Outstanding Questions in Atmospheric Composition, Chemistry, Dynamics and Radiation for the Coming Decade

NASA Workshop held at NASA-Ames
May 2014

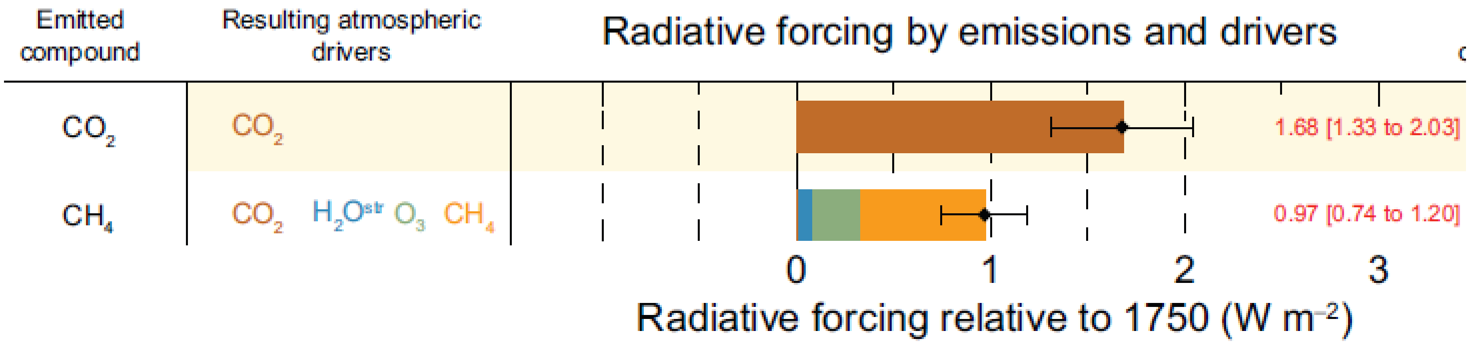
Tropospheric composition: research questions for the next decade

1. What is the future of air quality in a changing world?
2. How are the sources of methane changing?
3. How do convection and precipitation scavenge and redistribute trace species?
4. How do biogenic, anthropogenic, and pyrogenic organics affect oxidants and aerosols?
5. What controls the concentrations of radical oxidants?
6. How are humans and climate perturbing the nitrogen cycle?
7. What drives long-term trends in tropospheric ozone?
8. How do boundary layer dynamics, surface exchange and chemistry interact to determine the vertical distribution and evolution of trace species?
9. What controls background ozone concentrations in surface air?

with thanks to B. Alexander, J. Al-Saadi, E. Atlas. K. Bowman, W. Brune, A. Carlton, R. Cohen, O. Cooper, J. Dibb, B. Duncan, J. DeGouw, A. Fiore, J. Fishman, C. Heald, D. Henze, D. Jaffe, P. Kasibhatla, C. Kolb, J. Neu, M. Newchurch, D. Parrish, K. Pickering, M. Prather, W. Simpson, H. Singh, D. Streets, P. Wennberg, Y. Wang

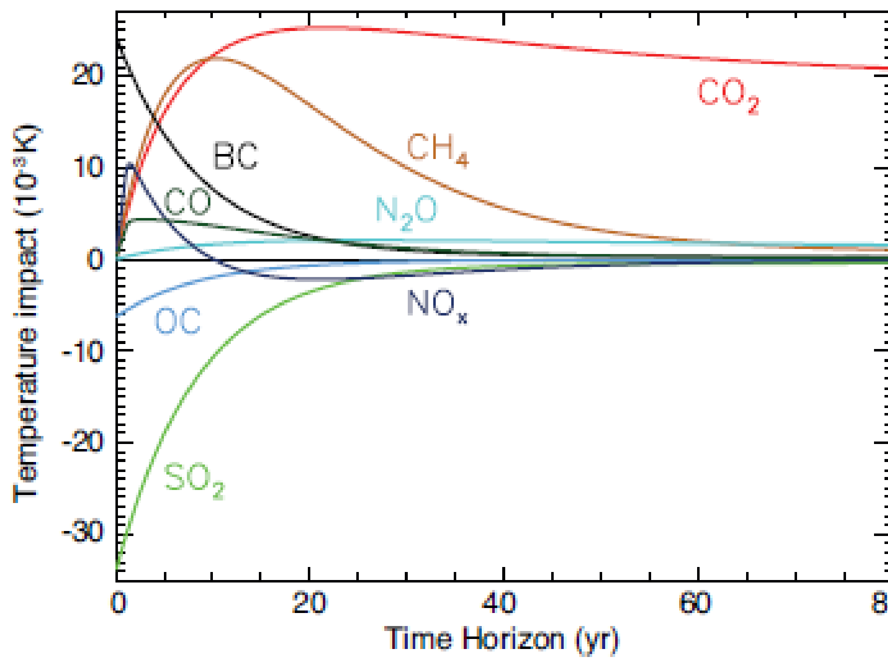
Importance of methane as near-term climate forcer

- **Present-day emission-based radiative forcing of methane is 0.97 W m^{-2}, compared to 1.68 W m^{-2} for CO_2**



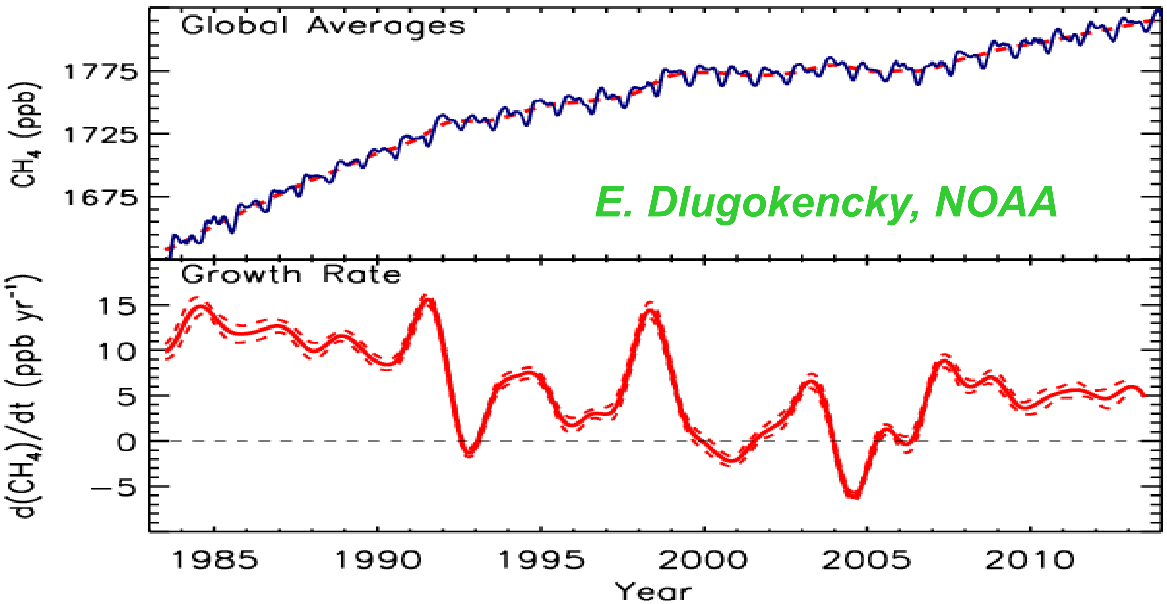
- **On 20-year time horizon methane is of comparable importance to CO_2**

Temperature response to 1-year pulse (2008) of emissions



IPCC [2014]

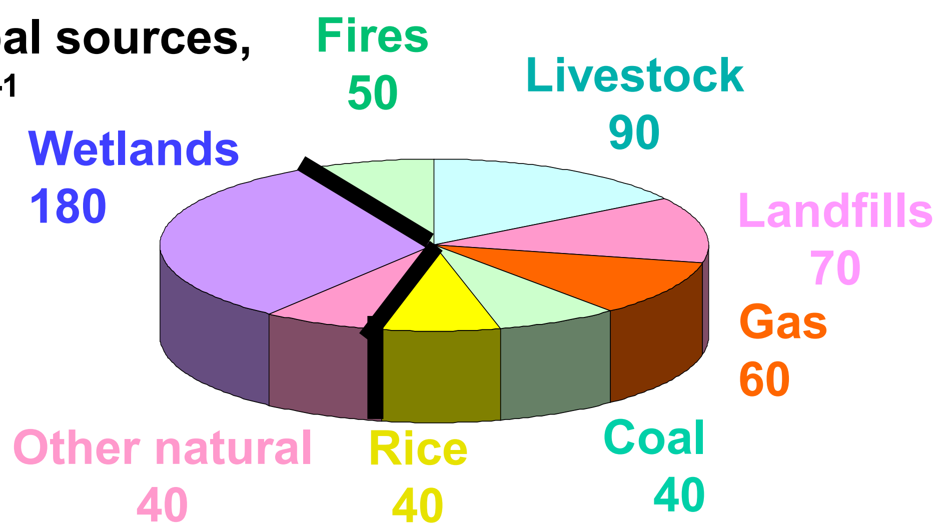
Methane trends over past 30 years are not understood



Why did methane level off in the 00s and then resume its increase?

Source attribution is difficult due to diversity, complexity of sources

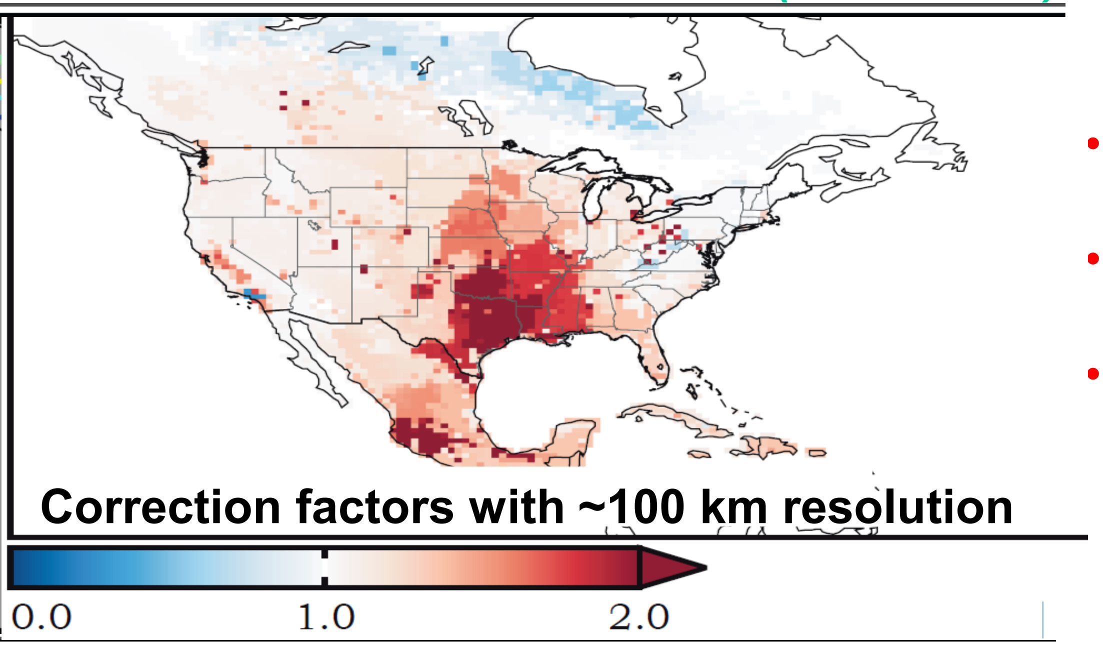
Global sources, Tg a^{-1}



Individual sources uncertain by at least factor of 2; emission factors are highly variable, poorly constrained

Satellites offer powerful constraints on methane sources

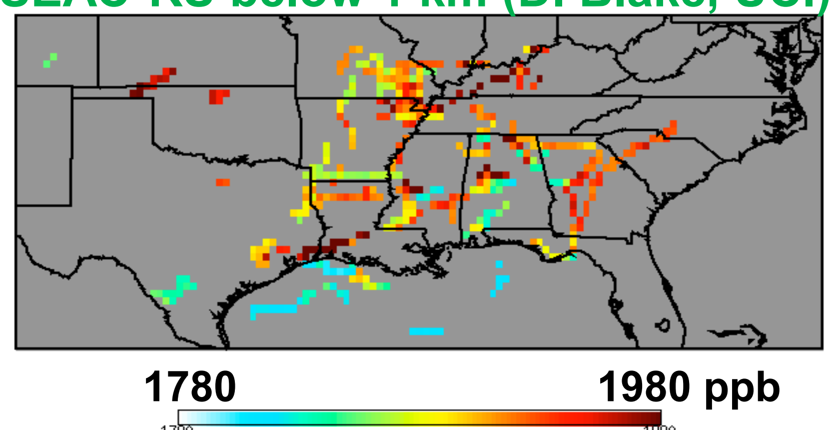
GOSAT observations, 6/09-1/12 (*n = 590,000*)



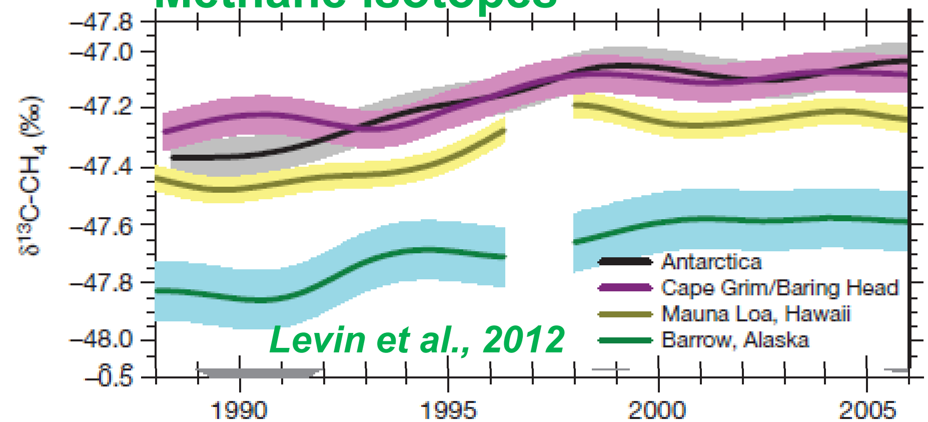
- GOSAT launched in 2009: sparse but high-quality
- TROPOMI to be launched in 2015 7x7 km^2 daily
- Future candidate missions: CarbonSat, geostationary

Need to complement with suborbital observations

SEAC^{4}RS below 1 km (D. Blake, UCI)



Methane isotopes



Methane: relating top-down constraints to processes

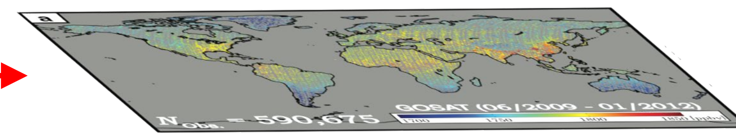
- **Need better connection between bottom-up and top-down estimates: bottom-up is essential for understanding and policy**

Top-down:
atmospheric observations + CTM inversion

Bottom-up:
activity rate
emission factor

concentrations

emissions



- **Need better understanding of wetlands source**

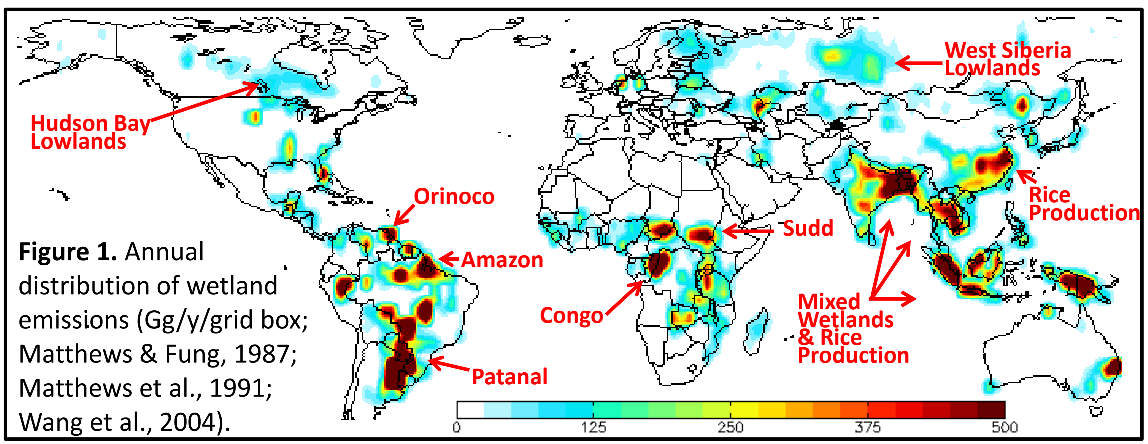
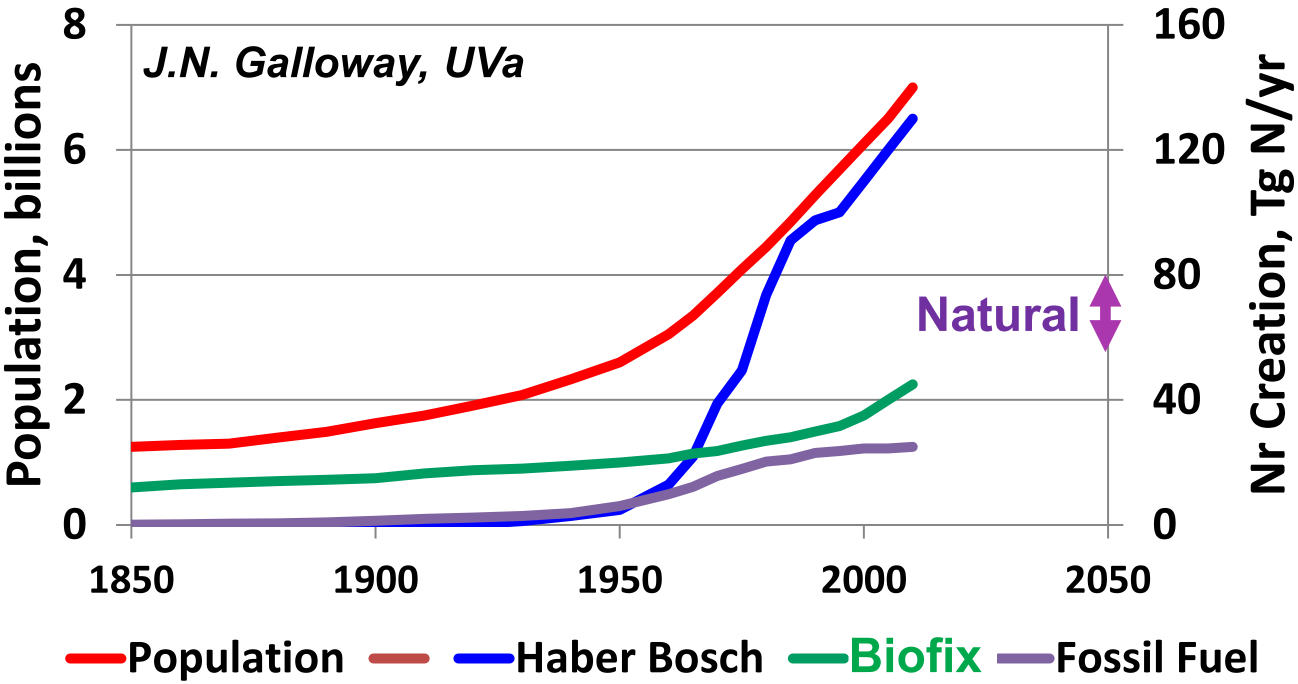


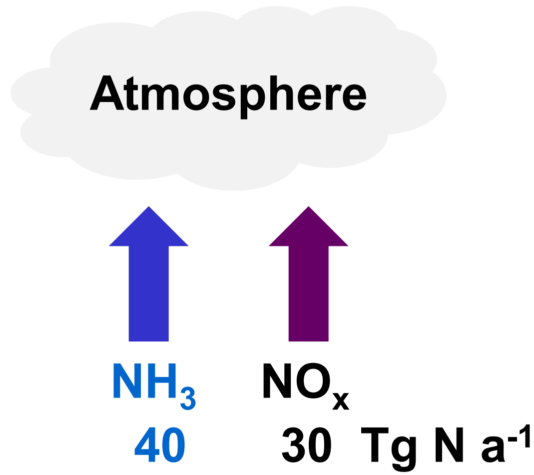
Figure 1. Annual distribution of wetland emissions (Gg/y/grid box; Matthews & Fung, 1987; Matthews et al., 1991; Wang et al., 2004).

- **Major contributor to interannual variability**
- **Complex response to climate change**
- **information on wetland properties from GRACE, SMAP**

Global human perturbation of nitrogen cycle

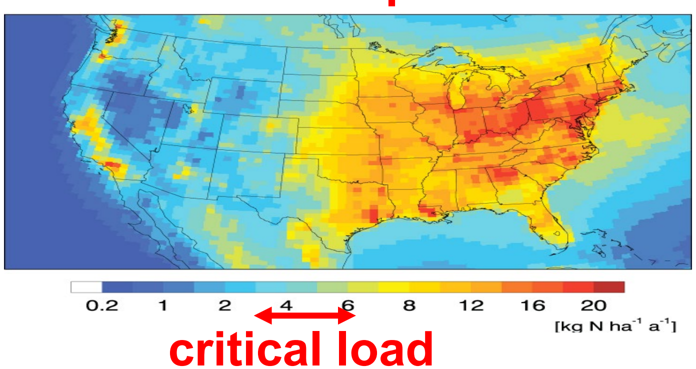
Global anthropogenic N fixation now greatly exceeds natural:





Resulting N deposition (NH_4^+, NO_3^-) modifies ecosystem function, C storage

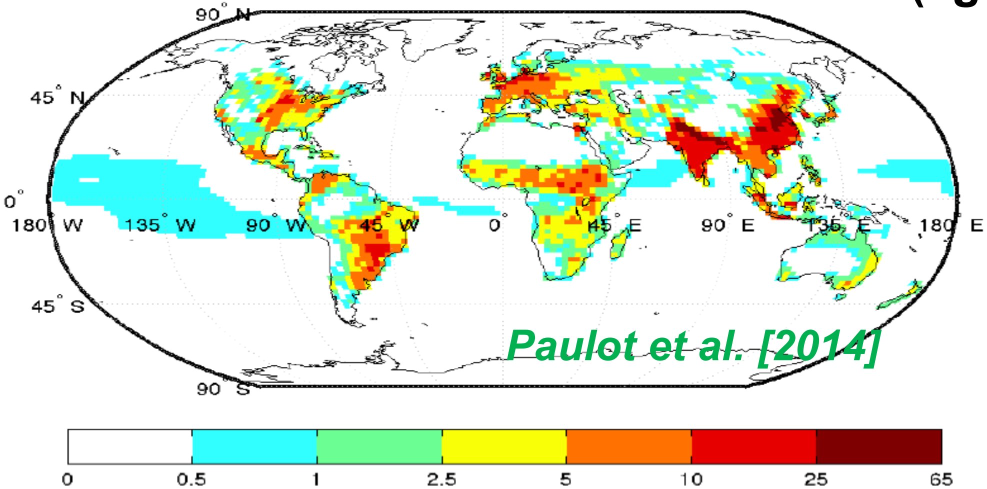
Annual N deposition



Zhang et al. [2012]

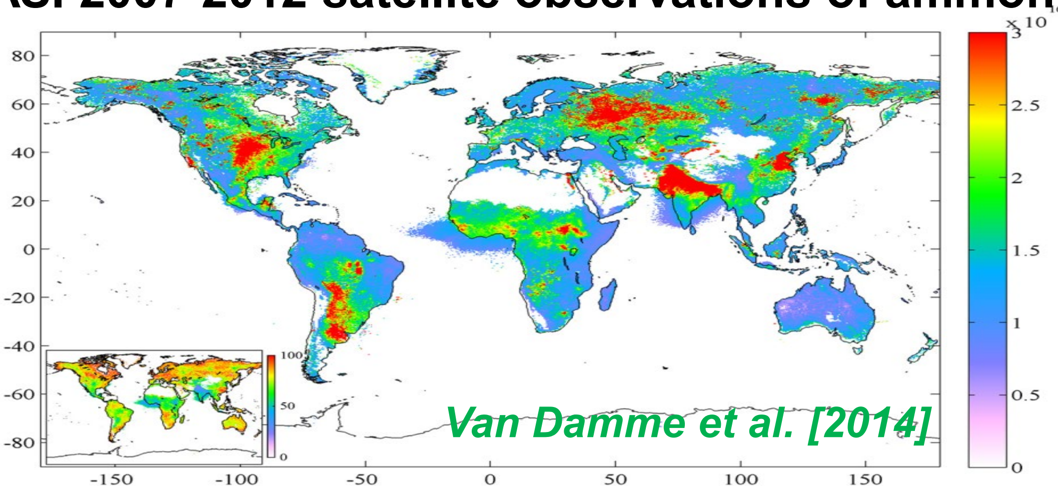
Need better understanding of ammonia emissions

Global ammonia emissions 2005-2008 (kg N ha^{-1} a^{-1})



- Agriculture is 75% of global source
- Ocean is dominant natural source but poorly constrained

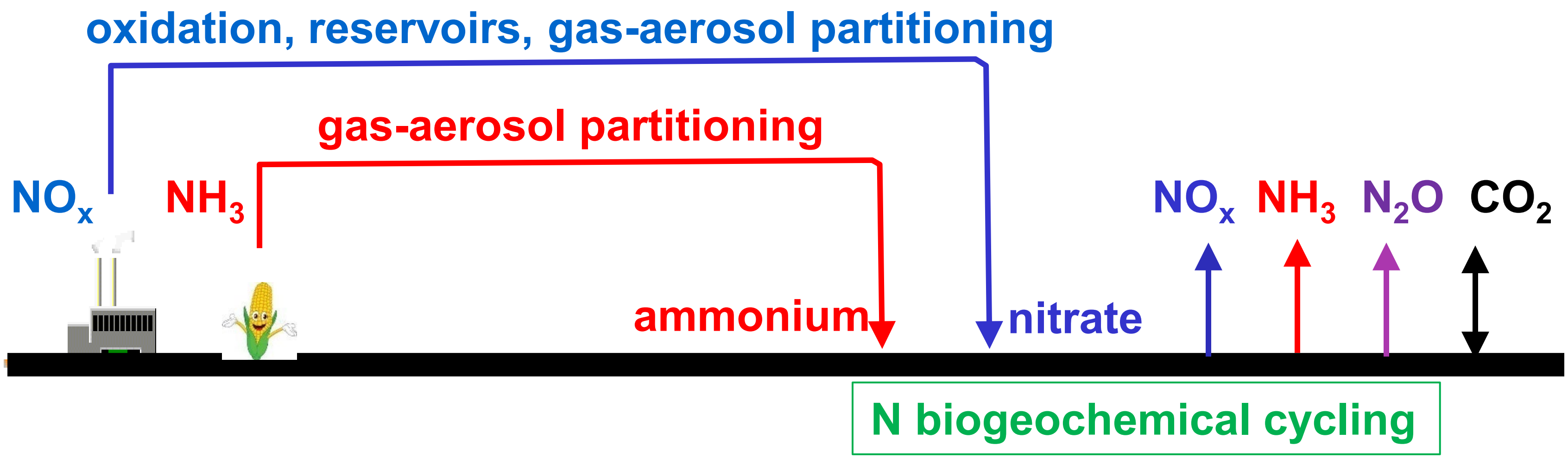
IASI 2007-2012 satellite observations of ammonia



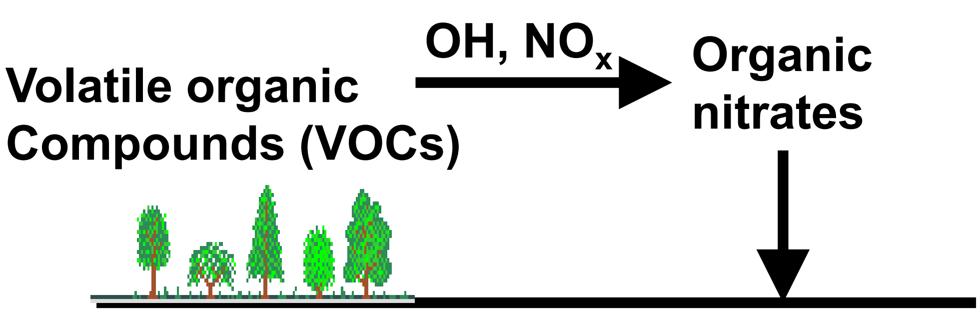
- Ammonia observed in TIR, highly sensitive to vertical distribution and gas-aerosol partitioning
- Need supporting aircraft campaigns to improve interpretation

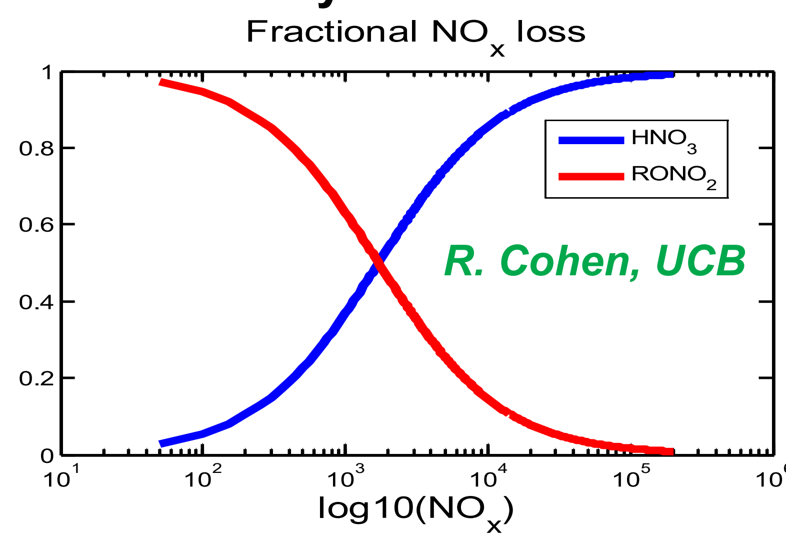
Coupling atmospheric chemistry to N biogeochemical cycle

- **Grasshopper effect, link to N_2O**



- **Need better understanding of organic nitrate chemistry**

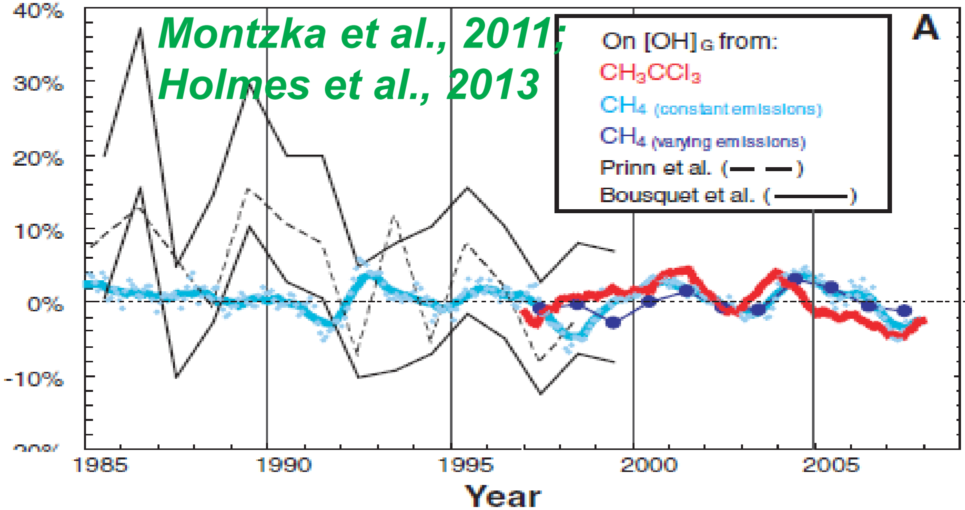




R. Cohen, UCB

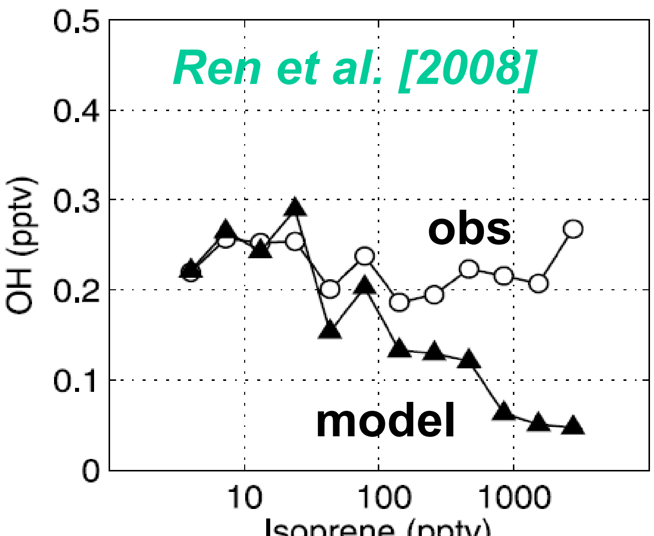
What factors control OH concentrations?

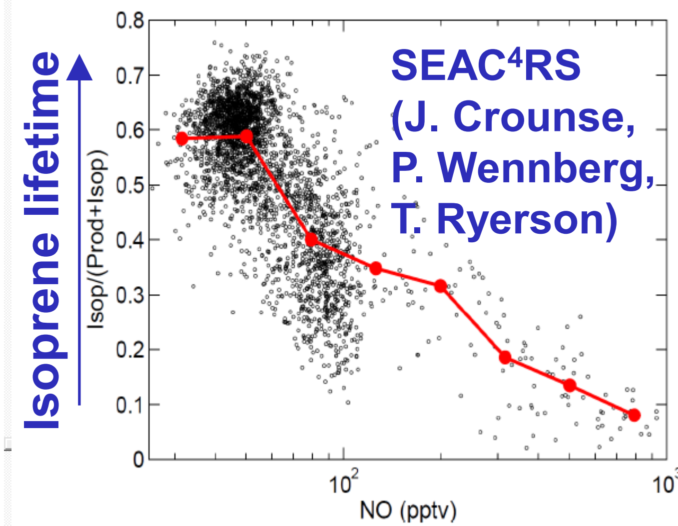
- **Trend in OH concentration inferred from methylchloroform**



- **No trend in past 30 years, IAV 2% - but this could well change in the future**
- **How do we maintain a global OH proxy record as methylchloroform disappears?**

- **Fundamental uncertainties in OH sensitivity to VOC-NO_x system**

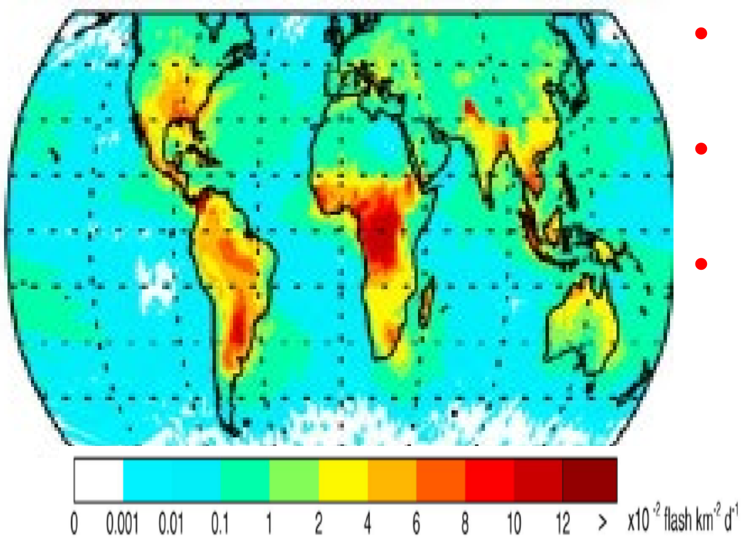




Need more confidence in OH measurements to test models

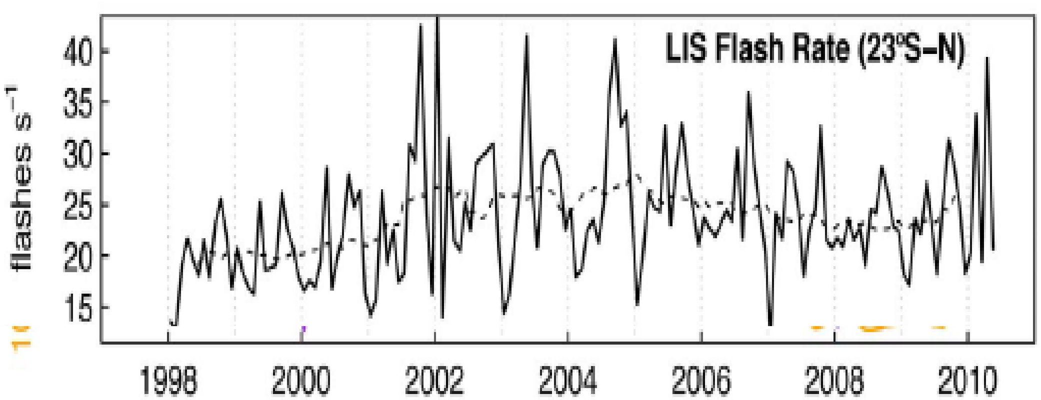
Lightning NO_x: poorly constrained source of ozone and OH

LIS/OTD lightning flashes, 1995-2005



- Lightning NO_x is the most powerful lever to change ozone and OH in models
- Yet there is no successful mechanistic model to predict lightning flash distributions
- Lightning NO_x source in range 1-20 Tg N a^{-1}; narrower range in models (3-8 Tg N a^{-1}) to fit ozone data

Interannual variability in LIS lightning flash rates

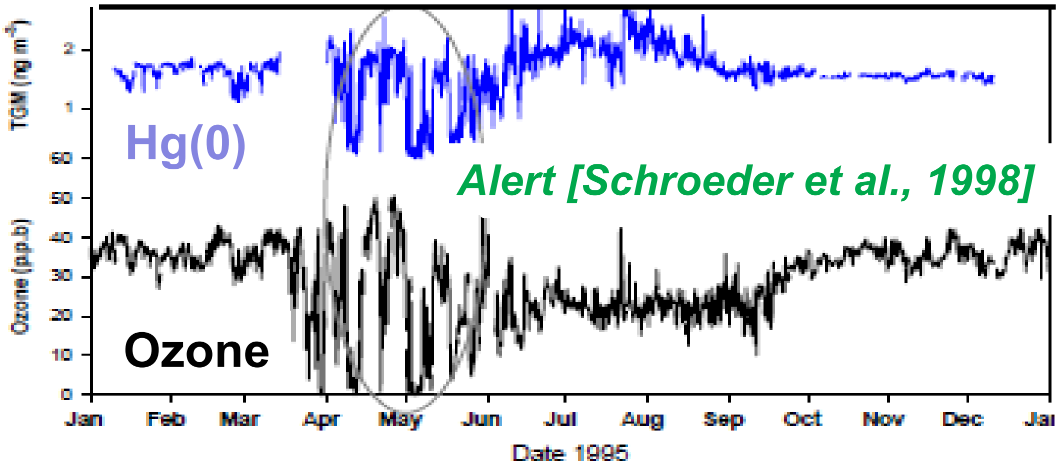


- What drives this variability and the associated NO_x source?
- What are the implications for the response of lightning to climate change?

Murray et al. [2012]

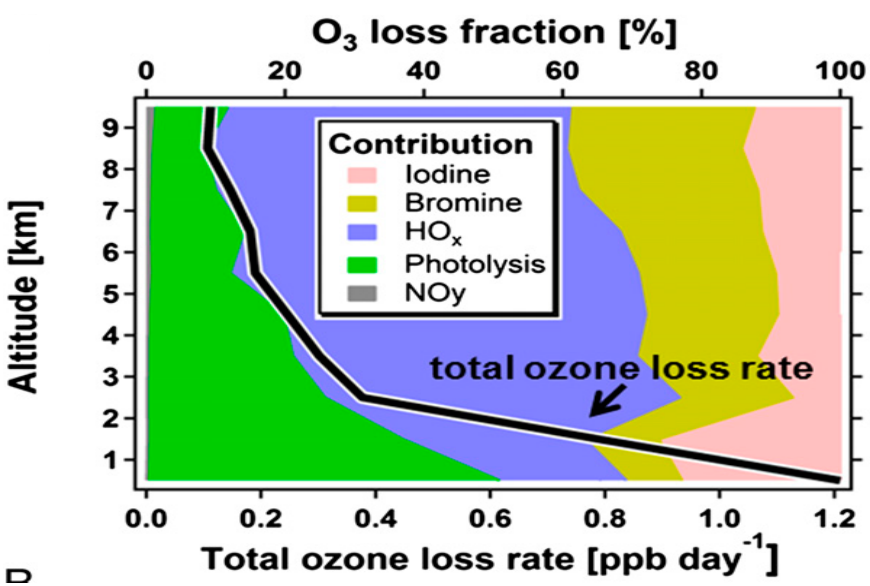
Importance of tropospheric halogen radicals

- **Br-driven ozone and mercury depletion in Arctic spring**



- **Also fast VOC oxidation by Cl**
- **What is the mechanism for BrO_x generation from sea ice, are there broader implications?**

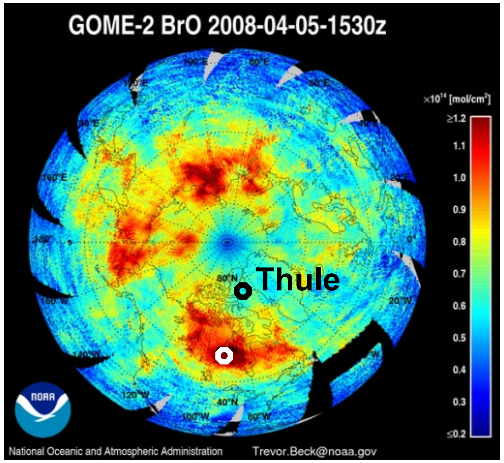
- **Ozone budget over SE Pacific from observed BrO and IO [*Dix et al., 2014]***



- **Br and I radicals at ppt levels are major sinks for ozone, NO_x, mercury**
- **But what are their sources, and could Cl also be important?**

Satellites have unexploited potential for tropospheric halogens

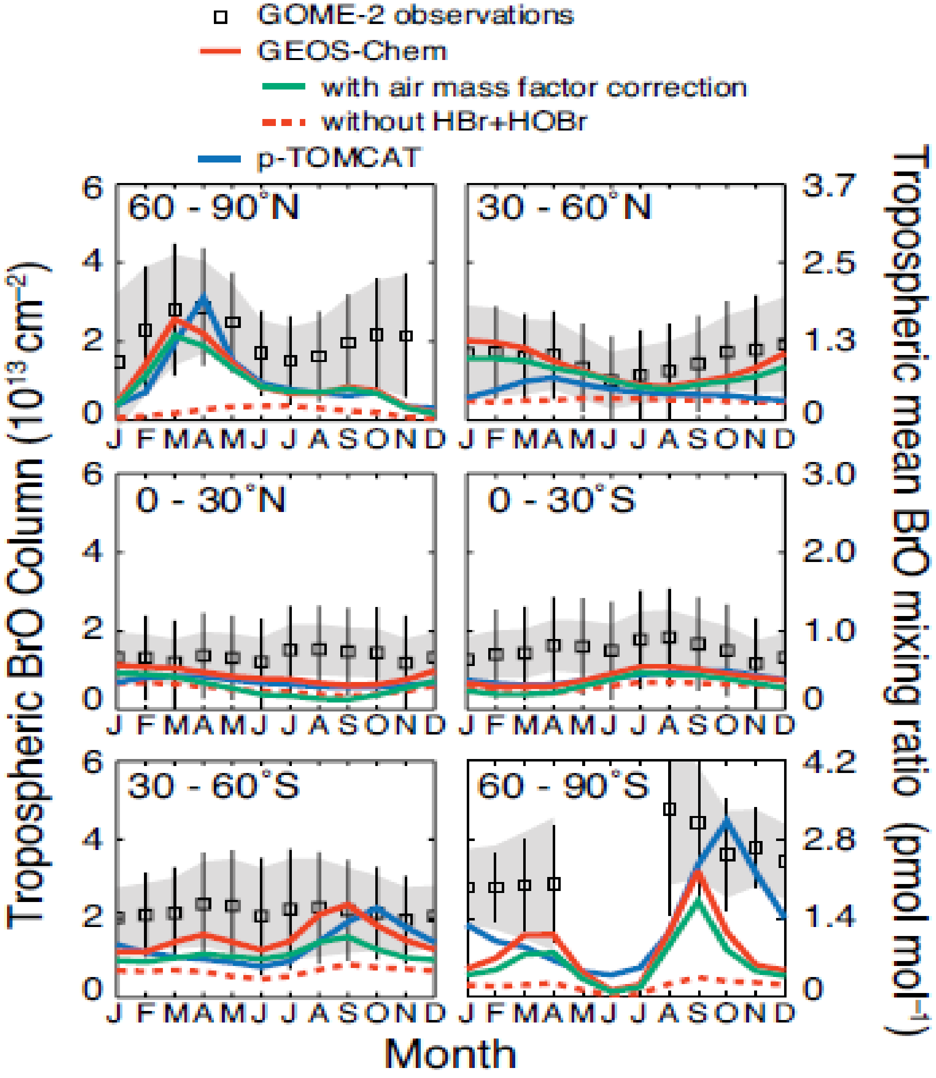
GOME-2 BrO in Arctic spring: Hot spots are not tropospheric



Salawitch et al. [2010]

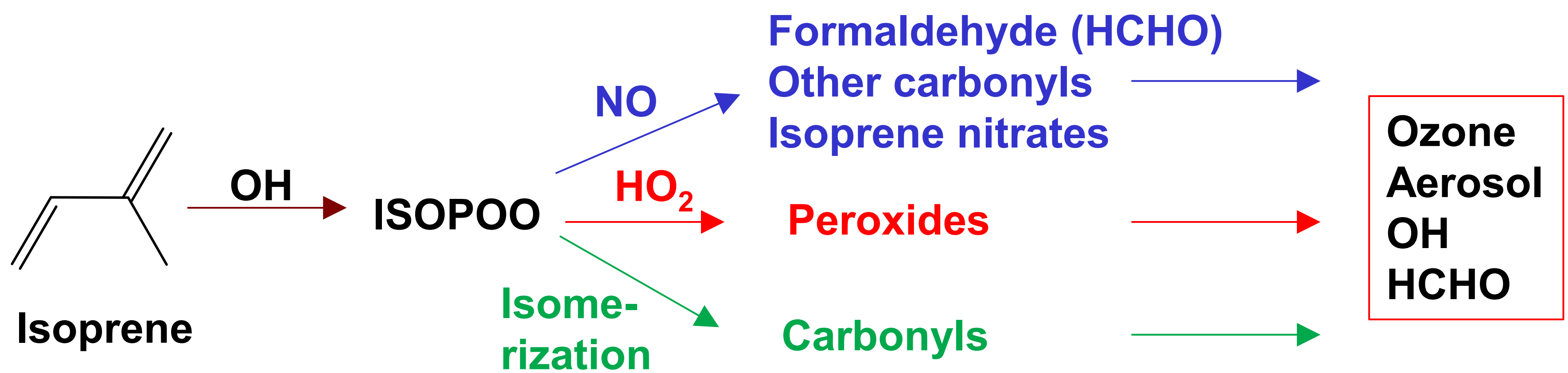
- More work is needed on retrievals of tropospheric BrO, IO, ClO
- Better understanding is needed of sources and chemistry through field and lab studies

Tropospheric BrO from GOME-2

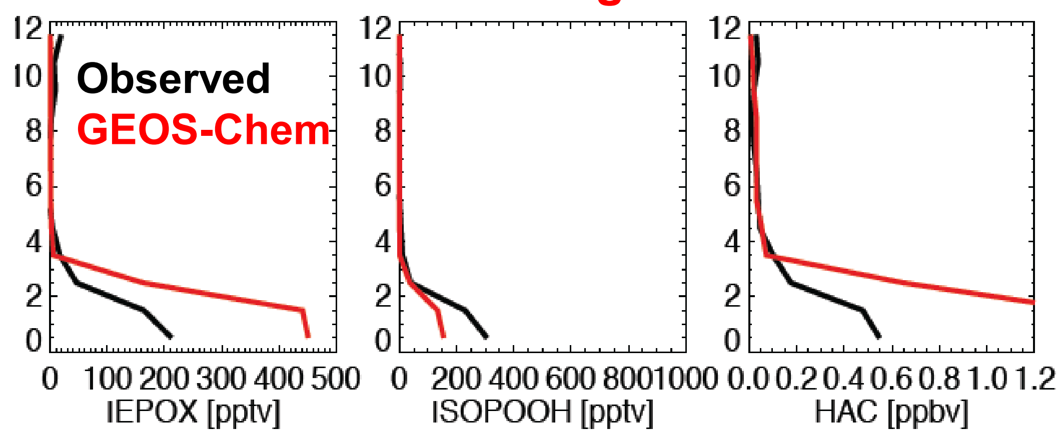


Theys et al. [2011]; Parrella et al. [2012]

Need to understand the cascade of products from oxidation of biogenic VOCs across NO_x regimes



Products of low-NO_x isoprene oxidation over Southeast US during SEAC[4]RS

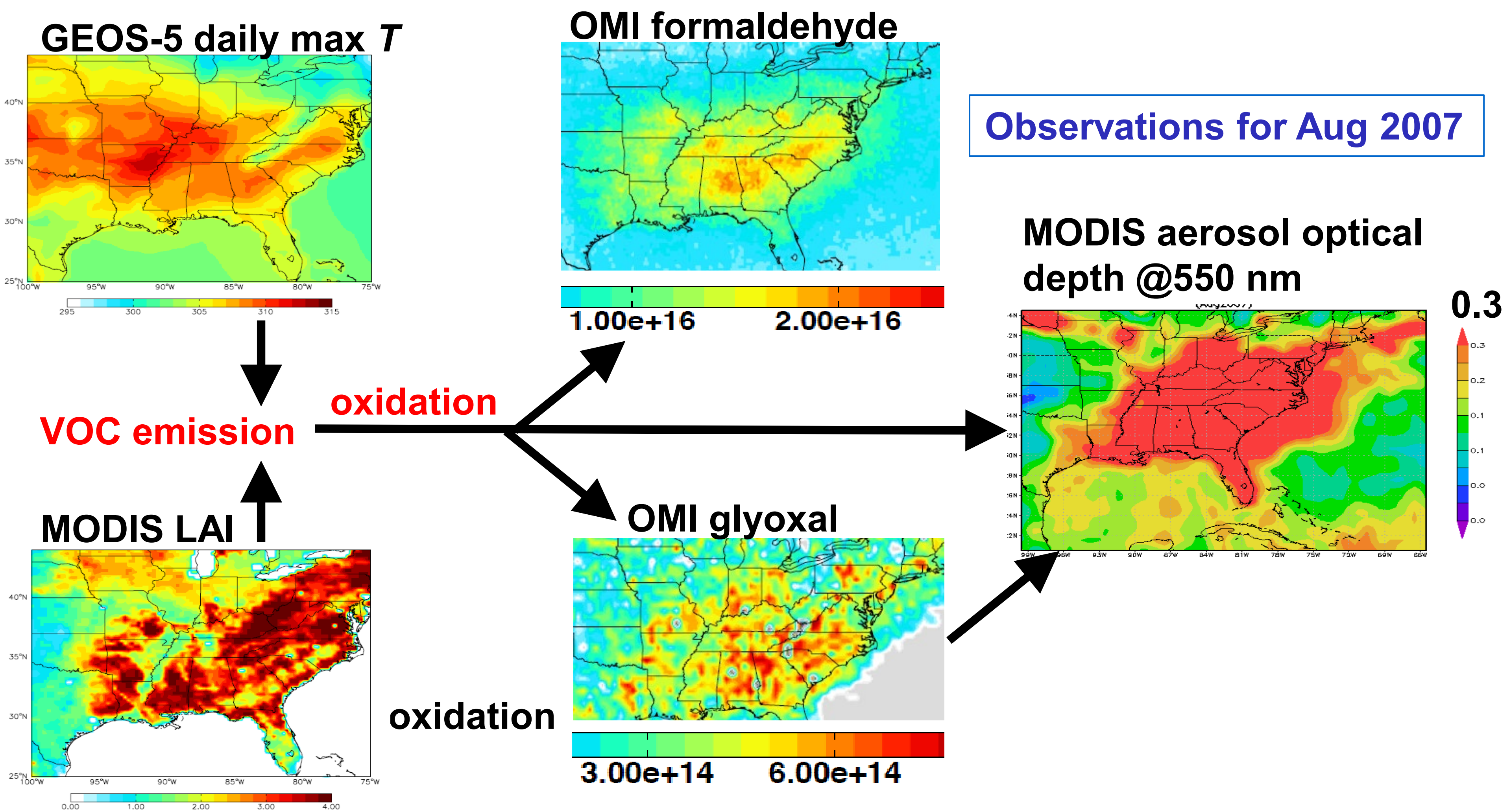


K. Travis, Harvard

PI: P.Wennberg

Oxidation cascade of other biogenic VOCs (e.g., terpenes) is even less understood

Satellites link VOC emissions, chemistry, aerosol formation

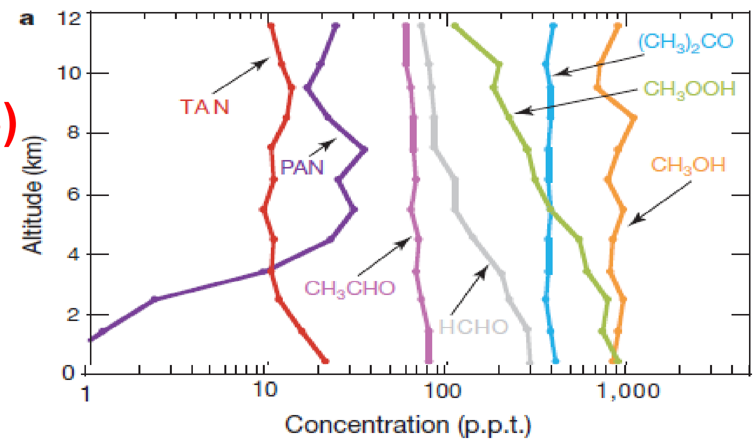


- Can we use formaldehyde and glyoxal to monitor changing emissions?
- What complementary information do they provide for VOC chemistry?
- How can we combine with aerosol data to constrain organic aerosol formation?

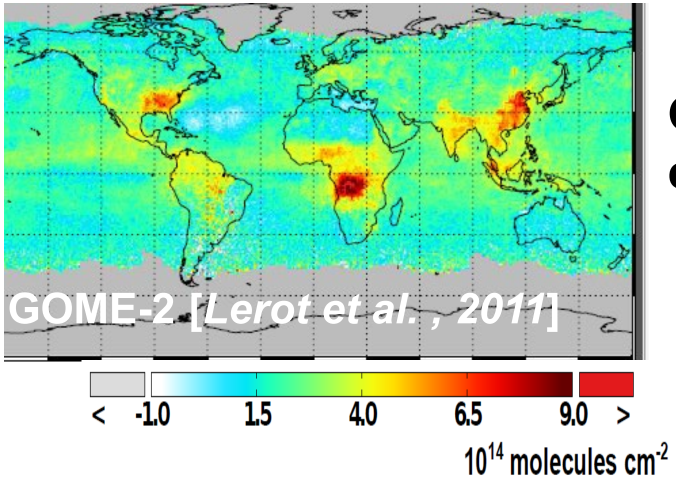
Oxygenated organics over oceans much higher than expected

South Pacific (PEM-TB)

Singh et al., 2001



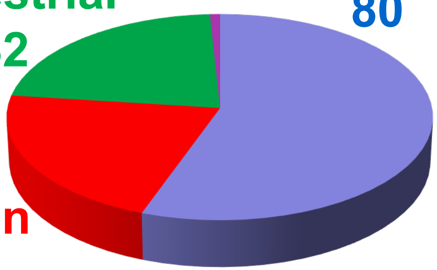
JJA 2007



Glyoxal column

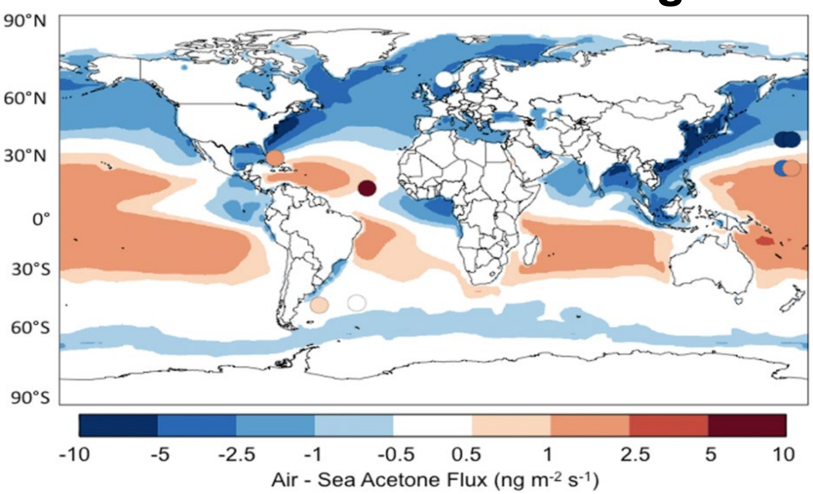
Need to better understand role of ocean as source and sink

Acetone sources (Tg a^{-1})



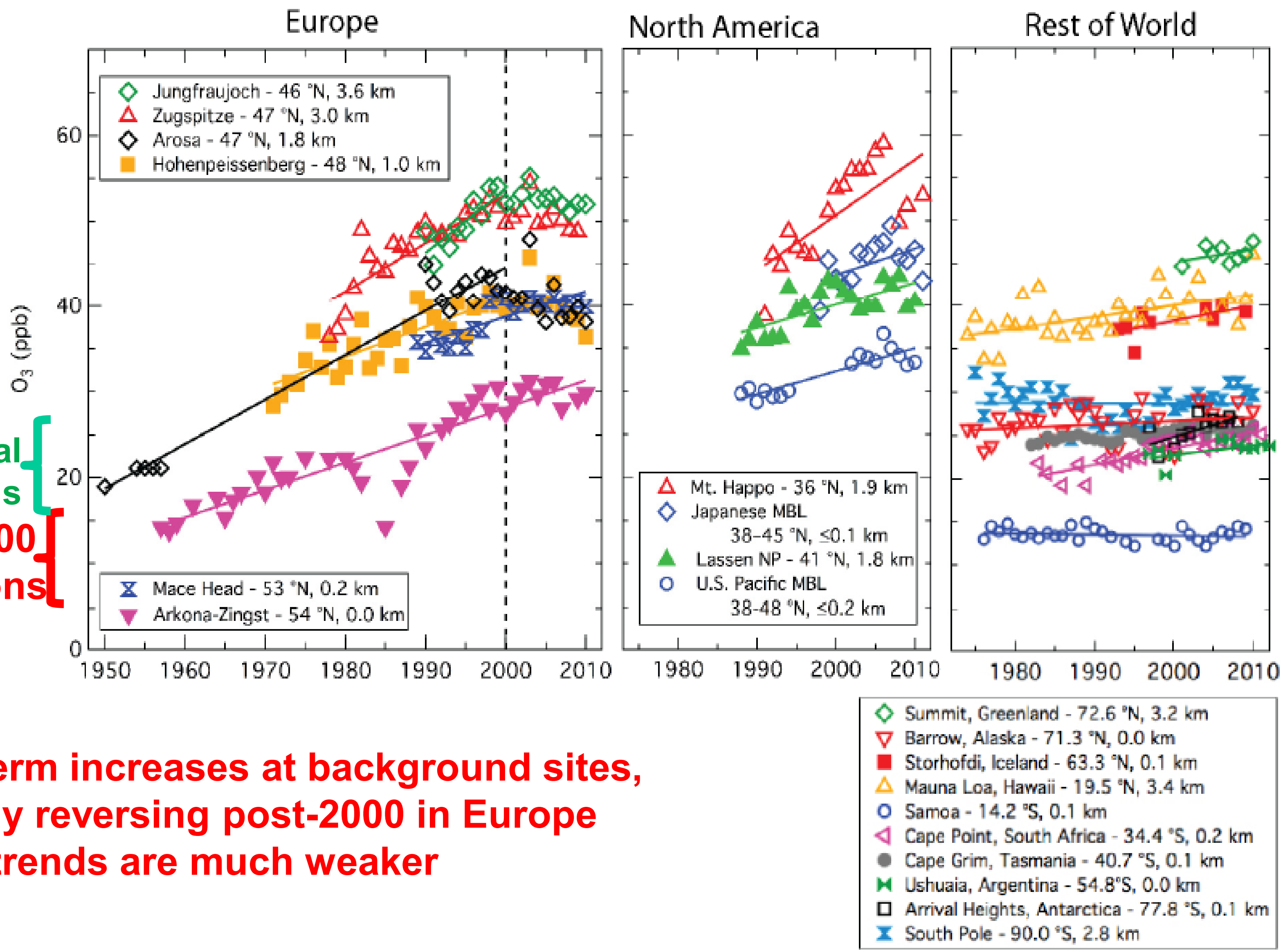
Fischer et al. [2012]

Acetone air-sea exchange



net uptake **net evasion**

What drives long-term trends in tropospheric ozone?



preindustrial models

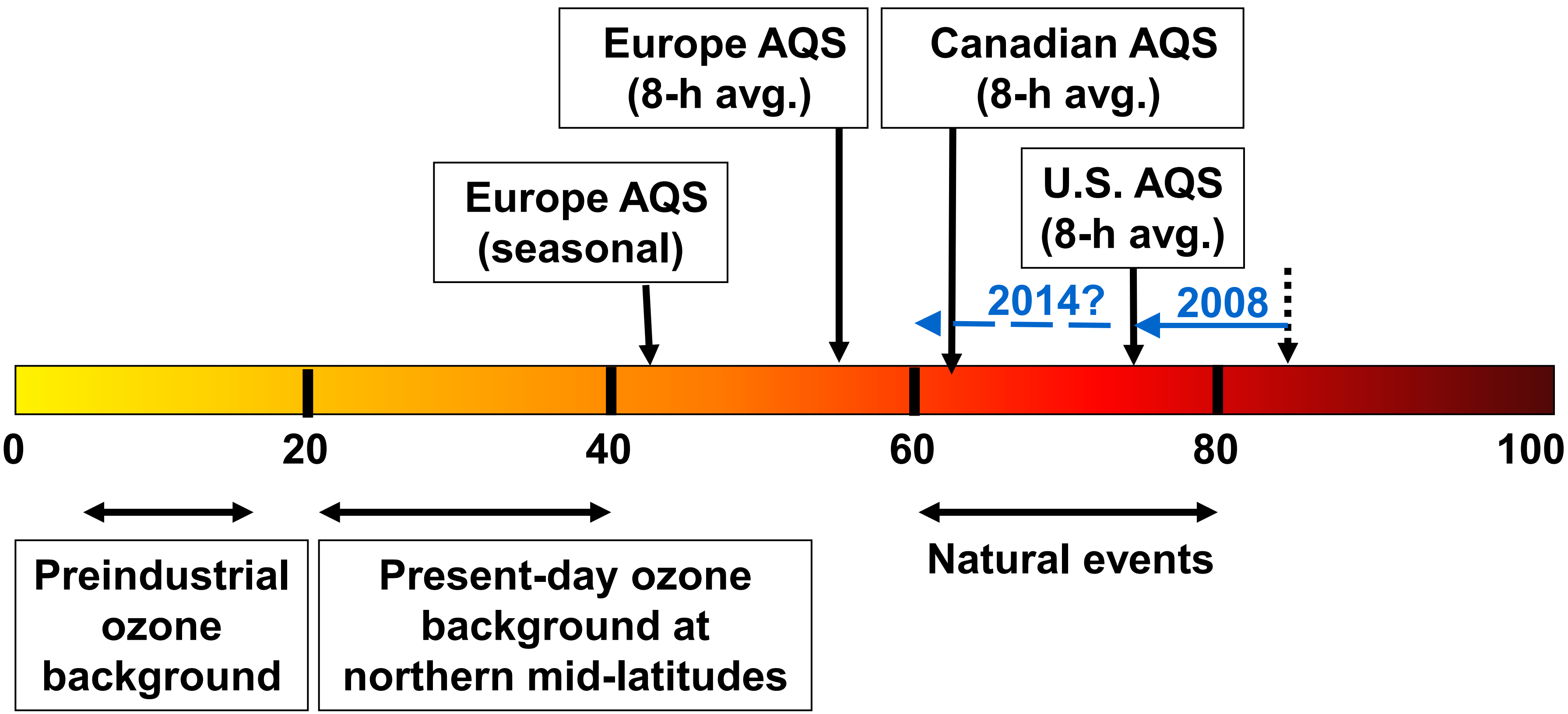
ca. 1900 observations

- **Long-term increases at background sites, possibly reversing post-2000 in Europe**
- **Model trends are much weaker**

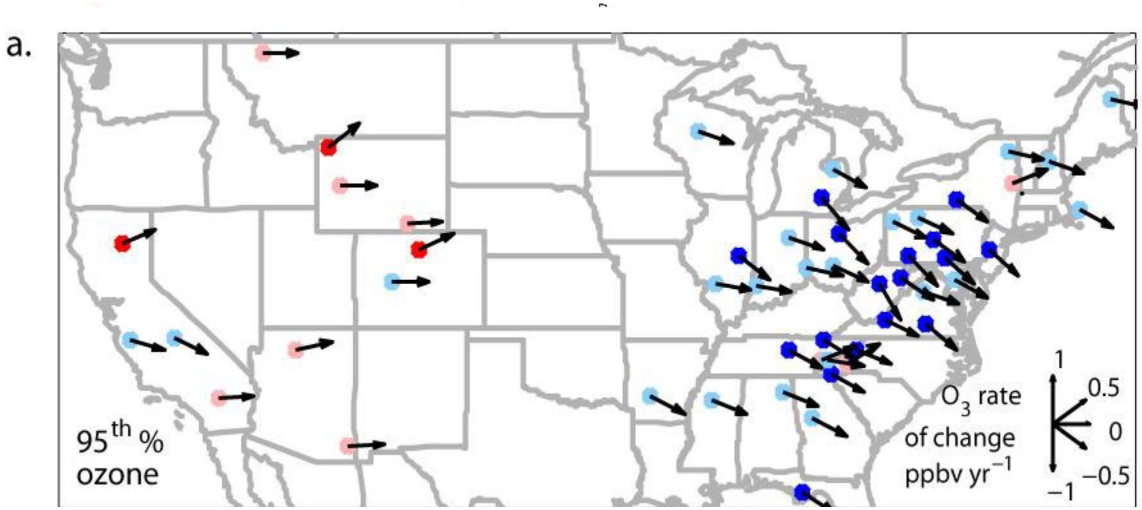
Cooper et al. [2014]; Parrish et al. [2014]

Implications of background ozone for air quality

What used to be a local/regional pollution problem is now becoming global

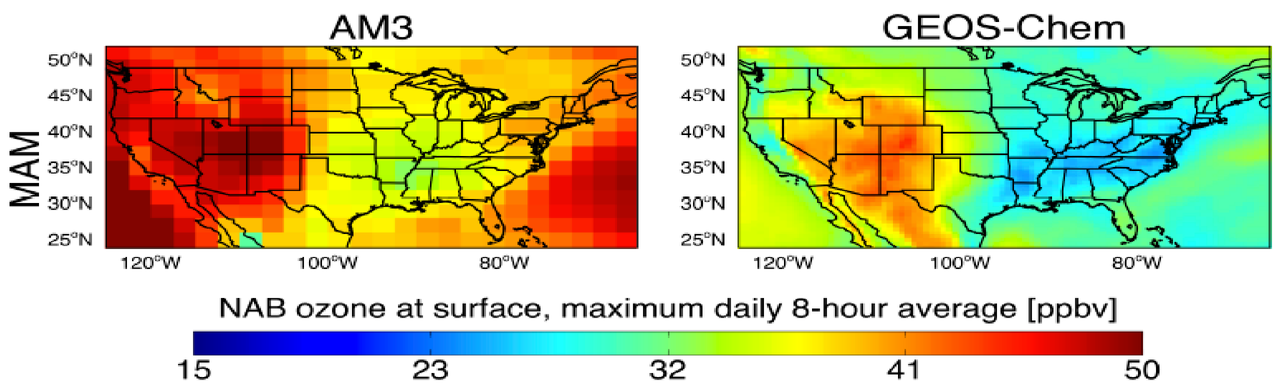


Intermountain West: new frontier for US ozone air quality



Spring 1990-2010 ozone trend *[Cooper et al., 2012]*

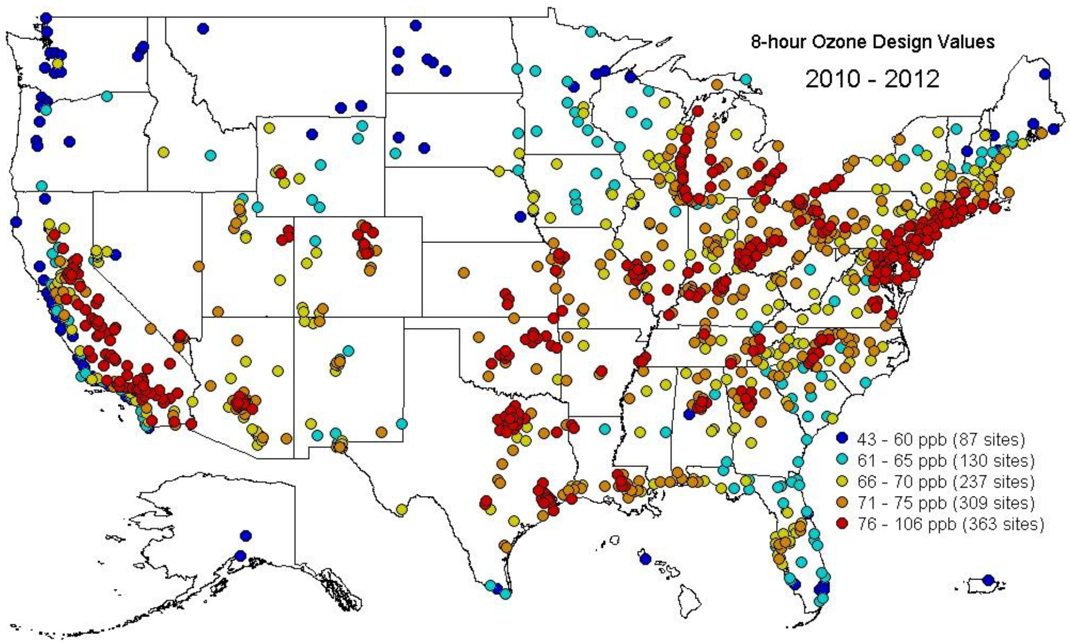
Large difference between models in estimates of N American background

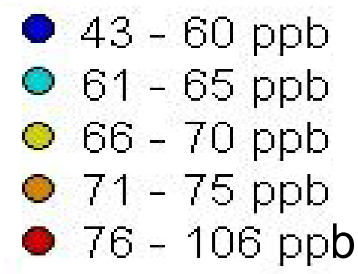


Fiore et al. [2014]

Need to better understand/quantify influences from stratospheric intrusions, fires, lightning, and ability of models to simulate high-ozone events

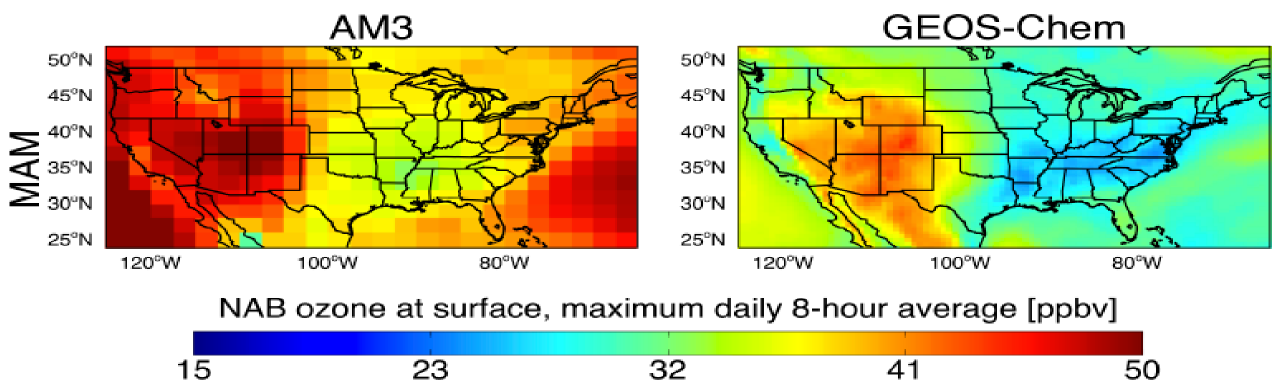
Intermountain West: new frontier for US ozone air quality





4th highest annual 8-h average ozone, 2010-2012 [EPA, 2014]

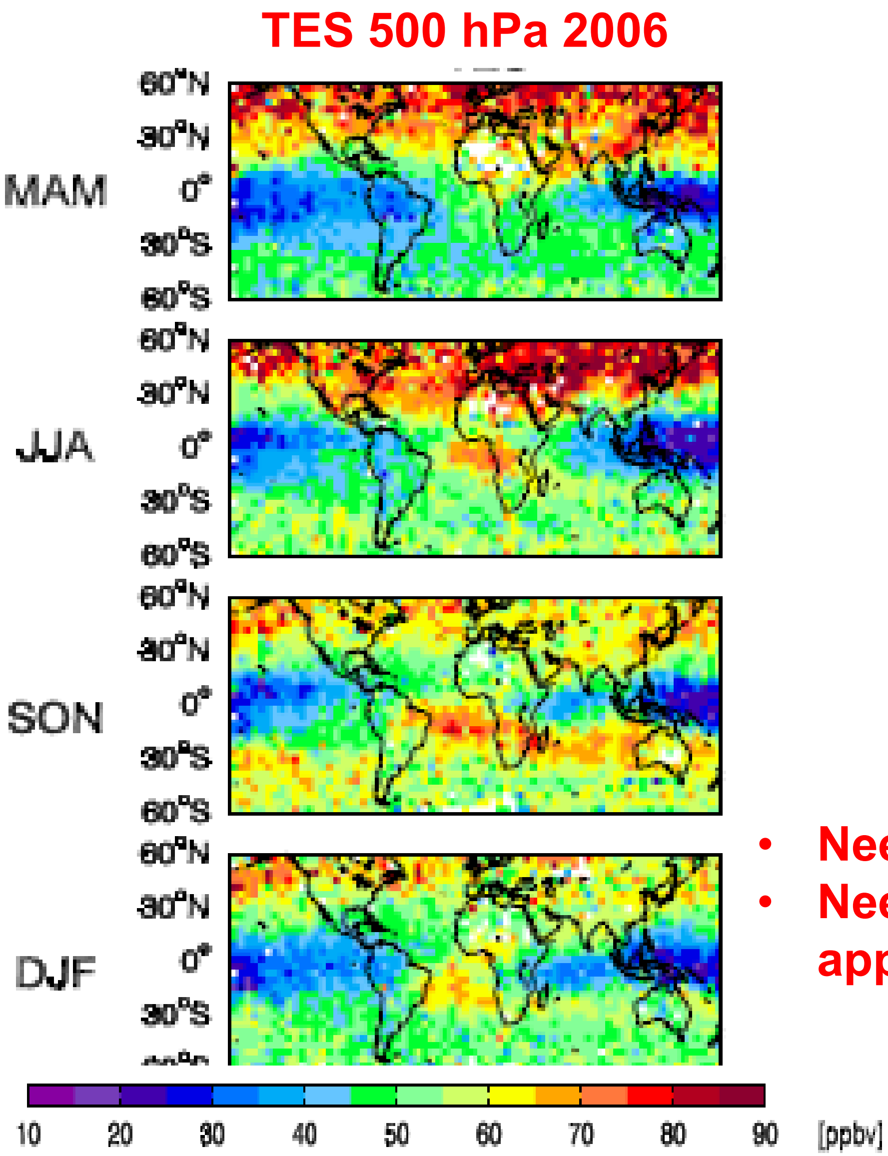
Large difference between models in estimates of N American background

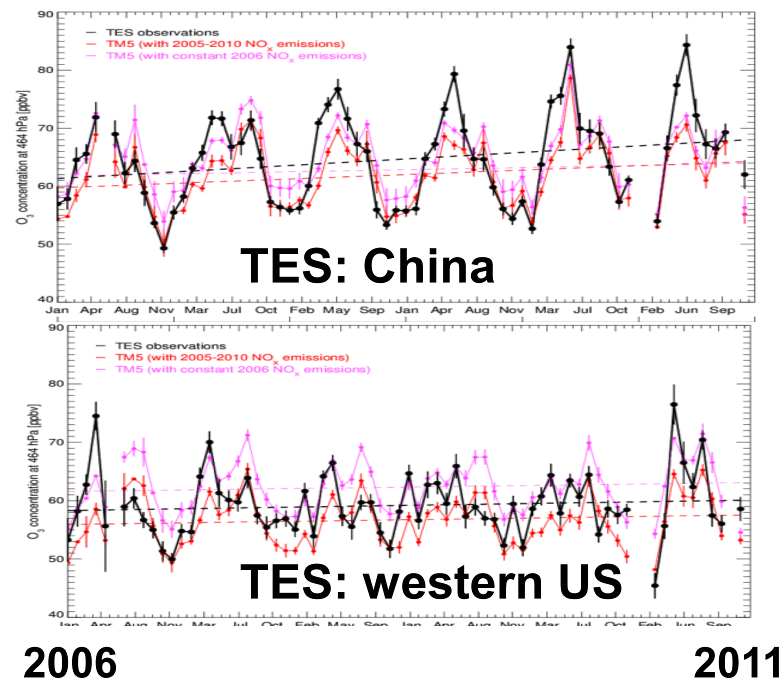


Fiore et al. [2014]

Need to better understand/quantify influences from stratospheric intrusions, fires, lightning, and ability of models to simulate high-ozone events

Tropospheric ozone measurement from TES and OMI is an Aura success story



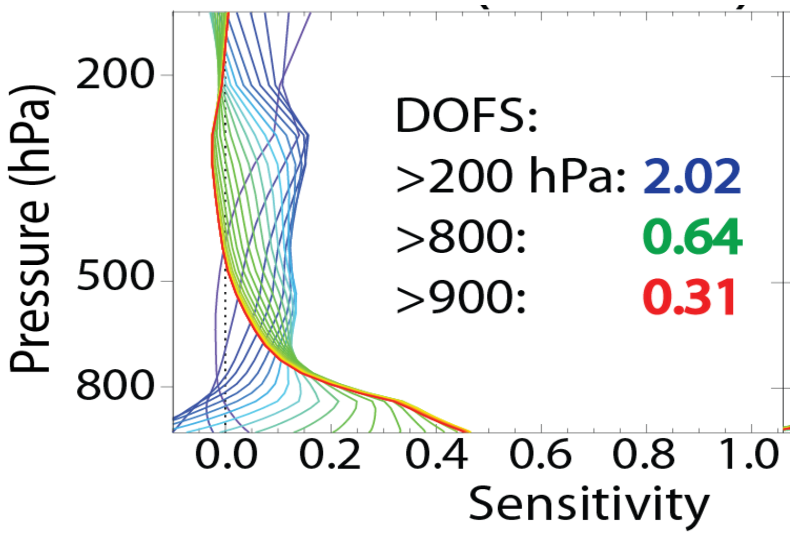


- Need to establish long-term record of trends
- Need better S/N, boundary layer sensitivity for application to events and air quality

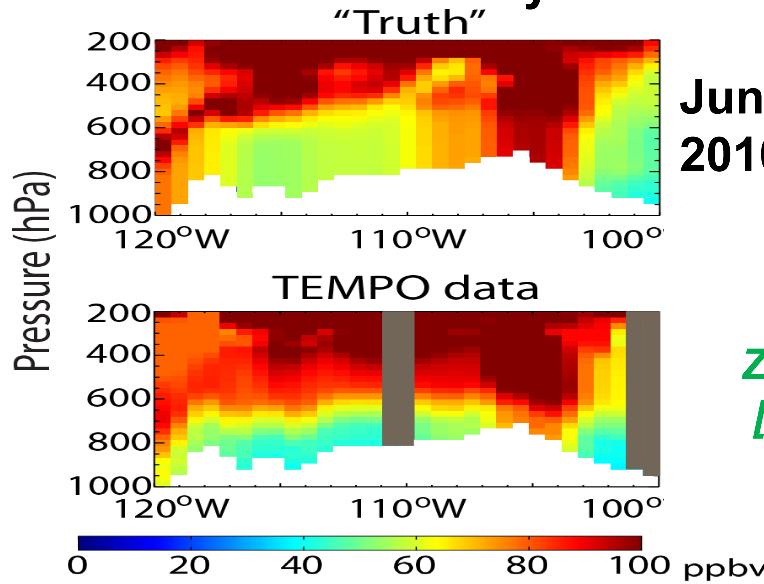
Zhang et al. [2010]; Verstraeten et al. [2014]

Geostationary observations and chemical data assimilation

Theoretical TEMPO sensitivity to surface ozone using UV+Vis



Simulation of stratospheric intrusion as would be seen by TEMPO

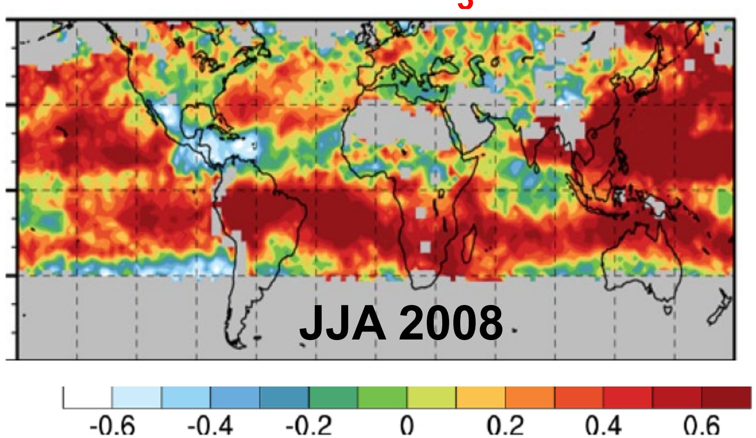


June 13, 2010

Zoogman et al. [2014]

Multi-constituent chemical data assimilation: a new frontier

Correlation of OMI O_3 w/ AIRS CO

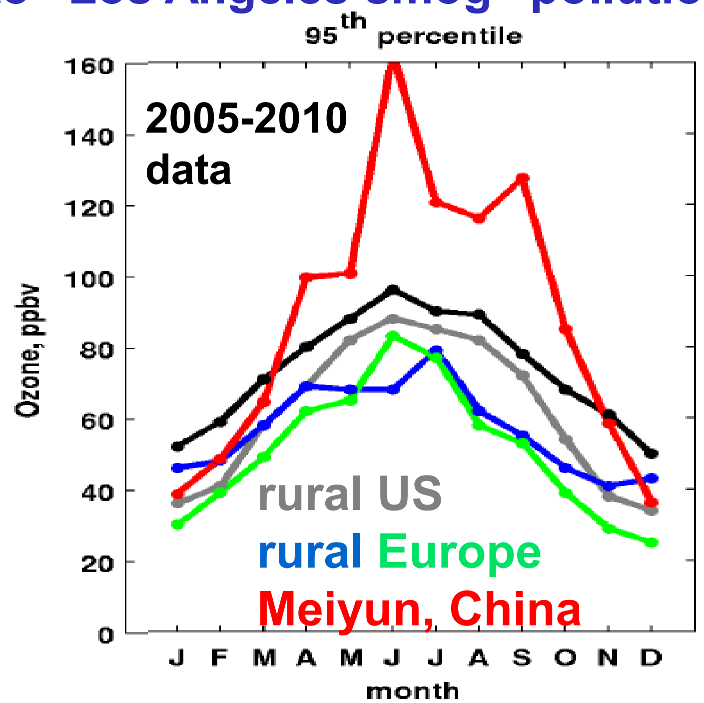


Need to develop capabilities for multi-platform assimilation of ozone and related species

Kim et al. [2013]

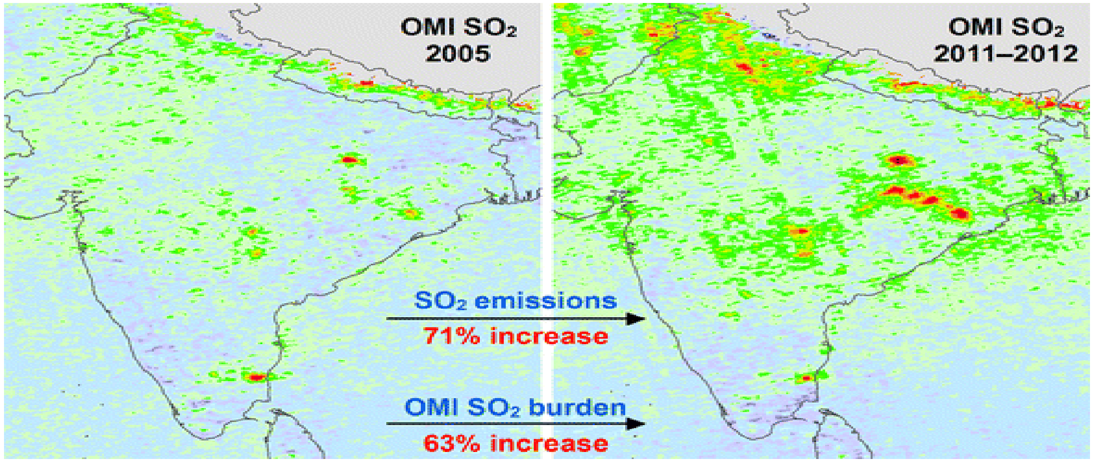
What is the future of air quality in the developing world?

China moving from "London fog" to "Los Angeles smog" pollution

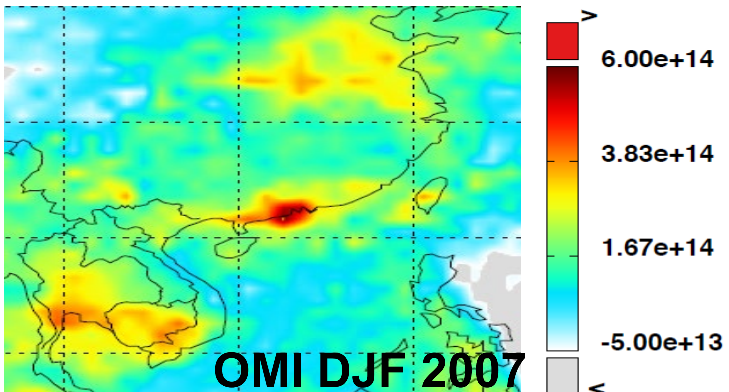


Satellites detect unexpected sources, monitor trends

OMI detects rising SO_2 emissions in India



Glyoxal hotspot in Guangzhou

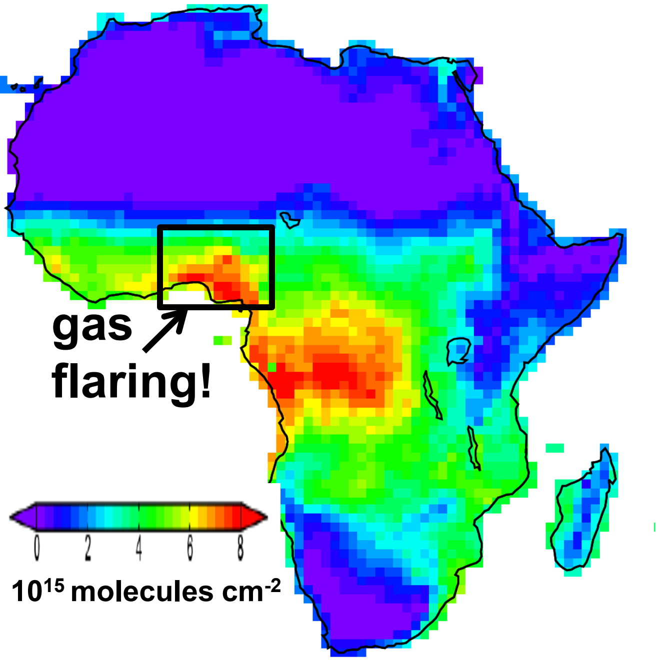


Planned coal power plants in SE Asia (T. Nace)

Cooper et al. [2014], Lu et al. [2013], C. Miller (Harvard)

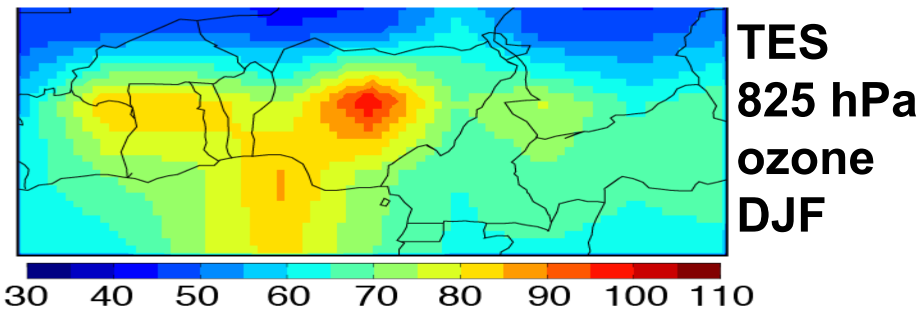
Nigerian air pollution revealed by satellite

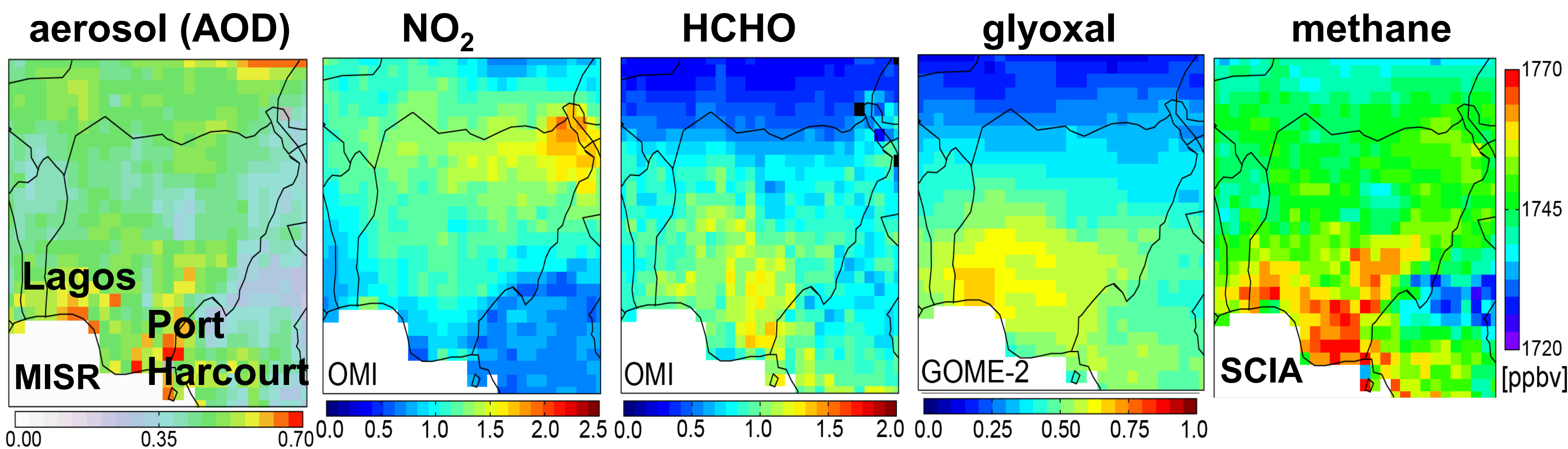
OMI formaldehyde 2005-2009



- Population: 170 million (+3% a^{-1})
- GDP: $270 billion (+7% a^{-1}) – oil!
- Most natural gas is flared
- >80% of domestic energy from biofuel, waste

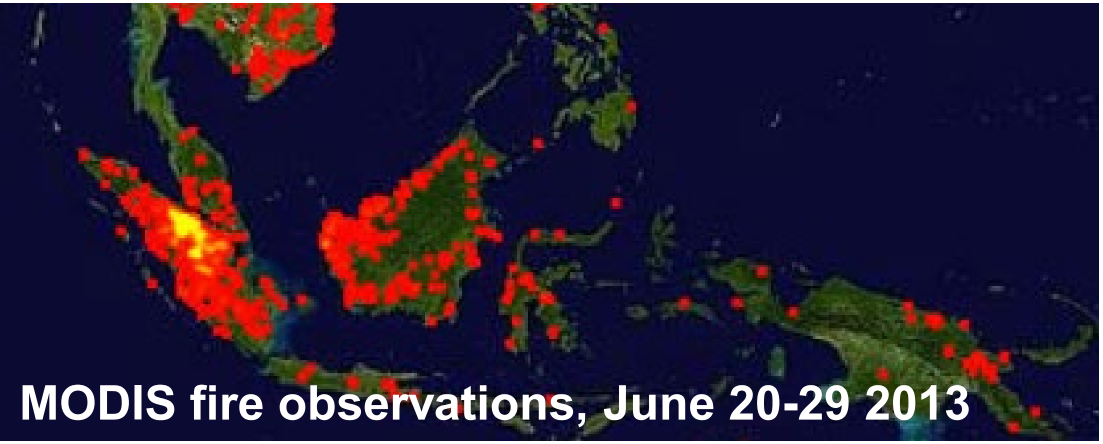
An unusual mix of very high VOCs, low NO_x –
What will happen as infrastructure develops?

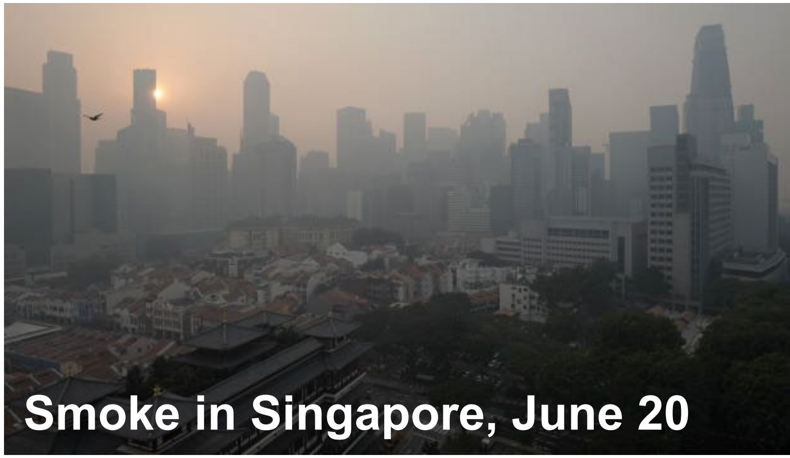




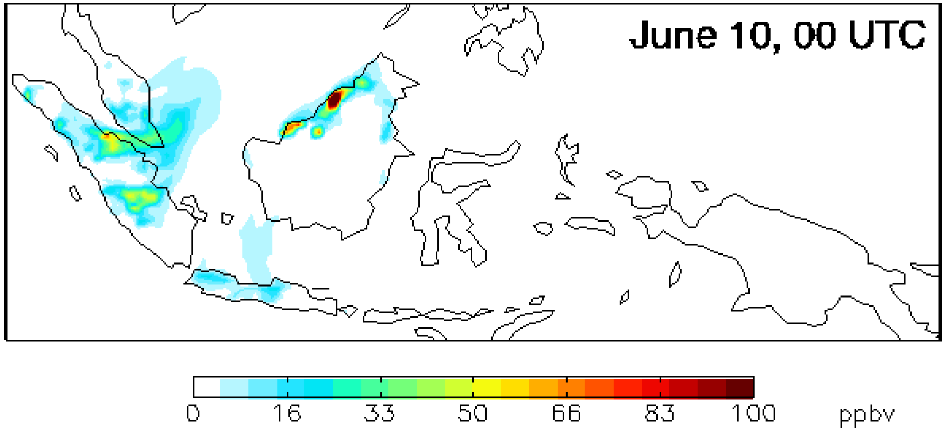
Marais et al., 2014

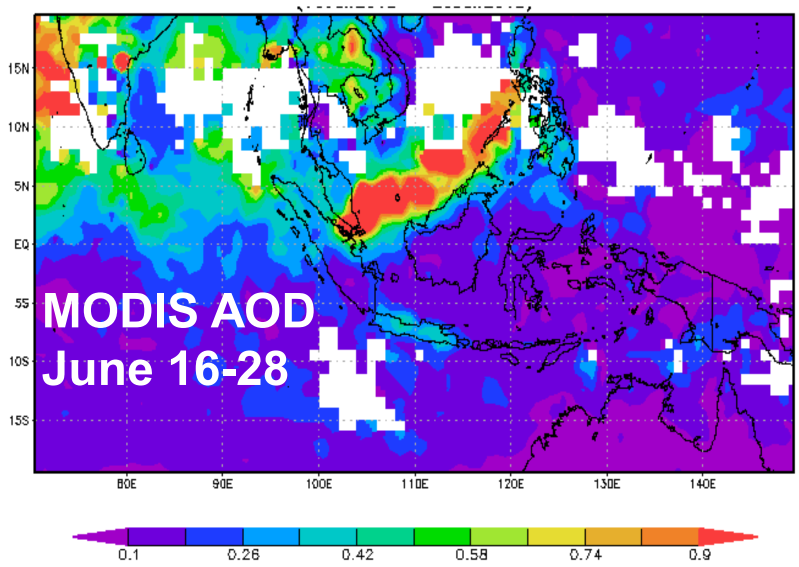
Tropical biomass burning: a major threat to public health





Smoke transport simulated by GEOS-Chem





Can we use assimilated satellite data to quantify surface aerosol concentrations, Issue air quality warnings?

Patrick Kim and Shannon Koplitz, Harvard

Geostationary constellation (2018-2020)

Addresses air quality problems of the developed world but gives short shrift to the rising problems of the developing world