dangerous interference with the climate system.

• Within present uncertainty of climate sensitivity, allowable future emissions range from about $-30$ years to $+30$ years.

• Climate sensitivity must be known with much greater accuracy for effective developing energy strategies.

• Atmospheric aerosols offset an unknown fraction of the warming forcing of incremental greenhouse gases.

• *The present uncertainty in aerosol forcing greatly limits accuracy in determining climate sensitivity.*

• *Much fundamental aerosol research is essential and urgent.*