Atmosphere-surface (ocean/ecosystem) exchange

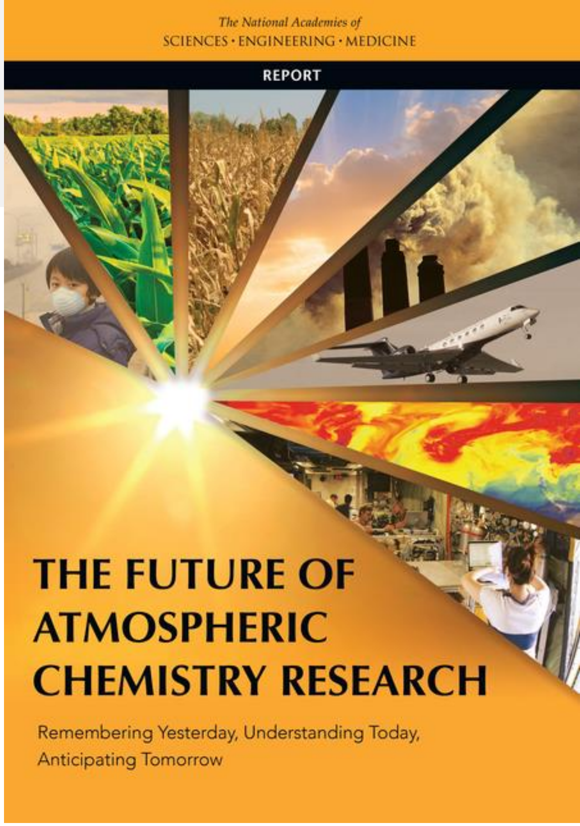
Emission sources

- Energy, Industry
- Agriculture
- Biomass burning
- Marine natural
- Terrestrial natural

Deposition processes

- Wet
- Dry (stomatal, non-stomatal)

Atmosphere-surface exchange
State of Science: 2016 NAS report

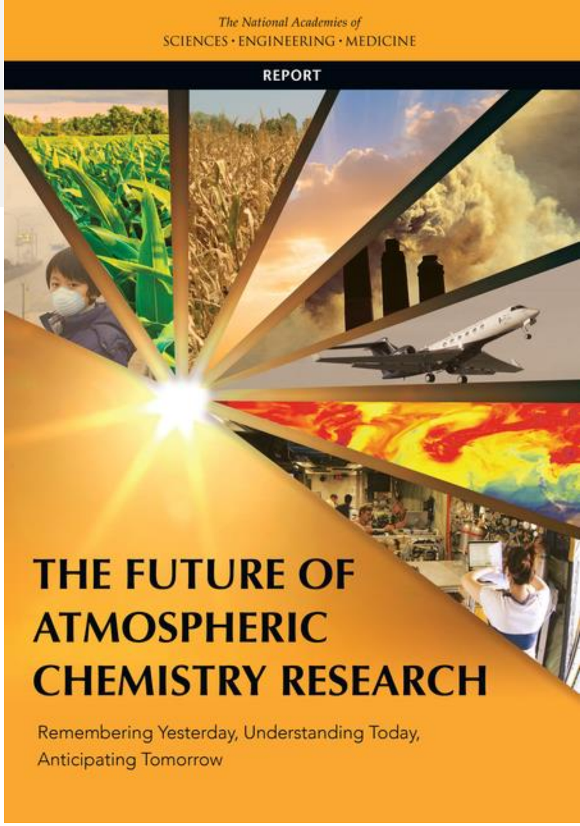


Priority Science Area 2: Quantify emissions and deposition of gases and particles in a changing Earth system.
Priority Science Area 5: Understand the feedbacks between atmospheric chemistry and the biogeochemistry of natural and managed ecosystems.

- **need accurate fluxes to get atmospheric distributions right**
- **fluxes are changing due to human activities**
- **feedbacks and interactions can be important**

Atmosphere-surface exchange
State of Science: 2016 NAS report

Recommendation 2: The National Science Foundation should take the lead in coordinating with other agencies to identify the scientific need for long-term measurements and to establish synergies with existing sites that could provide core support for long-term atmospheric chemistry measurements, including biosphere–atmosphere exchange of trace gases and aerosol particles.



Recent workshops summarizing state of science and open questions

- September 2016 Breckinridge CO: IGAC side meeting on “Removal processes involved in the lifecycle of organic aerosols”
- May 2017 Corsica France: SOLAS-WCRP-ESA “Frontiers in Ocean-atmosphere exchange” workshop
- July 2017 Boulder CO: IGAC Interdisciplinary Biomass Burning Initiative workshop
- October 2017 Lamont-Doherty Columbia University: Ozone dry deposition
- November 2017 UC Irvine: Long-term measurements of biosphere-atmosphere chemical interactions

September 2016 Breckinridge CO: IGAC side meeting on "Removal processes involved in the lifecycle of organic aerosols"

"The consensus was achieved that the community was severely lacking in observations specific to organic deposition and that the community needs to focus on developing an observational strategy first. There was a little discussion on model developments and intercomparisons, that would come in the second step. Discussion focused on long-term flux measurements although there was some discussion of aircraft fluxes as proof of concept."

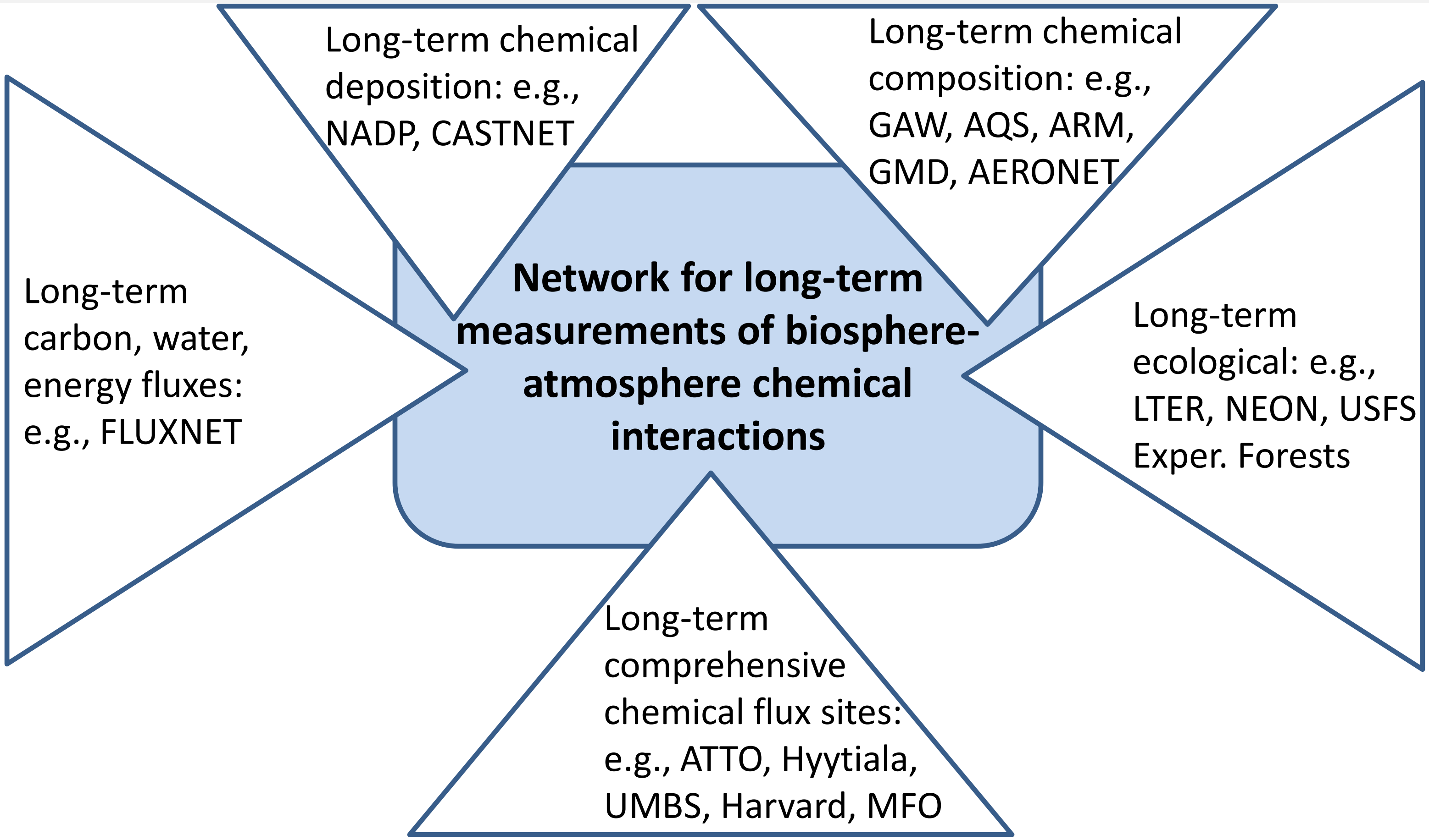
From report organized by
Alma Hodzic Roux and Alex Guenther

October 2017 Lamont-Doherty: Ozone dry deposition workshop

- How do individual deposition pathways vary, & how do they contribute to variability in the total observed deposition?
- What is the contribution of in-canopy-air chemical destruction of ozone by NO &/or BVOCs to the observed ozone flux?
- How does turbulence above & in the canopy impact the magnitude & variability of observed ozone deposition velocity?
- How "regionally representative" are flux-tower observations? What are the controls on spatial variability?

Slide from Olivia Clifton and Arlene Fiore

November 2017 Irvine Workshop on "Long-term measurements of biosphere-atmosphere chemical interactions"

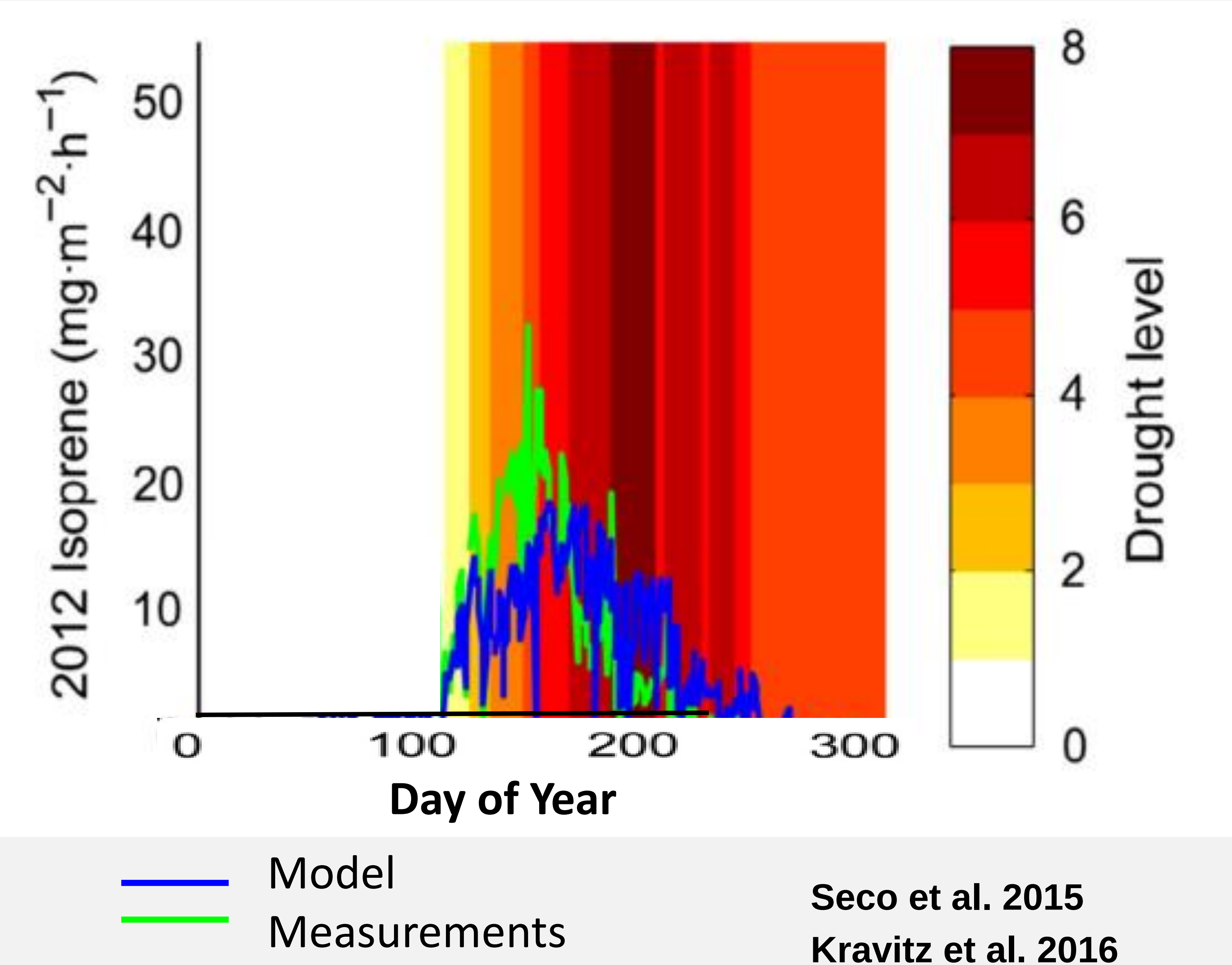


agreement that important scientific questions could be addressed by long-term chemical flux and ancillary measurements, and that a coherent network of sites are needed. The role of this network would be to facilitate data archiving and distribution, develop protocols, support existing sites, and prioritize needs for additional sites.

November Irvine Long-term flux network workshop: Selected Open Questions

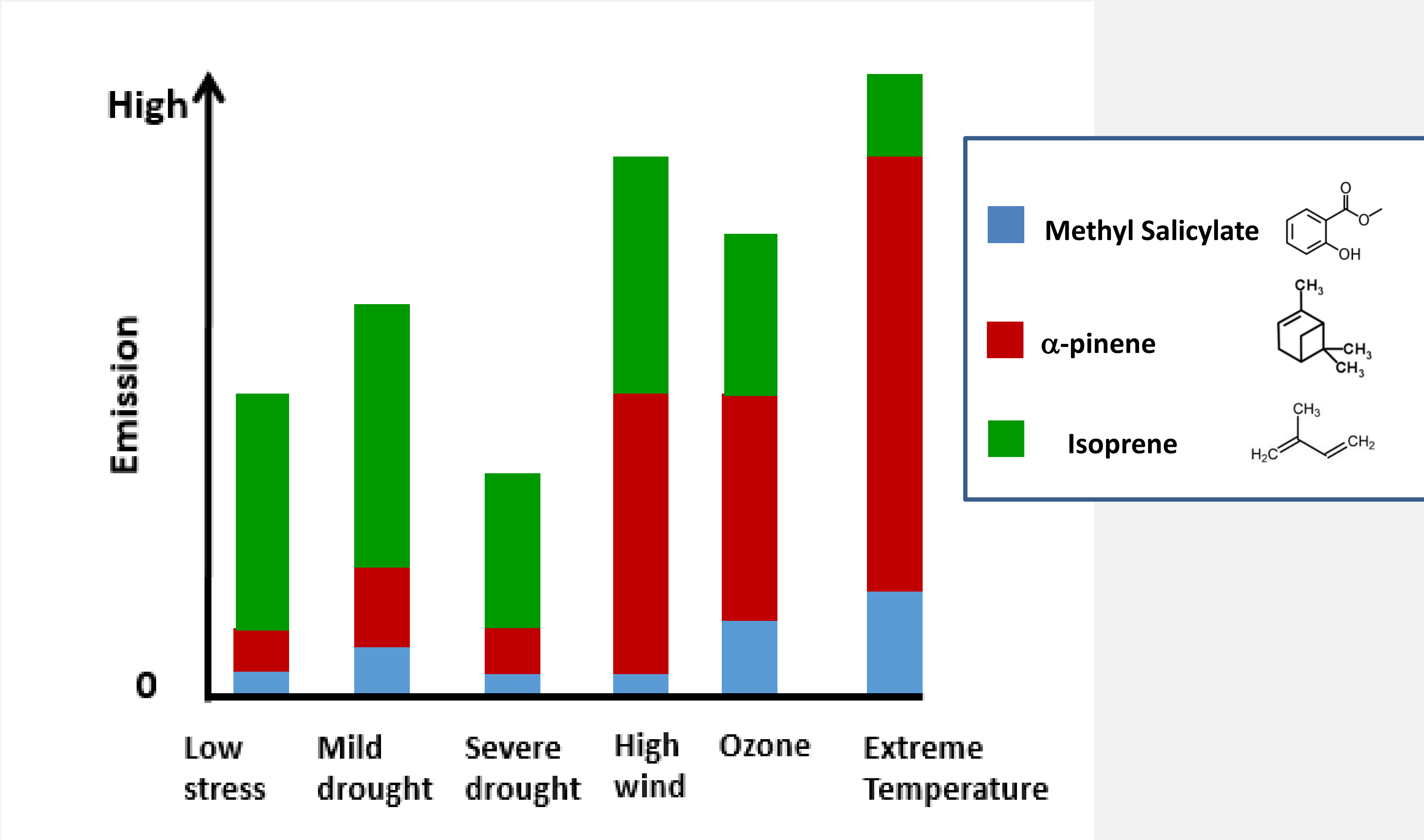
- What is the atmospheric reactivity of **missing biogenic carbon fluxes** and can we close the biosphere-atmosphere exchange budget of total OH reactivity?
- Under what conditions are terrestrial ecosystems a source or sink of particles including Cloud Condensation Nuclei and Ice Nucleating Particles?
- What are the most important pathways by which atmospheric pollutants are transferred to ecological endpoints?
- What is the inter-relationship between biogenic VOC and ozone in **extreme events** and how do these feedback on plant sensitivity to these events?
- What is the impact of **Arctic climate change**, including permafrost thawing, on biogenic emissions and atmospheric chemistry?
- How have emissions from the biosphere and their impact on climate-relevant parameters changed from 1750 to present?
- How do biogenic emissions respond to **invasive species and other ecological disturbances** associated with human activities?
- Can the ozone and PM2.5 bias in regulatory models be reduced by including **explicit canopy processes**?
- How does **changing emissions** and fate affect societally relevant issues including human health, ecosystem services and agricultural productivity?

Isoprene flux from Missouri USA Broadleaf Forest in 2012



Seco et al. 2015
Kravitz et al. 2016

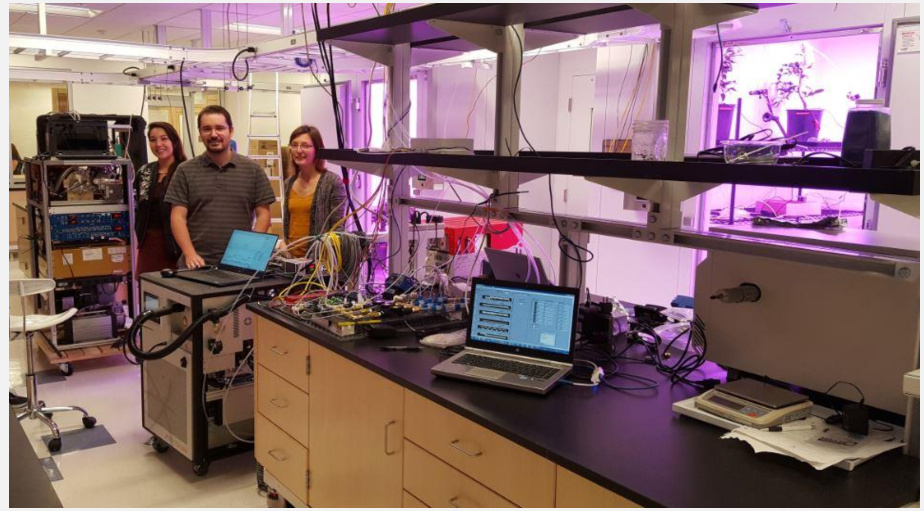
MEGANv3 biogenic emission model predicts plant emission response to stress



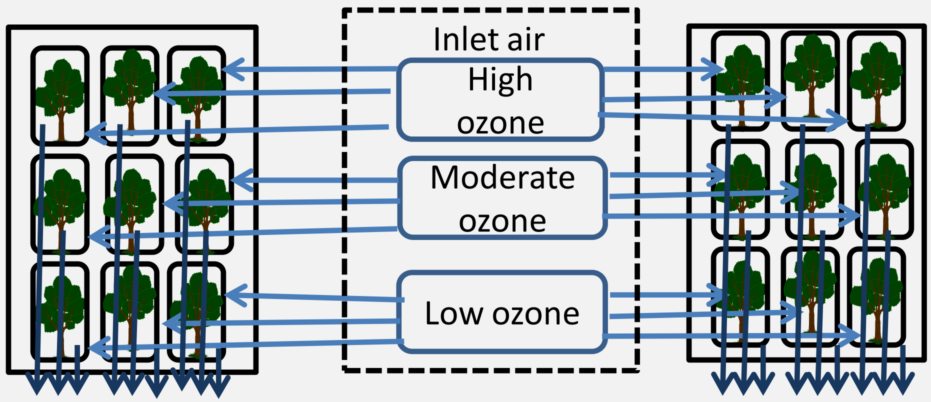
Jiang et al. 2018

Acknowledgement: National Science Foundation (NSF) Atmospheric Chemistry program award AGS1643042

UCI FLUXTRON: Ecosystems in a bottle



Control Room



Manipulation Room: Ozone, drought, Temperature, etc.

Automated, continuous measurement of plant physiology and multi-modal chemical analysis

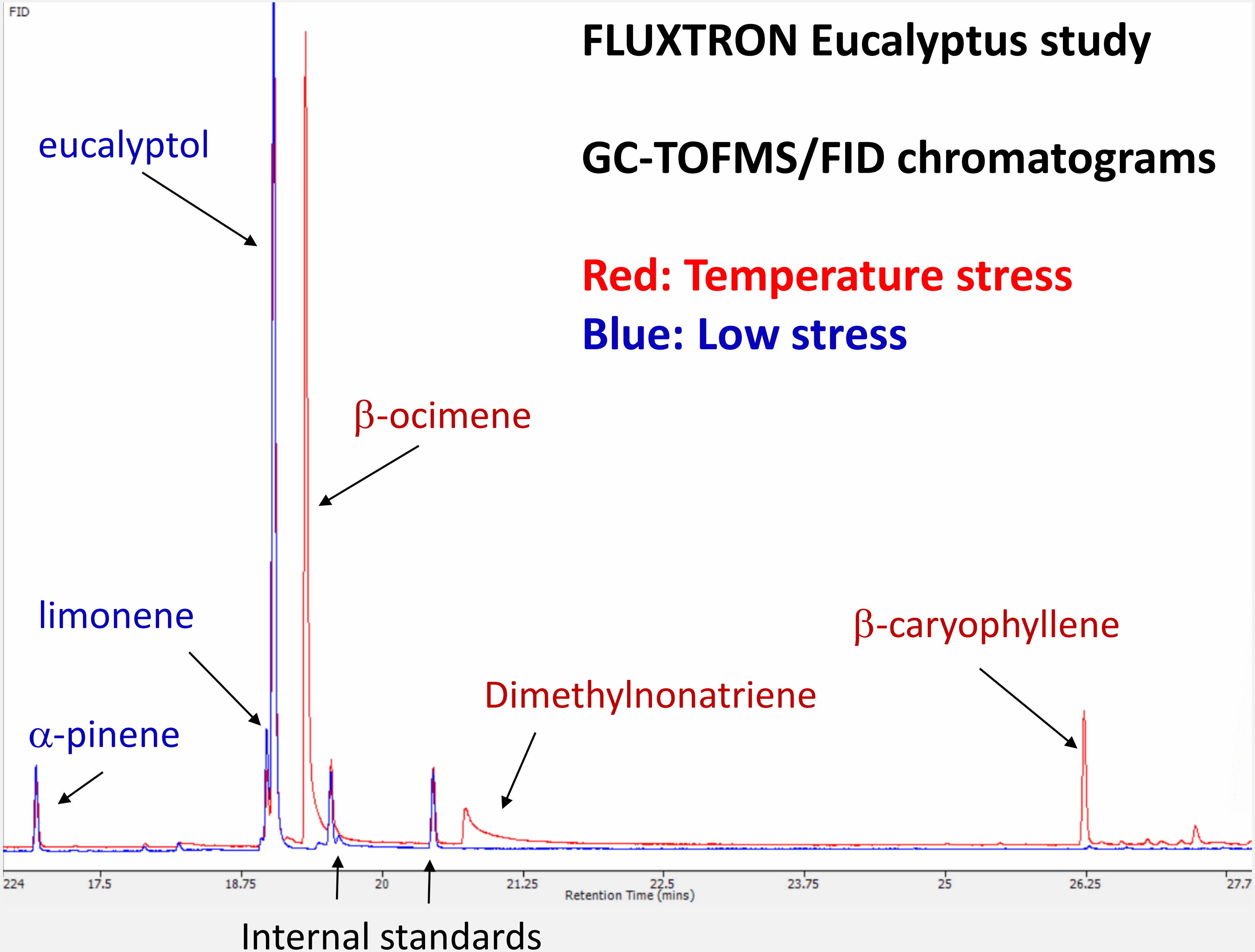
Acknowledgement: National Science Foundation (NSF) Atmospheric Chemistry program award AGS1643042

FLUXTRON Eucalyptus study

GC-TOFMS/FID chromatograms

Red: Temperature stress

Blue: Low stress



Acknowledgement: National Science Foundation (NSF) Atmospheric Chemistry program award AGS1643042

COALA: Characterizing the Organics and Aerosol Loading of Australia

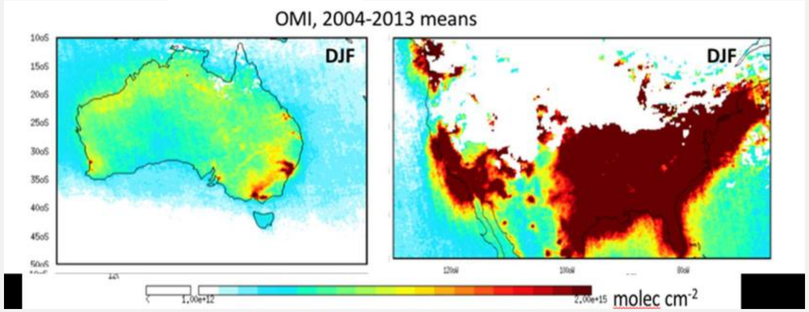
Louisa Emmons, Eric Apel, John Orlando, Alex Guenther, Saewung Kim
Kathryn Emmerson, Clare Murphy, Jenny Fisher, Neil Harris

Motivation and science questions

The motivation for COALA is two-fold: Australia as a clean chemical observatory and constrain BVOC emissions and chemistry.

Through targeted observations and modelling COALA will study:

1. How have emissions changed since pre-industrial conditions?
2. How are BVOC emissions chemically processed?
3. How will climate change and extreme weather (e.g., drought and temperature extremes) impact biogenic emissions?



KNMI/TEMIS Tropospheric NO_2 column from OMI satellite for Australia (left) compared to the US (right), averaged over 10 years for Dec. to Feb.

COALA-TALENT (UK)

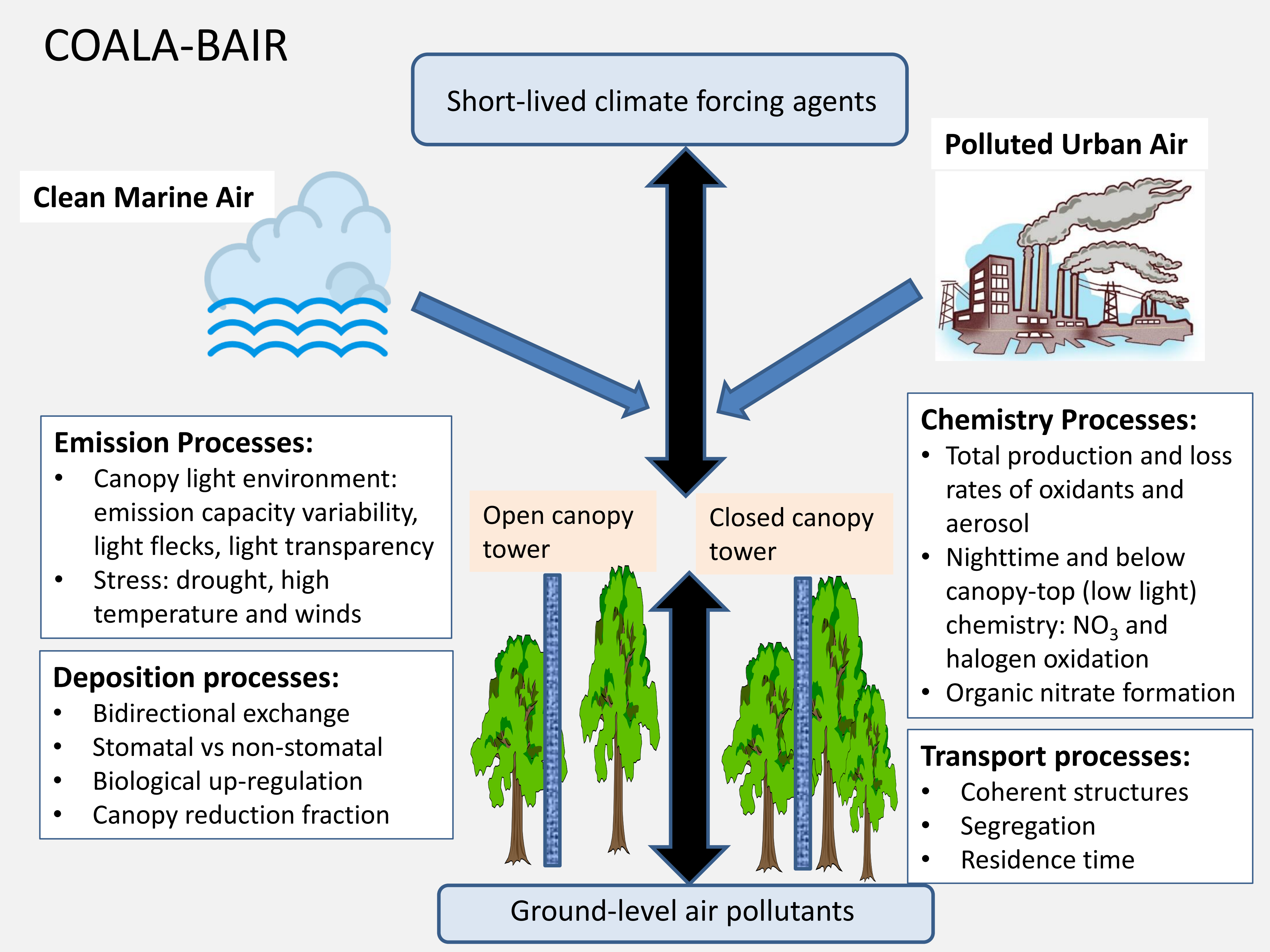
Testing Analogues for the Low Emission of NOx Transition

COALA-JOEYS (Australia)

Joint Organic Emissions Year-round Study

COALA-BAIR (USA)

Biogenic Anthropogenic Interactions Research



COALA-BAIR
Short-lived climate forcing agents
Polluted Urban Air
Clean Marine Air
Emission Processes:
• Canopy light environment: emission capacity variability, light flecks, light transparency
• Stress: drought, high temperature and winds
Deposition processes:
• Bidirectional exchange
• Stomatal vs non-stomatal
• Biological up-regulation
• Canopy reduction fraction
Open canopy tower
Closed canopy tower
Chemistry Processes:
• Total production and loss rates of oxidants and aerosol
• Nighttime and below canopy-top (low light) chemistry: NO_3 and halogen oxidation
• Organic nitrate formation
Transport processes:
• Coherent structures
• Segregation
• Residence time
Ground-level air pollutants

Open Questions addressed by COALA-BAIR

1) Can stress and canopy environment processes explain measurement vs model discrepancies in Australian forests?
2) Are we missing any biogenic compounds that have a significant impact on atmospheric chemistry?
3) How do organic nitrates form in high-BVOC, low-NOx environments?; implications for NOx, ozone, and aerosols?
4) How important is halogen oxidation in a coastal forest?
5) How does the simultaneous emission of isoprene and terpenes affect ozone and aerosol formation?

Timeline

July 1, 2018: Submit proposal to OFAP for EOL (ISFS, ISS) and ACOM (TOGA, HARP, O3/NOx) instruments and UCI (leaf to canopy VOC, organic halogen, OH reactivity).
After July 1: University proposals
Jan/Feb 2020: Field campaign at Cataract Park, New South Wales, Australia

Contact: Louisa Emmons (emmons@ucar.edu) or Alex Guenther (alex.guenther@uci.edu)

Recent Advances in Modeling Atmosphere-Surface Exchange

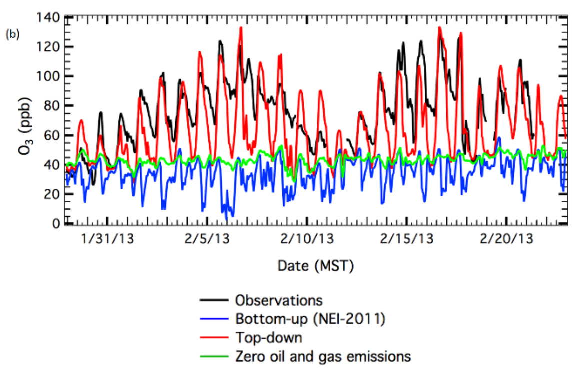
- Discuss recent advances in the priority science areas detailed in the 2016 NAS report:
  - **2: Quantify emissions and deposition of gases and particles in a changing Earth system.**
  - **5: Understand the feedbacks between atmospheric chemistry and the biogeochemistry of natural and managed ecosystems.**
- Highlight current and past NCAR projects including collaborative projects.
- Present some lingering science discussion questions

2: Quantify emissions and deposition of gases and particles in a changing Earth system.

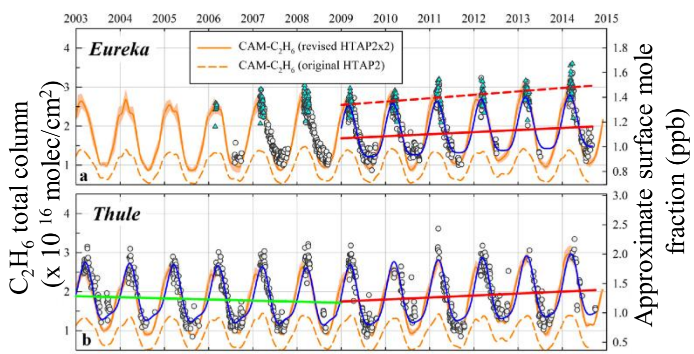
Anthropogenic Emissions (VOCs)

- In general, the atmospheric chemistry field is good at identifying new/changing emissions sources caused by “a changing Earth system”. Examples include:

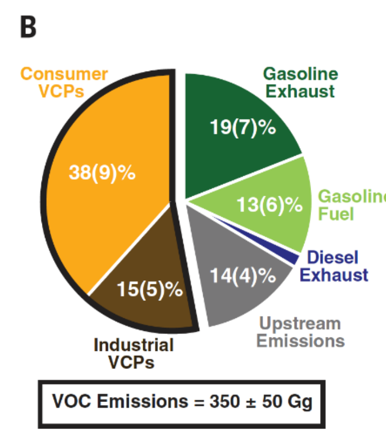
- Increased VOC emissions from oil and gas sector lead to high O_3 events in Utah (e.g., Ahmadov et al. 2014)



- **NCAR + University:** Increased emissions from oil and gas sector lead to increases in ethane at northern latitude long-term monitoring stations (e.g., Franco et al. 2016). Jim Hannigan at NCAR supports the long-term FT-IR measurements made at Thule.



- Volatile chemical products are now contributing a large fraction to total VOC emissions in LA (e.g., McDonald et al. 2018)



Discussion Questions: Are there new/changing sources of emissions that the atmospheric chemistry community is not studying or should be studying more?

2: Quantify emissions and deposition of gases and particles in a changing Earth system.

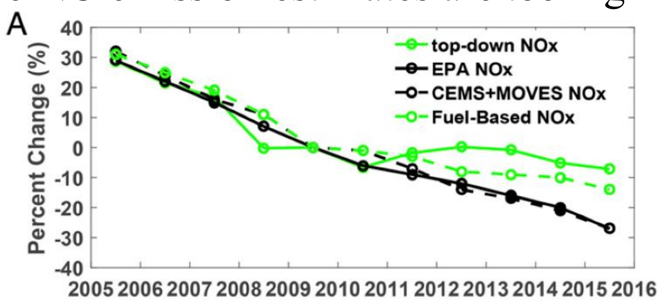
Anthropogenic Emissions (NOx)

- In general, there are still large uncertainties in emission inventories that greatly impact the ability of models to accurately simulate ozone and secondary organic aerosol?

Example model studies of NO_x:

Study	**Model/Location**	**NEI NO_x Emissions Evaluation**
Anderson et al. 2014	CMAQ/Maryland	51-70% overestimate in mobile
Kota et al. 2014	CMAQ/Texas	15-25% reduction in mobile
Canty et al. 2015	CMAQ/NE US	50% reduction in mobile
Travis et al. 2016	GEOS-Chem/SE US	60% reduction in all non-power plant

- Fuel based/tunnel studies suggest the MOVES model mobile NO emission estimates are too high by 40-60% (McDonald et al. 2012 and Fujita et al. 2012).
- **NCAR + University:** Satellite measurements suggest NO_x is not decreasing over the last decade in the US as substantially as expected by US EPA inventory estimates (Jiang et al. 2018).



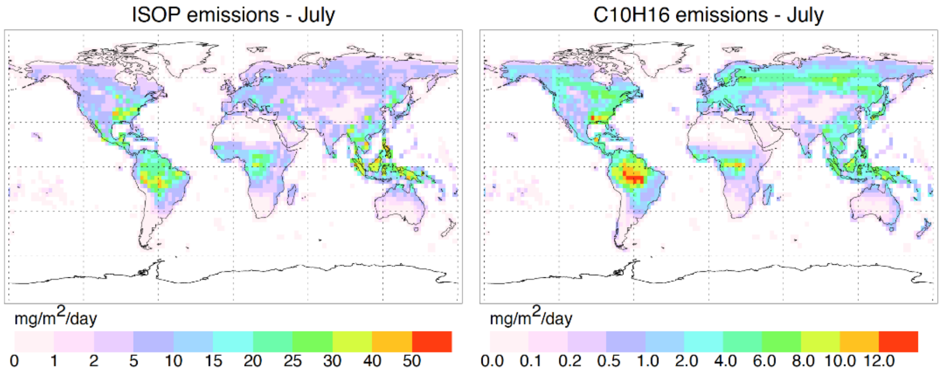
- **Discussion questions:** What techniques can best be used to better reduce uncertainties in emissions? How can the atmospheric chemistry field better communicate and collaborate with the US EPA to improve the NEI inventory?

2: Quantify emissions and deposition of gases and particles in a changing Earth system.

Biogenic Emissions from Land

- Globally biogenic VOC emissions are substantially greater than anthropogenic VOC emissions.
- The Model of Emissions of Gases and Aerosols from Nature (MEGAN) emissions modeling framework developed by Alex Guenther (Guenther et al. 2012) has been tested in many models.
- **NCAR + University:** For example, in CAM-chem (atmospheric chemistry component of NCAR's Climate Earth System Model) MEGAN emissions can be run with the coupled land model or with satellite leaf area index (Emmons et al. 2012).

July Isoprene and Monoterpene emissions from online MEGAN



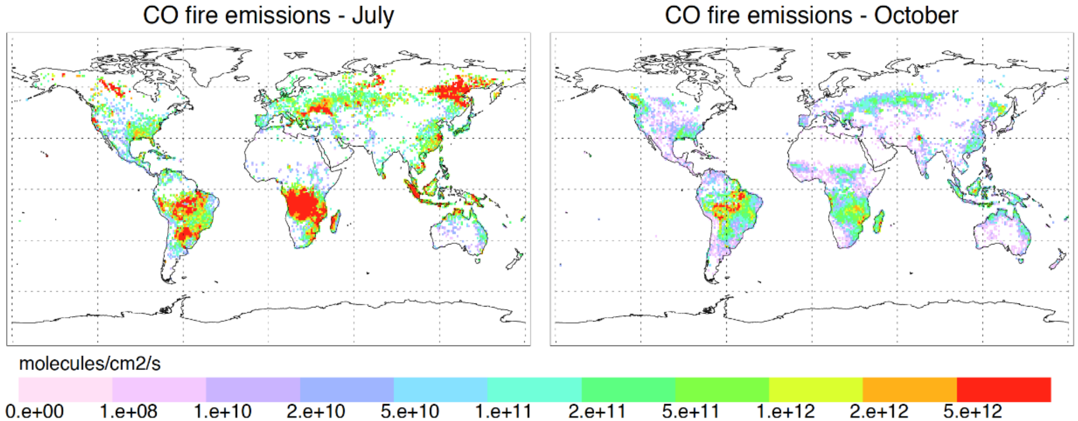
- **Discussion questions**: Based on satellite measurements and observations some models suggest MEGAN emissions are too high (e.g., GEOS-chem, Kaiser 2018). How can we better compare biogenic emissions in models and determine what causes the differences? How can we improve these biogenic emissions further (e.g., canopy models, improve emission factors, improve vegetation type maps…)?

2: Quantify emissions and deposition of gases and particles in a changing Earth system.

Biomass Burning Emissions

- **NCAR + University:** The Fire INventory from NCAR (FINN) generates fire emissions (daily, 1km resolution) for gases and particles from wildfires, agricultural fires, and prescribed burns (Wiedinmyer 2011).

Monthly CO emissions from FINN



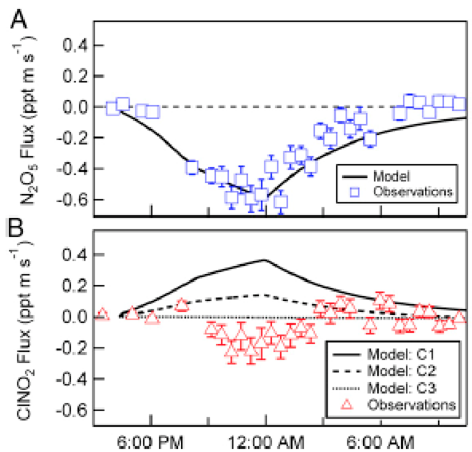
- **NCAR + University**: The emissions from FINN are also evaluated against MOPITT CO data (e.g., Gaubert et al., 2016) (CO retrievals from MOPITT are produced by NCAR)
- **Future NCAR Plans:** ACOM is planning to conduct laboratory studies examining kinetics and oxidation products from key reactive carbon compounds emitted from wildfires. ACOM will also improve the representation of wildfires in CAM-chem in coordination with upcoming campaigns.
- **Discussion questions:** Biomass burning emissions factors vary between emissions inventories and are dependent on a wide variety of fire characteristics (e.g, fuel type, fire temperature, combustion efficiency, etc.). How can we better reduce the uncertainties in these emissions factors?

2: Quantify emissions and deposition of gases and particles in a changing Earth system.

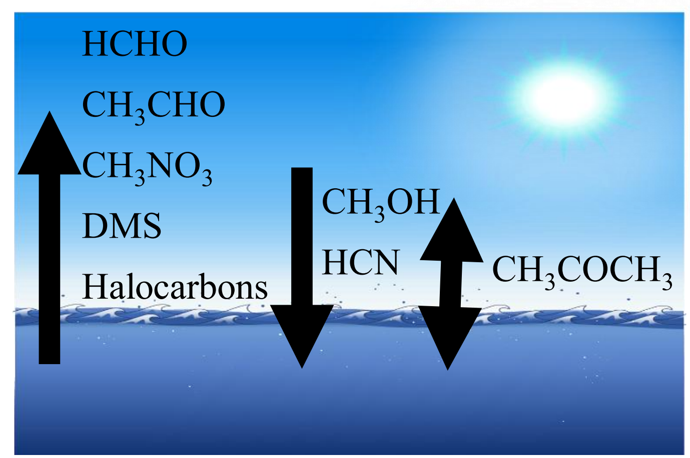
Ocean/Remote Atmosphere Emissions

- Recently, the NASA Atmospheric Tomography Mission (ATom) has provided a comprehensive dataset of the composition of the remote atmosphere.
- ATom and other field campaign observations provide constraints on emissions over the ocean.

- Eddy covariance measurements of fluxes are critical for constraining models. For example, Kim et al. 2014 determined that air-sea exchange of N_2O_5 and $ClNO_2$ removes up to 15% of nocturnal NO_x in costal regions.



- **NCAR + University:** Siyuan Wang is developing an online air-sea exchange module for CESM, which predicts the bi-directional oceanic fluxes of trace gases.

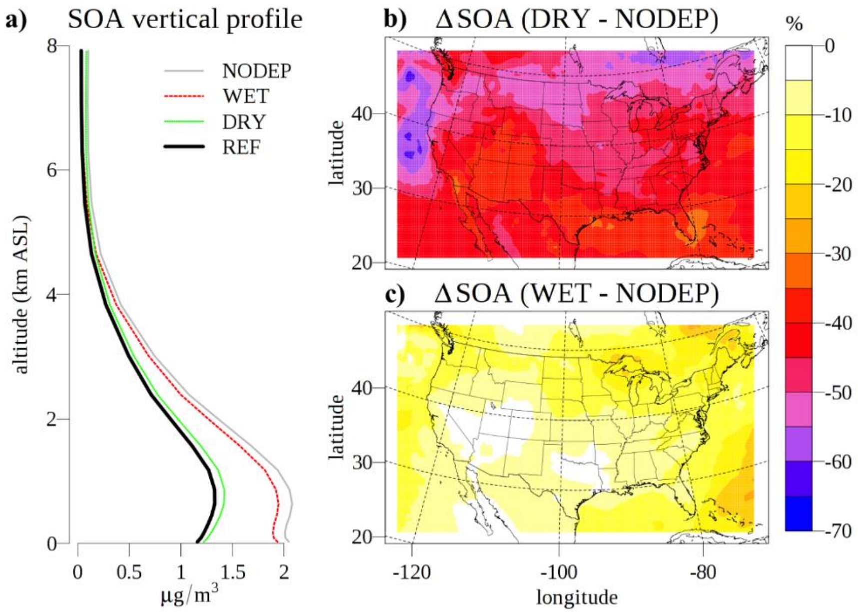


Discussion Questions: How can we facilitate better communication and more collaborative projects between the atmospheric chemistry and oceanography communities to better constrain concentrations of chemical compounds in the ocean and better represent biogeochemistry in models?

2: Quantify emissions and deposition of gases and particles in a changing Earth system.

Wet/Dry Deposition of OVOCs, low-volatility OVOCs, and SOA

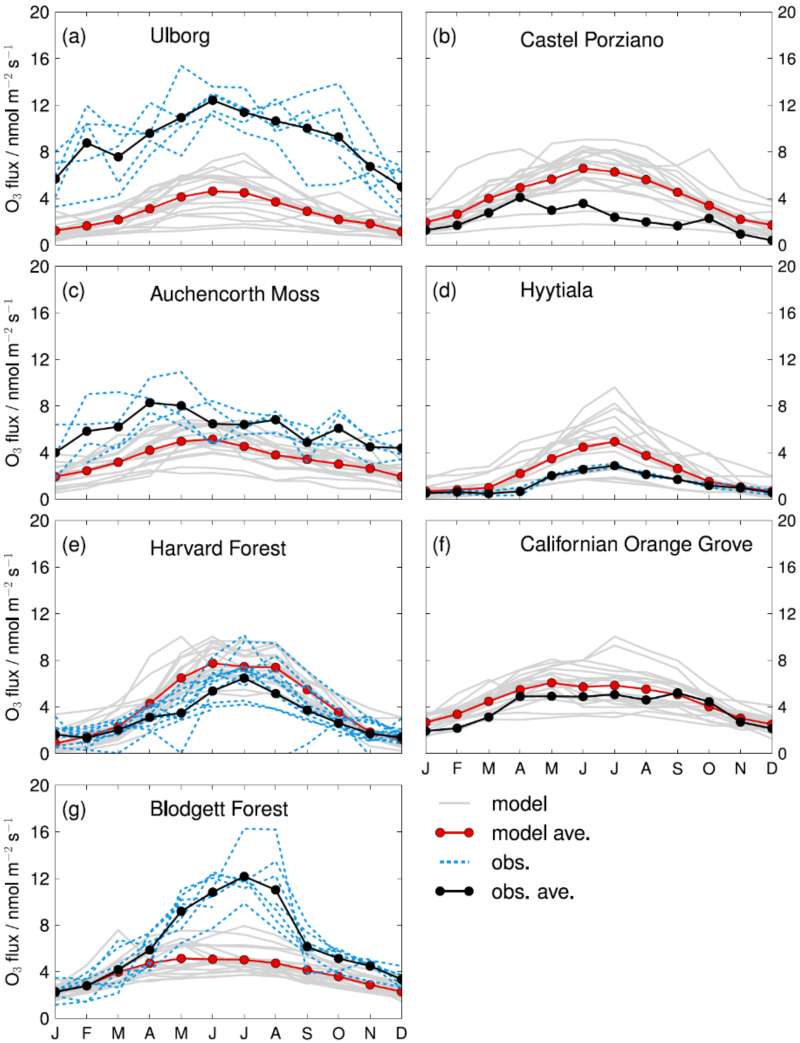
- There are large uncertainties in the wet/dry deposition of OVOCs, low-volatility OVOCs, and SOA.
- **NCAR + University:** Measurements are critical for constraining the models. For example, Karl et al. 2010 determined that dry deposition of OVOCs is larger than previously thought due to enhanced reactivity on the surface of plants.
- **NCAR + University:** Barth at al. 2016 determined hydrogen peroxide and methyl hydrogen peroxide scavenging efficiencies in thunderstorms during the DC3 field campaign.
- **NCAR + University:** Knote et al. 2014 using WRF-chem determined that semi-volatile organic compounds lost through wet/dry deposition significantly reduced SOA concentrations.



Discussion Questions: How can the atmospheric chemistry community further improve wet/dry deposition schemes used within models and better evaluate and reduce uncertainties in trace gases or aerosols caused by wet/dry deposition in models?

5: Understand the feedbacks between atmospheric chemistry and the biogeochemistry of natural and managed ecosystems.

Dry Deposition of Ozone (O_3)



- O_3 deposition both impacts the ecological system causing harm to vegetation and is an important loss of surface O_3.
- Hardacre et al. 2015 compared O_3 deposition in 15 different global models through the TF HTAP intercomparison project. Annual global deposition fluxes varied between 818-1256 Tg/yr.

Discussion Questions: How can the atmospheric chemistry community further reduce the O_3 deposition uncertainty/variability in models? Possibilities include (Hardacre 2015):

- Better understanding O_3 deposition over oceans, grasslands, and tropical forests
- Flux measurements with full seasonal cycle from sites with land cover classes that are representative of large regions of the model.

5: Understand the feedbacks between atmospheric chemistry and the biogeochemistry of natural and managed ecosystems.

Deposition of Nitrogen and Dust Impact on Ocean and Land Ecosystems

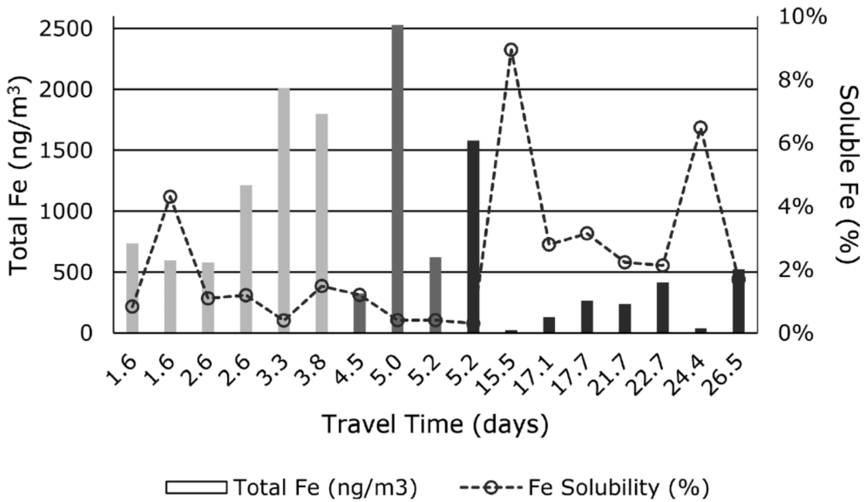
- Deposition of atmospheric nitrogen and mineral dust provides nutrients to ocean and land ecosystems.
- Excessive nitrogen deposition can harm ecosystems and threaten biodiversity (e.g., Ellis 2013, impact on U.S. national parks).
- **NCAR + University:** In the future, atmospheric nitrogen (nitrogen oxides and ammonia) deposition will change depending on emissions scenarios and the region (Lamarque et al. 2013).

Multi-model mean

Region	2000	RCP2.6 2100	RCP4.5 2100	RCP8.5 2100
Canada	203	150	148	201
USA	613	416	412	550
Mexico	412	464*	351	503*
South Asia	728	1550*	1023*	1318*
Korea region	1058	921	751	894
East Asia	756	1021*	690	888*
Southeast Asia	661	827*	569	752*

(Abbreviated table)

- Accurately incorporating dust deposition into models relies heavily on observations of the solubility of iron and phosphorus in dust particles, which is dependent on mineralogy, transportation time through the atmosphere, etc. (e.g., Schroth et al. 2009, Longo et al. 2016)



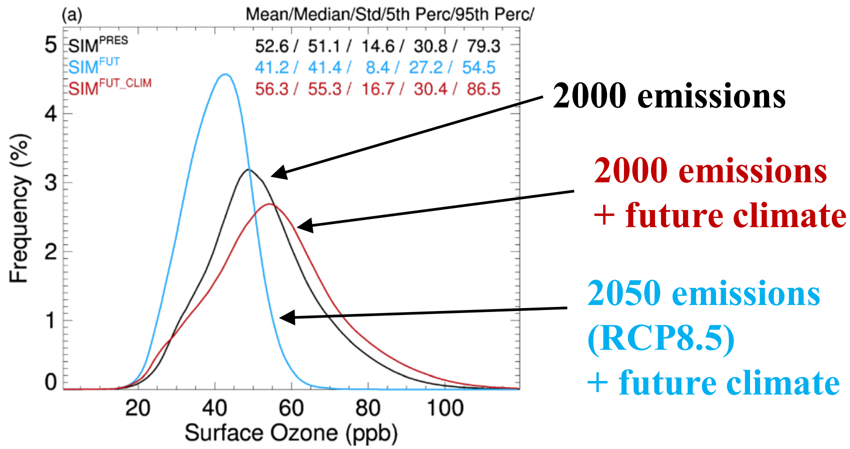
Discussion Questions: How do we encourage more communication and collaboration between the atmospheric chemistry community and ecologists/oceanographers to more completely address how dust and atmospheric nitrogen impact ecosystems?

5: Understand the feedbacks between atmospheric chemistry and the biogeochemistry of natural and managed ecosystems.

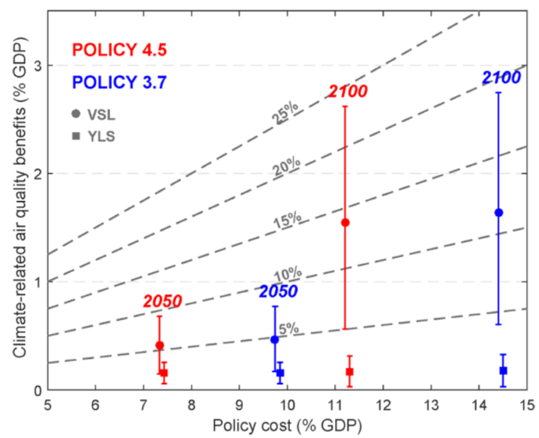
Air Quality in the Future

- Air quality is expected to change in the future due to climate change and climate policy. Models are important for quantifying and predicting these changes. Examples include:

- **NCAR + University:** Pfister et al. 2014 using a regional coupled chemistry-transport model (NRCM-Chem) determined that climate change will increase surface ozone and further degrade air quality unless stringent emissions controls are adopted.



- Garcia-Menendez et al. 2015 demonstrated that climate policy can improve air quality (O_3 and $PM_{2.5}$) and that air quality health benefits can offset part of the climate mitigation costs.



Discussion Questions: How can the atmospheric chemistry community facilitate more communication and collaboration with the climate community to better constrain how the changing climate will impact air quality and encourage more awareness of the significant air quality impacts that will occur by mitigating climate change (e.g., reducing coal, oil, and gas and increasing solar and wind energy)?

Potential Discussion Questions

2: Quantify emissions and deposition of gases and particles in a changing Earth system.

- Are there new/changing sources of emissions that the atmospheric chemistry community is not studying or should be studying more?
- What techniques can best be used to better reduce uncertainties in emissions? How can the atmospheric chemistry field better communicate and collaborate with the US EPA to improve the NEI inventory?
- Biomass burning emissions factors vary between emissions inventories and are dependent on a wide variety of fire characteristics (e.g, fuel type, fire temperature, combustion efficiency, etc.). How can we better reduce the uncertainties in these emissions factors?
- How can we better compare biogenic emissions in models and determine what causes the differences? How can we improve these biogenic emissions further (e.g., canopy models, improve emission factors, improve vegetation type maps…)?
- How can we facilitate better communication and more collaborative projects between the atmospheric chemistry and oceanography communities to better constrain concentrations of chemical compounds in the ocean and better represent biogeochemistry in models?
- How can the atmospheric chemistry community further improve wet/dry deposition schemes used within models and better evaluate and reduce uncertainties in trace gases or aerosols caused by wet/dry deposition in models?

5: Understand the feedbacks between atmospheric chemistry and the biogeochemistry of natural and managed ecosystems.

- How can the atmospheric chemistry community further reduce the O_3 deposition uncertainty/variability in models?
- How do we encourage more communication and collaboration between the atmospheric chemistry community and ecologists/oceanographers to more completely address how dust and atmospheric nitrogen impact ecosystems?
- How can the atmospheric chemistry community facilitate more communication and collaboration with the climate community to better constrain how the changing climate will impact air quality and encourage more awareness of the significant air quality impacts that will occur by mitigating climate change (e.g., reducing coal, oil, and gas and increasing solar and wind energy)?