Thunderstorms and atmospheric composition:
A meeting of cloud physics, dynamics, lightning and chemistry

Mary C. Barth

Atmospheric Chemistry Observations and Modeling
Mesoscale and Microscale Meteorology
National Center for Atmospheric Research, Boulder, Colorado
Thunderstorms and atmospheric composition: A meeting of cloud physics, dynamics, lightning and chemistry

Deep Convective Clouds and Chemistry (DC3) Field Project
- Supported by NSF, NASA, DLR
- DC3 co-Principal Investigators: Chris Cantrell, Steve Rutledge, Bill Brune, Jim Crawford, Heidi Huntrieser (DLR)
- DC3 Science Team and Logistical Support

Nitrogen Oxides ($\text{NO}_x =\text{NO} + \text{NO}_2$)
Volatile Organic Compounds (VOCs)
Ozone ($\text{O}_3$)
A meeting of cloud physics, dynamics, lightning and chemistry

Important ozone precursors that are soluble:
CH$_2$O = formaldehyde
H$_2$O$_2$ = hydrogen peroxide
CH$_3$OOH = methyl hydrogen peroxide
A meeting of cloud physics, dynamics, lightning and chemistry

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CH$_2$O, H$_2$O$_2$, CH$_3$OOH, and other soluble trace gases and aerosols

Photochemically-active plumes
CH$_2$O, H$_2$O$_2$, CH$_3$OOH, other VOCs, NO$_x$
\[ \rightarrow \] O$_3$  GHG in Upper Troposphere

Dynamics
Transport
Wet deposition
A meeting of cloud physics, dynamics, lightning and chemistry

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- CH$_2$O = formaldehyde
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Photochemically-active plumes:
- CH$_2$O, H$_2$O$_2$, CH$_3$OOH, other VOCs, NO$_x$
- O$_3$ (GHG in Upper Troposphere)

Lightning-production of NO$_x$

Wet deposition

Transport
Deep Convective Clouds and Chemistry (DC3) Field Experiment
Aimed to Learn How Thunderstorms Affect the Composition of the Troposphere

Sampled Storms in
- Northeast Colorado
- West Texas to Central Oklahoma
- Northern Alabama

- Weather balloons to characterize storm environment
- Radars and Lightning Mapping Arrays to characterize storm and lightning
- Aircraft to characterize composition in the inflow and outflow regions of storm

Overview Paper: Barth et al. (2015) BAMS
DC3 Special Issue in JGR
Example of DC3 Experimental Design

June 22, 2012
NE Colorado / SW Nebraska storm

GV and DC-8 aircraft sampled 3 isolated severe convective storms

Two storms were side-by-side, with one ingesting the High Park smoke plume

Smoke plume was at ~7 km altitude

Aircraft sampled smoke plume just to west of storms AND in convective outflow

Photo by Armin Sorooshian; Annotated satellite photo from Apel et al. (2015)
22 June 2012 DC3 Case

NEXRAD Composite Reflectivity valid 20120622T1950Z

Cameron R. Homeyer, NCAR (chomeyer@ucar.edu)
Sample PBL Air South of Both Storms and Outflow of First Storm
Sample PBL Just South of North Storm and Outflow of North Storm

NEXRAD Composite Reflectivity valid 20120622T2335Z

DC-8 in PBL

GV in outflow

Cameron R. Homeyer, NCAR (chomeyer@ucar.edu)
Sample Outflow of Both Storms

NEXRAD Composite Reflectivity valid 20120623T0130Z

DC-8 in outflow
GV in outflow

Cameron R. Homeyer, NCAR (chomeyer@ucar.edu)
Cloud Physics Effects on Soluble Trace Gases

Solubility of CH$_2$O, H$_2$O$_2$, CH$_3$OOH

H$_2$O$_2$: very soluble
CH$_2$O: moderately soluble
CH$_3$OOH: mildly soluble

CH$_2$O, H$_2$O$_2$, CH$_3$OOH and other soluble trace gases and aerosols

Wet deposition

Photochemically-active plumes
CH$_2$O, H$_2$O$_2$, CH$_3$OOH, other VOCs, NO$_x$ → O$_3$

CH$_2$O, H$_2$O$_2$, CH$_3$OOH, NO$_x$, O$_3$
1. Find entrainment rate into storm from surrounding environment
2. Use entrainment rate to determine amount of soluble trace gas transported to top of storm
3. Compare measured mixing ratio in outflow to estimated value transported to top of storm
   → Scavenging Efficiency
Scavenging Efficiencies
Six storms analyzed from Oklahoma and northeast Colorado regions
Barth et al. (2016), *Fried et al. (2016) JGR

<table>
<thead>
<tr>
<th>Date</th>
<th>Location</th>
<th>H$_2$O$_2$</th>
<th>*CH$_2$O</th>
<th>CH$_3$OOH</th>
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</thead>
<tbody>
<tr>
<td>18 May 2012</td>
<td>Colorado</td>
<td>84% ± 23%</td>
<td></td>
<td>68% ± 28%</td>
</tr>
<tr>
<td>29 May 2012</td>
<td>Oklahoma</td>
<td>88% ± 11%</td>
<td>51% ± 6%</td>
<td>77% ± 20%</td>
</tr>
<tr>
<td>02 June 2012</td>
<td>Colorado</td>
<td>94% ± 13%</td>
<td></td>
<td>12% ± 67%</td>
</tr>
<tr>
<td>06 June 2012</td>
<td>Colorado</td>
<td>97% ± 5%</td>
<td>50% ± 7%</td>
<td>84% ± 15%</td>
</tr>
<tr>
<td>16 June 2012</td>
<td>Oklahoma</td>
<td>89% ± 15%</td>
<td></td>
<td>30% ± 50%</td>
</tr>
<tr>
<td>22 June 2012</td>
<td>Colorado</td>
<td>79% ± 19%</td>
<td>38% ± 23%</td>
<td>44% ± 47%</td>
</tr>
<tr>
<td>Previous Studies</td>
<td></td>
<td>55-90%</td>
<td>11-57%</td>
<td>7% or less</td>
</tr>
</tbody>
</table>

- >80% removal of H$_2$O$_2$
- 12-84% removal of CH$_3$OOH
- Large uncertainties

Why would the less soluble CH$_3$OOH have scavenging efficiencies as high as the very soluble H$_2$O$_2$?
Retention of Dissolved Trace Gases in Freezing Drops

When cloud drops freeze, what happens to trace gases dissolved in those drops?

1) They stay in the frozen drop
2) They go back to the gas phase
3) They are partially retained in the frozen particles
Retention of Methyl Hydrogen Peroxide in Freezing Drops

WRF-Chem sensitivity simulations
1) Retention factor = 1.0
2) Retention factor = 0.5
3) Retention factor = 0.25
4) Retention factor = 0.0

Simulations of three storms
1) Airmass storm in Alabama (May 21)
2) Severe convective storm in northern Oklahoma (May 29)
3) Mesoscale convective system (June 11)

Example Domain Used

Double-moment cloud physics (Morrison scheme)
Simulation time: 12Z to 03Z
Simulations by Megan Bela
WRF-Chem simulation also predicts CH$_3$OOH scavenging efficiencies greater than expected (<10%).

CH$_3$OOH scavenging efficiency varies with ice retention factor:
- 50-100% CH$_3$OOH retained
- H$_2$O$_2$ retention factor < 25%
- CH$_2$O retention factor < 10%

Observation results from Fried et al. (2016) and Barth et al. (2016):
- May 21 airmass
- May 29 severe
- June 11 MCS

Bela et al. (2016 and in preparation)
Cloud Physics Could Enhance Scavenging of some VOCs

1. How much of the aldehydes and peroxides are lofted versus rained out?
   - $\text{H}_2\text{O}_2 > 80\% \text{ SE, } \text{CH}_2\text{O} \sim 50\% \text{ SE, } \text{CH}_3\text{OOH} 12-84\% \text{ SE}$

2. Are aldehydes and peroxides being retained in freezing drops?
   - CH$_3$OOH may be, H$_2$O$_2$ and CH$_2$O are likely not
   - Recent wind tunnel measurements find high retention coefficients for H$_2$O$_2$ and CH$_2$O
   - Differences between our aircraft study and the lab study could be explained by the complex cloud physical processes in the real world

$\Rightarrow$ More work to be done
Cloud Physics Effects on Soluble Trace Gases

Solubility of CH$_2$O, H$_2$O$_2$, CH$_3$OOH
- H$_2$O$_2$: very soluble
- CH$_2$O: moderately soluble
- CH$_3$OOH: mildly soluble

Photochemically-active plumes
CH$_2$O, H$_2$O$_2$, CH$_3$OOH, other VOCs, NO$_x$ → O$_3$

Transport

Wet deposition
CH$_2$O, H$_2$O$_2$, CH$_3$OOH and other soluble trace gases and aerosols
Cloud Physics Effects on Soluble Trace Gases

CH$_2$O, H$_2$O$_2$, CH$_3$OOH, other VOCs, NO$_x$ → O$_3$
Lightning and Chemistry

Predicting Lightning-NO\(_x\) Production

1. Predict the lightning flash rate
2. Estimate the amount of NO produced per flash
3. Find the location of the lightning-NO source in the storm

Photochemically-active plumes

VOCs, NO\(_x\) \(\rightarrow\) O\(_3\)

CH\(_2\)O, H\(_2\)O\(_2\), CH\(_3\)OOH, NO\(_x\), O\(_3\)

soluble trace gases and aerosols
Predicting Lightning Flash Rate

Empirical Relations between Storm Parameters and Flash Rate Derived Mostly from Radar Data (but also satellite data)

<table>
<thead>
<tr>
<th>Storm Parameter</th>
<th>Relationship</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cloud Top Height (20 dBZ)</td>
<td>FR = 3.44×10^{-5} Z_{20dbz}^{4.9}</td>
</tr>
<tr>
<td>Maximum Vertical Velocity</td>
<td>FR = 5.00×10^{-6} w_{max}^{4.55}</td>
</tr>
<tr>
<td>Precipitating Ice Mass</td>
<td>FR = 3.4×10^{-8} PIM – 18.1</td>
</tr>
<tr>
<td>Ice Water Path</td>
<td>FD = 33.33 IWP – 0.17</td>
</tr>
<tr>
<td>Updraft Volume</td>
<td>FR = 3.37×10^{-11} UPVOL</td>
</tr>
<tr>
<td>Upward Ice Flux</td>
<td>FD = 39.48 \phi_{ice}</td>
</tr>
<tr>
<td>35 dBZ Echo Volume</td>
<td>FR = 0.072 VOL35</td>
</tr>
</tbody>
</table>

→ Tested these relationships in WRF simulations of the 22 June storm
WRF Simulations to Test Lightning Flash Rate Parameterizations

- WRF represents timing and location fairly well
- Simulation produces more vigorous storms
Lightning Flash Rate Predictions

- Parameterizations map differently from each other
- Some parameterizations favor lightning in/by updraft region ($w_{\text{max}}$, cloud top height, updraft volume, 35 dBZ volume, ice flux)
- Others include anvil region (Ice Water Path, precipitating ice mass)
Flash Rate in Storm Region

Observations

- Lightning flash rates are difficult to predict
- Need to test these parameterizations more extensively
Lightning Production of NO$_x$

- Lightning-NO$_x$ production estimates for DC3 are 200 moles NO/flash
- Lightning Mapping Arrays give lightning flash rate, location, and flash size
- Some correlation between Lightning-NO$_x$ production and flash size

Pollack et al. (2016)
Ozone is chemically produced in convective outflow regions

- Lightning-production of NO\textsubscript{x}
- Transport
- Photochemically-active plumes: CH\textsubscript{2}O, H\textsubscript{2}O\textsubscript{2}, CH\textsubscript{3}OOH, other VOCs, NO\textsubscript{x}
- Wet deposition: CH\textsubscript{2}O, H\textsubscript{2}O\textsubscript{2}, CH\textsubscript{3}OOH and other soluble trace gases and aerosols
- CH\textsubscript{2}O, H\textsubscript{2}O\textsubscript{2}, CH\textsubscript{3}OOH, NO\textsubscript{x}, O\textsubscript{3}
Ozone is chemically produced in convective outflow regions
21 June 2012 Dissipating MCS Case

O$_3$ increases from 55 to 70 ppbv in mesoscale convective system outflow from 7 am to 6 pm LT

O$_3$ data on GV from Weinheimer et al. (NCAR) and on DC-8 from Ryerson et al. (NOAA/ESRL/CSD)
Ozone from Stratosphere Mixed into Upper Troposphere by Storm

June 22, 2012
NE Colorado / SW Nebraska storm

High $O_3$ just to north of convective outflow indicating stratospheric air

Photo by Armin Sorooshian; Annotated satellite photo from Apel et al. (2015)
High Ozone Values to North of Anvil from UTLS Mixing

- WRF-Chem shows 90-100 ppbv $O_3$ to north and west of storm at 11 km altitude
Modeled UTLS Interactions for 22 June 2012 case

- West-East Vertical Cross Section shows air being brought down within the storm
- High resolution simulations can be used to analyze how the UTLS mixing occurs
Thunderstorms and atmospheric composition:
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1. In severe thunderstorms, some volatile organic compounds are scavenged by thunderstorms more than expected → Cloud physical processes could be responsible

2. Lightning flash rates are hard to predict, affecting estimates of lightning-NO\textsubscript{x} production

3. Lightning-NO\textsubscript{x} production could be influenced by the size of the lightning flash

4. Ozone is chemically produced in convective outflow regions, but small-scale mixing from the stratosphere also puts ozone into the upper troposphere

Ozone is a greenhouse gas in the upper troposphere
Stratosphere-Troposphere Exchange Associated with Storms

Best evidence of STE in satellite and aircraft observations (Pan et al., 2014)

30 May 2012

DC-8 flight segment colored by $O_3$ mixing ratio

DC-8 $O_3$ lidar scan and in situ measurements show stratosphere to troposphere transport that only fine resolution models can simulate
Flash Area

• New method for getting flash area (Bruning and Thomas, 2015)
  – Can be applied to studies on lightning-NO\textsubscript{x} production

- Polygon obtained via convex hull method
- Branching based on fractal nature of lightning

Saw tendency for flash extent to be inversely proportional to flash rate

x, y planar view of lightning flash (VHF sources)

• Polygon obtained via convex hull method
• Branching based on fractal nature of lightning