Regional Aerosol Effects on the Atmospheric Radiation Balance

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Works cited


Acknowledgments

Office of Naval Research Young Investigator Program
National Science Foundation CAREER Award
Satellite observations: Polluted aerosol effects on clouds

- Colorized AVHRR satellite images of
  - A) Turkey (Istanbul, Izmit, Bursa)
  - B) Manitoba, Canada (Hudson Smelting)
  - C) South Australia (Augusta, Pririe, Adelaide, refineries)

- Yellow indicates polluted clouds
  - Smaller drops -- polluted (yellow)
  - Large drops -- clean (purple)

- Rosenfeld, *Science*, 2000
Aerosol Satellite Observations

- 3.7µm channel shows cloud reflectance for high cirrus and low stratus (P. Durkee)

<table>
<thead>
<tr>
<th>Ship Track Observations</th>
<th>Remote/Optical</th>
<th>In situ/Aerosol</th>
</tr>
</thead>
<tbody>
<tr>
<td>Conover</td>
<td>1966</td>
<td>↑ albedo = 20%</td>
</tr>
<tr>
<td>Coakley, Bernstein, Durkee</td>
<td>1987</td>
<td>↑ R_{3.7\mu m} = 3.9%</td>
</tr>
<tr>
<td></td>
<td></td>
<td>↑ R_{0.63\mu m} = 1.6%</td>
</tr>
<tr>
<td></td>
<td></td>
<td>R_{11\mu m} = 0.0%</td>
</tr>
<tr>
<td>Radke, Coakley, King</td>
<td>1989</td>
<td>↑ R_{0.63\mu m} = 13.6%</td>
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<tr>
<td></td>
<td></td>
<td>↑ \tau = 260%</td>
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<tr>
<td>King, Radke, Hobbs</td>
<td>1993</td>
<td>↓ r_e = 21%</td>
</tr>
<tr>
<td></td>
<td></td>
<td>↑ I_{τ-1}(0.74\mu m) = 220%</td>
</tr>
<tr>
<td></td>
<td></td>
<td>↓ I_{τ-1}(2.20\mu m) = 87%</td>
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<tr>
<td></td>
<td></td>
<td>↑ N_{drop} = 220%</td>
</tr>
<tr>
<td></td>
<td></td>
<td>↑ CN = 250%</td>
</tr>
<tr>
<td></td>
<td></td>
<td>↑ LWC = 250%</td>
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</table>
Ship track formation processes

**Hypothesis**

<table>
<thead>
<tr>
<th>Author</th>
<th>Year</th>
<th>Hypothesis</th>
</tr>
</thead>
<tbody>
<tr>
<td>Radke, Coakley, King</td>
<td>1989</td>
<td>ship stacks ⇒ ↑CCN</td>
</tr>
<tr>
<td>Albrecht</td>
<td>1989</td>
<td>↓N_{precip} ⇒ ↑LWC</td>
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<tr>
<td>Hudson</td>
<td>1991</td>
<td>↓CCN ⇒ ↑N_{precip}</td>
</tr>
<tr>
<td>Ackerman, Toon, Hobbs</td>
<td>1994</td>
<td>↓CCN ⇒ ↓h, ↓τ</td>
</tr>
</tbody>
</table>

*Nature 400, 713 - 714 (1999) BARRY J. HUEBERT*
Ships are an important source of aerosol

Nature 400, 743 - 746 (1999)
KEVIN CAPALDO*, JAMES J. CORBETT†, PRASAD KASIBHATLA‡, PAUL FISCHBECK†§ & SPYROS N. PANDIS*†
Outline

- **Radiative transfer framework**
- **Ship tracks: “experiment” and “control”**
  - The “indirect effect” of aerosols on clouds
  - Satellite and *in-situ* observations
- **Cloud drops form on aerosol nuclei**
  - Detailed microphysical model provides an accurate estimate of aerosol-cloud interaction
  - Externally-mixed model components show role of plume particles and SO$_2$ vapors
  - Organic/water/salt VLE is used to study water uptake
- **Enhanced drop numbers increase cloud albedo**
  - Effect on cloud albedo is underpredicted by the so-called “Twomey effect”
  - Variations in liquid water content and particle composition affect the result
- **Explicit climate response calculation (???)**
Atmospheric Radiation Balance

\[ F_s = 0.25 \times S_0 (1 - R_p) \]

Incoming solar radiation

100 energy units

19
Absorbed by \( H_2O, O_3, \) dust

8
Back-scattered by air

17
Absorbed by clouds

6
Outgoing shortwave radiation (albedo)

Absorbed by clouds

115
7
Reflected by surface

Sensible heat flux

115
106
Absorbed by clouds

Emission by clouds

46
Absorbed by surface

9
Outgoing longwave radiation

40
20
Net emission by \( CO_2, O_3, H_2O \)

Absorption by clouds, \( H_2O \) vapor, \( CO_2, O_3 \)

F \(_L\) = \( \sigma T_e^4 \)

Outgoing longwave radiation

Emission by clouds

7
Latent heat flux

24
Global Radiative Equilibrium

\[ F_{\text{net}} = F_s - F_L \]

\[ F_{\text{net}} = 0 \]

\[ T_e = \left( \frac{(1 - R_p)S_0}{4\sigma} \right)^{\frac{1}{4}} \]

- The net flux of radiation \( (F_{\text{net}}) \) is given by the difference between the incoming shortwave flux from the sun and the outgoing longwave flux of infrared radiation.

- As albedo \( (R_p) \) increases, the equilibrium temperature of the earth \( (T_e) \) decreases.

- Climate sensitivity (the response of global circulation to the change in radiative forcing) may diminish or enhance this effect as a result of feedback effects on weather patterns.
The "Twomey Effect"

- Twomey et al. (1984) proposed a correlation between albedo and "pollution level"
Ship Track Observations

- Radke et al. (1989) measured in situ higher droplet numbers and liquid water concentrations in track than in surrounding clouds.
Albedo (\(R_p\)) - average global reflectance, which is calculated from the cloud fraction times the reflectance of each cloud corrected by transmittance in reaching the cloud

AVHRR - Advanced Very High Resolution Reflectance Satellite launched by NOAA (the National Oceanic and Atmospheric Administration) for weather monitoring and prediction

Effective Radius (\(R_{\text{eff}}\)) - the optically-active characteristic particle radius, which is proportional to the ratio of the liquid water over the optical depth; stratus clouds have 2\(\mu\)m\(<R_{\text{eff}}<20\mu\)m, raindrops fall at \(R_{\text{eff}}\sim20\mu\)m

Liquid Water Content (LWC) - mass loading of the most common liquid in the atmosphere, water; for scale, LWC\(~0.5\) g m\(^{-3}\) is typical for a thin, “wispy” stratus cloud, but serious storm clouds (called “cumulus”) have LWC\(>1.0\) g m\(^{-3}\)

Optical Depth (\(\tau\)) - the sum of absorption plus scattering (also known as “extinction”) of light integrated over the distance it travels; an optical depth of \(~0.05\) is typical of a clean troposphere, but a smoky forest fire might have a plume with optical depth near 1.0

Supersaturation (\(S_c\)) - the percentage by which the ambient vapor pressure (of water) exceeds the saturation pressure at the ambient temperature
• “I don’t know how clouds form, but the clouds know how to do it, and that is the important thing”
  – a 5th grader in Texas
Ship track formation in stratus clouds

Dynamic eddies and updrafts (w)
Thermodynamic structure (dT/dz)
Marine aerosol

Same eddies and updrafts (w)
Same thermodynamic structure (dT/dz)
Polluted aerosol

Marine aerosol

0.1 10
Diameter (µm)

0.1 10
Diameter (µm)
Cloud Thermodynamic Structure

- Lapse rate (-dT/dz) is measured
- Total Water Content (TWC) is measured
- Updraft Velocity (w) is *estimated*

<table>
<thead>
<tr>
<th></th>
<th>Clean</th>
<th>Continentally-Influenced</th>
</tr>
</thead>
<tbody>
<tr>
<td>dT/dz</td>
<td>-6.5 K/km</td>
<td>-6.1 K/km</td>
</tr>
<tr>
<td>TWC</td>
<td>10.25 g/m³</td>
<td>11.39 g/m³</td>
</tr>
<tr>
<td>w</td>
<td>0.3 m/s</td>
<td>0.3 m/s</td>
</tr>
</tbody>
</table>

**Profile**

![Clean Cloud Thermodynamic Structure](image1)

![Continentally-Influenced Cloud Thermodynamic Structure](image2)
Aerosol Dynamics Model

- Based on Russell and Seinfeld (1998)
- Sectional with “virtual” water allows accurate evaporation
  - Jacobson et al., 1994
- Number and mass conserved
  - Tzivion et al., 1987
- Multi-population tracks external mixtures
- Multi-component tracks internal mixtures
- Classical binary nucleation
  - Kulmala and Laaksonen, 1990
  - Coffman and Hegg, 1995
Particle Growth in Populations with Fixed-Grid Bins

Particle Number Sources and Sinks

\[
\frac{\partial N_{ijk}(t)}{\partial t} = J_{ik}^{\text{nucl}} + J_{ik}^{\text{flux}} - K_{ik}^{\text{depn}} N_{p_{ik}} + J_{ijk}^{\text{grow}} + \frac{1}{2} \sum_{i_1 \leq i_2} \sum_{k_1 \leq k_2} K_{i_1 j_1}^{\text{coag}} N_{p_{i_1 k_1}} N_{p_{i_2 k_2}}
\]

\[
- \sum_{i_1 \leq i_{\text{max}}} \sum_{k_1 \leq k_{\text{max}}} K_{i_1 i}^{\text{coag}} N_{p_{i_1 k_1}} N_{p_{i k}}
\]

Particle Mass Sources and Sinks

\[
\frac{\partial M_{p_{ik}}(t)}{\partial t} = \sum_j \frac{M_{p_{ik}}}{M_{p_{jk}}} m_{ik} J_{ik}^{\text{nucl}} + \sum_j \frac{M_{p_{jk}}}{M_{p_{ik}}} m_{jk} J_{jk}^{\text{flux}} - \sum_j \frac{M_{p_{ik}}}{M_{p_{jk}}} m_{jk} K_{ik}^{\text{depn}} N_{p_{jk}}
\]

\[
+ \sum_j \frac{M_{p_{ik}}}{M_{p_{jk}}} m_{ik} J_{ik}^{\text{grow}} + \frac{2\pi D_j D_{p_{i k}}^{\text{amb}} F(K n_{i k}) A(K n_{i k}) (P_{j'} - P_{i j}^{\text{surf}})}{2}
\]

\[
+ \frac{1}{2} \sum_j \frac{M_{p_{ik}}}{M_{p_{jk}}} m_{ik} \sum_{i_1 \leq i_2} \sum_{k_1 \leq k_2} K_{i_1 i_2}^{\text{coag}} N_{p_{i_1 k_1}} N_{p_{i_2 k_2}} - \sum_j \frac{M_{p_{ik}}}{M_{p_{jk}}} m_{jk} \sum_{i_1 \leq i_{\text{max}}} \sum_{k_1 \leq k_{\text{max}}} K_{i_1 i}^{\text{coag}} N_{p_{i_1 k_1}} N_{p_{j k}}
\]
Ship Track in “Clean” Air

[Map showing ship tracks]

C131

Star Livorno
Comparison of Background and Track Supersaturation Profiles

- Lapse rate, updraft velocity, and total water content are identical
- Lower maximum supersaturation in track
- Higher cloud-average liquid water content in track

![Graph showing comparison of background and track supersaturation profiles.](image)

- Background LWC: \( \text{LWC}_{\text{Cloud}} = 0.385 \)
- Track LWC: \( \text{LWC}_{\text{Cloud}} = 0.412 \)
- Background RH: \( S_{\text{max}} = 0.91\% \)
- Track RH: \( S_{\text{max}} = 0.22\% \)
Role of updraft velocity in particle activation

- Decreasing updraft velocity lowers the maximum supersaturation reached.
Clean Conditions

Background

Track

Below Cloud

In Cloud
No Gas-to-Particle Growth

- Is condensation of $SO_2$ and its products needed to form CCN?
  - few plume particles activate in the absence of condensational growth
Ship Track in “Continently-Influenced” Air
Comparison of Background and Track Supersaturation Profiles

- Lapse rate, updraft velocity, and total water content are identical
- Lower maximum supersaturation in track
- Higher cloud-average liquid water content in track
Comparison of Tracks

- Clean case has larger decrease in effective radius than continentally-influence case

<table>
<thead>
<tr>
<th></th>
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<th>Continentally-Influenced</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Background</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$R_{\text{eff}}$</td>
<td>10.8 $\mu$m</td>
<td>5.7 $\mu$m</td>
</tr>
<tr>
<td>LWC</td>
<td>0.19 g/m$^3$</td>
<td>0.22 g/m$^3$</td>
</tr>
<tr>
<td>$S_{\text{max}}$</td>
<td>0.68%</td>
<td>0.26%</td>
</tr>
<tr>
<td>$N_d$</td>
<td>53 cm$^{-3}$</td>
<td>293 cm$^{-3}$</td>
</tr>
<tr>
<td><strong>Track</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$R_{\text{eff}}$</td>
<td>3.4 $\mu$m</td>
<td>4.1 $\mu$m</td>
</tr>
<tr>
<td>LWC</td>
<td>0.20 g/m$^3$</td>
<td>0.22 g/m$^3$</td>
</tr>
<tr>
<td>$S_{\text{max}}$</td>
<td>0.18%</td>
<td>0.15%</td>
</tr>
<tr>
<td>$N_d$</td>
<td>2130 cm$^{-3}$</td>
<td>985 cm$^{-3}$</td>
</tr>
</tbody>
</table>
Microphysical Sensitivity

Effective Radius ($\mu$m)

Liquid Water Content (g m\(^{-3}\))

Droplet (cm\(^{-3}\))

Supersaturation (%)

-0.1 0.1 0.2 0.3 0.4 0.5 0.6 0.7 0.8 0.9 1.0

Updraft Velocity

Updraft Area

Plume Emissions

Clean Marine Track
Clean Marine Background
Continental Track
Continental Background
Sensitivity to Chemical Composition

- Water uptake (and efficiency as CCN) is determined by particle composition

\[ RH = \frac{P_w}{P_{w,\text{sat}}} = x_w \gamma_w \exp \left[ \frac{4M_w \sigma_w}{RT \rho_w D_p} \right] \]

where

\[ \gamma_w, \rho_w, \sigma_w = \text{fcn}^s(P, T, x_w, x_i, \ldots x_n) \]

- Seawater consists of water plus
  - NaCl (77% to 42% of solute mass)
  - Other inorganic ions (13% to 8% of solute mass)
  - Organic compounds (10% to 50% of solute mass; Middlebrook et al., 1998; Gagosian et al., 1981)
    - Malic acid, citric acid, amino acids: ~70%
    - Monosaccharides, polysaccharides: ~16.5%
    - Alkanes, fatty acids, alcohols: ~13.5%
Hygroscopic Growth of Particles with Organic Species

Hygroscopic Growth Factor = \frac{\text{Diameter at Ambient RH}}{\text{Diameter at Dry RH (<40%)}}

Ming and Russell, 2000
Comparison of Predicted Drop Number to Parameterizations

- Parameterizations use empirical data sets to fit a relationship between droplet and particle numbers of unknown composition.
Aerosol Scattering Model (Erlick et al.)

- **Mie scattering algorithm** (Bohren and Huffman, 1983)
  - Linear-by-volume mixing for non-absorbing species
  - Maxwell-Garnett mixing for absorbing species

- **Radiative transfer algorithm** (Friedenreich and Ramaswamy, 1999)
  - Solar parameterization for inhomogeneous scattering and absorbing atmospheres
  - Developed from Line-by-Line reference computations for clouds and water vapor
  - Exponential sum-fit technique for water vapor
  - Surface albedo from Taylor et al., 1996
Clean Marine Case Absorption and Reflectance

![Graphs showing ambient and track cloud absorption and albedo with wavelength on the x-axis and absorption and albedo on the y-axis.](a) JDT178a

- Ambient Cloud Absorption (W m$^{-2}$)
- Track Cloud Absorption (W m$^{-2}$)
- Albedo

- Ambient Cloud Absorption (37.7 W m$^{-2}$ total difference)
- In cloud absorption (65.8 W m$^{-2}$ total difference)
- Ambient cloud albedo

- Above track absorption (44.3 W m$^{-2}$ total difference)
- In track absorption (97.4 W m$^{-2}$ total difference)
- Track albedo

Wavelength (µm)
Predicted and observed albedos

Clean marine case

Continentally-influenced case
Optical depth dependence on particle size and composition

Clean marine case
Background

Track

Continently-influenced case
Background

Track
# Clean Marine Case

## Radiative Sensitivity

<table>
<thead>
<tr>
<th>Albedo</th>
<th>0.2</th>
<th>0.3</th>
<th>0.5</th>
<th>45%</th>
<th>55%</th>
<th>65%</th>
<th>Comp. Fluxes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Updraft Velocity</td>
<td>Background</td>
<td>Track</td>
<td>Background</td>
<td>Track</td>
<td>Background</td>
<td>Track</td>
<td>Background</td>
</tr>
<tr>
<td>0.2</td>
<td>0.2</td>
<td>0.3</td>
<td>0.5</td>
<td>0.6</td>
<td>0.7</td>
<td>0.8</td>
<td>0.9</td>
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## Optical Depth

<table>
<thead>
<tr>
<th>Optical Depth</th>
<th>0.2</th>
<th>0.3</th>
<th>0.5</th>
<th>45%</th>
<th>55%</th>
<th>65%</th>
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<td>Background</td>
<td>Track</td>
<td>Background</td>
<td>Track</td>
<td>Background</td>
</tr>
<tr>
<td>0.2</td>
<td>0.6</td>
<td>0.3</td>
<td>0.5</td>
<td>0.4</td>
<td>0.2</td>
<td>0.1</td>
<td>0.0</td>
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</table>
Predicted and parameterized albedos

Clean marine case

Continentally-influenced case
Model Evaluation

• **Aerosol observations**
  - Ship tracks provide a good example of the effect of pollution in perturbing cloud properties
  - Model and observations agree within 10% for albedo predictions (changes are within 5%)

• **Aerosol evolution processes**
  - Detailed microphysical predictions of kinetic activation of cloud droplet exceeded literature parameterizations by 20% to 50%
  - Droplet activation varies almost 50% with updraft velocity in polluted cases, less than 5% in clean cases
  - Externally-mixed model shows role of smog particles in modifying clean clouds, resulting in more than 3 times the drop number, 50% decrease in effective radius, and 30% reduction in effective supersaturation

• **Aerosol radiative effects**
  - Changes in albedo exceeded Twomey’s estimates by up to ~20%
Key Uncertainties

• **Aerosol observations**
  – Updraft velocity and entrainment
  – Droplet vertical distribution
  – Longer (>3 hr) time history of evolution
  – Particle composition (especially black carbon fraction)
  – Particle shape (and alignment)
  – Emissions (!) and plume dilution

• **Aerosol evolution processes**
  – Aerosol droplet activation
  – Hygroscopic growth of particles
  – Microscale dynamics and eddies
  – Supersaturation profile and entrainment

• **Aerosol radiative effects**
  – Optical properties of organic particles (especially mixtures and organics)
  – Scattering of asymmetric particles
Estimated radiative effects of aerosols

- The Report of the Intergovernmental Panel on Climate Change (1995) summarized the uncertainty associated with the cooling of aerosols by the indirect effect.
Climate Forcing Framework

Emissions (g/m³)

Aerosol Forcing $\Delta F$ (W/m²)

Climate Response $\Delta T = k \times \Delta F$

Global Temperature Change $\Delta T$ (K)
Explicit Climate Response Framework

Emissions (g/m³)

explicit Climate Response

regional Temperature Change \( \Delta T \) (K)
Acknowledgements

• Carynelisa Erlick,
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  Princeton University

• Venkatachalam Ramaswamy,
  Geophysical Fluid Dynamics Laboratory,
  National Oceanic and Atmospheric Administration

• Yi Ming,
  Department of Chemical Engineering,
  Princeton University
Continental Case Absorption and Reflectance

Ambient Cloud Absorption (W m⁻²)

Track Cloud Absorption (W m⁻²)

Ambient Cloud Albedo

Track Cloud Albedo

Wavelength (µm)
Continental Case
Radiative Sensitivity

Albedo

Optical Depth
Secondary Particle Sources and Light Scattering

WITHOUT AEROSOL

no particles

WITH AEROSOL

particles

"sun"

"atmosphere"

"polluted atmosphere"

"terpenes"

"photochemical smog"

\( + O_3 \)
Clouds nucleate better on some particles than others

**Beer**
- Liquid (H₂O/EtOH) supersaturated with vapor (CO₂) nucleates on salt (but not pepper) to form bubbles

**Clouds**
- Vapor (air) supersaturated with liquid (H₂O) nucleates on hygroscopic particles (but not hydrophobic ones) to form droplets

Bohren, 1987
Mass- and Number-Conserving Growth

- **Conservation of Mass**
  \[
  M_{p_{jk}}(t) + M_{p_{(i+1)jk}}(t) + \Delta t \left( \frac{\partial M_{p_{jk}}(t)}{\partial t} + \frac{\partial M_{p_{(i+1)jk}}(t)}{\partial t} \right) = M_{p_{jk}}(t + \Delta t) + M_{p_{(i+1)jk}}(t + \Delta t),
  \]

- **Conservation of Number**
  \[
  N_{p_{ik}}(t) + N_{p_{(i+1)k}}(t) + \Delta t \left( \frac{\partial N_{p_{ik}}(t)}{\partial t} + \frac{\partial N_{p_{(i+1)k}}(t)}{\partial t} \right) = N_{p_{ik}}(t + \Delta t) + N_{p_{(i+1)k}}(t + \Delta t),
  \]

- **Fixed Mean Diameter**
  \[
  \frac{D_{p_{(i+1)}{\text{dry}}}}{D_{p_{i}{\text{dry}}}} = \text{constant}
  \]

**Variables**

- \(D_{p_{ik}}\)  equivalent dry diameter of section \(i\) in population \(k\)
- \(N_{p_{ik}}\)  number of particles in section \(i\) in population \(k\)
- \(M_{p_{ijk}}\)  particulate mass of species \(j\) in section \(i\) in population \(k\)
Water Condensation

- **Fixed grid (for solutes)**

  ![Diagram of fixed grid](image)

- **Moving grid (for water)**

  ![Diagram of moving grid](image)

![Graph showing water condensation](image)
Continently-Influenced Conditions

**Background**

**Track**

In Cloud

Below Cloud