

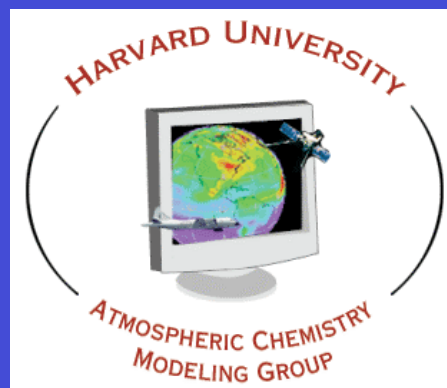
AIR QUALITY STUDIES USING OBSERVATIONS FROM SPACE

PM, NO₂, formaldehyde, glyoxal, CO, ozone

Daniel J. Jacob

with Easan Drury (now at NERL), Folkert Boersma (now at KNMI),
Dylan Millet (now at U. Minnesota), Tzung-May Fu (now at Hong Kong Polytech U.),
Monika Kopacz, Lin Zhang

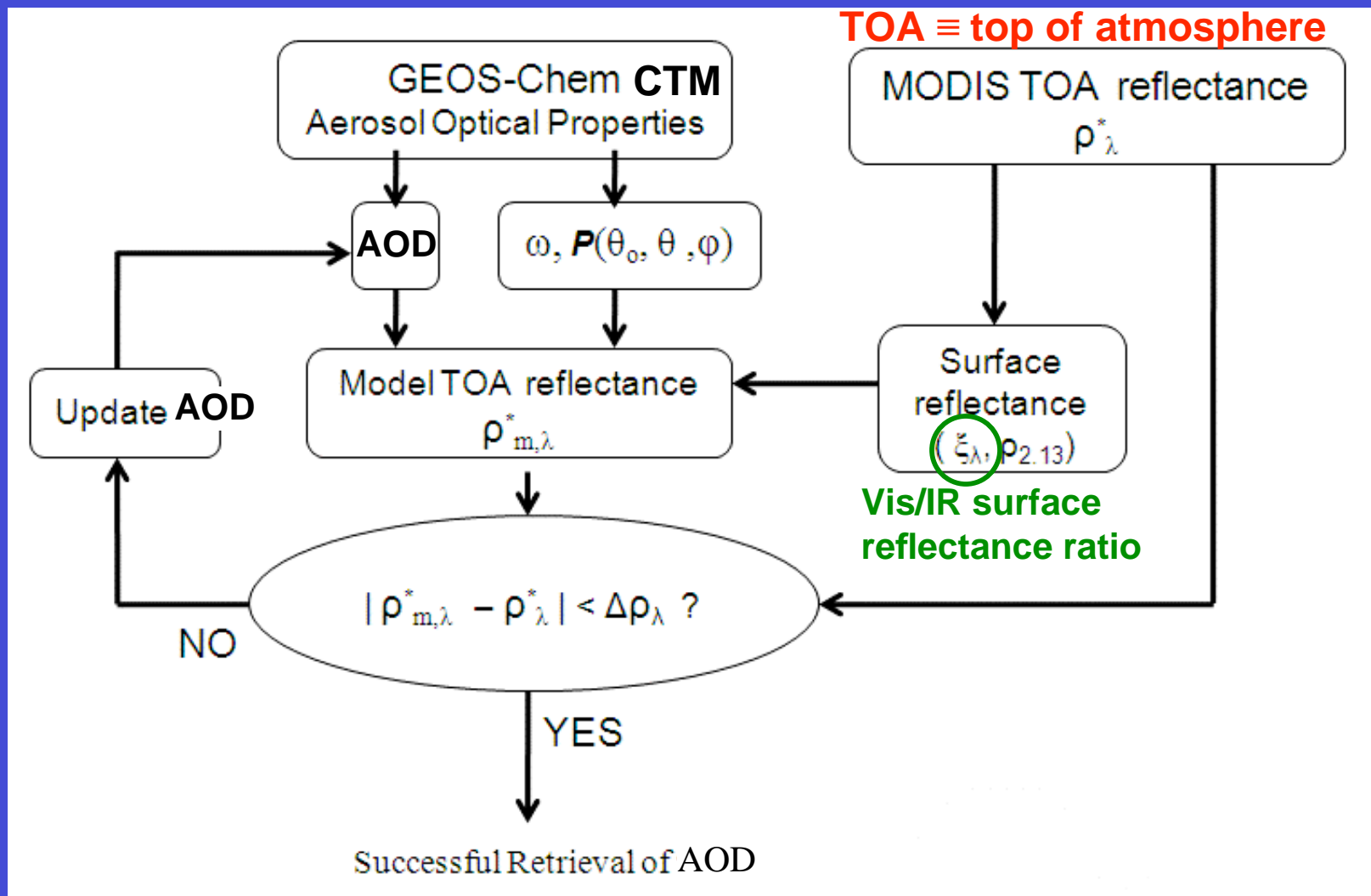
and collaborations with Harvard/SAO: Kelly Chance, Thomas Kurosu,
Xiong Liu, Rob, Spurr



and funding from NASA ACPMAP, NASA GTCP, EPRI

IMPROVED MODIS AOD RETRIEVAL ALGORITHM

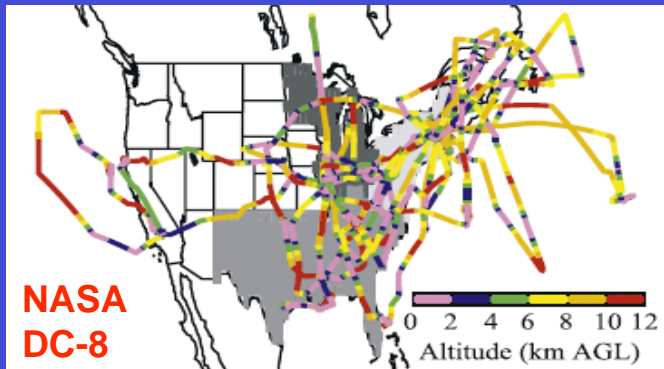
resolves local variability in surface reflectance, aerosol properties;
enables quantitative comparison to CTM



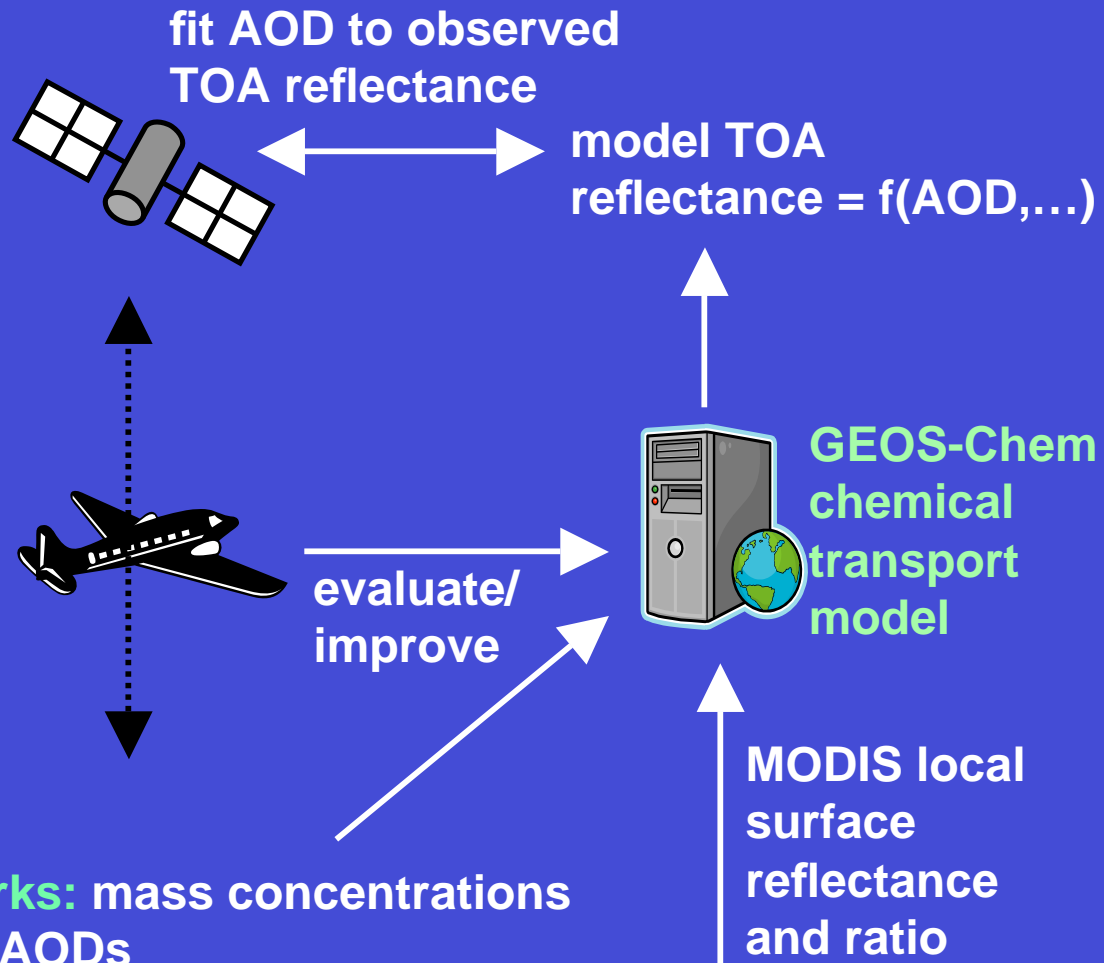
APPLICATION TO ICARTT AIRCRAFT MISSION PERIOD (Jul-Aug 2004)

MODIS satellite instrument:
TOA reflectance

NASA, NOAA, DOE aircraft:
speciated mass concentrations,
microphysical & optical properties



EPA AQS/IMPROVE surface networks: mass concentrations
NASA AERONET surface network: AODs



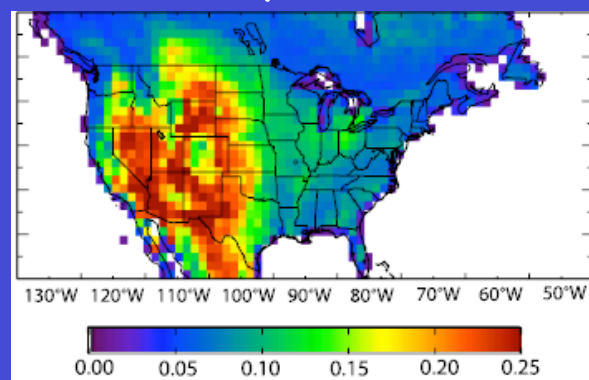
EASTERN U.S.

Drury et al. [JGR , submitted]

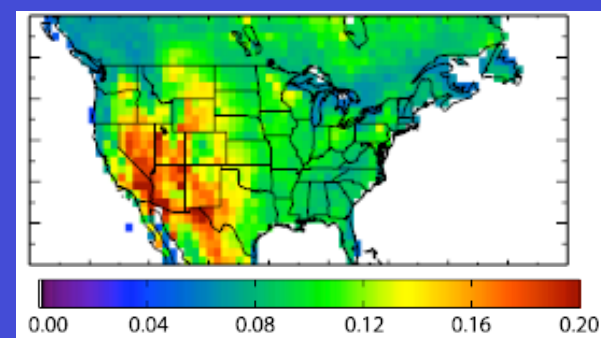
IMPROVING THE SURFACE REFLECTANCE CORRECTION FOR MODIS AEROSOL RETRIEVALS

Measured top-of-atmosphere (TOA) reflectances (Jul-Aug 2004)

2.13 μm

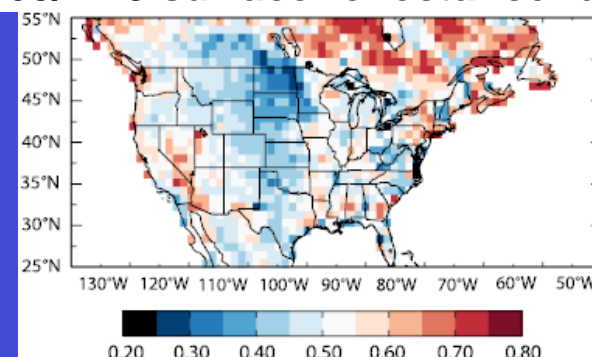
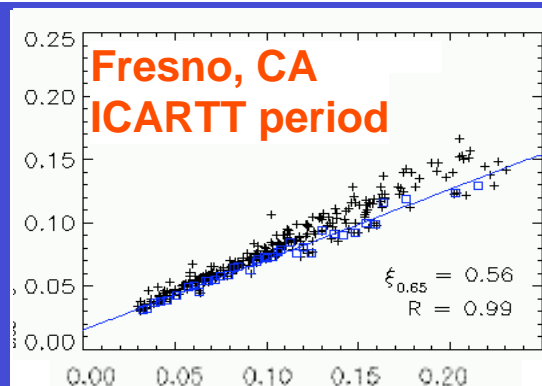


0.65 μm



Measured 0.65 vs. 2.13 TOA reflectances: take lower envelope for given location to derive surface reflectance ratio

0.65/2.13 surface reflectance ratio

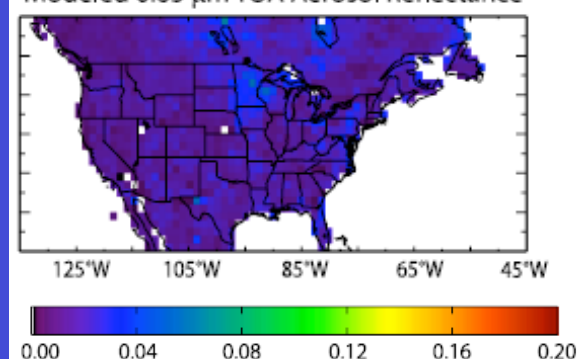


Remove molecular reflectance

Derive aerosol reflectance at 0.65 μm (same procedure for 0.47 μm)

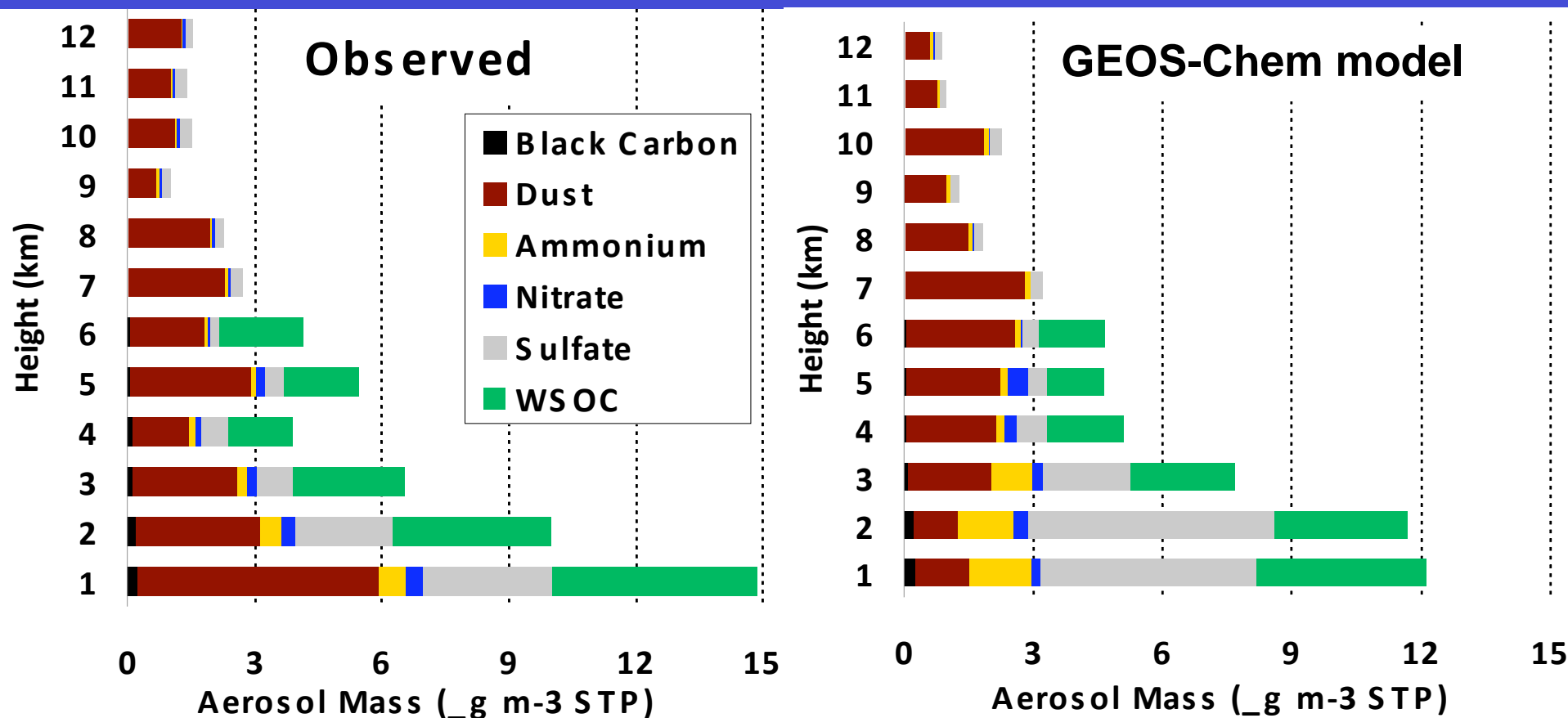
Drury et al. [JGR 2008]

Modeled 0.65 μm TOA Aerosol Reflectance



MEAN AEROSOL VERTICAL PROFILES IN ICARTT

NASA DC-8 and NOAA WP-3D

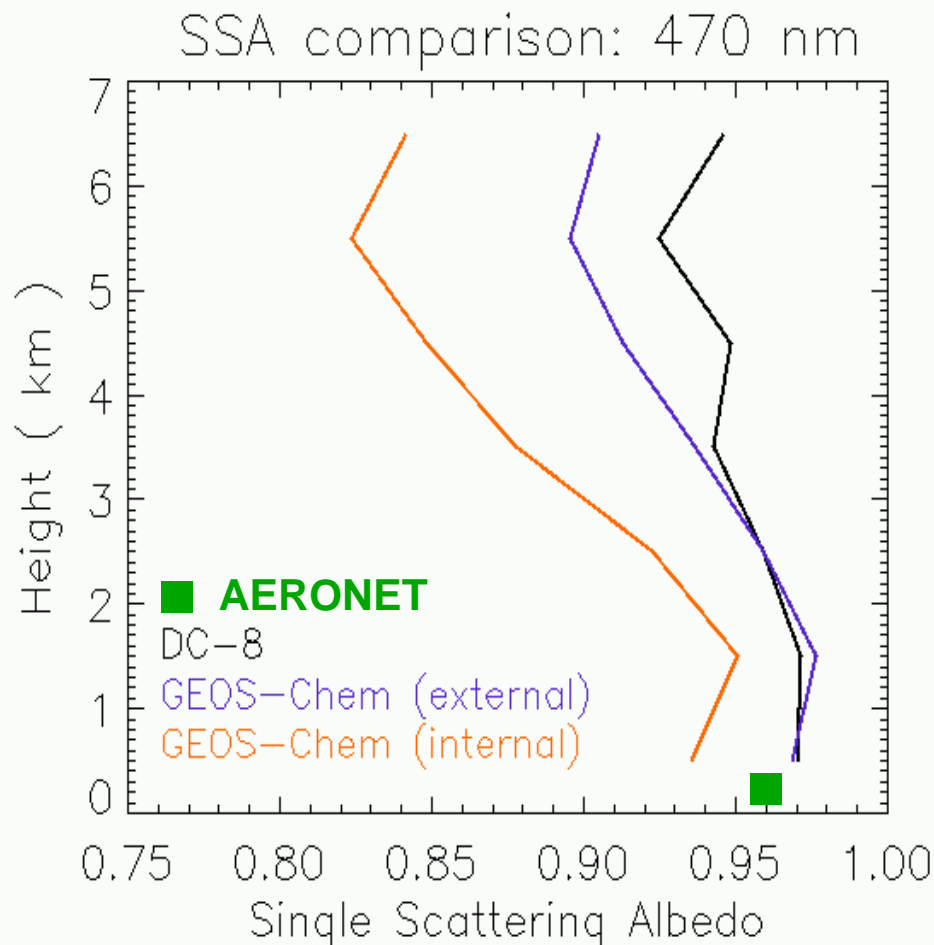


- Bulk of mass is in boundary layer below 3 km: mostly sulfate, organic
- Dust, organic dominate above 3 km

Drury et al. [JGR , submitted]

AEROSOL OPTICAL PROPERTIES IN ICARTT

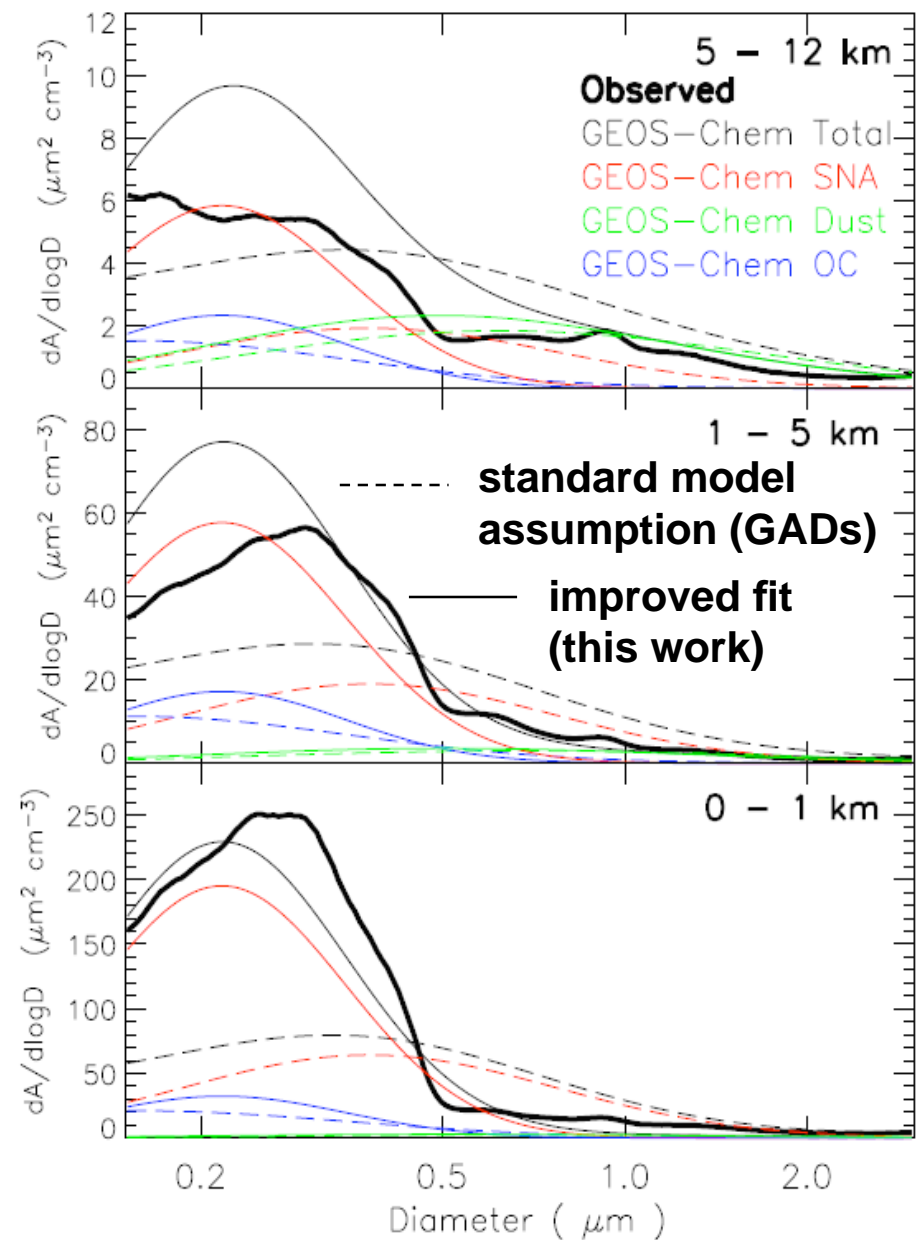
Single-scattering albedo



- External mixture is better assumption
- Narrow sulfate and OC size distributions relative to GADS (σ 2.2 \rightarrow 1.6); decreases 180° backscatter

Drury et al. [JGR , submitted]

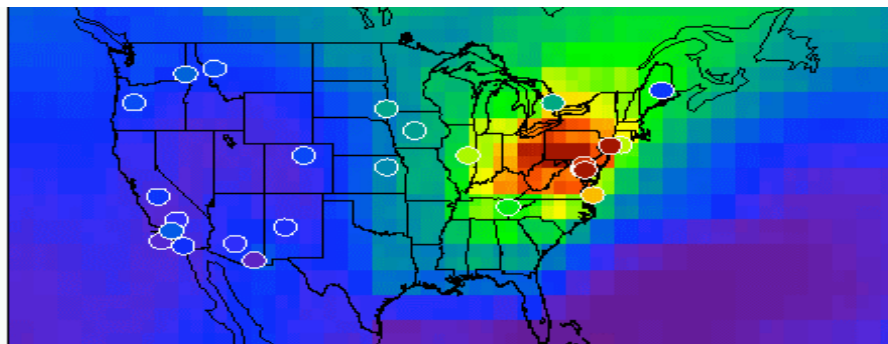
Size distributions



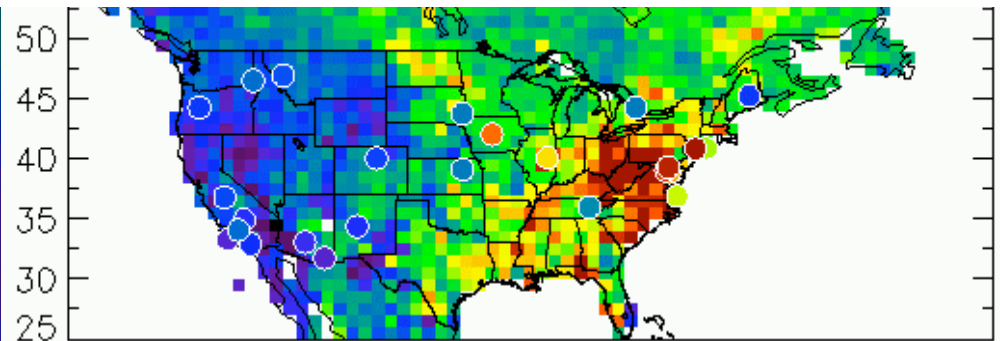
AEOSOL OPTICAL DEPTHS ($0.47\ \mu\text{m}$), JUL-AUG 2004

c004 and c005 are the MODIS operational data; AERONET data are in circles

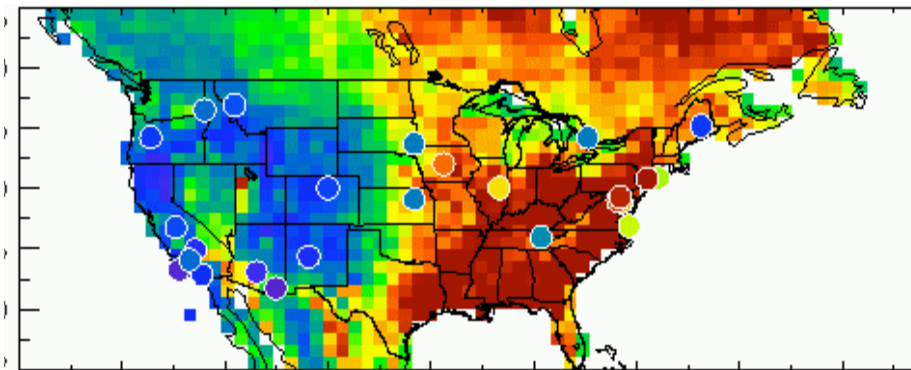
GEOS-Chem model



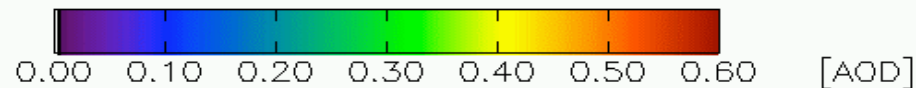
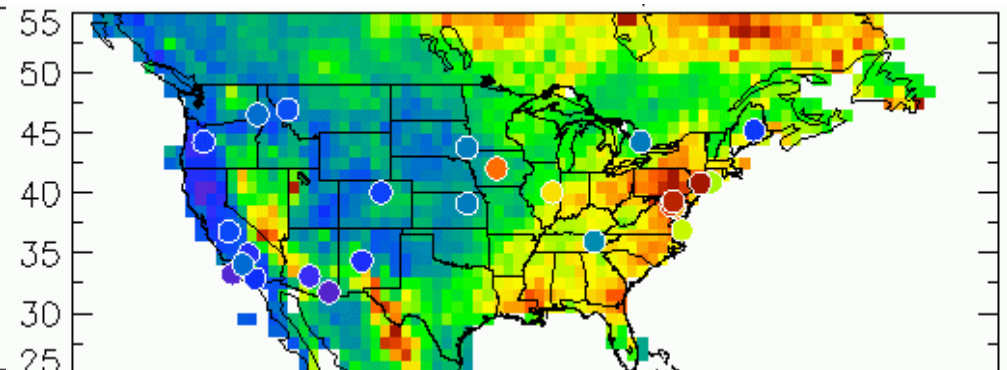
MODIS (this work)



MODIS (c004)



MODIS (c005)



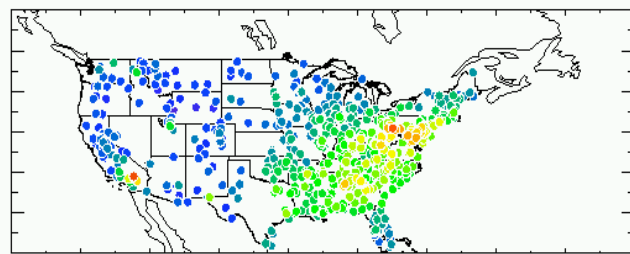
- Beyond improving on the operational products, our MODIS retrieval enables quantitative comparison to model results (consistent aerosol optical properties)
- Results indicate model underestimate in Southeast US – organic aerosol

Drury et al. [JGR , submitted]

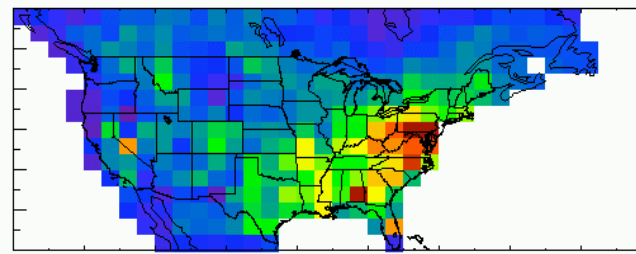
INFERRING $PM_{2.5}$ FROM MODIS AODs

Infer $PM_{2.5}$ from AOD by $PM_{2.5} = AOD * \left[\frac{PM_{2.5}}{AOD} \right]_{\text{GEOS-Chem}}$

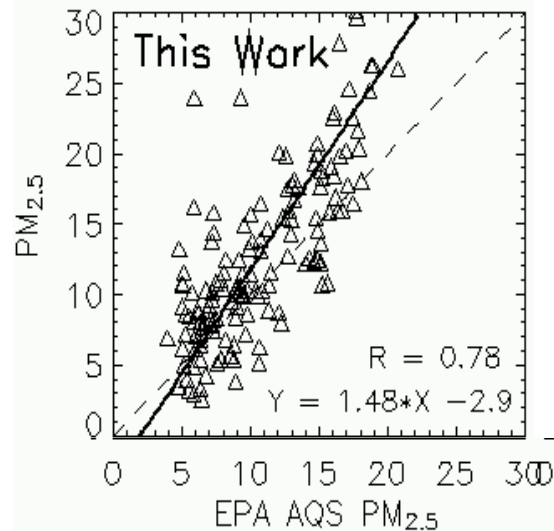
EPA AQS surface network data



MODIS $PM_{2.5}$ (this work)



Jul-Aug 2004

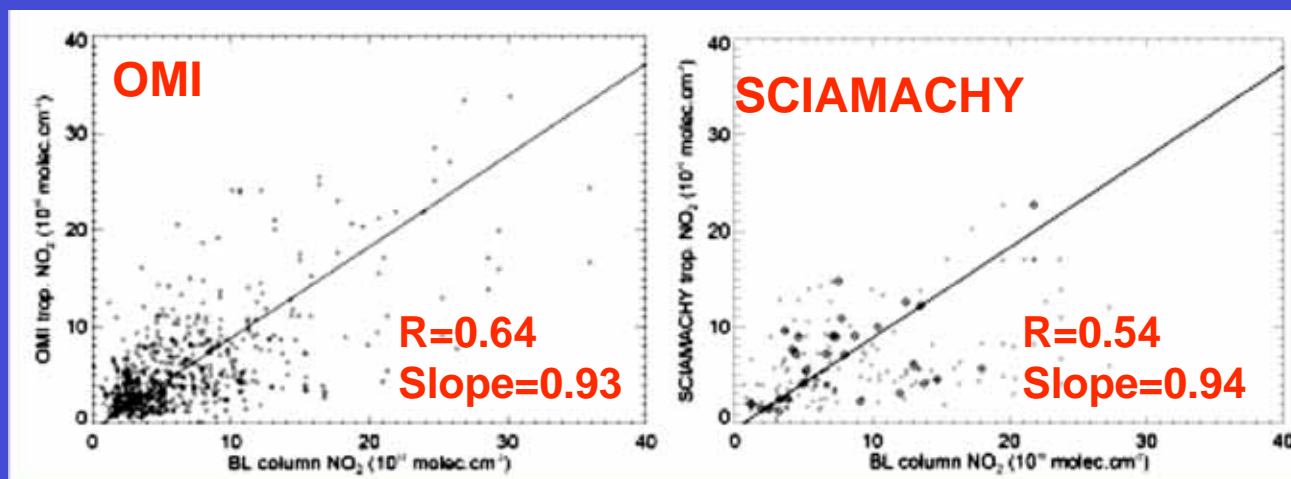
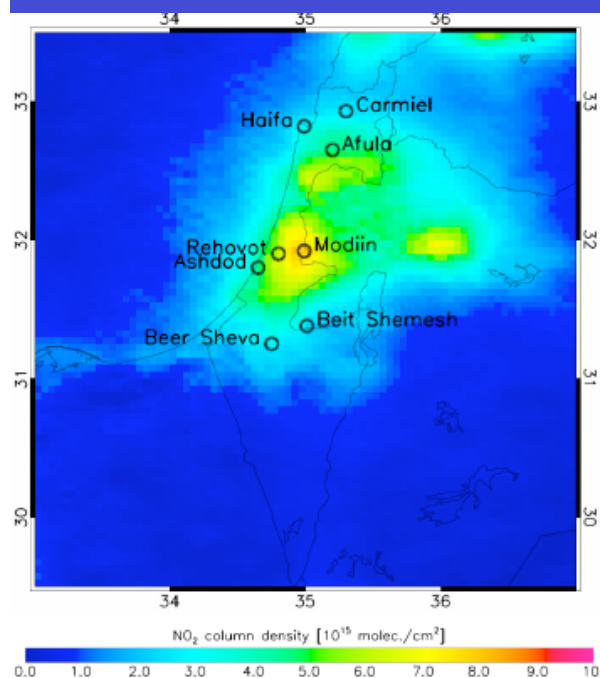


Bias in source regions due to clear-sky sampling?

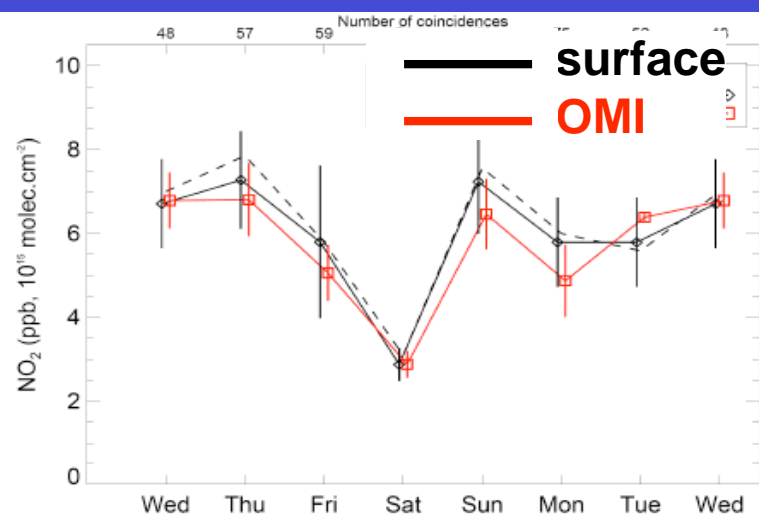
Drury et al. [JGR , submitted]

VALIDATING TEMPORAL NO₂ VARIABILITY IN SATELLITE DATA USING SURFACE NO₂ DATA FROM URBAN ISRAEL SITES

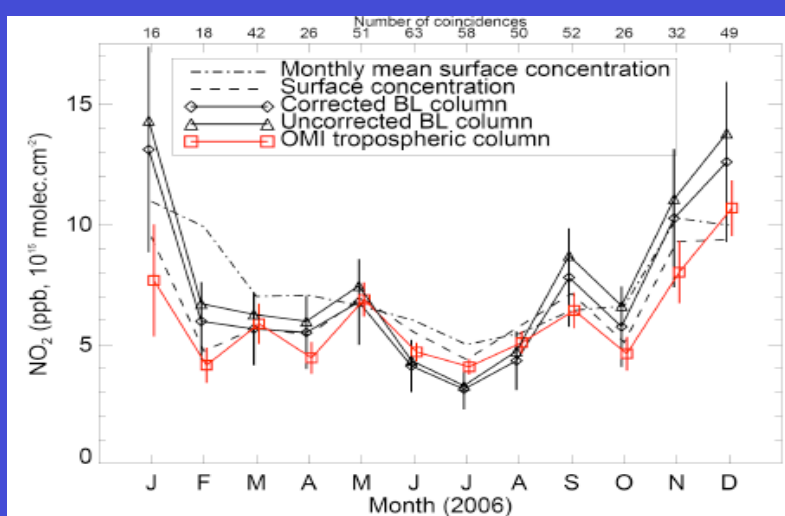
Satellite vs. in situ daily data (2006)



Weekly cycle



Seasonal cycle

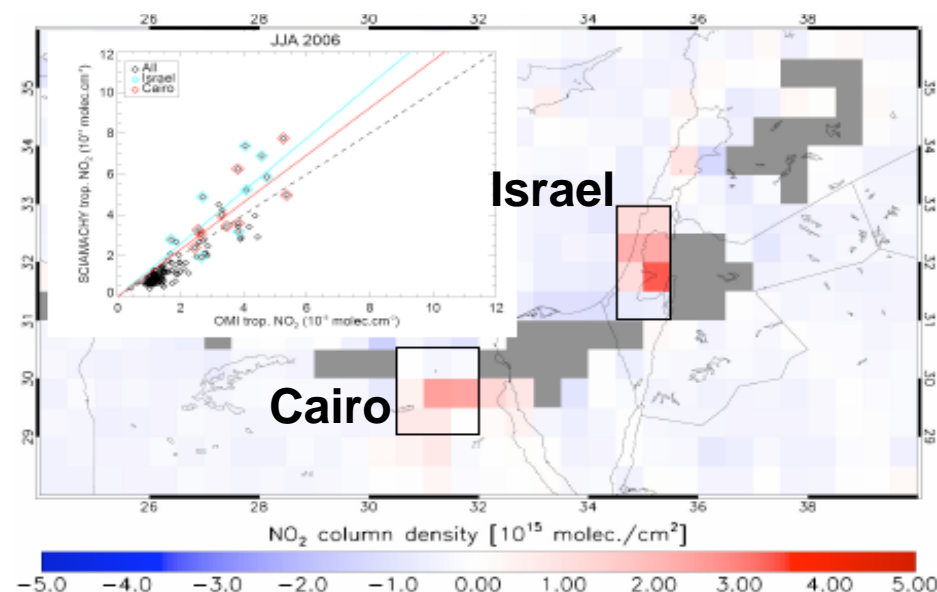
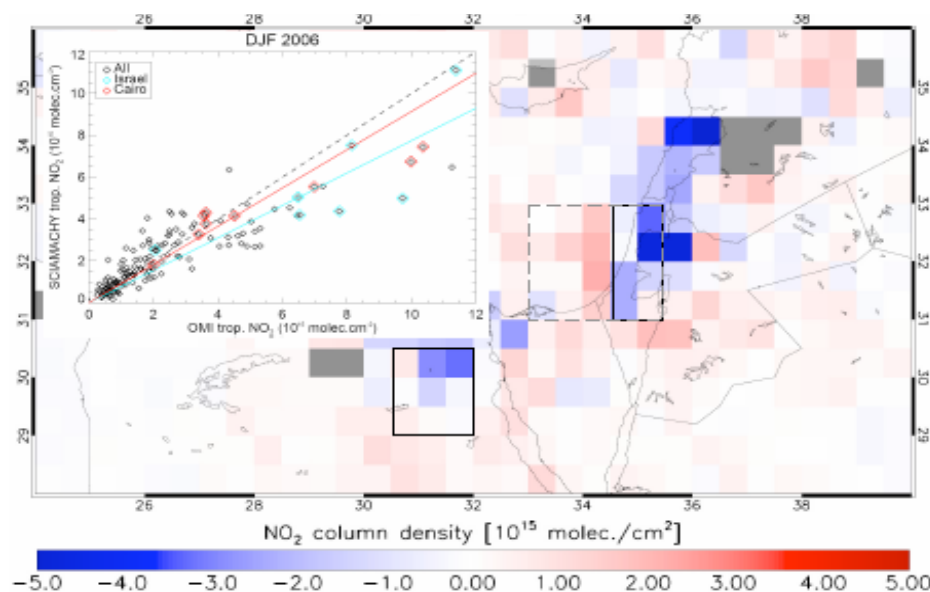


Boersma et al.
[ACP, in press]

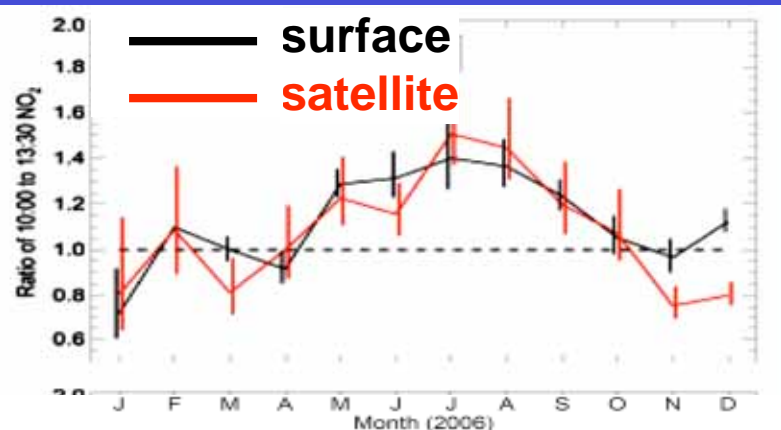
SEASONAL VARIATION OF THE DIURNAL DIFFERENCE BETWEEN SCIAMACHY (10 am) AND OMI (1:30pm)

SCIAMACHY – OMI difference: DJF 2006

JJA 2006



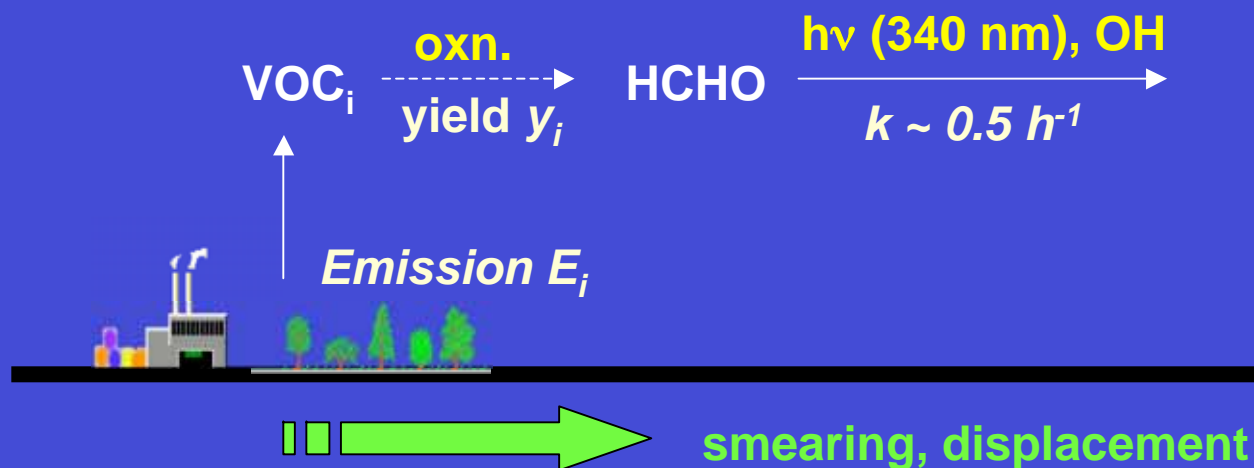
Seasonal variation of 10:00 to 13:30 NO₂ ratio at urban Israel sites



- maximum vehicle use in daytime would result in higher NO₂ at 13:30 than at 10:00;
- This is more than compensated in summer by high chemical loss in daytime

Boersma et al. [ACP in press]

RELATING FORMALDEHYDE COLUMNS TO VOC EMISSION

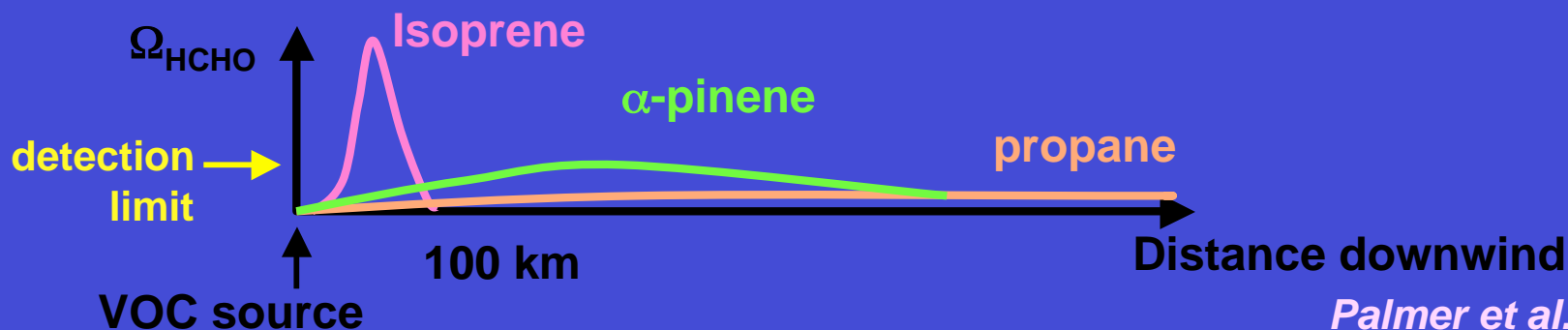


In absence of horizontal wind, mass balance for HCHO column Ω_{HCHO} :

$$\Omega_{\text{HCHO}} = \frac{\sum_i y_i E_i}{k}$$

Local linear relationship
between HCHO and E

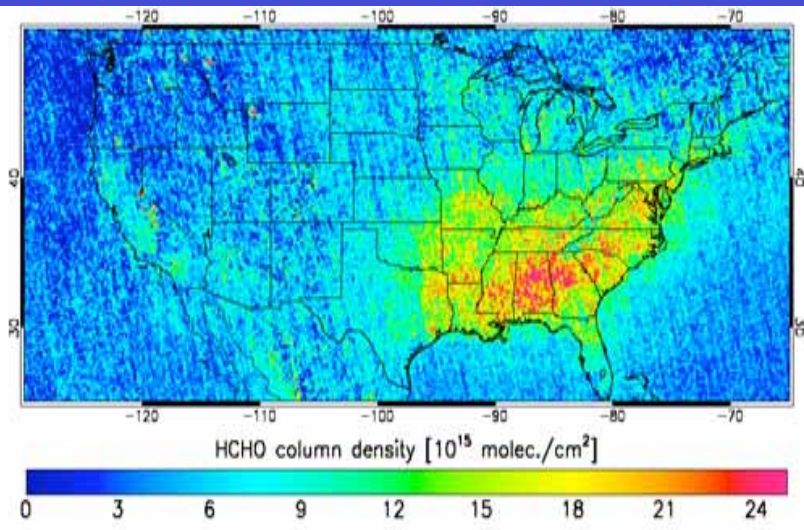
... but wind smears this local relationship between Ω_{HCHO} and E_i depending on the lifetime of the parent VOC with respect to HCHO production:



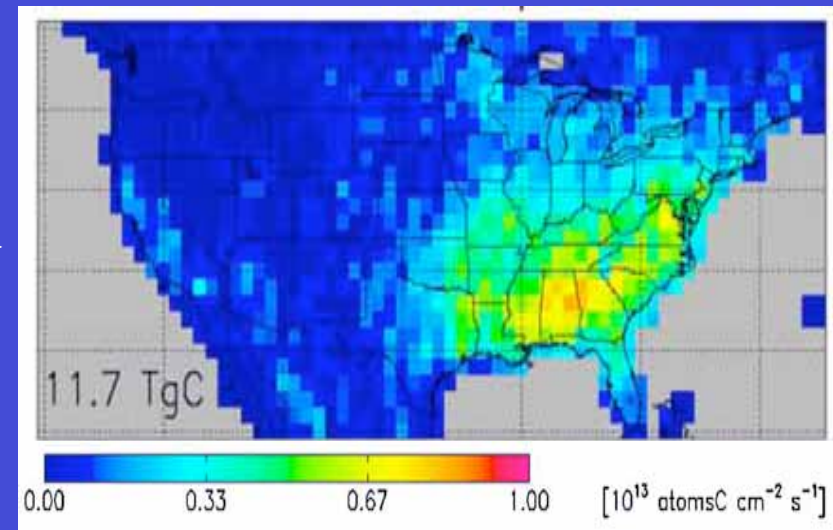
Palmer et al. [2003]

ISOPRENE EMISSION INFERRED FROM OMI

Mean OMI formaldehyde column (JJA 2006)

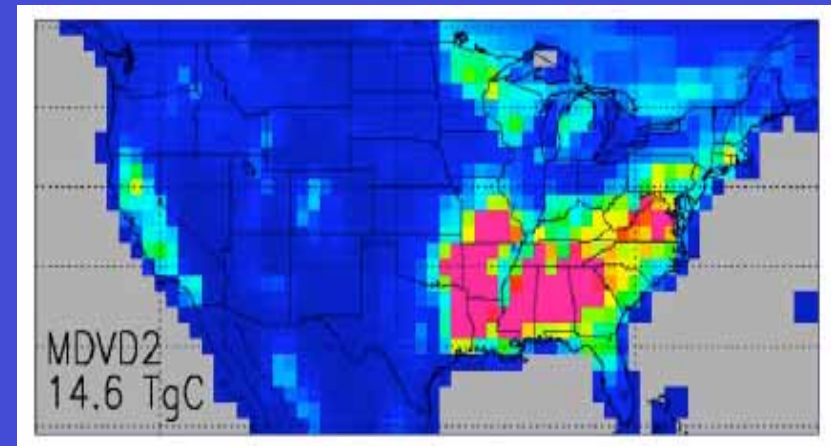


Isoprene emission



...compare to MEGAN bottom-up inventory:

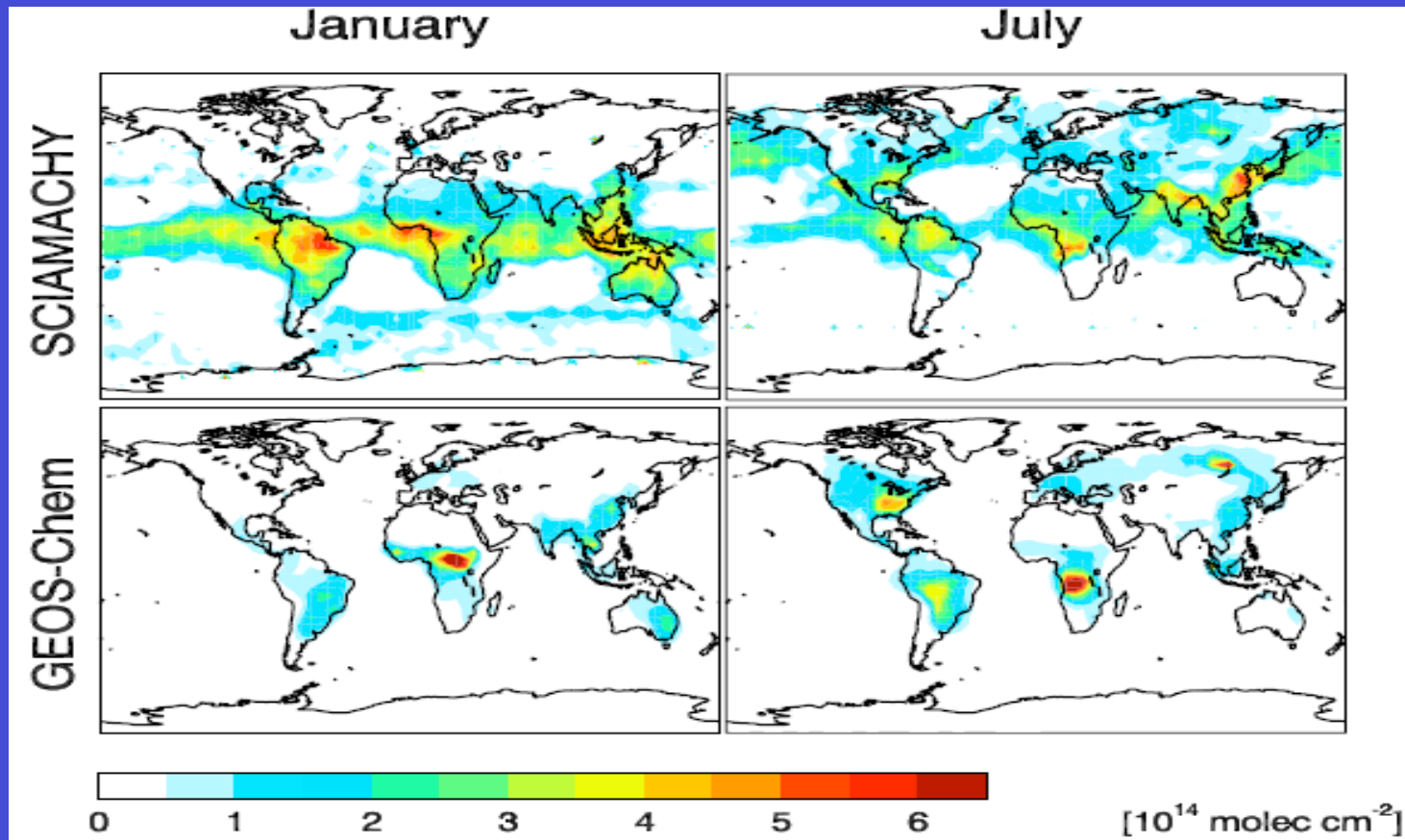
- OMI constrains isoprene emission with uncertainty of 40% (single retrieval)
- We find that MEGAN isoprene emissions are too high over US by 25% ...implies 40% downward correction of broadleaf source
- “Isoprene volcano” in the Ozarks appears to be a myth



Millet et al. [2008]

GLYOXAL COLUMNS: WHAT DO THEY MEAN?

Monthly mean 10 LT glyoxal columns for Jan and Jul 2006



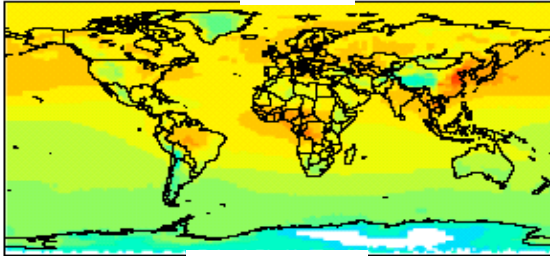
- In model, main sources of glyoxal are isoprene oxidation and biomass burning; glyoxal is of most interest for SOA formation
- SCIAMACHY data are roughly consistent (higher by 50%) but show high values over tropical oceans that suggest a large marine source

Fu et al. [2008]

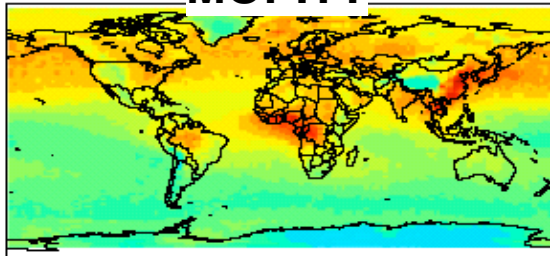
Annual mean CO column
May 2004- April 2005

Using the GEOS-Chem model adjoint to optimize CO sources using multi-sensor data

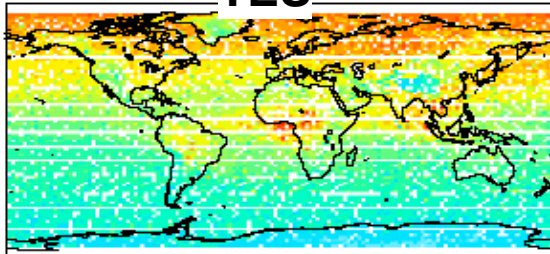
AIRS



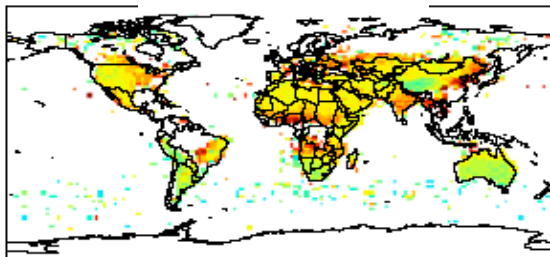
MOPITT



TES

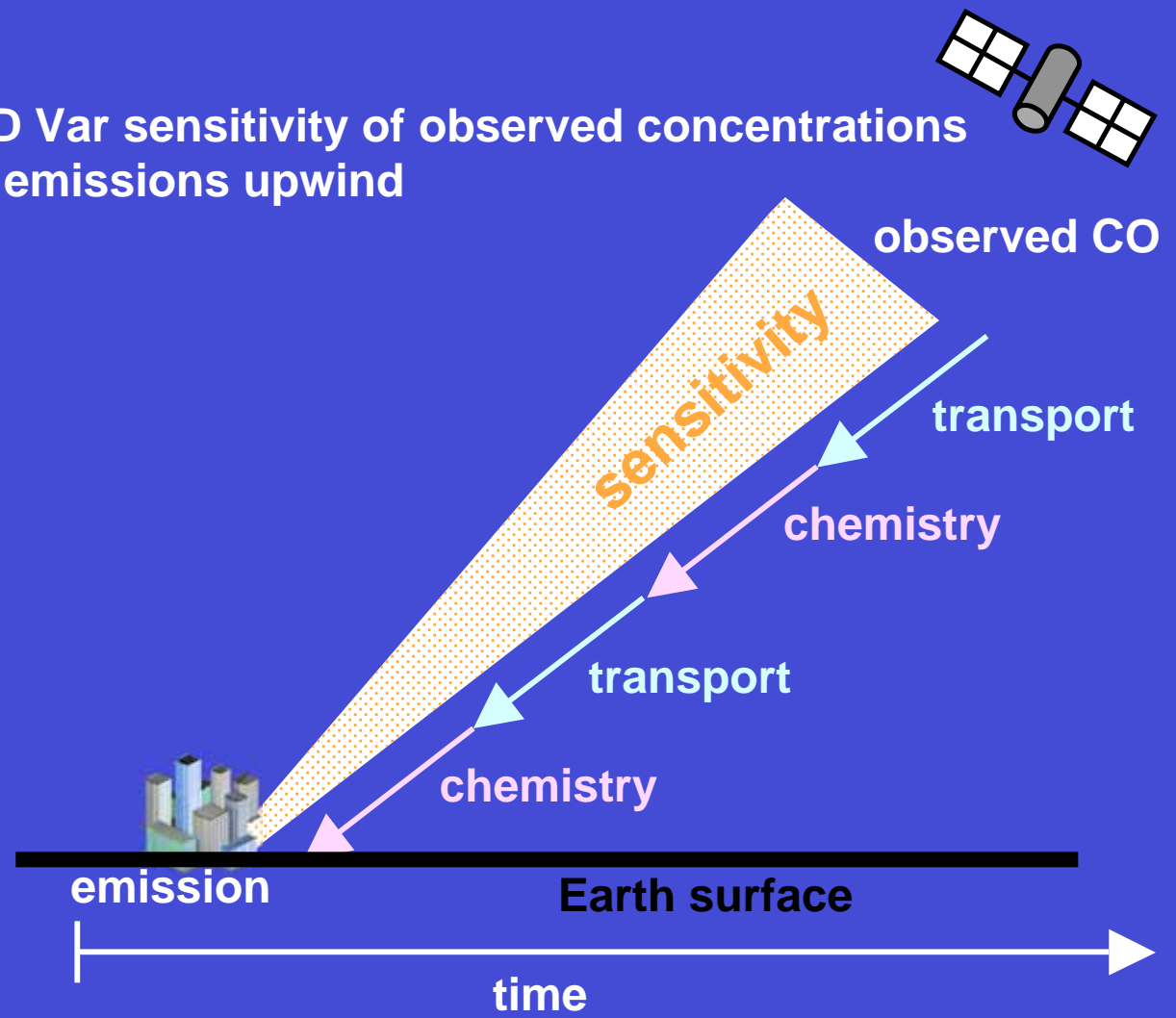


SCIAMACHY



0.00 0.88 1.75 2.62 3.50 10^{18} molec/cm²

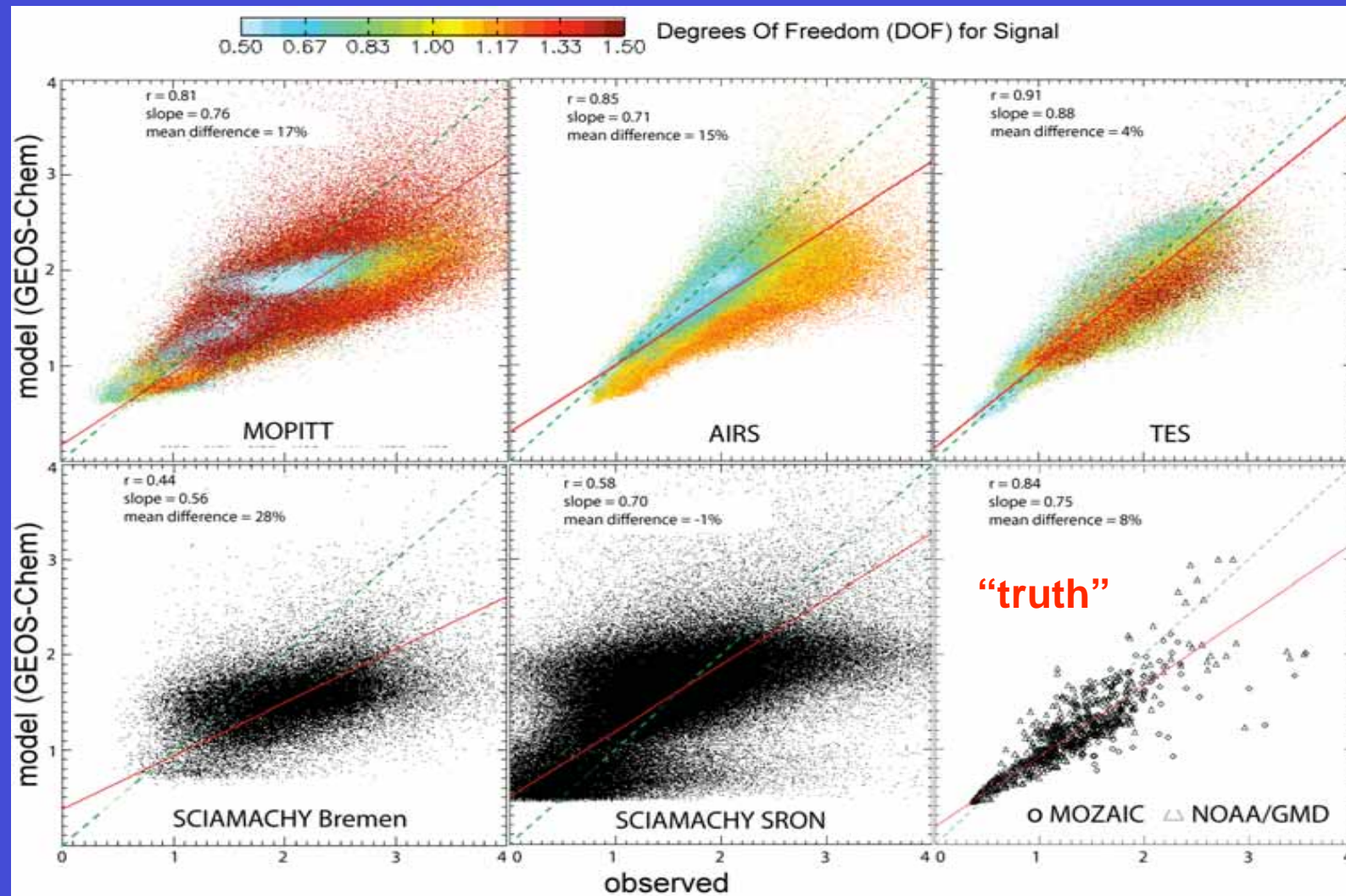
4-D Var sensitivity of observed concentrations
to emissions upwind



Kopacz et al. (in prep.)

EVALUATING CONSISTENCY BETWEEN SATELLITE DATA SETS

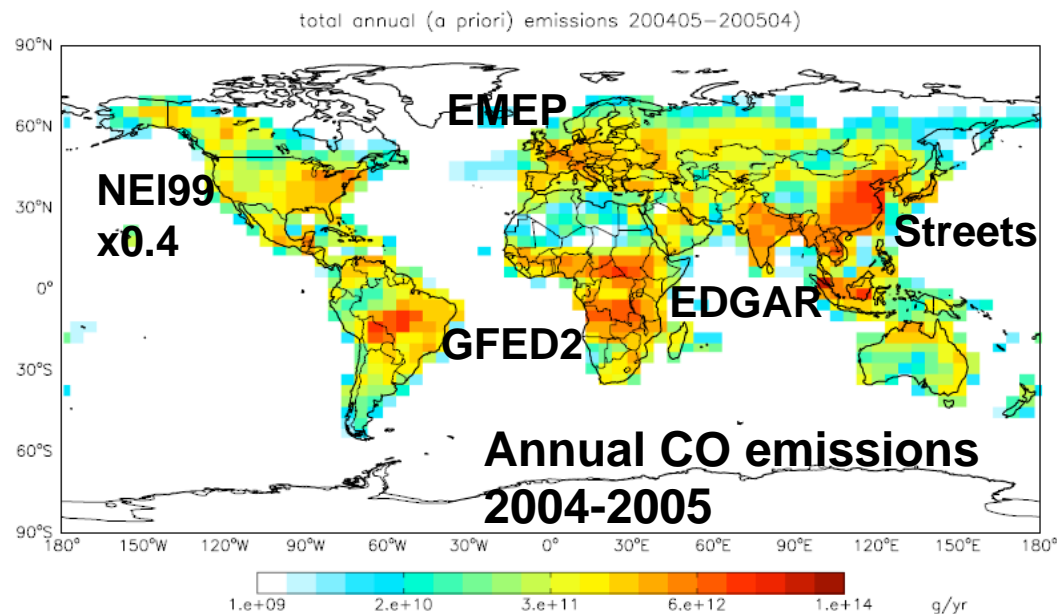
Global (2°x2.5°) correlation of daily data with GEOS-Chem, May 2004 –April 2005



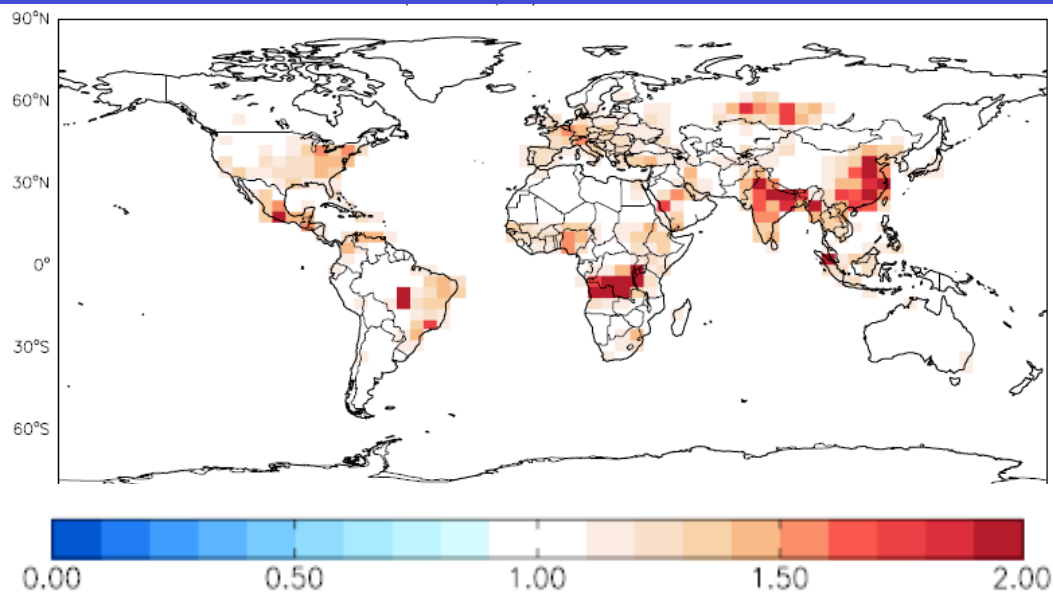
Model allows intercomparison of instruments with different sensitivities and viewing cycles; results show good consistency

Kopacz et al., in prep.

Best prior estimate from current inventories



Correction factors from adjoint inversion



INVERSE MODEL RESULTS

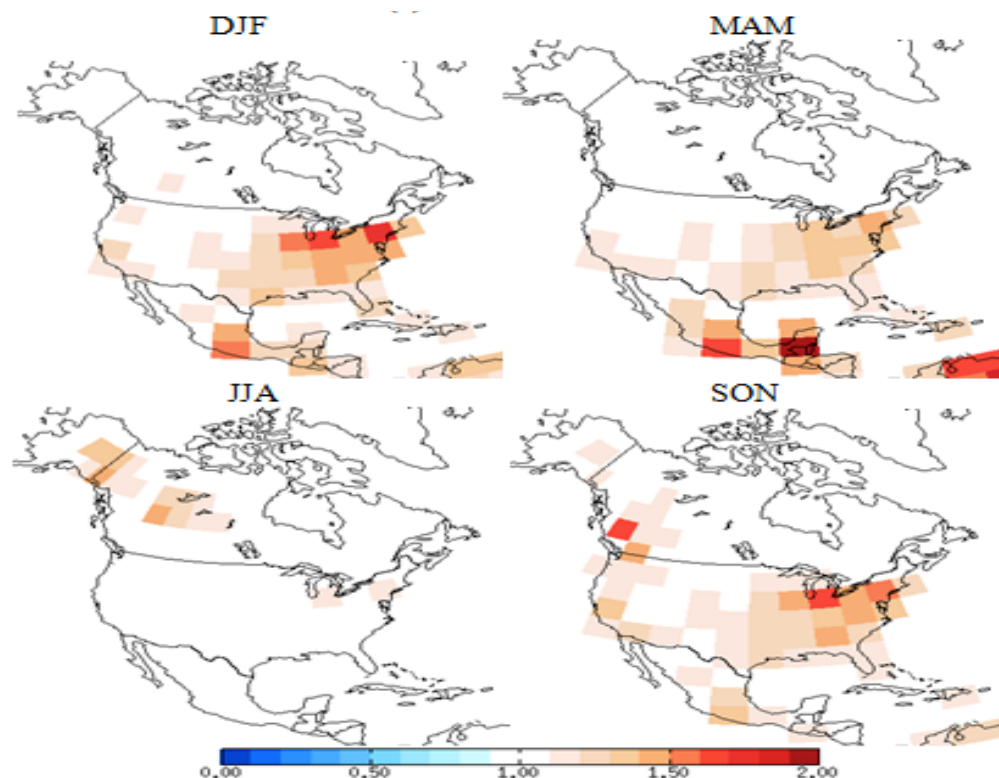
1. Use AIRS, MOPITT, SCIAMACHY-Bremen in adjoint inversion;

2. Use TES, NOAA/GMD, MOZAIC for independent evaluation of inversion results

Prior inventories underestimate emissions, particularly at northern mid-latitudes in winter

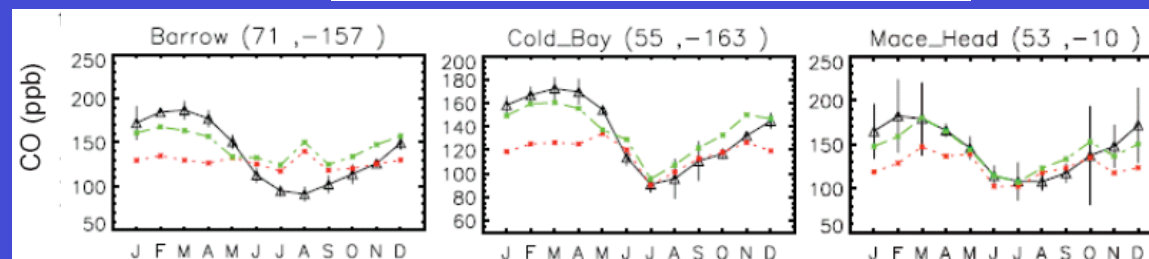
Kopacz et al. (in prep.)

CORRECTION FACTOR IN US: SEASONAL VARIATION



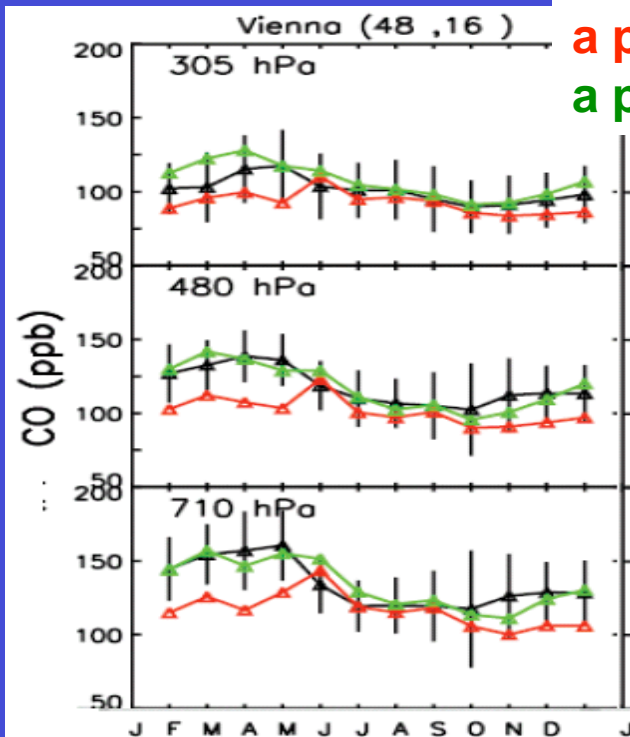
Kopacz et al., in prep.

**GMD
data**



MOZAIC data

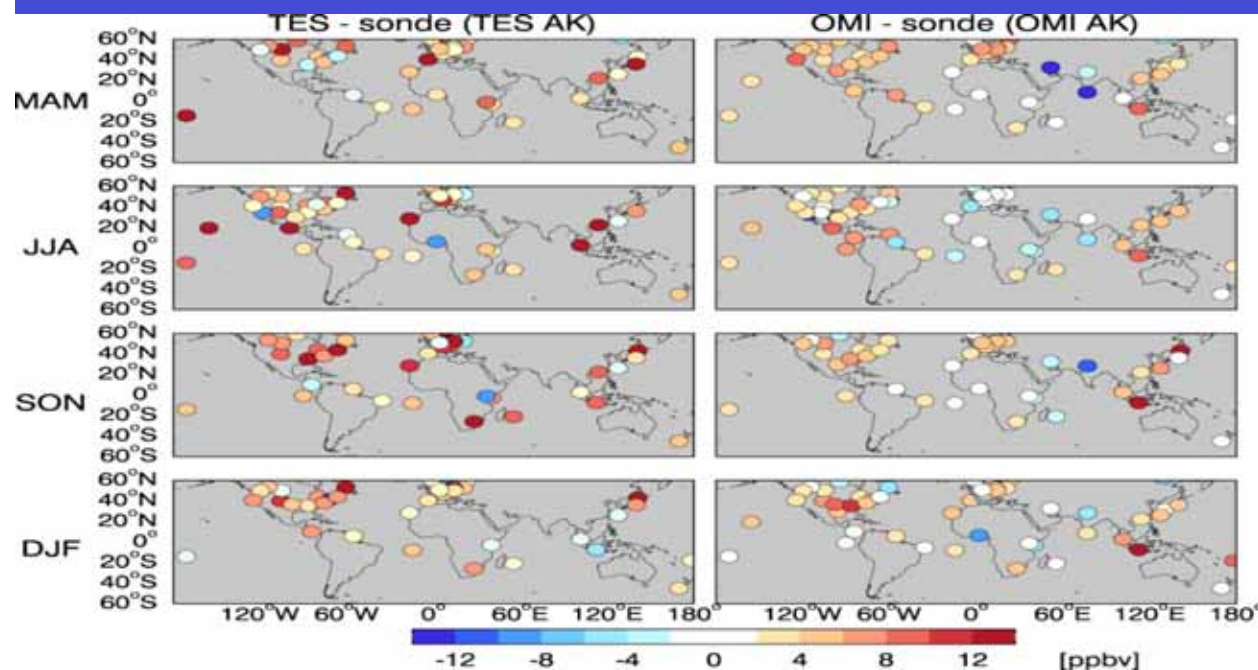
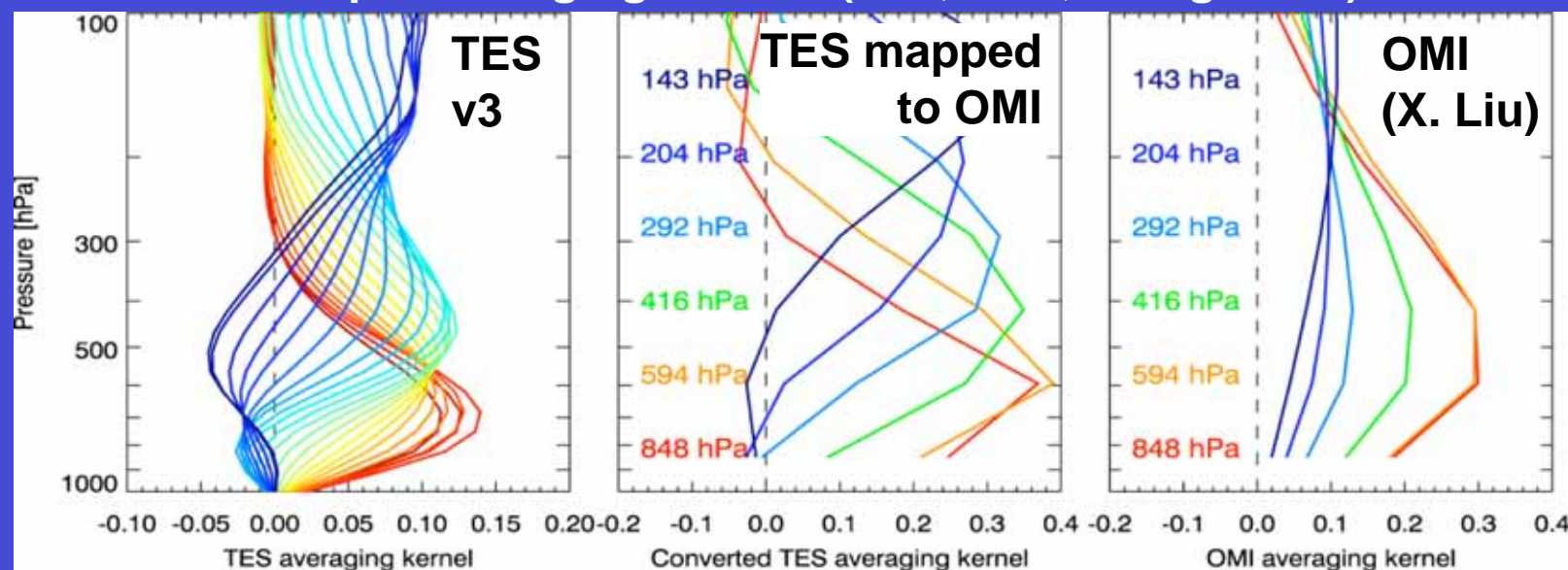
observed
a priori
a posteriori



- Best prior estimate (EPA NEI99 reduced by 60% on basis of ICARTT) is OK in summer when ICARTT was flown
- Underestimate of emissions from cold vehicle starts in winter?

INTERCOMPARISON OF TES AND OMI TROPOSPHERIC OZONE

Sample averaging kernels (28N, 58W, 6 Aug. 2006)



500 hPa bias relative to ozonesondes (2005-2007):

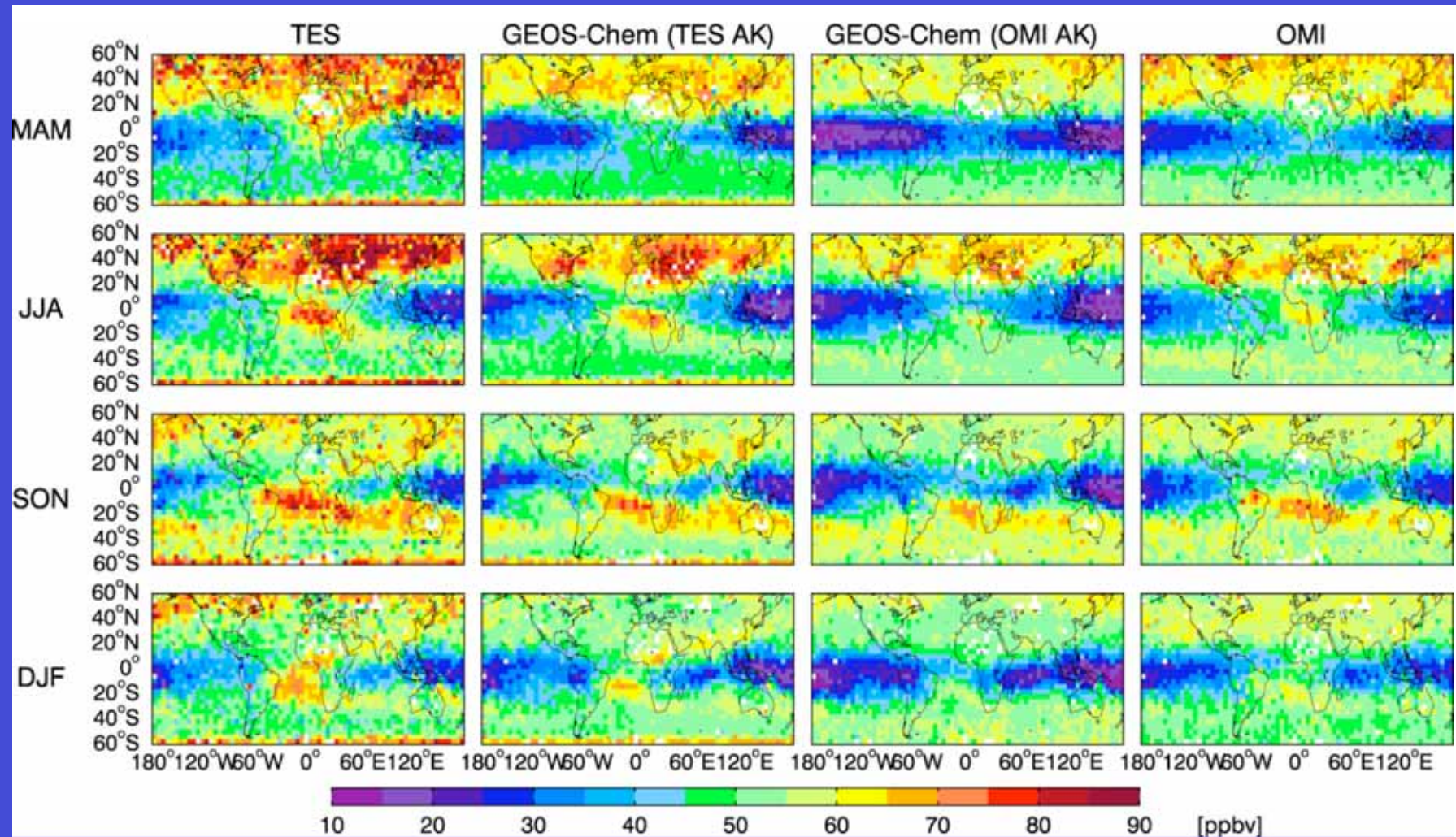
TES: 5.4 ± 7.0 ppbv

OMI: 2.5 ± 5.5 ppbv

Zhang et al. , in prep.

GEOS-Chem AS INTERCOMPARISON PLATFORM

2006 data at 500 hPa; TES and OMI reprocessed to fixed *a priori*

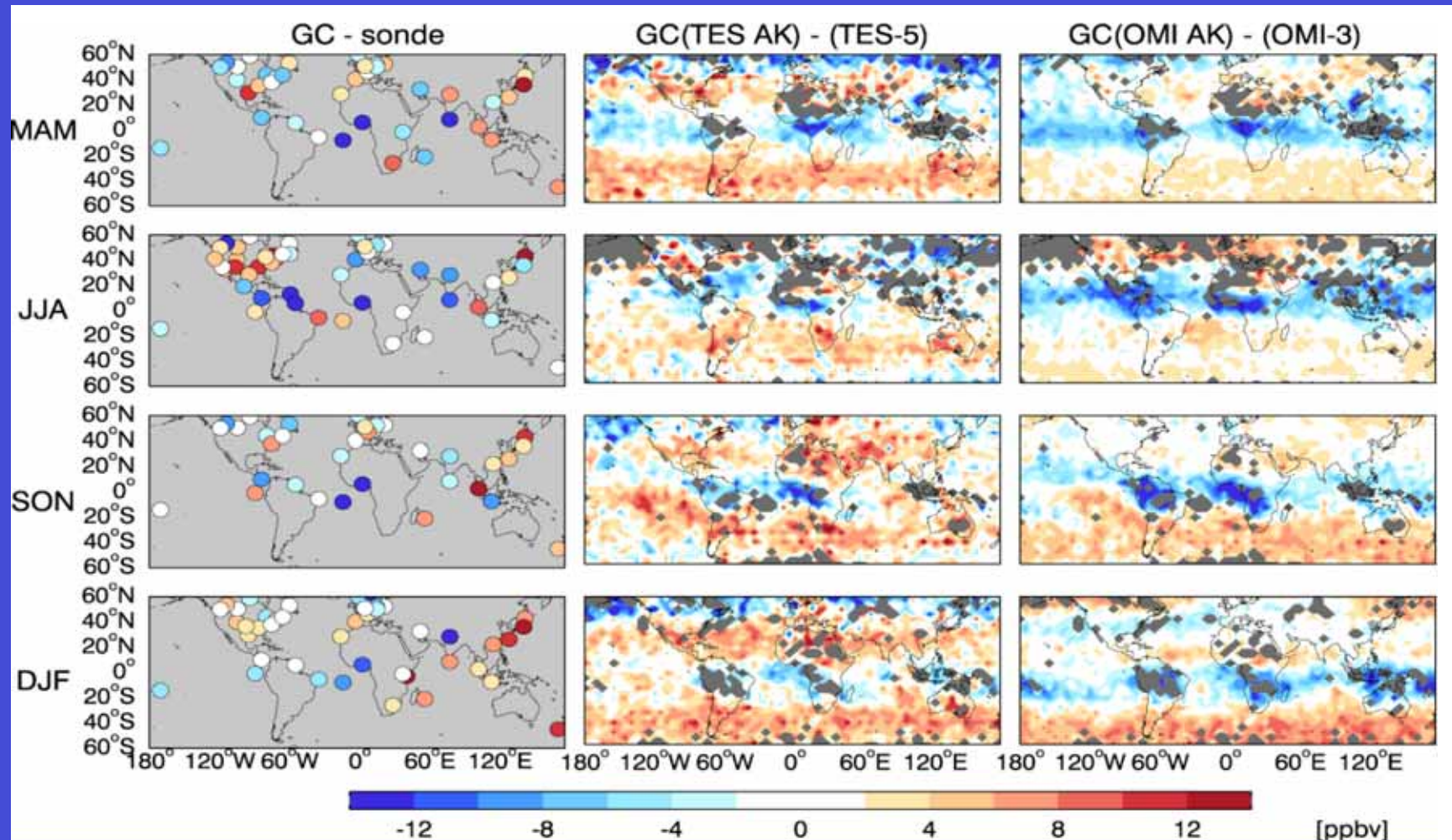


- Higher sensitivity of TES at low altitudes – cf. N. African burning signal
- TES and OMI inconsistent in summer mid-latitudes, some tropical regions

Zhang et al. , in prep.

USING SATELLITE DATA DIAGNOSE MODEL ERRORS

Comparison of GEOS-Chem (GC) 500 hPa data for 2006 to ozonesondes and to bias-corrected satellite data



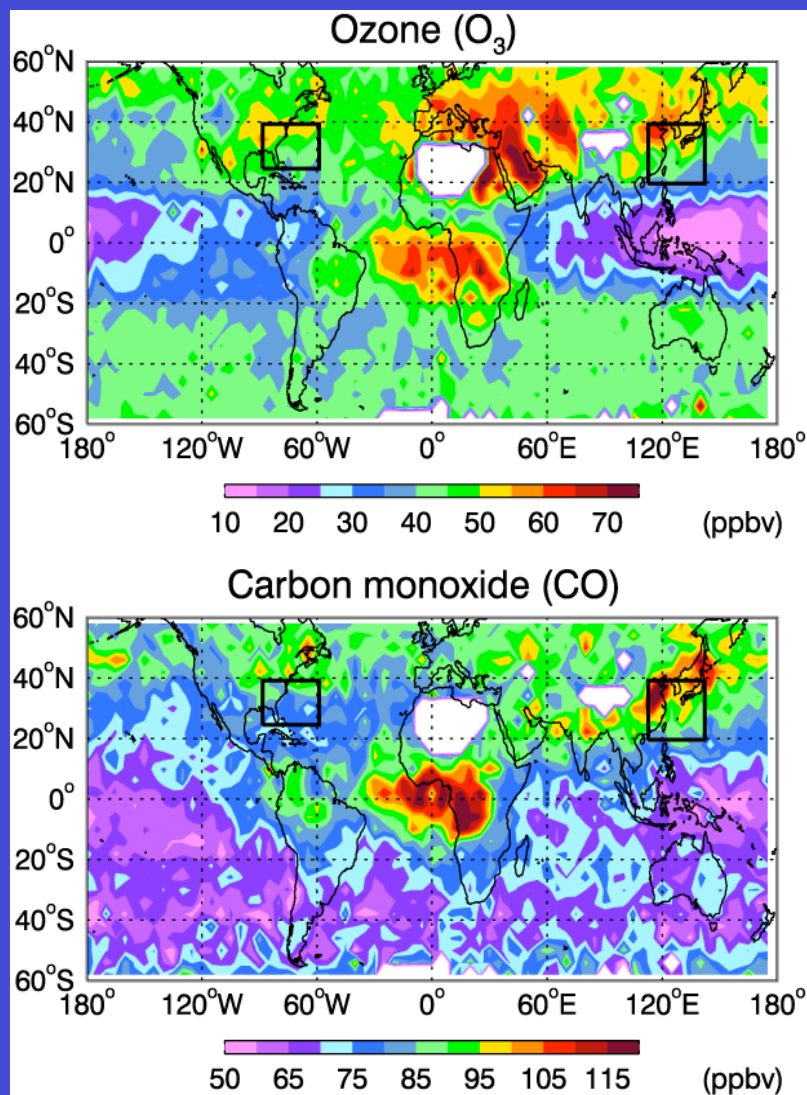
- Model too low in large regions of tropics – in part due to biomass burning
- Model too high in extratropical SH: excessive STE?

Zhang et al. , in prep.

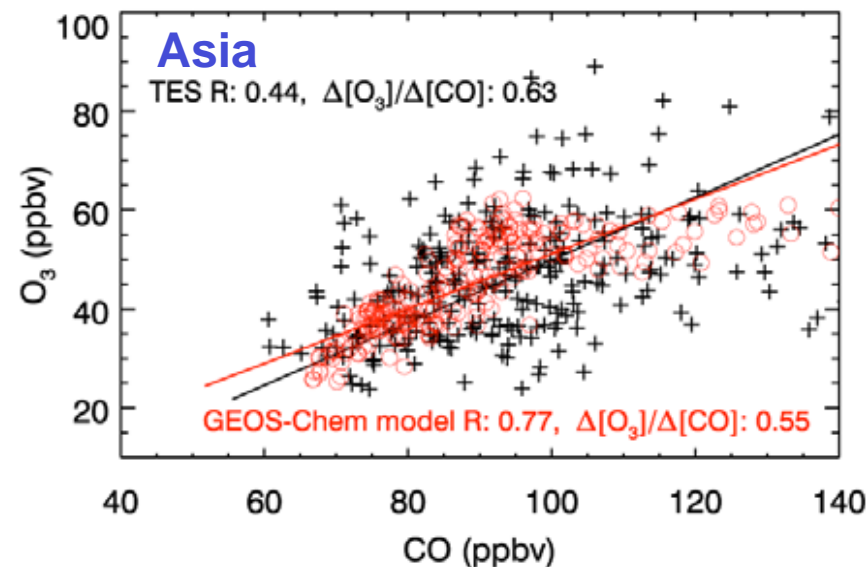
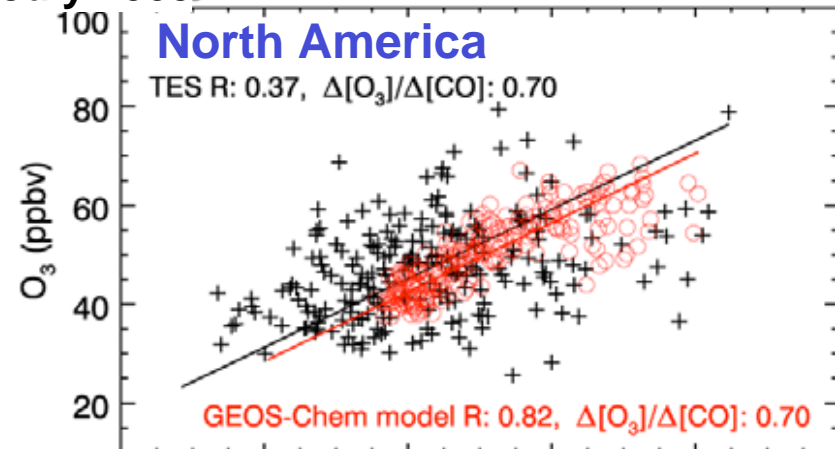


Ozone and CO at 4 km from TES satellite instrument

Ozone- CO correlation demonstrates ozone pollution export in continental outflow, tests global models of atmospheric composition (here GEOS-Chem)



July 2005₃-CO correlations in continental outflow



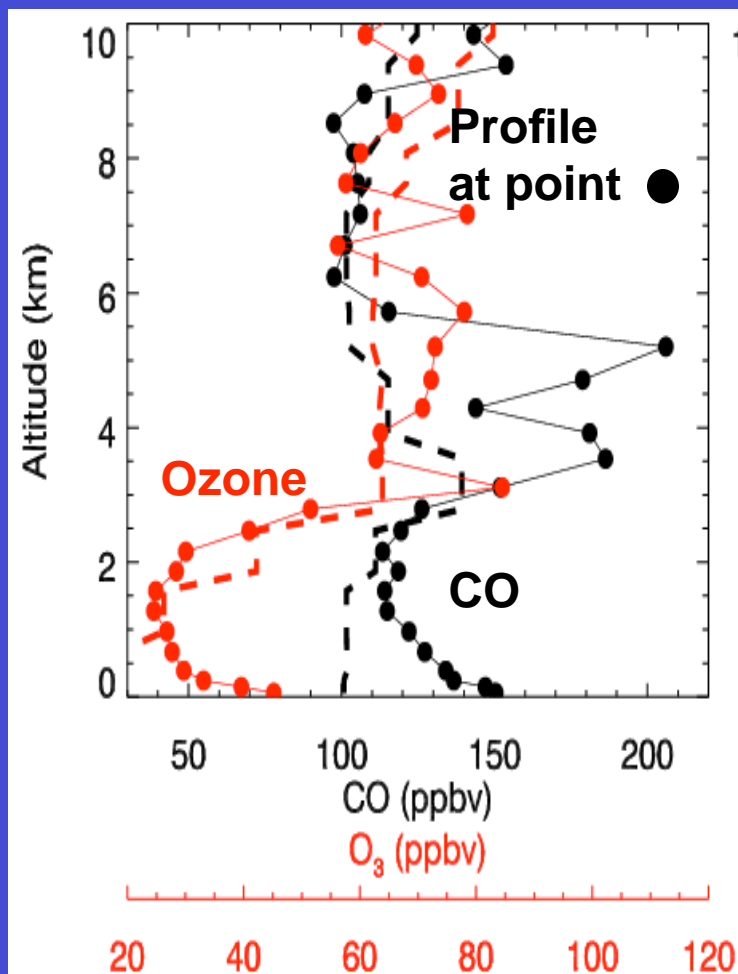
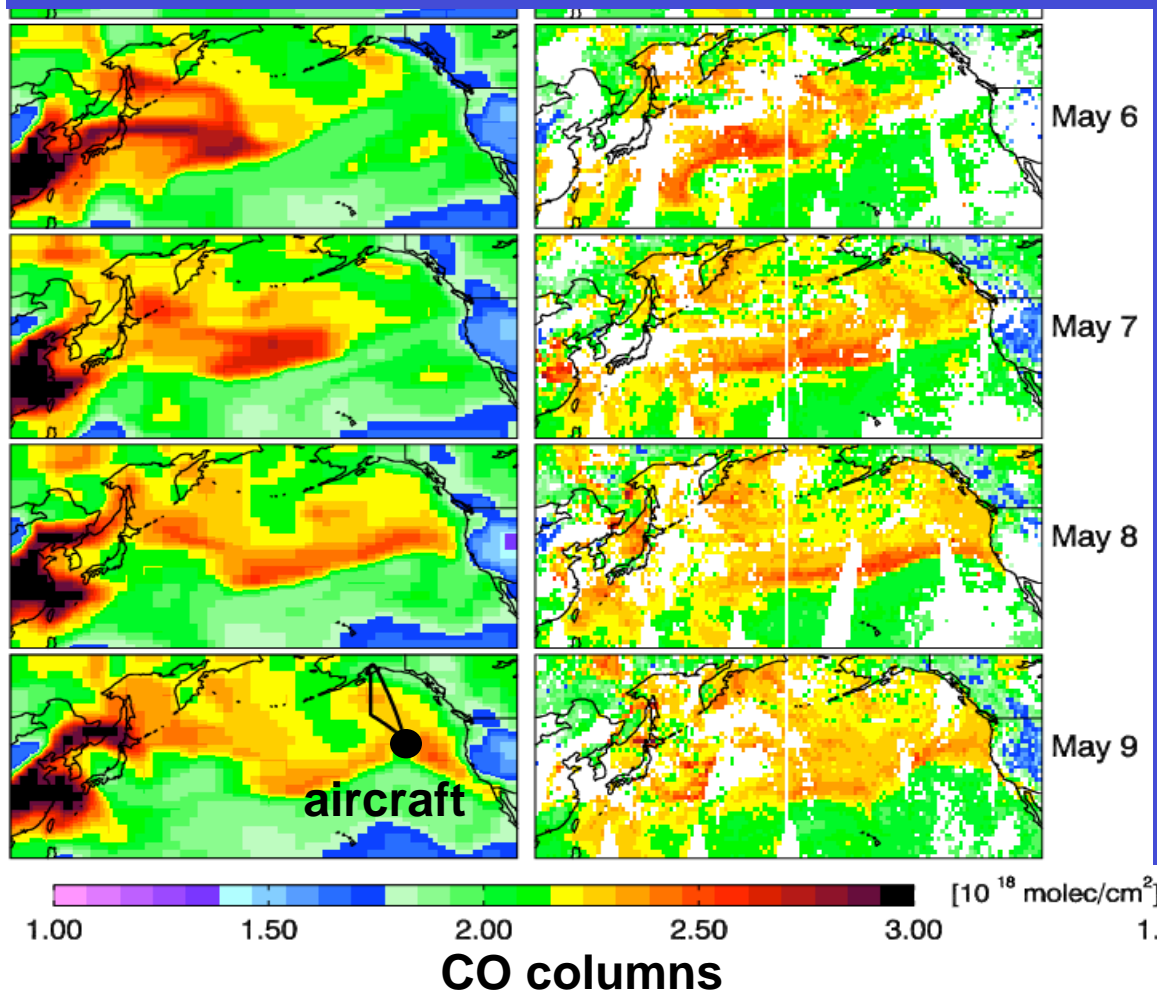
Linking satellite and aircraft to understand transpacific pollution

NASA/INTEX-B campaign over Northeast Pacific (spring 2006)

GEOS-Chem model

AIRS satellite instrument

Aircraft (May 9)



Asian pollution plumes show sustained ozone production across Pacific;
mean surface ozone enhancement in western US from Asian pollution is 5-7 ppb

Zhang et al. [2008]

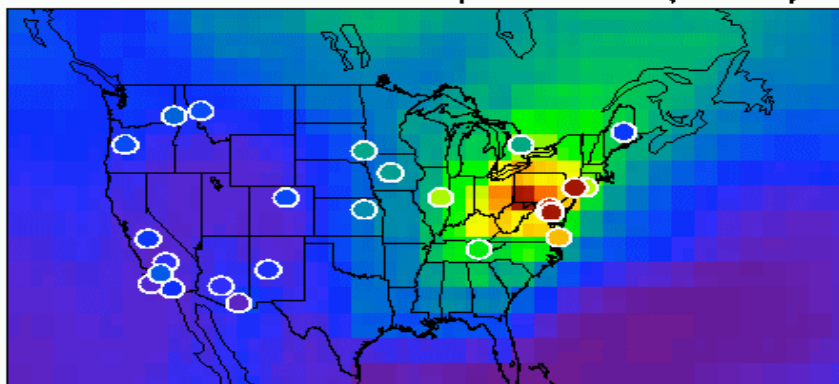
MEAN AEROSOL OPTICAL DEPTHS DURING ICARTT

Model results compared to observations from AERONET network (circles)

Model w/ GADs size distributions

Model w/improved size distributions

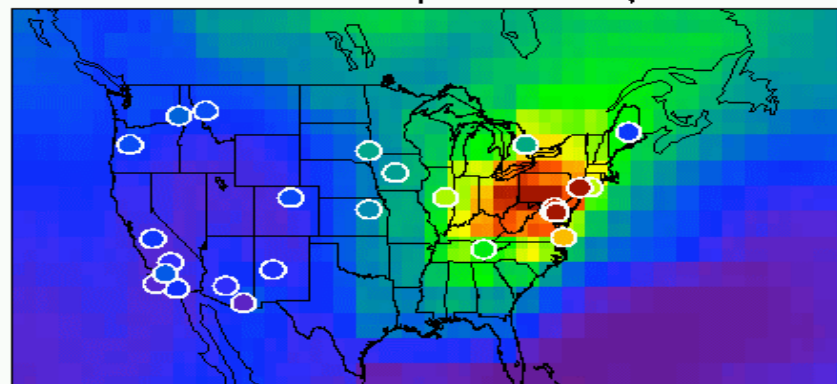
GEOS-Chem 0.47 μm AOD (GADs)



0.0 0.1 0.2 0.3 0.4 0.5 0.6 [AOD]

$r = 0.89$ bias = -21%

GEOS-Chem 0.47 μm AOD (this work)



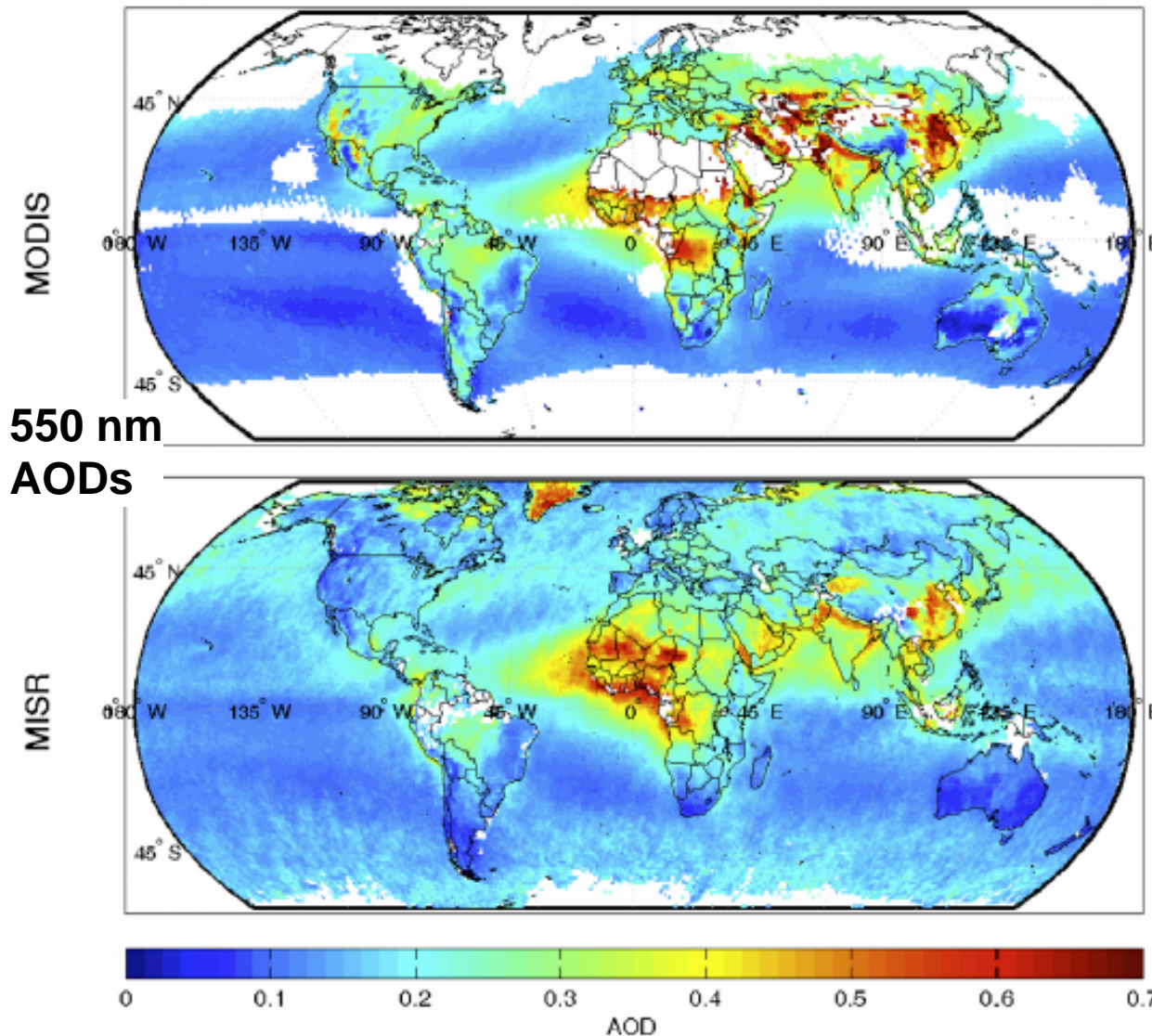
$r = 0.89$ bias = -7%

Main improvement was to reduce the geometric standard deviation in the log-normal size distributions for sulfate and OC from 2.2 to 1.6

Easan Drury, in prep.

Aerosol optical depths (AODs) measured from space

Jan 2001 – Oct 2002 operational data



MODIS (c004)

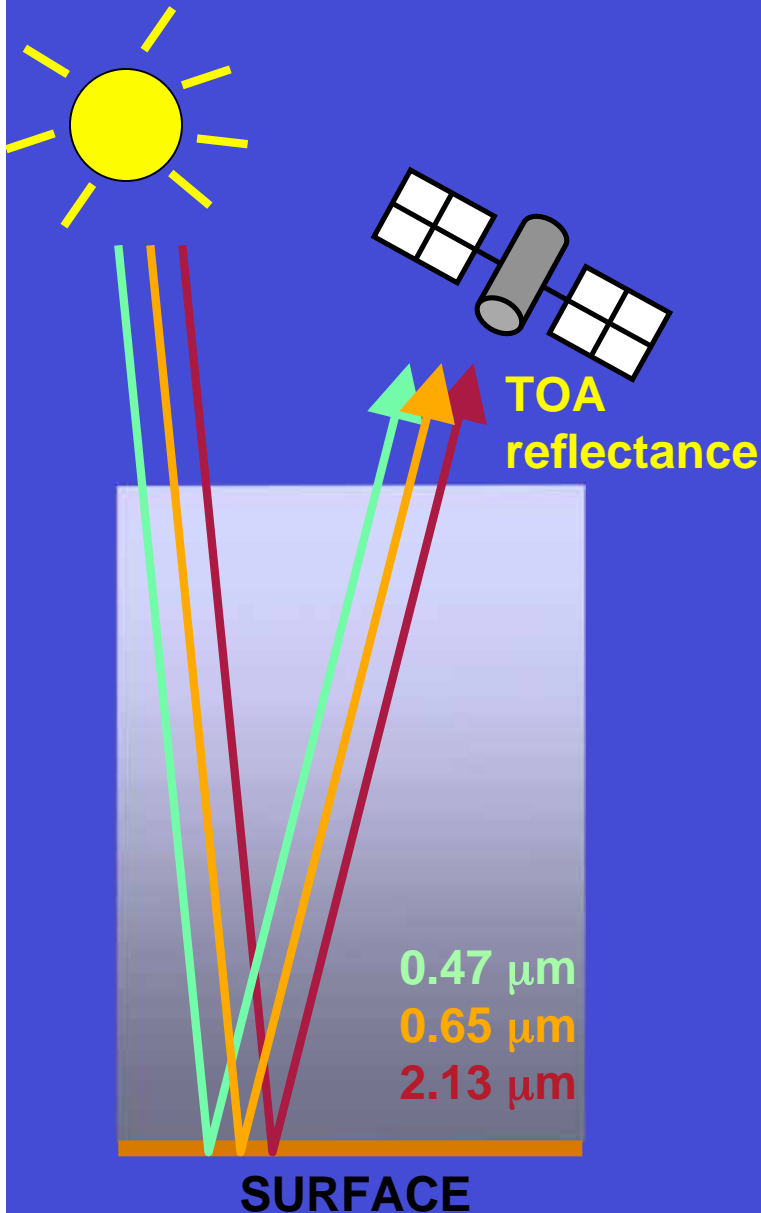
return time 2x/day;
nadir view
known positive bias
over land

MISR

9-day return time;
multi-angle view
better but much sparser

van Donkelaar et al. [2006]

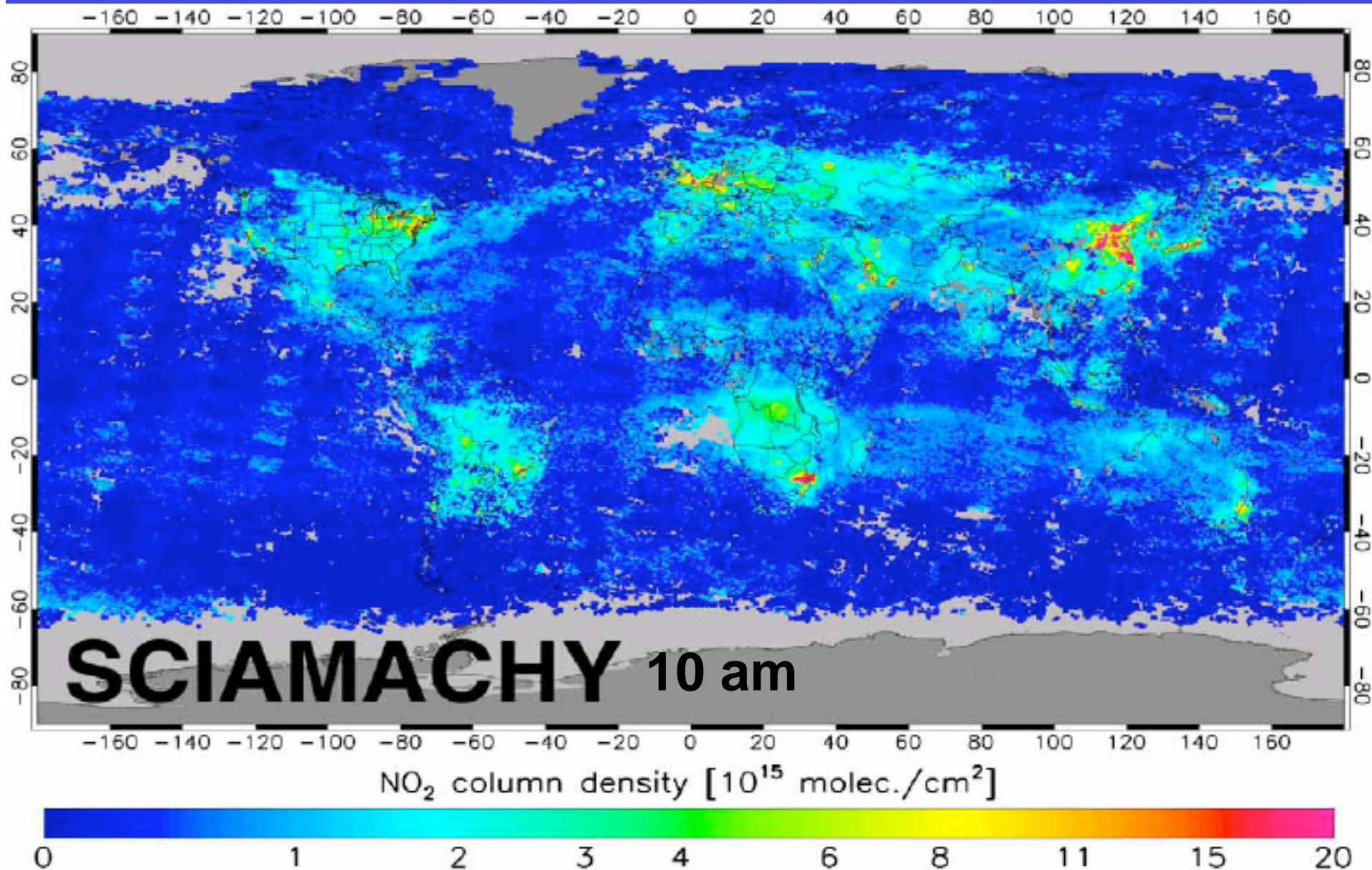
MODIS OPERATIONAL RETRIEVAL OVER LAND



- Use top-of-atmosphere (TOA) reflectance at 2.13 μm (transparent atmosphere) to derive surface reflectance
- Assume 0.47/2.13 and 0.65/2.13 surface reflectance ratios to derive atmospheric reflectances at 0.47 and 0.65 μm by subtraction
- Assume generic aerosol optical properties to convert atmospheric reflectance to AOD

Y. Kaufman, L. Remer, and MODIS Science Team

DIURNAL VARIATION OF BOUNDARY LAYER NO_2 as observed by the SCIAMACHY and OMI satellite instruments (Aug 2006)



Boersma et al. [2008]