Anthropogenic and biogenic contributions to intercontinental transport of ozone pollution at northern mid-latitudes

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TF HTAP Modeling Team & Many Measuring PAN

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Observing intercontinental transport at northern mid-latitudes: July 2004 Alaskan and Canadian Fires

Mean 500 mb carbon monoxide (combustion effluent) retrieved from the AIRS instrument (http://airs.jpl.nasa.gov)

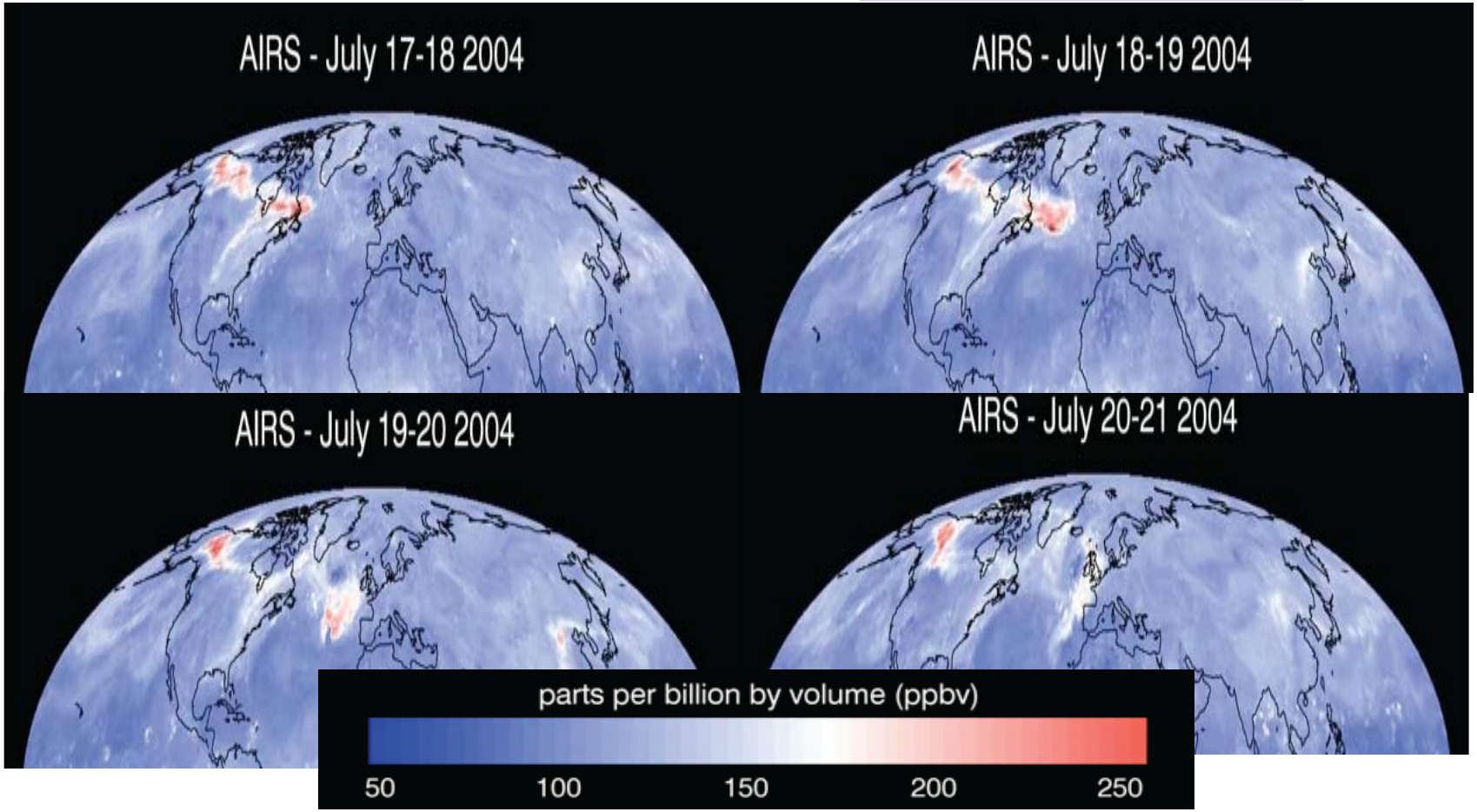
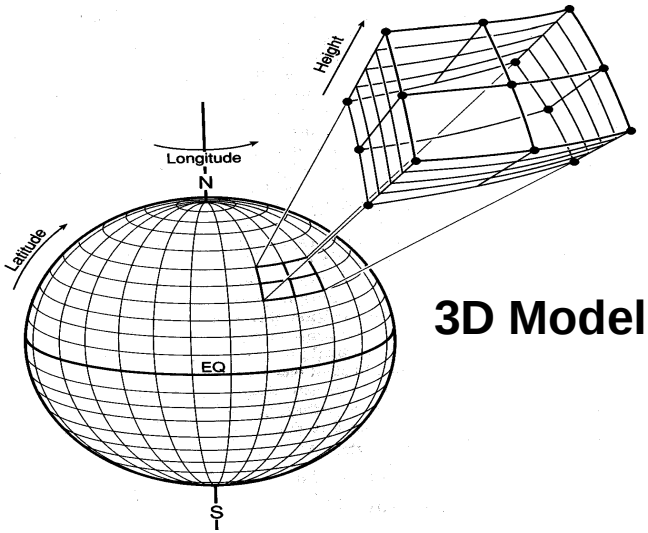


Image credit: NASA/JPL; http://photojournal.jpl.nasa.gov/catalog/PIA11034
Frames c/o Yuanyuan Fang, Princeton/GFDL

Intercontinental influence on surface ozone may occur mainly through "hemispheric background"

Intercontinental ozone transport is difficult (impossible?) to observe directly at surface ***[e.g., Derwent et al., 2003; Fiore et al., 2003; Goldstein et al., 2004; Jonson et al., 2005]***

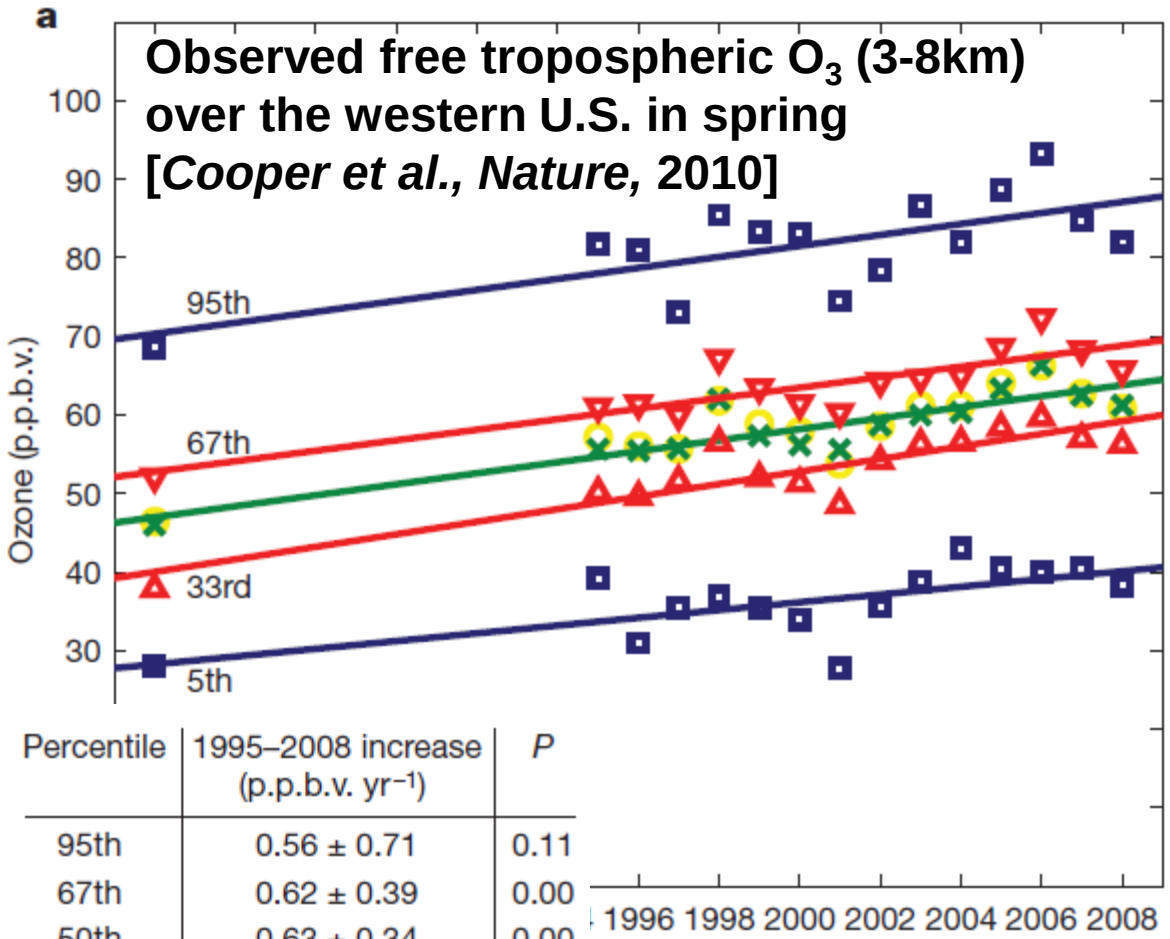
→ Estimates rely on models (evaluated with observations)



3D Model Structure

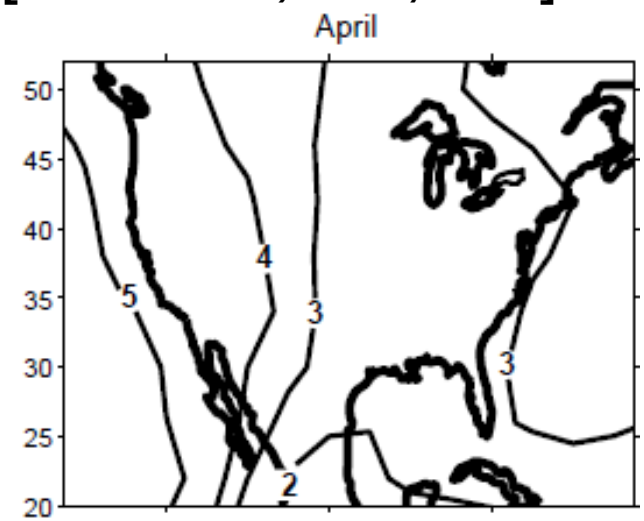
→ Long-term measurements should contain information on tropospheric (and surface) O_3 response to trends in emissions (key tests for models?)

Observational and modeling evidence indicates rising baseline tropospheric O_3 from human activities



Percentile	1995–2008 increase (p.p.b.v. yr^{-1})	*P*
95th	0.56 ± 0.71	0.11
67th	0.62 ± 0.39	0.00
50th	0.63 ± 0.34	0.00
33rd	0.64 ± 0.35	0.00
5th	0.48 ± 0.53	0.07

Modeled increase in surface O_3 due to tripling Asian anthropogenic emissions from 1985 to 2010 [*Jacob et al., GRL*, 1999]



How much free tropospheric Asian pollution mixes into surface air?

Rising background O_3 has implications for attaining ever-tightening air quality standards

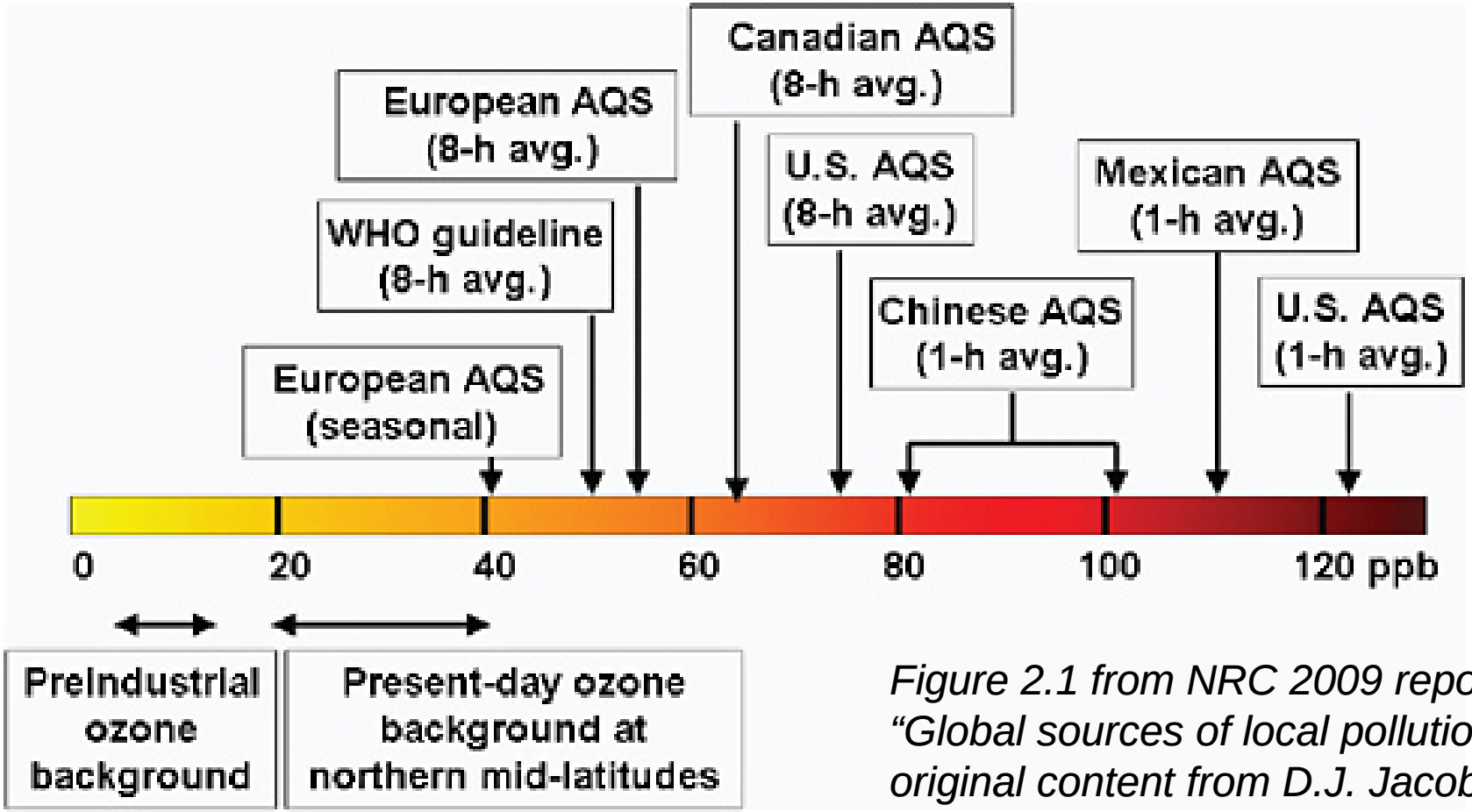
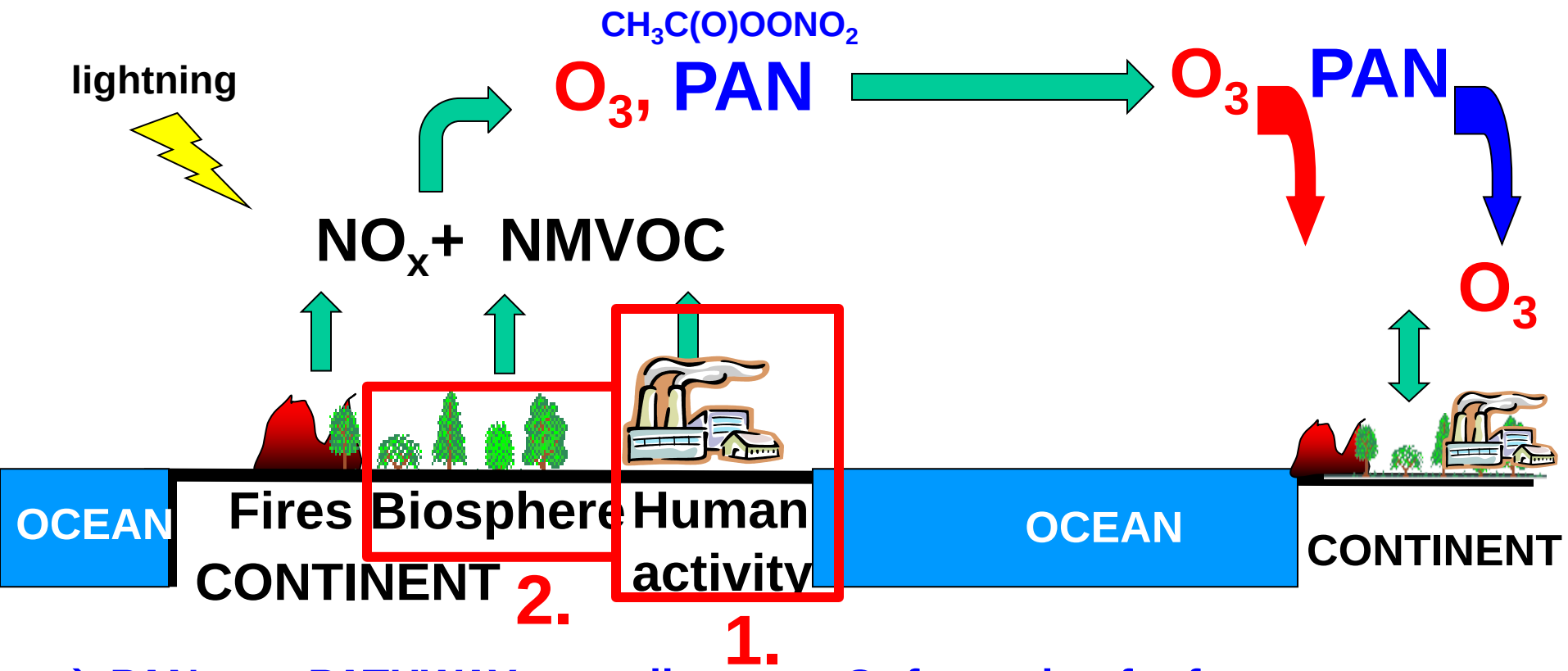


Figure 2.1 from NRC 2009 report, "Global sources of local pollution"; original content from D.J. Jacob

Observational evidence for increasing surface O_3 background [e.g. *Lin et al*., 2000; *Jaffe et al*., 2003, 2005; *Vingarzan*, 2004; *EMEP/CCC-Report 1/2005 ; Derwent et al., 2007; Jaffe and Ray*, 2007; *Royal Society*, 2008; *NRC, 2009*]

Human activities and "natural" sources influence intercontinental O_3 transport



→ **PAN as a PATHWAY: contributes to O_3 formation far from source region** [*e.g., Moxim et al.*, 1996; *Heald et al., JGR, 2003; Hudman et al., JGR, 2004;Zhang et al., 2008; Fischer et al., 2010*]

→ **PAN as a PROXY: may reflect changing O_3 precursor sources better than O_3 itself** [*Jaffe et al.*, 2007; *Fischer et al.*, 2010]

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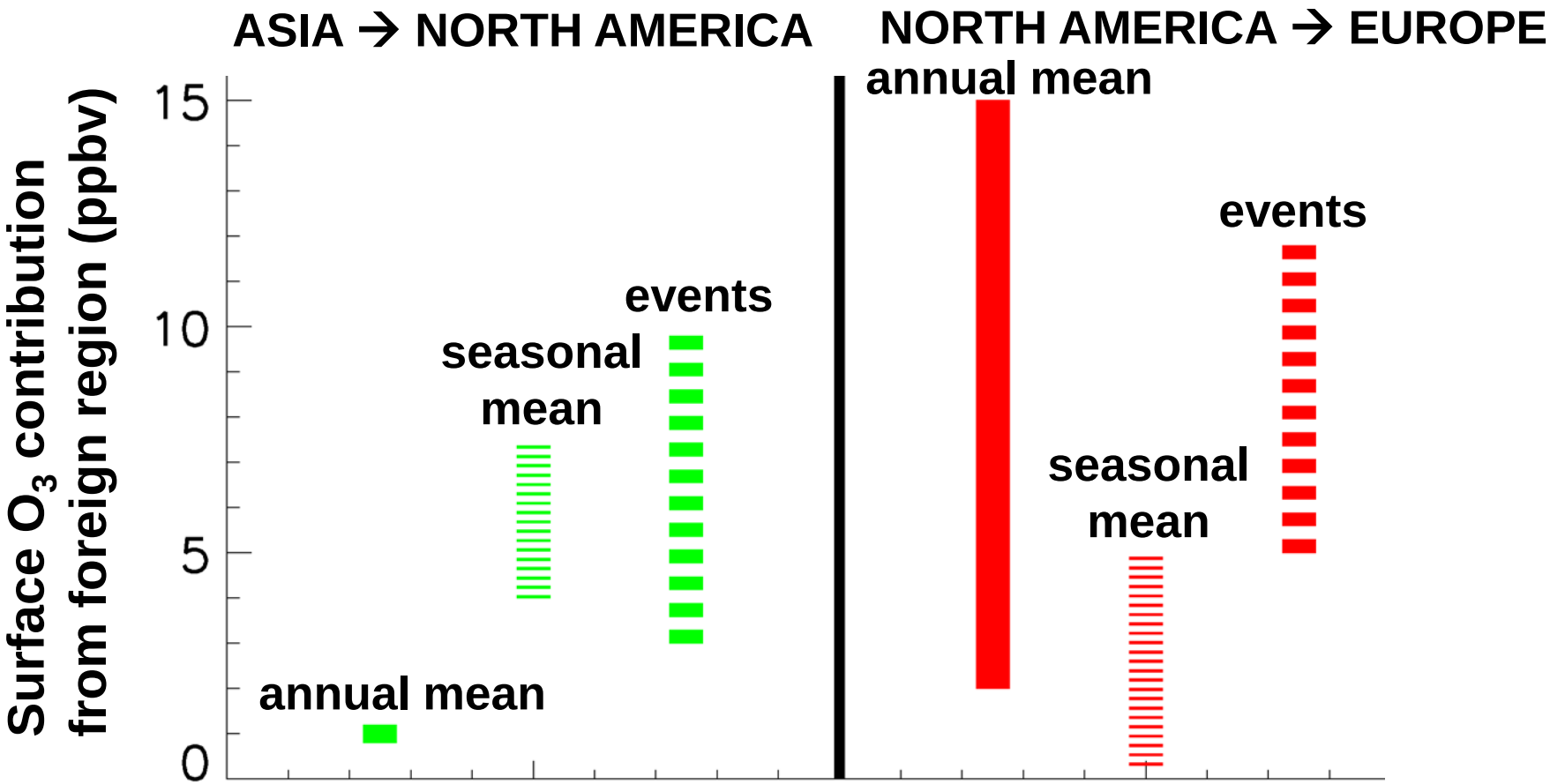
Wide range in prior estimates of intercontinental surface ozone source-receptor (S-R) relationships



Studies in TF HTAP [2007] + *Holloway et al*., 2008; *Duncan et al*., 2008; *Lin et al*., 2008

Assessment hindered by different (1) methods, (2) reported metrics, (3) meteorological years, (4) regional definitions

Few studies examined all seasons

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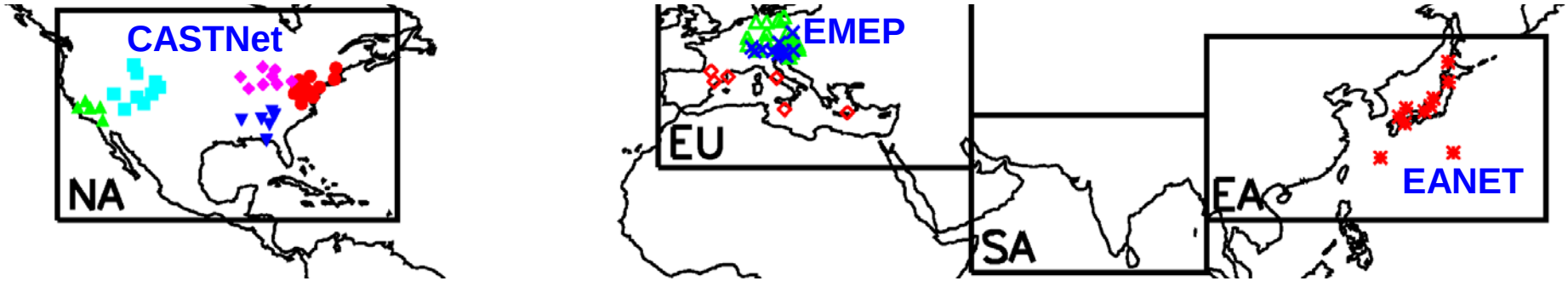
Multi-model effort to quantify & assess uncertainties in N. mid-latitude hemispheric O_3 transport (www.htap.org)

Task Force on Hemispheric Transport of Air Pollution (TF HTAP) to inform policy negotiations under CLRTAP



BASE SIMULATION (21 models):

- → horizontal resolution of 5 x5° or finer
- → 2001 meteorology
- → each group's best estimate for 2001 emissions
- → methane set to 1760 ppb

SENSITIVITY SIMULATIONS (13-18 models):

- → -20% regional anthrop. NO_x, CO, NMVOC emissions, individually + all together (=16 simulations)

TF HTAP, 2007, 2010; Sanderson et al., GRL, 2008; Shindell et al., ACP, 2008; Fiore et al., JGR, 2009, Reidmiller et al. ACP, 2009; Casper Anenberg et al., ES&T, 2009; Jonson et al., ACPD,2010

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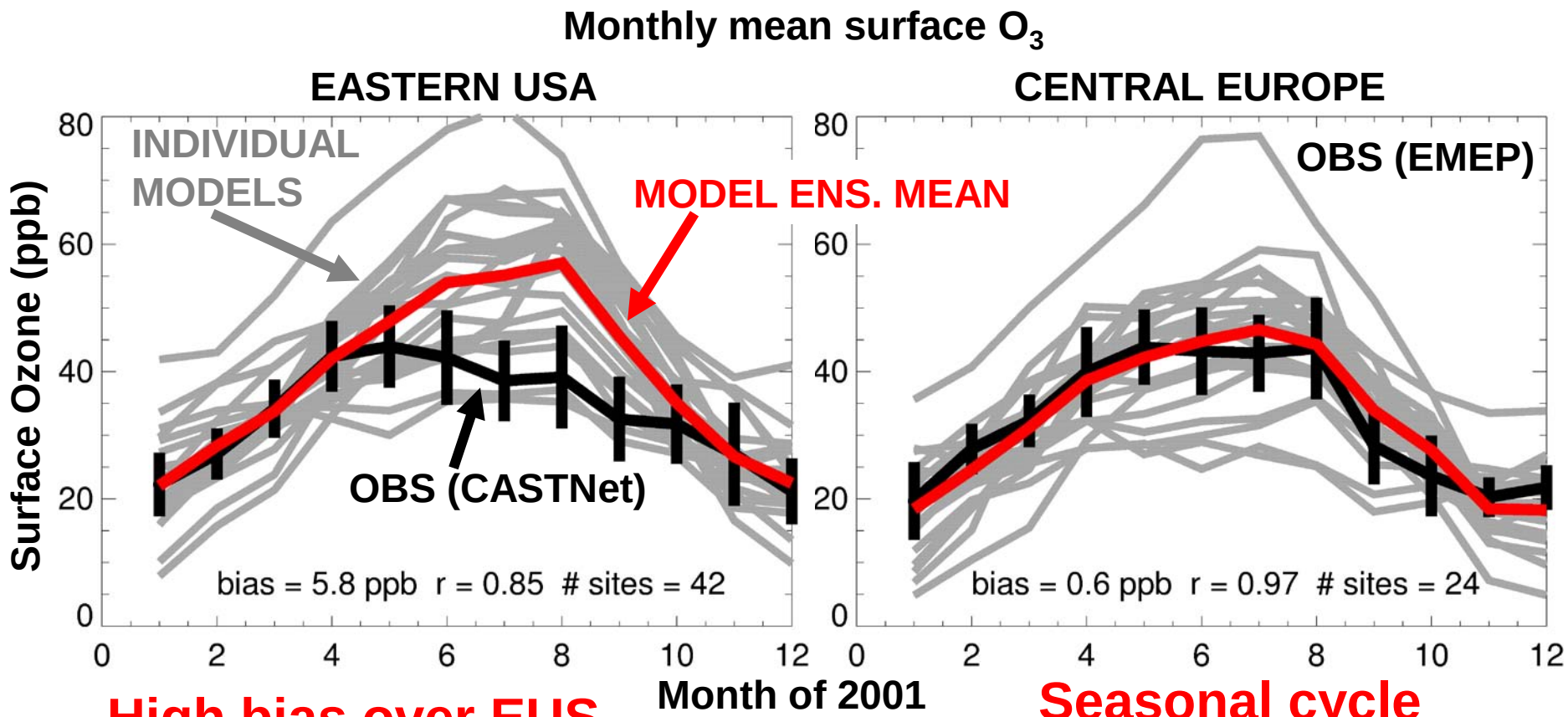
Multi-model mean captures obs in spring, when hemispheric transport at N. mid-latitudes peaks



High bias over EUS in summer and early fall

Seasonal cycle captured over Europe

What observational constraints would best test model suitability for intercontinental transport?

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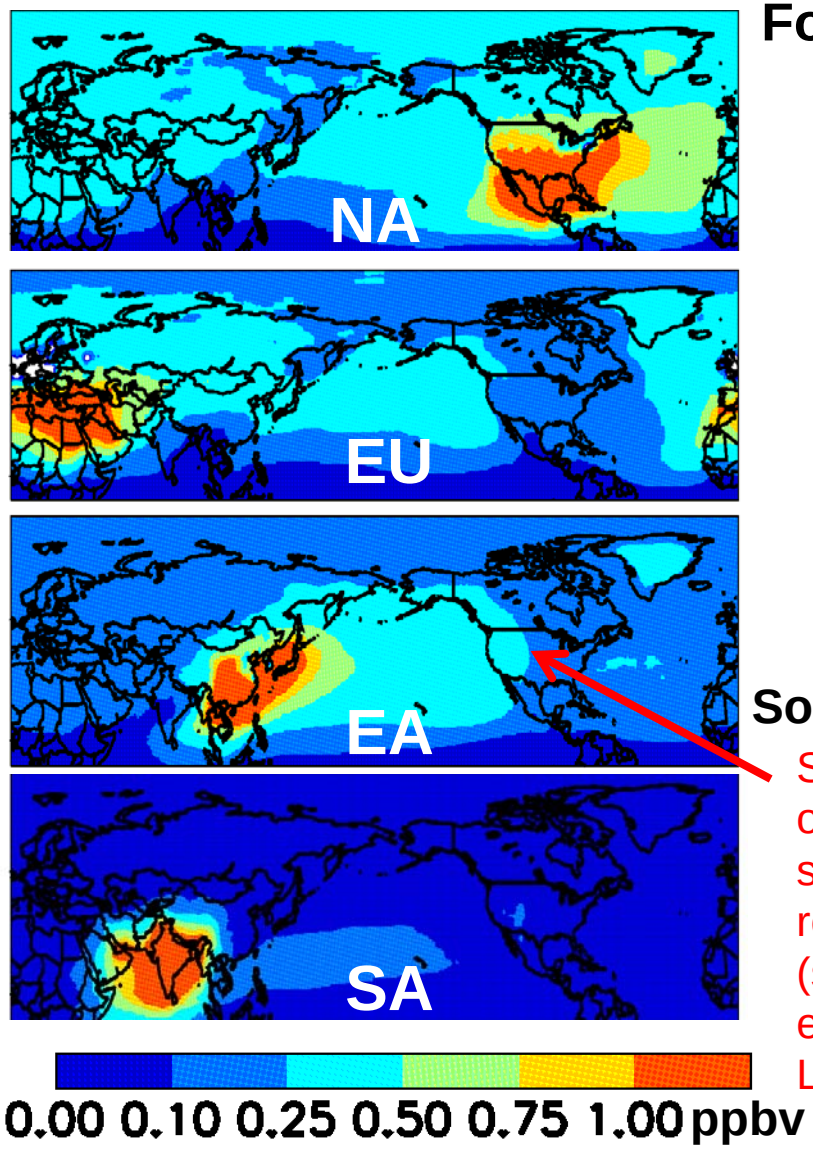
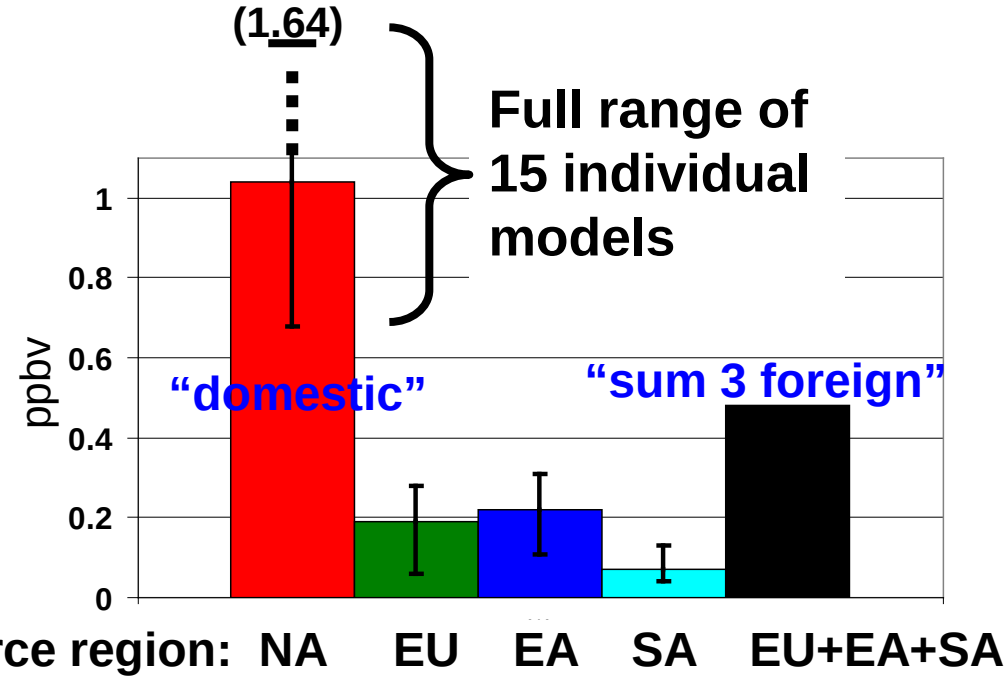
Model ensemble annual mean decrease in surface O_3 from 20% reductions of regional anthrop. O_3 precursors



Spatial variability over continental-scale receptor region (NA) (see also Reidmiller et al, 2009; Lin et al., 2010)

Foreign vs. "domestic" influence over NA:



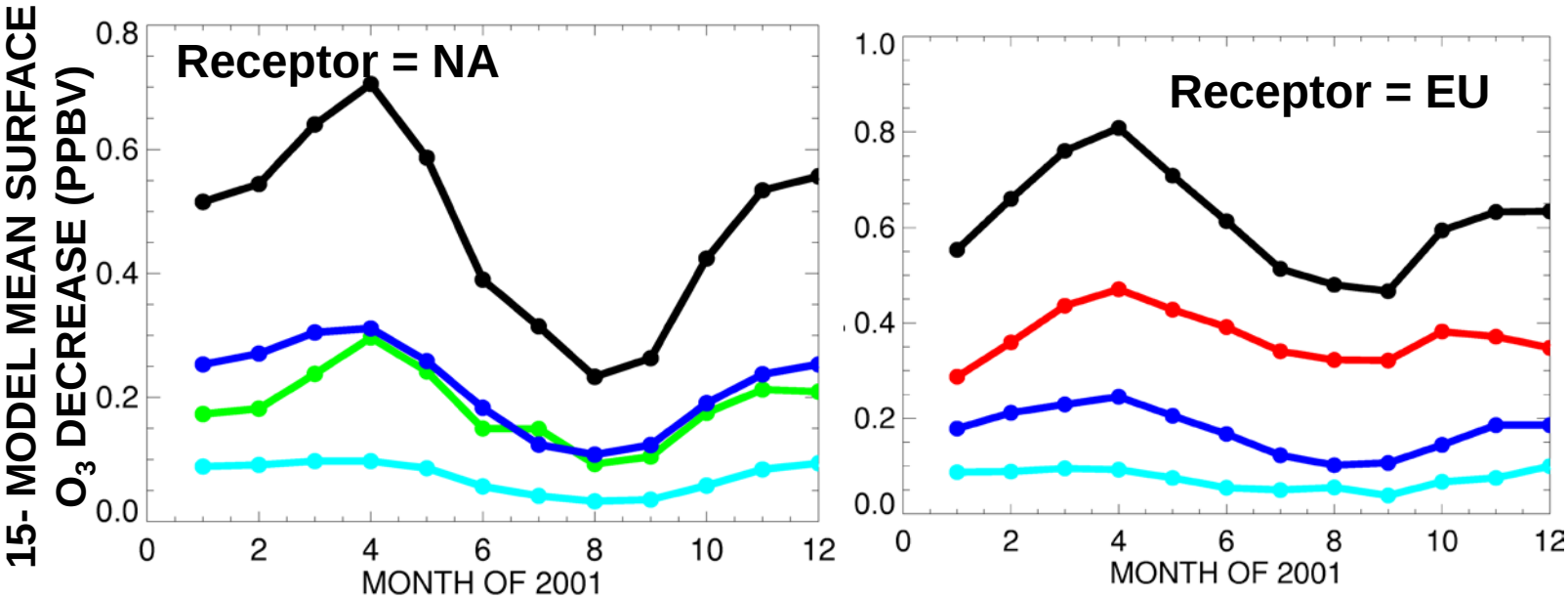
$$\frac{\sum 3\ \text{foreign}}{\text{domestic}} = 0.45$$

"import sensitivity"

Fiore et al., JGR, 2009

Seasonality of surface ozone response over North America and Europe to -20% foreign anthrop. emissions

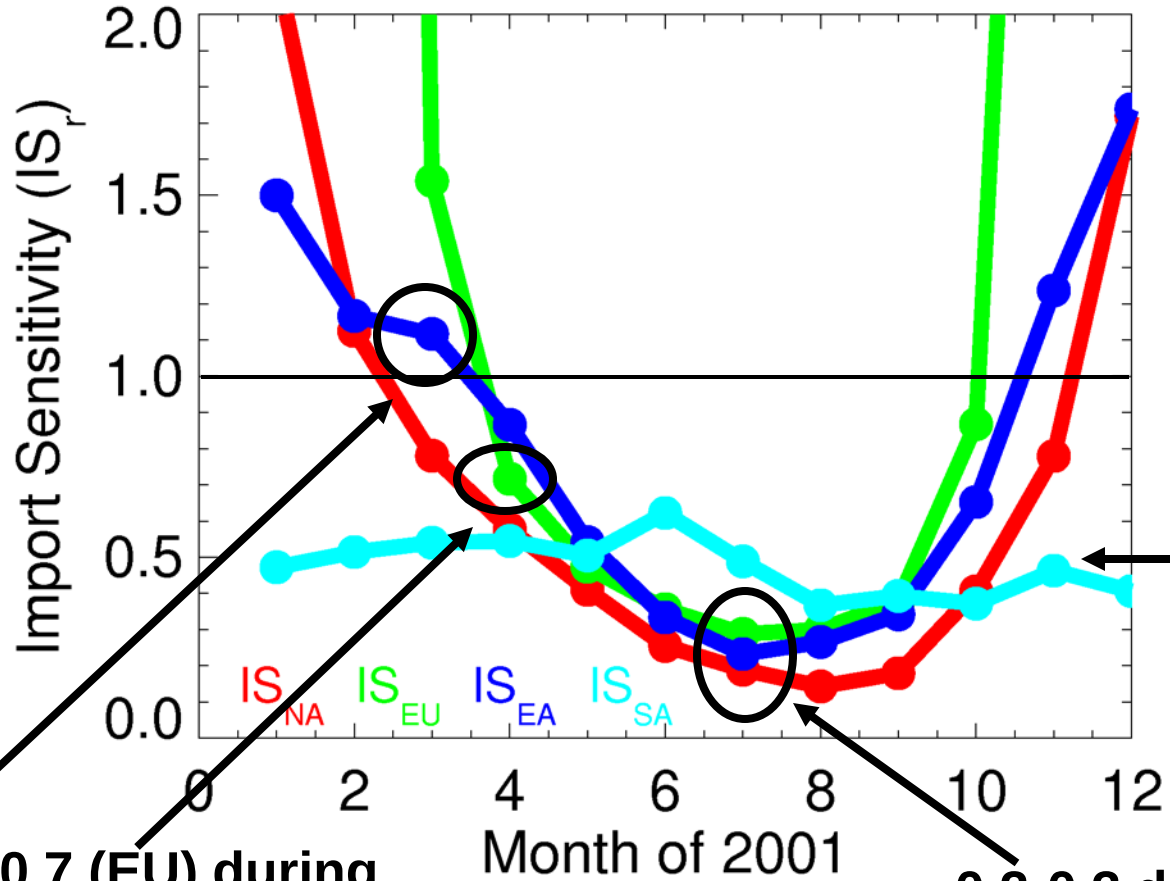
Source region: SUM3 NA EA EU SA



1. **Spring max due to longer O_3 lifetime, efficient transport [e.g., Wang et al., 1998; Wild and Akimoto, 2001; Stohl et al., 2002; TF HTAP 2007]**
2. **Response typically smallest to SA emissions (robust across models)**
3. **Similar response to EU& EA emissions over NA Apr-Nov (varies by model)**
4. **NA>EA>SA over EU (robust across models)**

Fiore et al., JGR, 2009

Monthly mean import sensitivities (surface O_3 response to foreign vs. domestic emissions)



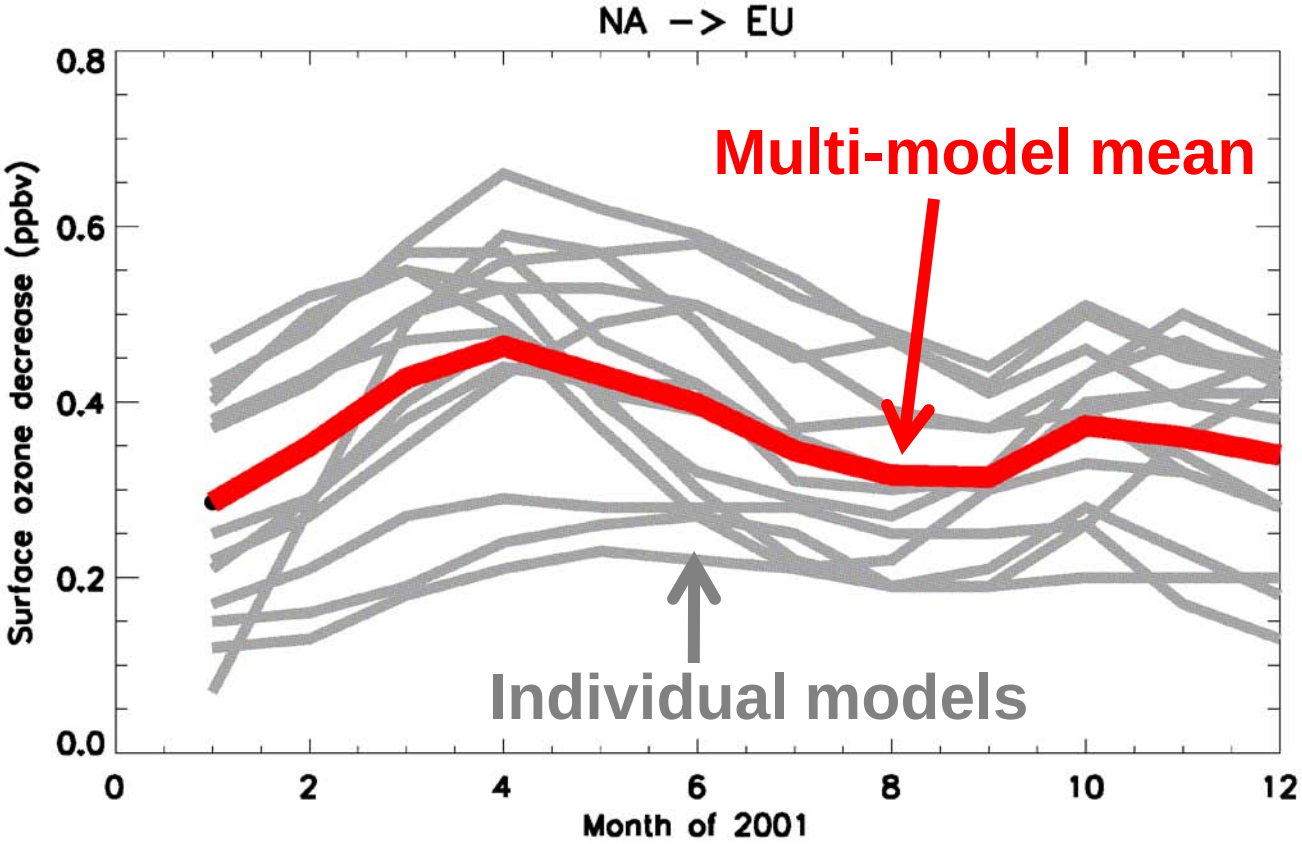
SA fairly constant ~0.5

1.1 (EA), 0.7 (EU) during month with max response to foreign emissions

0.2-0.3 during month of max response to domestic emissions

Fiore et al., JGR, 2009

UNCERTAINTIES IN ESTIMATES:
Surface O_3 response to foreign anthrop. emissions varies widely across individual models



→No obvious correlation of strength of foreign influence with individual model biases relative to O_3 observations [*Fiore et al., JGR, 2009; Reidmiller et al., ACP 2009;Jonson et al, ACPD 2010]*

→ Generally not due to model differences in anthropogenic emissions (one exception)

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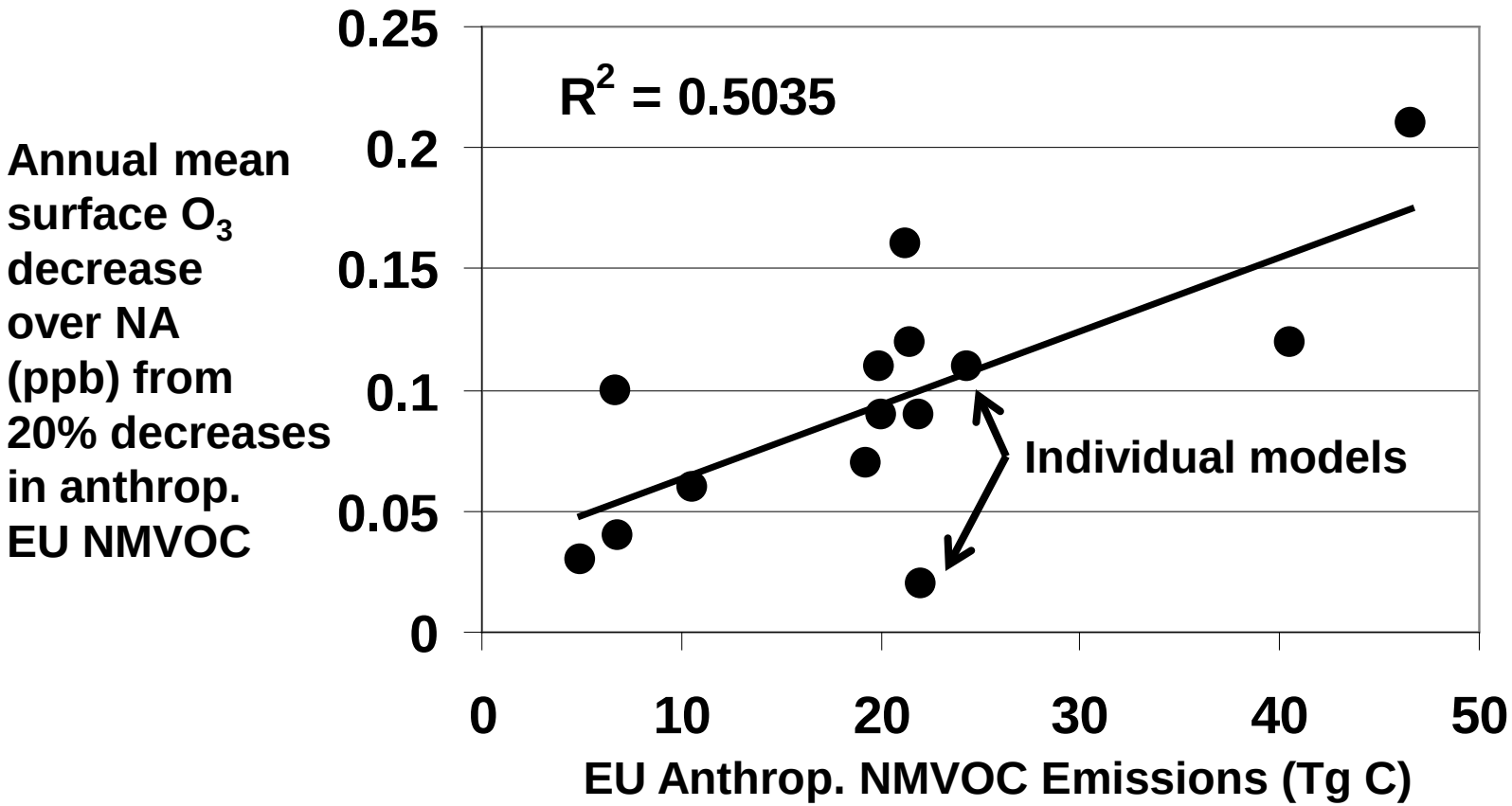
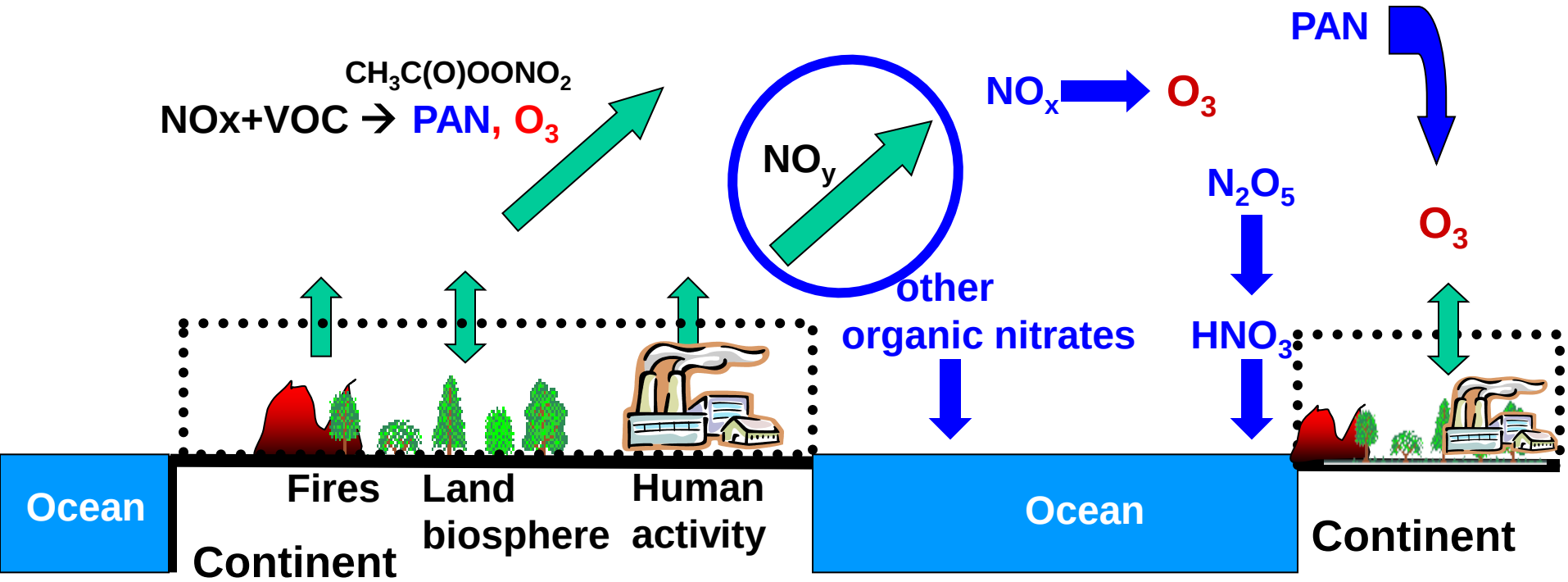
Strong sensitivity of exported EU O_3 to large spread in EU NMVOC inventories (anthrop. NO_x fairly similar across models)



Fiore et al., JGR, 2009

Models likely differ in export of O_3 + precursors, downwind chemistry (PAN [Emmerson and Evans, ACP, 2009]), and transport to receptor region



NO_y partitioning (e.g., PAN vs. HNO_3) influences O_3 formation potential far from source region

How well do the models used to estimate intercontinental O_3 transport simulate observed distributions of PAN?

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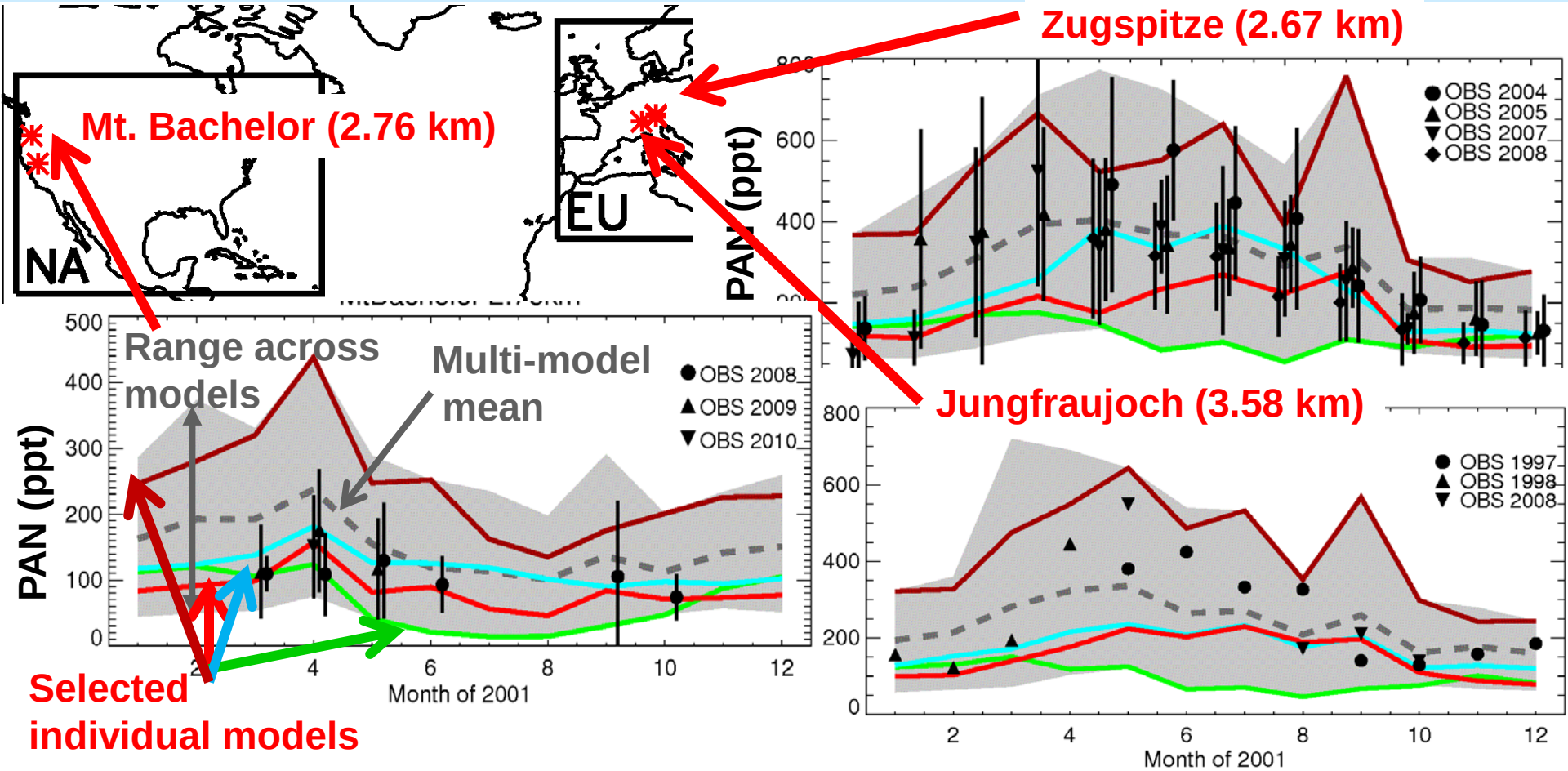
Comparison of models with PAN measurements at mountain sites in Europe and Western USA



- No measurements for year 2001; observed interannual variability precludes definitive conclusions as to which models are best
- Multi-model mean generally within range of observations
- Some models consistently high/low; others vary site to site

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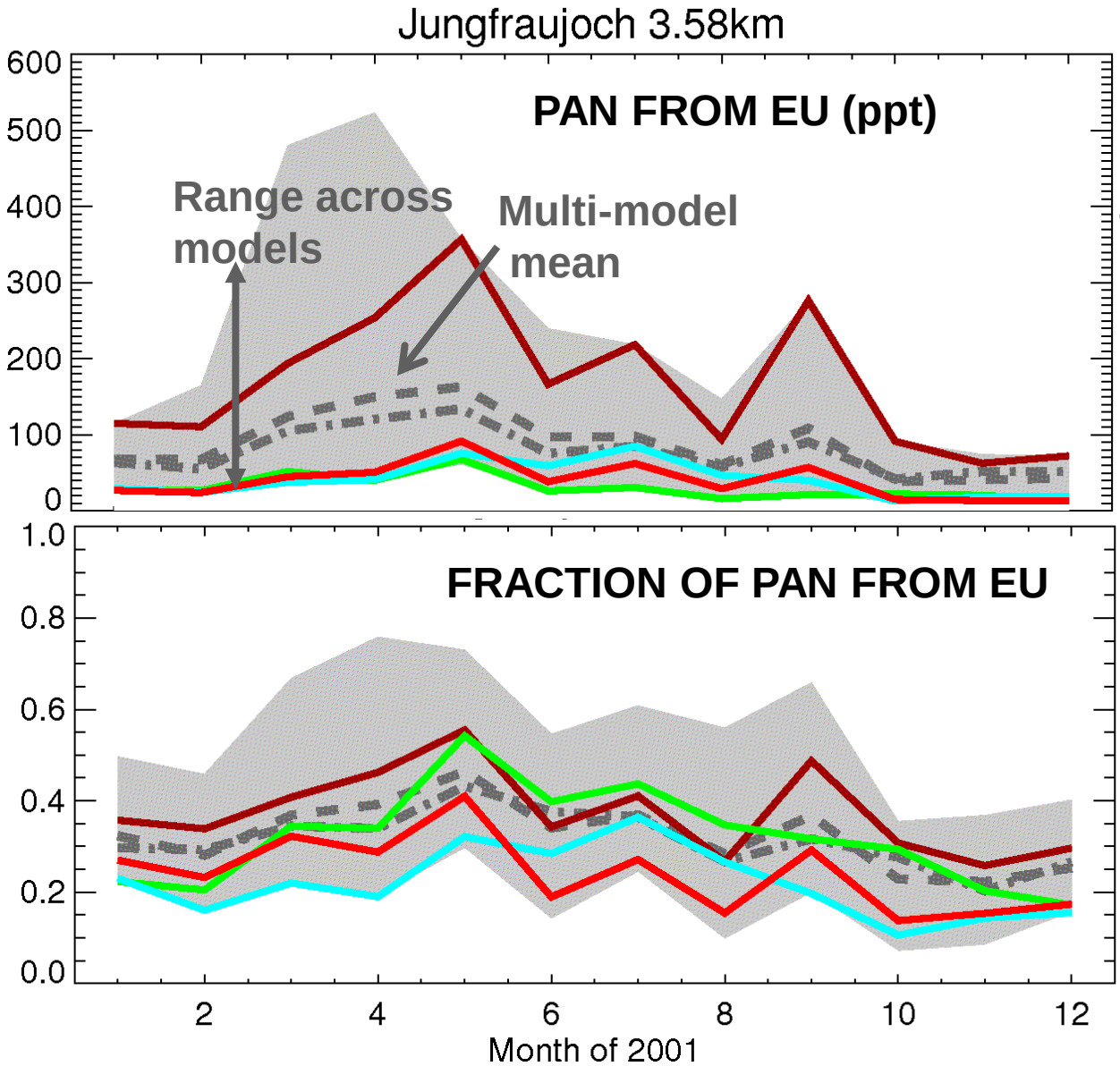
Individual models rank differently in terms of absolute vs. relative source attribution



Determined by differencing simulations with -20% EU anthrop. emissions from the base case, then x5

Relative attribution matters from a policy perspective.

Evaluating models with total observed PAN won't address relative importance.

Observational constraints?

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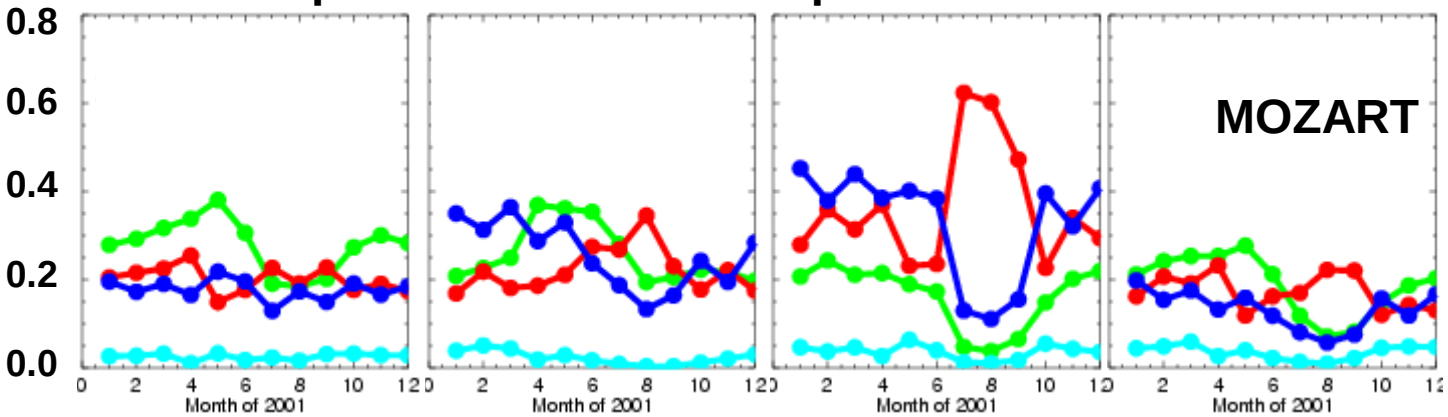
Relative influence of regional O_3 precursors on PAN at Mount Bachelor (OR), as estimated by the models

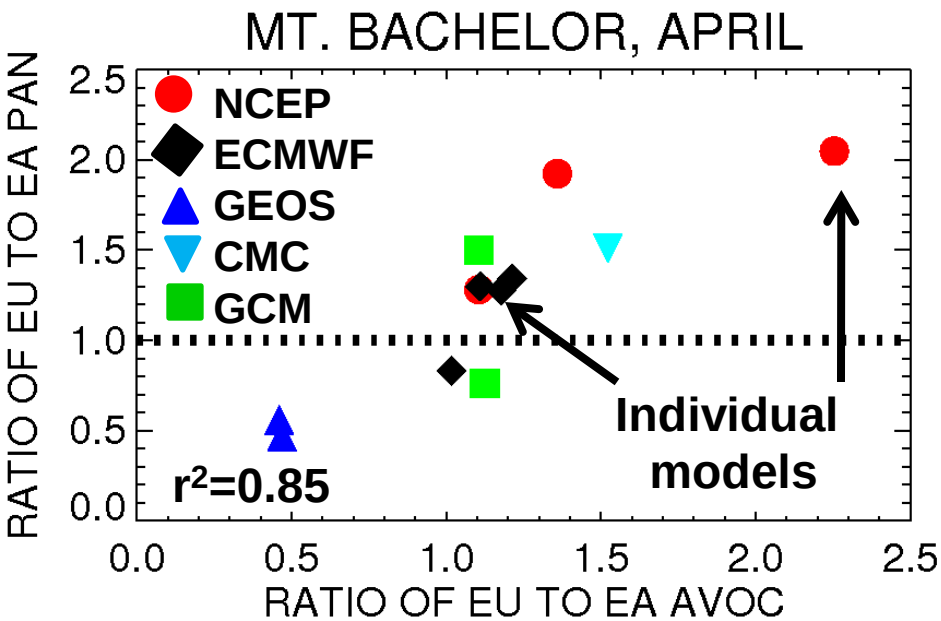
Fraction of total PAN from source region
EU NA EA SA

4 example HTAP models sampled at Mount Bachelor



More EU influence

More EA influence



EU/EA PAN influence correlates with EA/EU AVOC emissions;

…Or meteorology?

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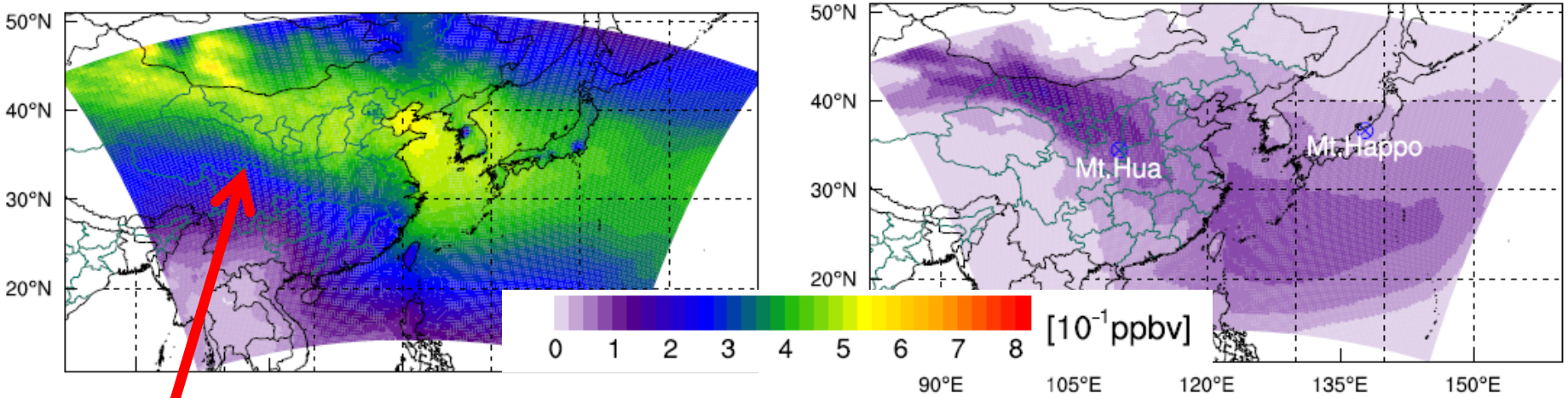
PAN as a pathway for intercontinental O_3 transport: How important?

MARCH 2001 SURFACE O_3 DECREASE (from Base simulation) in CMAQ regional model with boundary conditions (BCs) from the global MOZART-2 model:

BCs for all species reflect -20% EU anthropogenic emissions

Only PAN BCs reflect -20% EU anthropogenic emissions



Large spatial gradients within continental-scale "HTAP" regions

PAN decomposition contributes ~20% of spatial average total O_3 response; up to ~50% in subsiding plumes at mountain sites [*Lin et al*., ACP, 2010]

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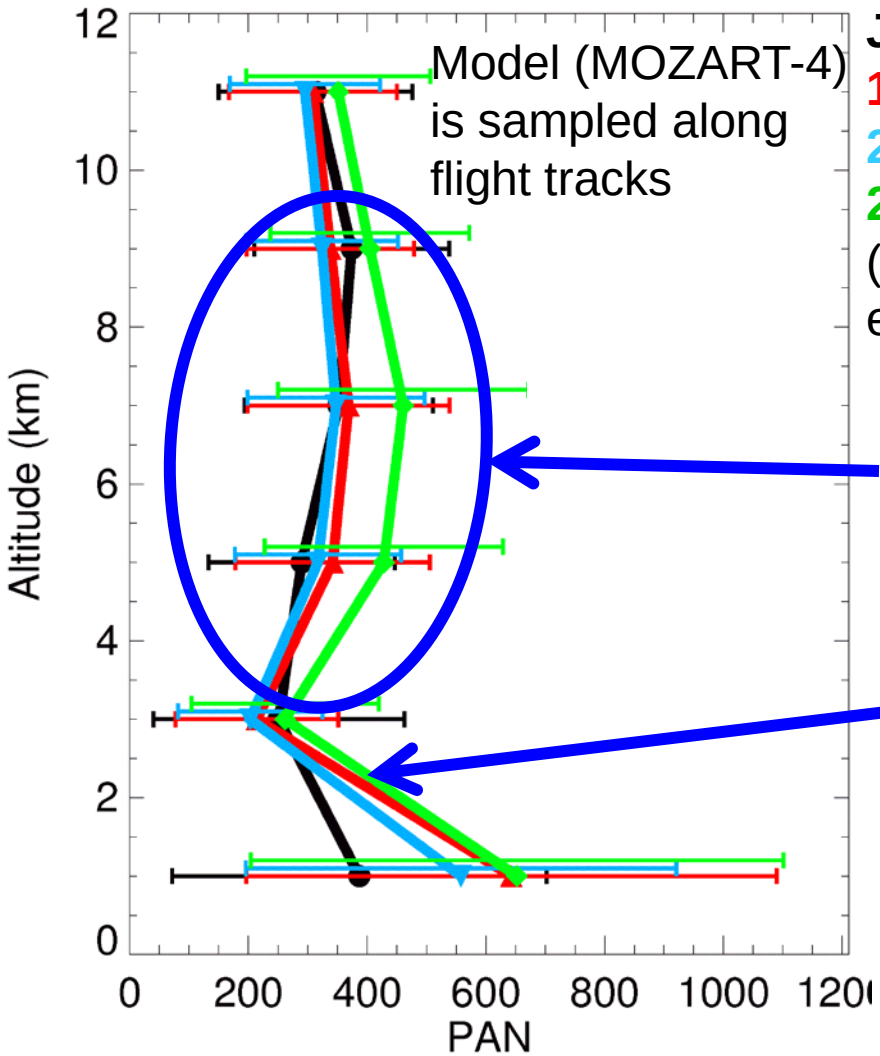
PAN as a proxy: Contributions to PAN from anthropogenic versus lightning NO_x sources [*Fang et al.*, JGR, in press]

PAN over the eastern U.S. during summer 2004 (INTEX-NA field campaign)



July MOZART-4 simulations with:

1999 U.S. Anthro NO_x; "low" lightning

2004 U.S. Anthro NO_x (-23%); "low" lightning

2004 U.S. Anthro NO_x; "high" lightning (10x)

(anthro and lightning NO_x perturbations are equivalent, in each case changed by 0.16 Tg N)

Cross-model differences in lightning NOx may contribute to differences in PAN in The middle and upper troposphere;

Little lightning influence in lower troposphere where PAN is more sensitive to anthropogenic NO_x changes

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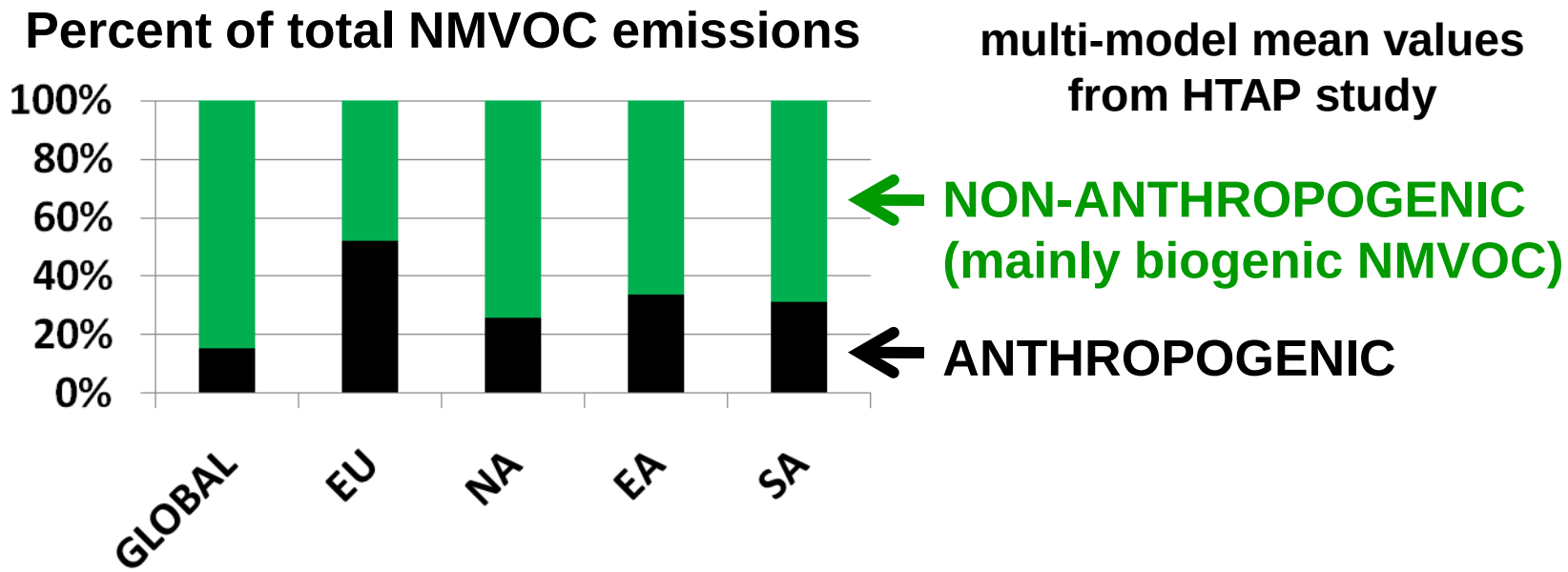
What is the role of BVOC in hemispheric transport of ozone and PAN?



"the treatment of isoprene in global models can only be seen as a first order estimate at present, and points towards specific processes in need of focused future work" [*von Kuhlmann et al.*, 2004]

→ Rapidly evolving knowledge from lab and field studies

[e.g., Lelieveld et al., 2008; Hofzumahaus et al., 2009; Paulot et al., 2009ab; Perring et al., 2009; Brown et al., 2009; and many, many others!]

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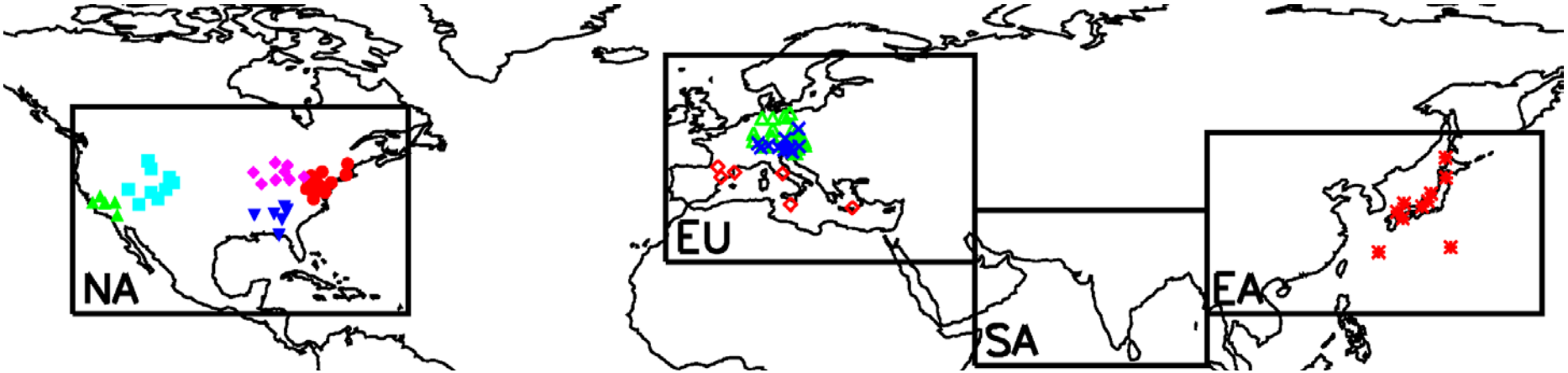
Explore role of isoprene vs. anthropogenic emission changes in North America with MOZART-2 global model



BASE SIMULATION:

- → horizontal resolution of 1.9°x1.9°
- → 2001 meteorology
- → best estimate for 2001 emissions
- → methane set to 1760 ppb

SENSITIVITY SIMULATIONS:

- → -20% regional anthrop. NO_x, CO, NMVOC emissions, individually + all together (FOCUS ON NA)
- → +20% NA isoprene emissions

Interannual variation in NA isoprene emissions ~20-30% [*Palmer et al*. 2006]

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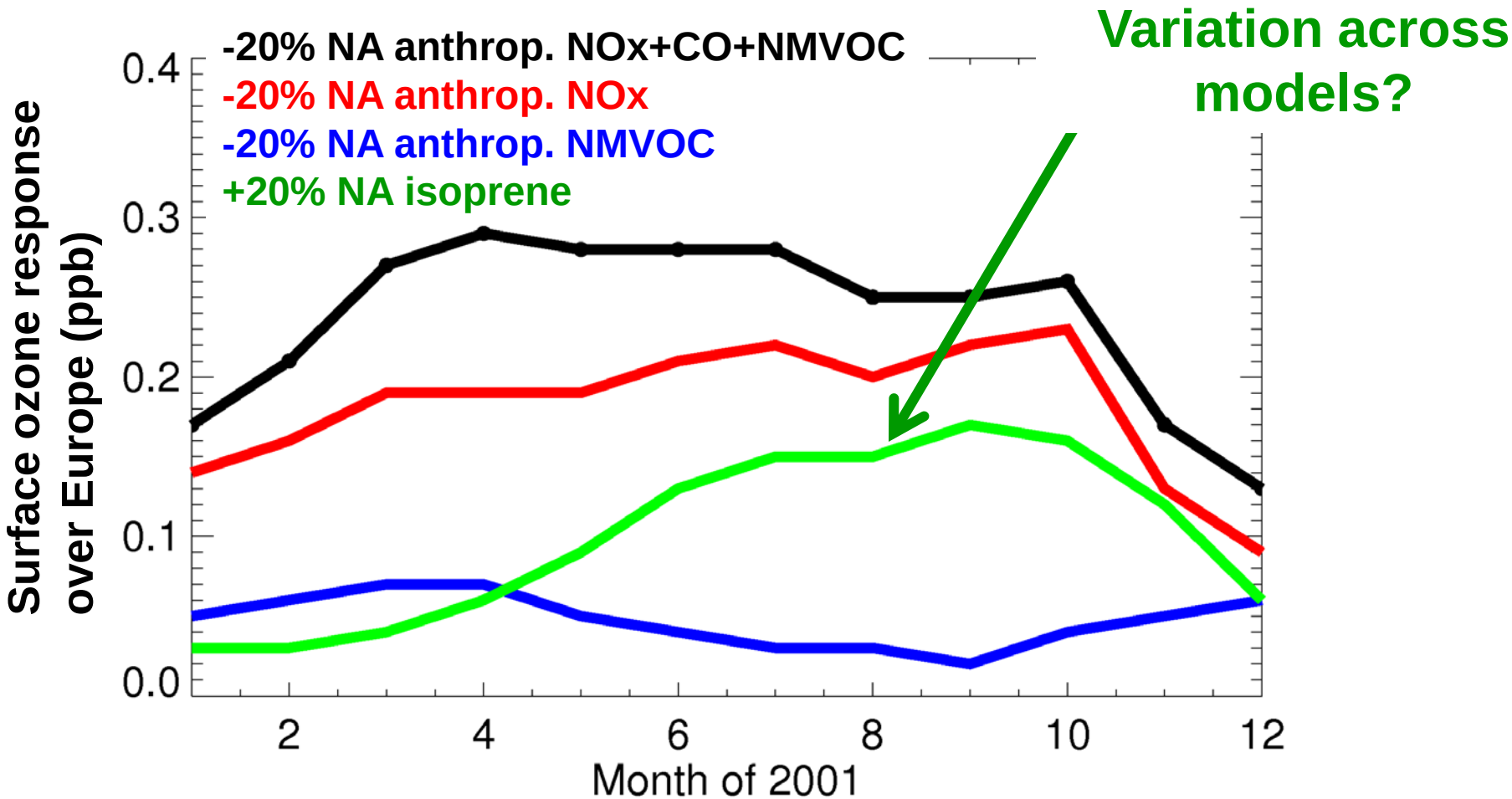
Surface O_3 response over Europe (spatial average) to North American emission perturbations



O_3 response over EU to NA isoprene comparable to NA anth. NO_x in summer/fall; larger than NA anth. NMVOC

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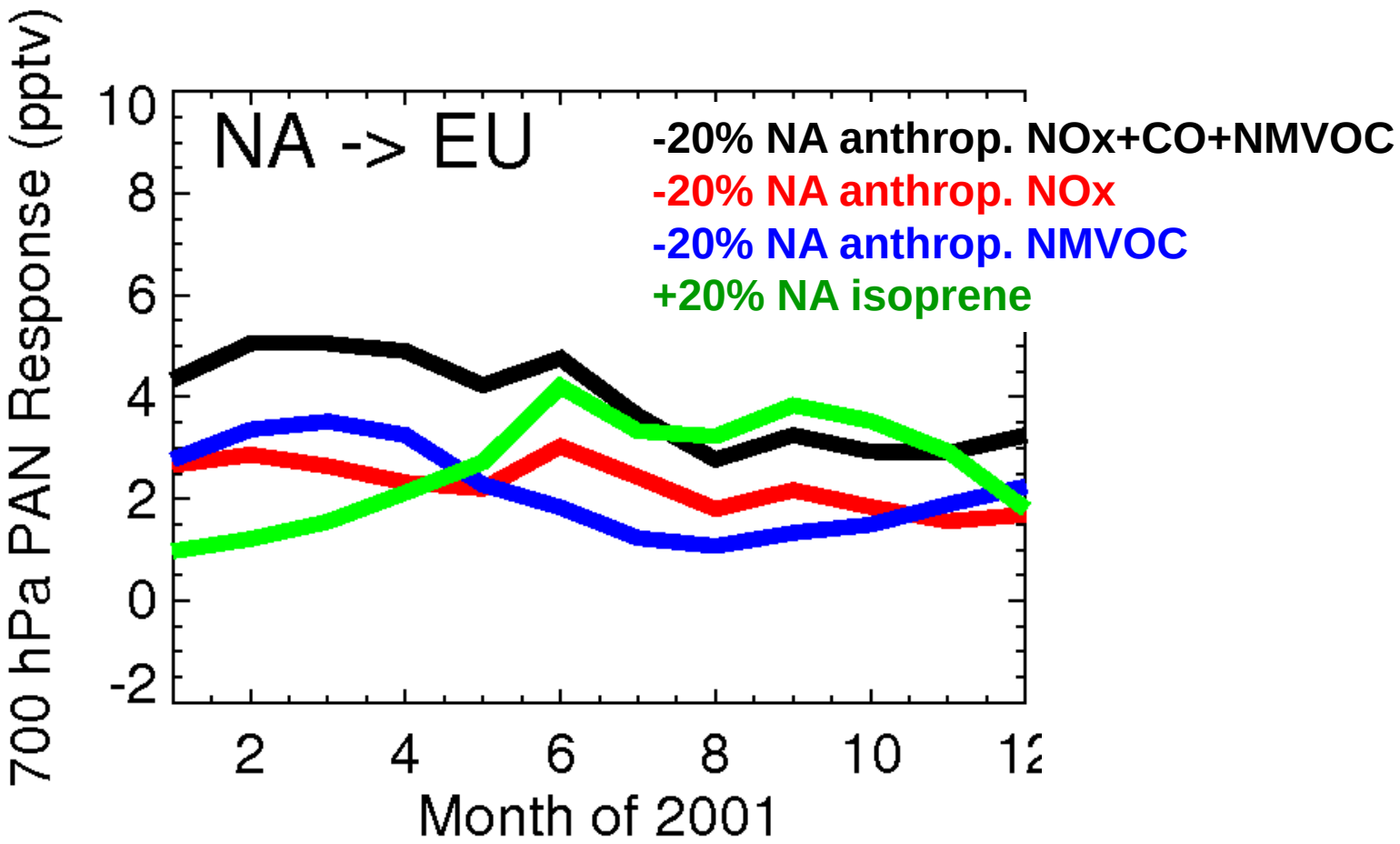
PAN (700 hPa) response over Europe (spatial average) to North American emission perturbations



PAN response over EU to NA isoprene comparable to that to all NA anthrop. emissions in summer and fall

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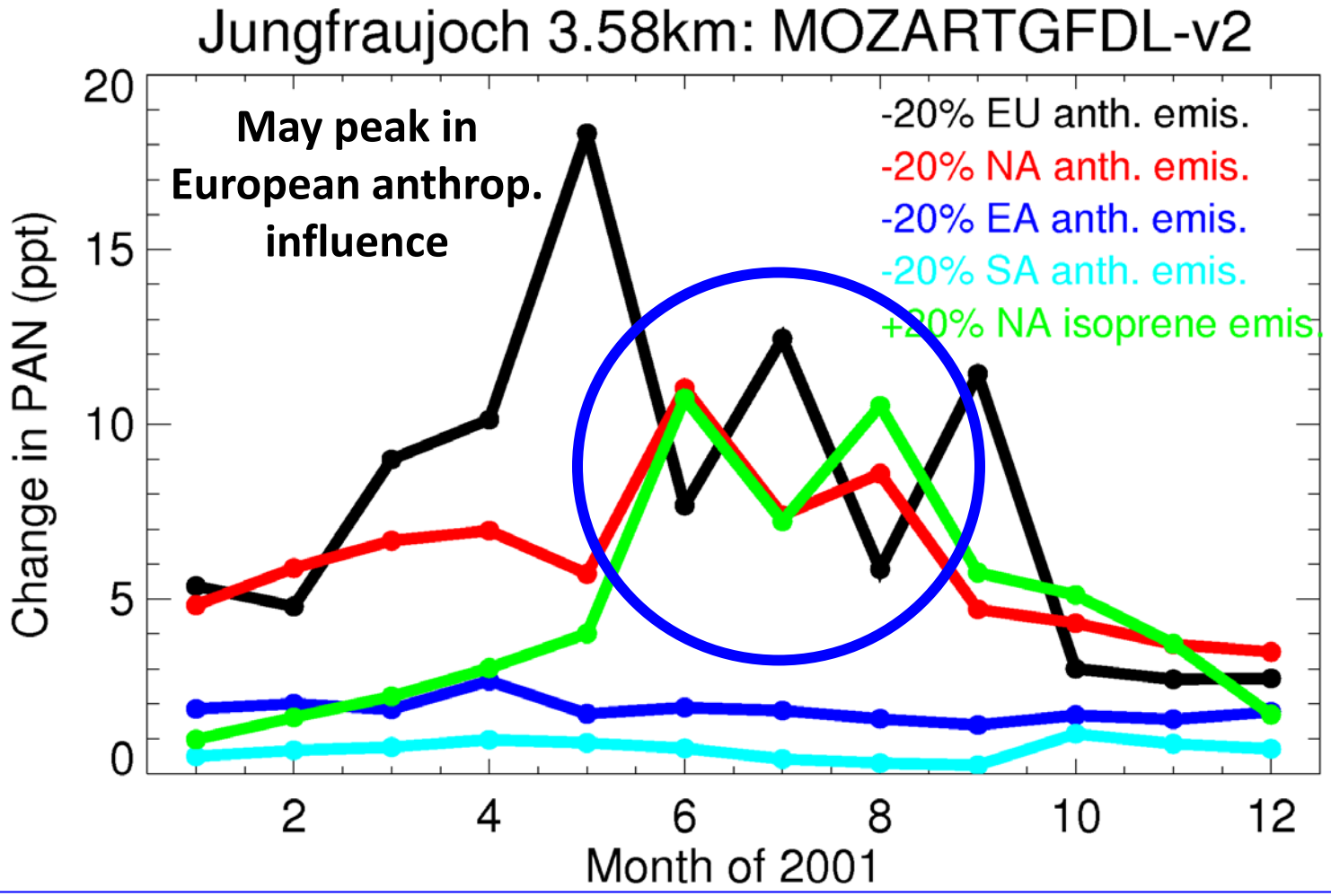
PAN response to regional precursor emission changes at a European mountain site (model is sampled at the site)



NA isoprene influence on PAN at European mountain site similar to that from NA or EU anth. emissions in summer

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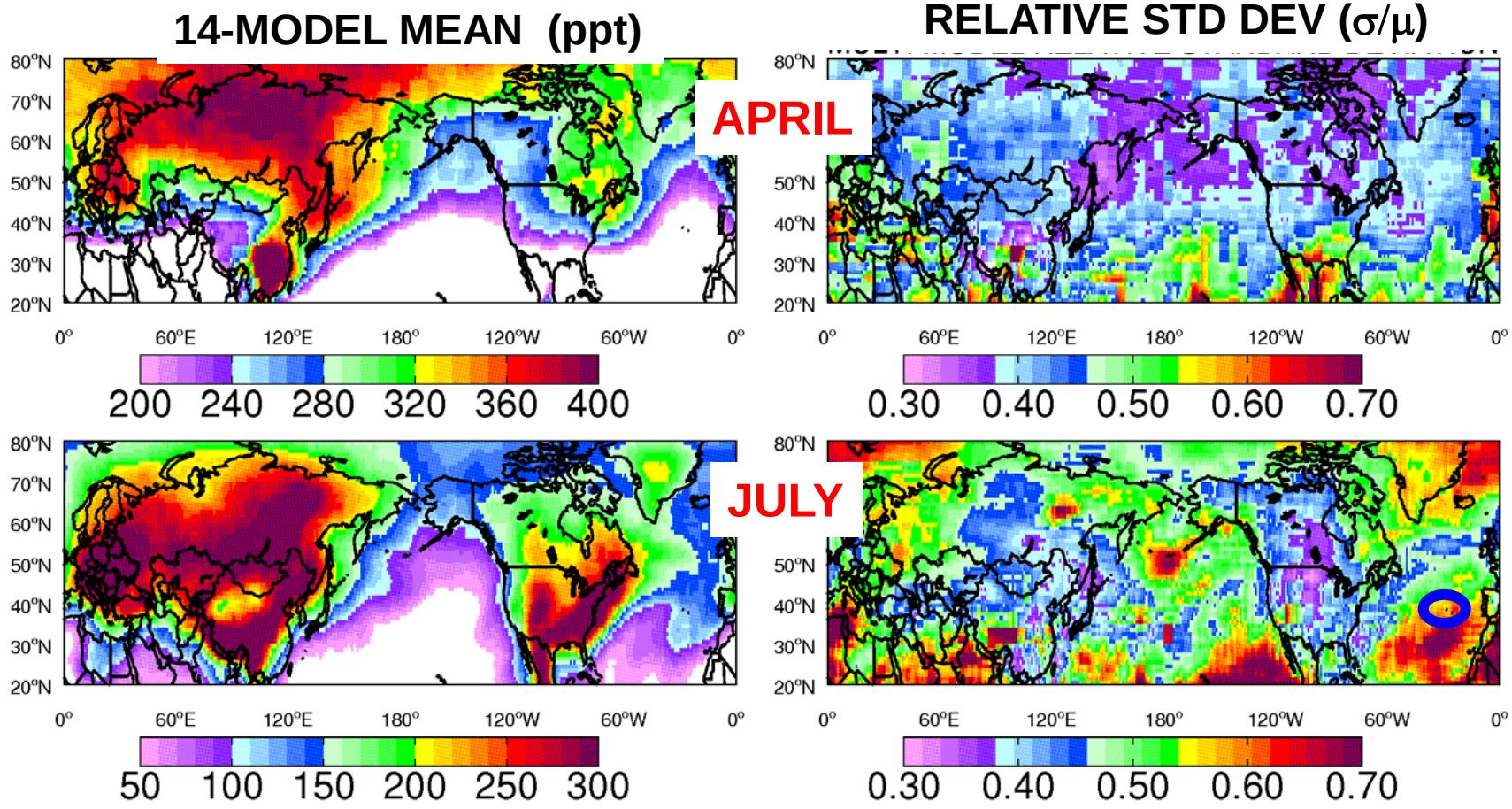
Returning to multi-model ensemble… where do models show largest relative discrepancies in PAN?

PAN AT 650 hPa



Observations at PICO-NARE [*e.g., Honrath et al., JGR*, 2004] may help in identifying "most useful" models for NA → EU intercontinental transport

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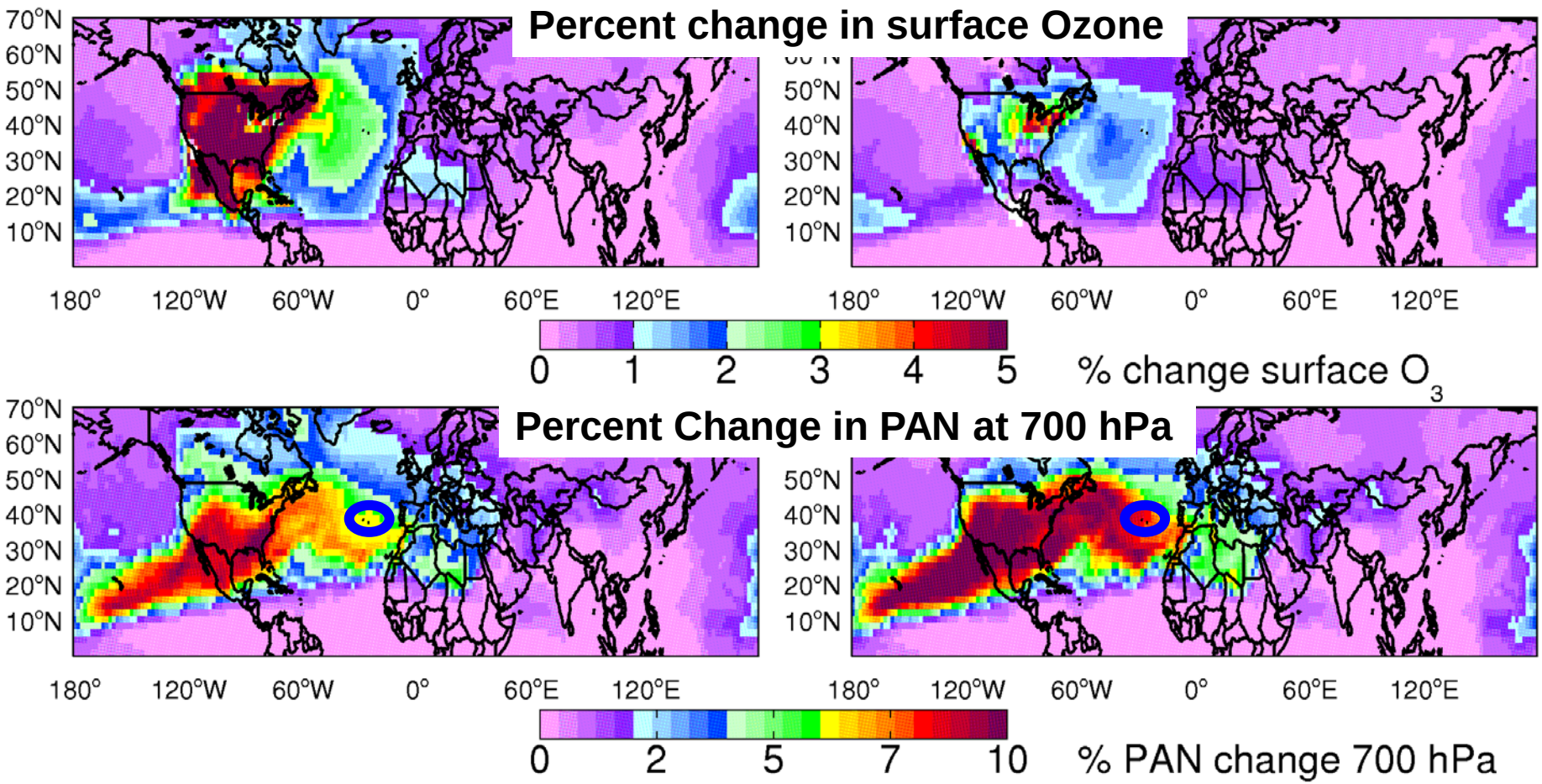
PAN is relatively more sensitive than ozone to anthropogenic and isoprene emission changes

Sensitivity simulations in the global MOZART-2 model:

-20% NA anthropogenic emissions **-20% NA isoprene emissions**



PICO-NARE site in the Azores well-positioned for sampling N. American outflow
[*e.g., Honrath et al., JGR*, 2004; *val Martin et al., JGR*, 2008.]

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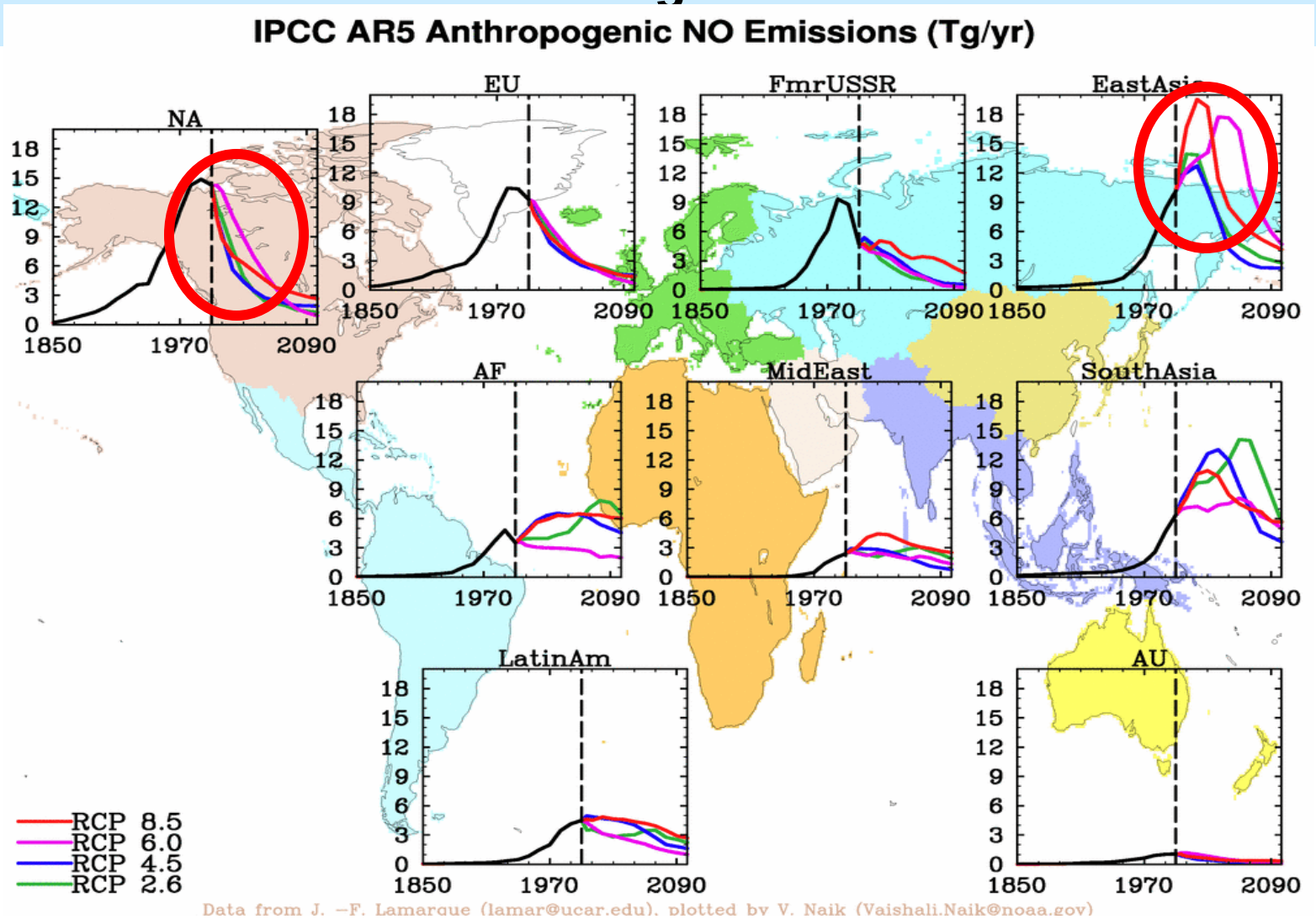
How will global emissions (and intercontinental O_3 transport) change in the future?



Implications for U.S. background O_3 and attainment of standards?

c/o Vaishali Naik, GFDL

What about climate change?

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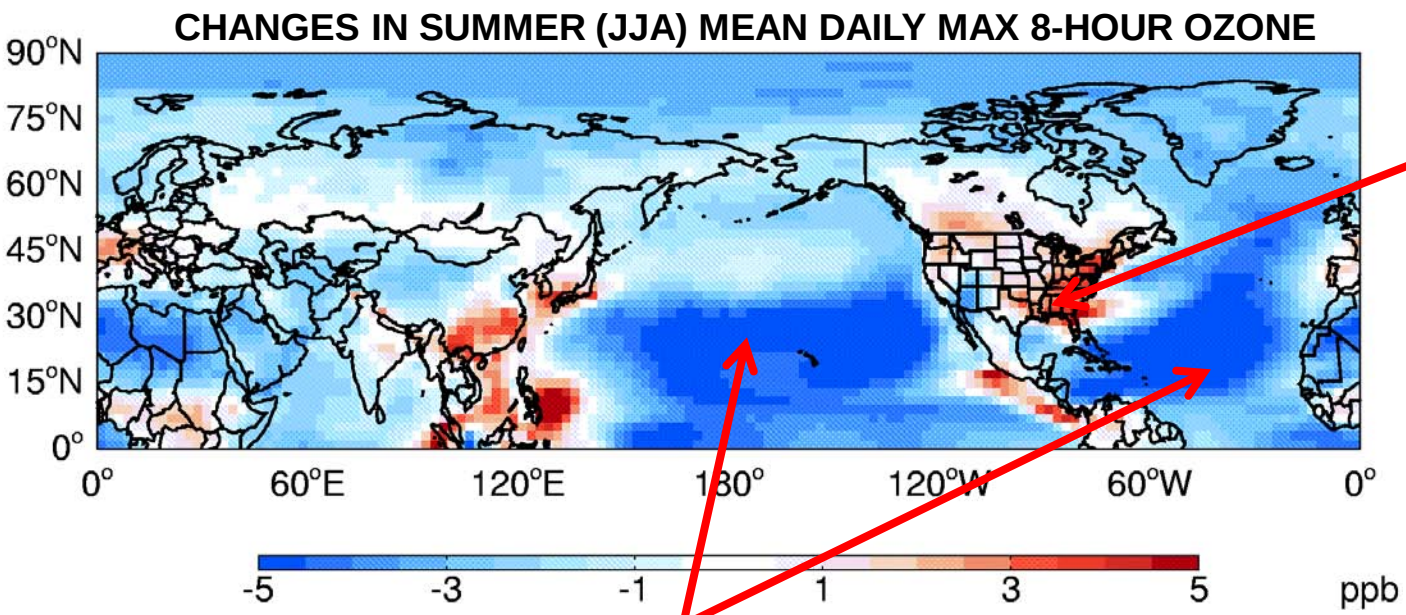
Summertime surface O_3 changes in a warmer climate in the new GFDL chemistry-climate model (AM3)

20-year simulations with annually-invariant emissions of ozone and aerosol precursors

Present Day Simulation ("1990s"): observed SSTs + sea ice (1981-2000 mean)

Future Simulation ("A1B 2090s"): observed SSTs + sea ice + average 2081-2100 changes from 19 IPCC AR-4 models



Previously noted degradation of summertime EUS O_3 air quality ***e.g., reviews of Jacob and Winner, Atmos. Environ. 2009 and Weaver et al., BAMS, 2009***

Previously noted decrease of lower troposphere background O_3
e.g., Johnson et al., GRL, 2001; Stevenson et al., JGR, 2006

Climate change expected to reduce intercontinental influence, though net effect determined by climate plus emission changes (human + natural)

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Anthropogenic and biogenic influence on intercontinental surface O_3: Concluding thoughts and some questions raised

- **Benchmark multi-model effort: wide cross-model and spatiotemporal variability in intercontinental influence**

What is the role of mesoscale (i.e., sub-grid) processes in mixing air between the free troposphere and the surface?

- **PAN: Pathway and Proxy for intercontinental O_3 pollution**

Exploit differences across model ensembles to help guide observational sampling strategies?

Potential for constraints on O_3 source attribution via PAN?

- **NA isoprene influence on EU surface O_3 comparable to NA anthrop. NO_x in summer and fall**

Pay more attention to role of biogenic sources when assessing impacts of human activities on climate and air quality?

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