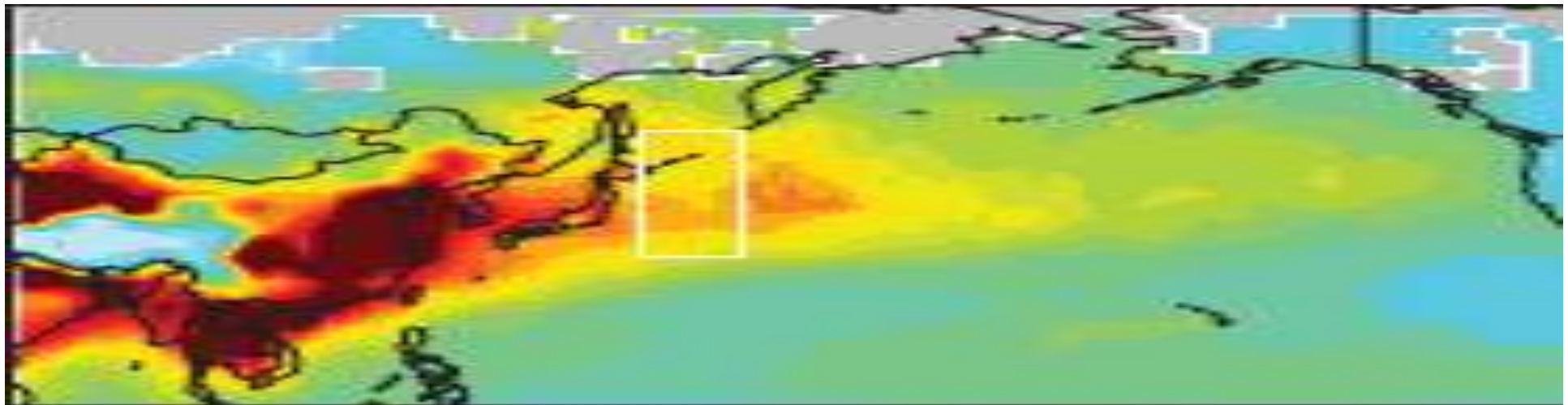
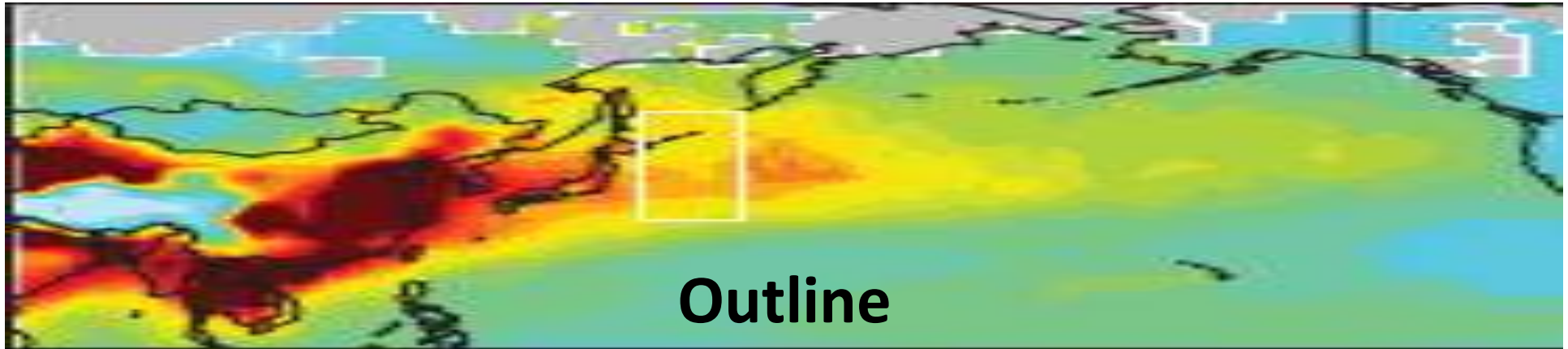


Hemispheric Ozone: Current Understanding & Future Directions

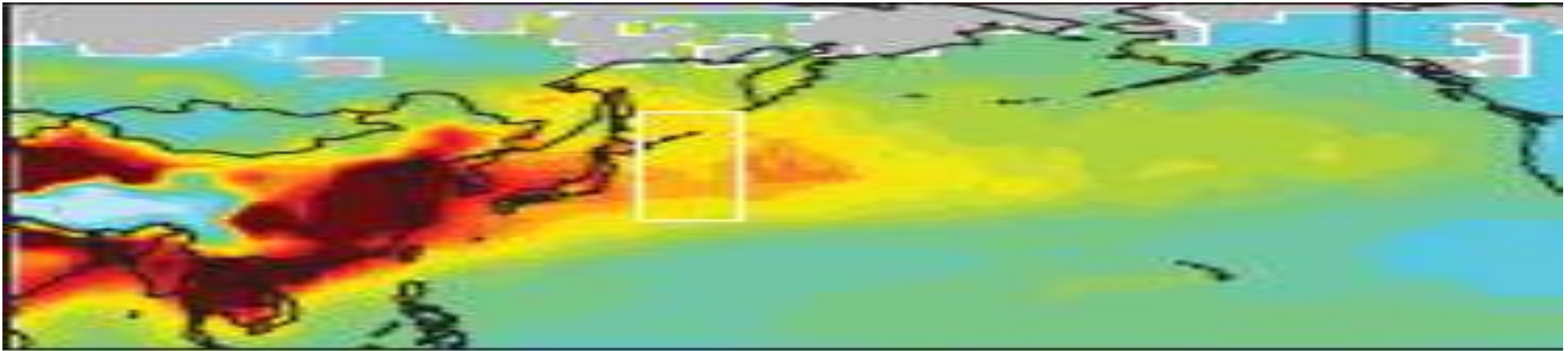
Kathy Law

LATMOS-CNRS, UPMC, Paris, France

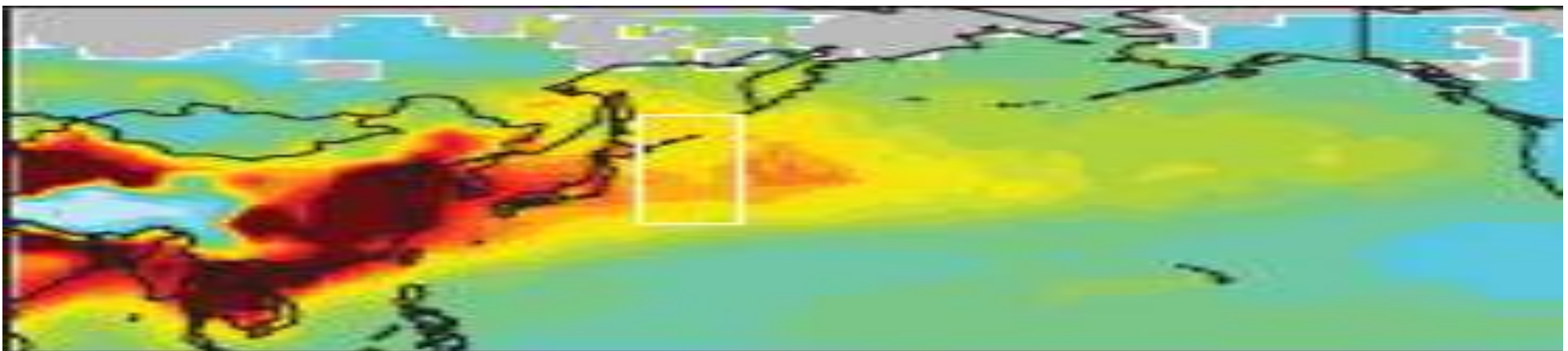




- Motivation
- Observational evidence: « background » vs episodes
- Hemispheric transport and trends
- Pollutant export, processing and import (deposition)
- Concluding remarks – future directions

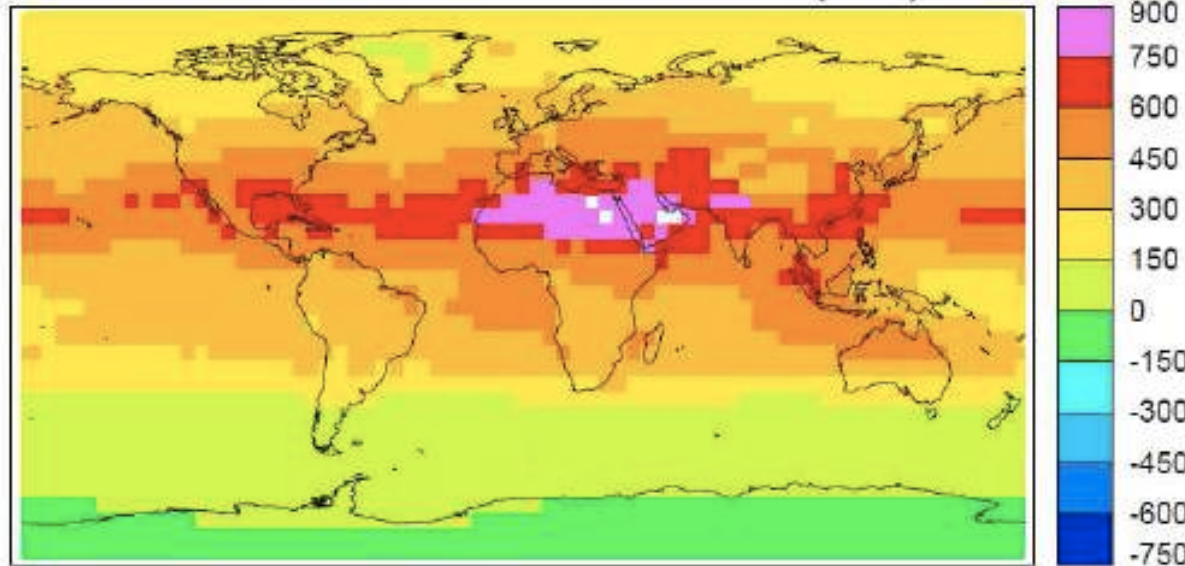


Motivation



Climate: Ozone Radiative Forcing

Multi-model mean 2000s-1850s O3t RF (355) mW/m2

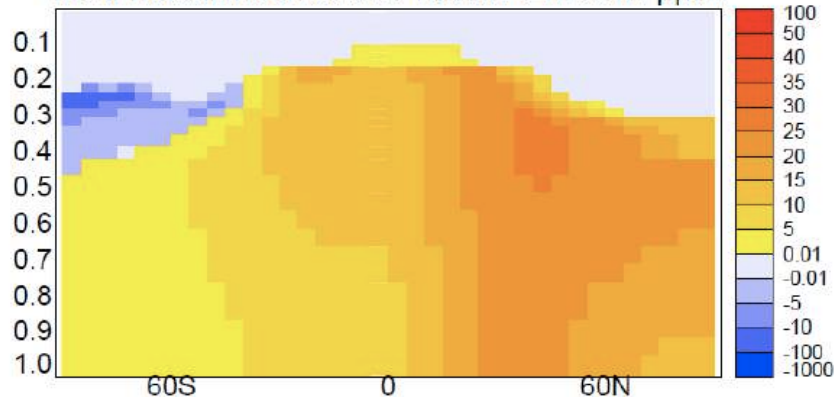


Total tropospheric RF

Maximum forcing is away from emission regions

Mainly LW (5-14km) + smaller SW especially over reflective surfaces

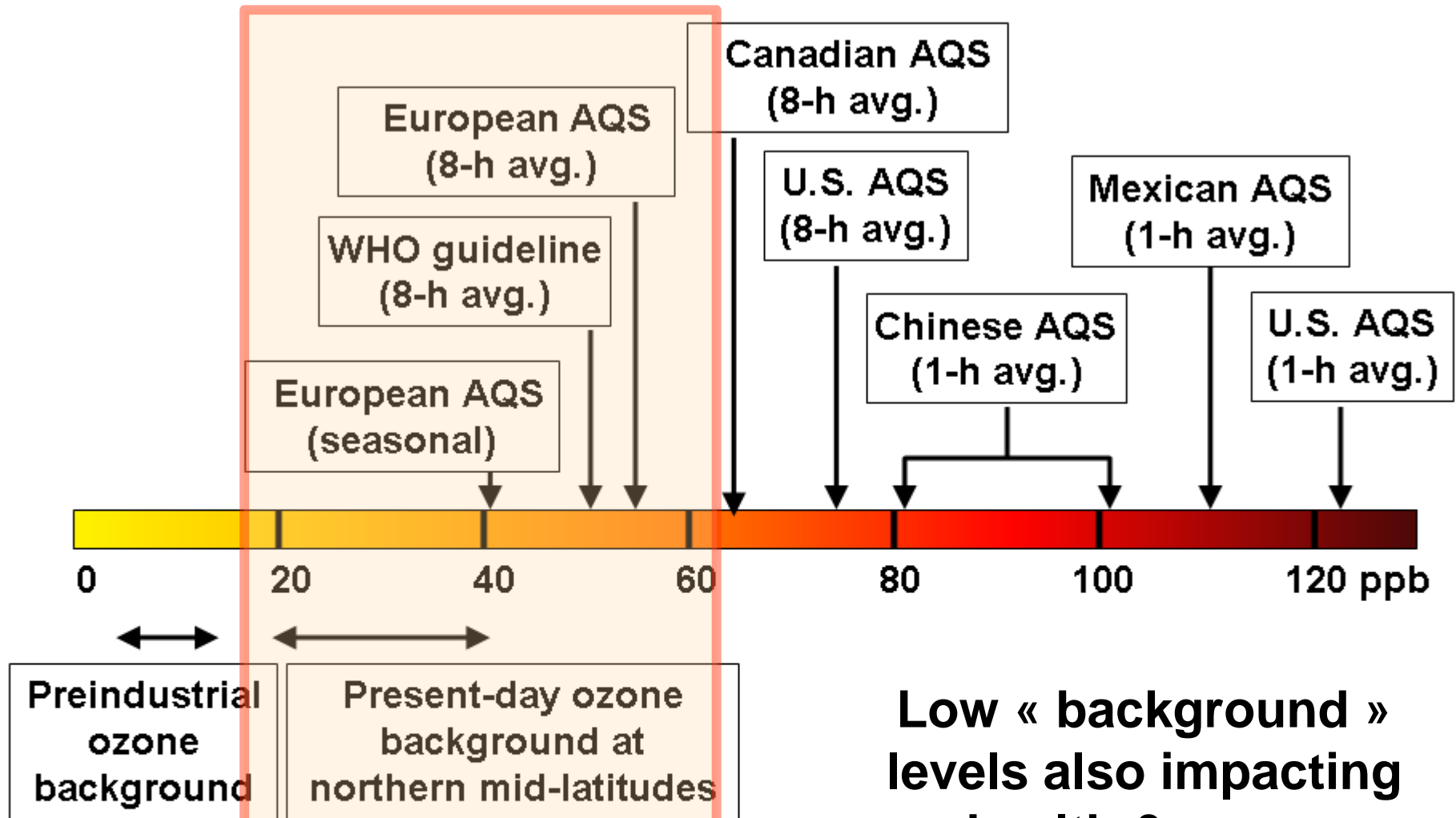
Multi-model mean 2000s-1850s AZM O3t / ppbv



Global mean RF = $0.4 \text{ Wm}^{-2} \pm 30\%$

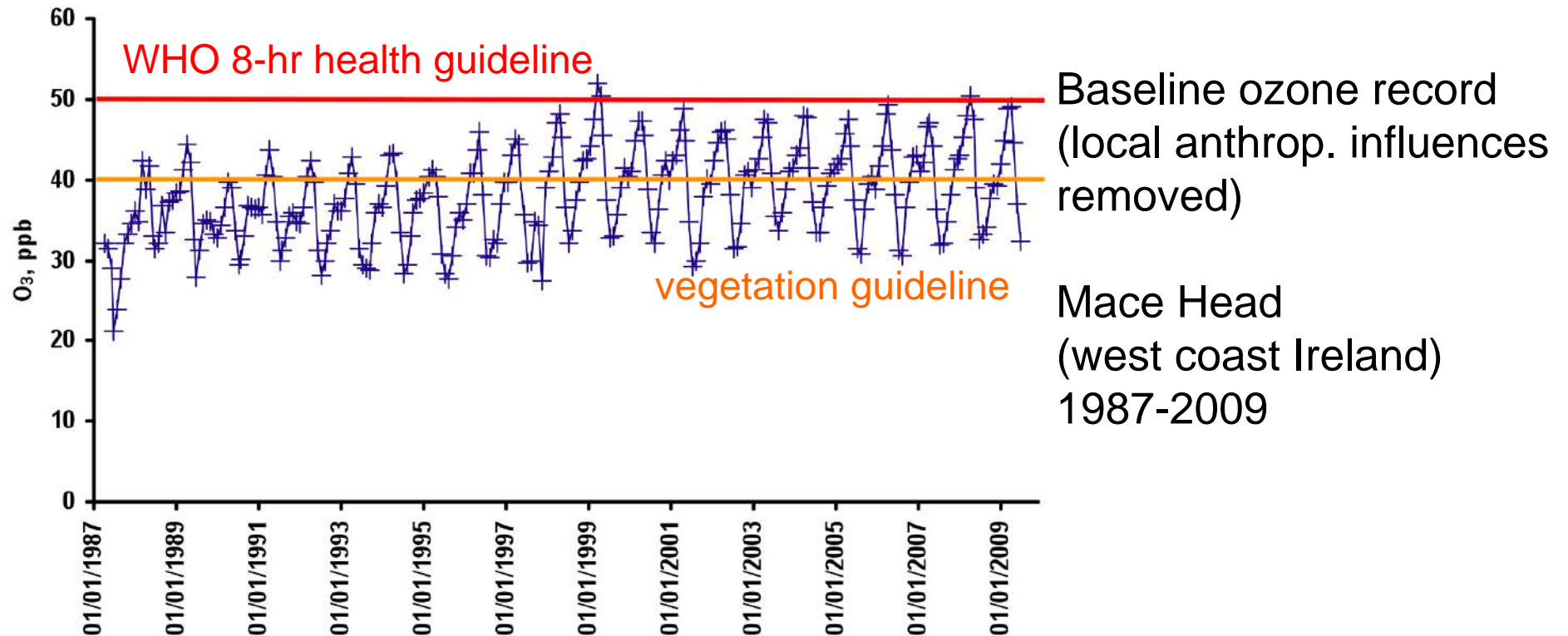
Stevenson et al. (2012)

HEMISPHERIC POLLUTION: OZONE REGIONAL AIR QUALITY STANDARDS



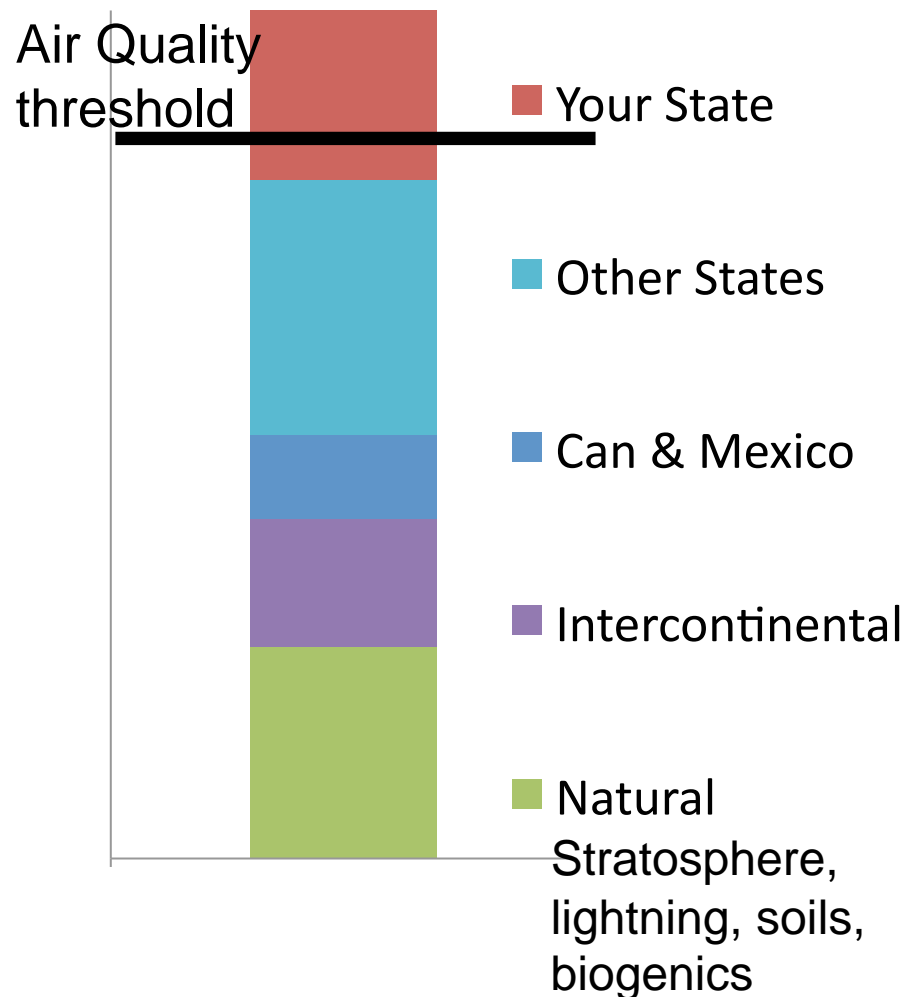
**Low « background »
levels also impacting
health & crops
(30 ppbv)**

O₃ import into receptor regions

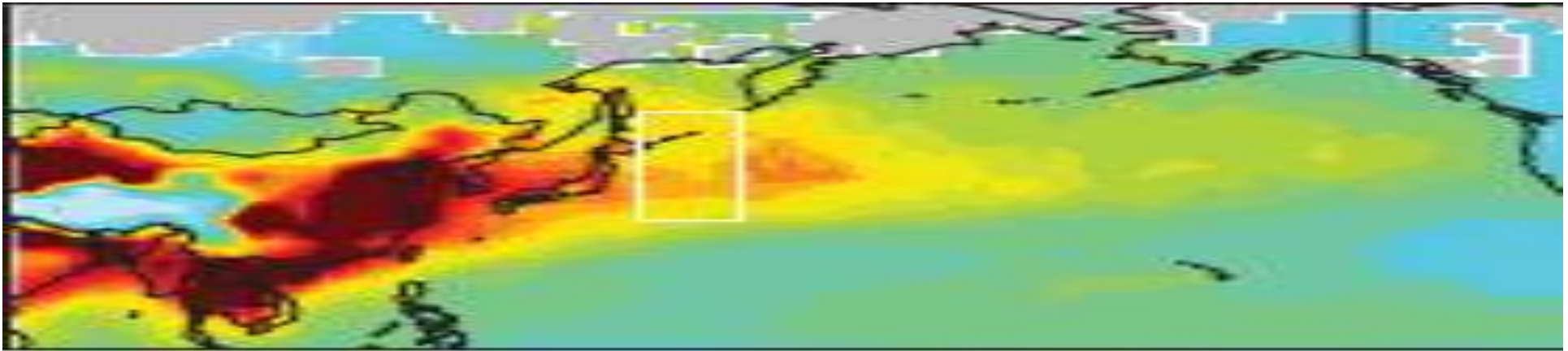


Ozone measurements on west coast of continents show levels approaching or exceeding levels of ambient standards

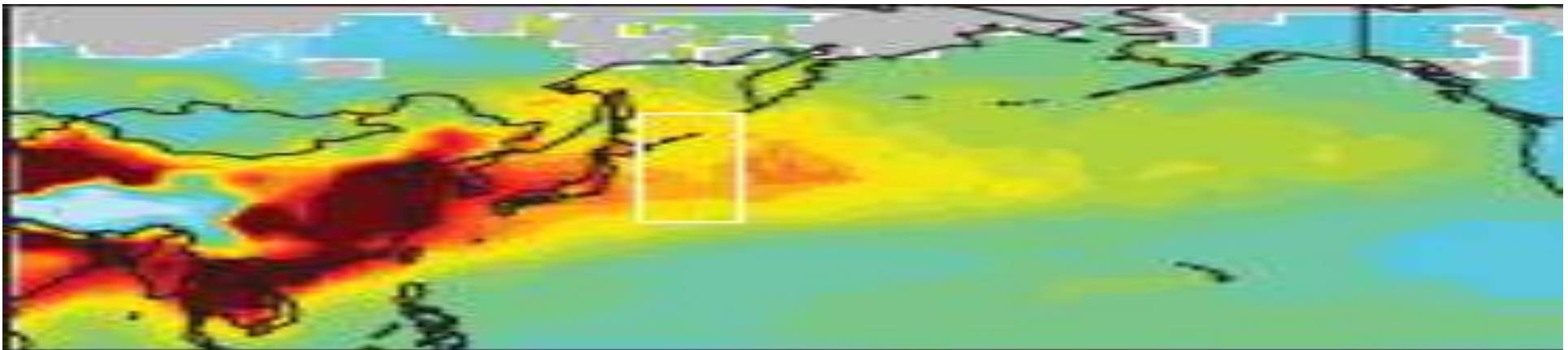
Long-Range Transport: Implications for AQ management



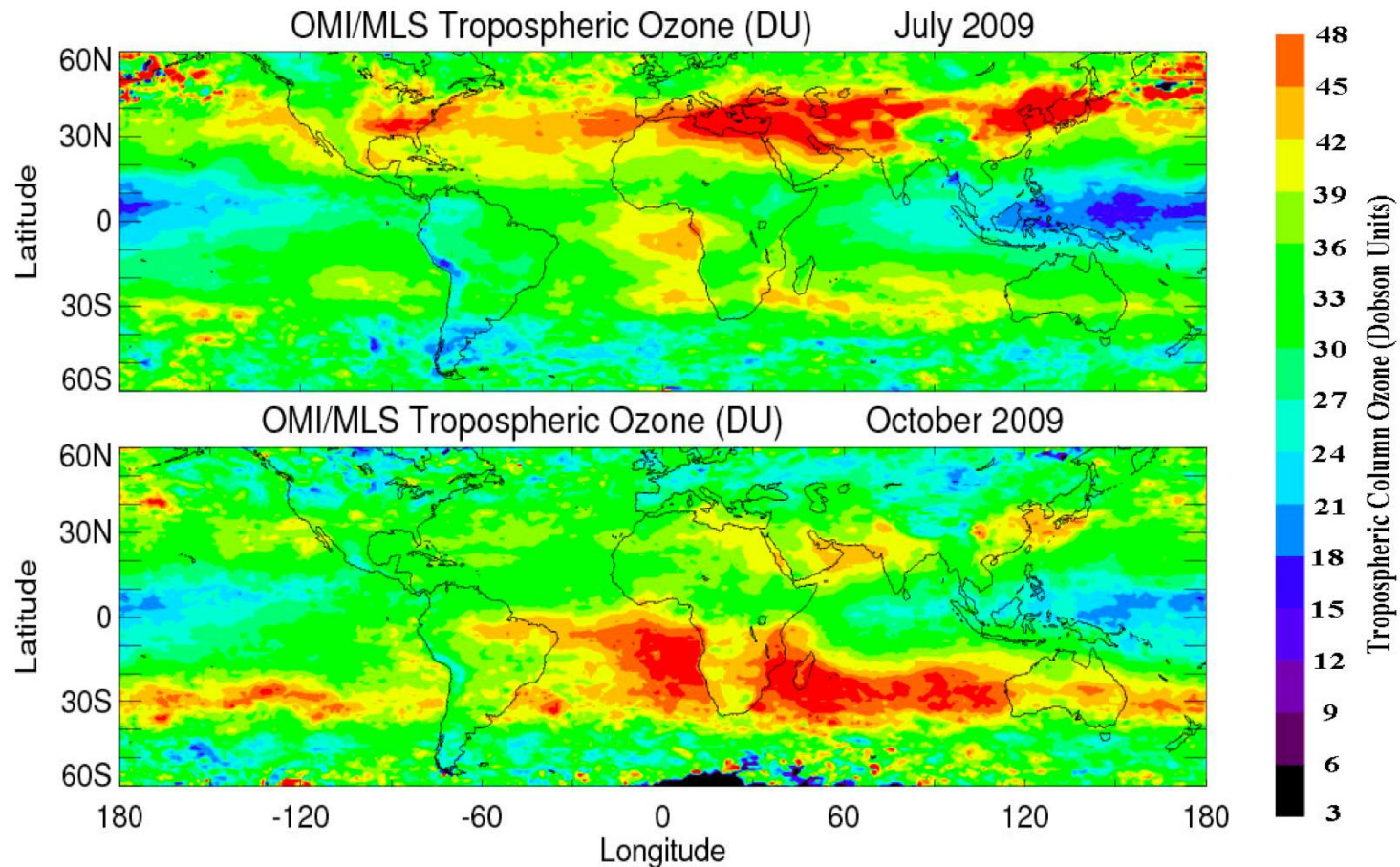
- What are the magnitudes of different fractions contributing to pollutant levels?
- How will fractions change in the future?
- How efficiently can each fraction be mitigated?
- What policies and programs are needed?



Observational Evidence

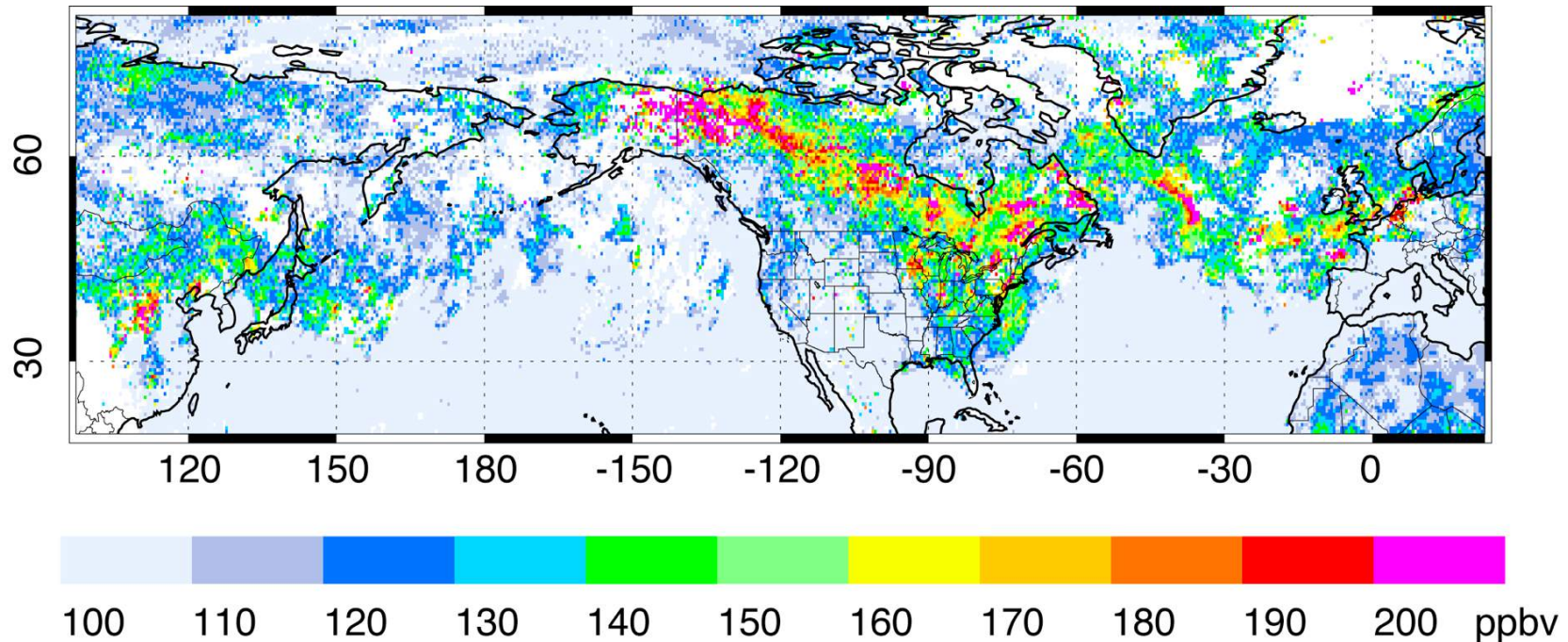


Hemispheric Ozone from Space



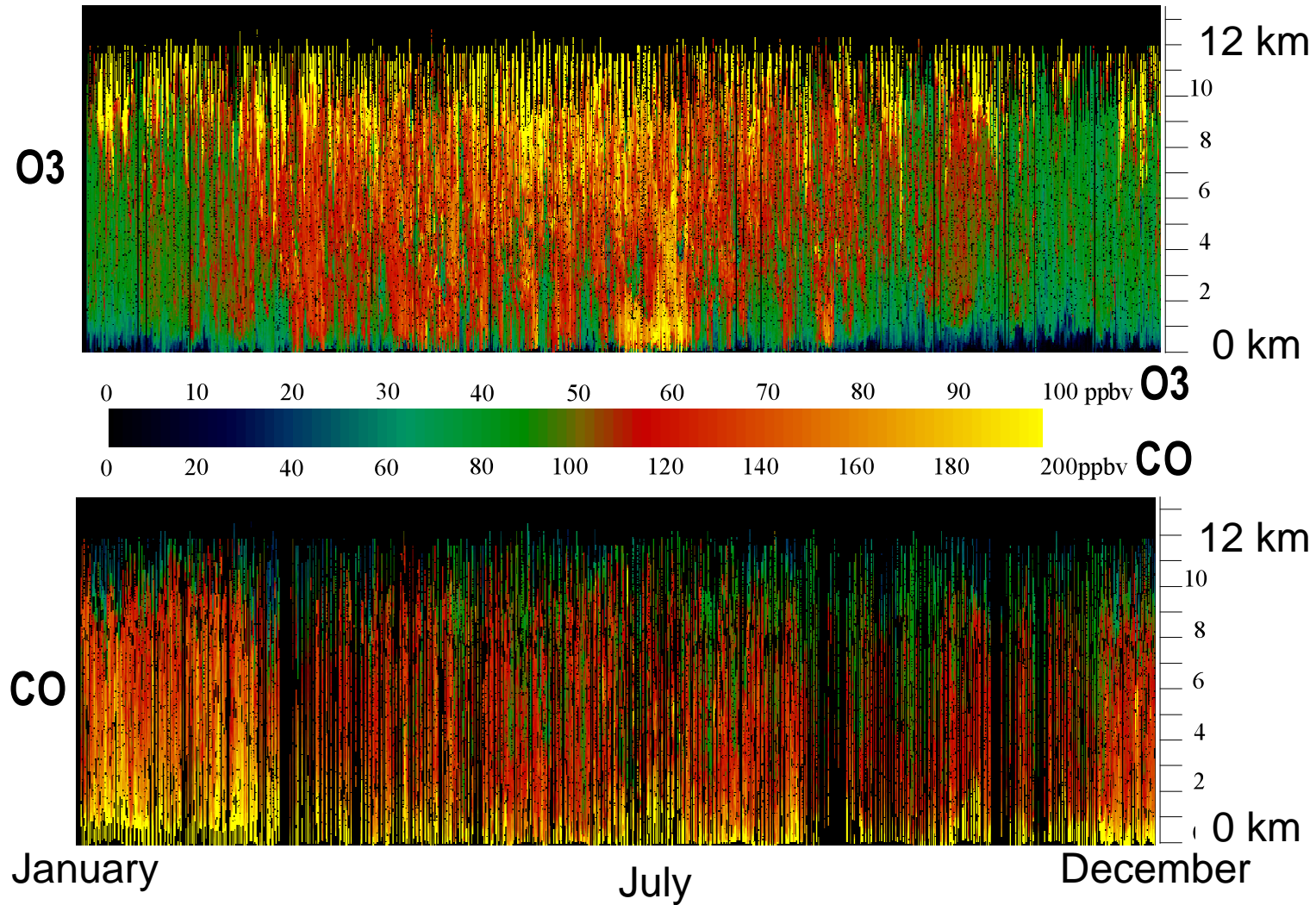
Tropospheric ozone column satellite data - mix of long-range transport, chemical production/loss and natural sources (stratosphere) [Ziemke et al., 2006]

O₃ precursors: plume events



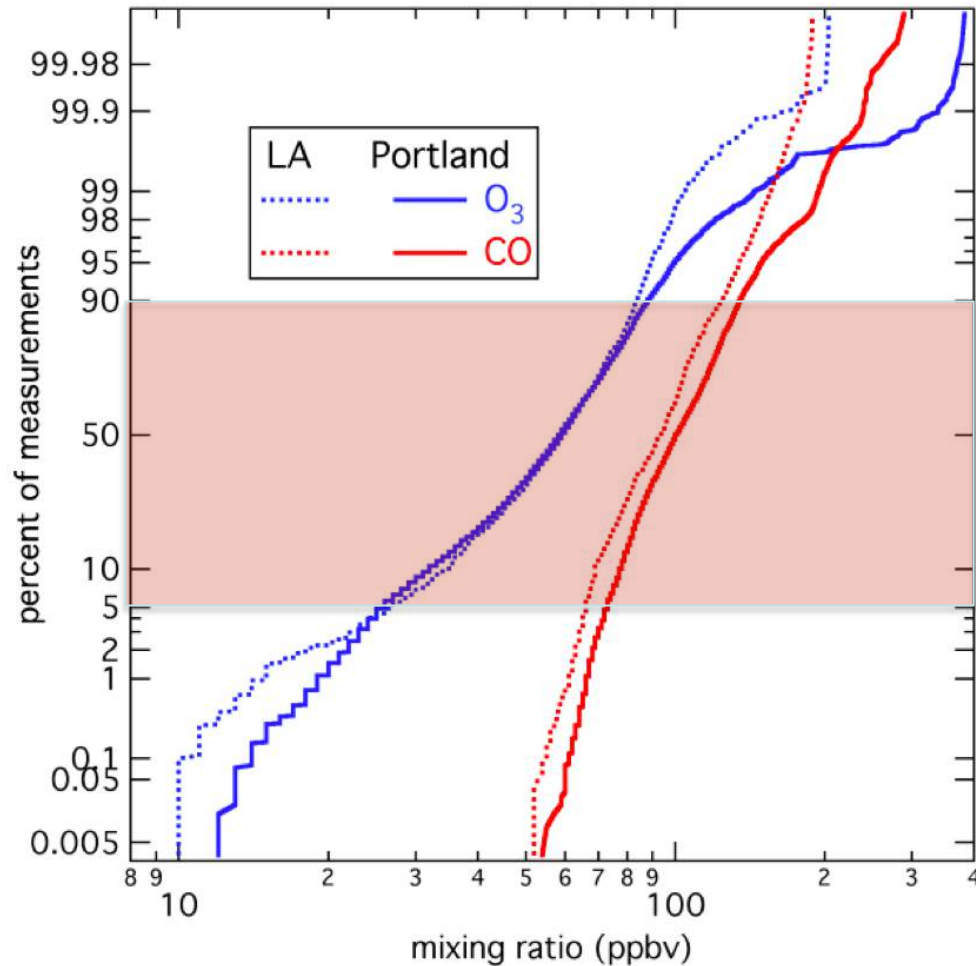
Forest fire and anthropogenic CO plumes seen by MOPITT satellite in July 2004 (*NRC, 2010*)

Troposphere: plumes of different ages



MOZAIC data collected on commercial aircraft at Frankfurt during 2003 from 0-12km (1080 profiles) [courtesy Zebinden, Thouret]

Receptor regions see broad spectrum of concentrations



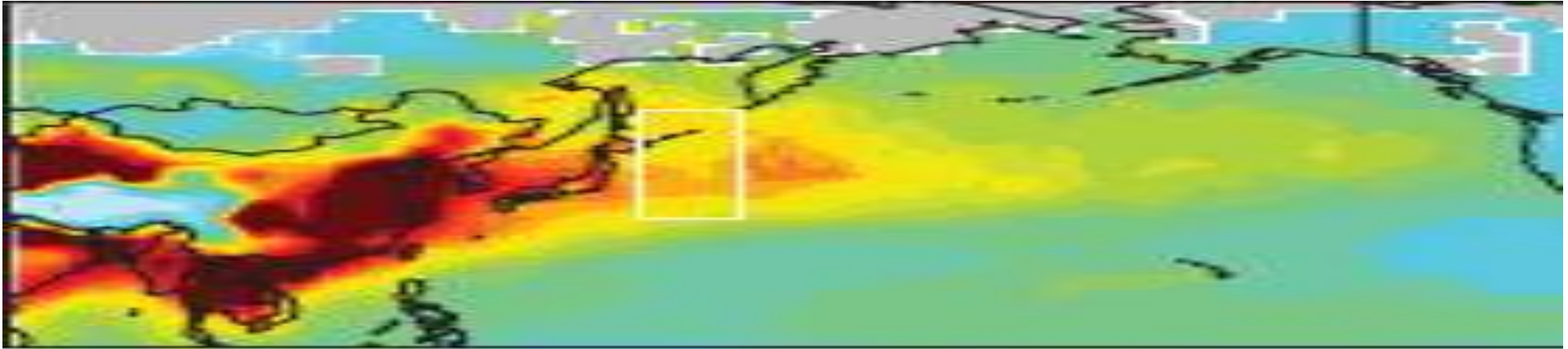
« **Plumes** » only account for 1-2% of air masses

All NH influenced by anthrop. pollution
(even at remote sites)

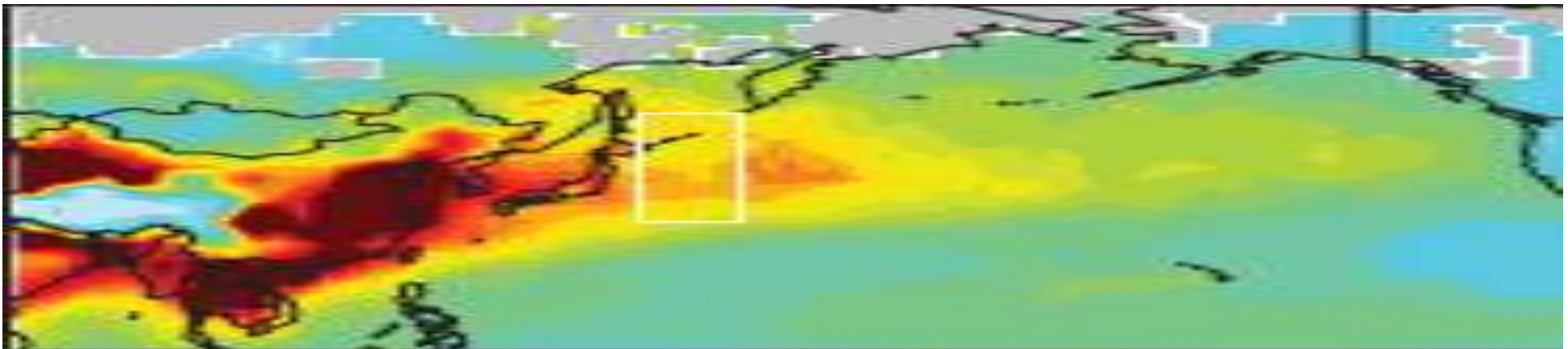
« **Background** » *not a very useful concept*

Pollution is global

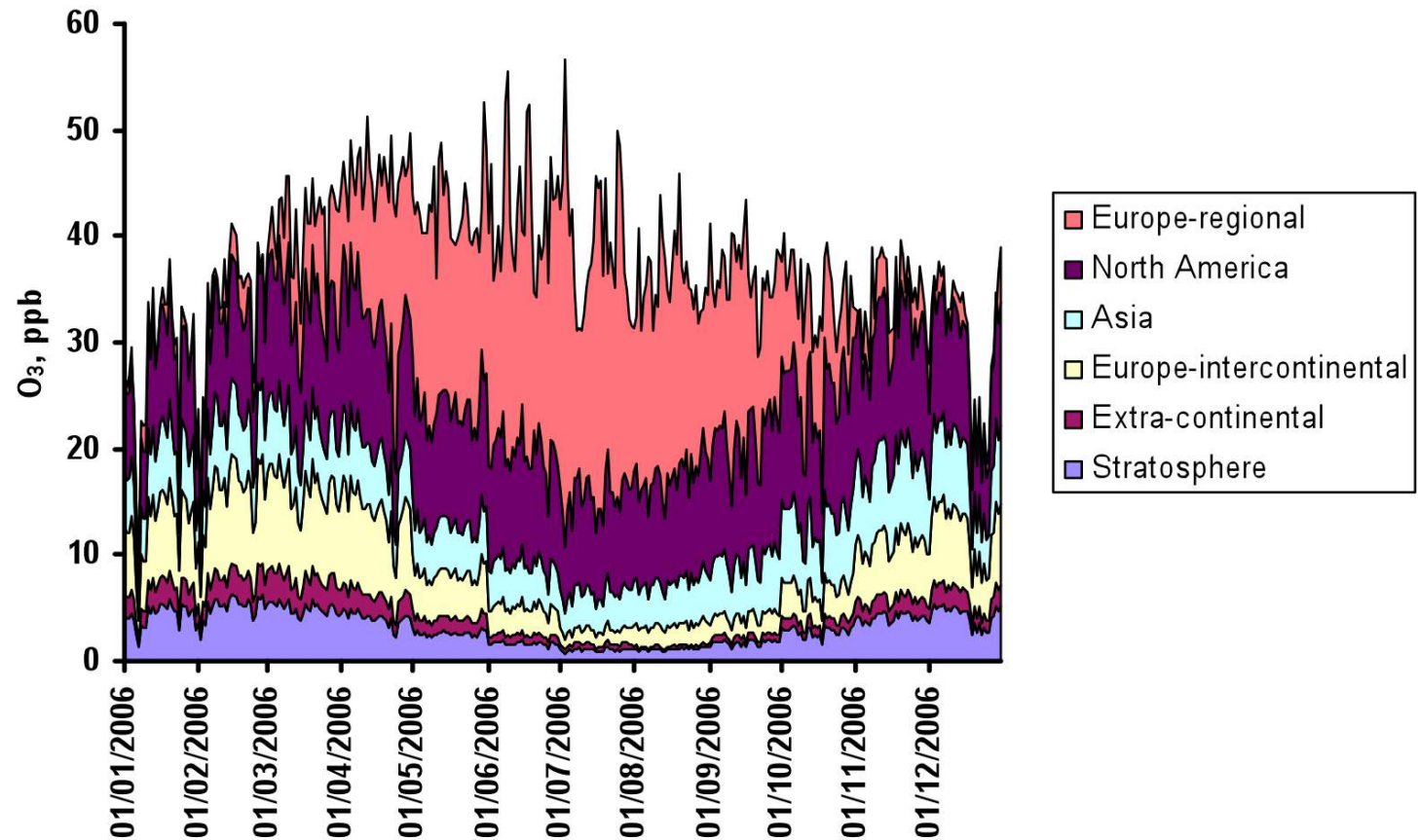
Vertical CO and O₃ profiles - MOZIAIC aircraft @ 2-10km over west coast USA, summer (HTAP, 2010)



Hemispheric Transport & Trends

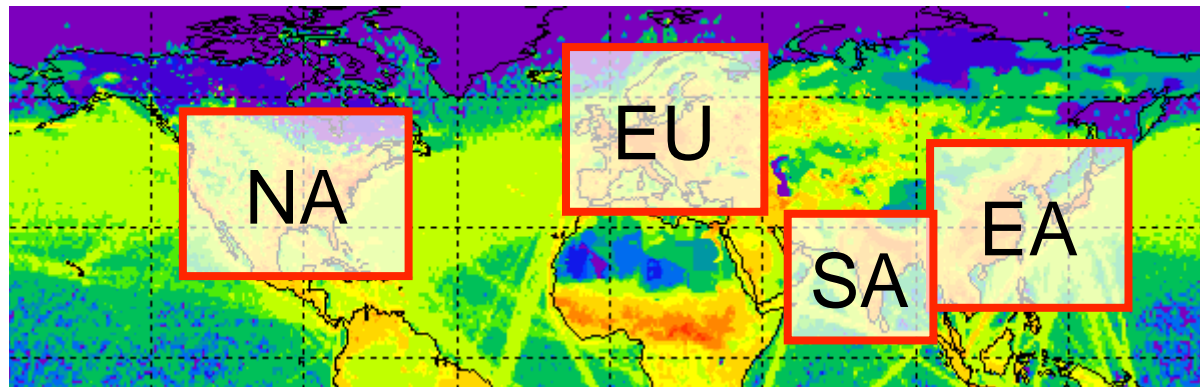
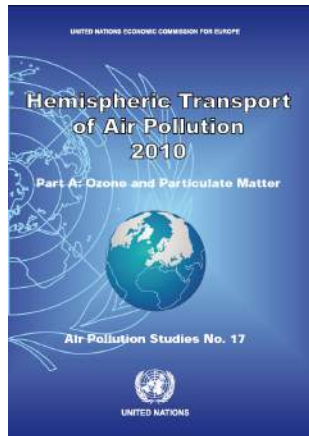


Observed Ozone: Mix of Origins



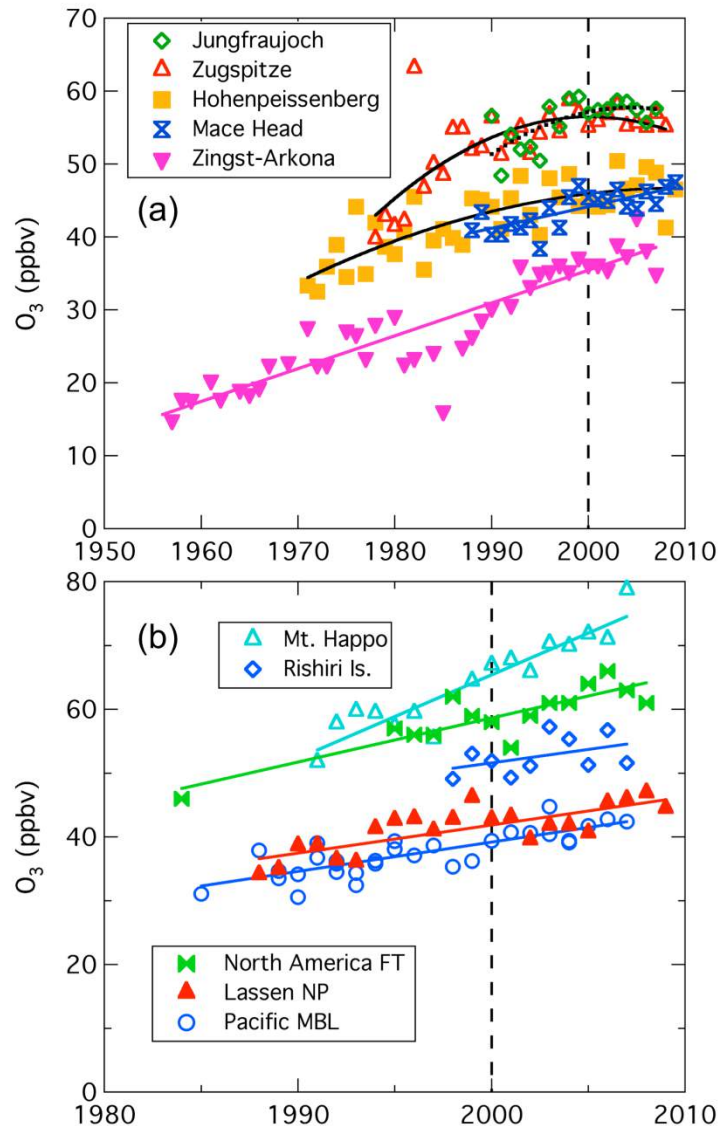
Analysis of natural, inter-continental and local contributions to surface O₃ (southern England)
Derwent et al. (2008)

Relative Annual Intercontinental Response (RAIR) Metric :
changes outside a region/ changes in all regions



- **At least 30% surface O₃ response in a receptor region is due to changes outside the region (NO_x, CO, VOCs) + 30-50% from CH₄**
- **External contribution more important when ‘local’ sources are more regulated and at lower concentrations**
- **Implications for ozone trends (past and future)**

Observed Ozone Trends

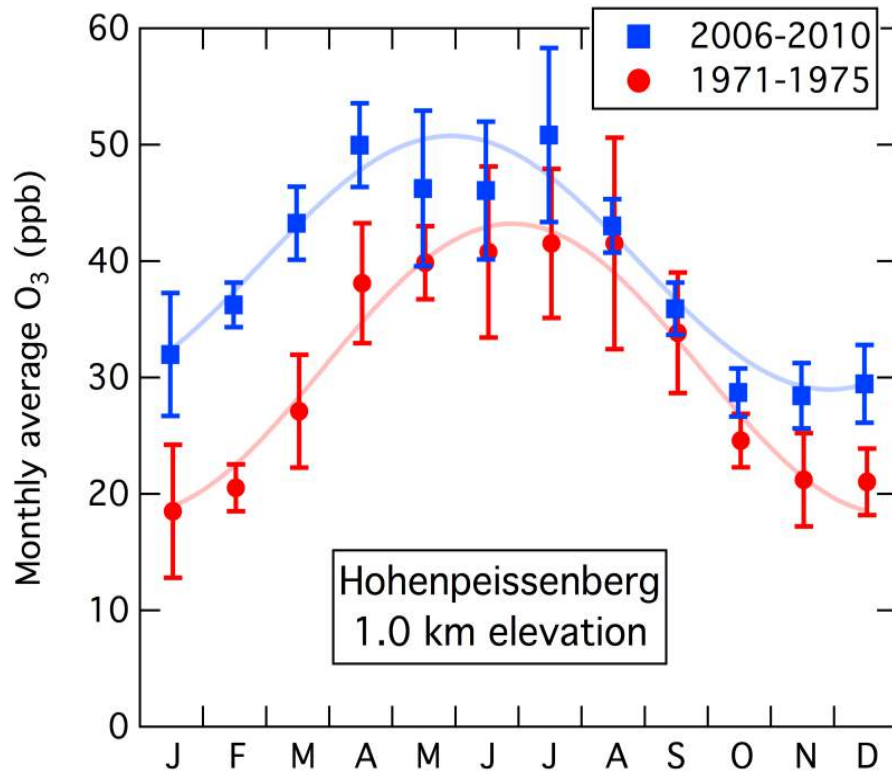


spring-time « background » ozone trends

- Local + inter-continental impacts reflected by changing average concentrations (anthrop. + natural)
- Since 1950 O₃ in northern mid-latitudes has increased by at least a factor of two – most likely due to anthrop. emissions
- O₃ continues to increase in certain regions (e.g. downwind Asia) – especially in winter and spring
- Slowing trends in central Europe and North America

*Parrish et al., 2012 +
also Logan et al. (2012) & other studies*

Changes in Observed Ozone Seasonal Cycle

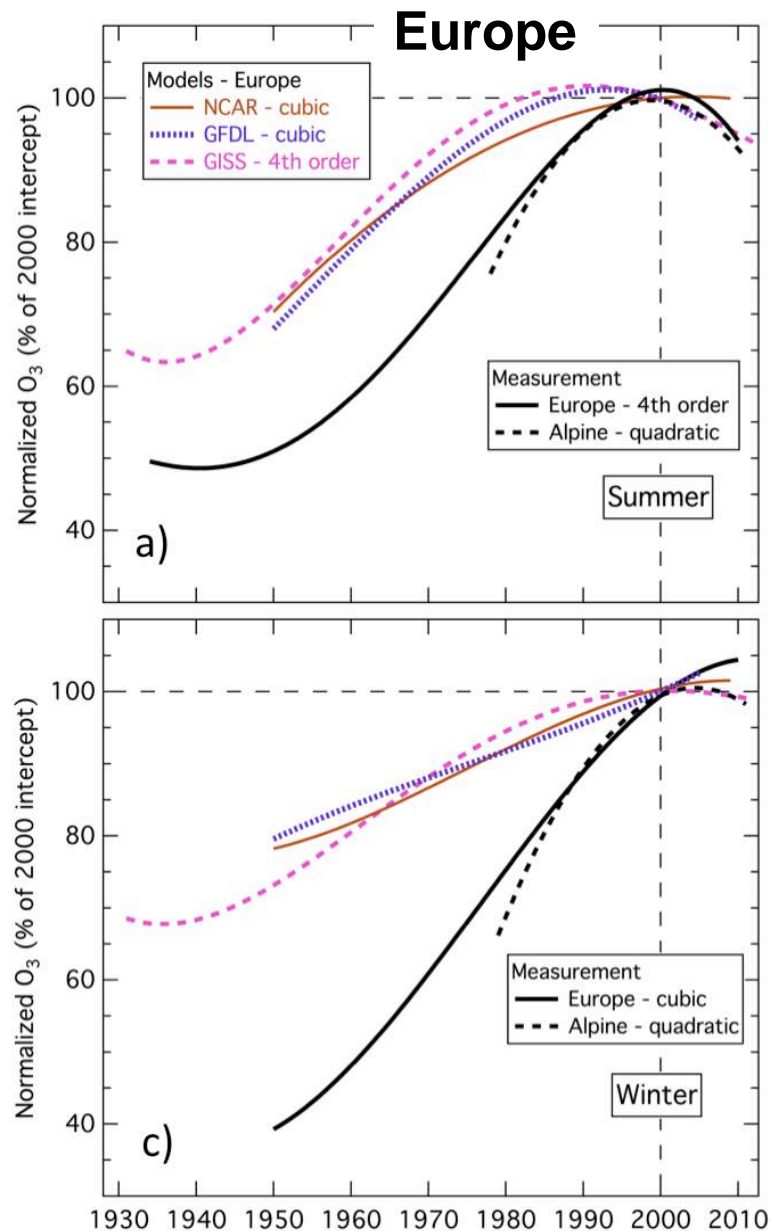


Parrish, Law, et al., 2013;

also Cooper et al., 2014

- Shift to earlier max. - ozone increases in winter/spring
- Implications for “chronic health effects” & crops (growing season)
- **Why?** (shifting emissions patterns (EU/NA↓, AS↑) coupled to large-scale transport (e.g. NAO), strat. flux, climate change?)
- Global models – first results suggest they have difficulties

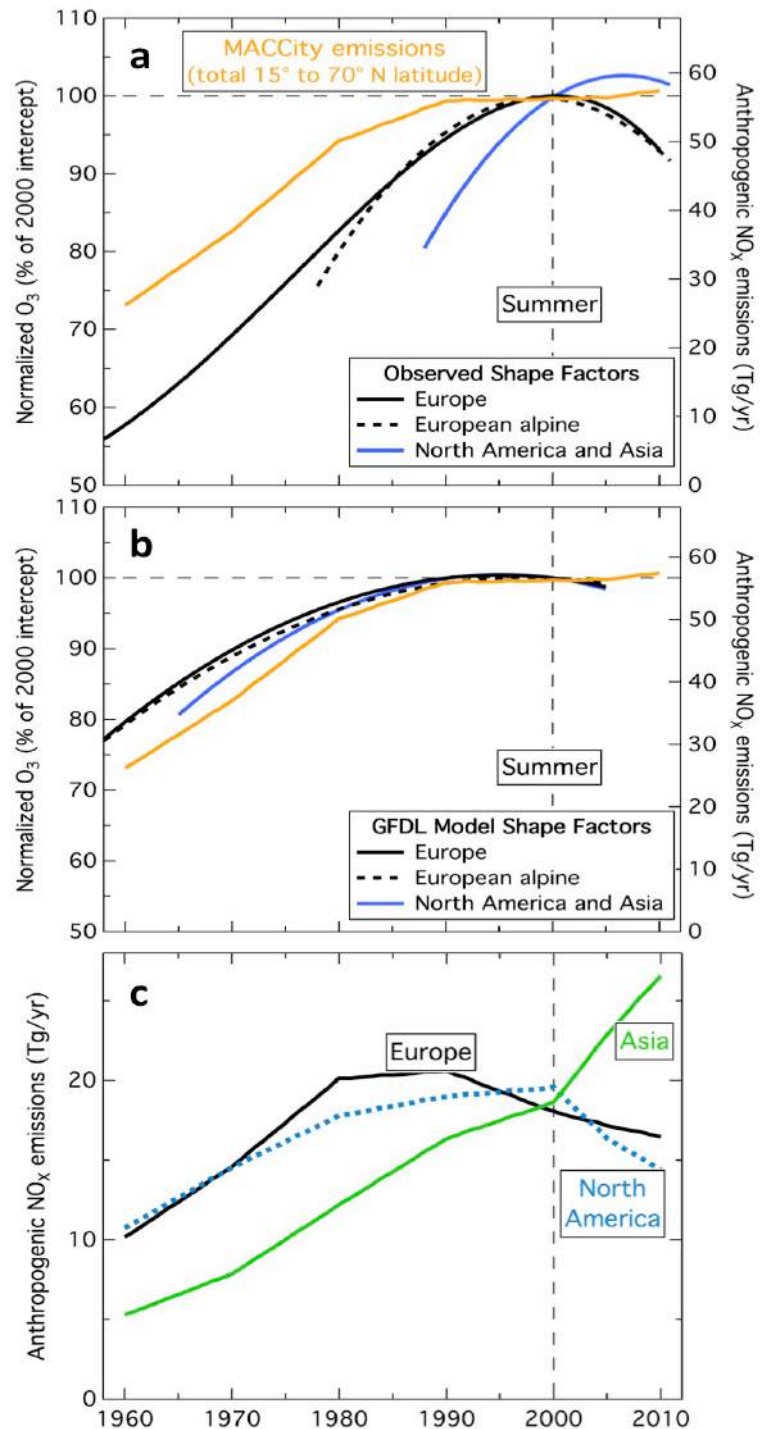
Northern Hemisphere Ozone Trends: Europe



Modelled trends:

- 50% of observed trends 1950-2000 (AR5 emissions)
- 25-40% of observed slowing (over Europe)
- Larger summer EU trends than winter (opposite to obs.)
- Overestimate 2000 concs.

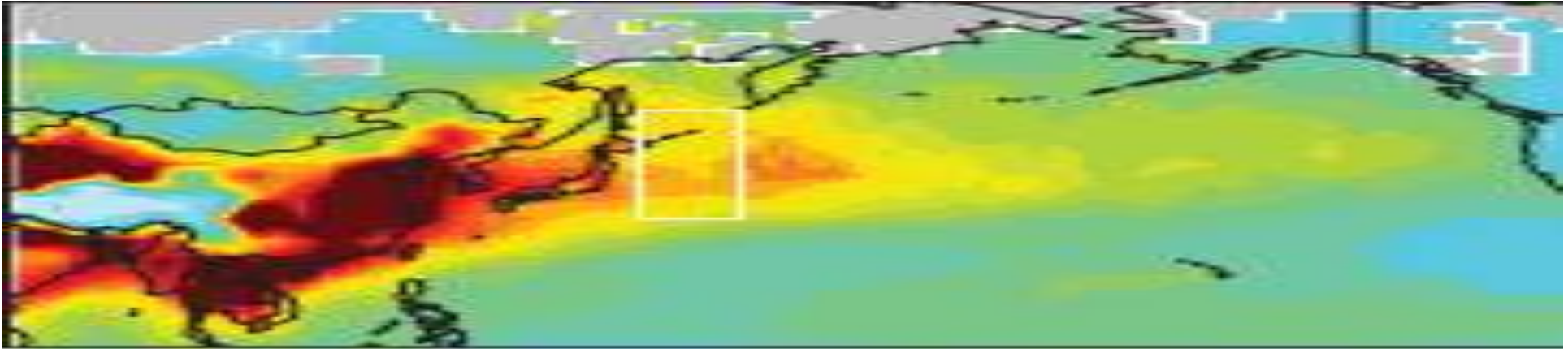
Parrish et al., (2014, JGR in revision)



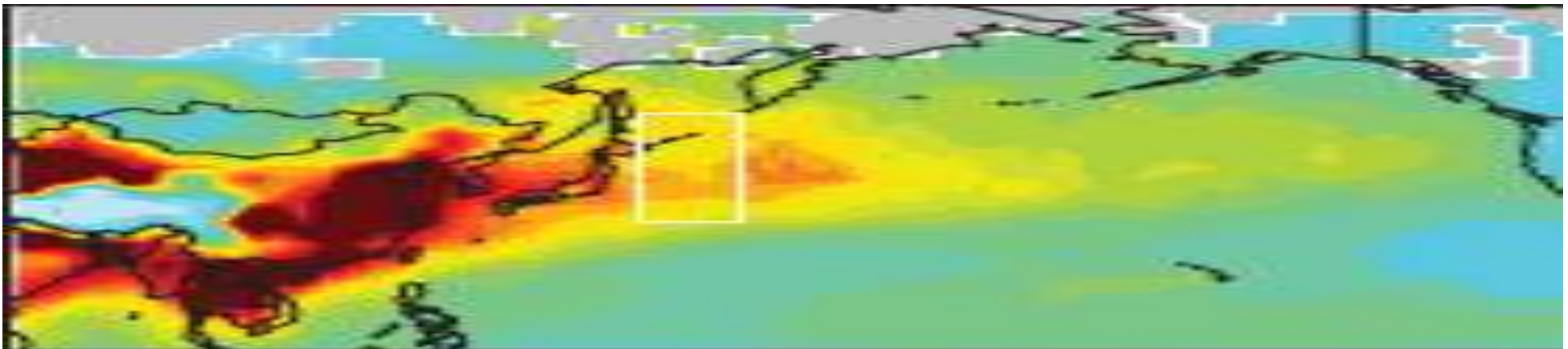
Modelled trends appear to be driven by precursor emission trends (e.g. NO_x)

Not capturing increases over Asia and N. America (west)

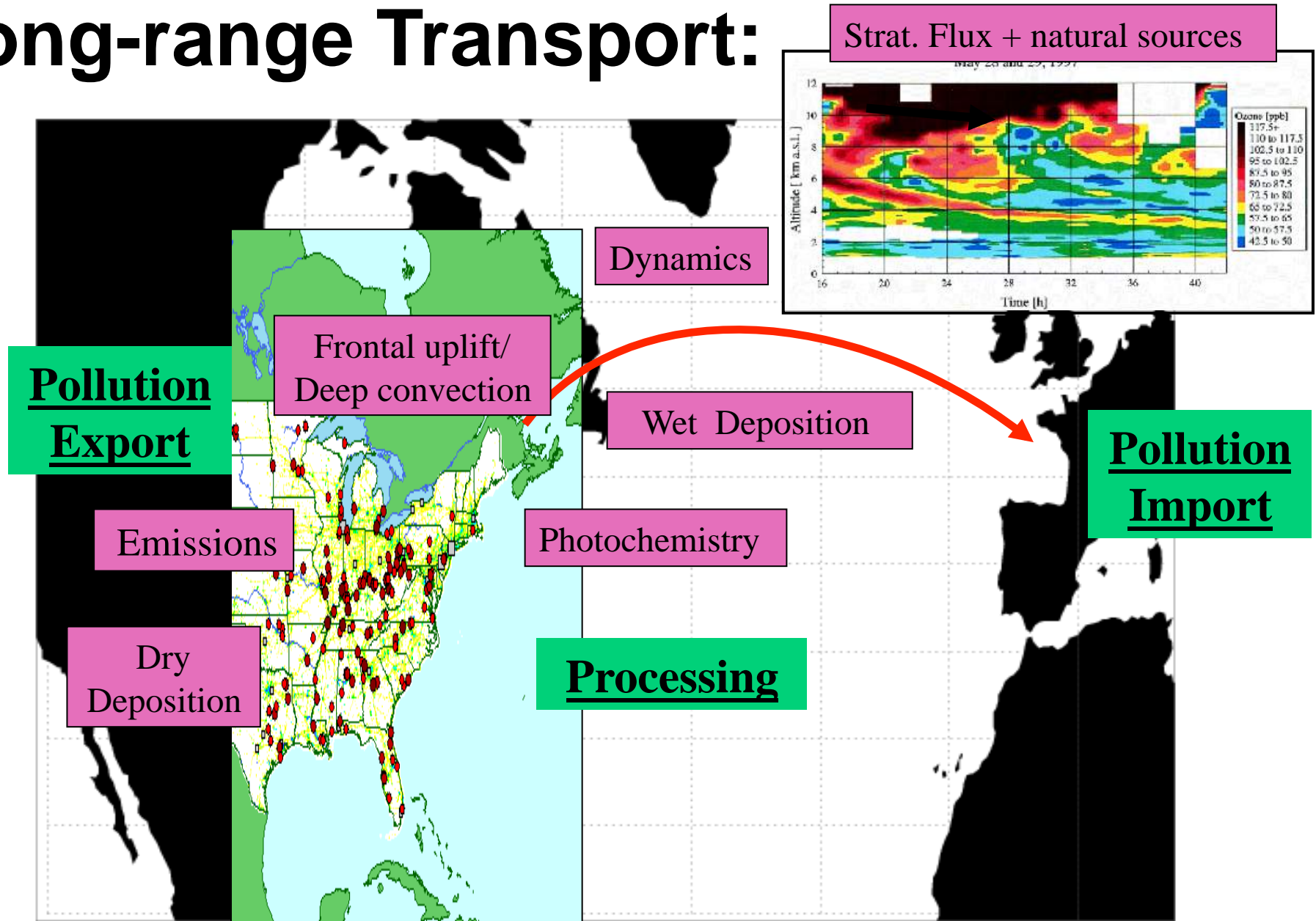
Parrish et al., (2014, JGR in revision)



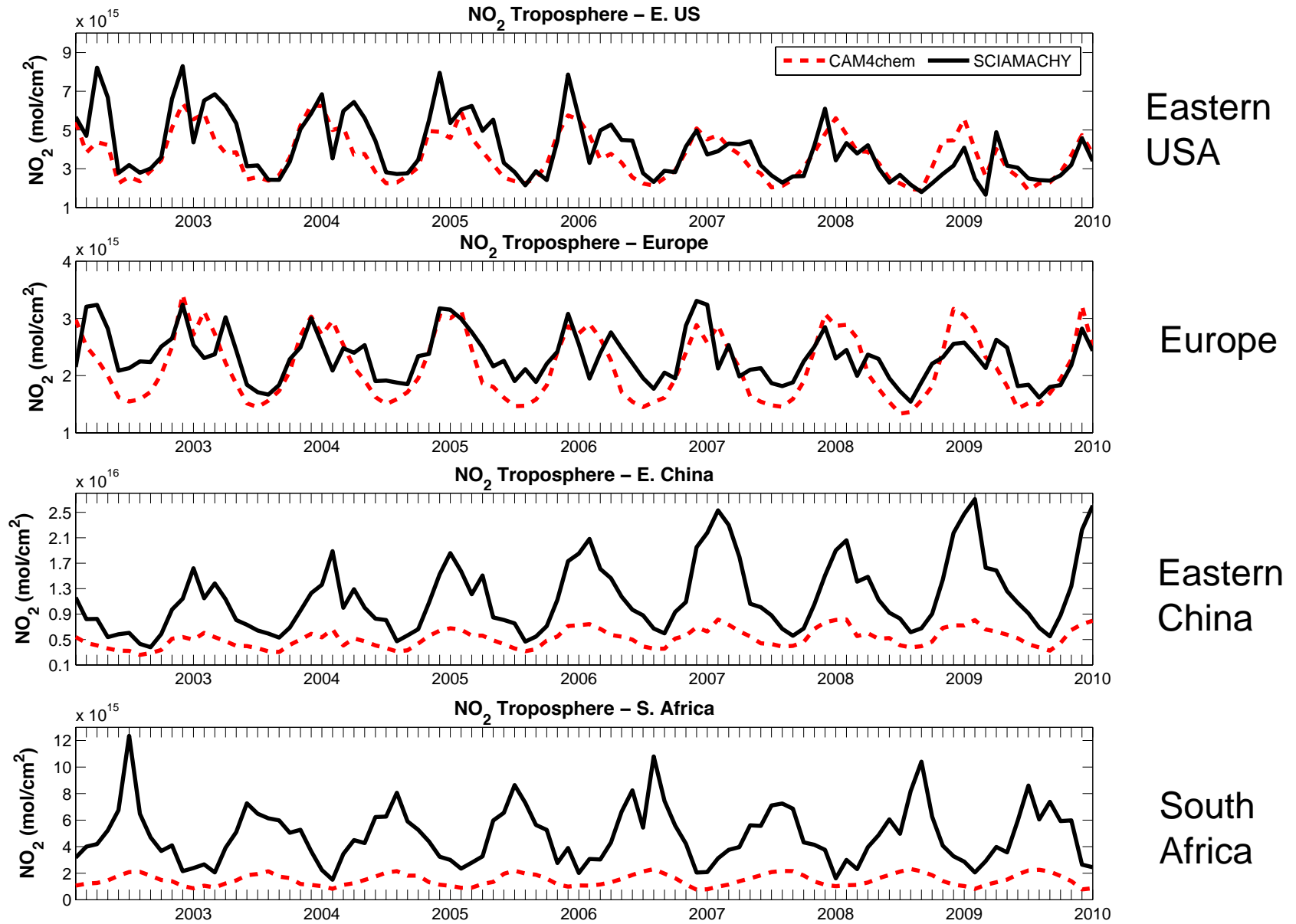
Pollutant (ozone) Export, Processing & Import



Long-range Transport:

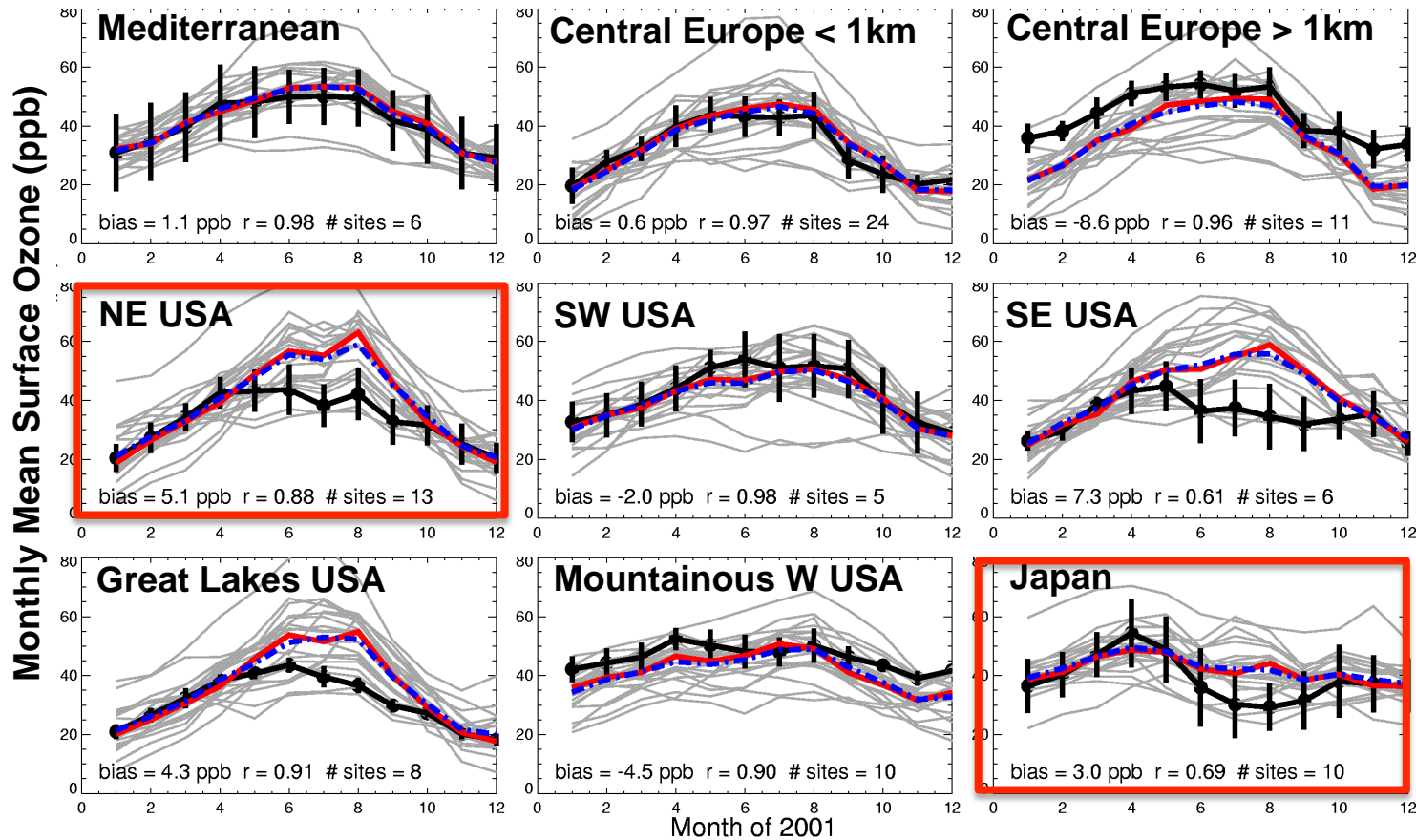


CAM4chem versus SCIAMACHY NO₂ (trop. columns)



HTAP 2010 Findings

Modelled Seasonal Cycles (2001)



Fiore et al. (2009)

Black-obs

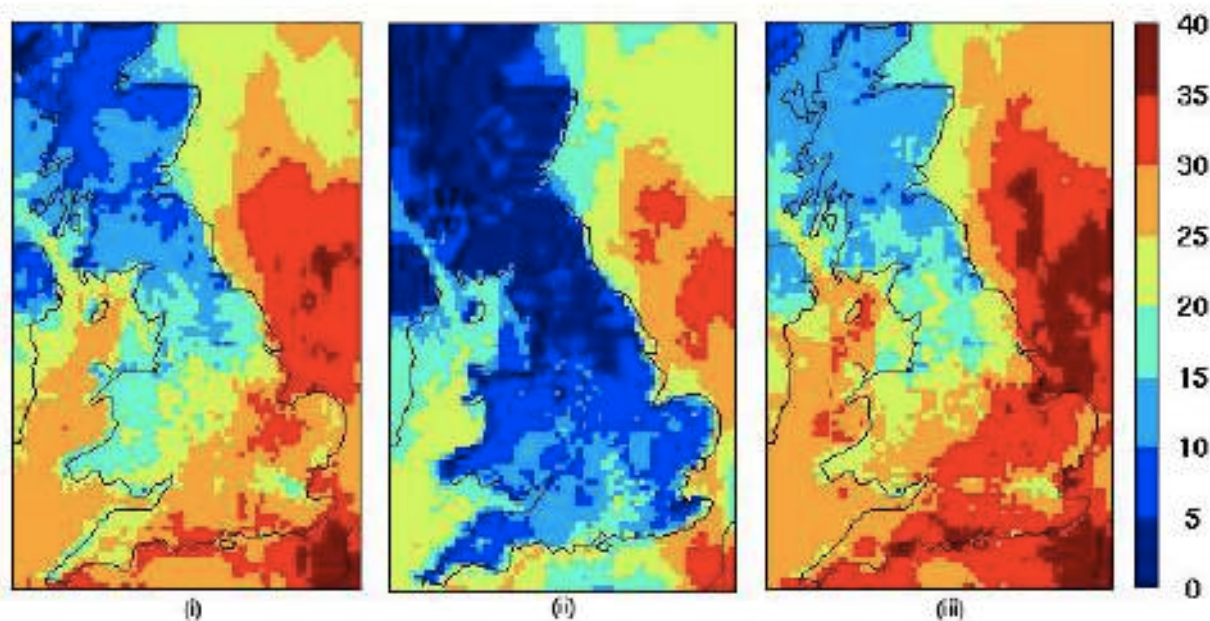
Large spread across models - ensemble mean generally captures observed monthly mean surface O₃ but **there are notable biases**

Ozone Deposition onto Vegetation

Ref: summer 2006

No stress

Drought stress

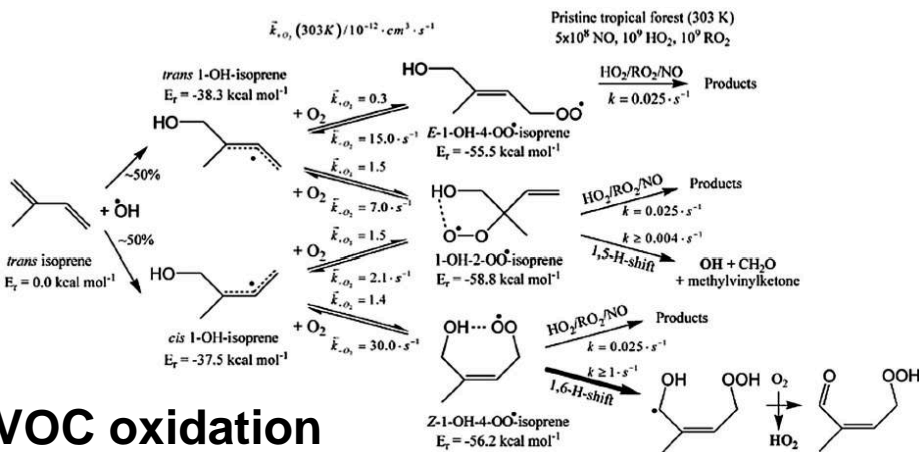


Emberson
et al.
(2013),
ACP

Fig. 3. Number of days when daily maximum 8 h average O₃ concentration > 100 µg m⁻³ during the June to July period in 2006 for (i) “reference”, (ii) “no stress” and (iii) “stress” scenarios.

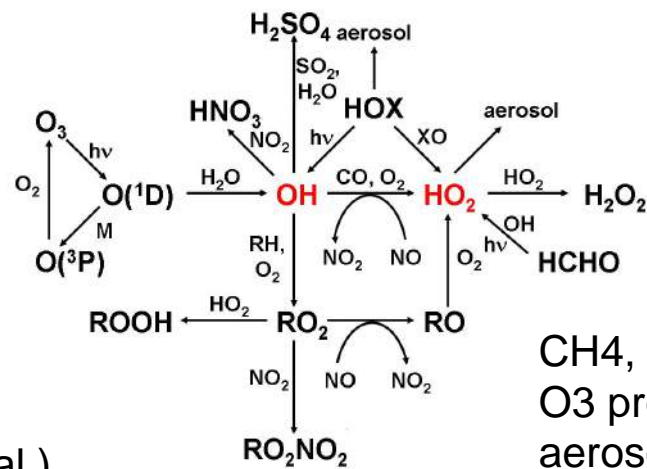
**ozone thresholds – chronic impacts even at
low concs (< 35 ppbv)**

Missing Processes?

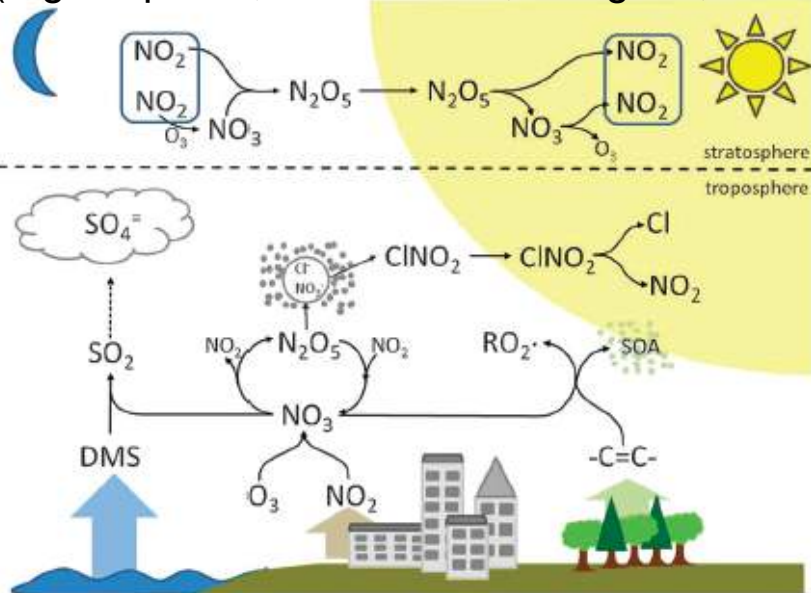


VOC oxidation

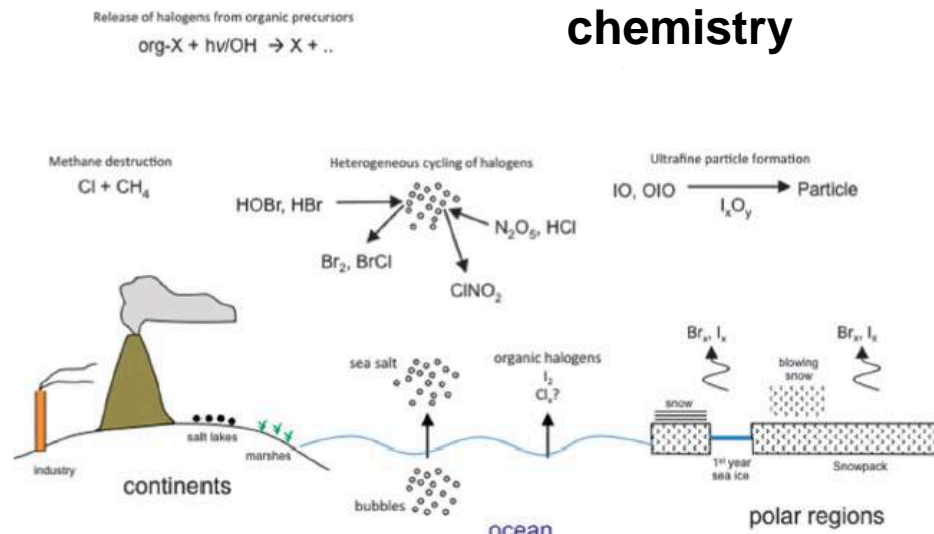
(e.g. isoprene, Peters et al.; Criegees, Maudlin et al.)



CH₄, CO oxidation,
 O₃ production,
 aerosol formation,
Heterogeneous chemistry



Night-time chemistry (NO₃, N₂O₅),
 Brown & Stutz (2012)

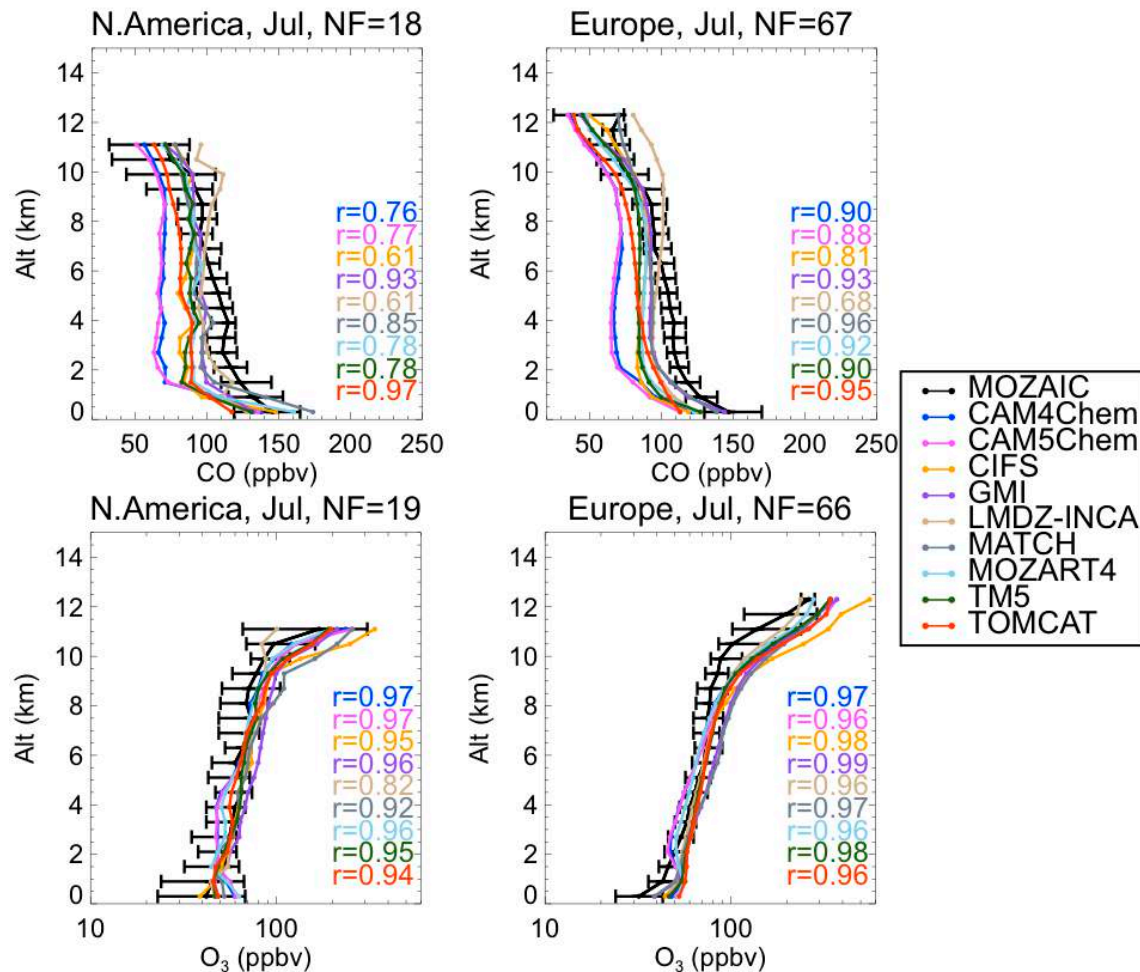


Halogen chemistry, (CINO₂, BrO)
 Saiz-Lopez & von Glasow (2012)



POLMIP: POLARCAT Model Inter-comparison Project

Models vs MOZAIC (July 2008)



Transport of ozone and precursors to the Arctic:

9 global models + WRF-Chem

Low biases in CO over source regions & Arctic driven by OH biases at mid-latitudes (+ vertical transport biases)

High ozone bias over source regions in UT (stratospheric ozone flux)

*Monks et al. (2014),
Arnold et al. (2014)*





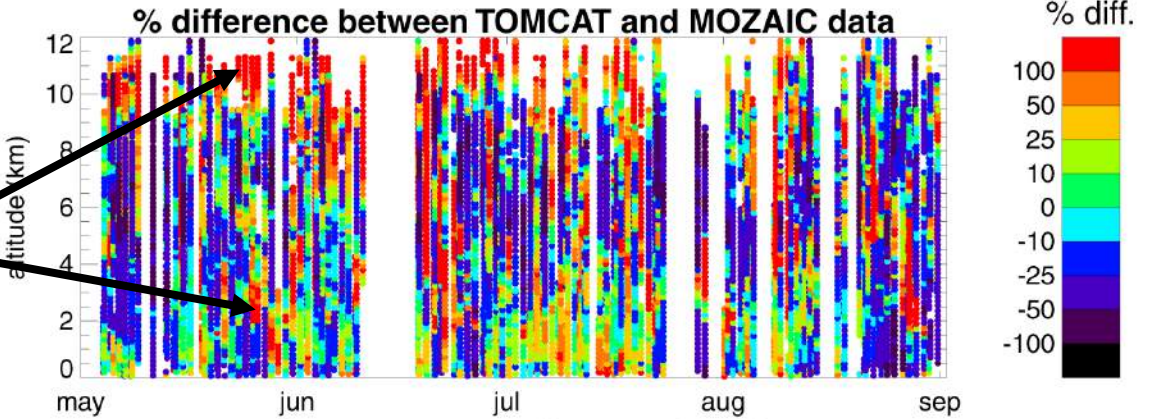
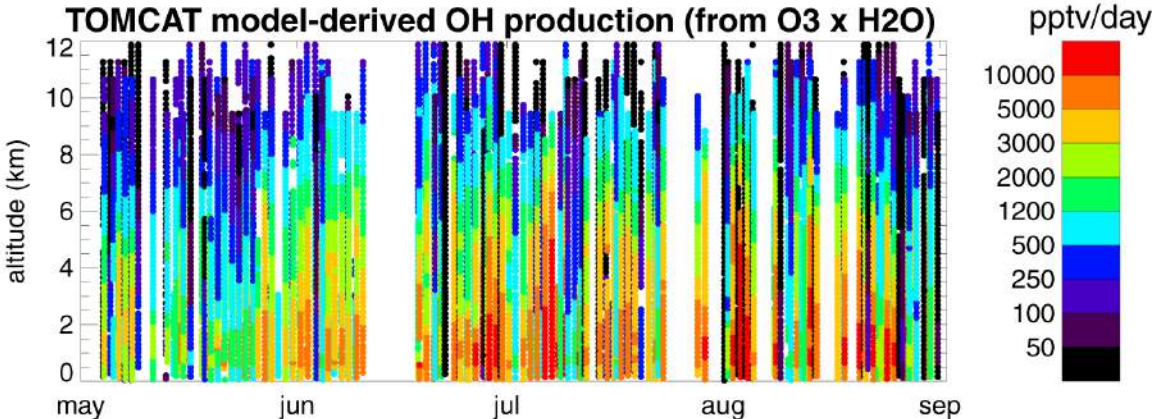
Global Oxidising Capacity = OH concentration

OH radicals control the lifetimes of many greenhouse gases (CH₄, O₃, HCFCs)

MOZAIC data used to validate modelled **primary photochemical production of OH** in the troposphere*

model overestimates OH production (from O₃ x H₂O) by more than 100%

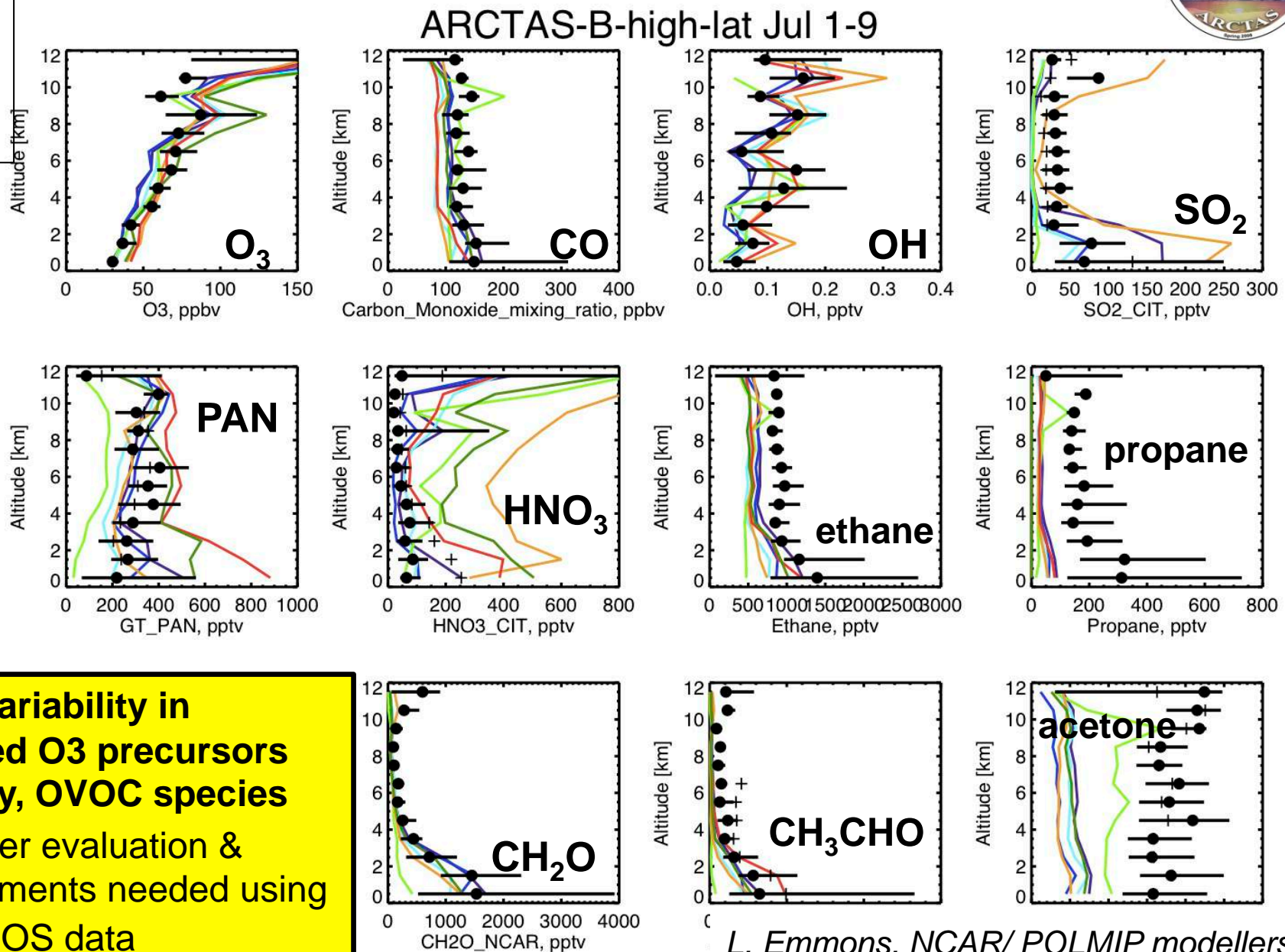
*JO1D, k's taken from TOMCAT



POLMIP: evaluation using ARCTAS data (July 2008)



Colours:
models
• obs

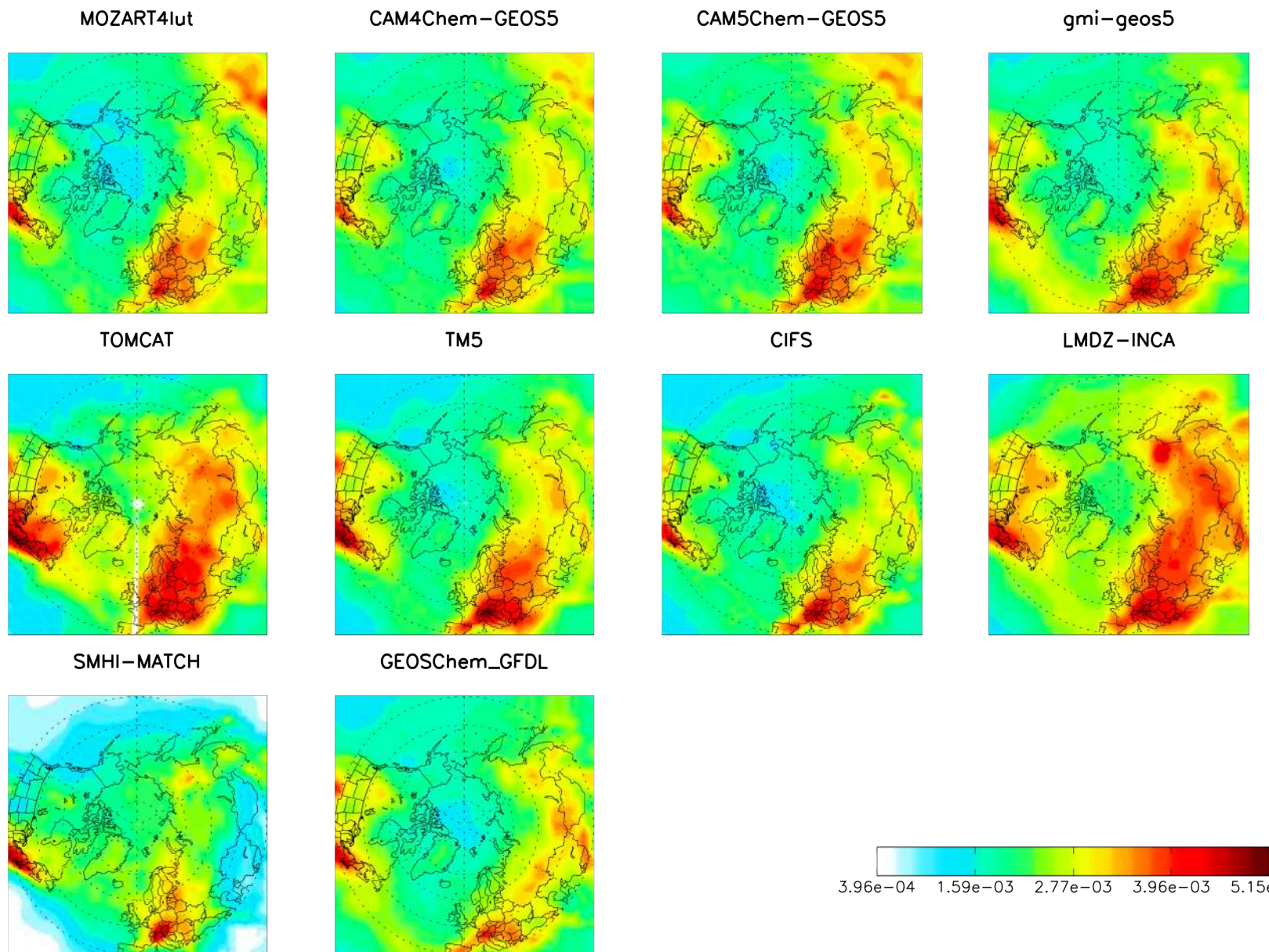



Large variability in modelled O₃ precursors and NO_y, OVOC species
 → Further evaluation & improvements needed using e.g. IAGOS data

L. Emmons, NCAR/ POLMIP modellers

POLMIP Model PAN:CO (July, column ratios)

pan/co

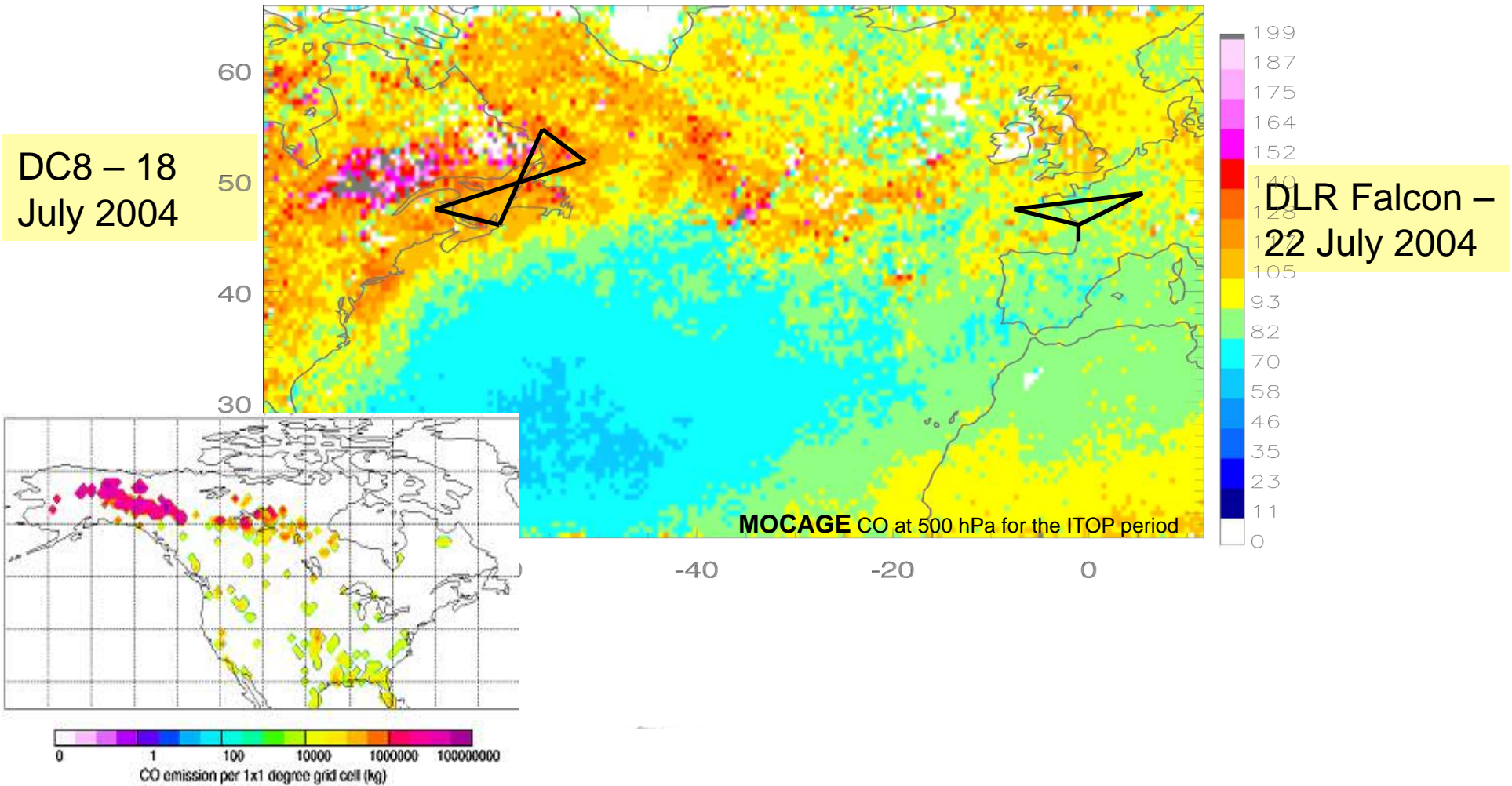


Courtesy Steve Arnold  UNIVERSITY OF LEEDS

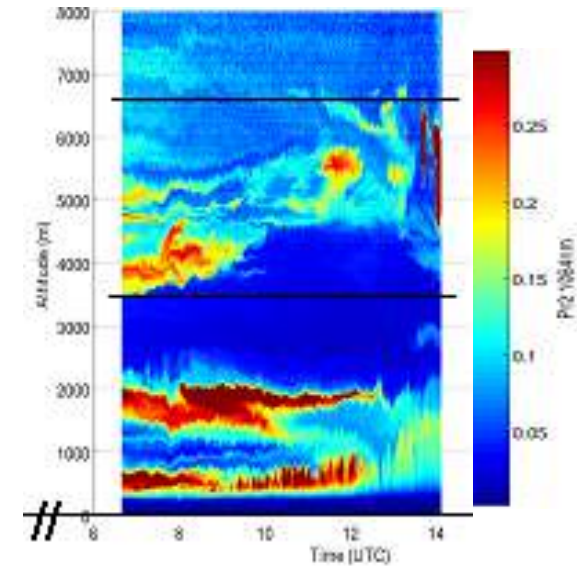
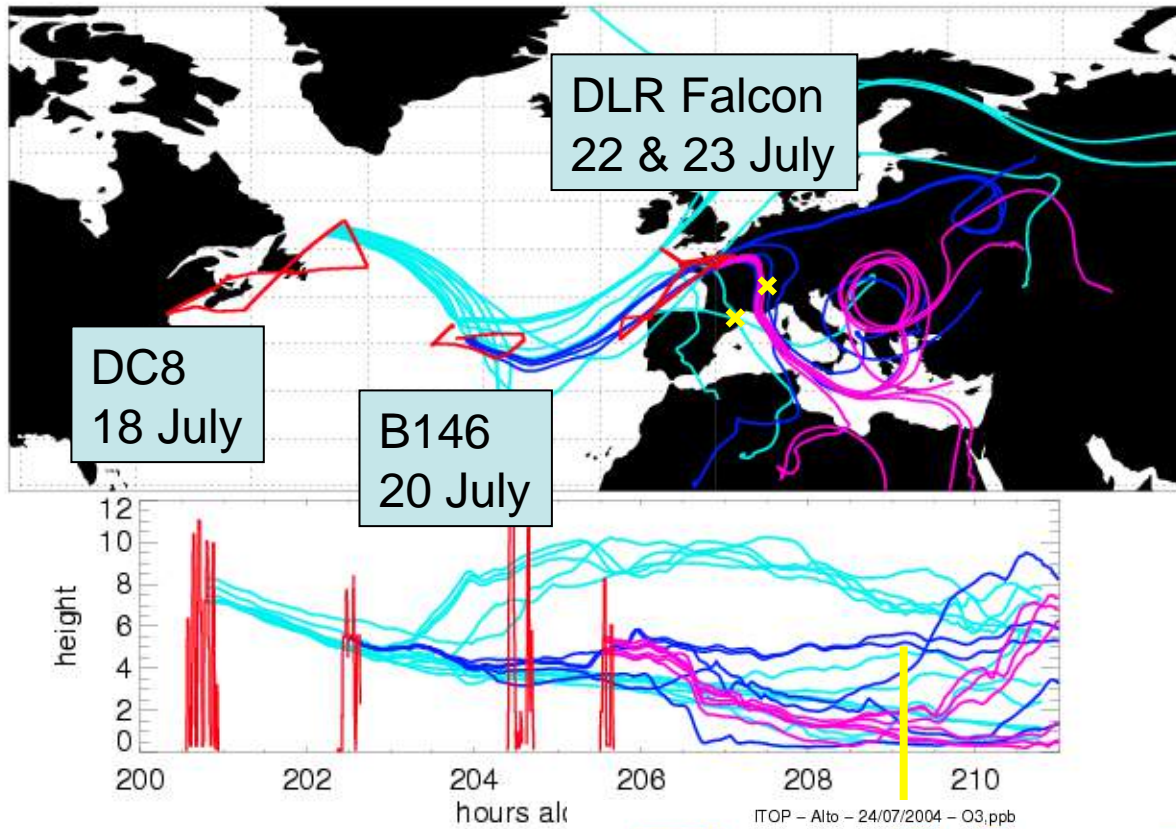
Plume processing: Alaskan Forest Fire Plume (July 2004)

DC8 – 18
July 2004

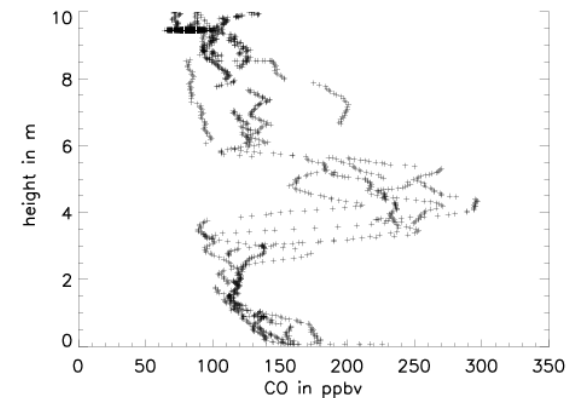
DLR Falcon –
22 July 2004



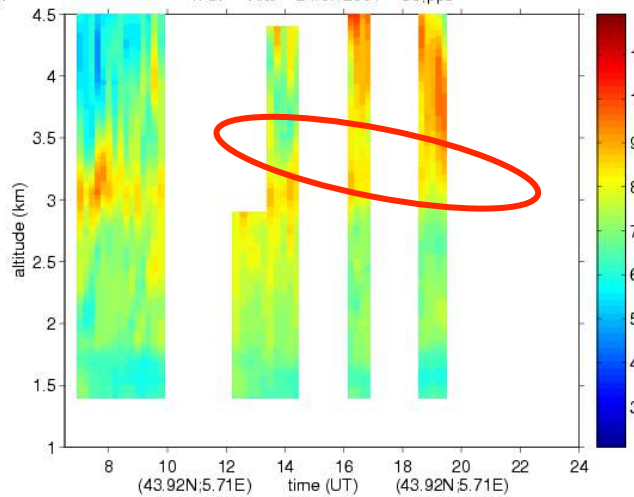
ICARTT Lagrangian Analysis: Import of Alaskan FF plume into European PBL



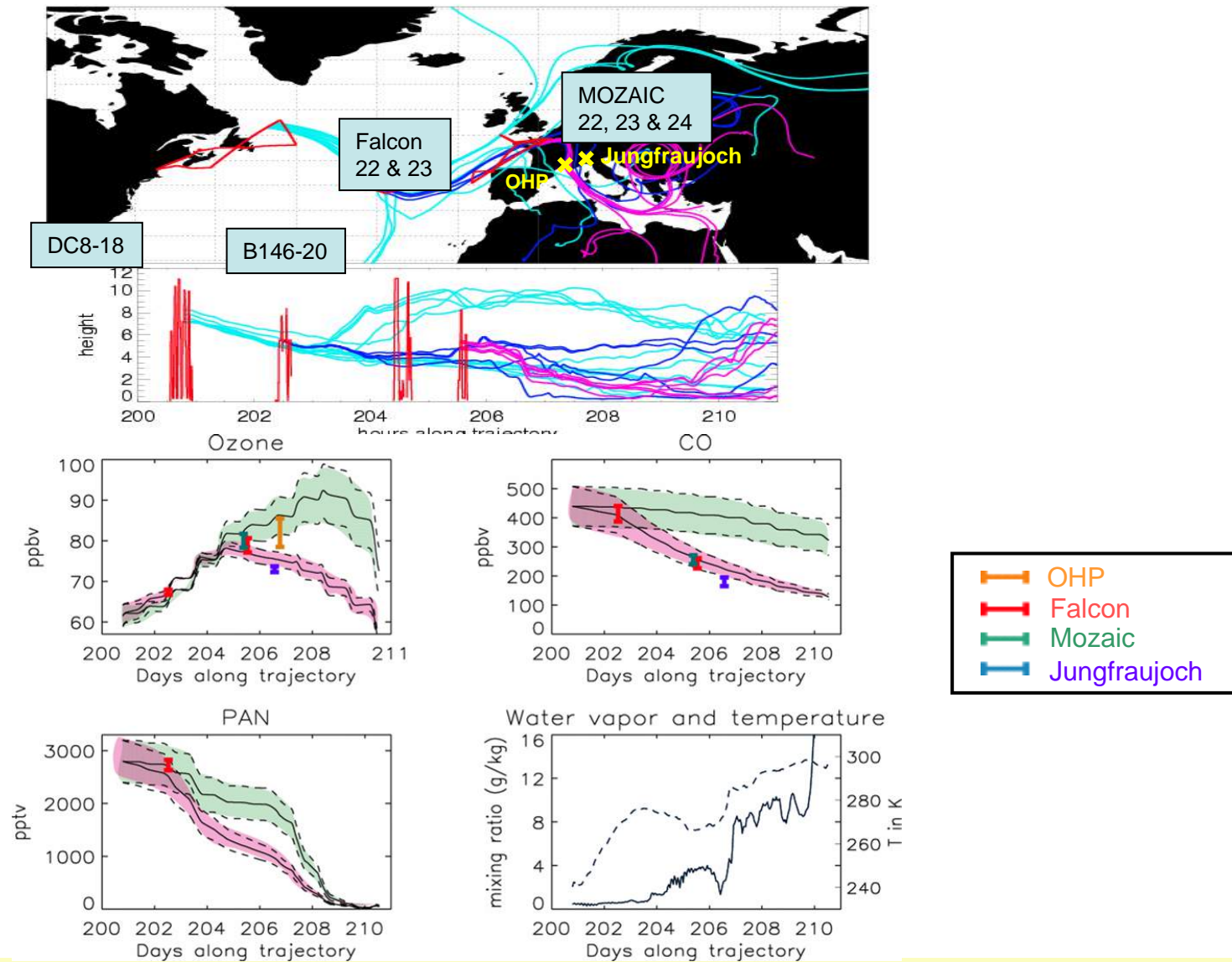
Paris aerosol lidar & MOZAIC 22/23/24 July



Ozone lidar - south France, 24 July



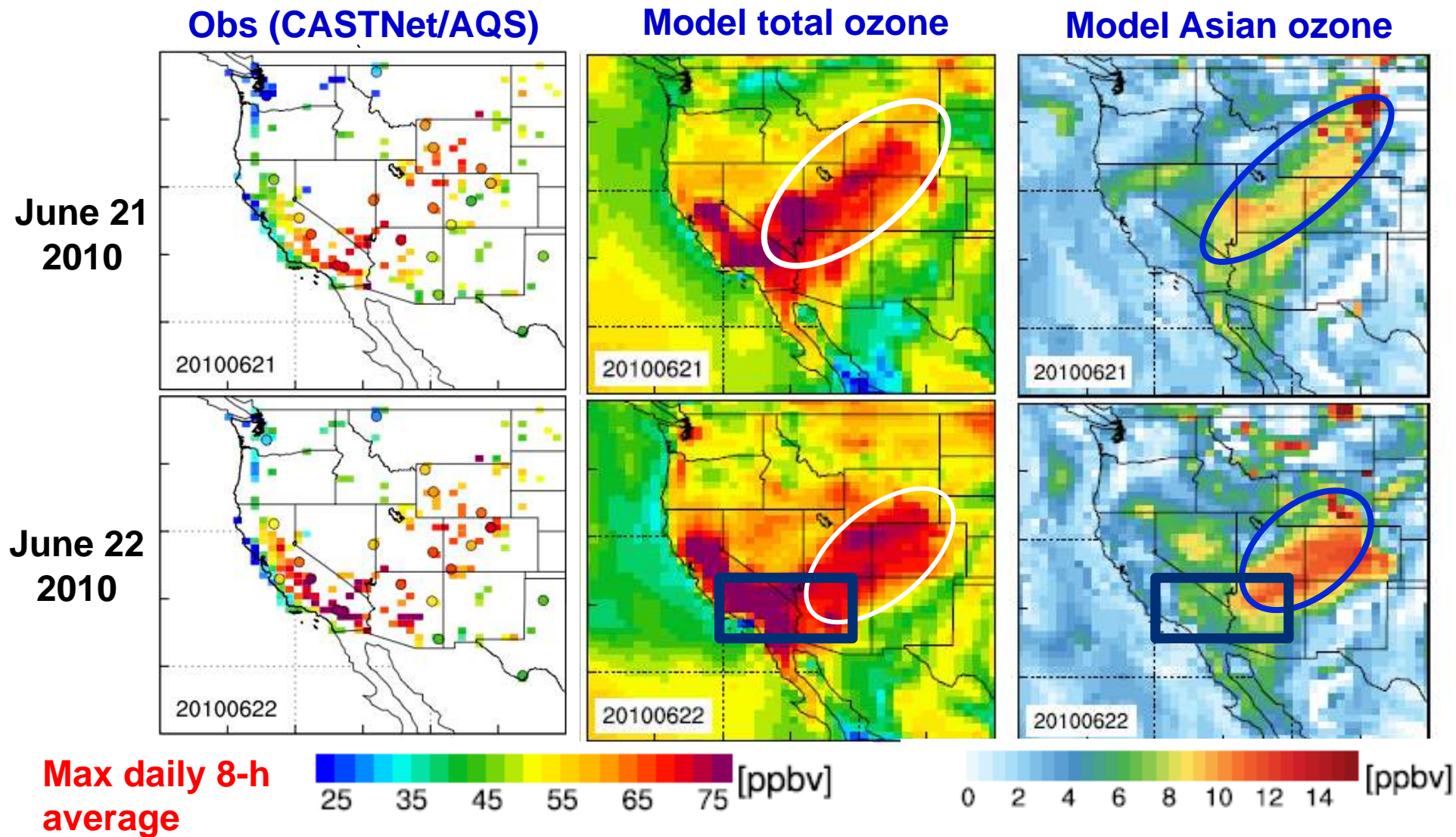
ICARTT Lagrangian Analysis: Import of Alaskan FF plume into European PBL



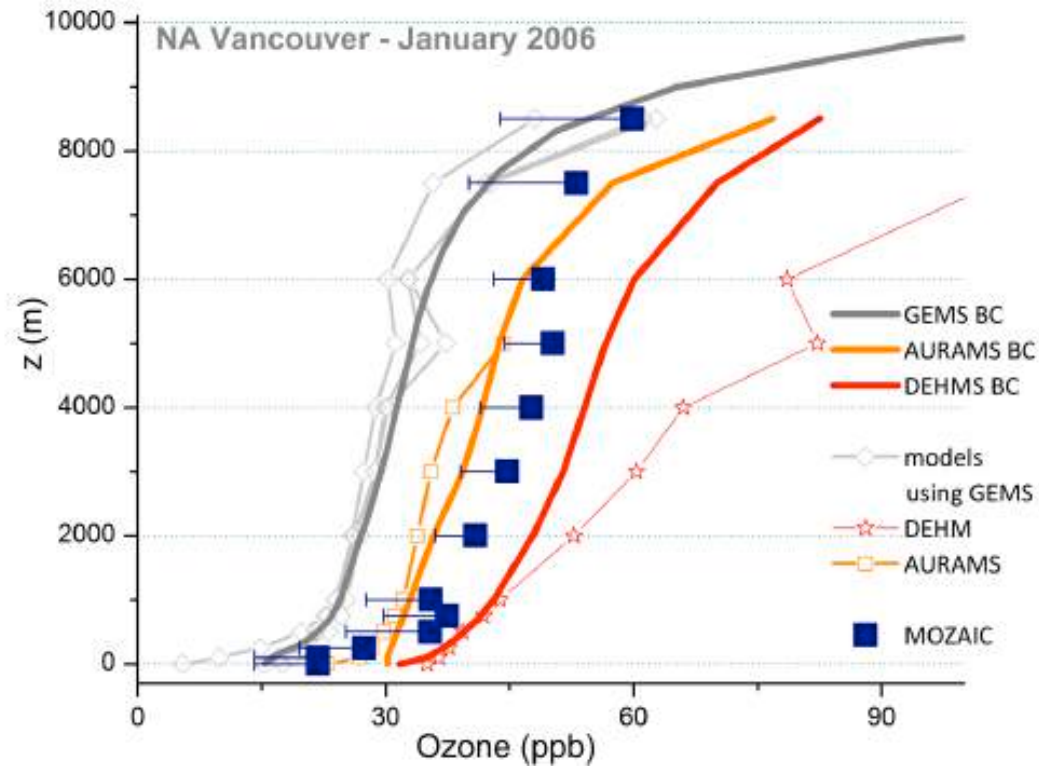
Net production during 8 days + net ozone import into European PBL

(Real et al., 2007) – Photochemical Lagrangian calculations

Asian pollution contributing to high surface O₃ events, Implication for attainment of tighter standards in WUS



Importance of Ozone Import



Winter (western N. America)

- Systematic low biases in ozone boundary conditions from global models lead to underestimation of regional model ozone at the surface

- Significant penetration of signal into the regional model domain

Summer

- Biases in FT ozone during summer but not affecting surface O₃ (large biases)

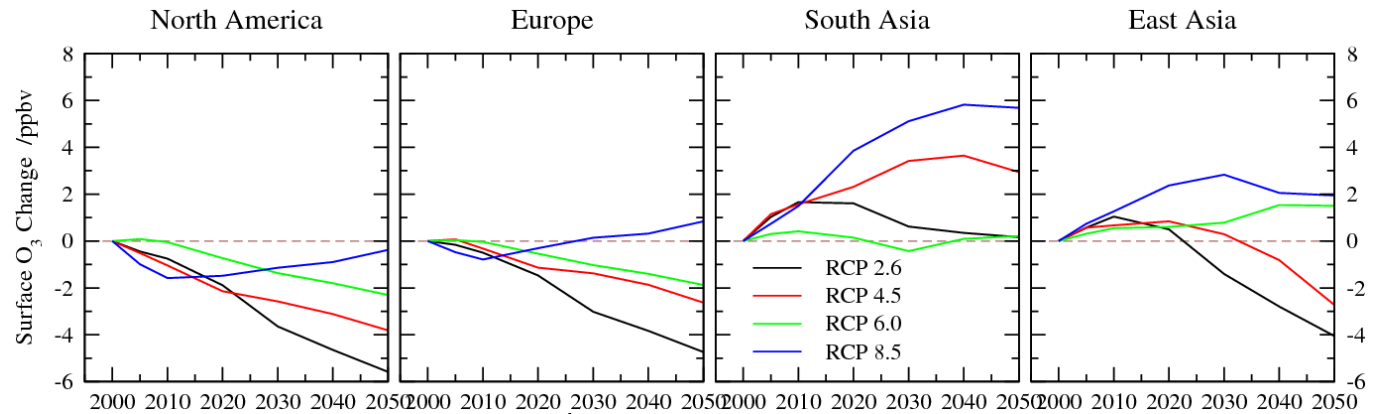
14 regional models:
12 used GEMS boundary conditions
& run using same emissions

AQMEII: Solazzo et al. (2013)

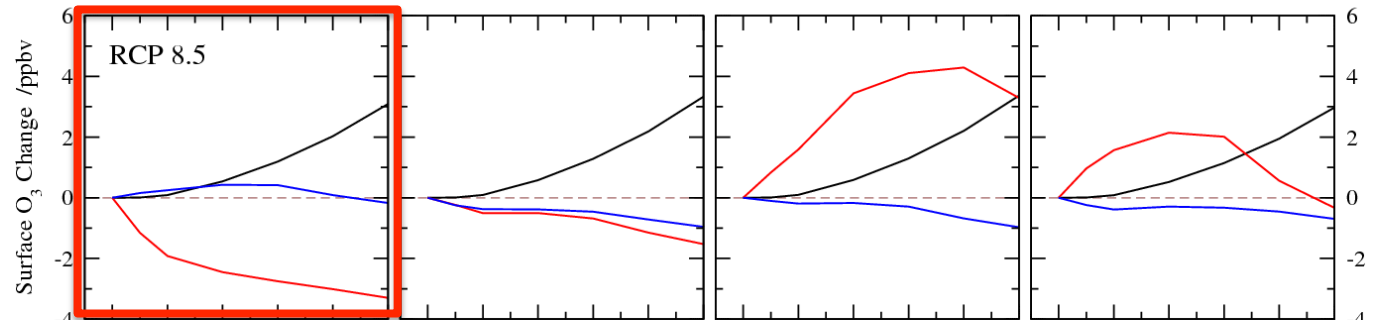
Linearized results of 6 models (Wild et al., 2012):

Role of CH₄ in Future O₃ Scenarios

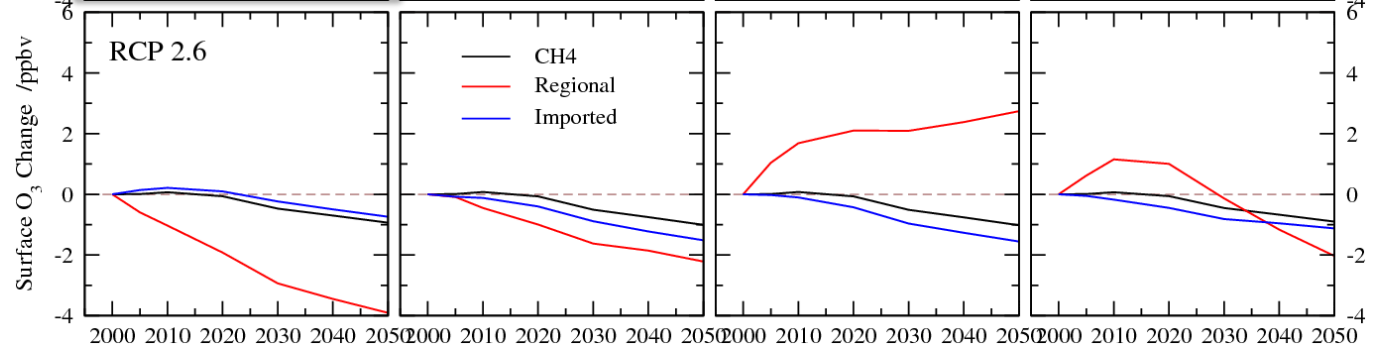
O₃ Change in 4 emission scenarios



CH₄, Regional, & Imported Components of O₃ Change in “High” Scenario



Components of O₃ Change in “Low” Scenario



CH₄ is an important determinant of future O₃ levels, offsetting benefits of regional controls.

Hemispheric Ozone: Challenges

- **Hemispheric ozone (increasingly) important** - depends on local (AQ) versus hemispheric control (CLRTAP) (CH₄) esp. as local emissions decrease and thresholds are lowered
- **Ozone source attribution + trends** – understanding still incomplete (datasets like IAGOS invaluable)
- **Pollutant import** much less well evaluated → focused area of research (*high time resolution vertical profiles, e.g. on regional carriers ...*)