UNIVERSITY OF CAMBRIDGE

National Centre for Atmospheric Science
NATURAL ENVIRONMENT RESEARCH COUNCIL

Long-Range Transport of Ozone and its Precursors

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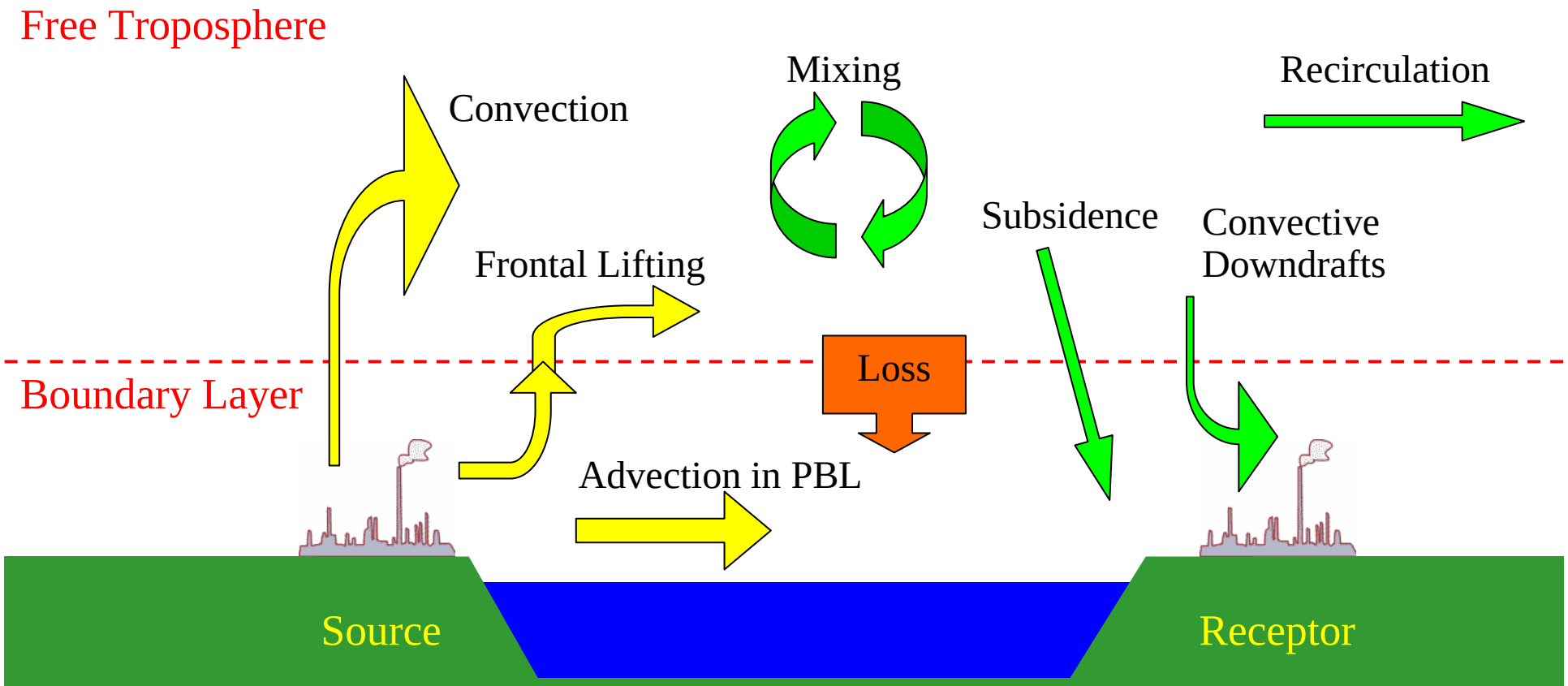
Long-Range Transport of Ozone

- What is the impact of remote sources on regional O_3?
  - Contributions to incoming O_3 over a region ("background O_3")
  - How much do remote sources contribute to high O_3 episodes?
  - How well do we understand long-range source-receptor relationships?
  - Can we perform source-attribution for surface O_3?

- How will these impacts change in future?
  - Changes in emissions (and chemistry)
  - Changes in meteorology

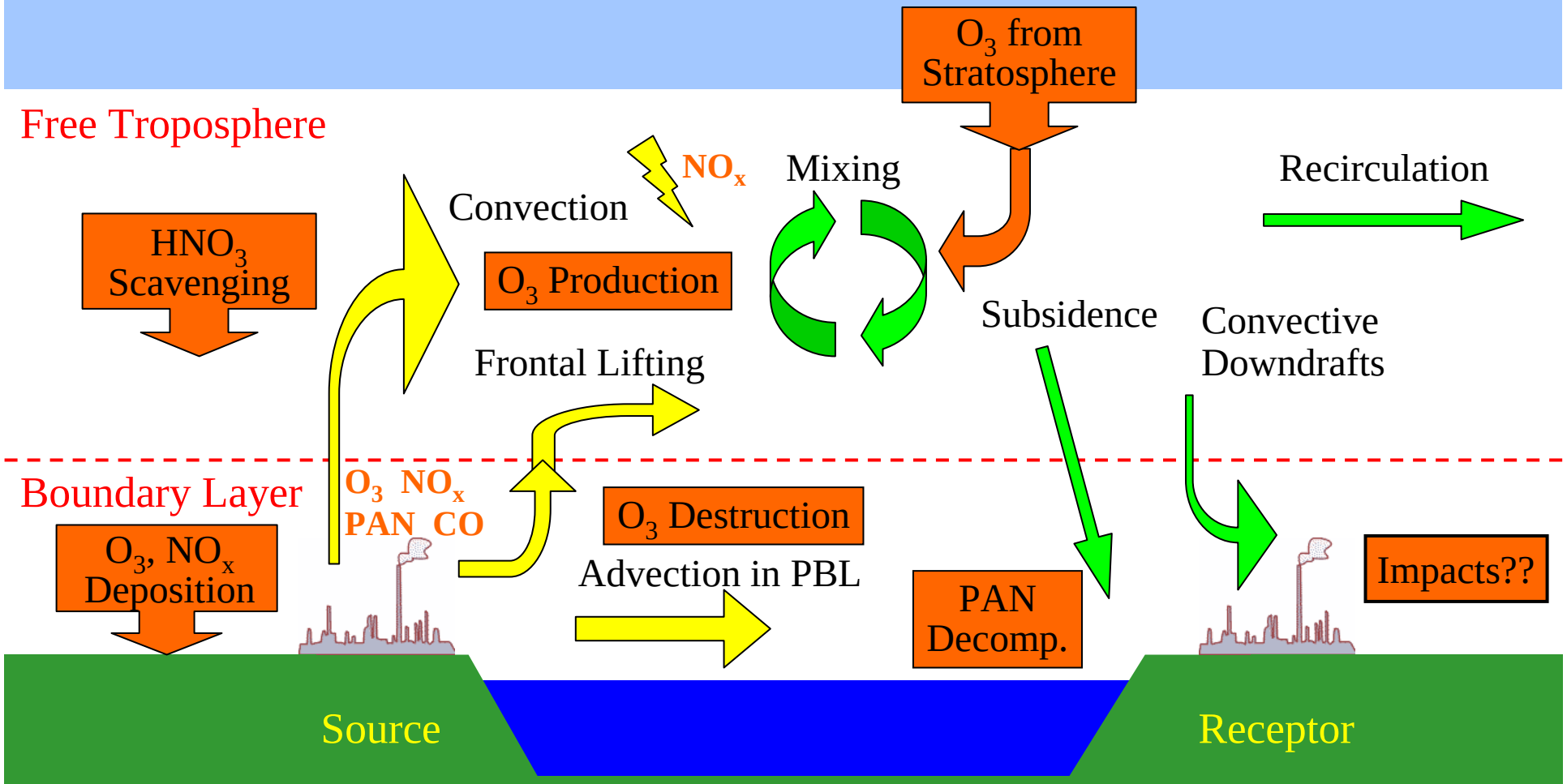
- How reliable are our estimates?
  - Examine sources of global model uncertainty

Mechanisms for Intercontinental Transport



- How important are the different pathways?
- How do they differ for different species? Or regions?

Mechanisms for Intercontinental Transport

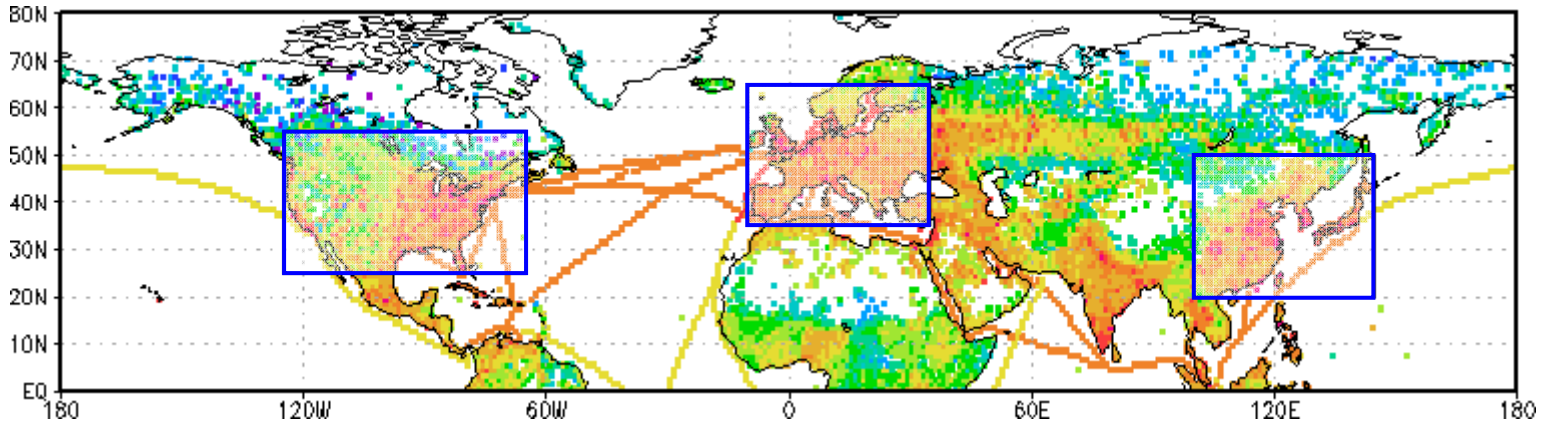


O_3

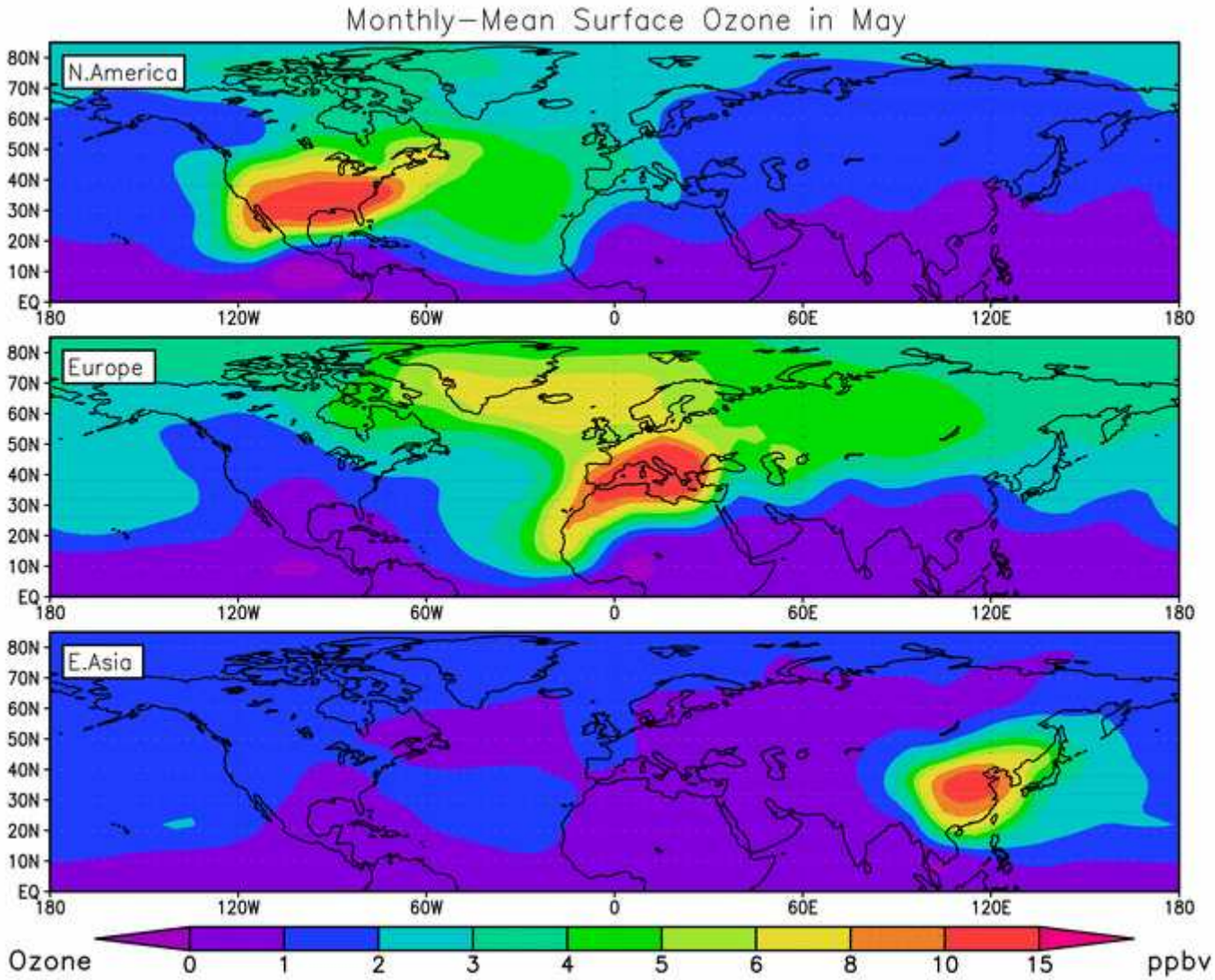
- Chemical changes sensitive to transport pathways
- Unraveling these processes remains a challenge!

Intercontinental Transport of Ozone

Current Industrial/Fossil Fuel NO_x Emissions



- Industrial emission regions located at similar latitudes
  - Transport times about 1 week; chemical lifetime 3-4 weeks
- How much do major emission regions affect each other?
  - How much do they contribute to background O_3?
  - Could this affect attainment of air quality standards?
- Explore O_3 production and transport with a 3-D global CTM
  - Single-region <u>anthropogenic</u> emissions perturbation experiments



- Photochemistry active in summer
- Transport most efficient in spring

} Largest O_3 impacts in late spring

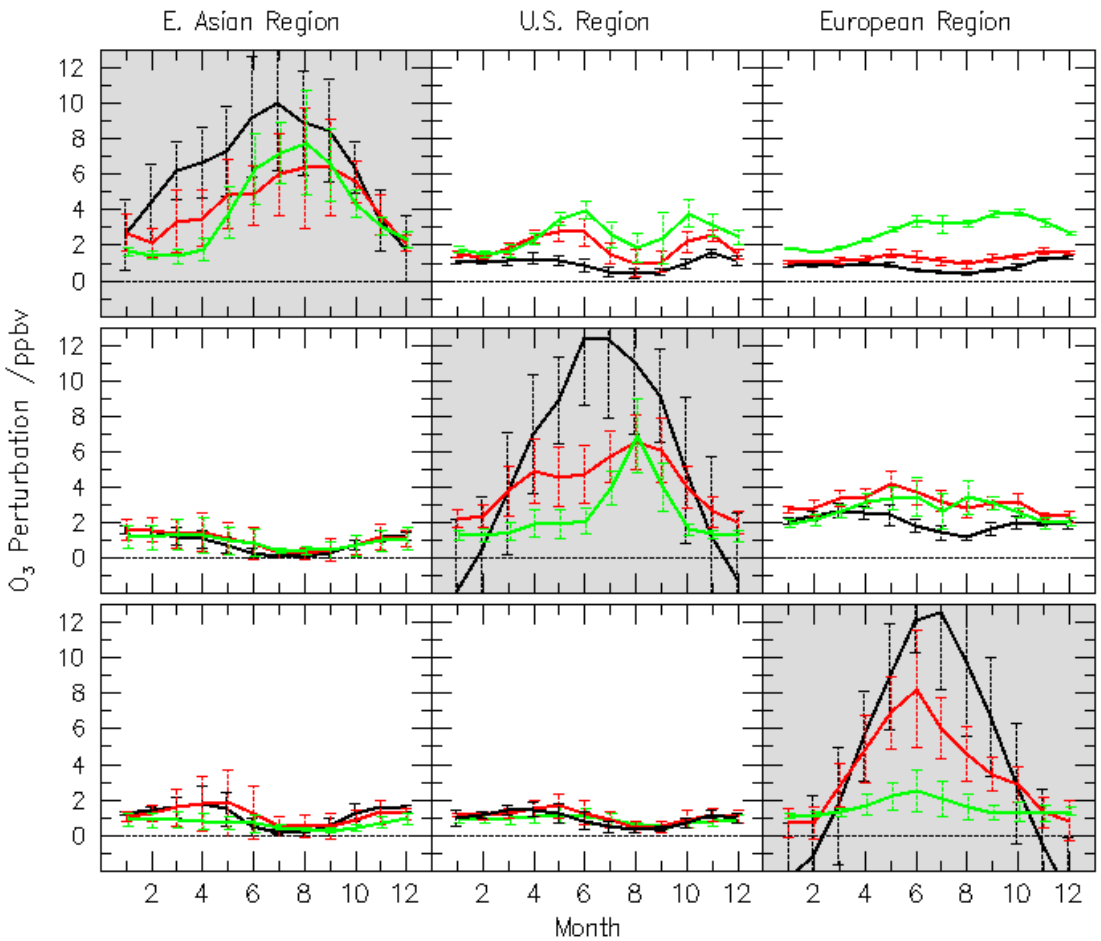
Wild and Akimoto [2001]

Source-Receptor Matrix

East Asian Emissions

US Emissions

European Emissions



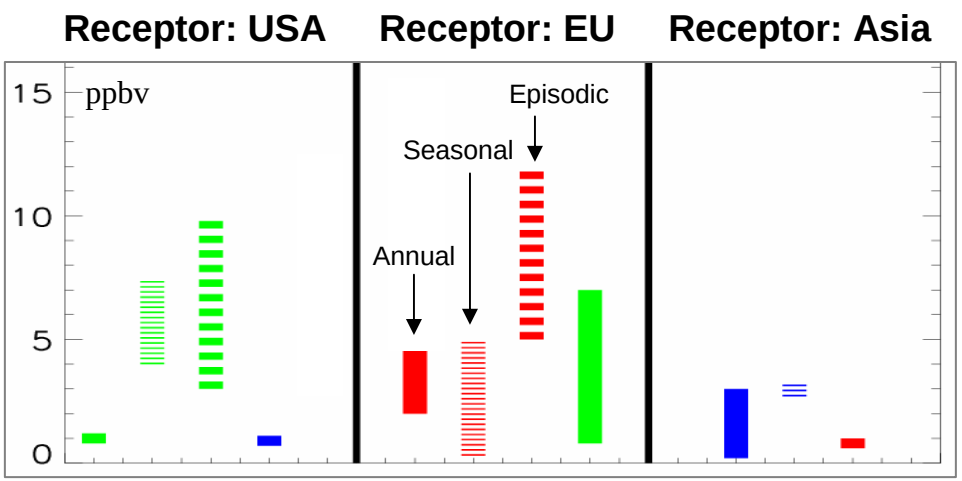
----- 200 hPa
----- 650 hPa
----- Surface

- Major emission regions directly affect each other
  - Upwind sources contribute 1-2 ppbv to surface background O_3
  - This is sufficient to affect attainment of air quality standards
  - Study now being repeated with many models within HTAP framework

Wild and Akimoto [2001]

Review of Recent LRT Studies

Receptor	Source	O3 enhancement (ppbv)	Method	Reference
Europe	U.S.	2	a	*Wild and Akimoto, 2001*
Europe	North America	2-4	b	*Auvray and Bey, 2005*
Europe	North America	5.8–12.2	c	*Derwent et al., 2004*
Europe	North America	3-5 (summer avg); 10-12 (events)	b	*Guerova et al., 2006*
Europe (summer)	North America	2-4 (daytime mean); 5-10 (events)	b	*Liu et al., 2002*
Europe (yearly mean)	North America	18 (Atlantic fringes)10-15 (Central Europe)	c	*Derwent et al., 2002*
Europe (high altitudes)	North America	10-12 (events)	d	*Huntreiser et al., 2005*
Mace Head, Ireland	North America	0.4 (winter); 0.2 (spring); -0.3 (summer); -0.9 (fall)	e	*Derwent et al., 1998*
Europe	Asia	0.8	a	*Wild and Akimoto, 2001*
Europe	Asia	9 (Atlantic fringes); 5-7 (central Europe)	c	*Derwent et al., 2002*
Europe	Asia	3.5–6.6	c	*Derwent et al., 2004*
Europe	Asia	1.5-4	b	*Auvray and Bey, 2005*
U.S.	Asia	1	a	*Wild and Akimoto, 2001*
Northwestern U.S. (spring)	Asia	4 (mean); 7.5 (max)	b	*Berntsen et al., 1999*
Northwestern U.S. (spring)	Asia	4	f	*Jaegle' et al., 2003*
Western U.S. (spring)	Asia	3-10 (range during Asian pollution events)	g	*Yienger et al., 2000*
U.S.	Asia (future)	2-6 (western U.S.); 1-3 (eastern U.S.); highest April-June	h	*Jacob et al., 1999*
Western U.S. (spring)	Asia (future)	30-40 (max during Asian pollution events)	I	*Yienger et al., 2000*
U.S.	Europe	0.9	a	*Wild and Akimoto, 2001*
U.S. (summer)	Asia + Europe	4-7 (typical afternoon range); 14 (max)	b	*Fiore et al. 2002*
U.S.	Background (1980-1998)	3-5 (spring, fall)	j	*Lin et al., 2000*
U.S. (Mar - Oct 2001)	Background (hemispheric pollution)	4-12 (typical range in afternoon mean)	k	*Fiore et al., 2003*
U.S. (summer)	Background (anthropogenic methane)	6 (afternoon mean)	l	*Fiore et al., 2002*
West Coast U.S.	Background (1984-2002)	10	m	*Jaffe et al., 2003*
Asia	Europe	1.1	a	*Wild and Akimoto, 2001*
East Asia (spring)	Europe	3 (daytime mean)	b	*Liu et al., 2002*



Source Regions: **N. AMERICA** **ASIA** **EUROPE**

Difficult to compare due to

- different methods
- different metrics
- different region definitions

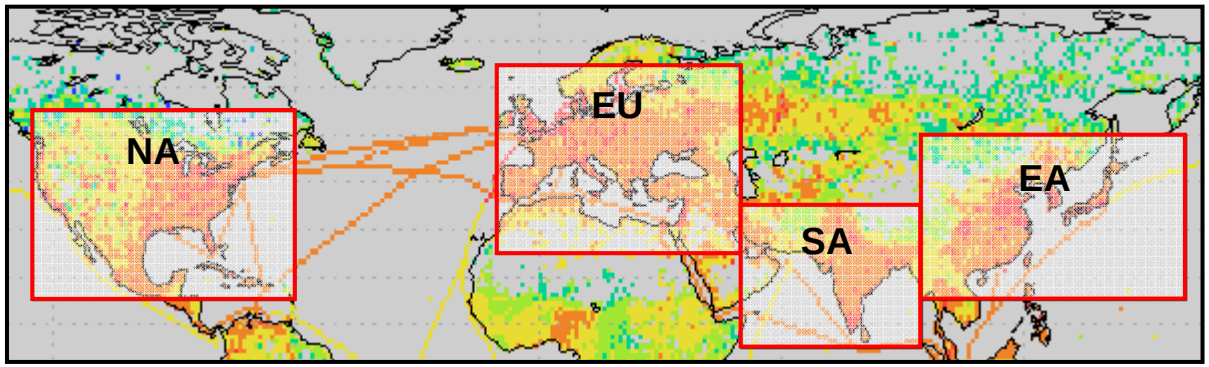
Estimates are from tables in *Holloway et al., ES&T, 2003* and *Fiore et al., EM, 2003*, updated by Tracey Holloway, 2006.

Task Force on Hemispheric Transport of Air Pollution

Model Intercomparison

(27 models involved)

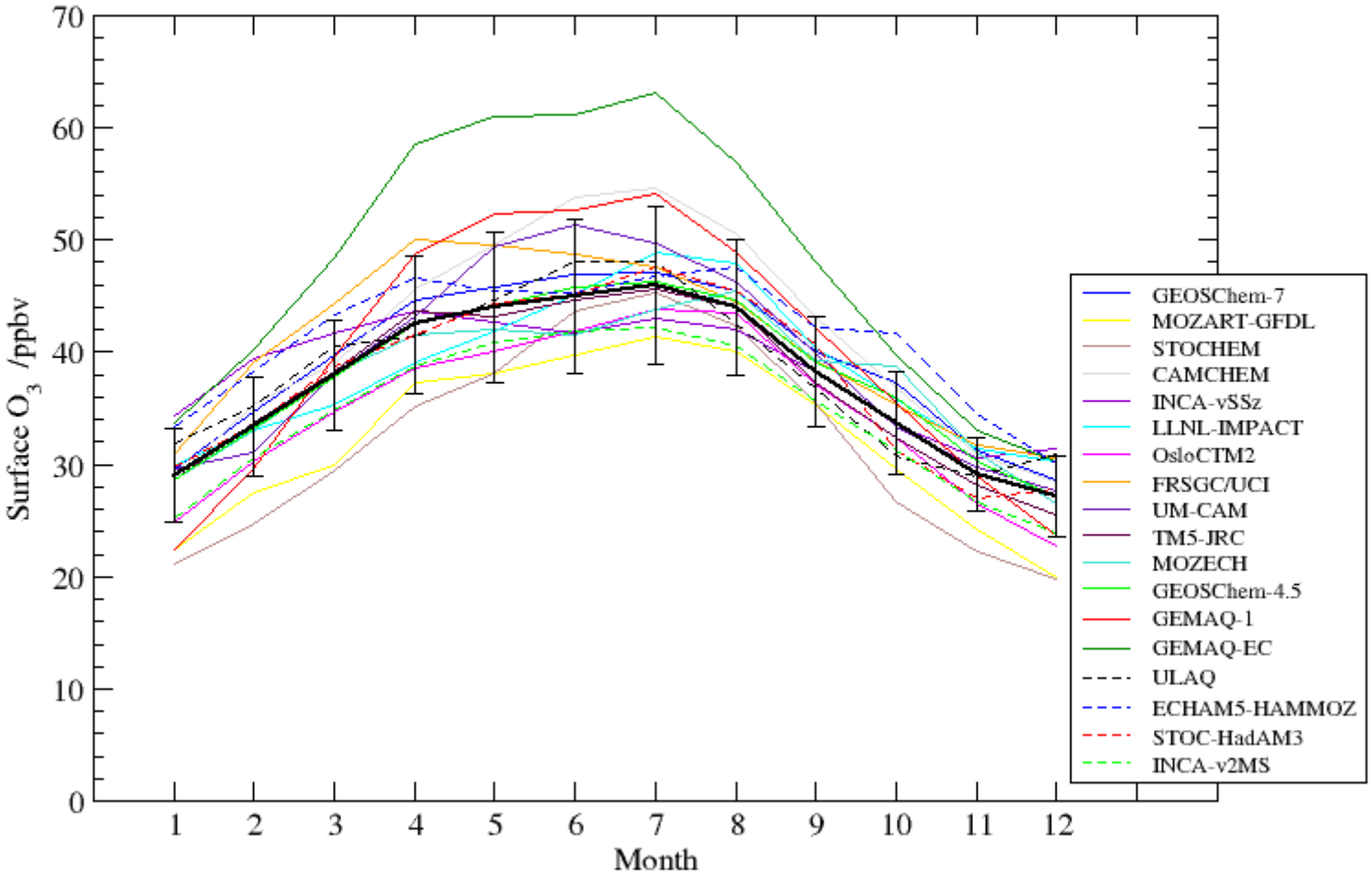
- Objectives
  - Quantify source-receptor relationships for given regions and assess uncertainties in these estimates
- Source-Receptor Regions



Focus Species
- Ozone and precursors
- Aerosols and precursors
- Mercury and POPs
- Idealised Tracers

- Products
  - **2007 Interim Report** to inform the review of the 1999 CLRTAP Gothenburg Protocol to abate acidification, eutrophication, and tropospheric ozone
  - **2009 Assessment Report** to inform the CLRTAP on hemispheric air pollution and source-receptor relationships

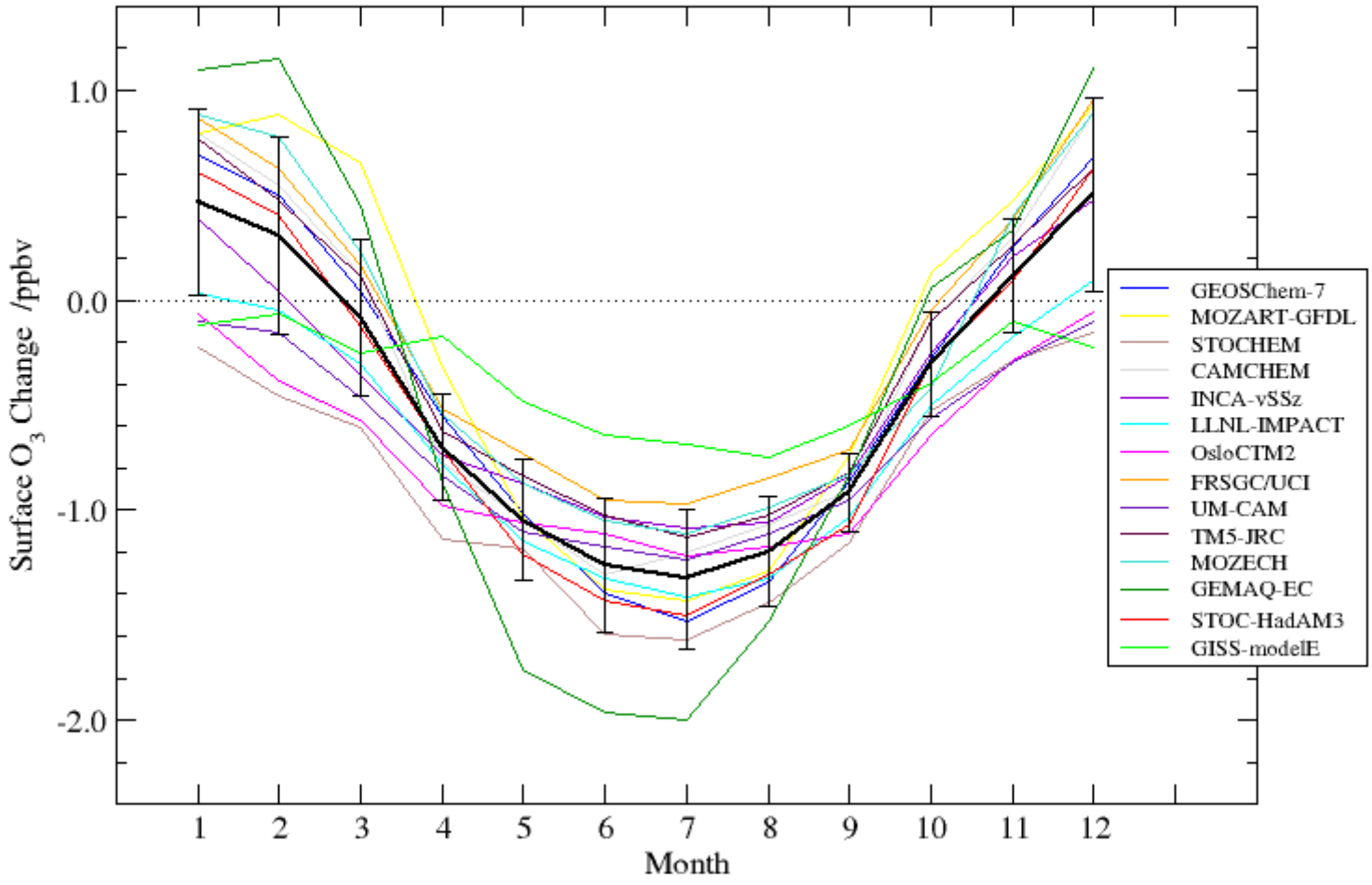
Monthly Mean Surface O_3 over Europe



- Each model using its own emissions and 2001 meteorology
- For surface O_3, model range often spans ~15-30 ppbv

HTAP

Monthly Mean Surface O_3 change in Europe from
20% Reduction in Regional NO_x Emissions



- Strong seasonality, but models reasonably consistent

HTAP

Source-Receptor Matrix for Surface O_3

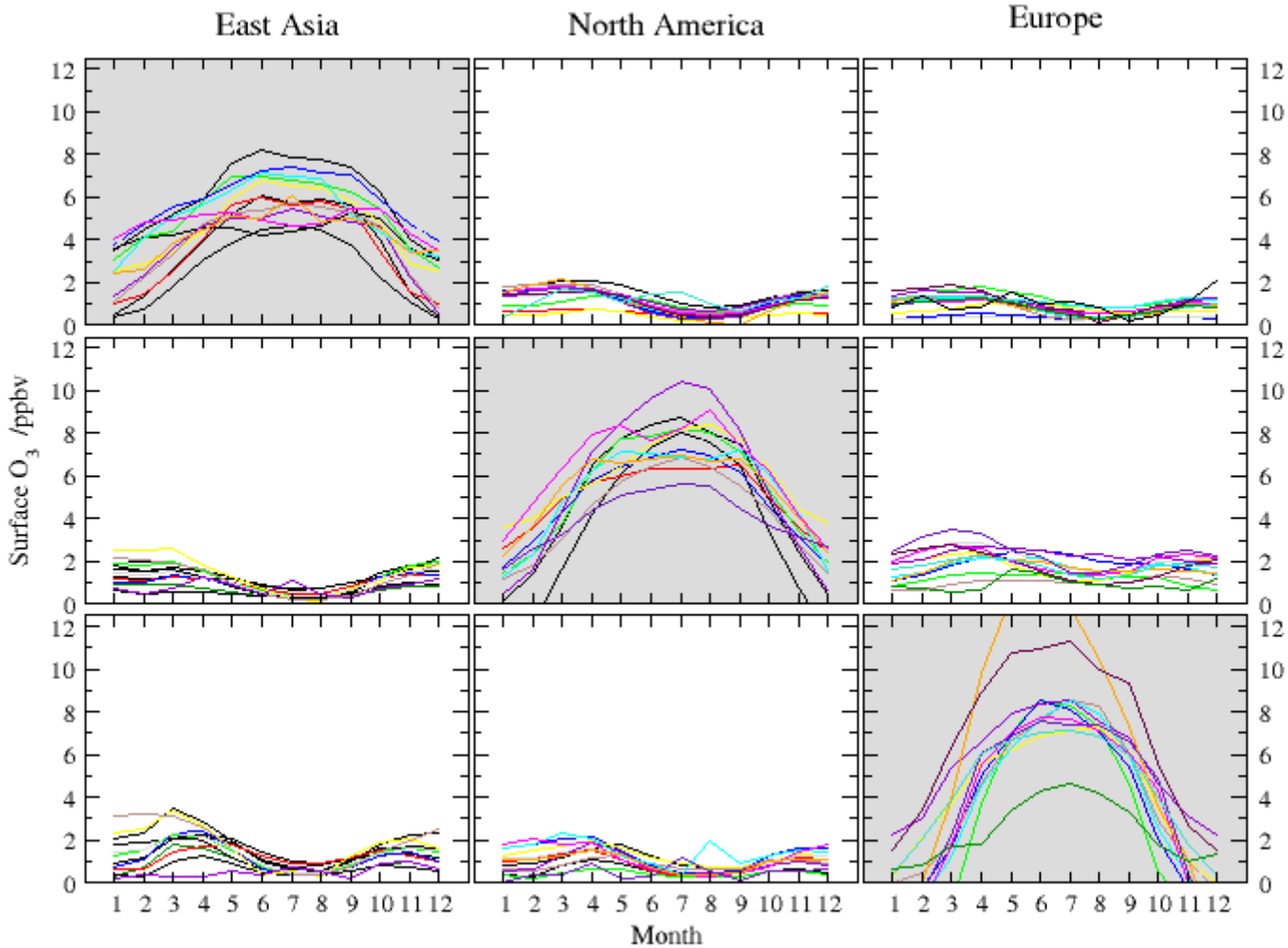
Receptors

Sources

East Asian Emissions

N. American Emissions

European Emissions



- Regional impacts scaled from 20% reduction in NO_x/CO/VOC emissions
  - Caveat: non-linearities in O_3 chemistry will increase these responses!

HTAP
14 models

Source-Receptor Matrix for Surface O_3

Receptors

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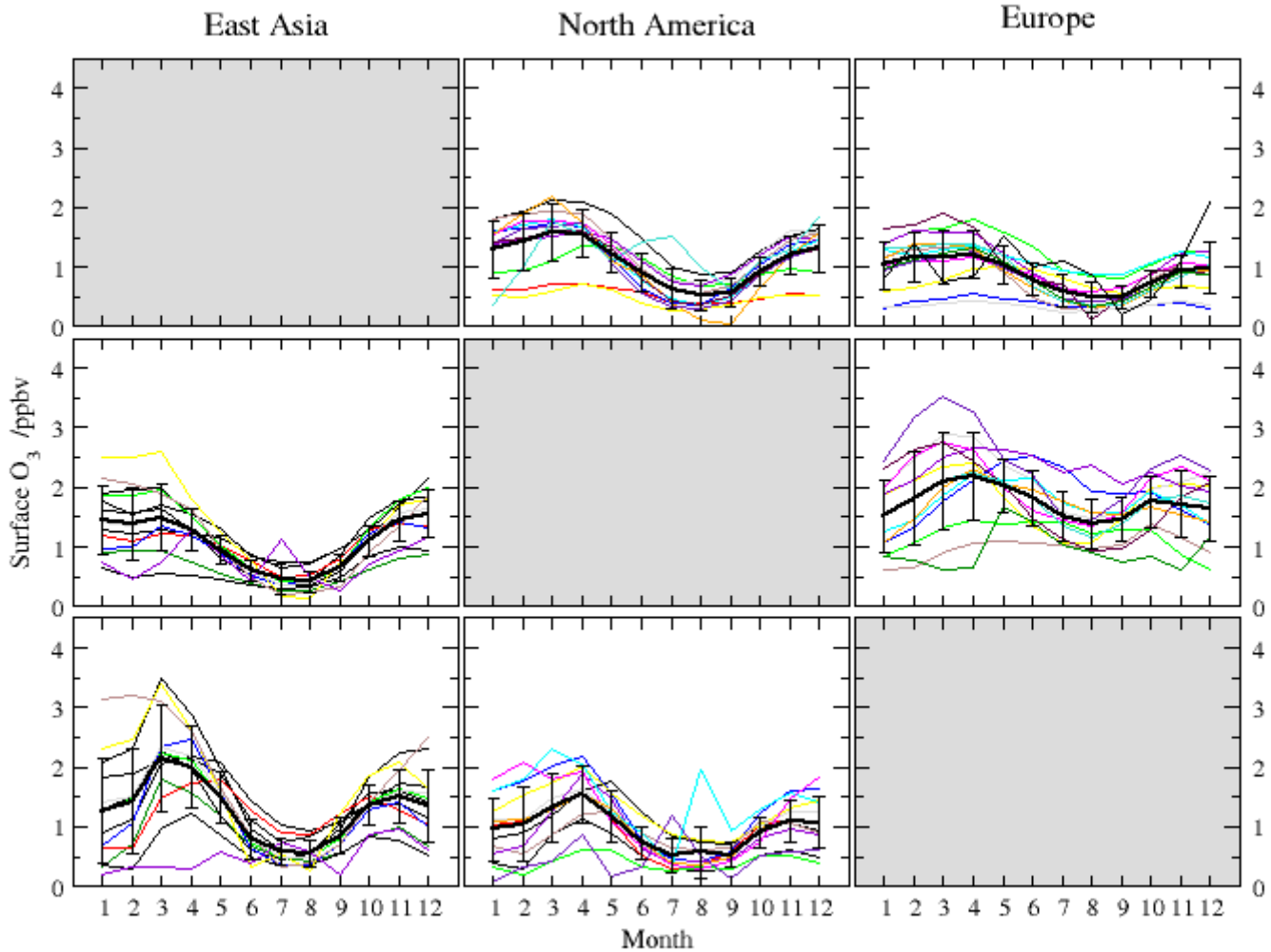

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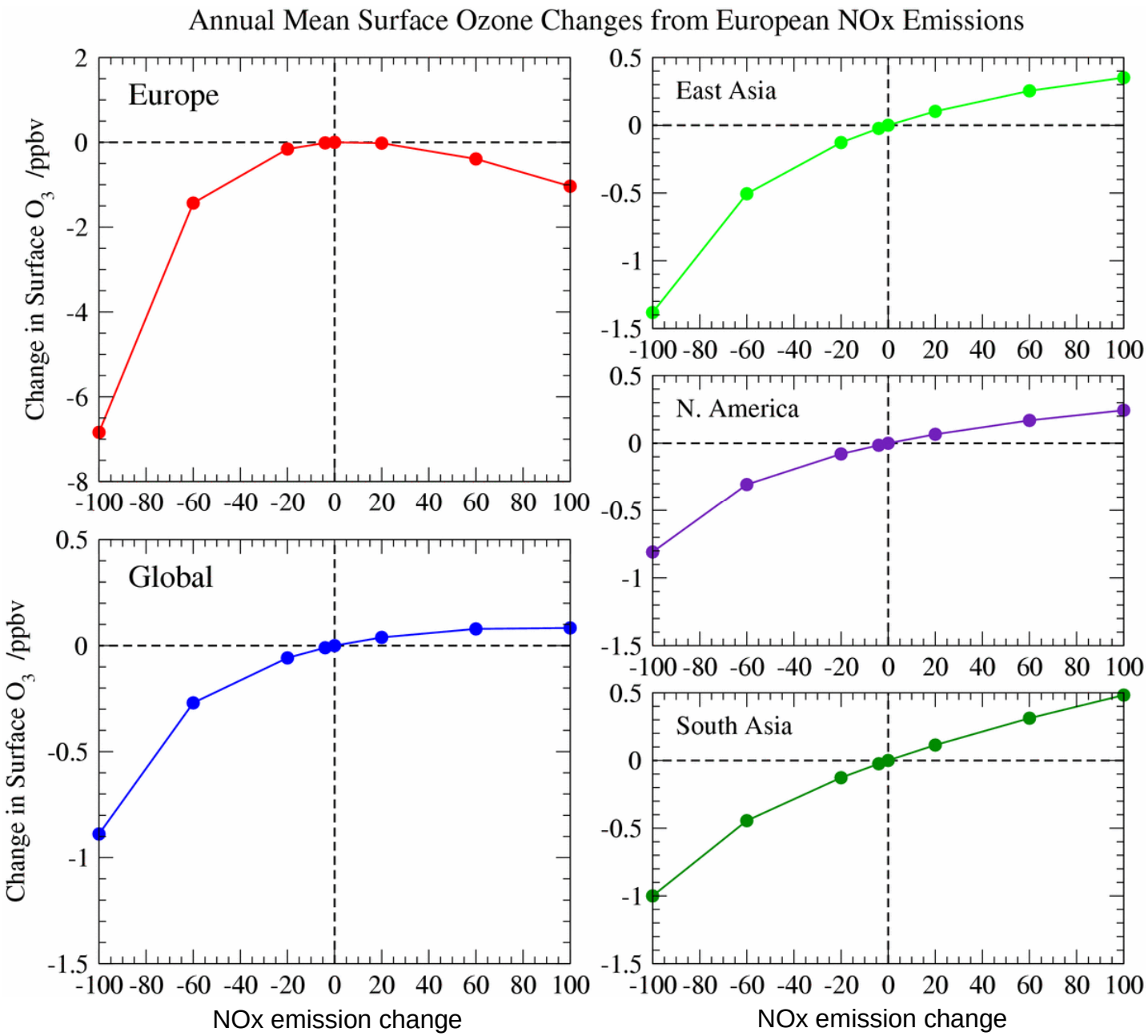
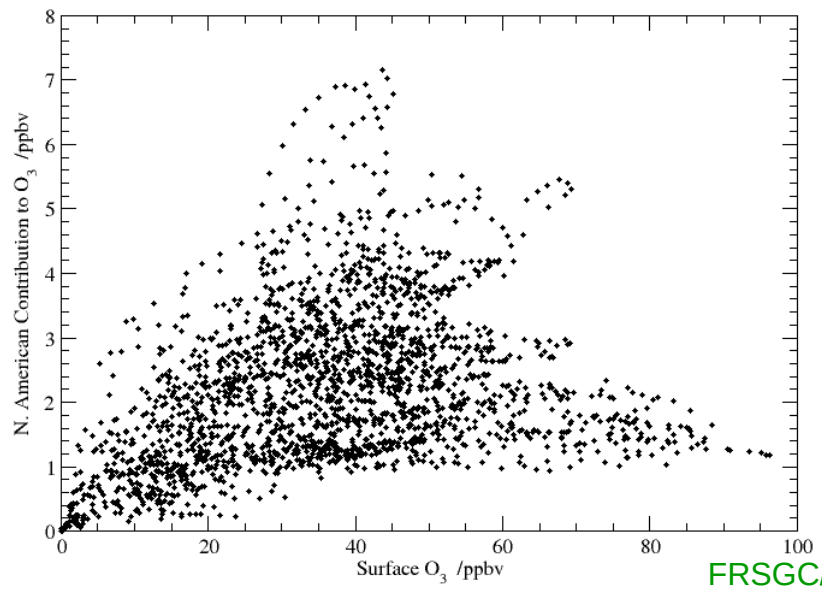
HTAP
14 models


Annual Mean Surface Ozone Changes from European NOx Emissions
Europe
East Asia
N. America
Global
South Asia
Change in Surface O_3 /ppbv
NOx emission change
NOx emission change

Transport Episodes

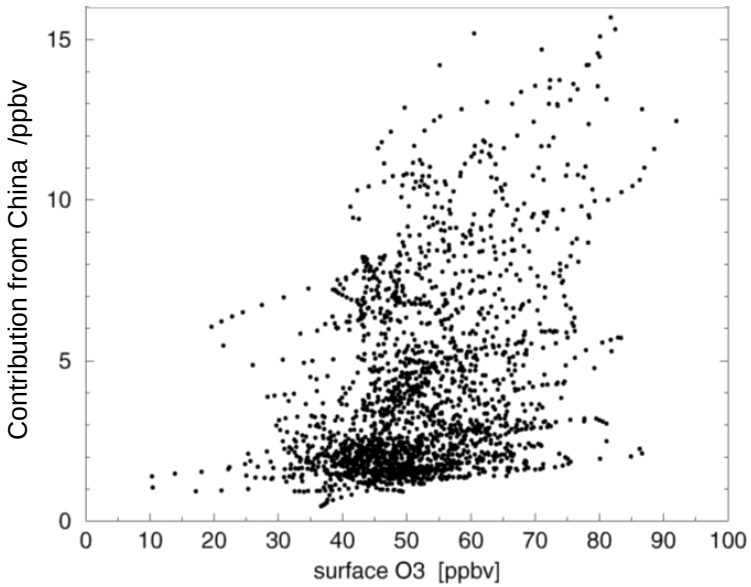
- Do transport episodes make a significant contribution to urban O_3?
  - Maximum enhancements occur at intermediate O_3 levels (40-60 ppbv)
  - Important for regional transport (location-dependent)

N. American O_3 over London (summer 2001)



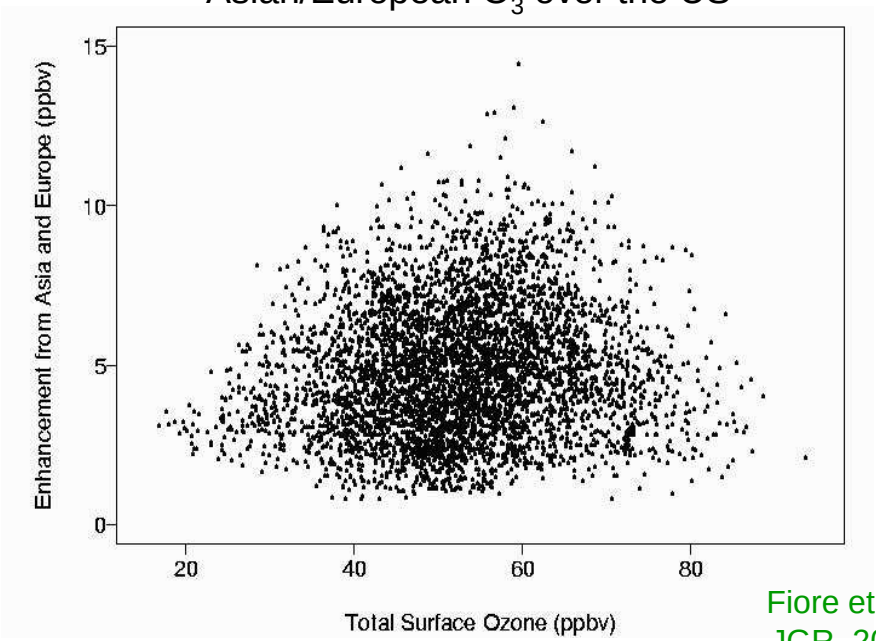
FRSGC/UCI
HTAP

Chinese O_3 over Tokyo (spring 2001)



FRSGC/UCI, Yoshitomi et al., in prep

Asian/European O_3 over the US



Fiore et al.,
JGR, 2002

Regional Meteorology and its Impacts on O_3

Key Questions and Challenges

- How are regional and global impacts influenced by meteorology?
  - What is the variability in O_3 production from a given source?
- How does meteorology govern climate impacts of an urban source?
  - How will future changes in meteorology affect climate impacts from cities?
- How well can models simulate the time scales for O_3 formation?

Model Approach

- Perturb fossil fuel NO_x/CO/NMHC emissions over one city for one day
  - Follow atmospheric perturbation for 1 month
- Repeat for each day of March 2001 (TRACE-P measurement campaign)
- Look at variability in magnitude and location of O_3 production

Ozone Responses

Look at regional and global O_3 from a single day's emissions over Shanghai

March 12

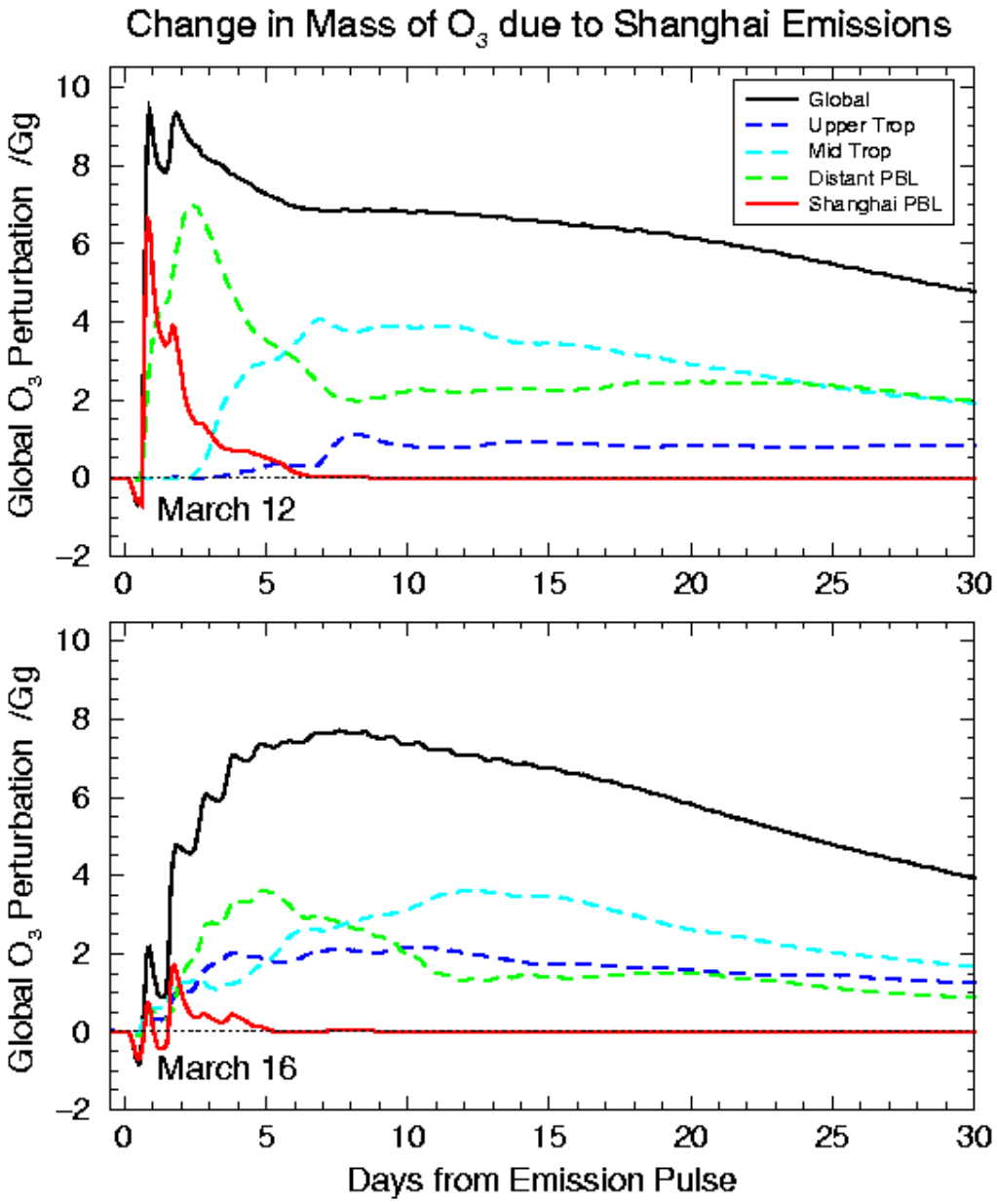
- Sunny, high pressure
- Strong regional $P(O_3)$

March 16

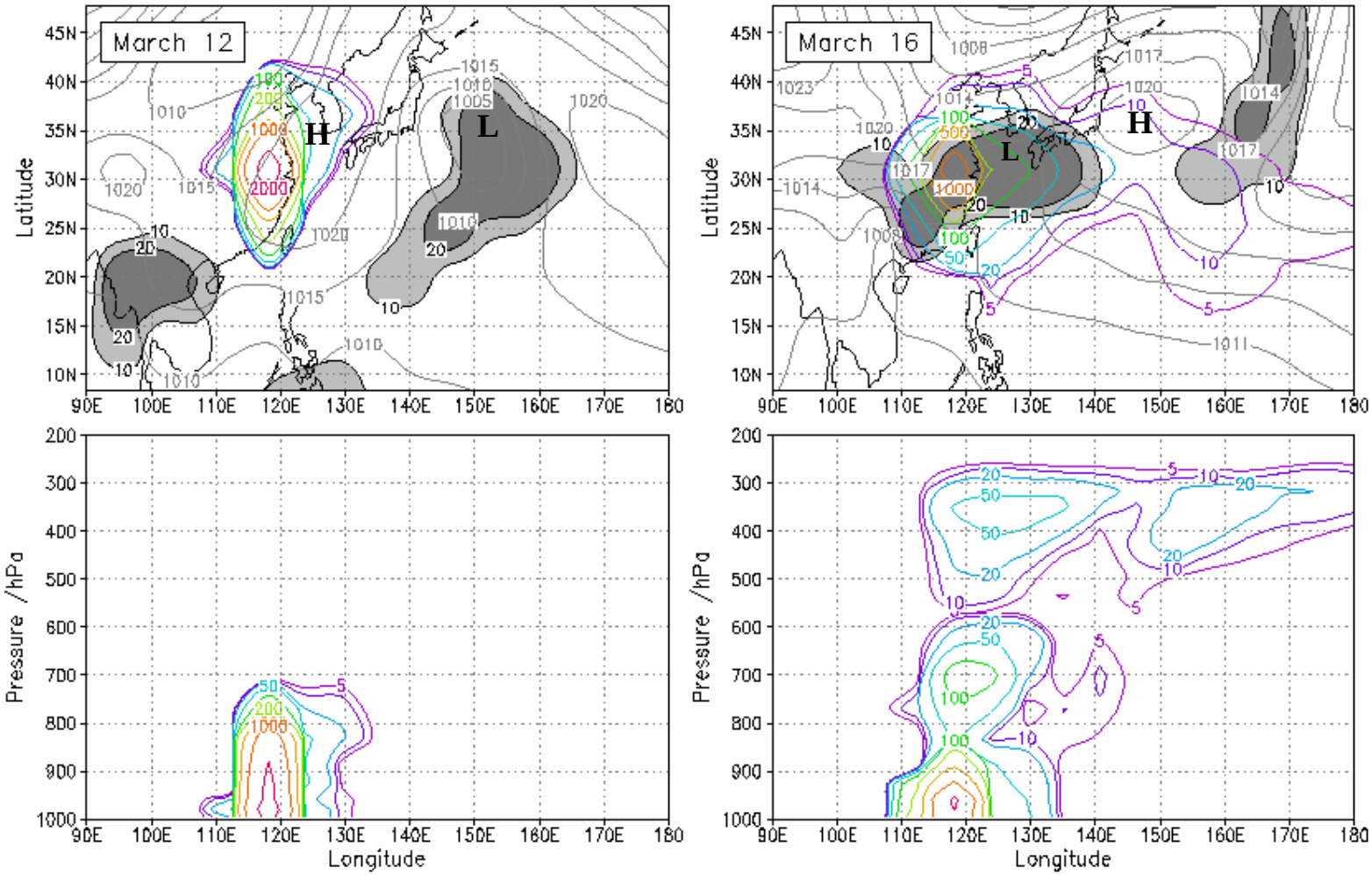
- Heavily overcast
- Little regional $P(O_3)$

Regional production different, Global production similar

- Evolution quite different
- Location of $P(O_3)$ different

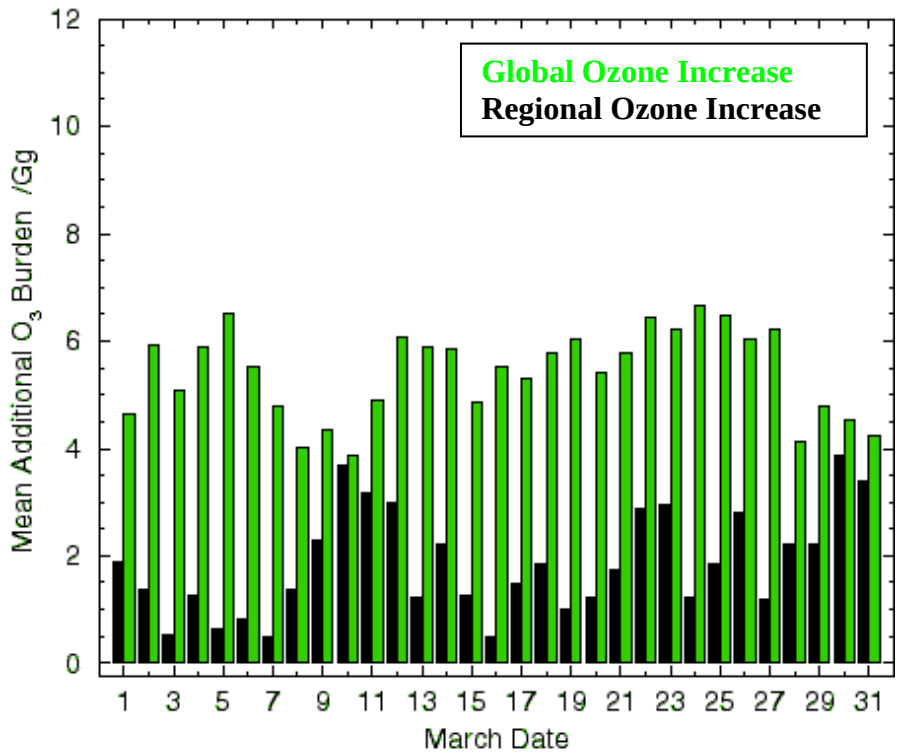


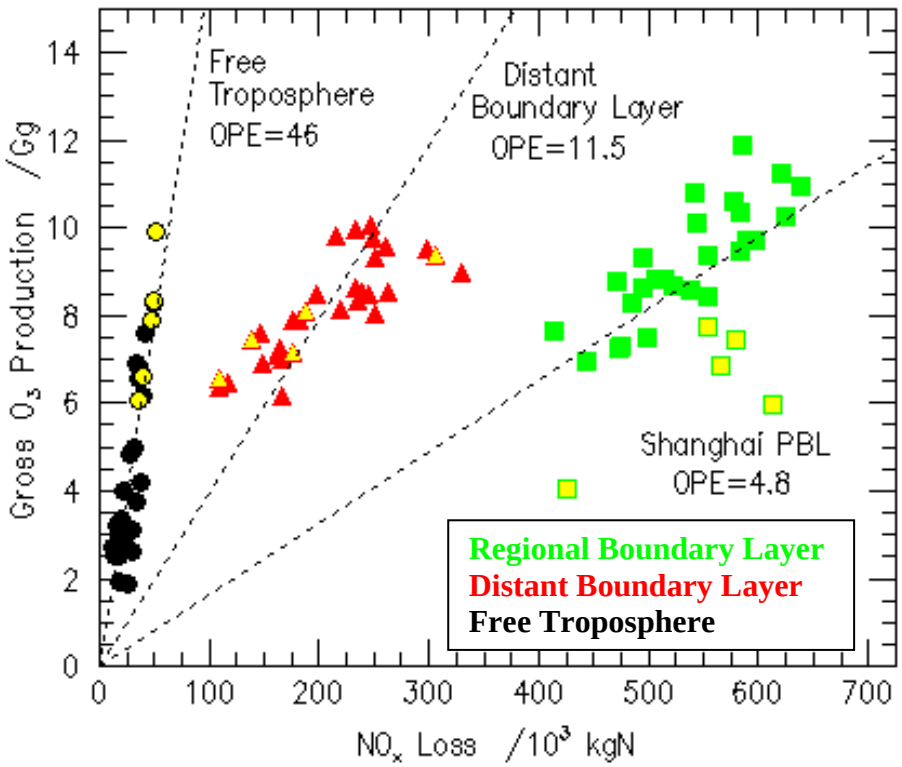
Meteorological Setting on March 12 and 16, 2001



Column- and latitude-integrated gross O_3 production over the first 3 days following 1 day of emissions over Shanghai

Ozone Response to Shanghai Emissions





- Effects on O_3 burden
  - Days with high regional O_3 (smog) have a reduced effect on global O_3
  - Regional meteorology strongly influences climate impacts

- $P(O_3)$ vs. NO_x loss for each day
  - O_3 production efficiency (OPE) strongly dependent on location
  - Good representation of lifting processes is required!

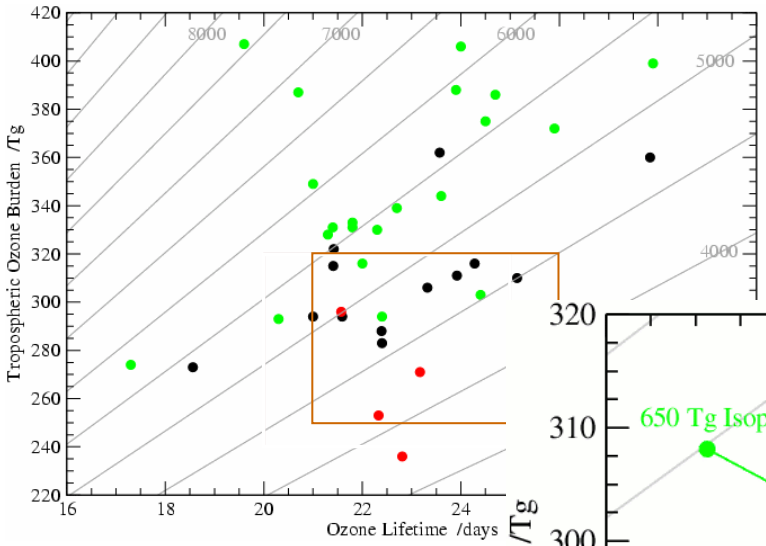
Summary of Long-Range Transport Issues

- What is the impact of remote sources on regional O_3?
  - Long-range continental contributions typically average 2-4 ppbv
  - May make a major contribution during transport episodes (10-15 ppbv)
  - Not usually a major contributor to urban O_3 exceedances
    - Except for regional transport (e.g., Chinese O_3 over Japan in Spring)
  - Importance of episodic vs. background transport not well understood
  - Source attribution for O_3 remains a challenge due to non-linear chemistry

- Emission region meteorology has a large effect on O_3
  - Regional O_3 build-up in stagnant anticyclonic conditions (air quality)
  - Regional export/lifting processes important (free troposphere, climate)
  - Climate change may alter the balance of these effects

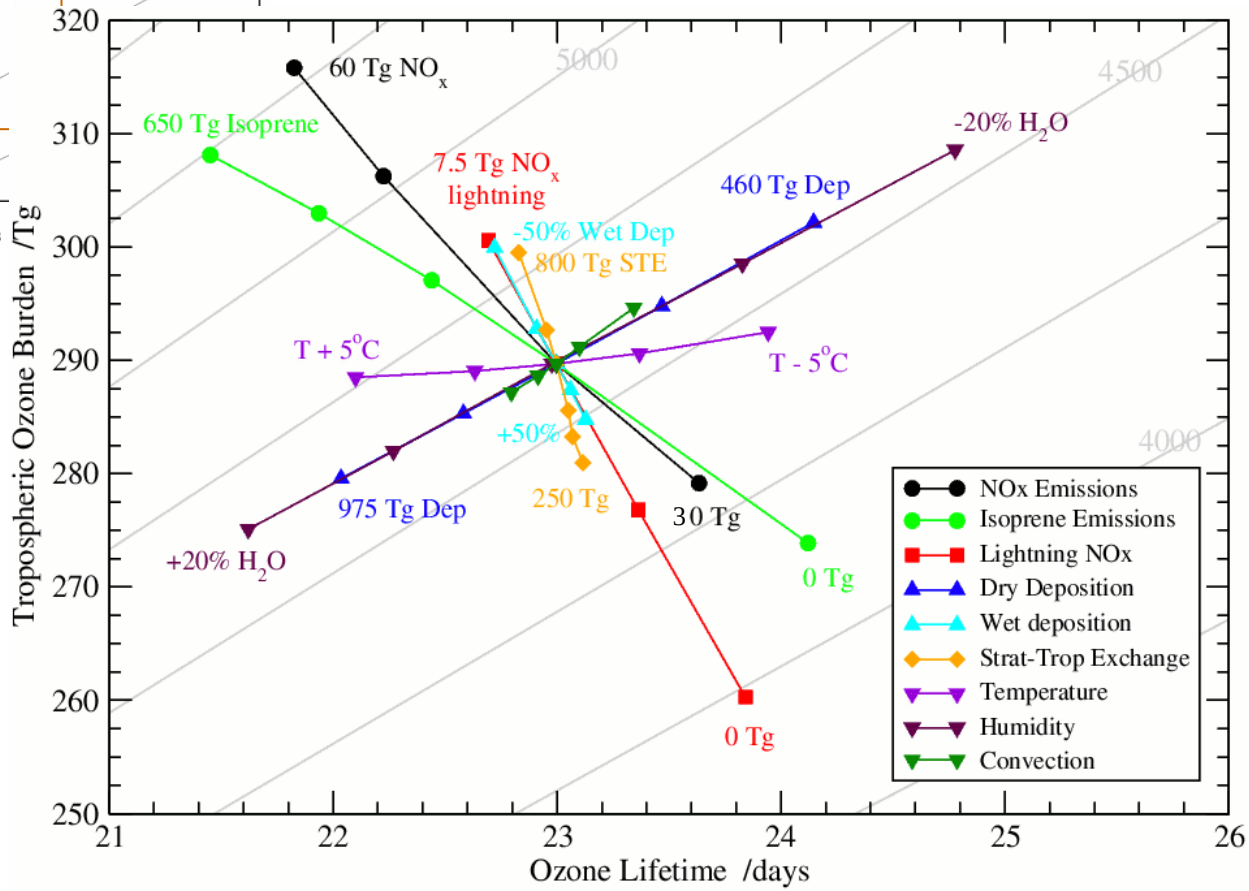
- Global model estimates remain rather uncertain
  - Need more thorough evaluation of model processes → model refinement

Accounting for Model Differences



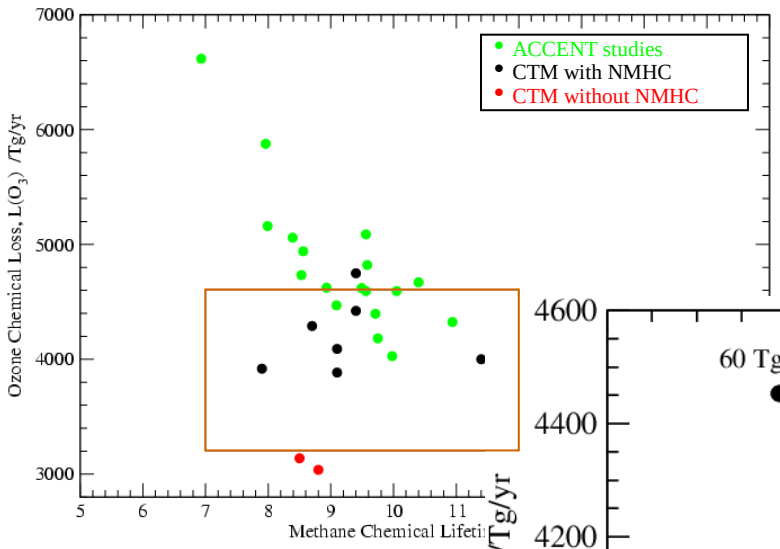
O_3 Burden vs. O_3 Lifetime

Diagonals in grey show O_3 loss rate (Tg/year)

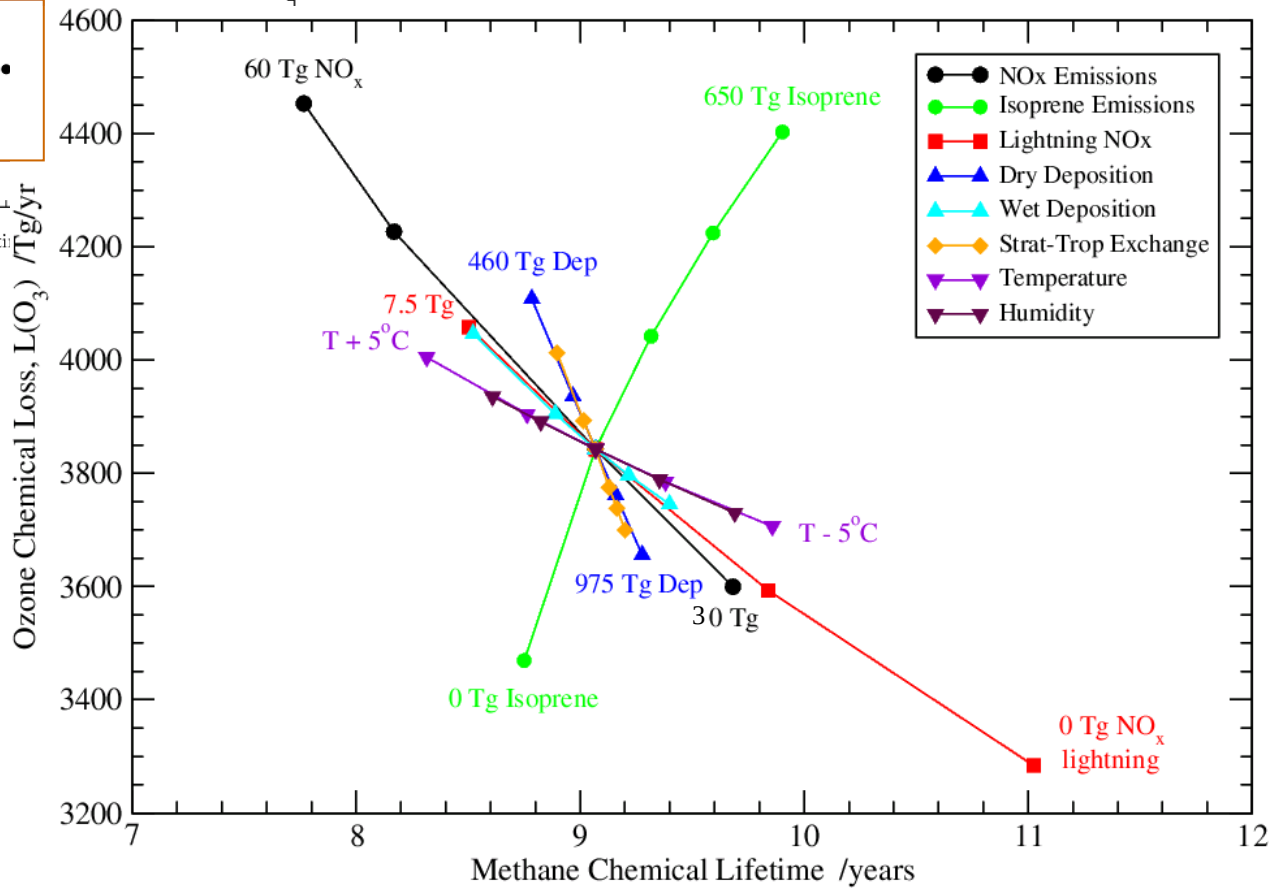


- Sensitivity to key variables
- Explains part of scatter

Accounting for Model Differences

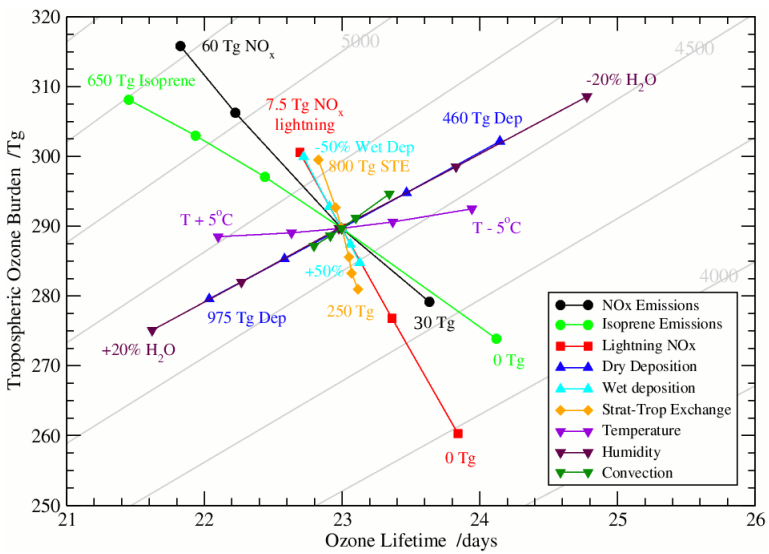


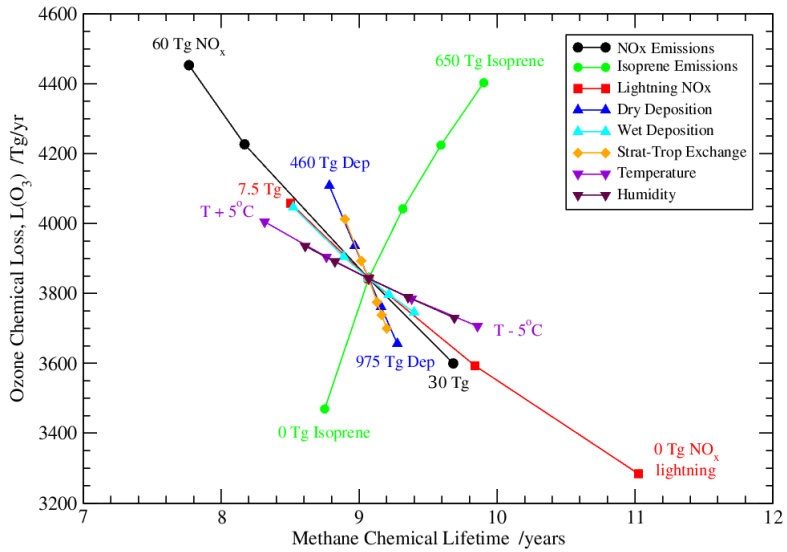
O_3 Loss Rate vs. CH_4 Lifetime



- Strong sensitivity to NOx and Isoprene emissions
- Strong sensitivity to lightning NOx (currently not well constrained)
- Influence of other factors rather smaller

Accounting for Model Differences





- Summary of key sensitivities
  - NOx emissions: more O_3, $P(O_3)$, more OH
  - Isoprene emissions: more O_3, $P(O_3)$, less OH
  - Lightning NOx: poorly constrained, large impact on O_3 and OH
  - STE: impact on O_3 burden and lifetime
  - Meteorology: particularly humidity
- Sensitivity to chemical schemes not explored here…