Global Sources of North American Ozone

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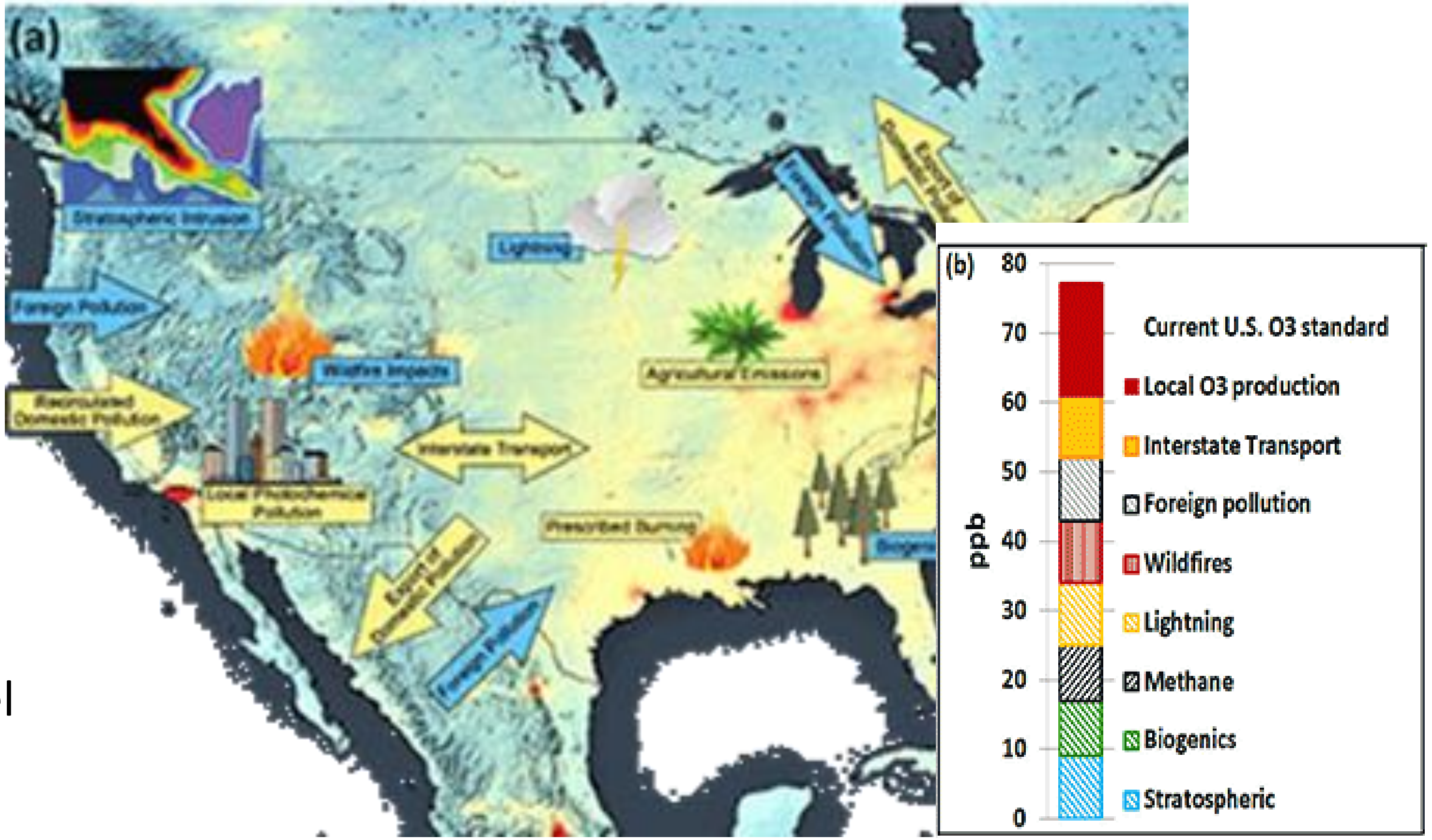
Part I - Outline

- Quick Overview of “Background”
- Part II – Model System Description and Evaluation
- Part III – Model attribution results

What are background concentrations?

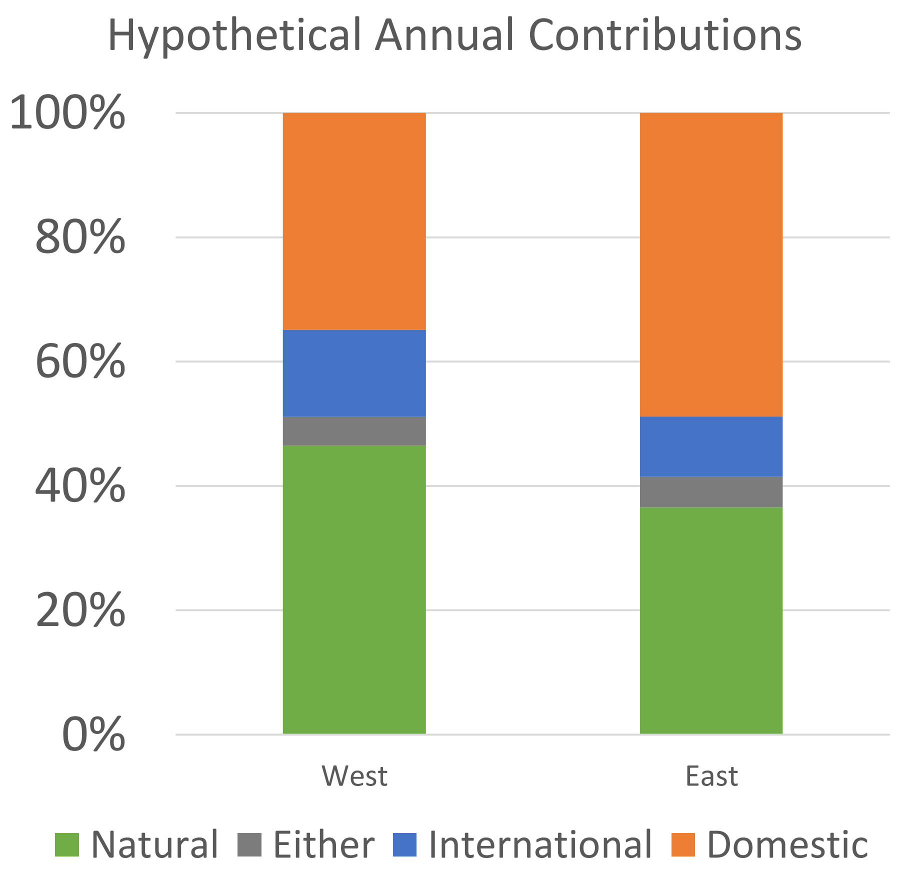
doi: 10.1525/elementa.309

- Jaffe et al. (2018) uses a source oriented definition
  - Non-Controllable *Ozone* Sources contribute to background ozone.
  - What is controllable, to some extent, depends on context.
- “Non-Controllable” Ozone Sources
  - Stratosphere
  - Lightning NOx
  - Wildfires, Biogenics
  - Seasonal uncertainty ±10 ppb
- “Controllable”
  - Depends on Context…
  - Non-Attainment Area
  - State, Country
  - International?
- ***Ambient air has all sources***
  - NCOS can be important
  - NCOS vary from year to year
  - NCOS vary from model to model



Zero-out estimates of ozone contributions

- *Motivations:*
  - Interannual variability (e.g., Lin et al., 2017)
  - Modeling system (e.g., Huang et al. 2017)
  - 2016 platform (α)
- *New Estimates:*
  - *Northern Hemispheric:* **Natural**
  - International anthropogenic: **Intl**
  - Domestic anthropogenic: **USA**
  - Nonlinear: **Residual**
    - Requires either
    - Requires both
- All starts with a modeling platform



Part I - Outline

System Description

- Global model versions and options
- Emissions
  - Natural
  - Anthropogenic

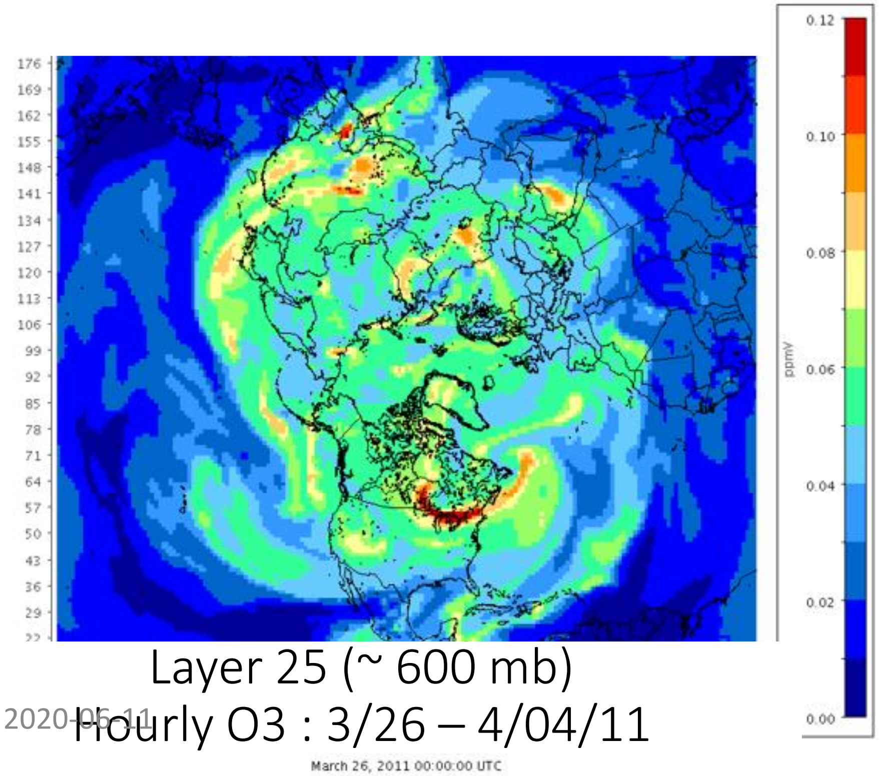
CMAQ Results and Evaluation

- Seasonal Average Ozone
- Sonde Evaluation
- CASTNet Evaluation
- Tropospheric Ozone Assessment Report Databases
- Satellite semi-quantitative

I won’t show results from GEOS-Chem results, but I will occasionally reference the performance from GEOS-Chem in the 2011 EPA modeling platform and preliminary 2016 GEOS-Chem.

Hemispheric CMAQ

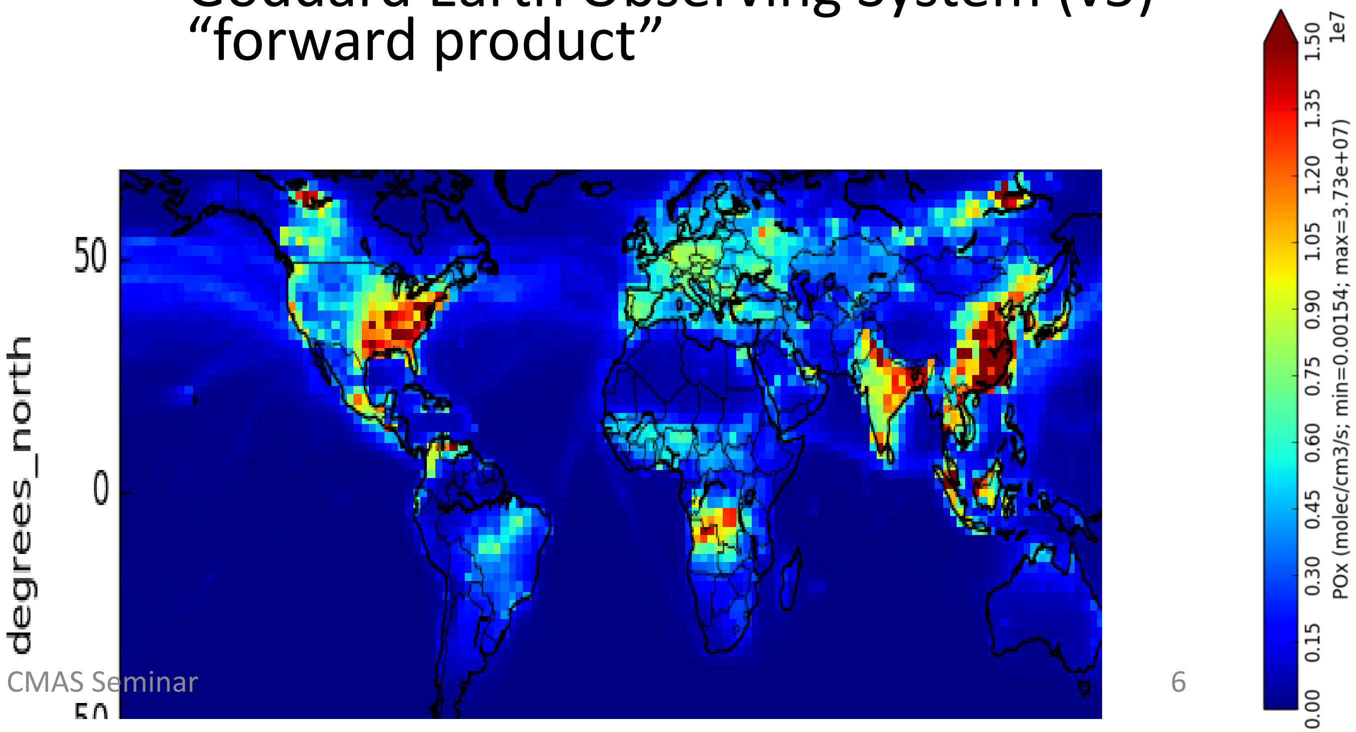
- v5.2.1 (IPV, dust, halogens)
- 8 month spinup period
- Polar stereographic (~1x1 deg)
- 44 Layers up to 50mb
- Weather Research and Forecasting



Layer 25 (~ 600 mb)
Hourly O3 : 3/26 – 4/04/11

GEOS-Chem

- Version 12.0.1
- 1-year spinup period
- 2x2.5 degree w/ half polar cells
- 72 vertical layers up to 0.01mb
  - ~38 up to 50mb
- Goddard Earth Observing System (v5) “forward product”



Natural Emissions

- Biogenics (plants and soils):
  - **BOTH:** Model of Emissions of Gases and Aerosols from Nature (MEGAN) v2.1
  - **H-CMAQ** North America Biogenic Emission Inventory System (BEIS)
- Wild and Prescribed Fires:
  - **GEOS-Chem**: 2011: GFED or 2-16: FINN v1.6
  - **H-CMAQ**: FINN v1.5 and over US 2016 platform
- Lightning:
  - **GEOS-Chem** with Lee Murray updates
  - **H-CMAQ** GEIA climatological averages by latitude & season
- Inline Dust:
  - **GEOS-Chem**: DEAD w/ current parameters
  - **HCMAQ**: Inline CMAQ algorithm
- Sea Salt: similar in-line schemes
- Dimethyl Sulfide
  - **GEOS-Chem** in-line
  - **H-CMAQ** not in present run
  - Relevant for aerosols and haze

Details in EPA 2019: epa.gov/sites/production/files/2019-12/documents/2016fe_hemispheric_tsd.pdf and Vukovich et al. CMAS 2018

Anthropogenic Emissions

Global

- EDGAR-HTAP base year 2010
  - ***BOTH interpolated to 2014 by CEDS sector/country scalars***
  - GEOS-Chem uses RETRO VOC
  - HCMAQ uses Pouliot sector-based speciation
- ***Shipping***:
  - HCMAQ: EDGAR-HTAP and 2016fe platform within Continental US modeling domain
  - GEOS-Chem: ARCTAS SO2, ICOADS CO, and over Europe from EMEP
- Aircraft:
  - HCMAQ: EDGAR-HTAP
  - GEOS-Chem: AEIC

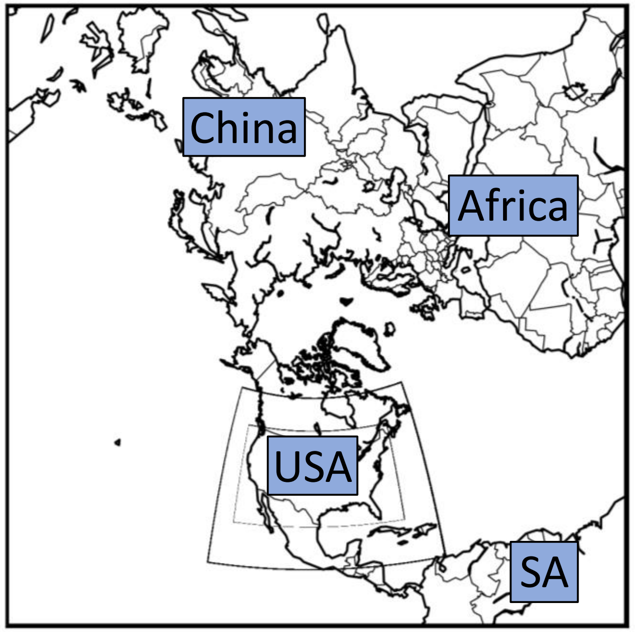
Regional

- US: 2016fe Platform
- Canada: EC 2013 interpolated
- Mexico
  - Mobile 2016 MOVES
  - Other scaled from 2008
- Asia (non-China): MIXv1
- ***China: Tsinghua University (THU)***
  - Lower sulfate than CEDS
  - Lower NOx than CEDS
  - Similar trends in power sector
  - Differences in metals where THU applies government required controls
  - Zhao et al. doi: 10.1073/pnas.1812955115

Details in EPA 2019: epa.gov/sites/production/files/2019-12/documents/2016fe_hemispheric_tsd.pdf and Vukovich et al. CMAS 2018

Results and Evaluation

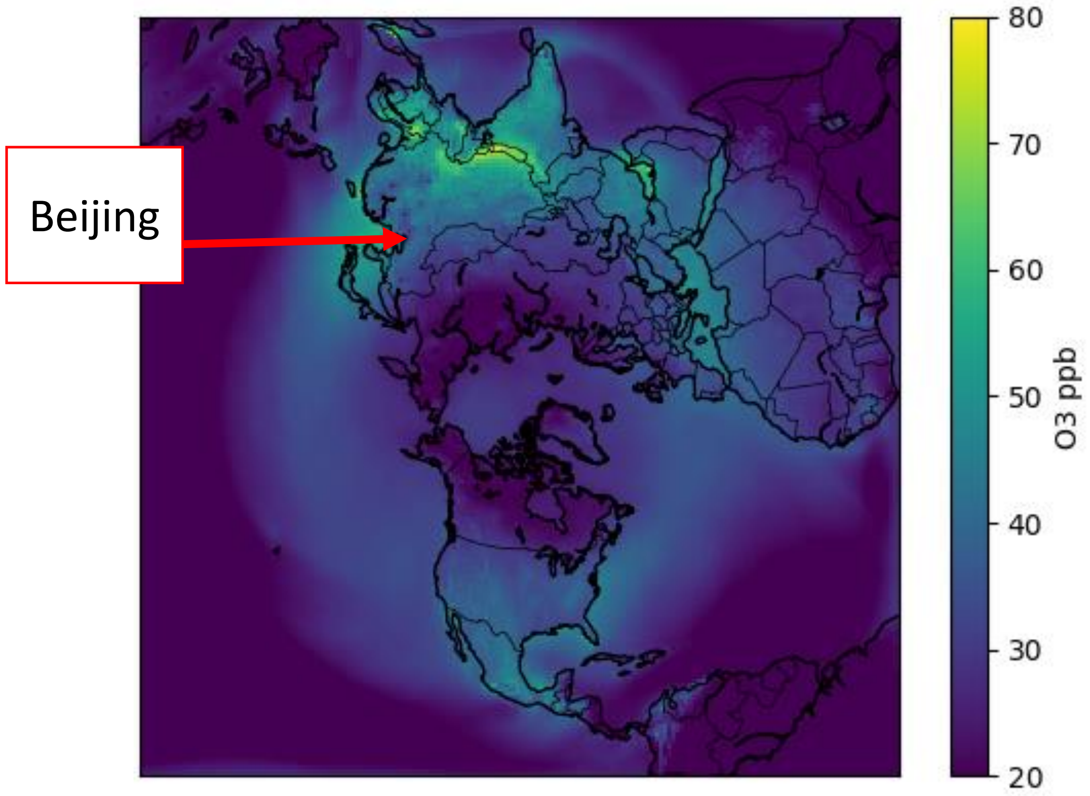
CMAQ-Only
Seasonal Averages for Ozone
Sonde and CASTNet Evaluation
TOAR Qualitative Evaluation



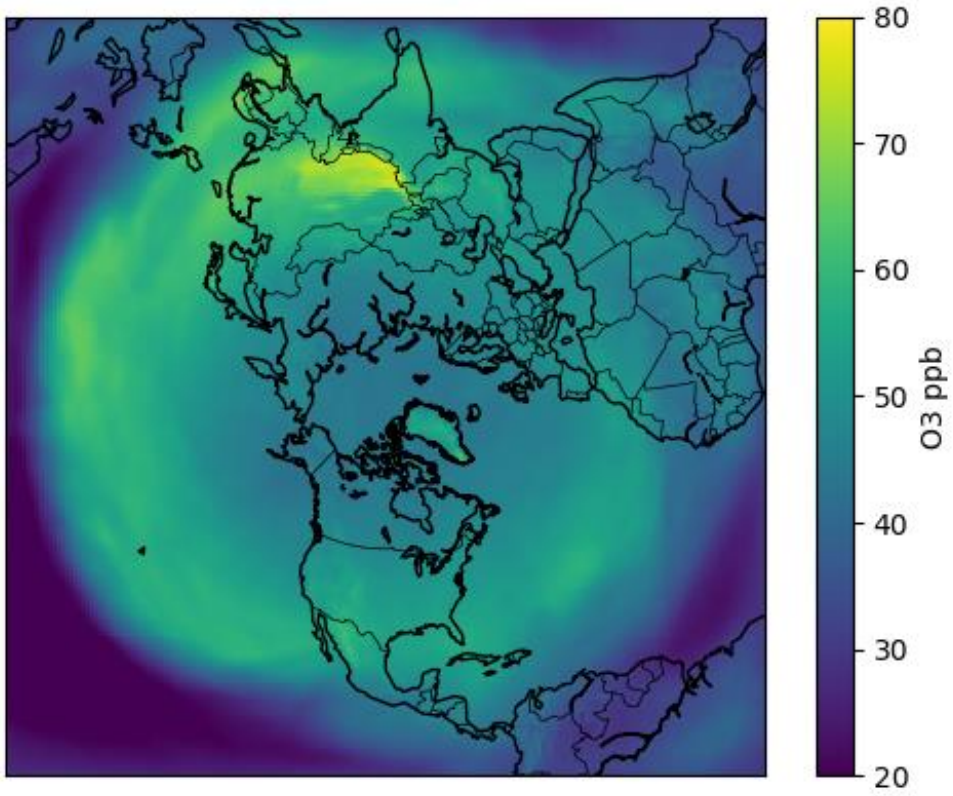
Ozone Surface and about 5km Spring

Northern Hemisphere Spring (March April May, MAM) concentrations are relatively low with clear transport in the mid-troposphere seen most strongly in the southern latitudes

Surface



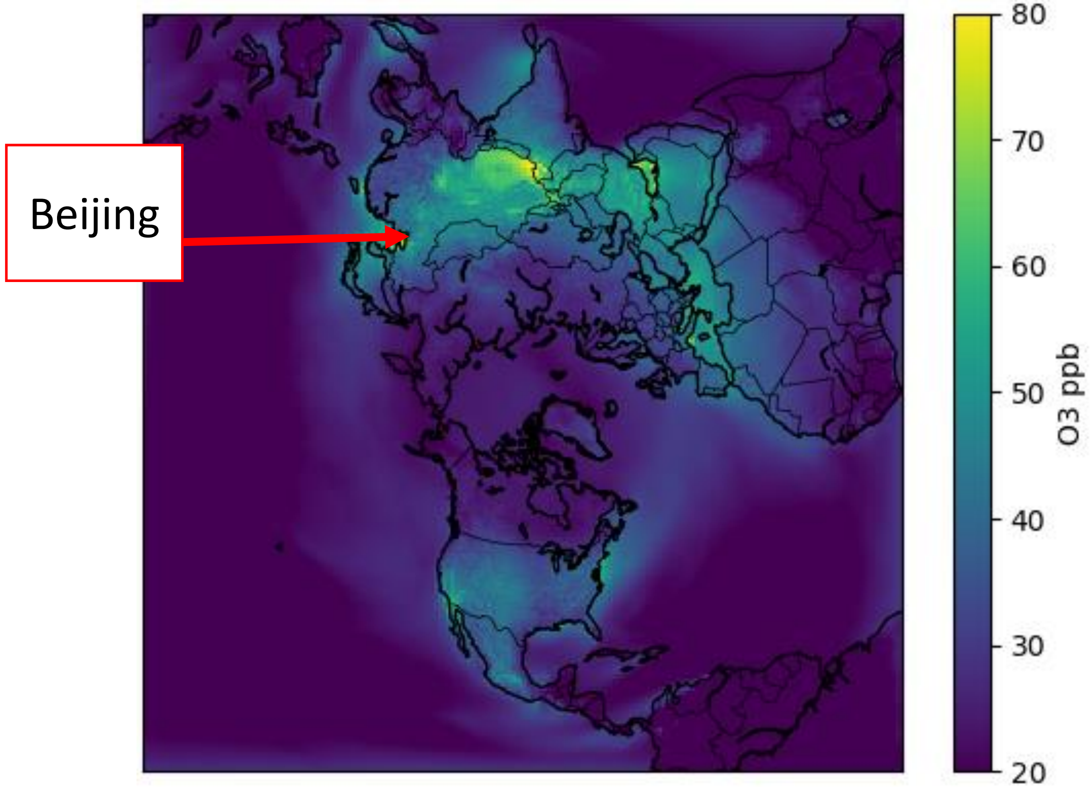
0.5 sigma or ~500hPa or 5km



Ozone Surface and about 5km Summer

Northern Hemisphere Summer (June-July-August, JJA) concentrations are higher both at the surface and aloft., but the transport patterns are less clearly defined than spring.

Surface



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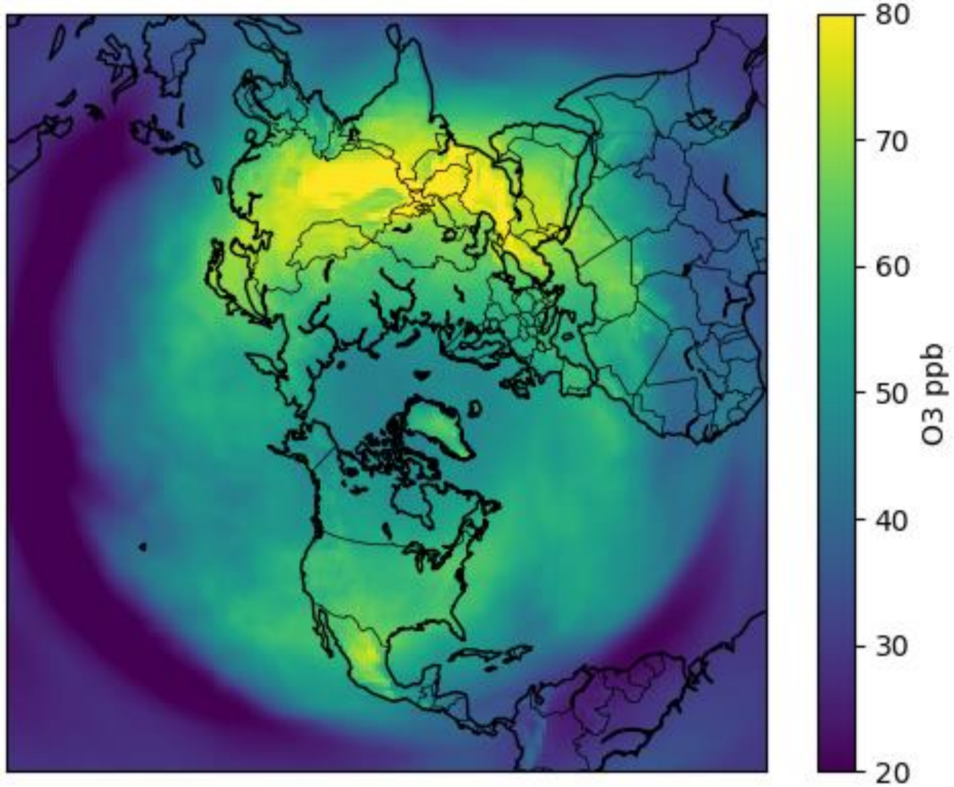
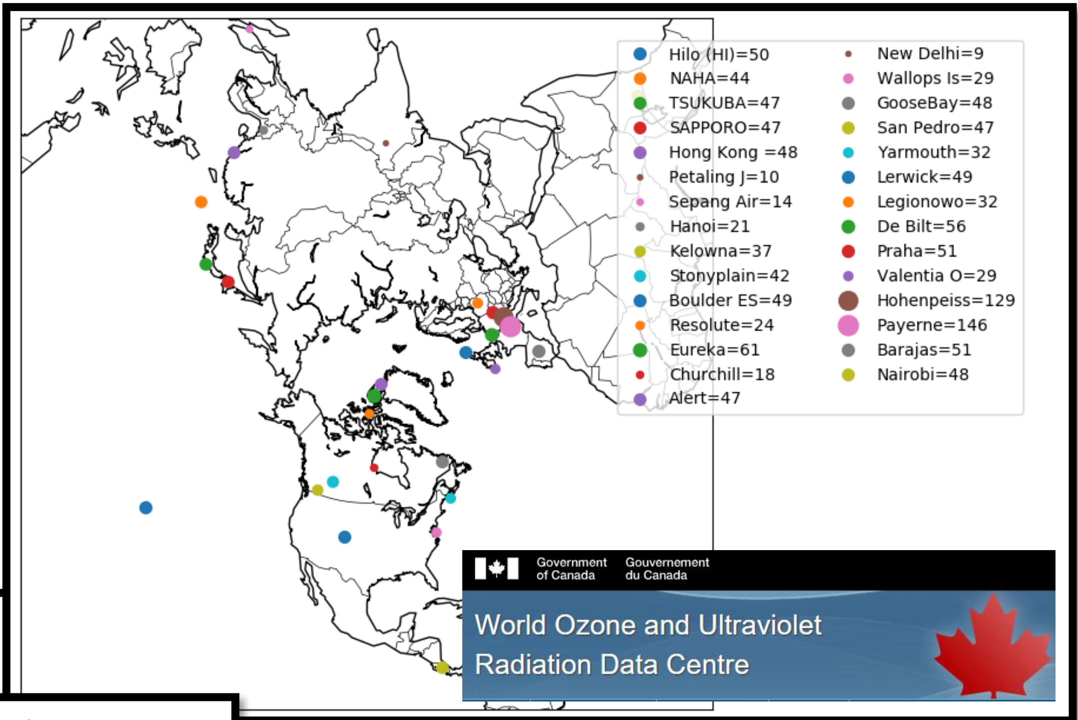
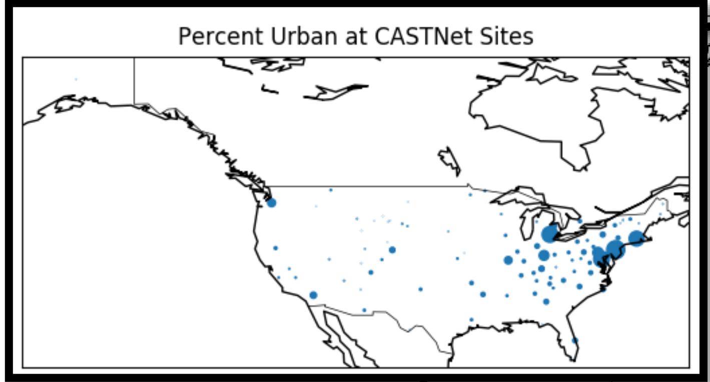
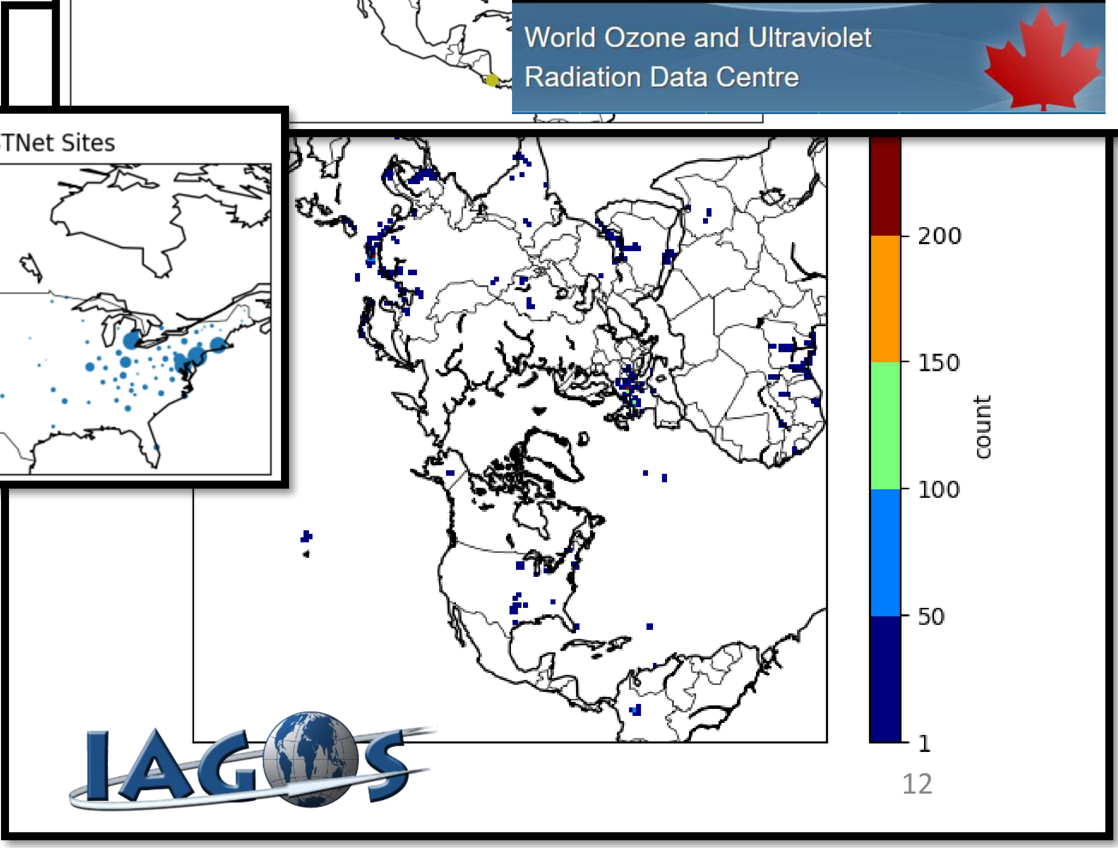


Evaluation Networks

- WOUDC
  - In domain sites: 29; launches: 1315
  - Many in NA and W EU
  - Averaging samples w/in CMAQ sigma levels
- CASTNet
  - Large scale simulations will not capture small-scale gradients
  - Not all CASTNet sites are rural
- In-service Aircraft for Global Observing System (IAGOS)
  - 333 grid cells covered
  - 3156 ascents or descents

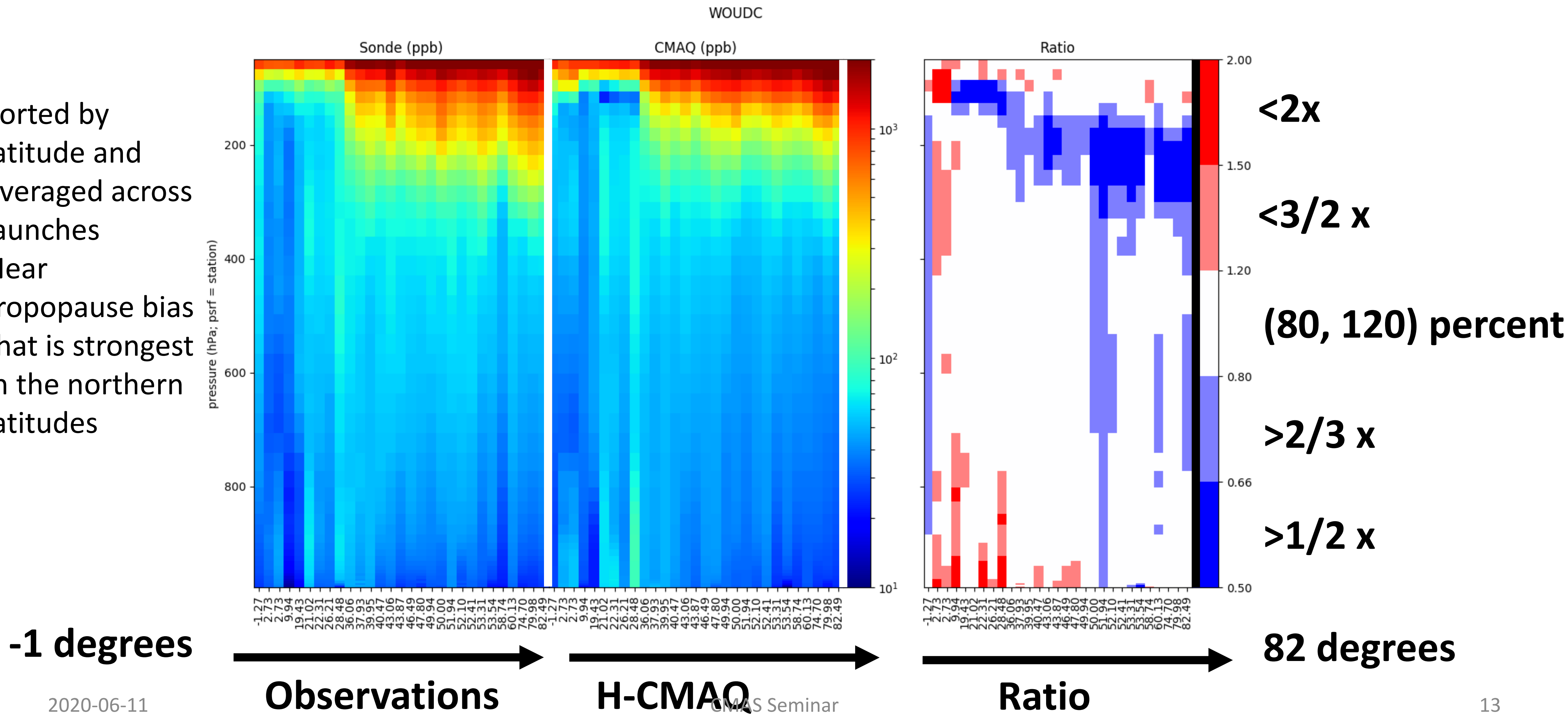






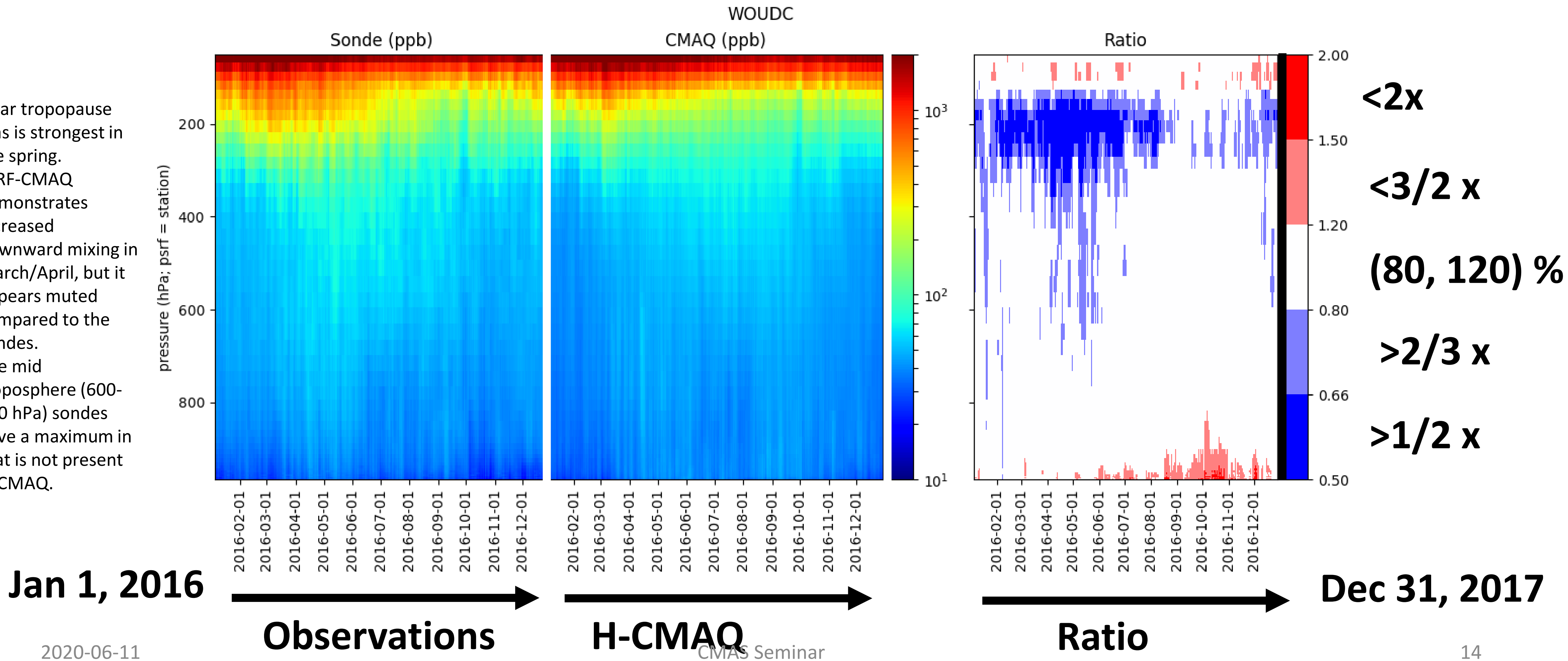
WOUDC Sondes: by Site (all Times)

- Sorted by latitude and averaged across launches
- Near tropopause bias that is strongest in the northern latitudes



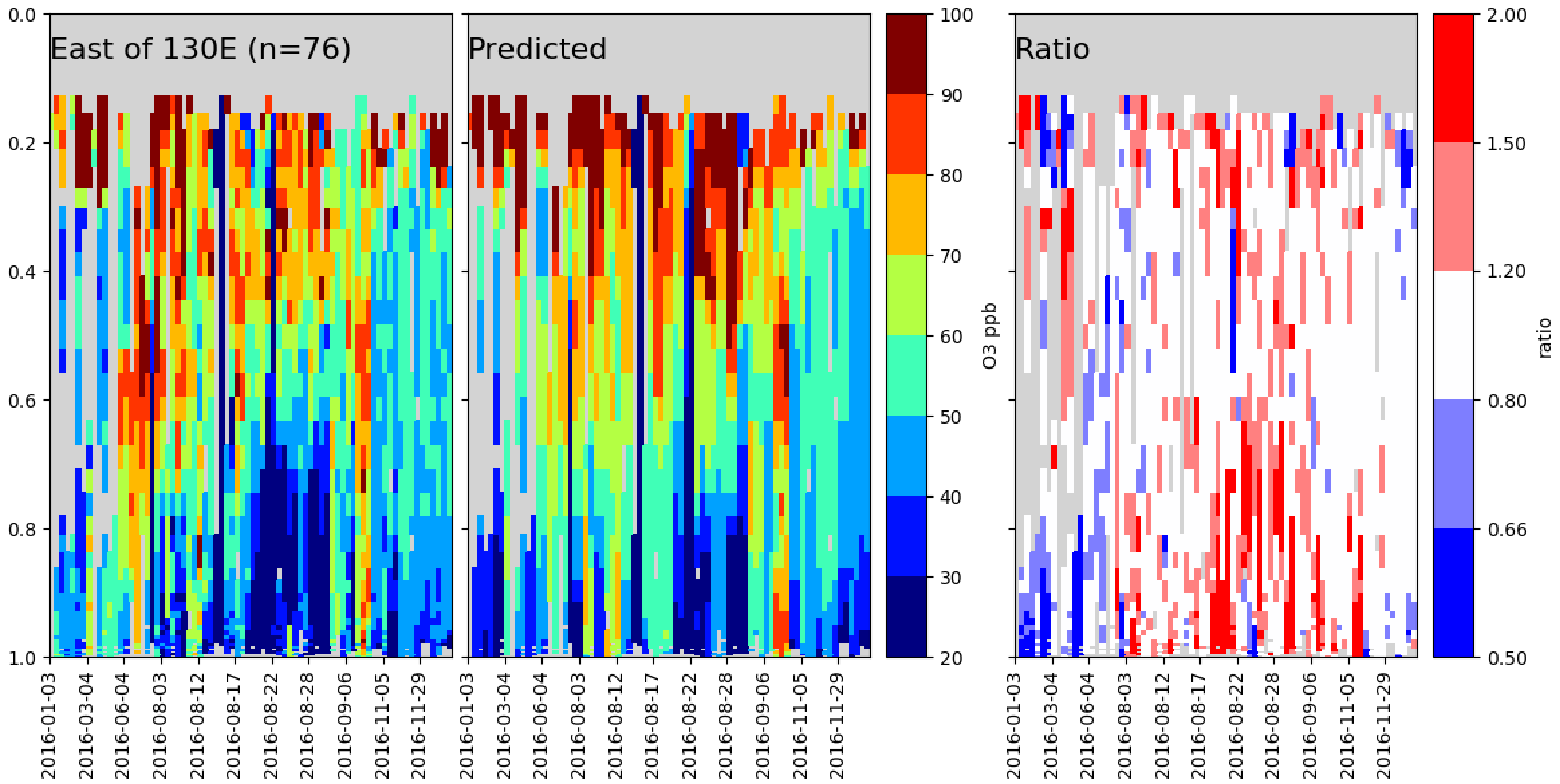
WOUDC Sondes: by Time (all Sites)

- Near tropopause bias is strongest in the spring.
- WRF-CMAQ demonstrates increased downward mixing in March/April, but it appears muted compared to the sondes.
- The mid troposphere (600-400 hPa) sondes have a maximum in that is not present in CMAQ.



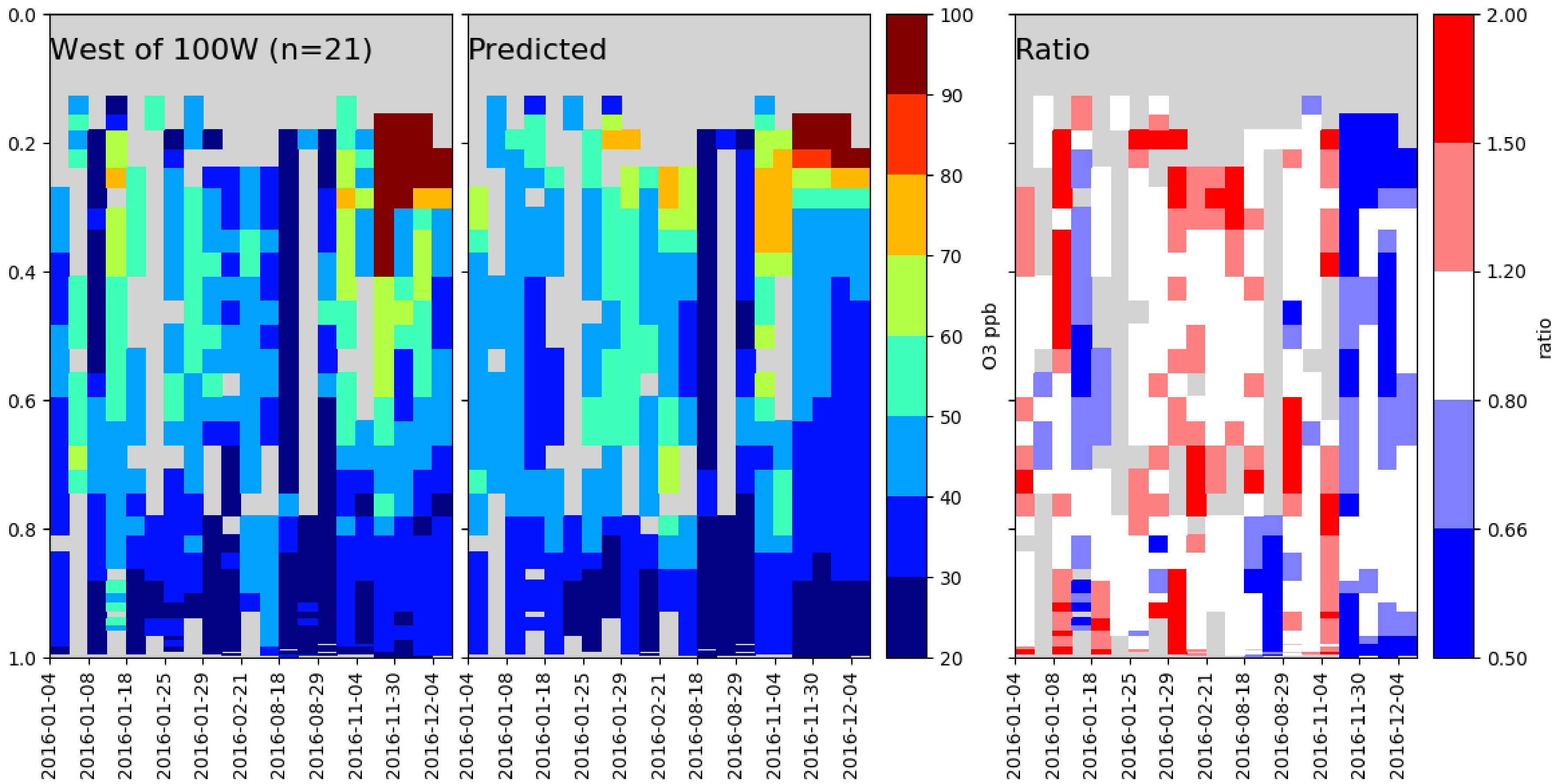
IAGOS Flights

- Focusing on east (Japan) for Asian outflow
- Missing Apr, Jul, and Oct flights
- Captures a few prominent upper air features
- Tends to be high biased
- Over the continent, tends to be higher biased

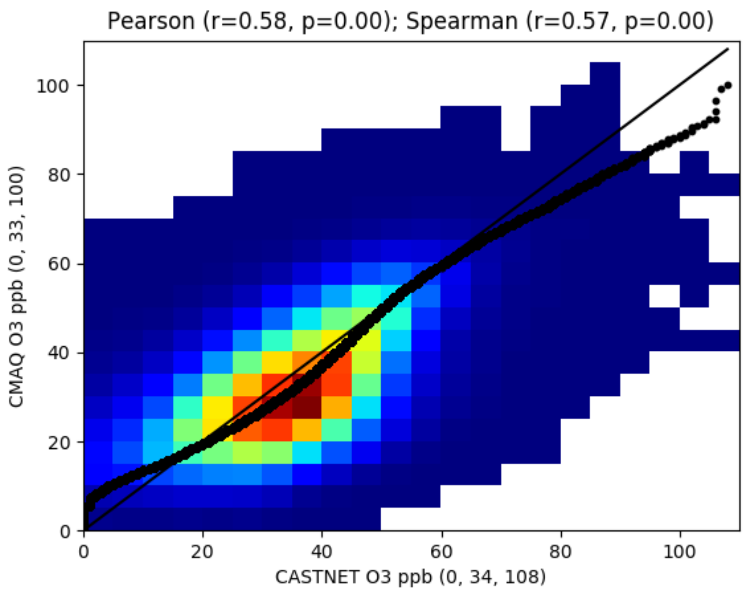


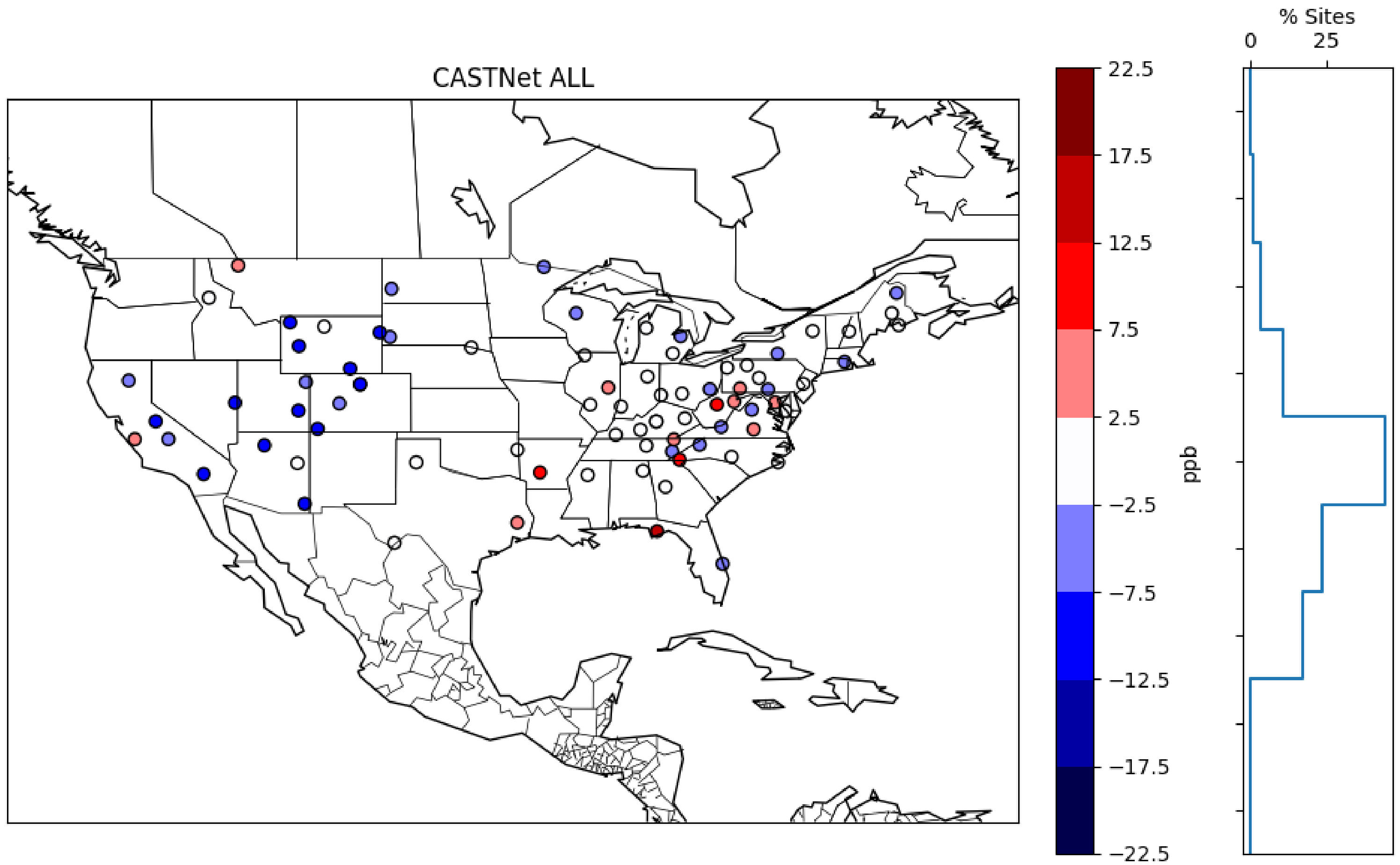
IAGOS Flights

- Focusing on west (Hawaii) for incoming air
- Missing Mar-Jul, and Oct flights…
- Captures several key features
- Mixed performance



CASTNet Monitors: All Year

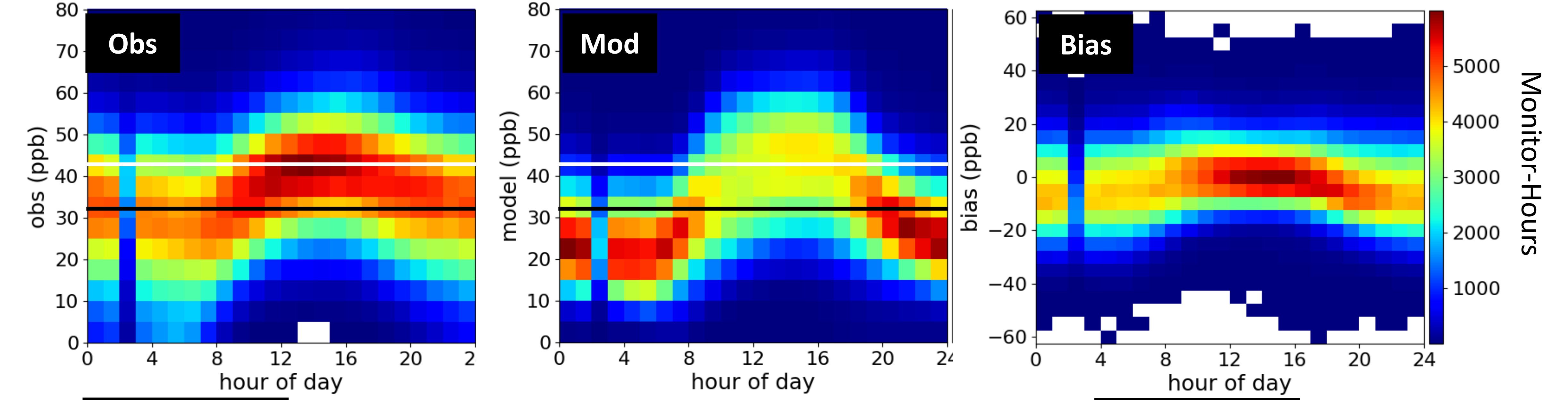


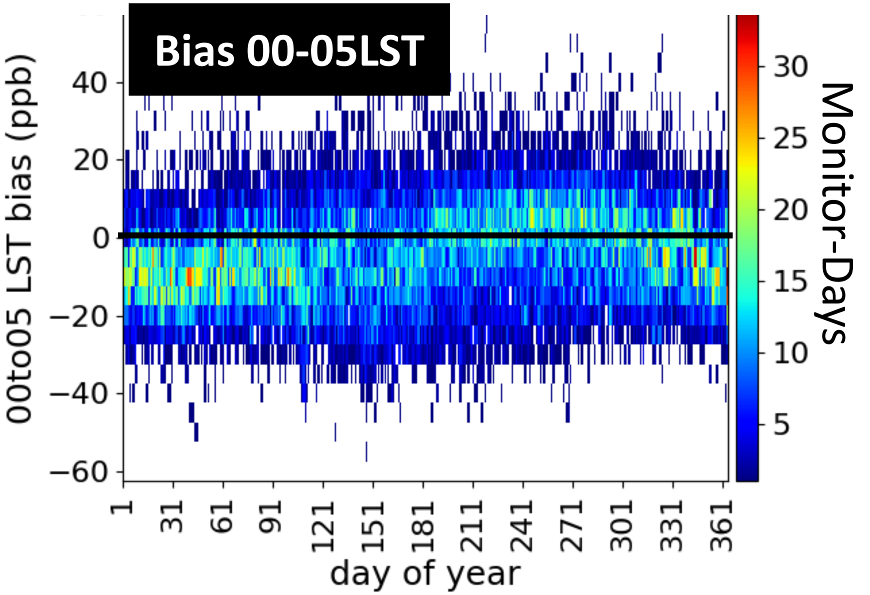


CASTNet monitors are not all rural, but they are frequently used as a proxy. Here we evaluate hourly ozone.

- 15LST has an r=0.67
- Performance at these monitors is within ±7.5 ppb at most monitors.
- There is a west-east bias divide

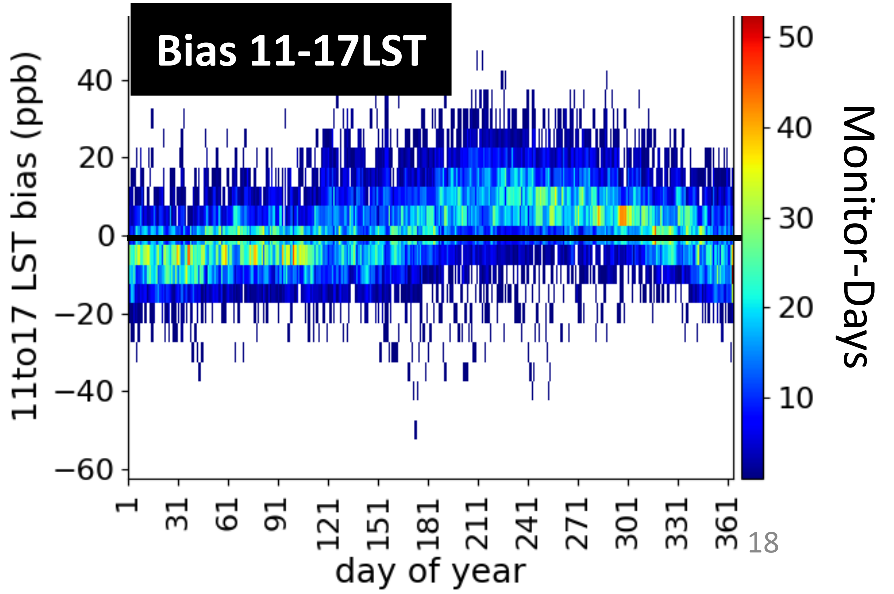
CASTNet Diurnal Performance



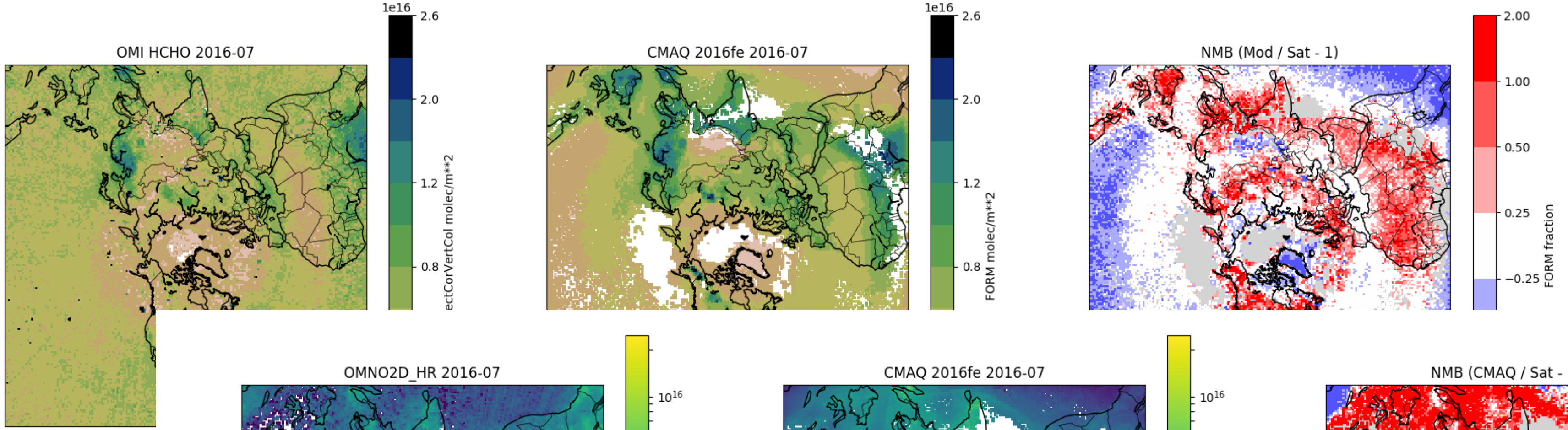


Plots show count of monitor-days with bias as a function of hour of day or day of year

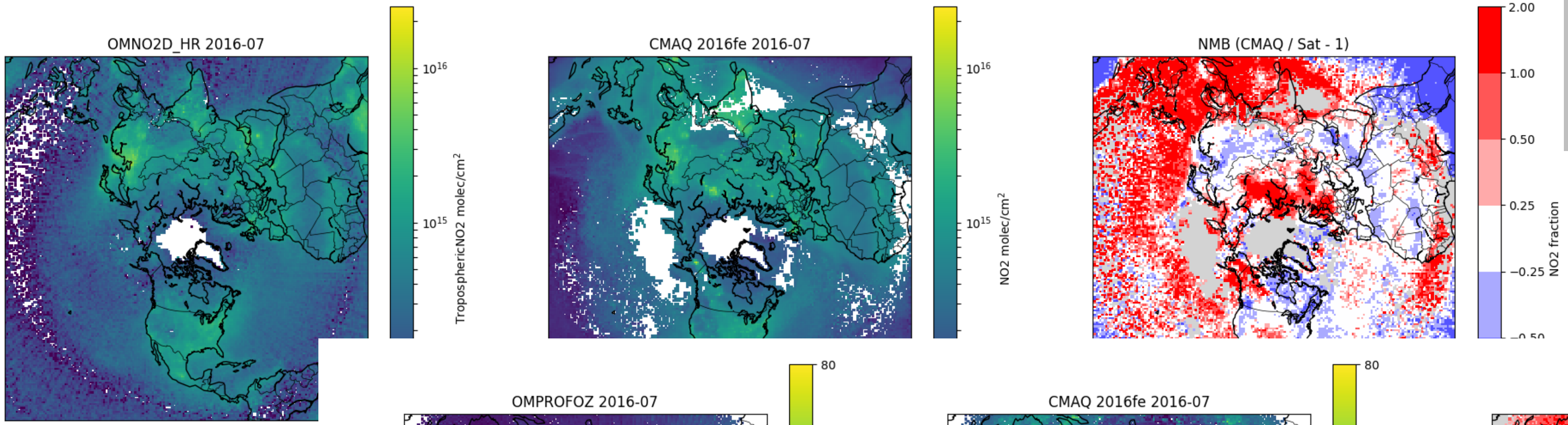
- Hour of day all year
- 0-5LST over the year
- 11-17LST over the year



Satellites and Sondes Evaluation avail elsewhere

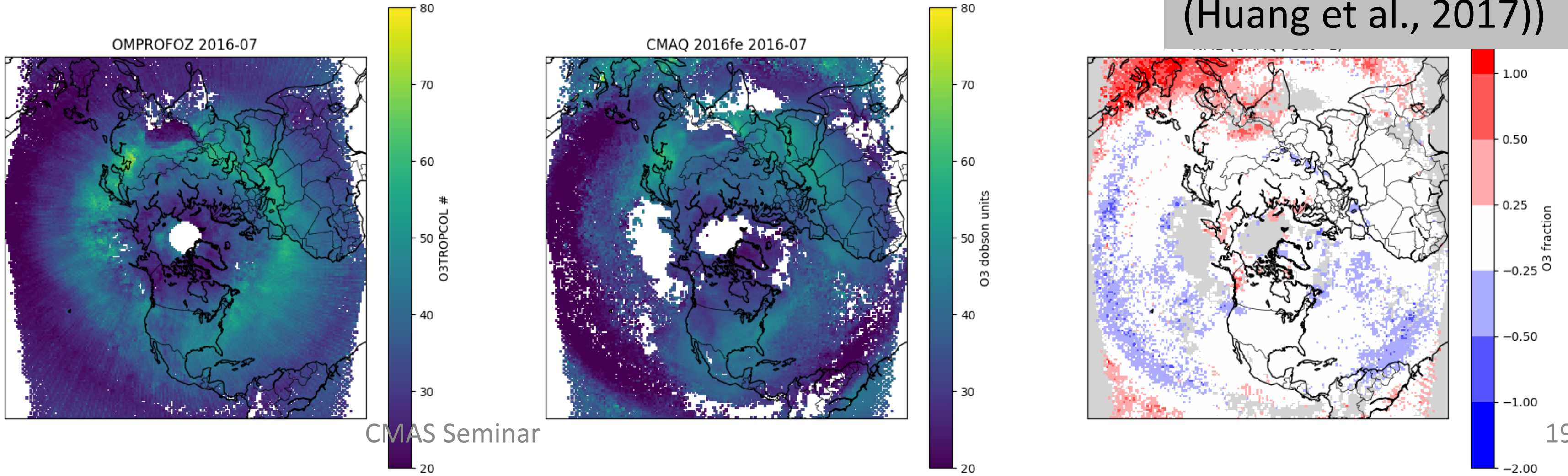


SAO Formaldehyde
(González Abad et al., 2015)



NASA Nitrogen Dioxide
(Krotkov et al., 2017,
Lamsal OMNO2D_HR)

SAO Ozone Profiles
(Huang et al., 2017))



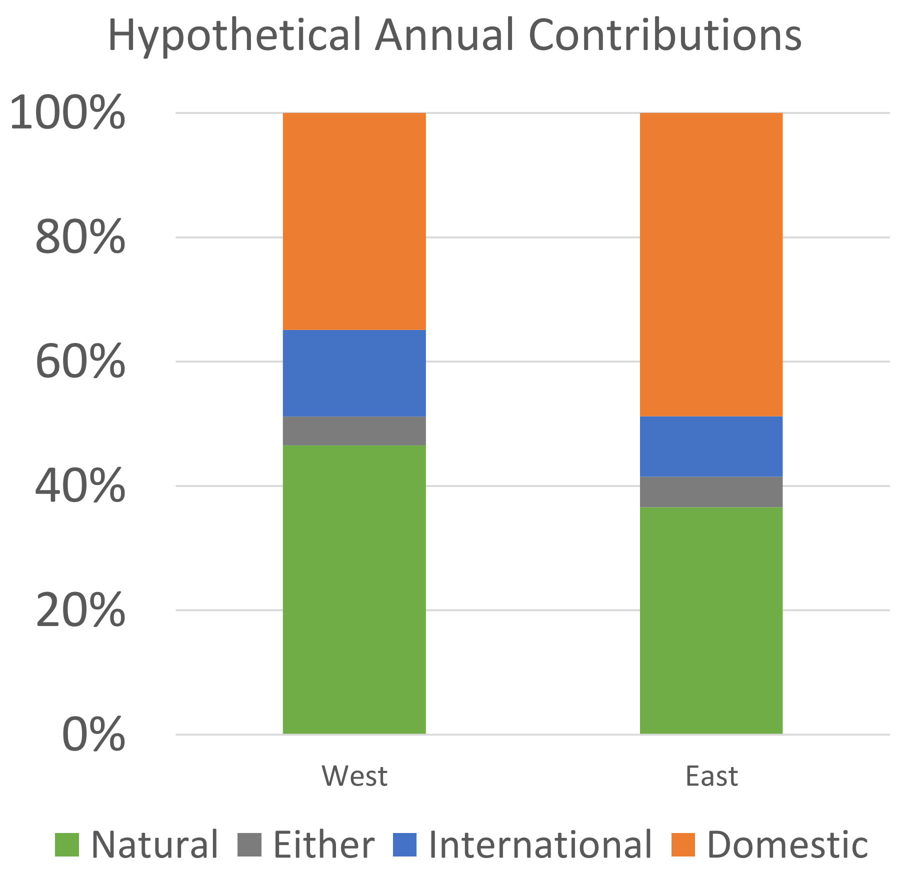
Summary

- Compares well to sondes in the mid troposphere
  - appears to have a near tropopause low-bias
  - low bias northward of 50 degrees Dec-May
  - performing similarly to GEOS-Chem used for the 2011 platform
- Routine aircraft measurements show mixed results.
- Performs best in JJA compared to CASTNet
  - Most data is within 10 ppb of observations
  - Clear West-East bias gradient
- TOAR evaluation suggests similar results with better performance at rural than urban monitors
- Compared to current test of GEOS-Chem v11-02* were less biased.
  - H-CMAQ was low-biased while GC was high-biased compared to sondes
  - Testing GC version (v12.0.1), considering meteorology

*w/FINN fires and 2016 lightning

Part II: Zero-out estimates of ozone contributions

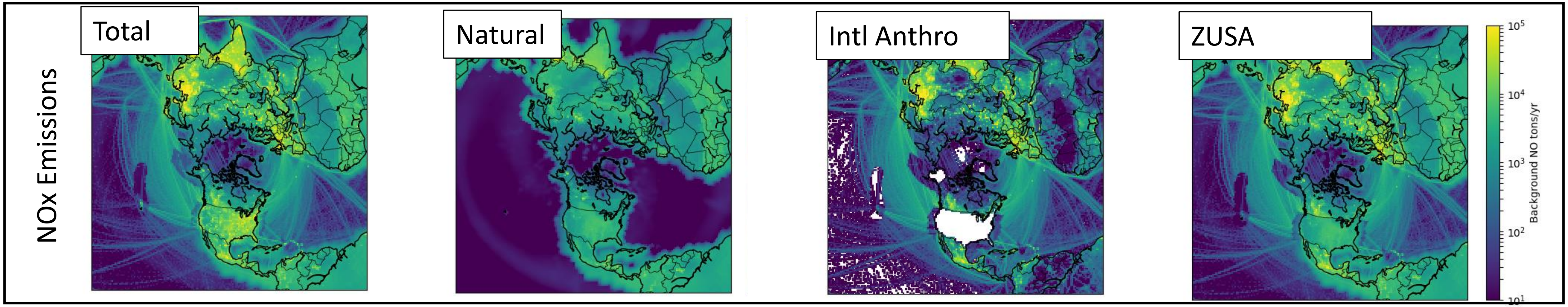
- *Motivations:*
  - Interannual variability (e.g., Lin et al., 2017)
  - Modeling system (e.g., Huang et al. 2017)
  - 2016 platform (α)
- *New Estimates:*
  - *Northern Hemispheric:* **Natural**
  - International anthropogenic: **Intl**
  - Domestic anthropogenic: **USA**
  - And either: **Residual**…
- Long range transport and aloft results
  - At 108km & Separating China and India
- Surface results
  - 108km and 12 km nested from 108 km LBC
  - Natural, Intl, USA



Estimates of 2016 Ozone Components

- **Predictions** = *F*(*M*, *E*)
  - Total : *E* = sum({*nat, usa, sum(intl)*})
  - Natural : *E* = sum({*nat*}); *soil NOx and methane are treated as natural*
  - ZINTL : *E* = sum({*nat, usa*}); *Prescribed fires are treated as anthropogenic*
  - ZUSA aka USB : E = sum({nat, sum(intl)}); Others…

Evaluations: Henderson CMAS 2018; IGC9 2019; CMAS-SA 2019

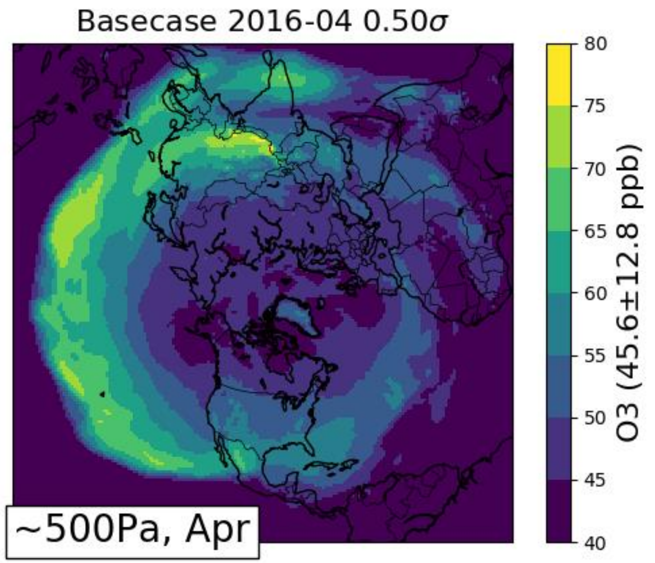


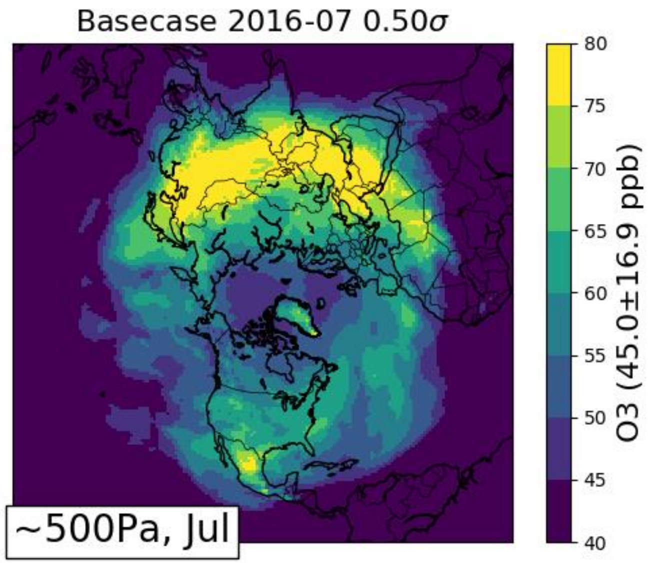
Contributions
- Natural = ZANTH
- USA = Total - ZUSA
- Intl = Total - ZINTL
- RES* = Total - USA - INTL – NAT

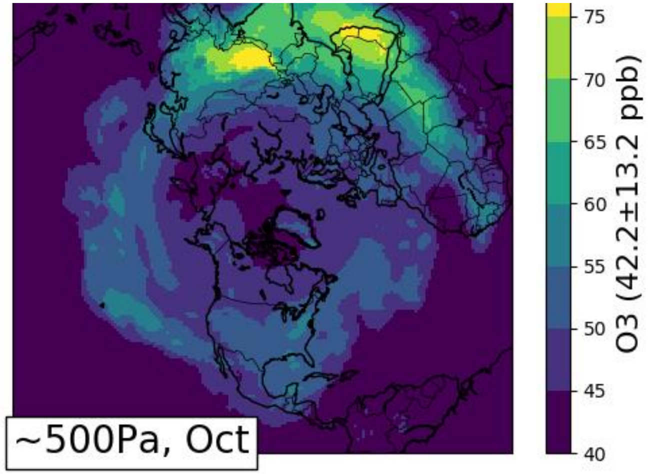
International Parts
- CHN = Total - ZCHN
- SHIP = Total - ZSHIP
- IND = Total - ZIND
- CANMEX = Total – ZCANMEX
- OTHER = Intl - CHN - IND - SHIP - CANMEX

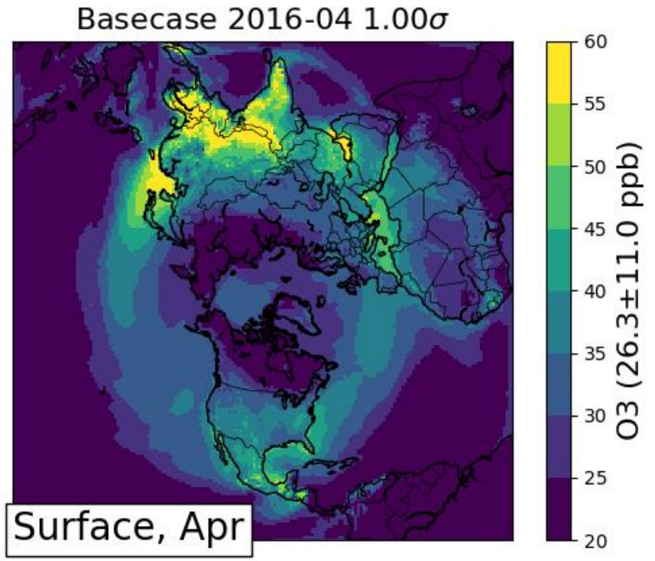
Monthly average ozone illustrate transport

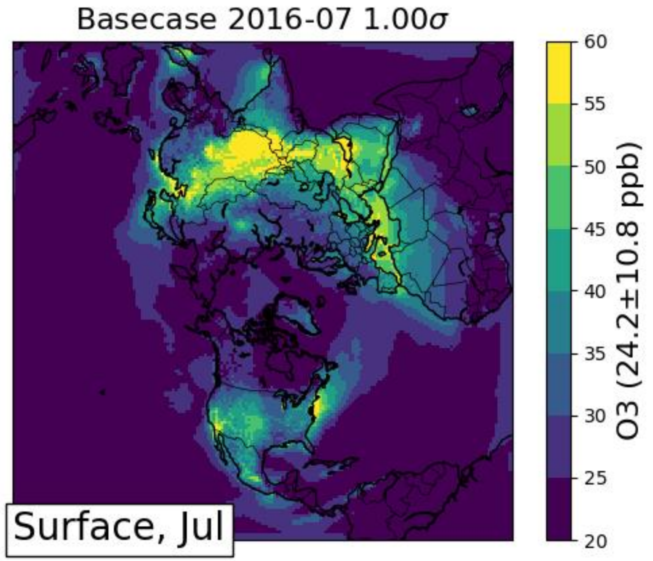
Evaluations: Henderson CMAS 2018; IGC9 2019; CMAS-SA 2019

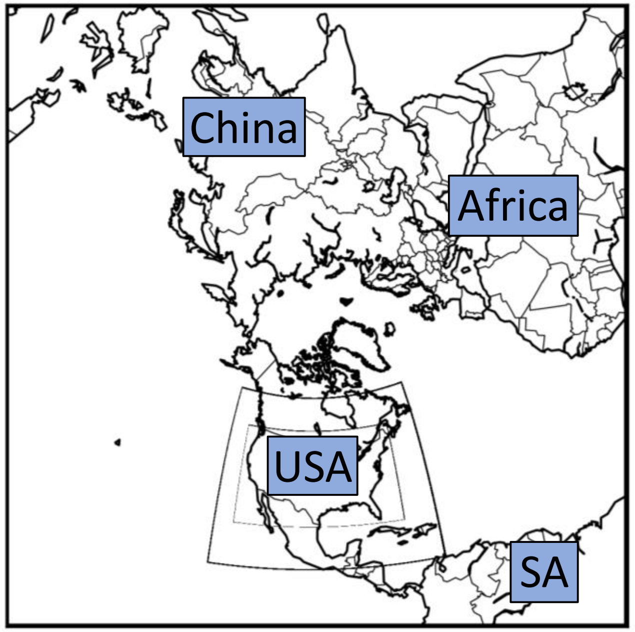




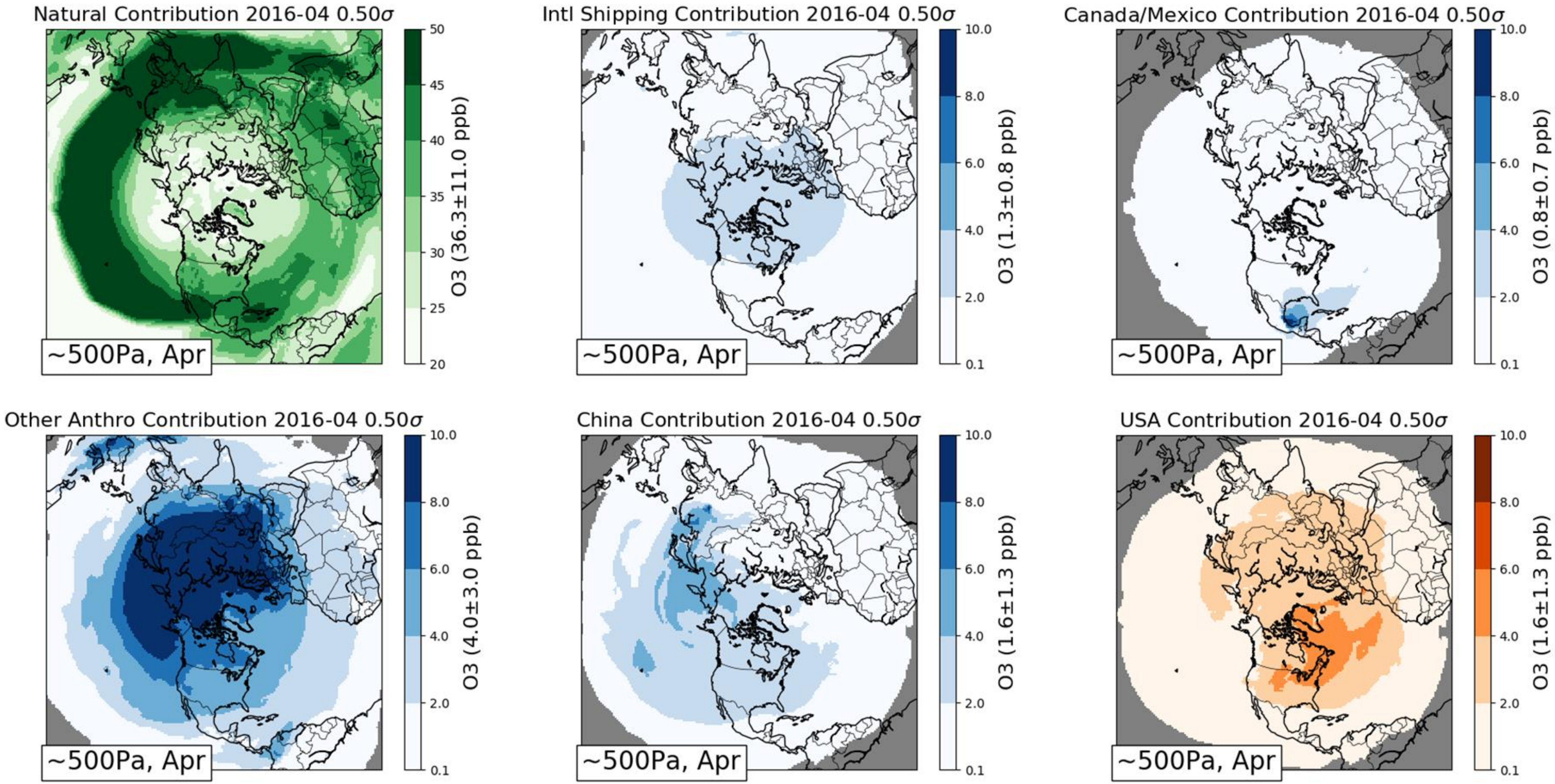




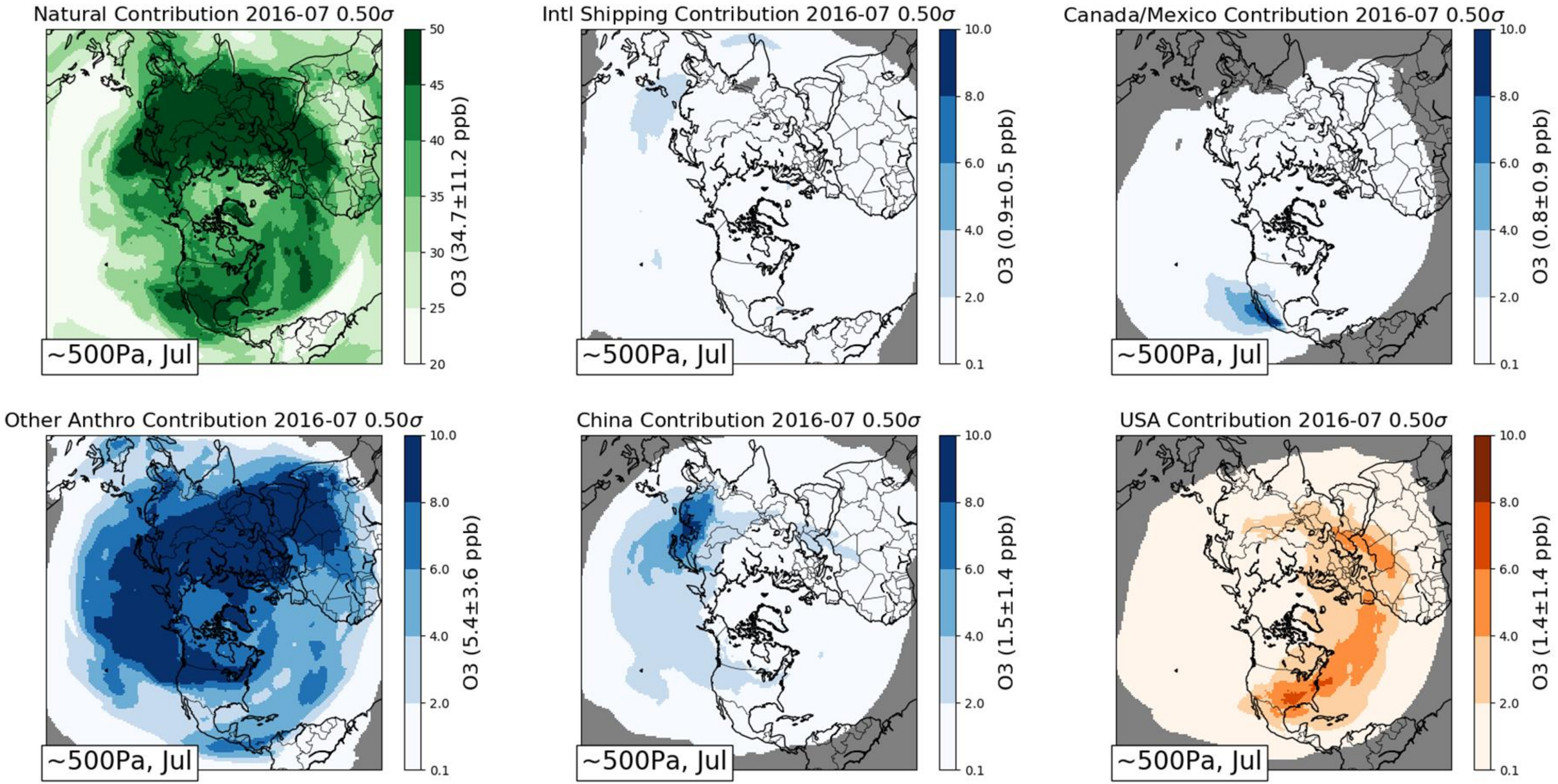




Ozone source contributions in April aloft

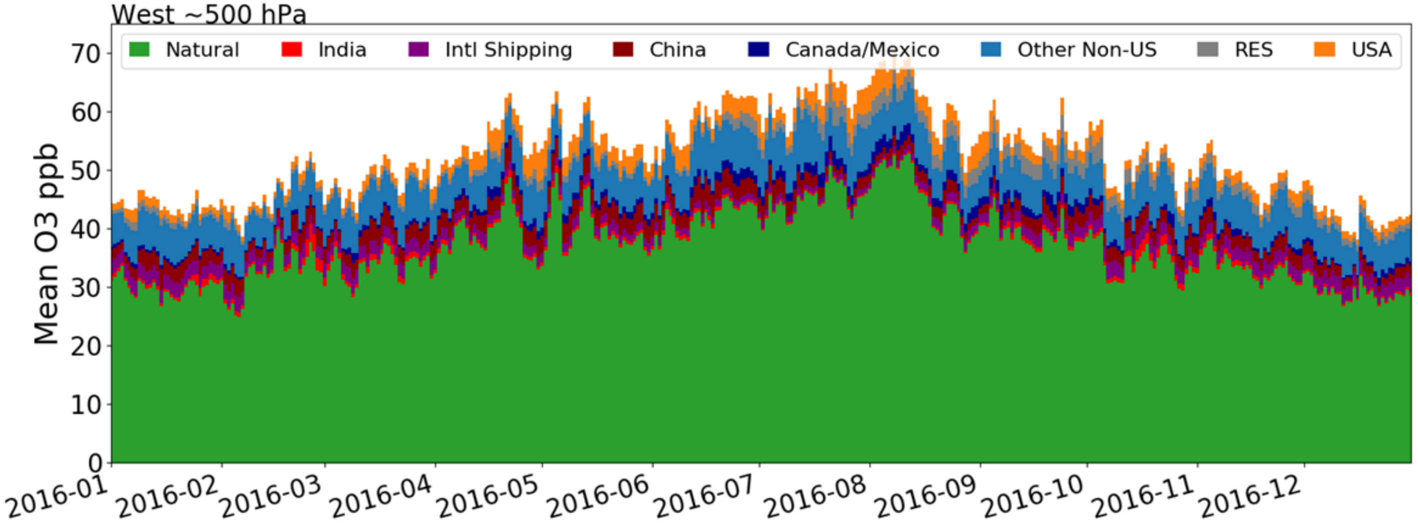


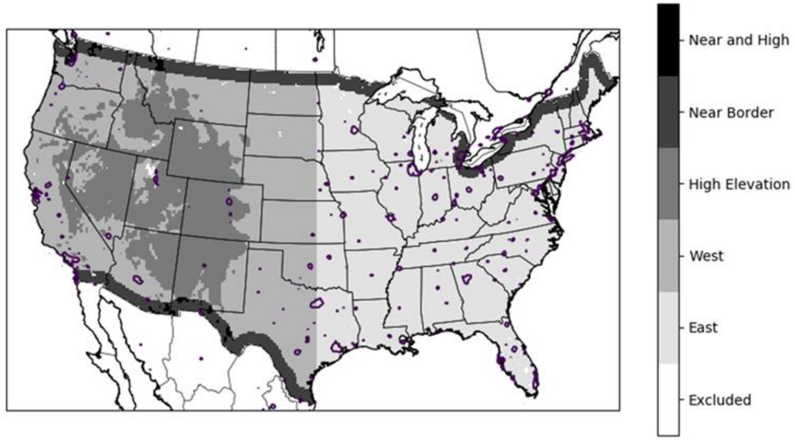
Ozone source contribution in July aloft



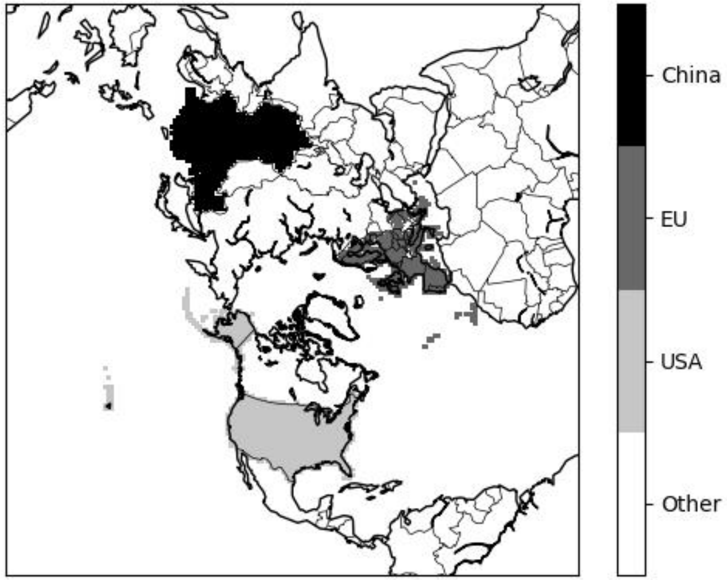
Difference between West and East aloft (108km)

West Contributions

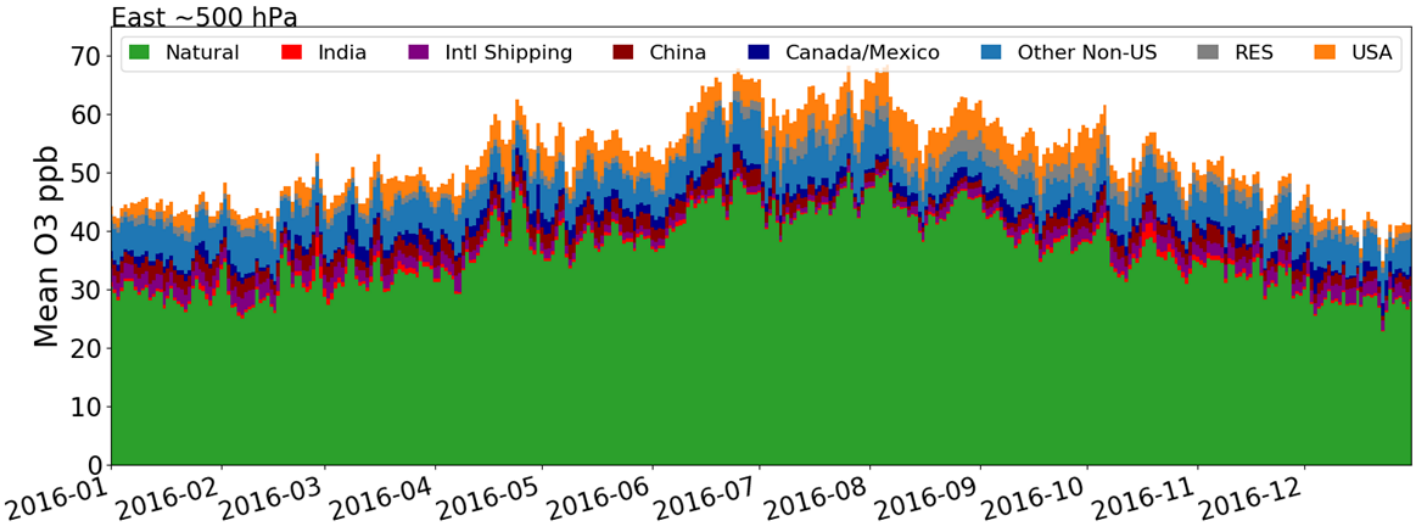




Other countries 10-15 ppb

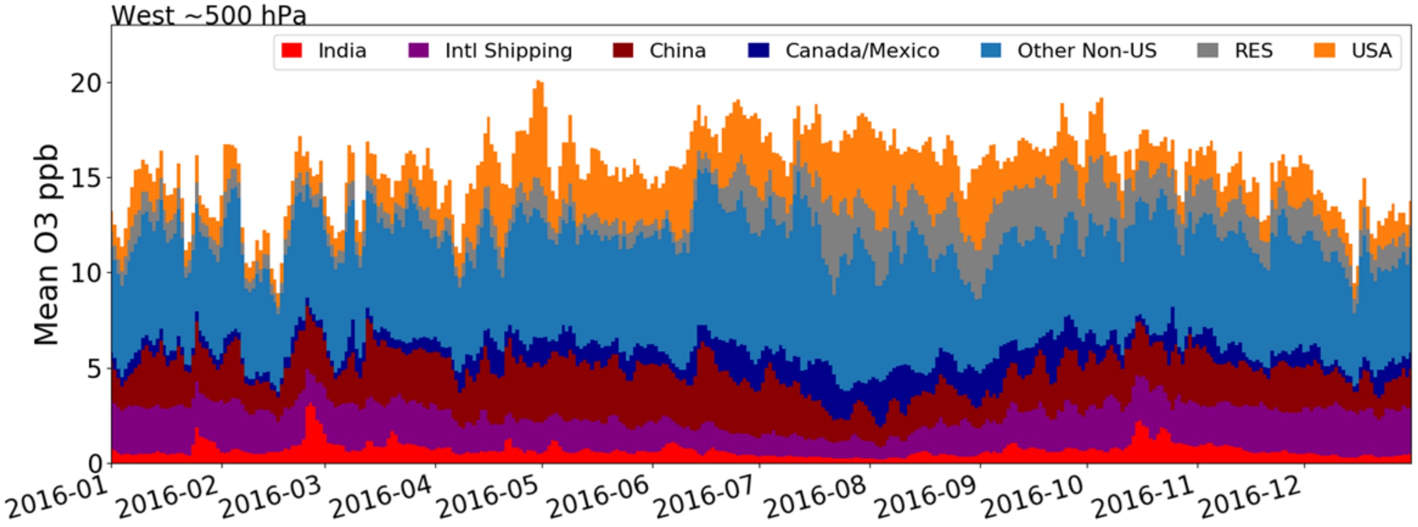


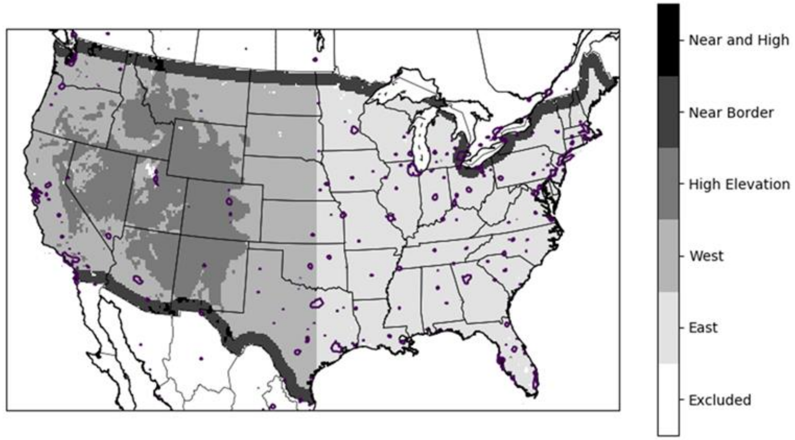
East Contributions



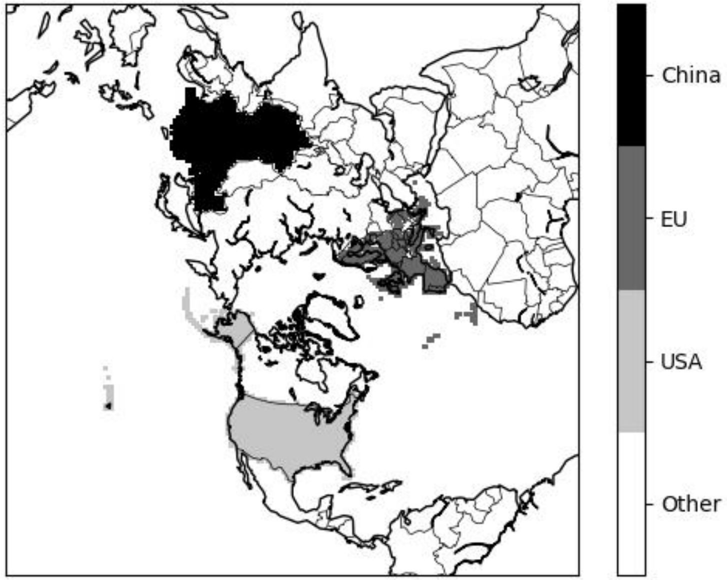
Difference between West and East Aloft (108km)

West Anthropogenic

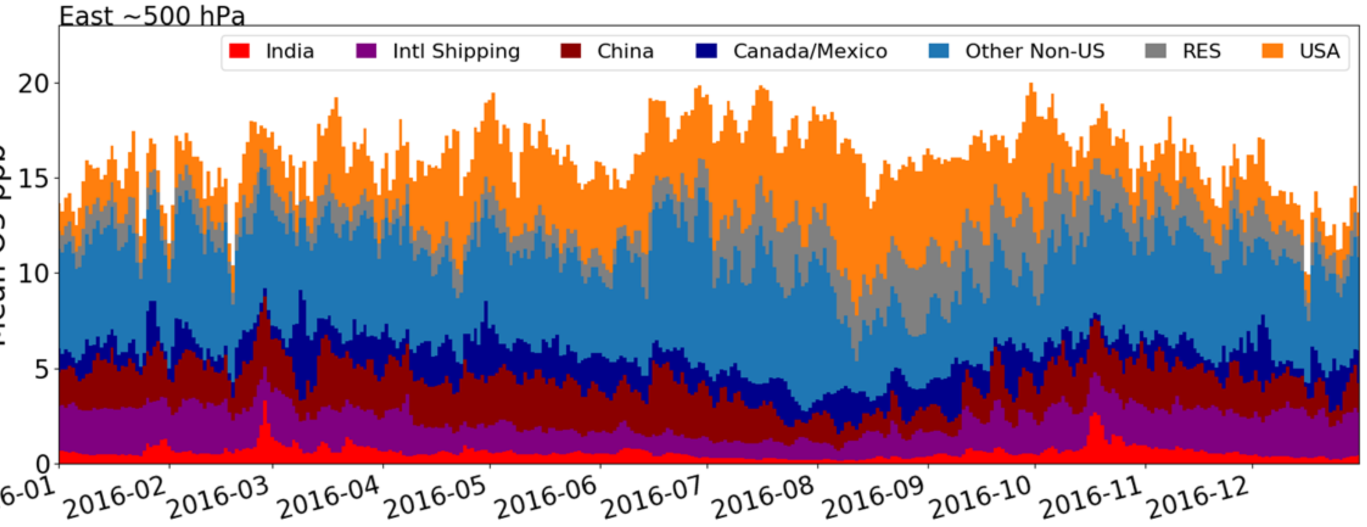




Other countries 10-15 ppb

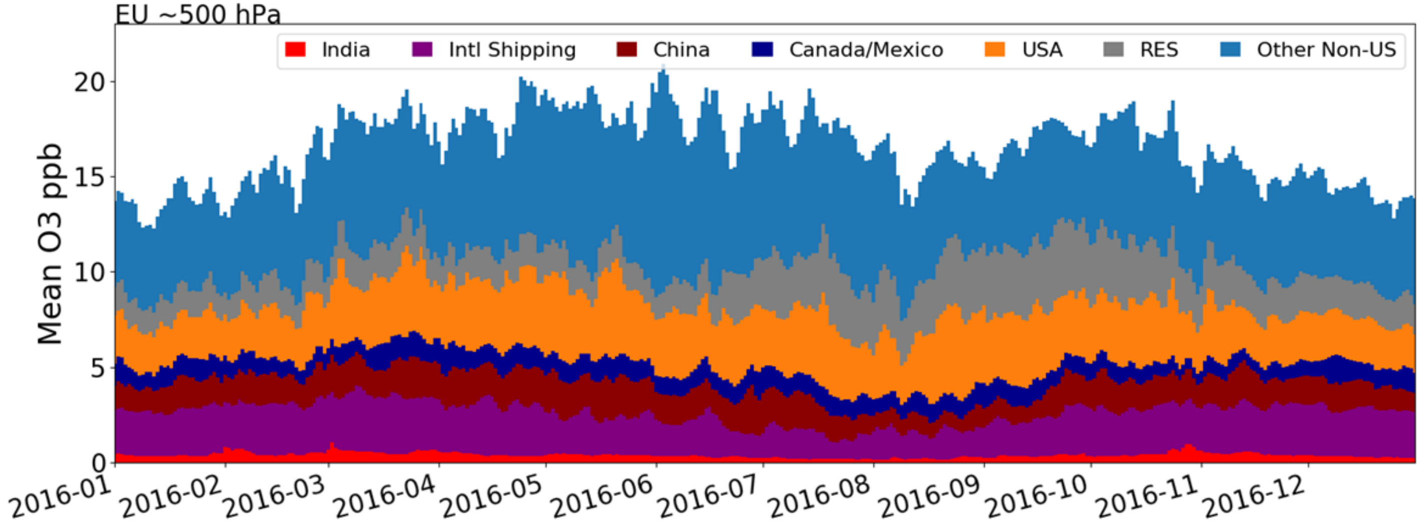


East Anthropogenic

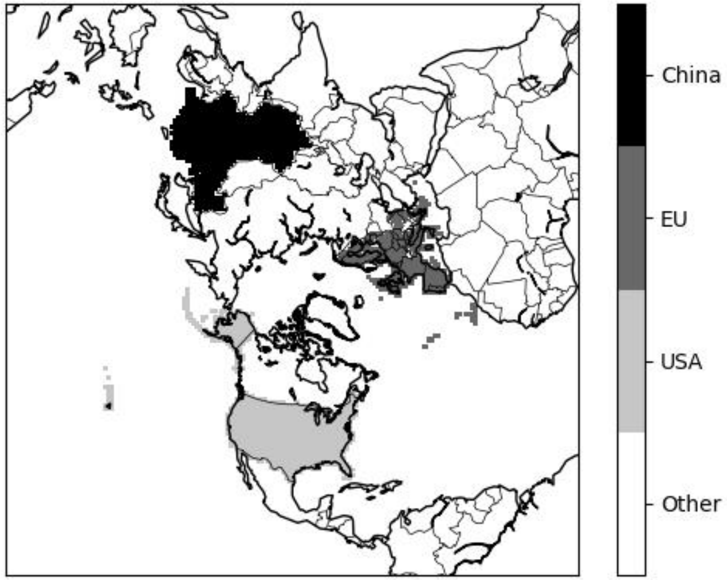


China and the European Union (108km)

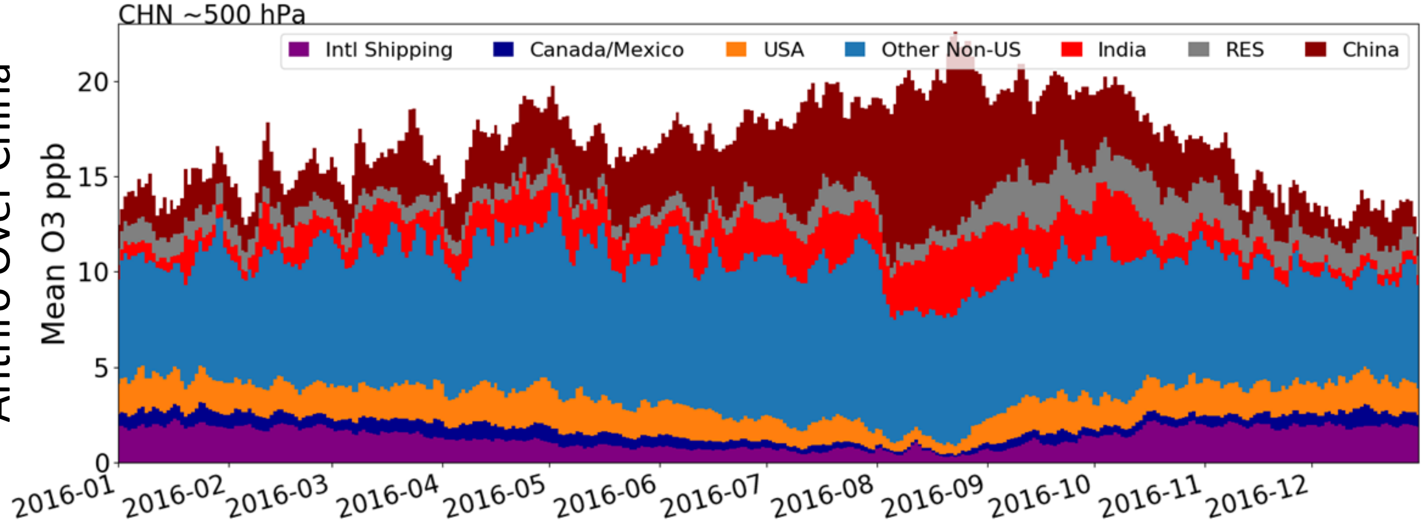
Anthro over the EU
(UK in EU at time of writing)



Other Non-US is not all EU, but this gives us a sense that upwind contributions similar in the EU as in the West.

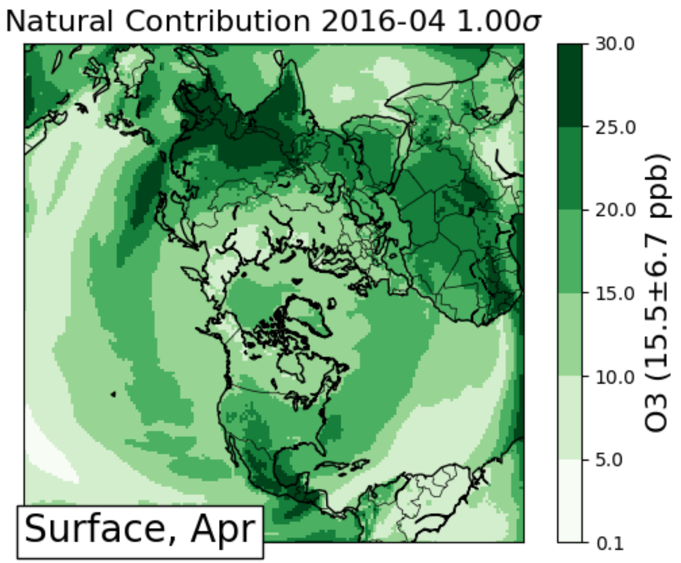


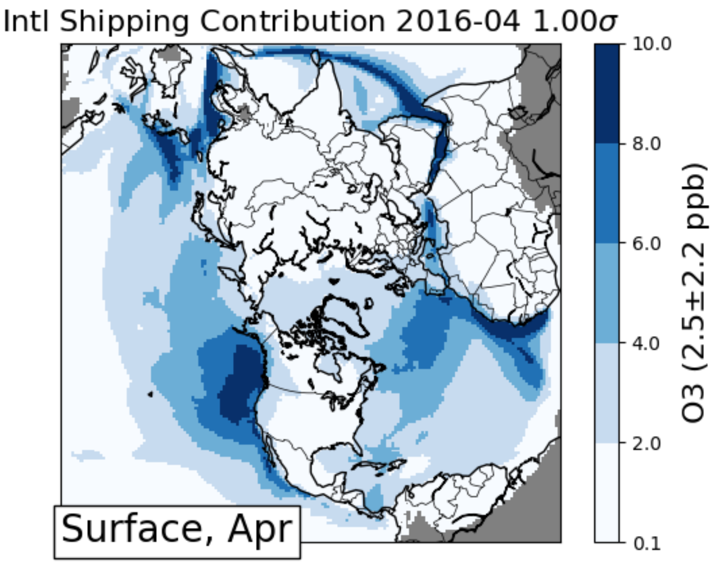
Anthro Over China

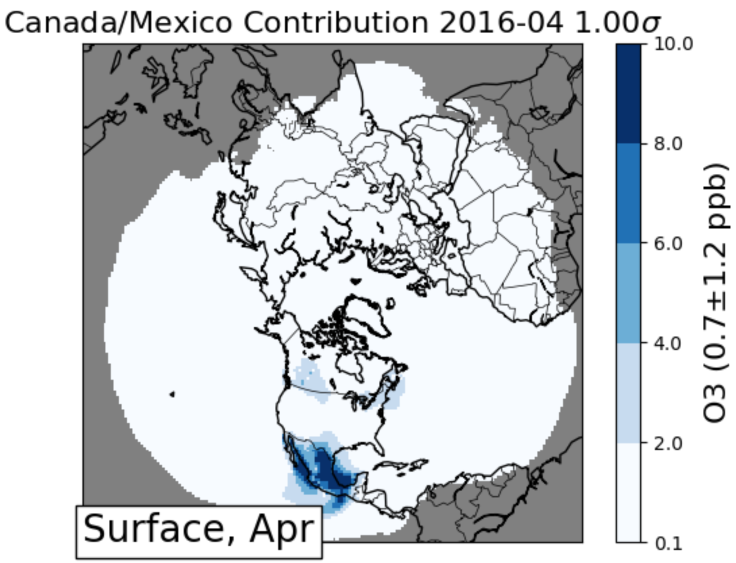


China Aloft ozone from other countries is 10-15 ppb

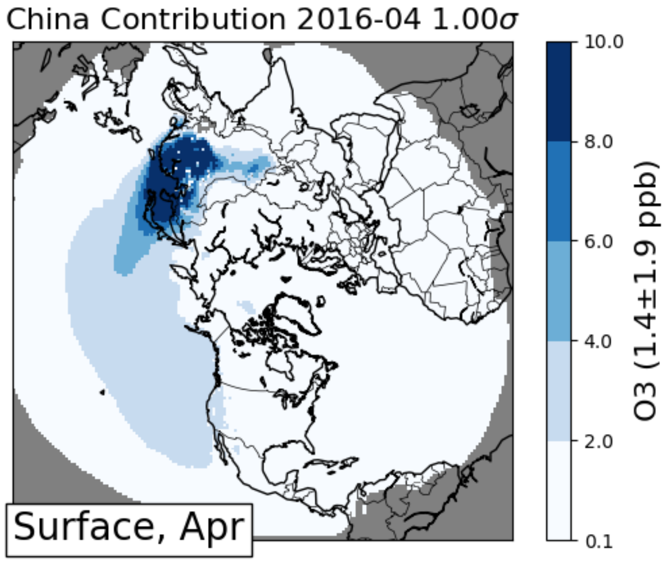
Ozone source contributions in April at the Surface

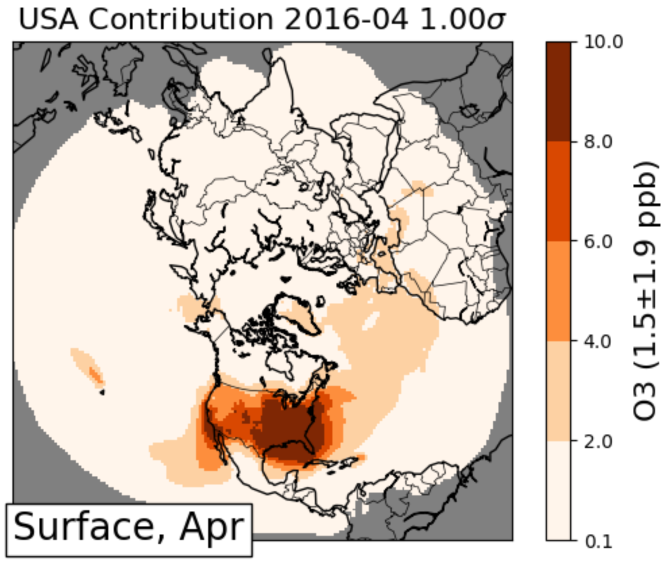




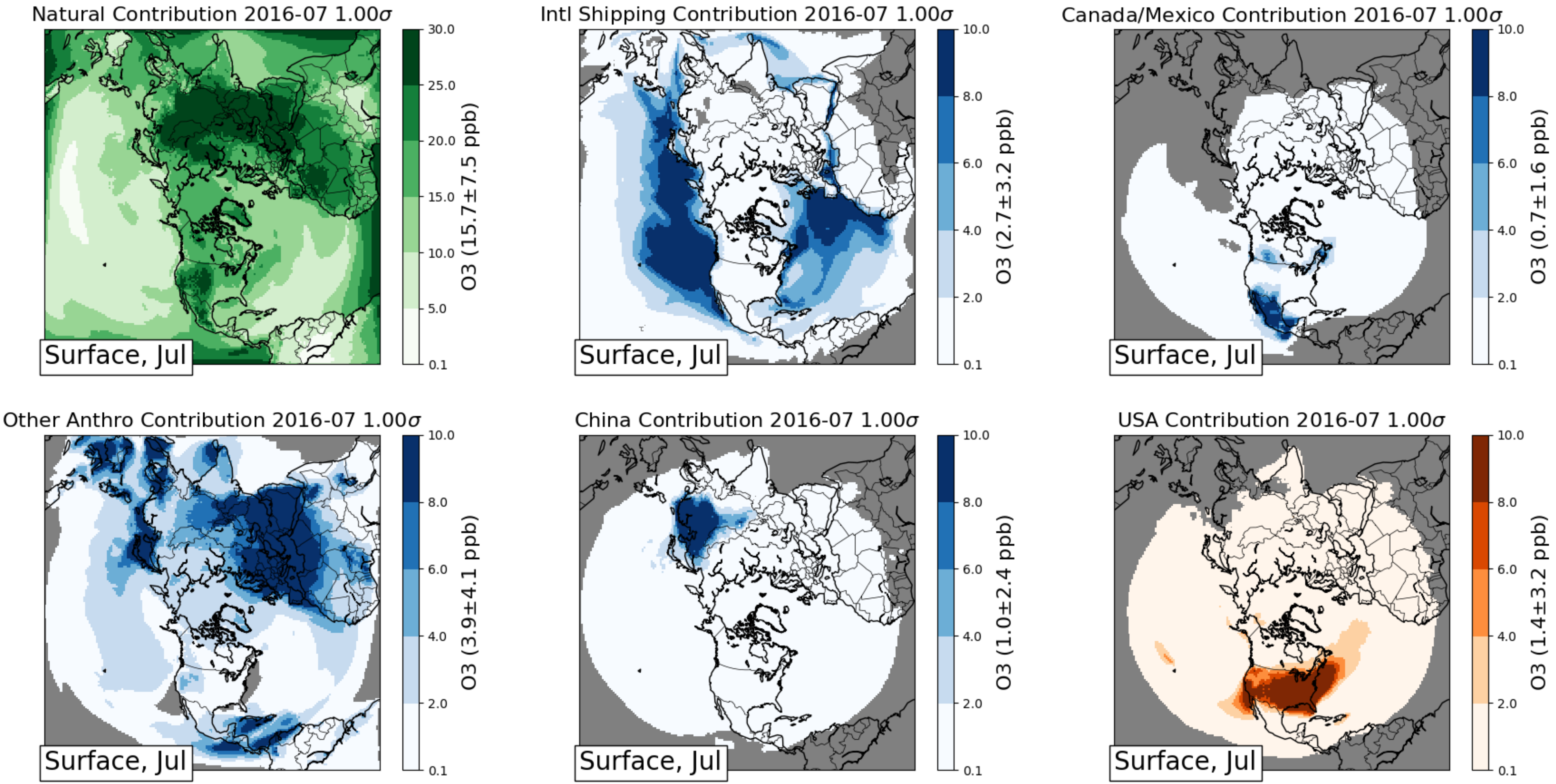


"The only reliable quantitative ozone measurements from the late 19th century were made at Montsouris near Paris where ozone averaged 11 ± 2 ppbv from 1876 to 1910." ... "While these measurements indicate that late 19th century ozone in western Europe was much lower than today, there is no way to know if these values were representative of other surface locations in the NH." - Cooper et al., 2014. doi: 10.12952/journal.elementa.000029



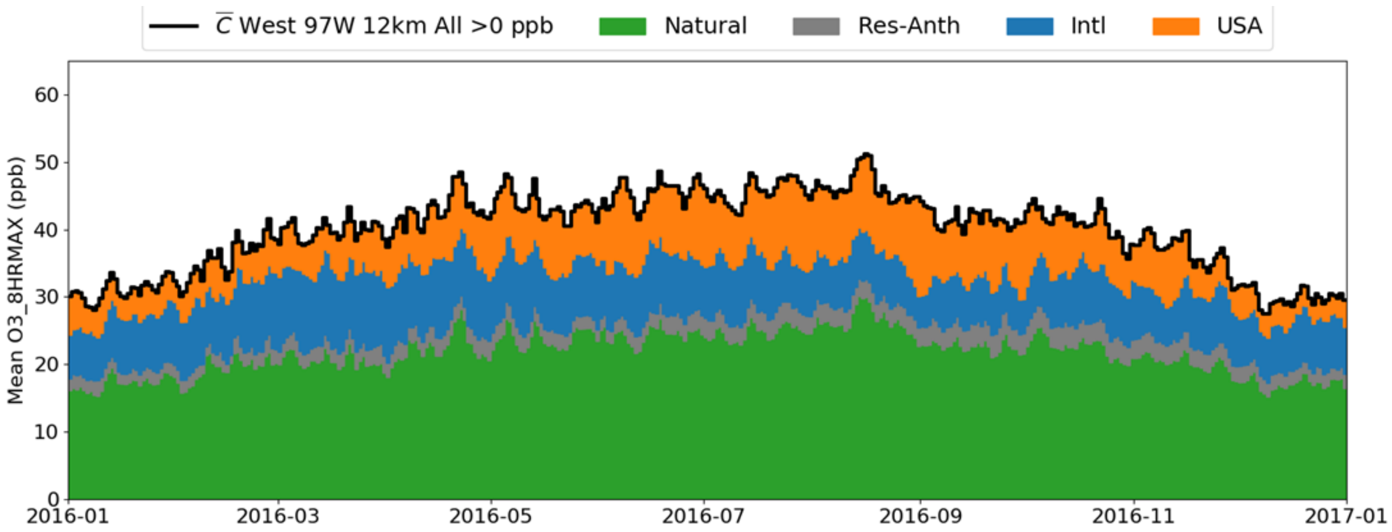


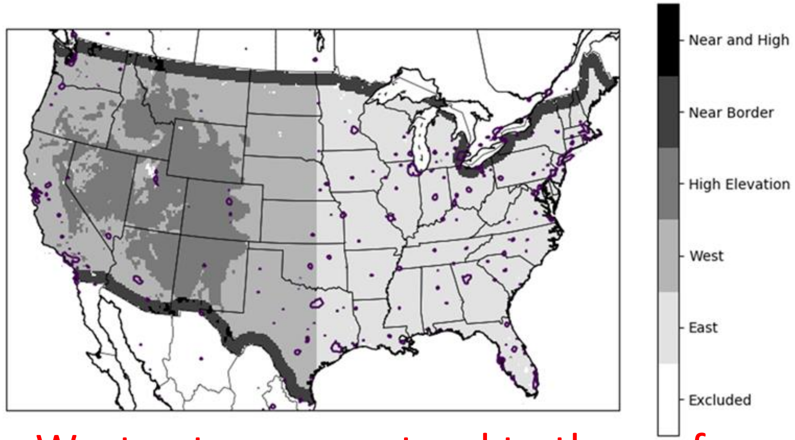
Ozone source contributions in July at the Surface



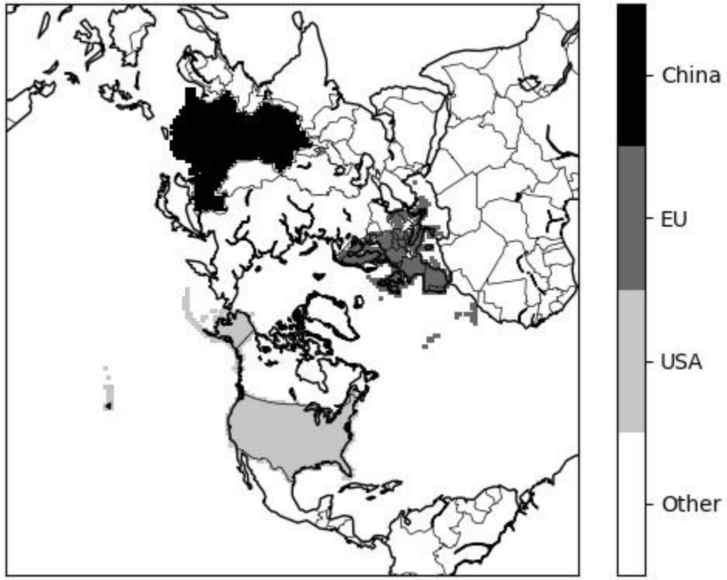
Difference between West and East Surface (12km)

West Contributions

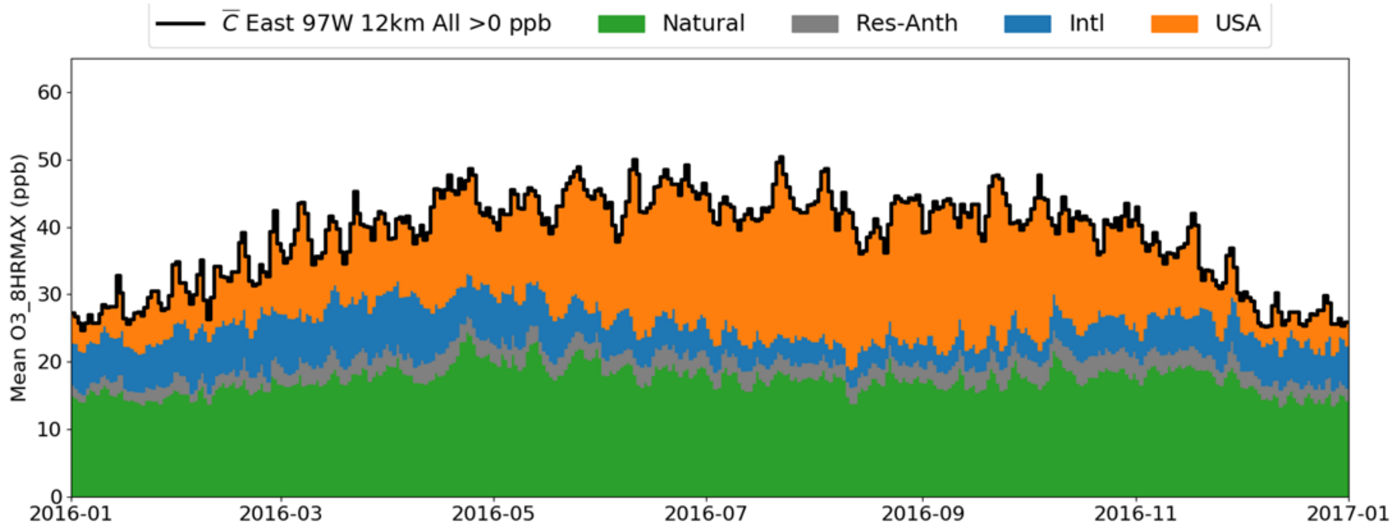




West gets more natural to the surface; think stratosphere.

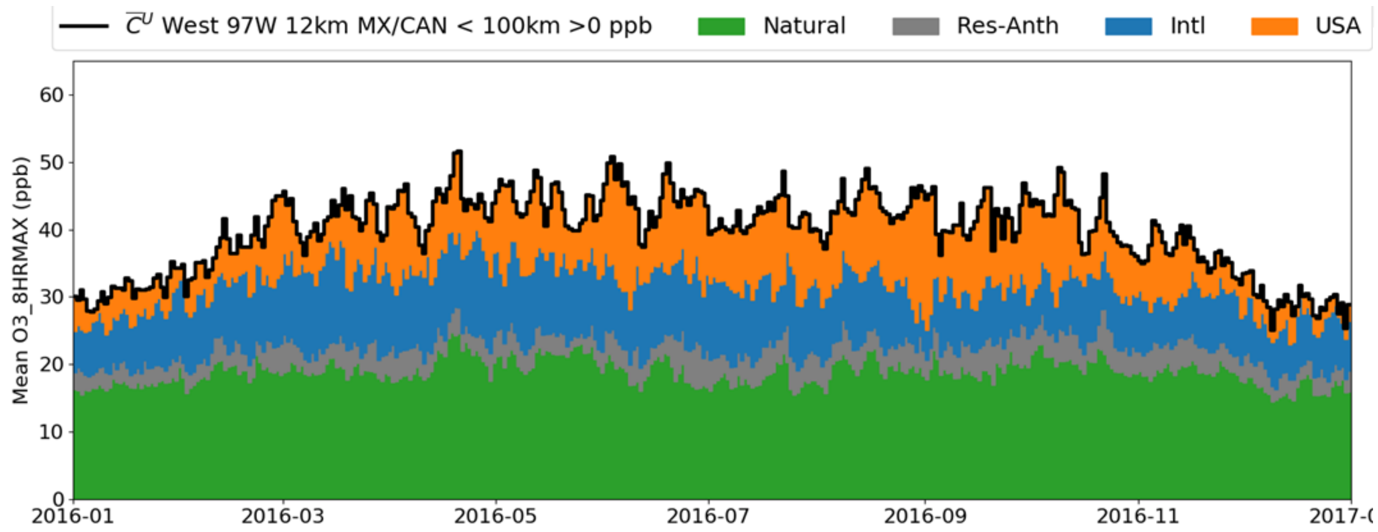


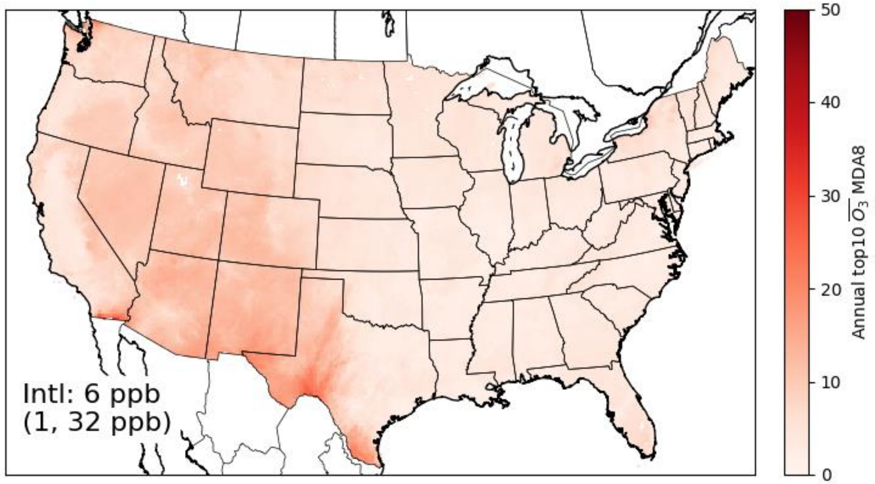
East Contributions

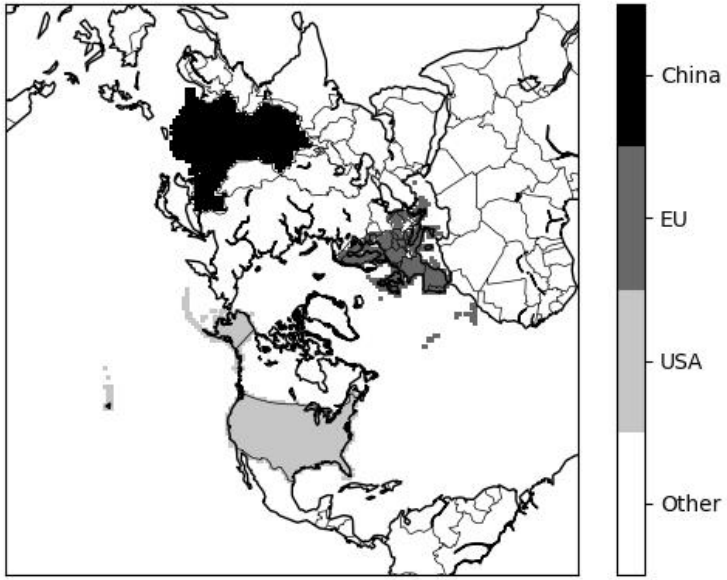


Differences within the West at the Surface (12km)

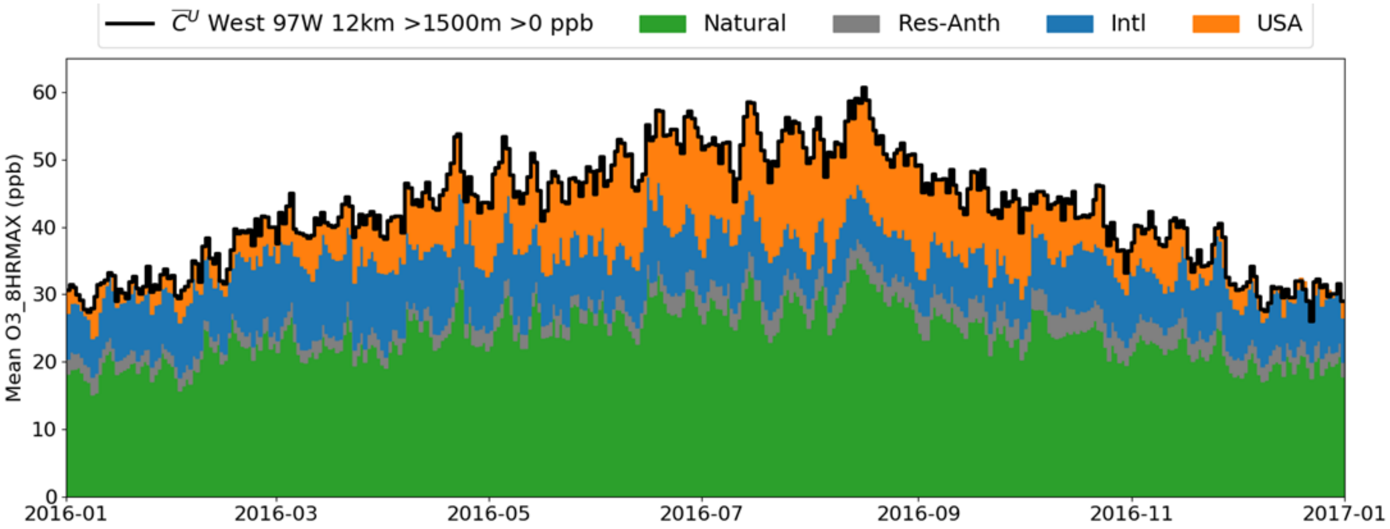
Near Border has consistent international





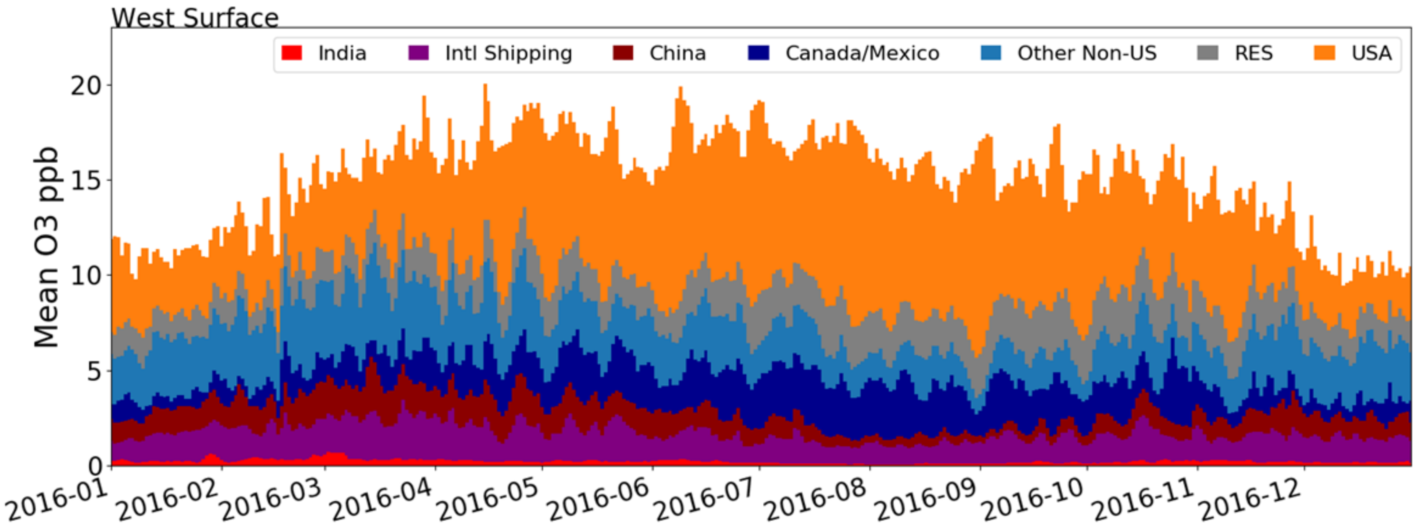


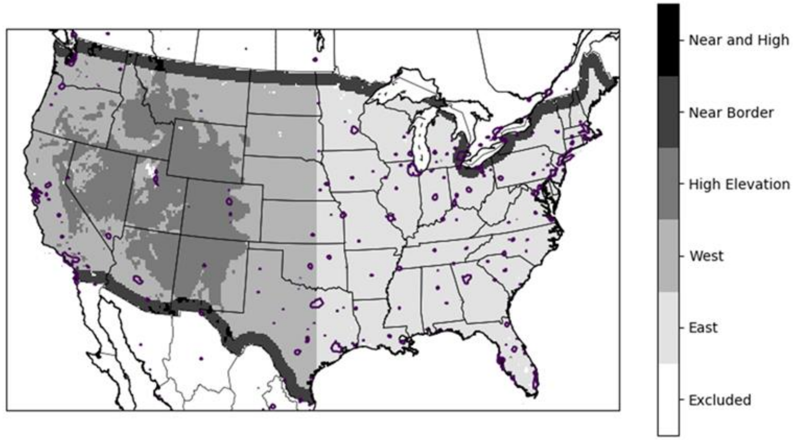
High Elevation more Natural



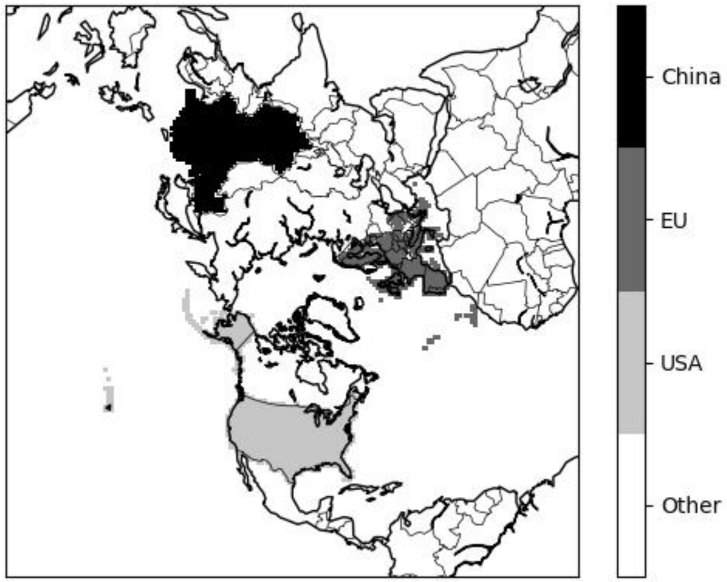
Difference between West and East Surface (108km)

West: Canada increases as long-range decrease

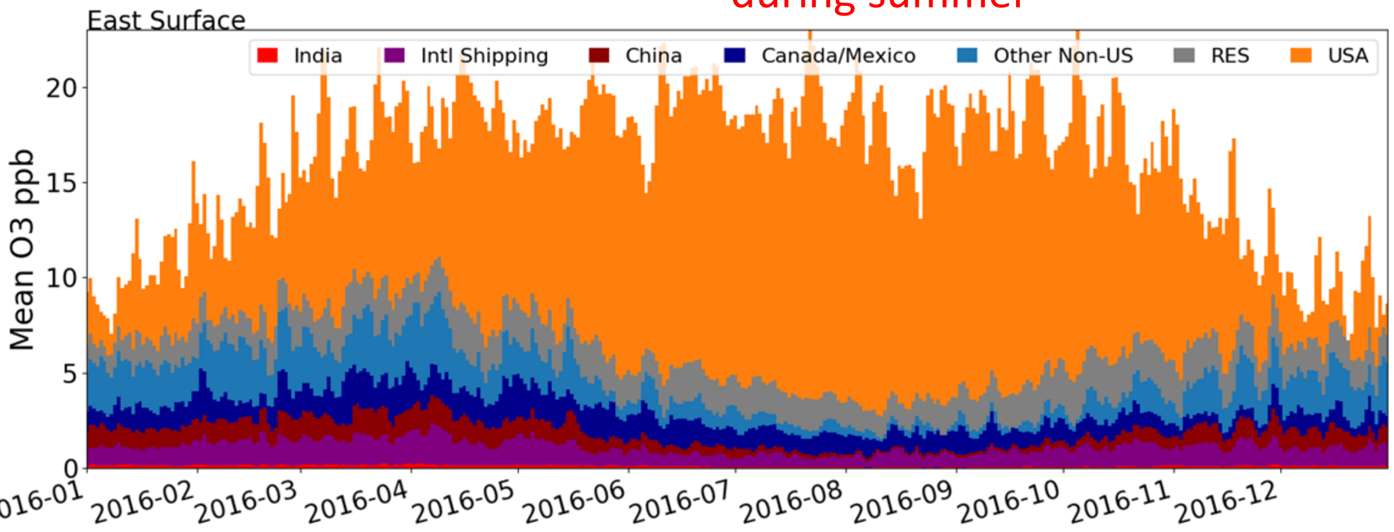




Other countries 2-5 ppb on average during summer



East: International decreases in summer



India/China pop-weighted impact higher, but consistent with West, Horowitz, Fiore doi:10.5194/acp-9-6095-2009, see supplement Tables S1

Summary

- Zero-out simulations provide estimates of contributions
  - Global Natural, International Anthropogenic, Domestic Anthropogenic
  - India, China, International Shipping, more to come
- Generally consistent with the literature
  - HTAP Phase I and Phase II; Jaffe et al. (2018)
  - USB is higher in the West than in the East, USB can be a significant contributor on high ozone days.
  - Long-range transport contributes more in the spring than summer
  - Canada and Mexico operate as short-range transport to most of the West
- Largest West/East difference at the surface was natural
- International Contribution on top 10 days at the surface
  - Summer most places: 1-15 ppb
  - Near-border: up to 30 ppb (no bias correction)
  - Eastern US decreases from all sources in summer
  - Western US increases from Canada/Mexico