Spectroscopic Needs for Atmosphere Pollution Measurements

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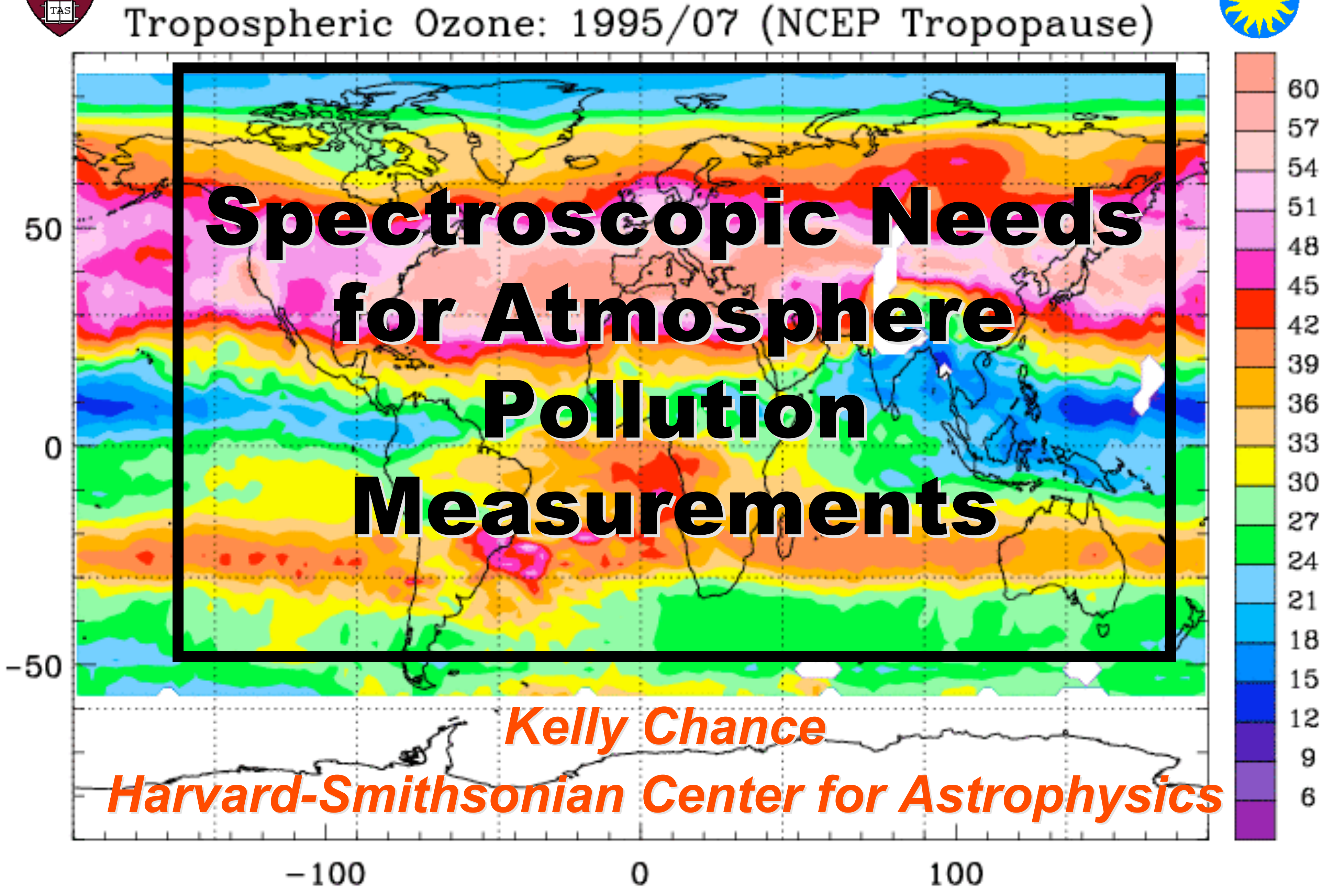
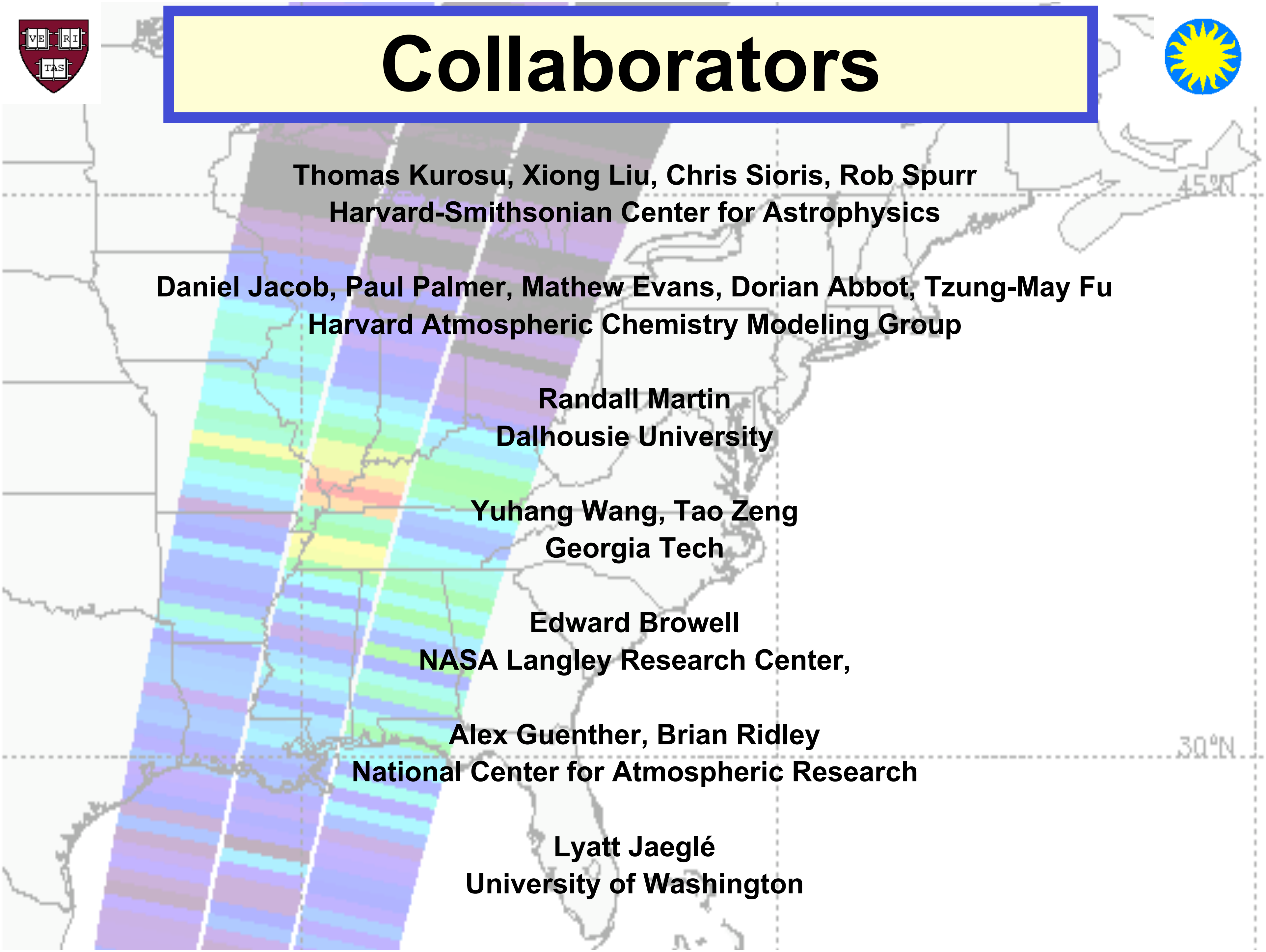


Collaborators



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Outline

- Introduction and motivation
- Descriptions of satellite instruments
- Outstanding spectroscopic needs for atmospheric pollution/air quality measurements
- O_3 profiles and tropospheric O_3
- NO_2: Inventories of nitrogen oxide emissions
- HCHO and CHOCHO: Volatile organic compound inventories
- BrO: destruction of tropospheric ozone in the polar spring, emissions from salt lakes and volcanoes
- Conclusions and future directions

Introduction and Motivation

- Target tropospheric gases are O_3, HCHO, CHO-CHO, NO_2, CO, SO_2, BrO.
- Our aims are:
  1. To retrieve tropospheric gases from GOME, SCIAMACHY, OMI, and future UV/visible/IR satellite instruments (*e.g.*, OMPS).
  2. To perform geophysical process studies with the results.
  3. To develop capability for air quality forecasts.
- Successful retrieval involves detailed development of algorithm physics coupled with chemistry and transport modeling and multiple-scattering radiative transfer calculations.

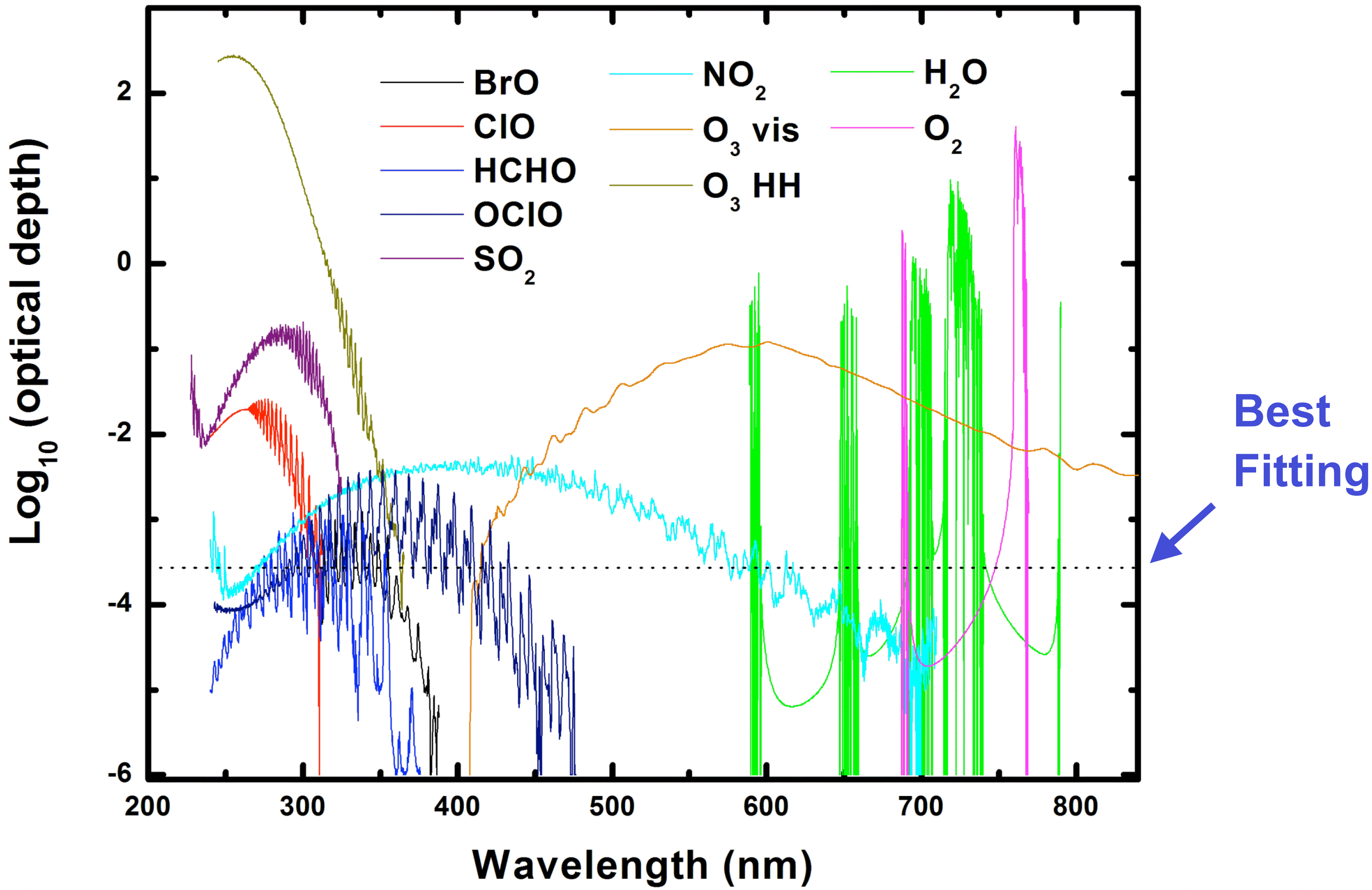
GOME/SCIAMACHY/OMI/GOME-2

Instrument	Detector	Spectral Coverage [nm]	Spectral Resolution [nm]	Ground Pixel Size [km^2]	Global Coverage
GOME (1995)	Linear array	240-790	0.2-0.4	40×320 (40×80 zoom)	3 days
SCIAMACHY (2002)	Linear array	240-2380	0.2-1.5	30×30 30×60 30 ×90 30×120 30× 240 (depending on product)	6 days
OMI (2004)	2-D CCD	270-500	0.42-0.63	15×30 - 42×162 (depending on swath position)	daily
GOME-2 (2006?)	Linear array	240-790	0.24-0.53	40×40 (40×80 wide-swath, 40×10 zoom)	1.5 to 3 days

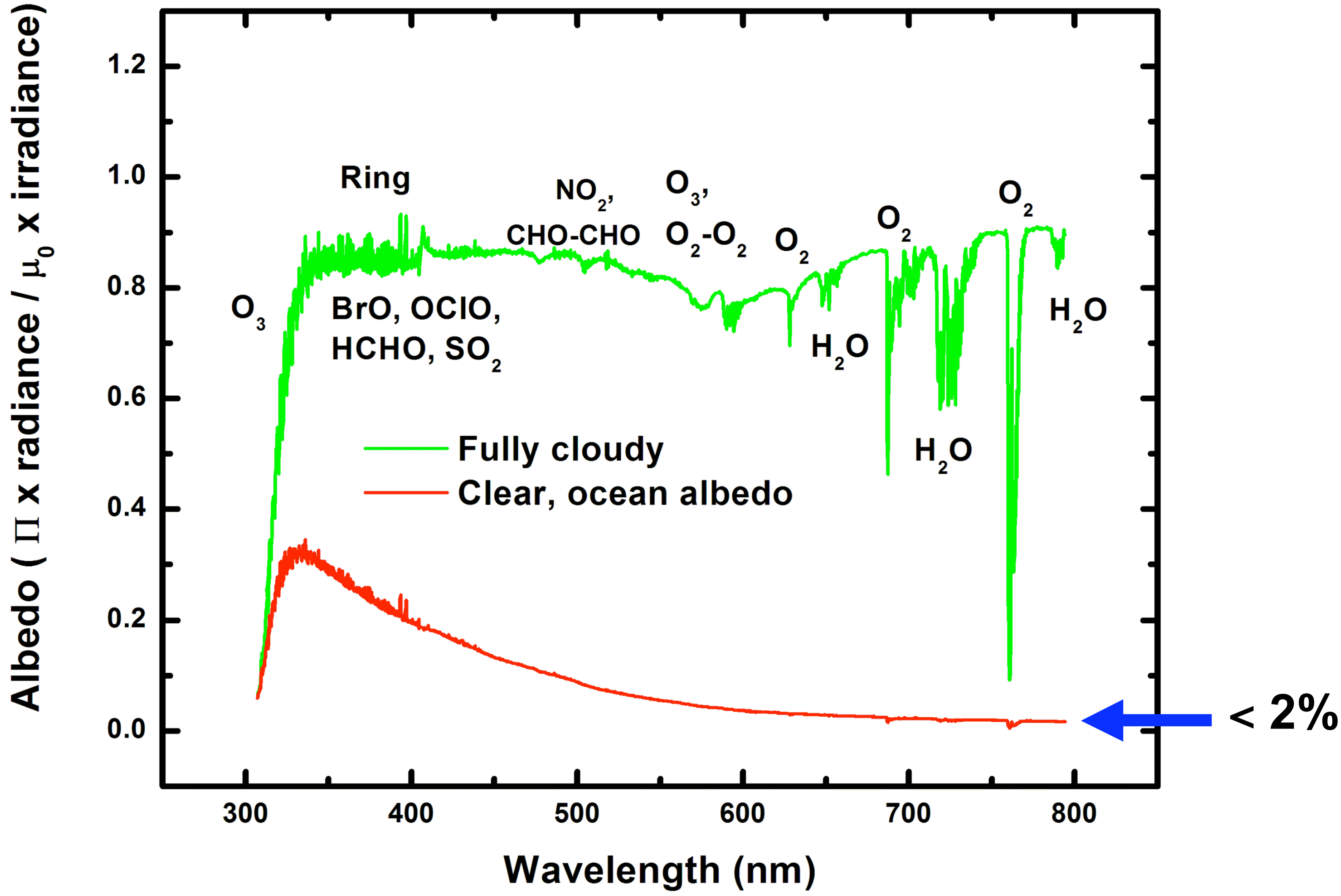
Major Spectroscopic Needs For Pollution Measurements

Gas/ Reference	Spectral Coverage [nm]	Spectral Resolution [nm]	Need
O_3	250-1000 + 9.6 µm IR	<0.01 (FTS)	FTS measurements over the range of stratospheric and tropospheric temperatures; simultaneous IR measurements @ 9.6 µm
Solar Irradiance	250-1000	<0.01 (FTS)	FTS extrasolar measurements
HCHO	300-365	<0.01 (FTS)	FTS measurements over range of tropospheric temperatures; resolve ~15% discrepancy in intensities; IR
SO_2	290-327	<0.01 (FTS)	FTS measurements over the range of tropospheric temperatures
O_2-O_2	330-460	<0.1	Improved intensities and wavelength calibration; trace gas fitting and cloud products / cloud correction
NO_2, CHOCHO, CO	Visible, IR		In reasonable shape

Optical Depths for Typical GOME Measurement Geometry



GOME Earth albedo spectra, clear and cloudy

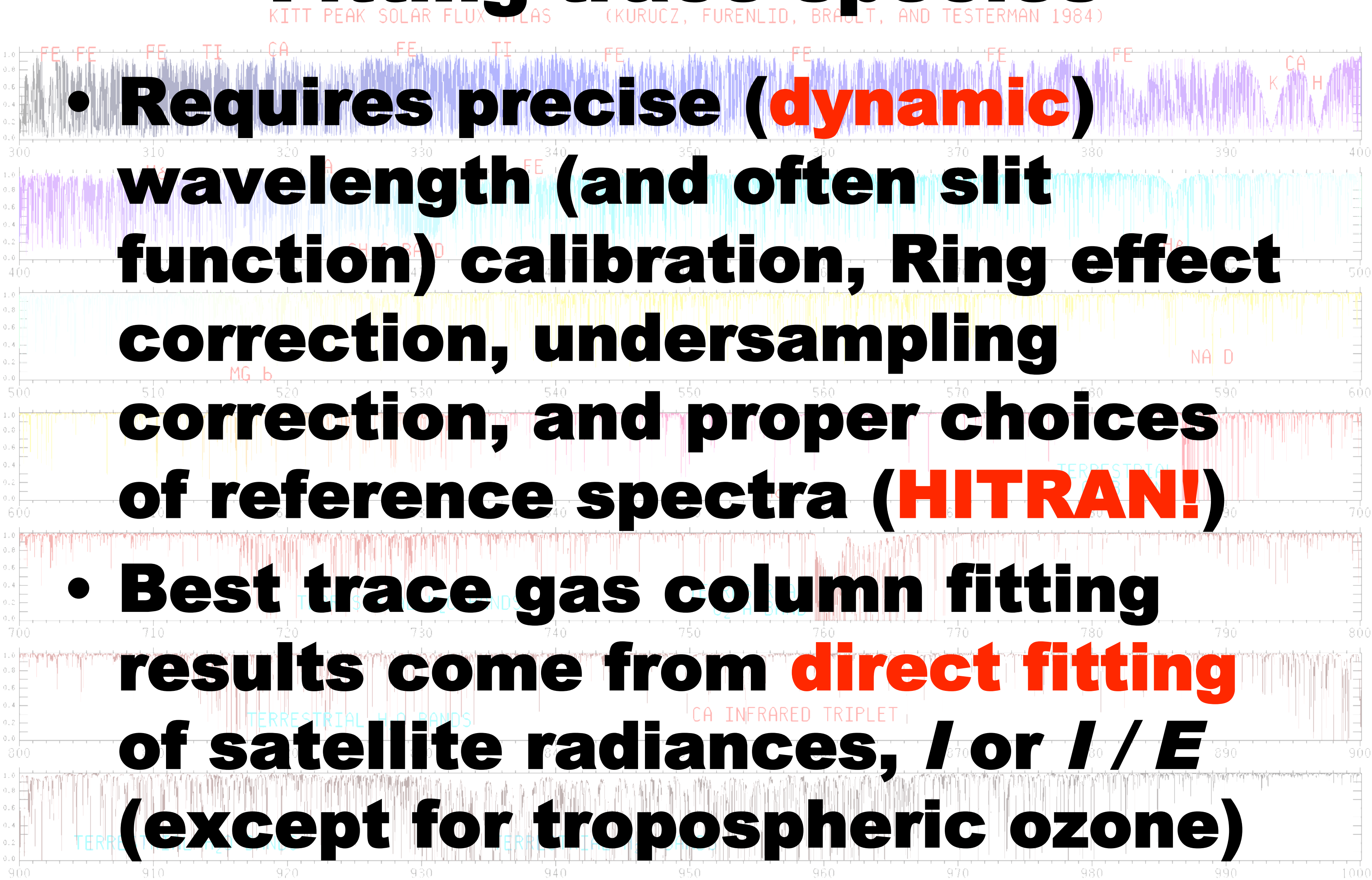


Spectrum fitting and radiative transfer correction

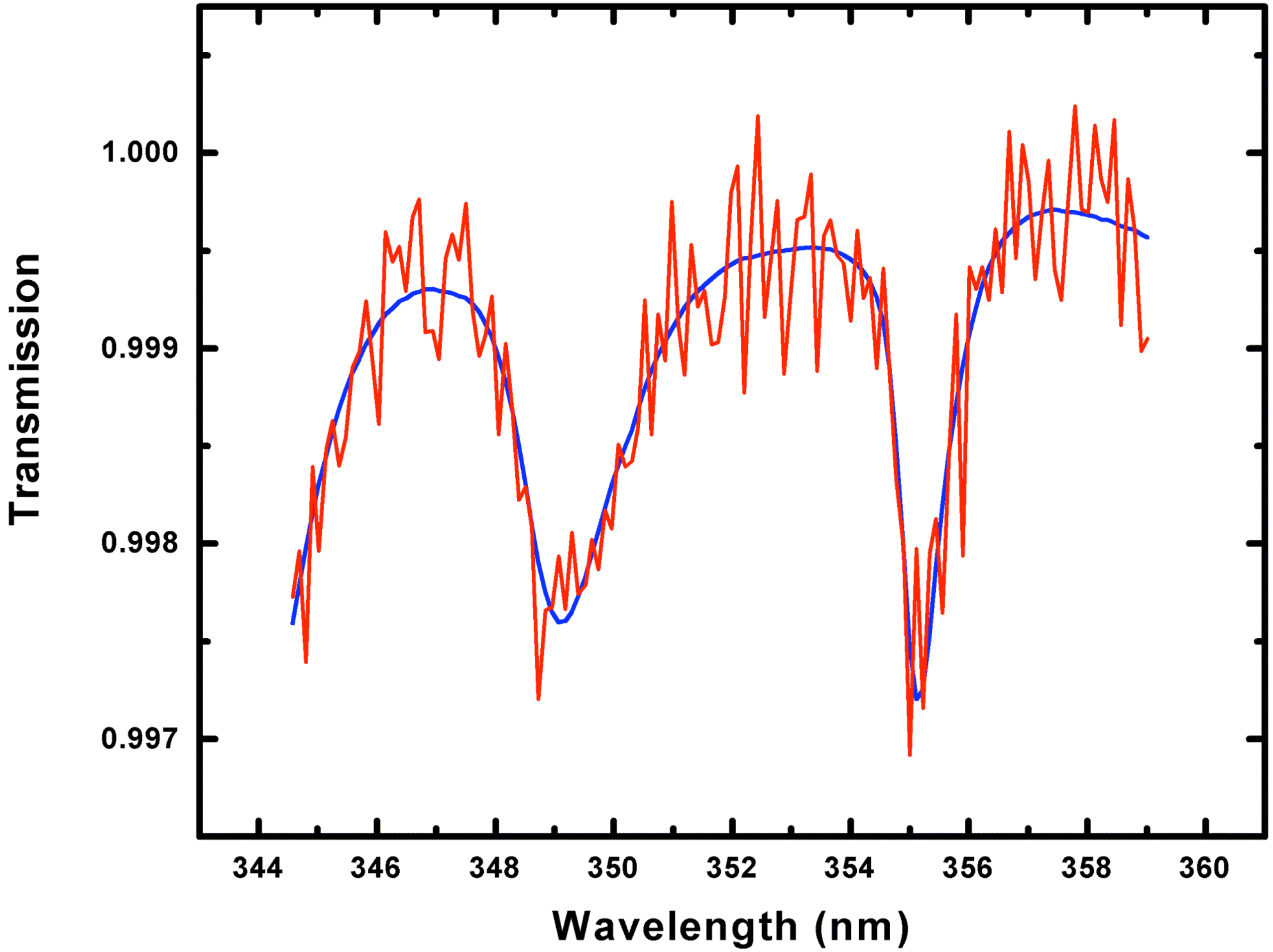
Frustra fit per plura, quod fieri potest per pauciora. Essentia non sunt multiplicada praeter necessitatem.

- *William of Occam*

Fitting trace species

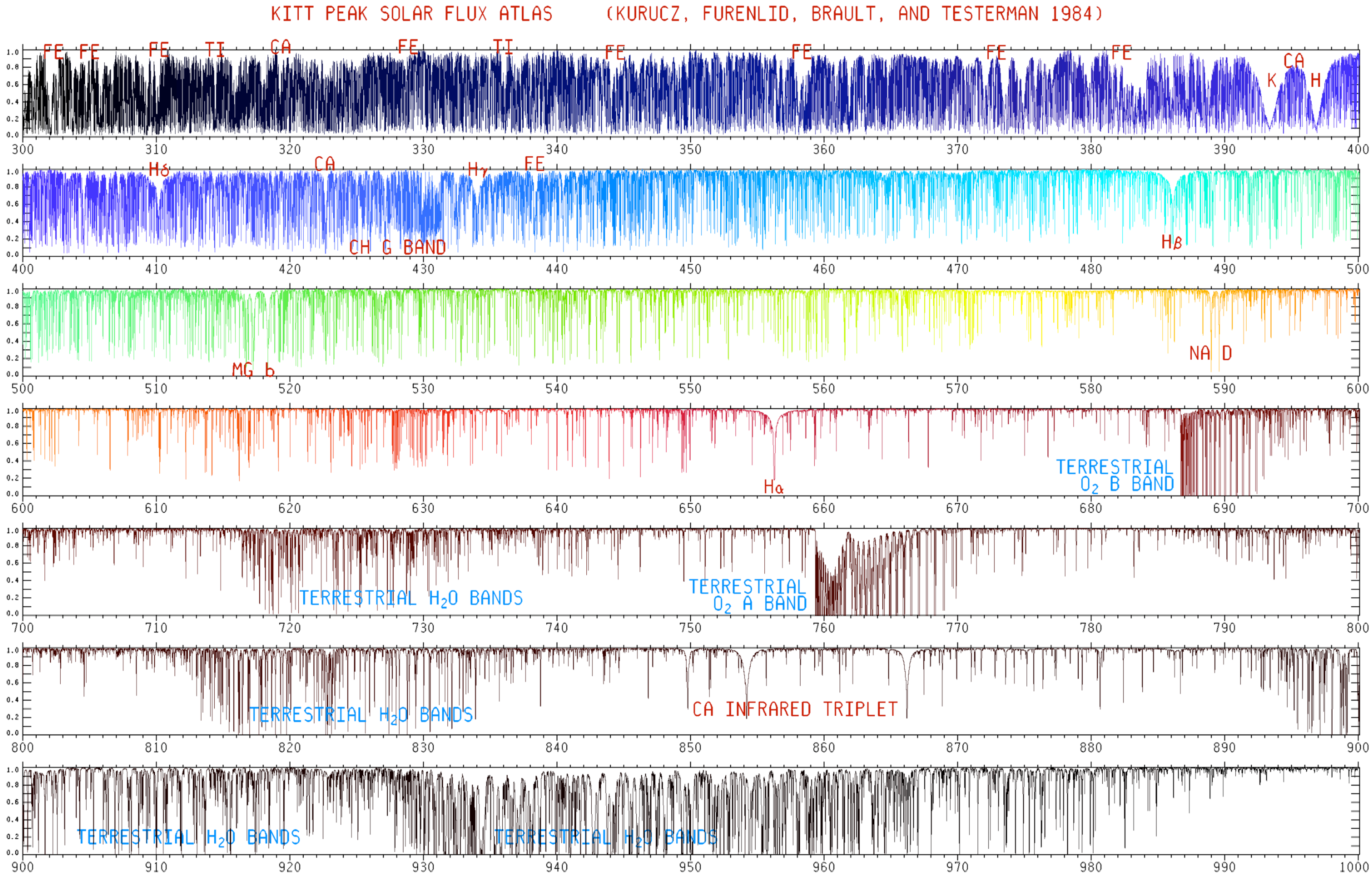


- **Requires precise (dynamic) wavelength (and often slit function) calibration, Ring effect correction, undersampling correction, and proper choices of reference spectra (HITRAN!)**
- **Best trace gas column fitting results come from direct fitting of satellite radiances, *I* or *I* / *E* (except for tropospheric ozone)**



GOME BrO fitting for the FIRS-2 overflight on April 30, 1997. The integration time is 1.5s. The fitting precision is 4.2% and the RMS is 2.7×10^{-4} in optical depth. Fitting and inversion give a vertical BrO column of 9.3×10^{13} cm^{-2}.

High resolution solar reference spectrum



Top-of-atmosphere solar spectral irradiance

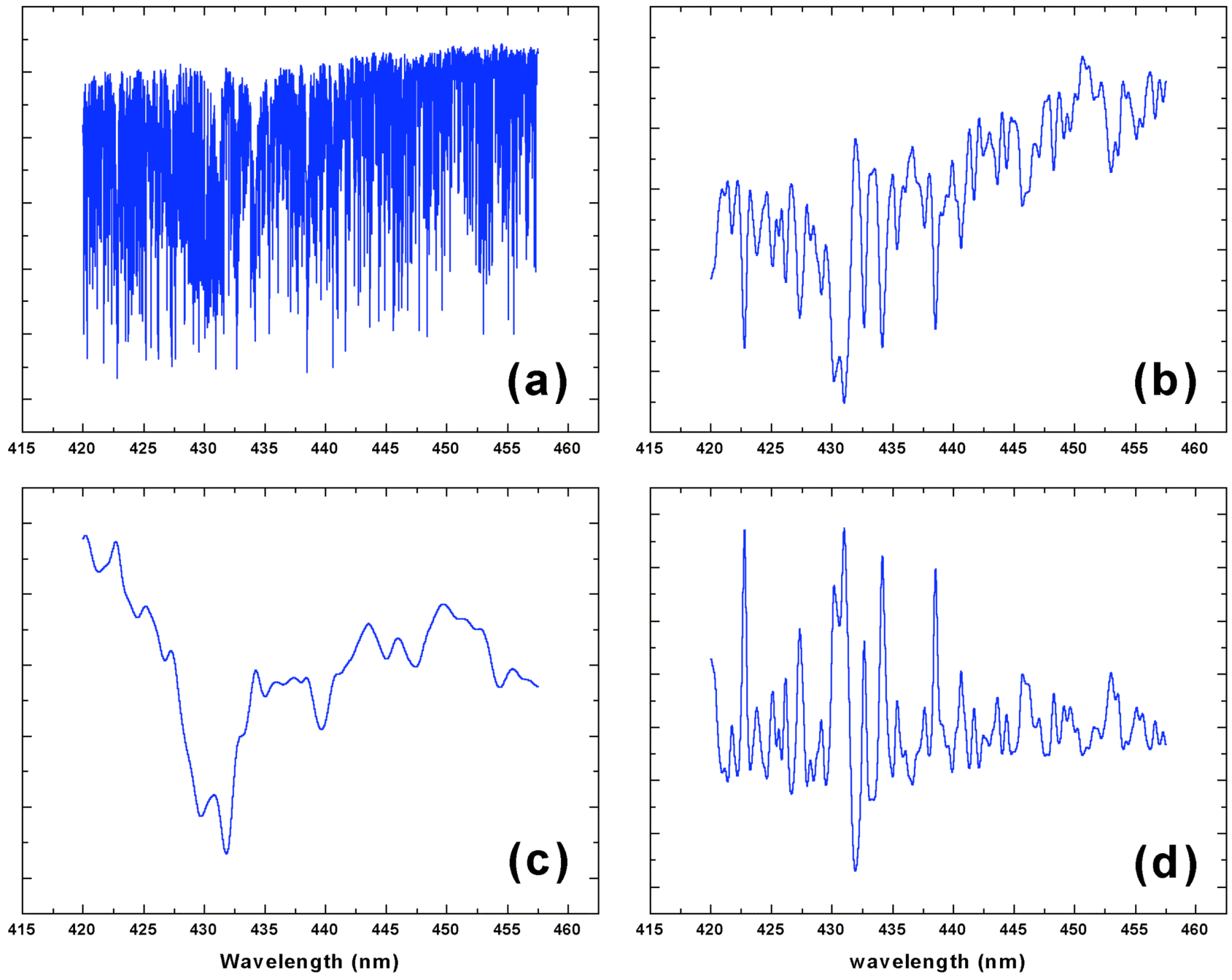
The high resolution solar spectral irradiance is critical in analyzing atmospheric trace gases:

- **Solar lines are source of accurate wavelength calibration (±0.0003-0.0004 nm for GOME!) – Our method now used operationally on GOME, SCIAMACHY, OMI, and OMPS**
- **Determination of the Ring effect**
- **Improved knowledge of instrument slit functions**
- **Correction for spectral undersampling**
- **Photochemistry of Schumann-Runge system**

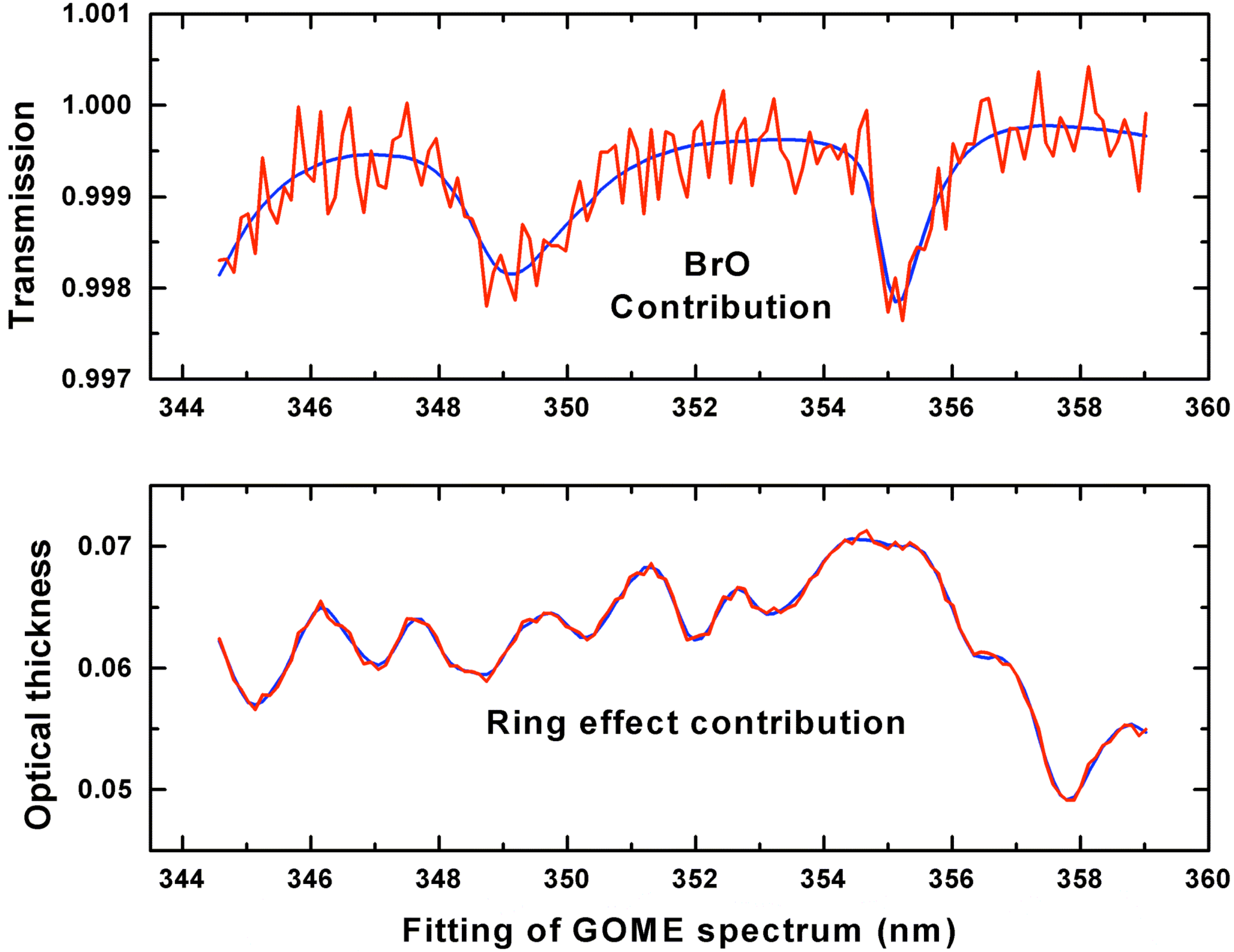
A space-based determination would be an ideal support mission for 12+ international atmospheric missions!

- ***Range: 240-1000+ nm***
- ***FWHM: 0.01 nm or better***
- ***Ideal FTS Space Shuttle Canadian European experiment***

Ring effect correction spectrum



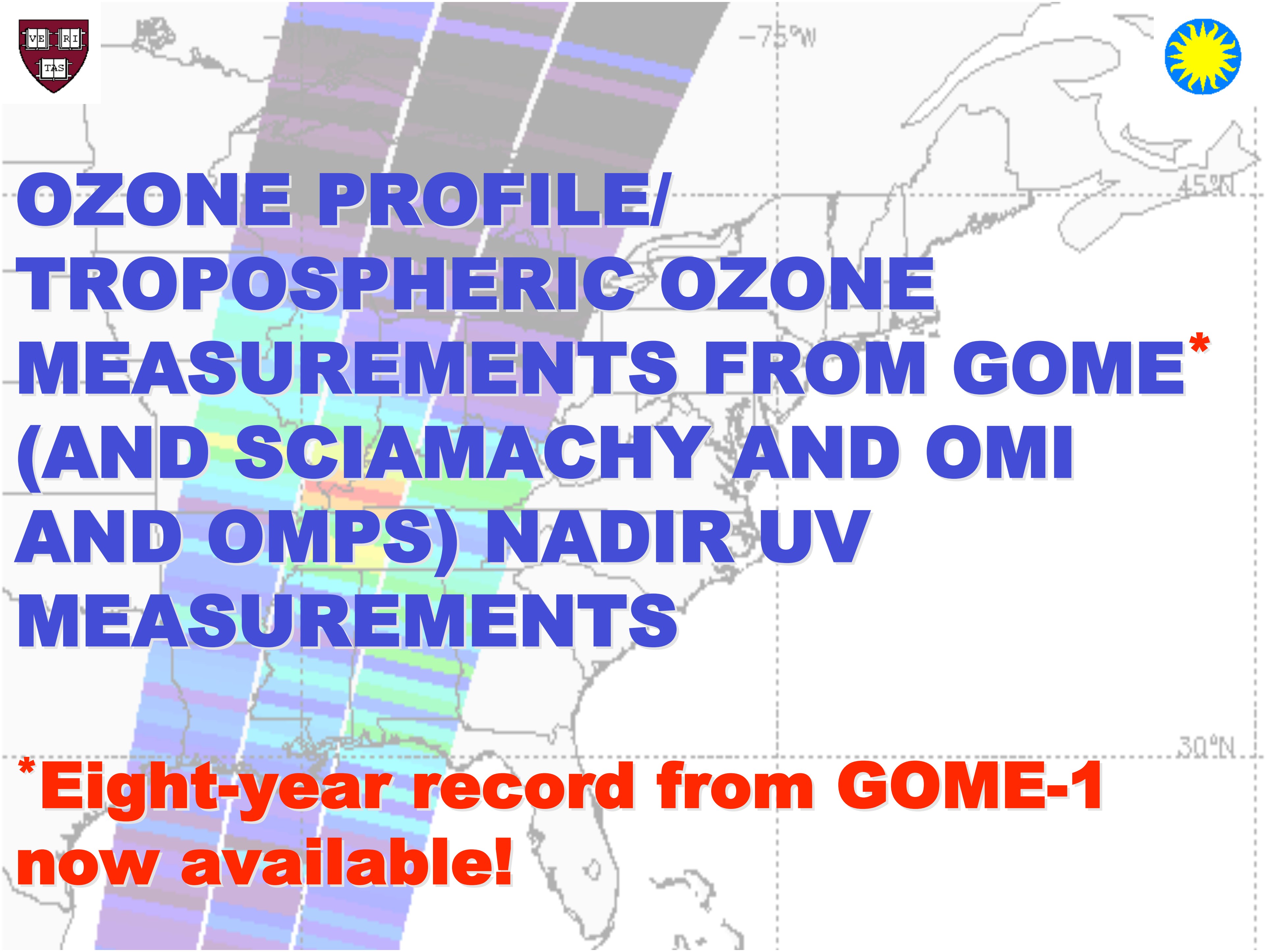
(a) Fraunhofer reference spectrum for the NO_2 fitting region; (b) Fraunhofer convolved to GOME spectral resolution; (c) = (b) convolved with rotational Raman cross-sections = Ring effect scattering source per molecule; (d) High-pass filtered version of (c) / (b) = DOAS "Ring effect correction."

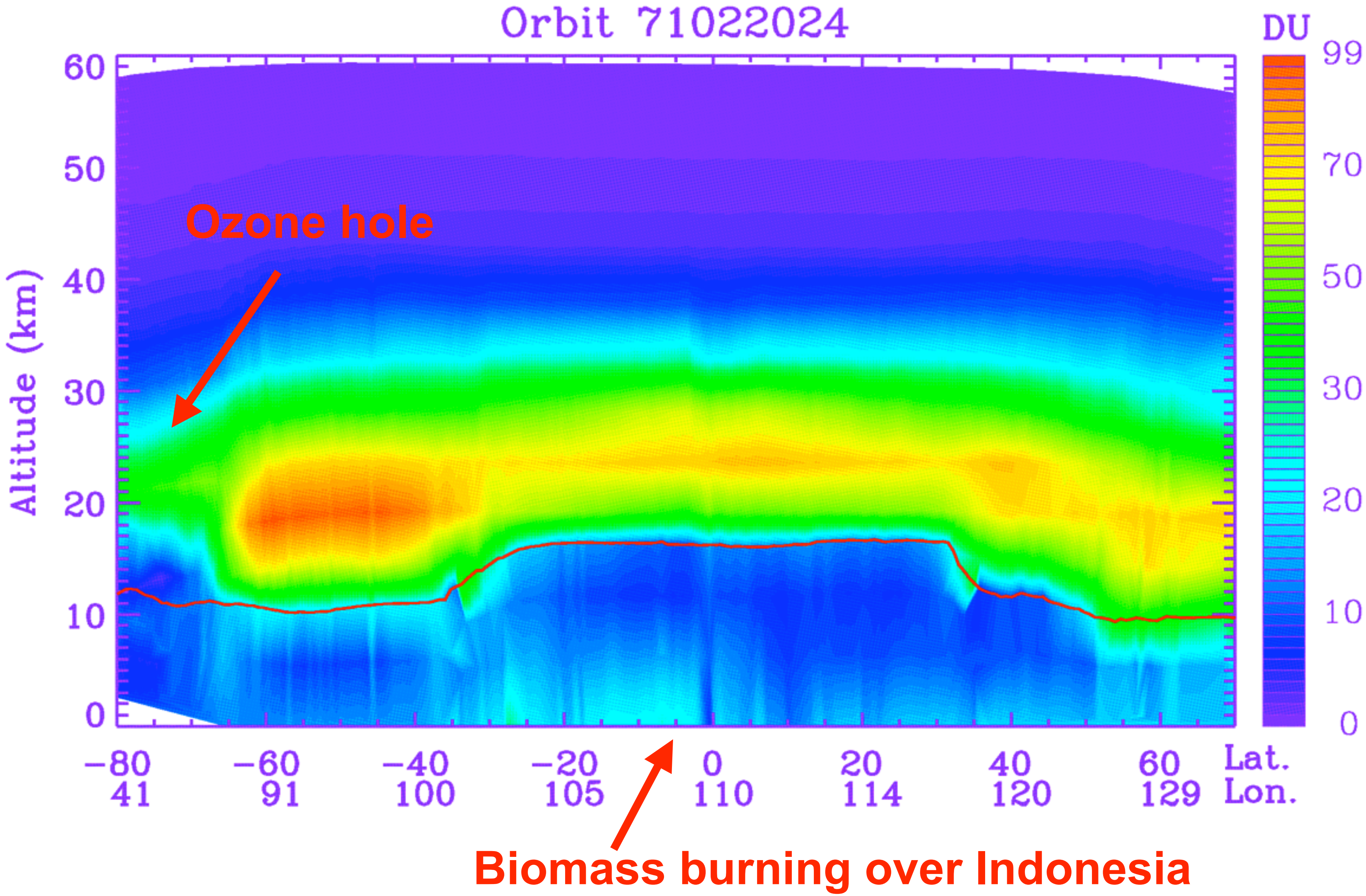


GOME BrO fitting: Relative contributions absorption by atmospheric BrO (top) and the Ring effect - the inelastic, mostly rotational Raman, part of the Rayleigh scattering – (bottom).

OZONE PROFILE/ TROPOSPHERIC OZONE MEASUREMENTS FROM GOME* (AND SCIAMACHY AND OMI AND OMPS) NADIR UV MEASUREMENTS

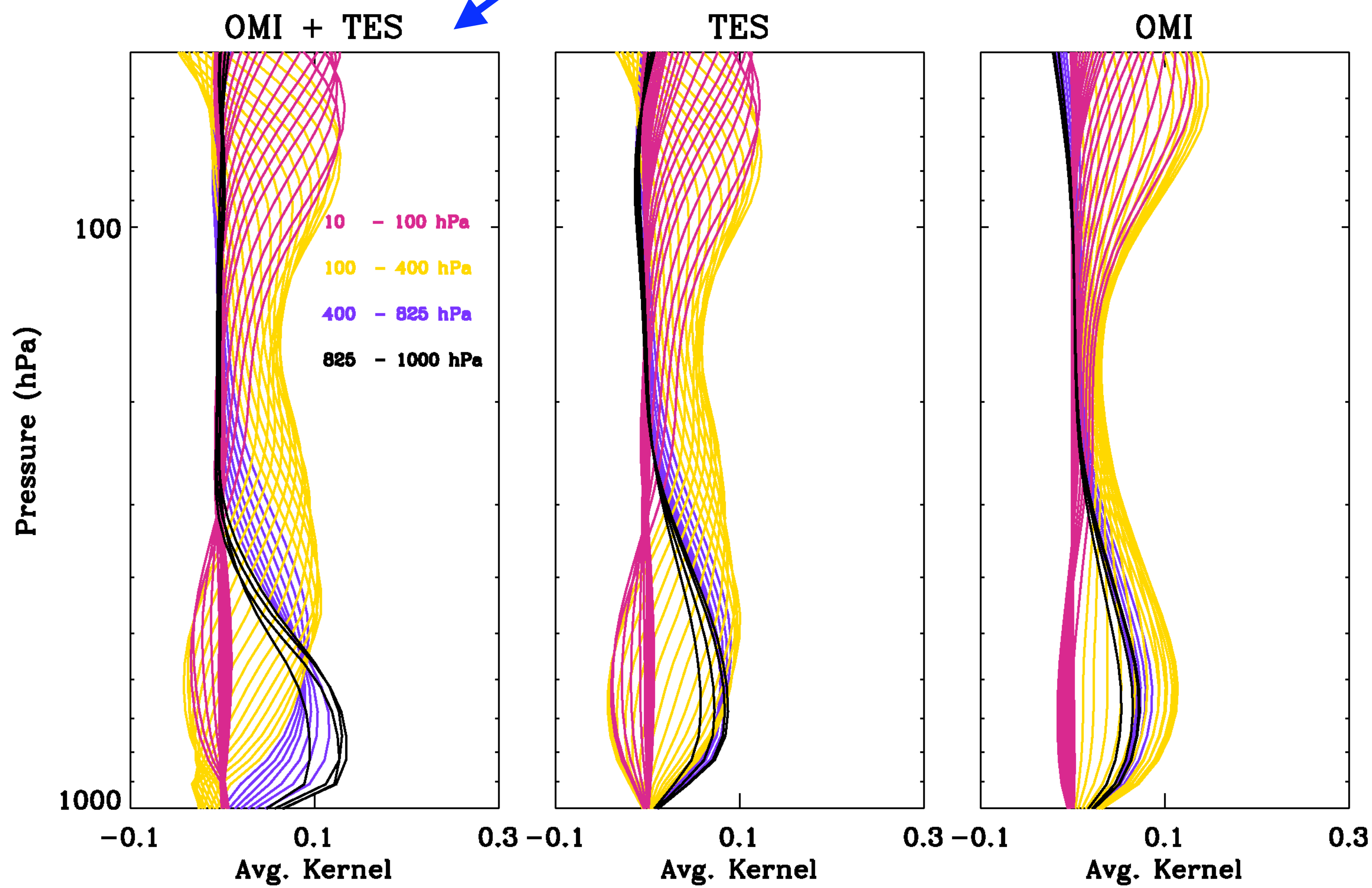
*Eight-year record from GOME-1 now available!





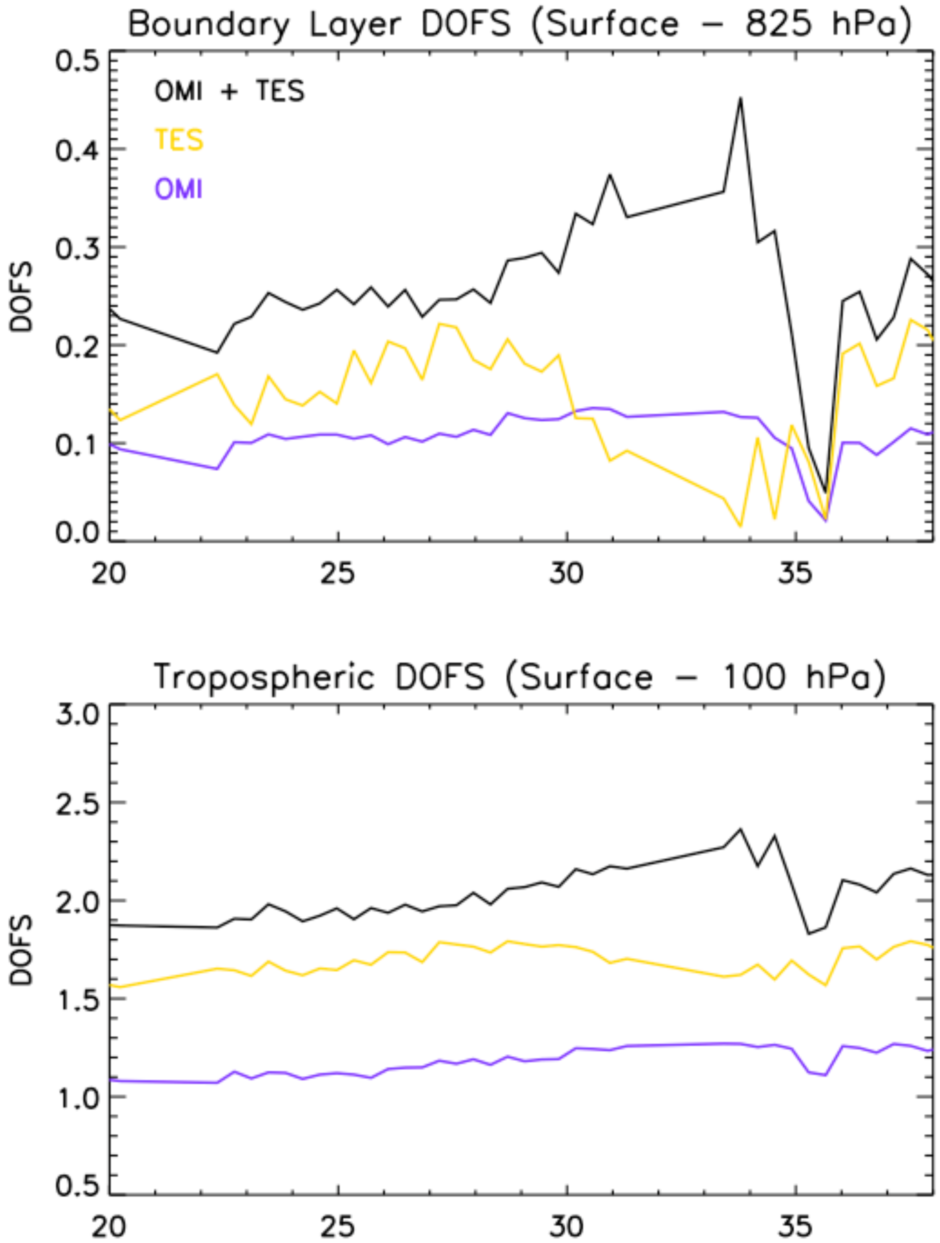
Orbit 71022024
Altitude (km)
60
50
40
30
20
10
0
Ozone hole
Biomass burning over Indonesia
DU
99
70
50
30
20
10
0
−80 41
−60 91
−40 100
−20 105
0 110
20 114
40 120
60 129
Lat.
Lon.

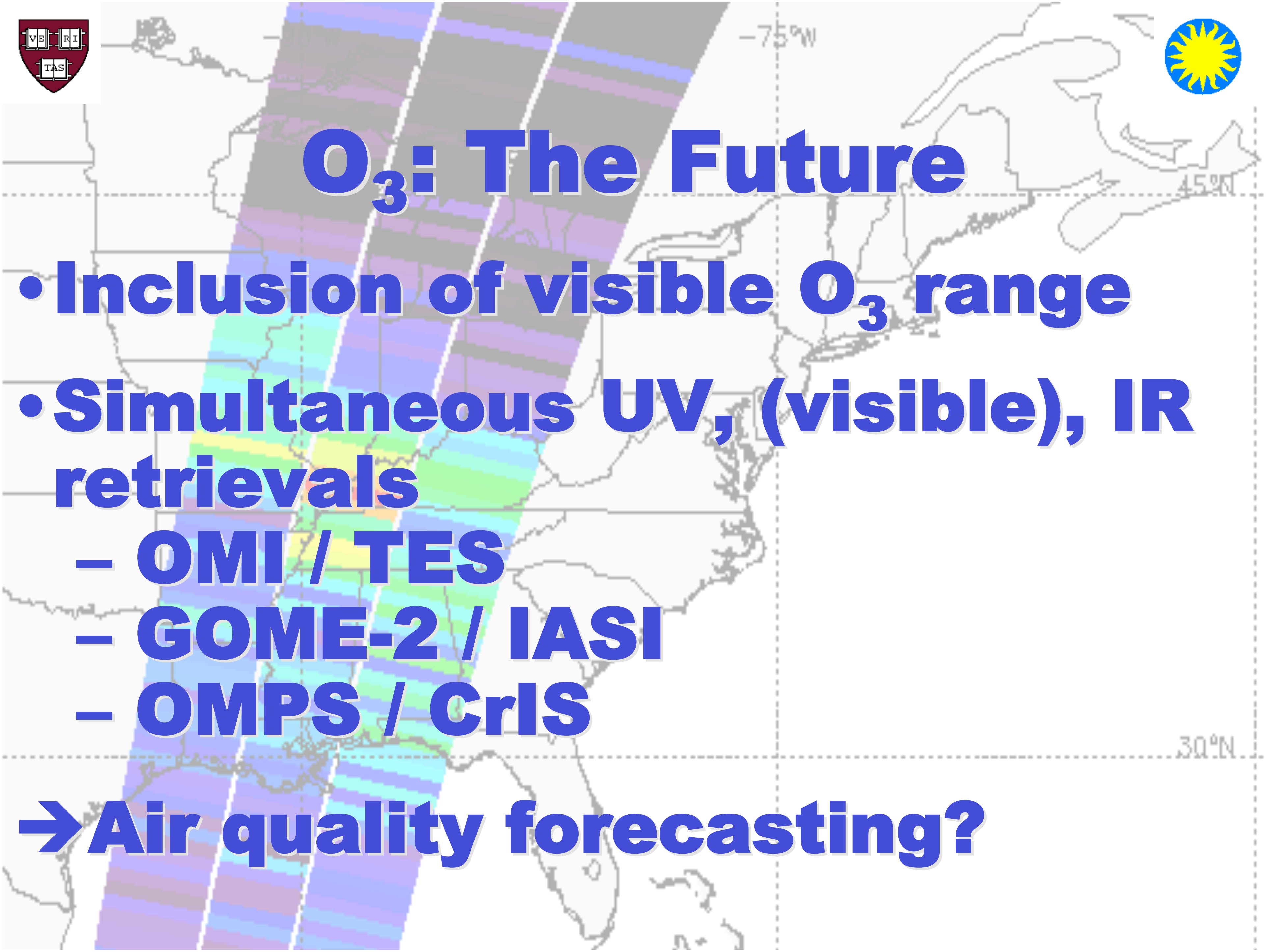
Needs spectral data!



Examples of Averaging Kernels. Left: averaging kernels for an OMI and TES synthetic ozone profile retrieval for an ozone estimate at 30.5 degrees latitude. The middle and right panels show the averaging kernels for this same scene but assuming a TES and OMI sounding of this scene respectively.

Top: The boundary layer DOFS for the previous set of ozone profiles as would be measured by OMI (purple line), TES (orange line) and OMI plus TES (black line). The DOFS are a metric for the vertical resolution or sensitivity of ozone sounding to the true ozone. Particularly striking is the non-linear increase in boundary layer sensitivity near 34 degrees latitude. This results from the more linearly independent averaging kernels of TES and OMI for this scene. Bottom: The total DOFS for the region between the surface and 100 hPa.



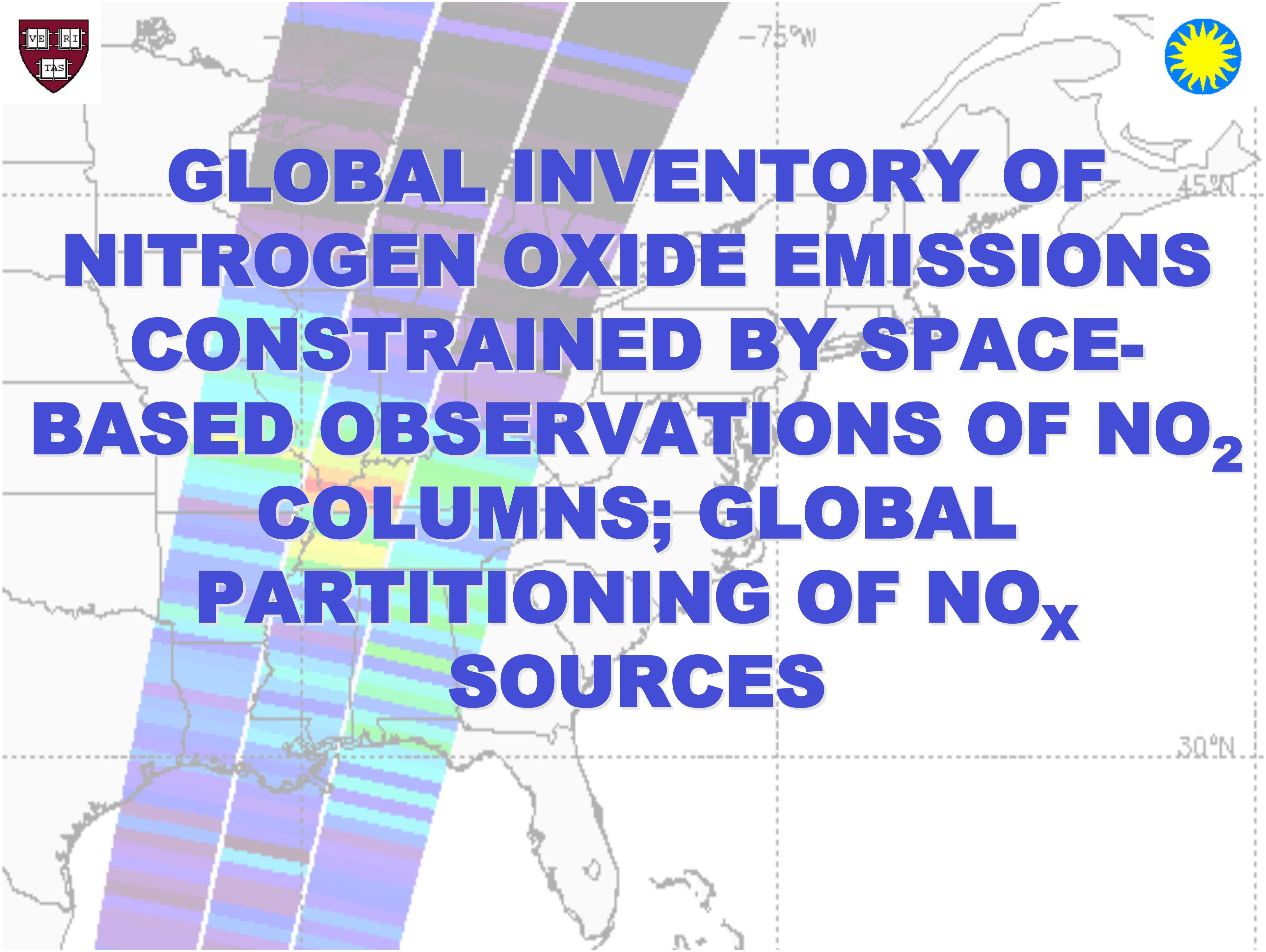


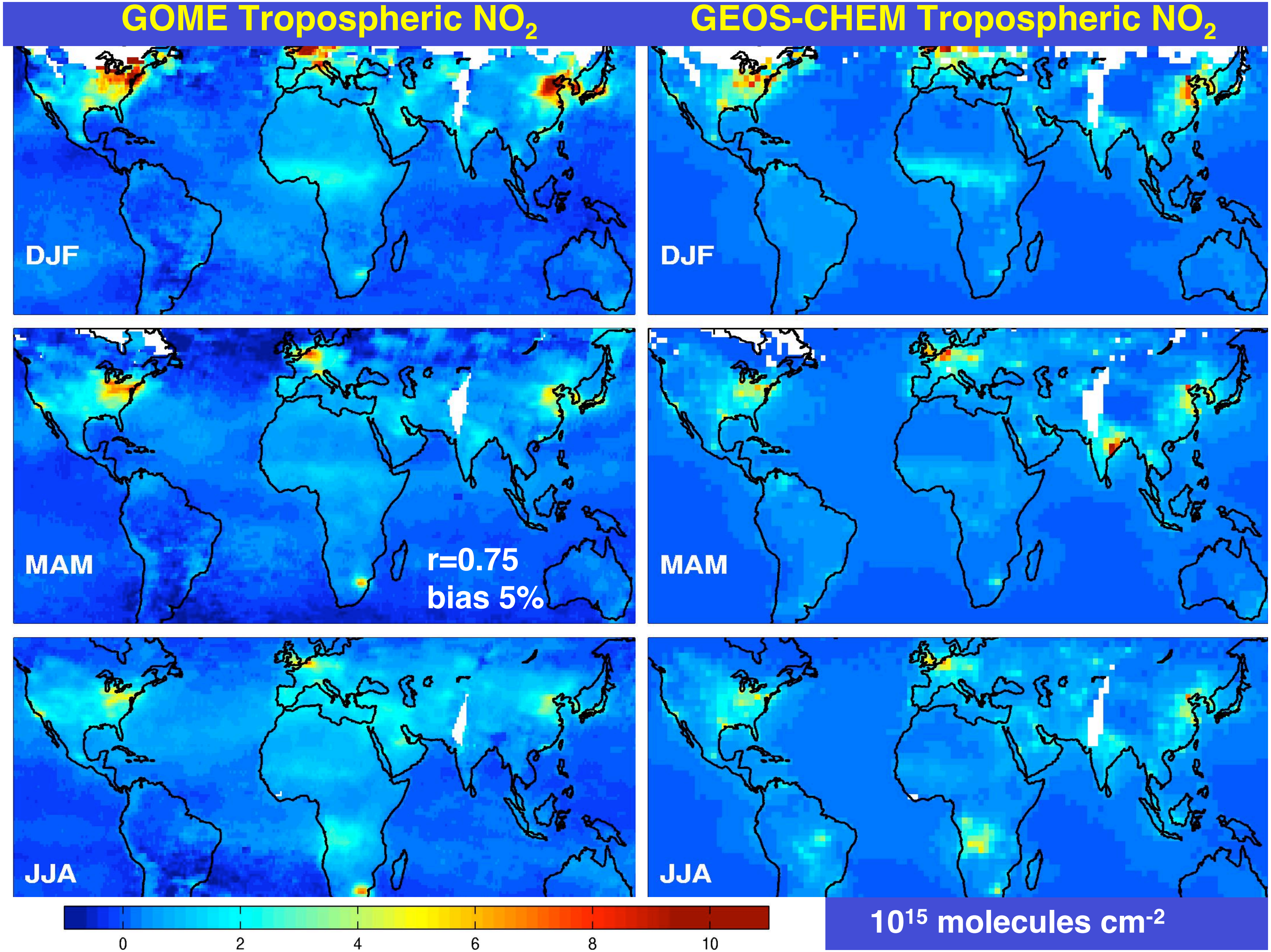
O_3: The Future

- Inclusion of visible O_3 range
- Simultaneous UV, (visible), IR retrievals
  - OMI / TES
  - GOME-2 / IASI
  - OMPS / CrIS

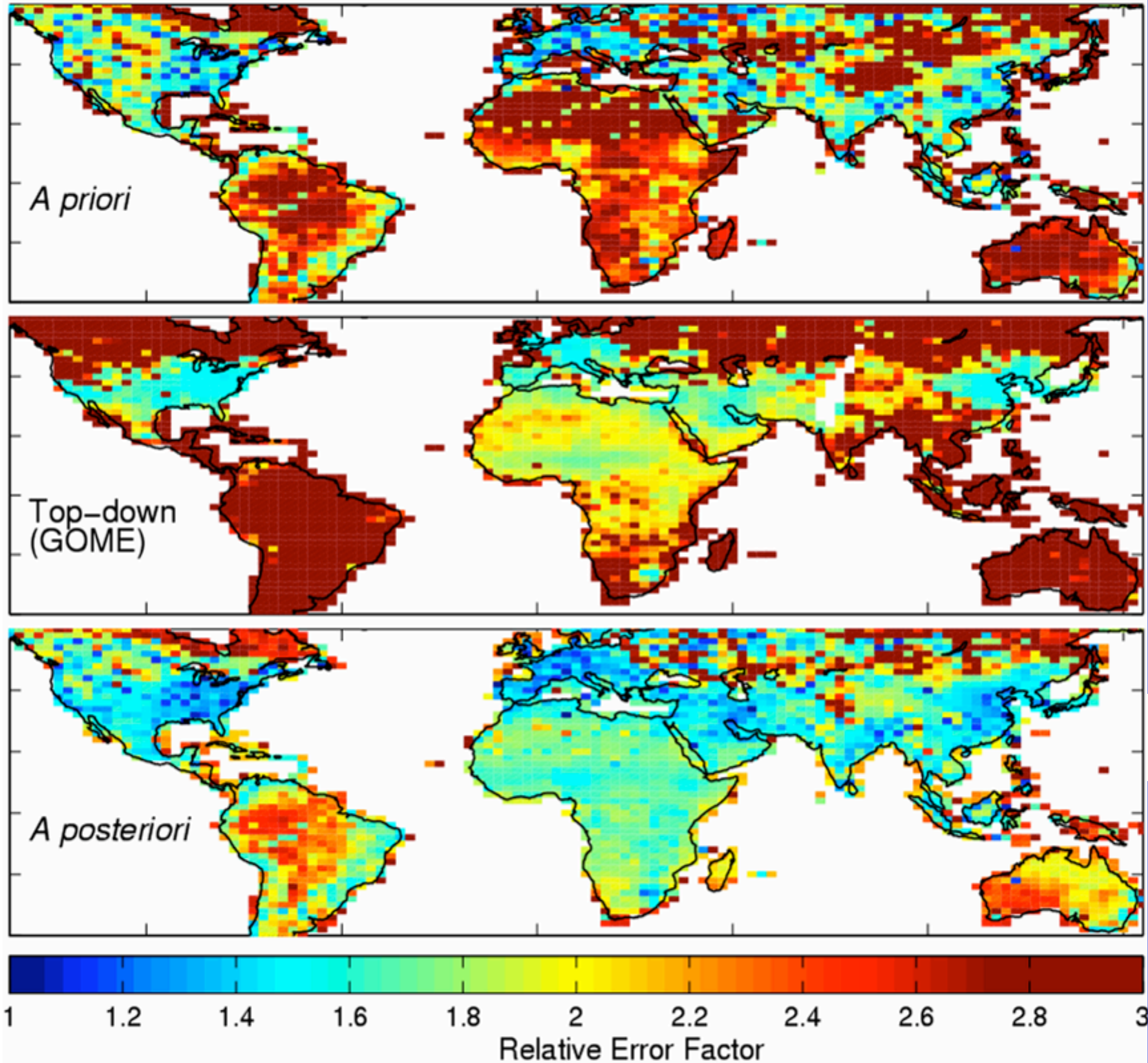
→Air quality forecasting?

GLOBAL INVENTORY OF NITROGEN OXIDE EMISSIONS CONSTRAINED BY SPACE-BASED OBSERVATIONS OF NO_2 COLUMNS; GLOBAL PARTITIONING OF NO_x SOURCES




GOME Tropospheric NO_2
GEOS-CHEM Tropospheric NO_2
DJF
DJF
MAM
r=0.75
bias 5%
MAM
JJA
JJA
0
2
4
6
8
10
10^{15} molecules cm^{-2}

TOP-DOWN INFORMATION FROM GOME REDUCES ERROR IN NO_X EMISSION INVENTORY



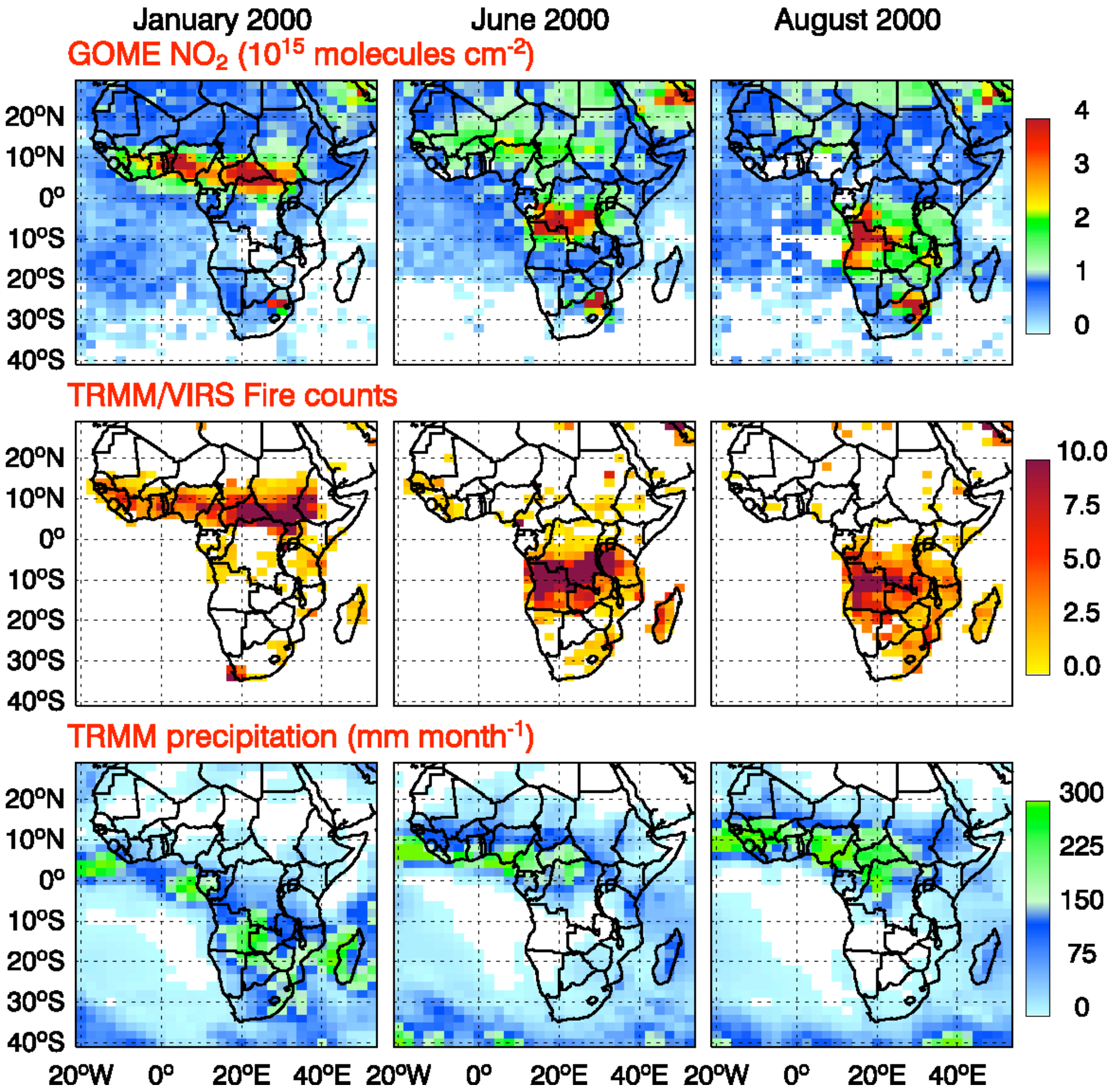
Bottom-up error ε_a
Mean=2.0

Top-down error ε_t
Mean=2.0

$$\ln^{-2}\varepsilon = \ln^{-2}\varepsilon_a + \ln^{-2}\varepsilon_t$$

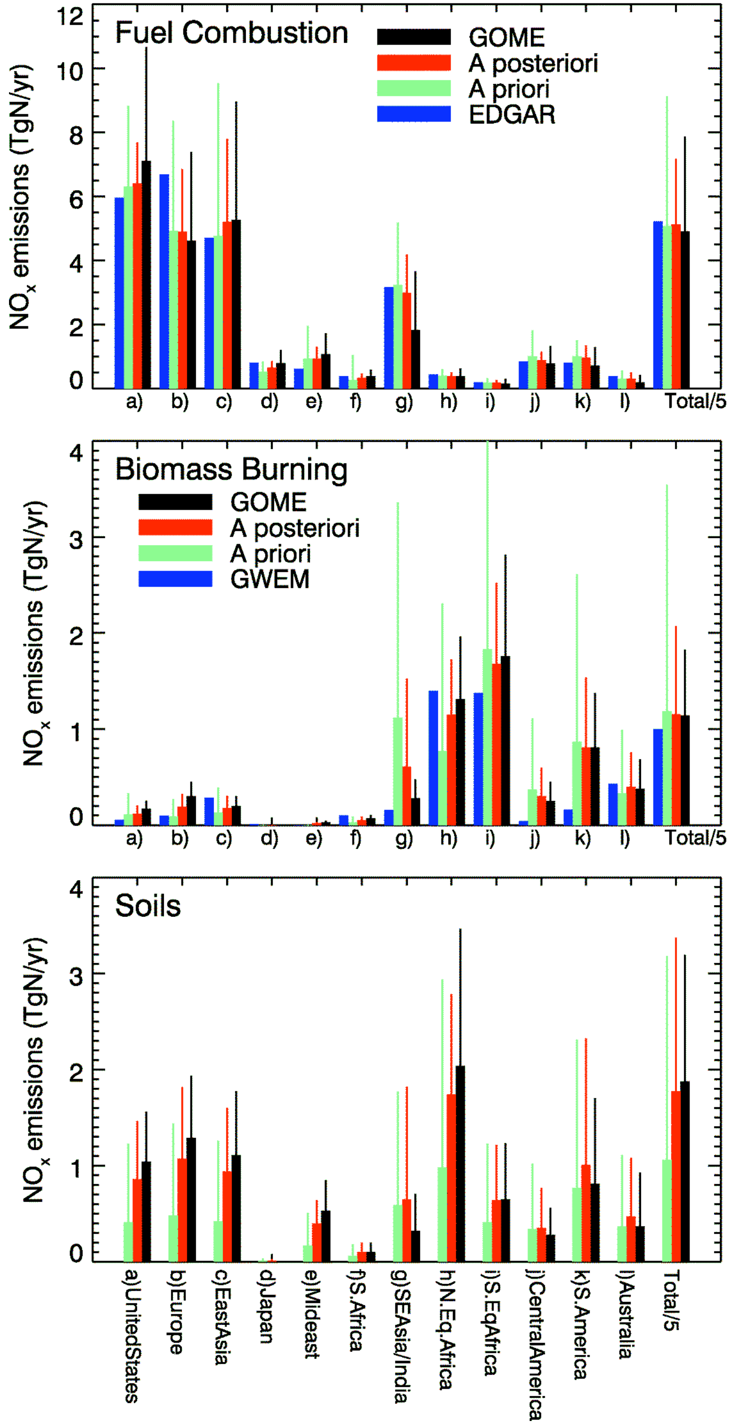
Mean=1.6

Soil NO_x



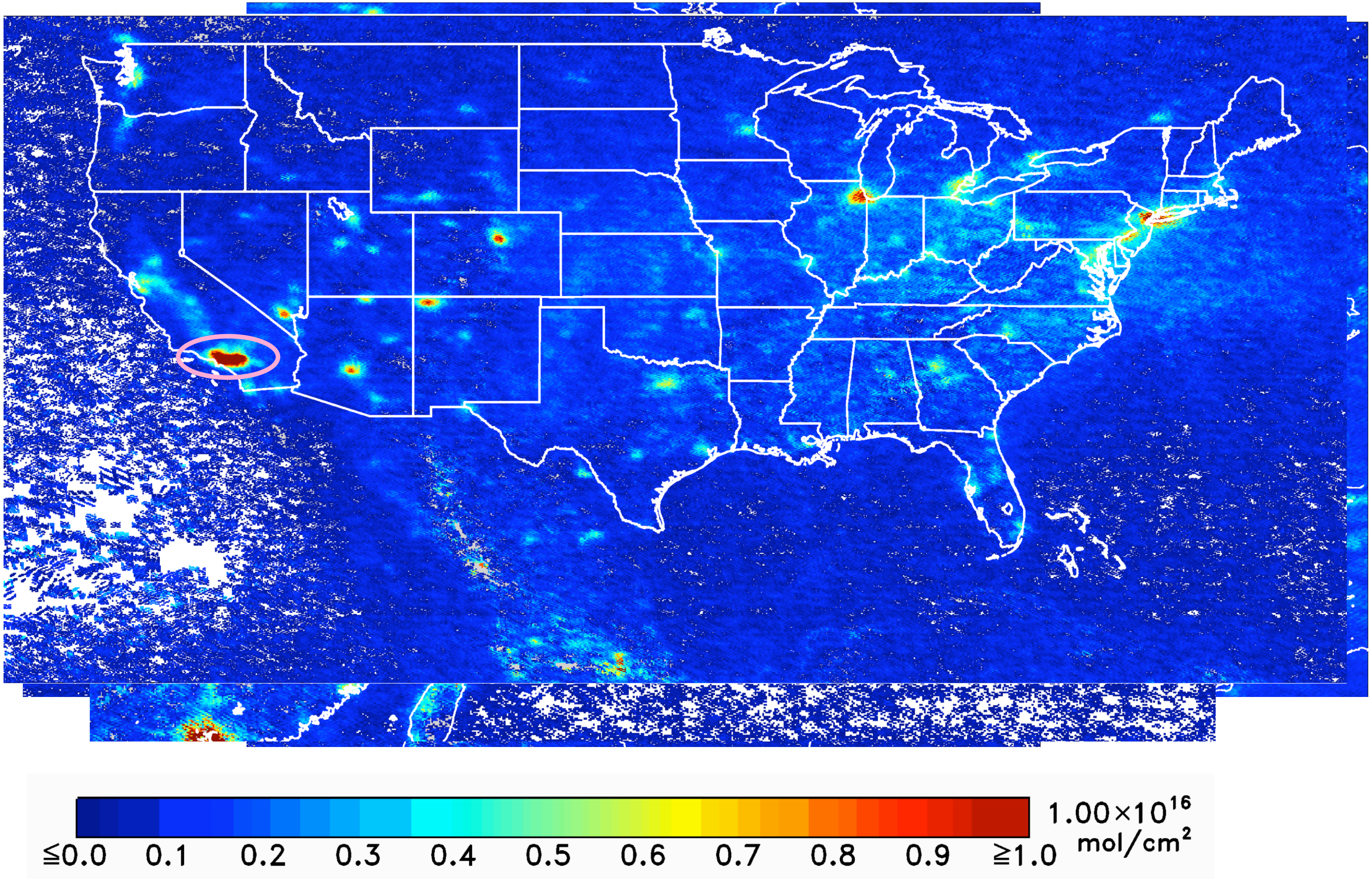
January 2000
June 2000
August 2000
GOME NO2 (10^15 molecules cm^-2)
TRMM/VIRS Fire counts
TRMM precipitation (mm month^-1)
20°N
10°N
0°
10°S
20°S
30°S
40°S
20°W
0°
20°E
40°E
4
3
2
1
0
10.0
7.5
5.0
2.5
0.0
300
225
150
75
0

Regional GOME top-down (black), a priori (green), and a posteriori (red) annual NO_x emissions in TgN / year for 2000. The thin lines on top of each bar display the absolute errors. Global emissions are divided by a factor of 5.

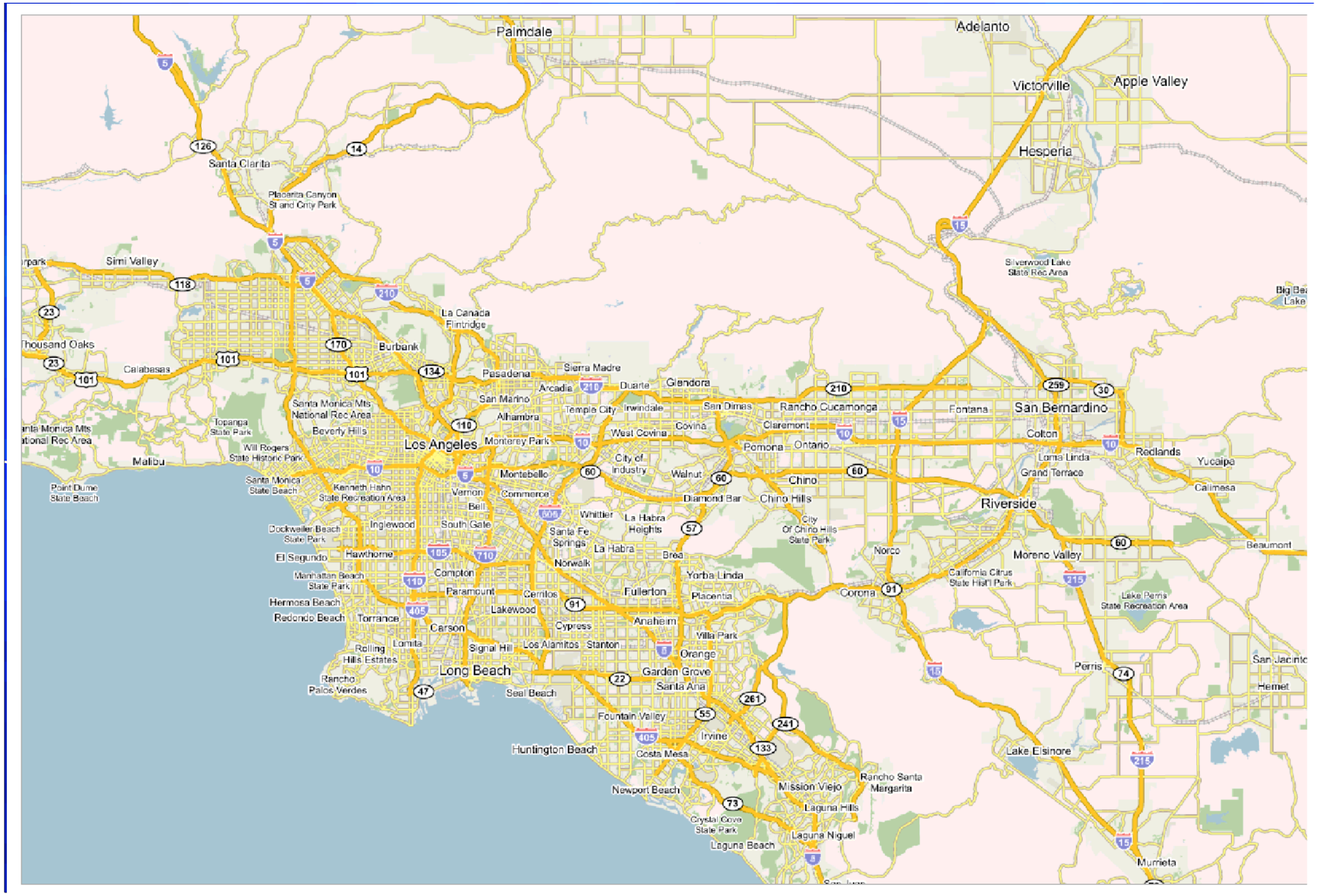


Global partitioning of NO_x sources using satellite observations: Relative roles of fossil fuel combustion, biomass burning and soil emissions, L. Jaeglé, L. Steinberger, R.V. Martin, and K. Chance, *Faraday Discuss*. 130, 407-423, 2005.

OMI Tropospheric NO_2 (July 2005)



OMI NO_2

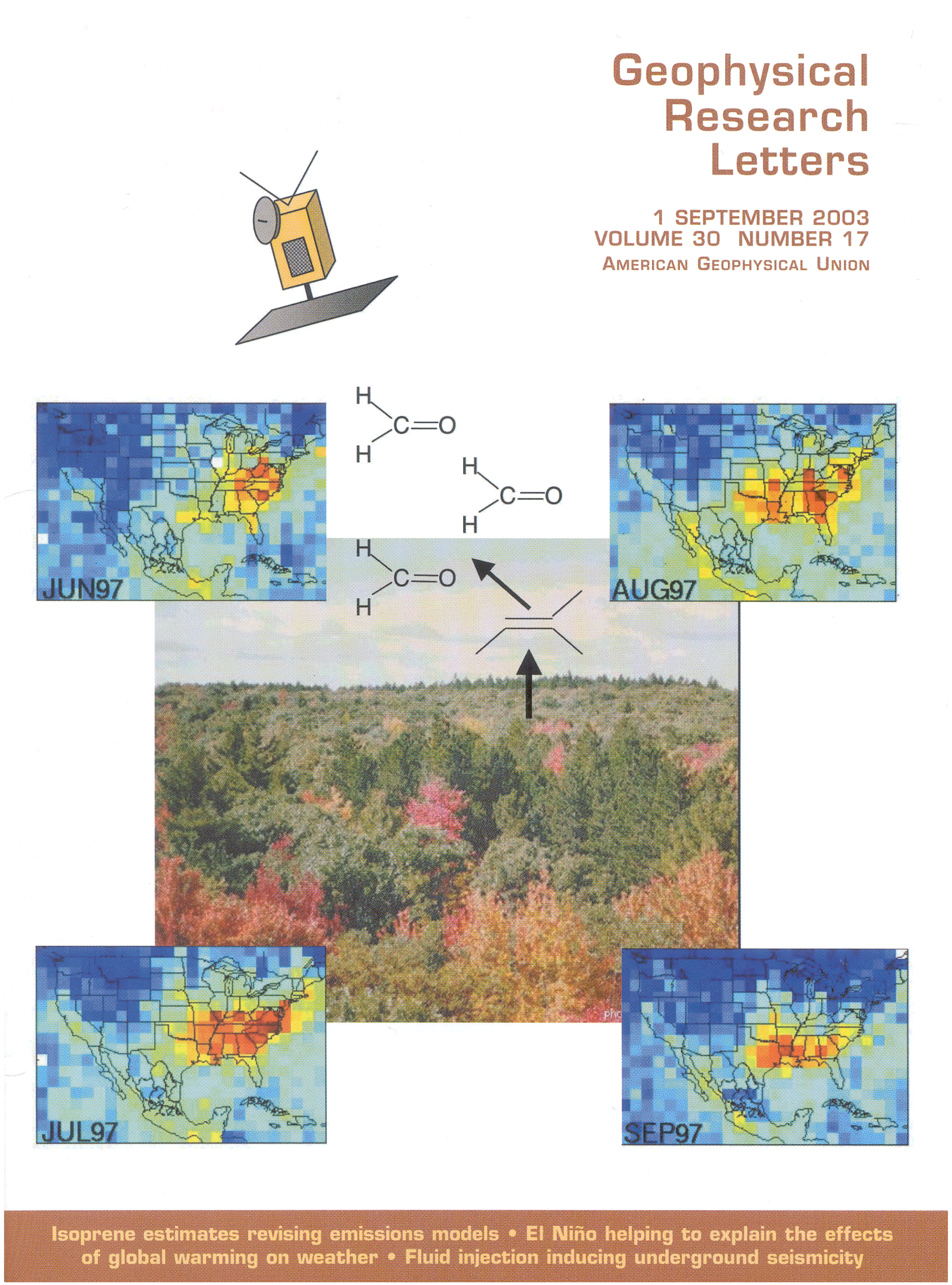

Palmdale
Adelanto
Victorville
Apple Valley
Hesperia
Santa Clarita
Simi Valley
Thousand Oaks
Calabasas
Malibu
La Canada Flintridge
Burbank
Pasadena
Sierra Madre
Arcadia
Duarte
Glendora
San Marino
Alhambra
Temple City
Irwindale
San Dimas
Rancho Cucamonga
Fontana
San Bernardino
Beverly Hills
Los Angeles
Monterey Park
West Covina
Covina
Claremont
Pomona
Ontario
Colton
Redlands
Loma Linda
Yucaipa
Grand Terrace
Calimesa
Santa Monica
Montebello
Walnut
Chino
Chino Hills
Diamond Bar
Riverside
Inglewood
South Gate
Whittier
La Habra Heights
La Habra
Norco
Moreno Valley
Beaumont
El Segundo
Hawthorne
Compton
Norwalk
Brea
Yorba Linda
Manhattan Beach
Paramount
Cerritos
Fullerton
Placentia
Corona
Hermosa Beach
Redondo Beach
Torrance
Lakewood
Anaheim
Cypress
Carson
Villa Park
Orange
Long Beach
Los Alamitos
Stanton
Garden Grove
Santa Ana
Seal Beach
Perris
San Jacinto
Hemet
Fountain Valley
Irvine
Huntington Beach
Costa Mesa
Lake Elsinore
Rancho Santa Margarita
Newport Beach
Mission Viejo
Laguna Hills
Laguna Niguel
Laguna Beach
Murrieta

NO_2 / NO_x: The Future

- Continually improved inventories and partitioning
  - Higher spatial resolution
- Trends in NO_x emissions
- Account for transport in the inversion of NO_2 to NO_x

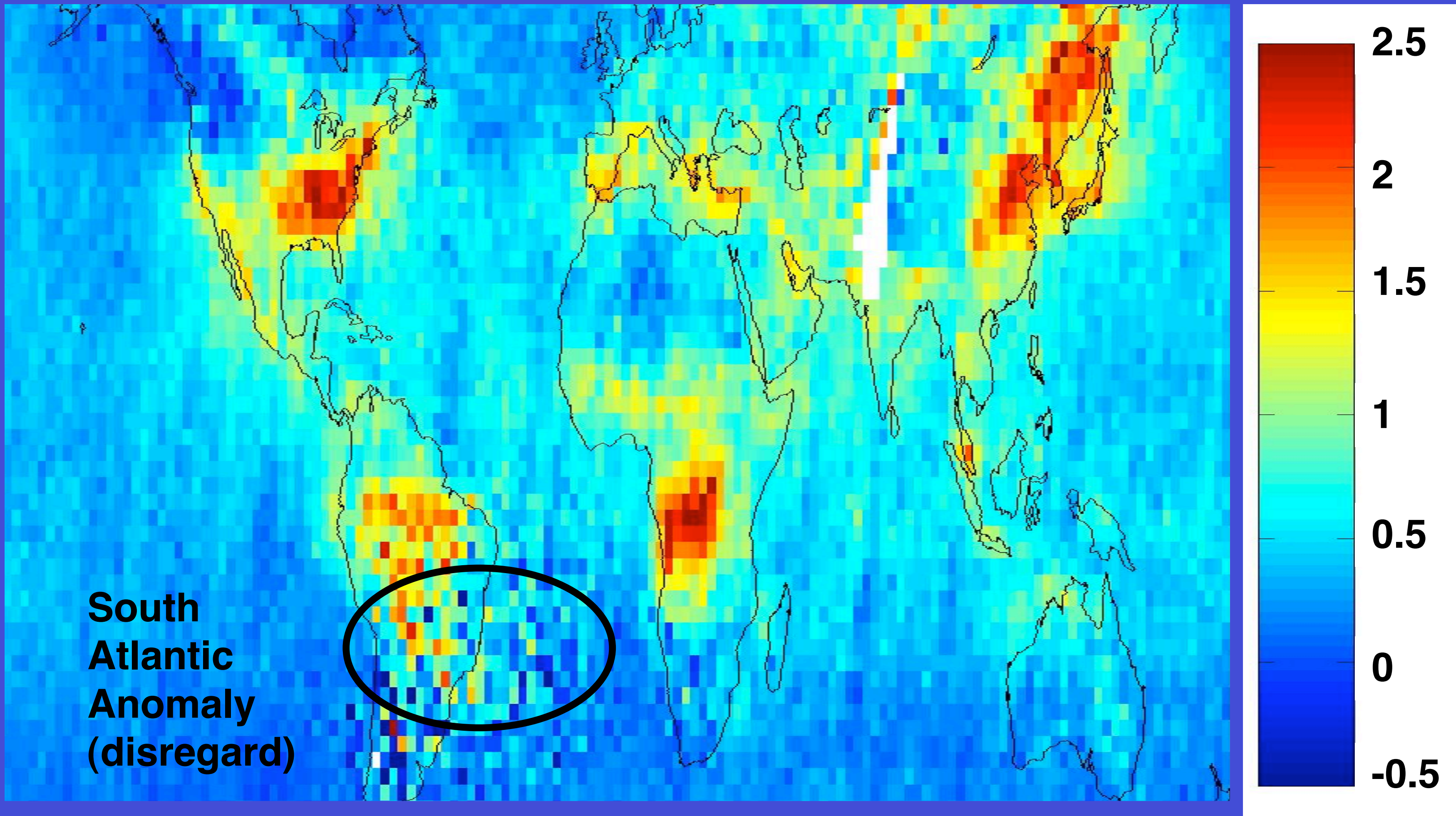
→ Air quality forecasting

VOC EMISSION INVENTORIES DERIVED FROM GOME FORMALDEHYDE MEASUREMENTS

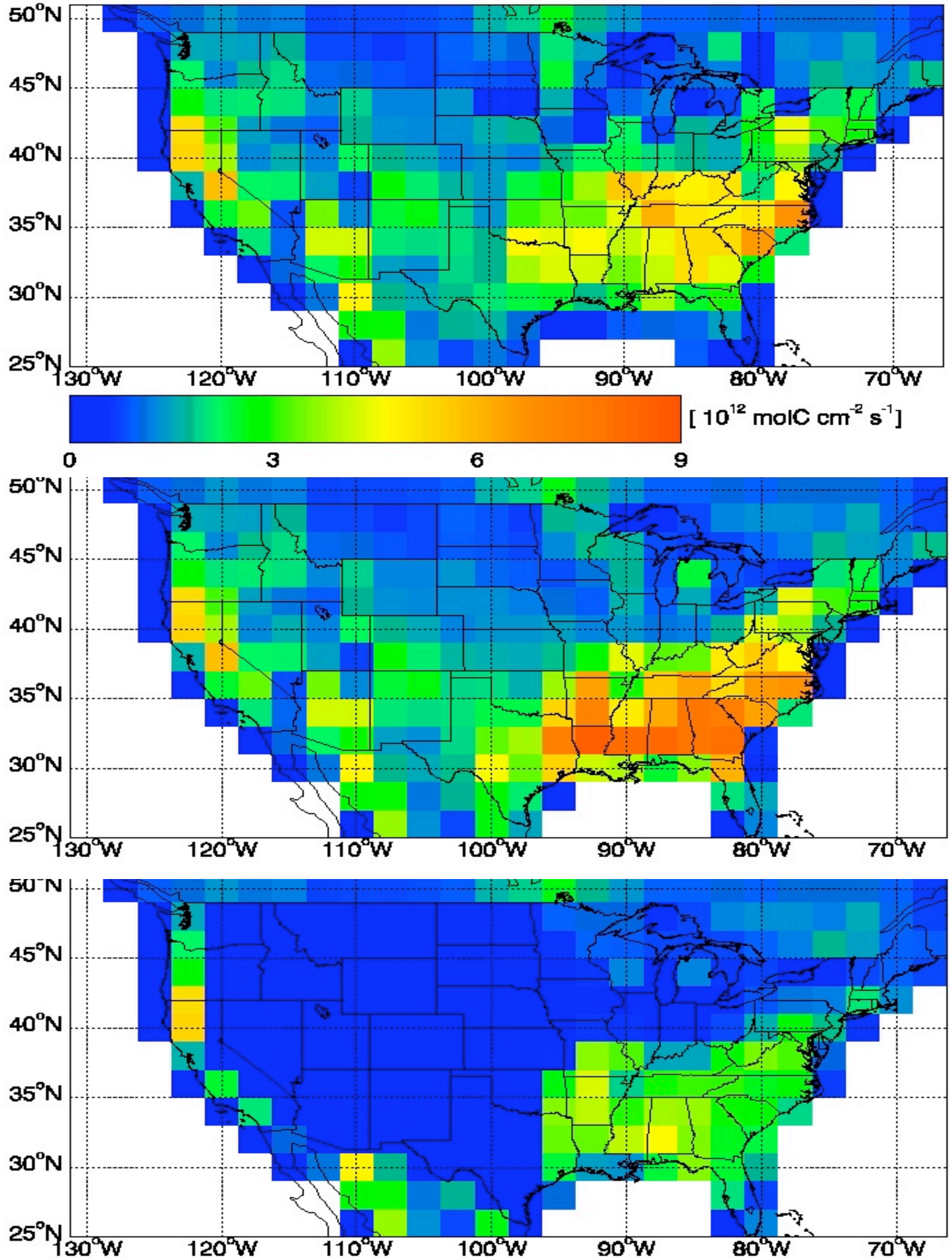


HCHO COLUMNS MEASURED BY GOME (JULY 1996)

Units of 10^{16} molecules cm^{-2};
Uncertainty ~ $1x10^{16}$ molecules cm^{-2} (1-2 ppbv in 2-km boundary layer)



High HCHO regions reflect VOC emissions from fires, biosphere, human activity



ISOPRENE EMISSION INVENTORIES, JULY1996

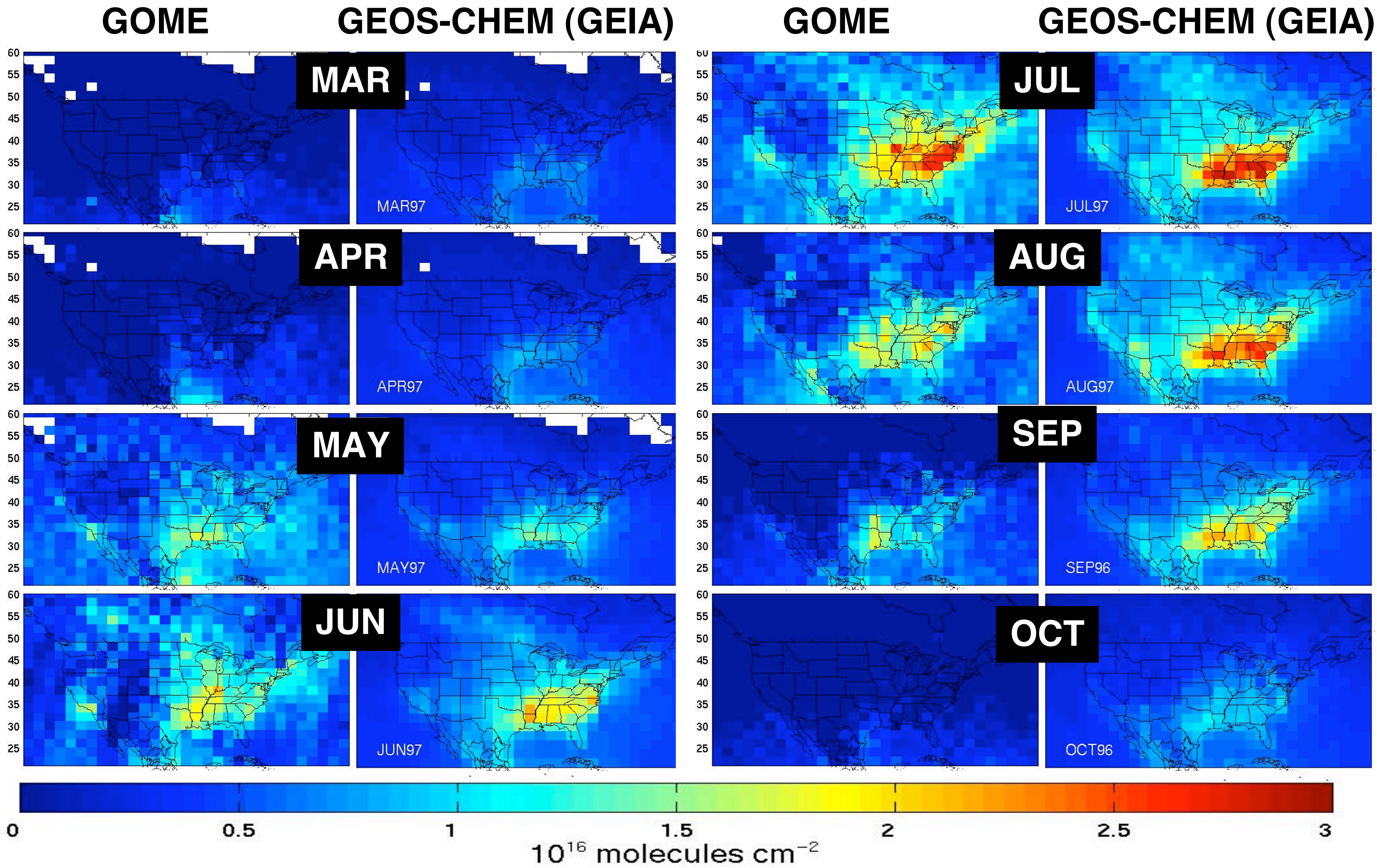
GOME top-down (5.7 Tg)

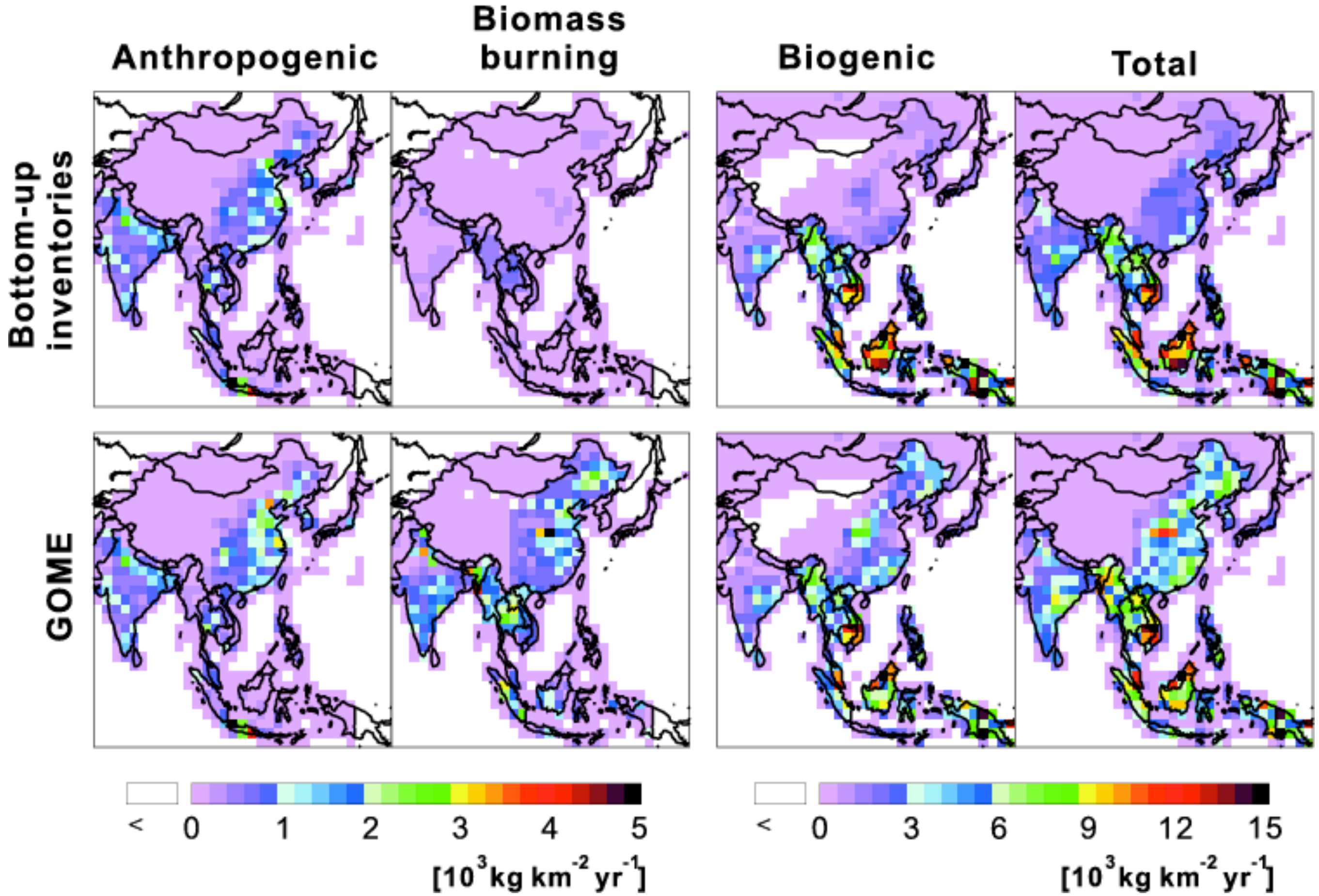
GEIA (7.1 Tg)

BEIS2 (2.6 Tg)

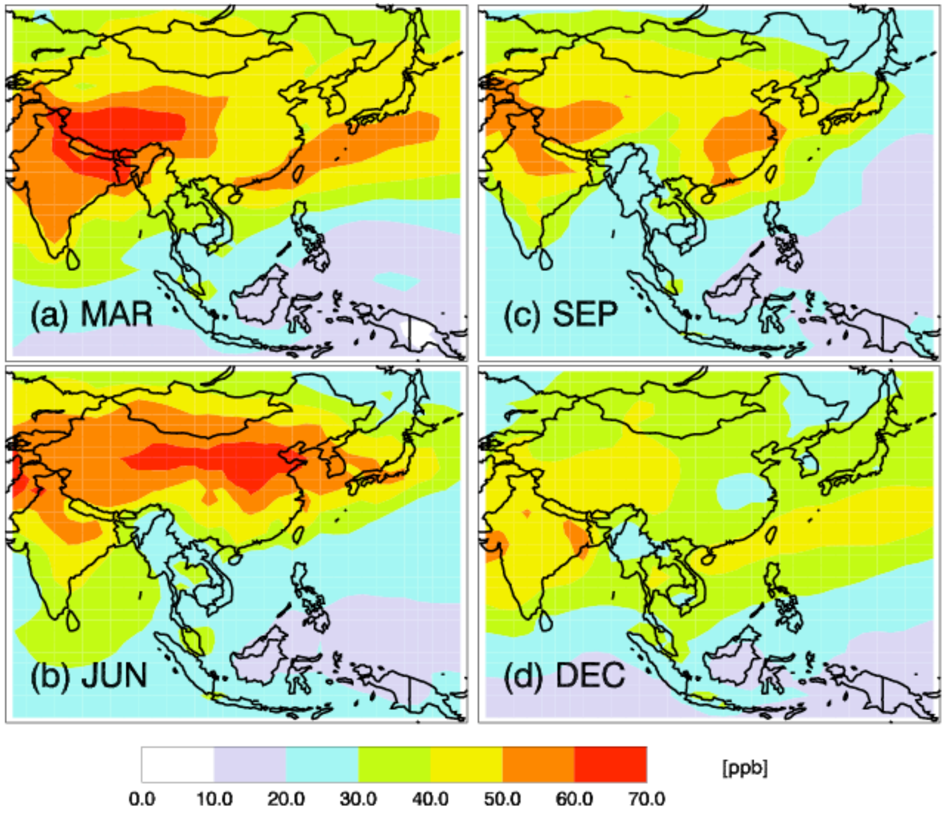
SEASONALITY OF GOME HCHO COLUMNS (9/96-8/97)

Largely reflects seasonality of isoprene emissions;
general consistency with GEIA but also some notable differences

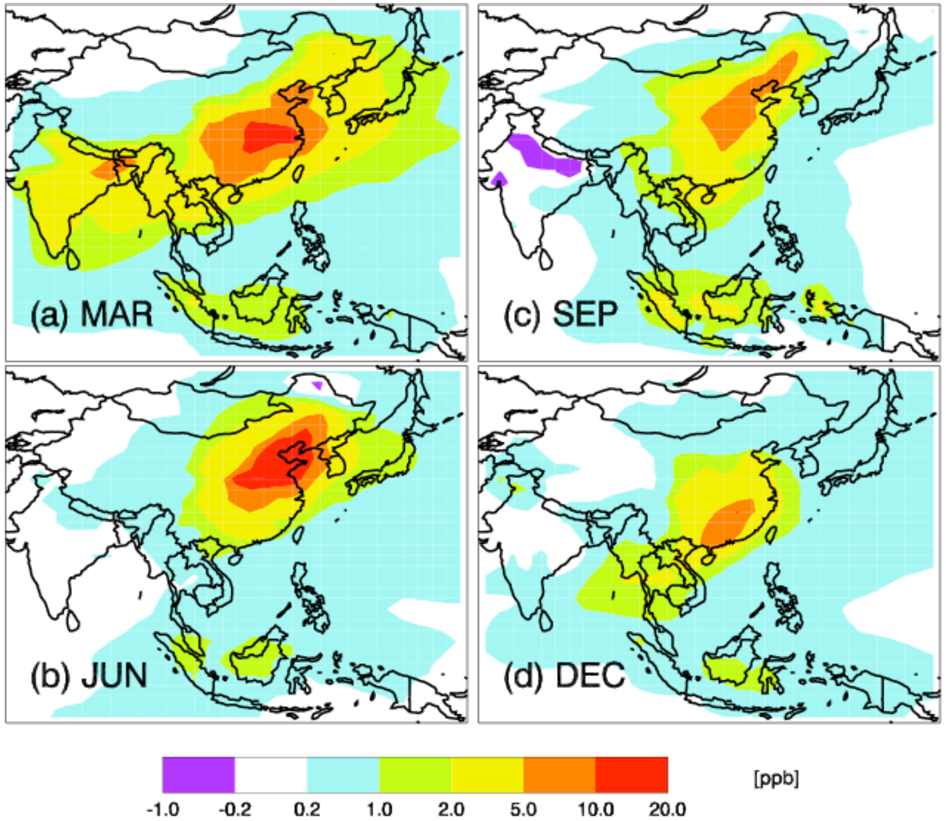




Reactive NMVOC emissions from East and South Asia. Upper panels: bottom-up inventories of Streets *et al*. [2003a] (anthropogenic, biomass burning) and Guenther *et al*. [2006] (biogenic). Bottom panels: emissions inferred from the GOME HCHO observations in this study. Color scales: left - anthropogenic and biomass burning; right - biogenic and total sources. (Courtesy T.-M. Fu)

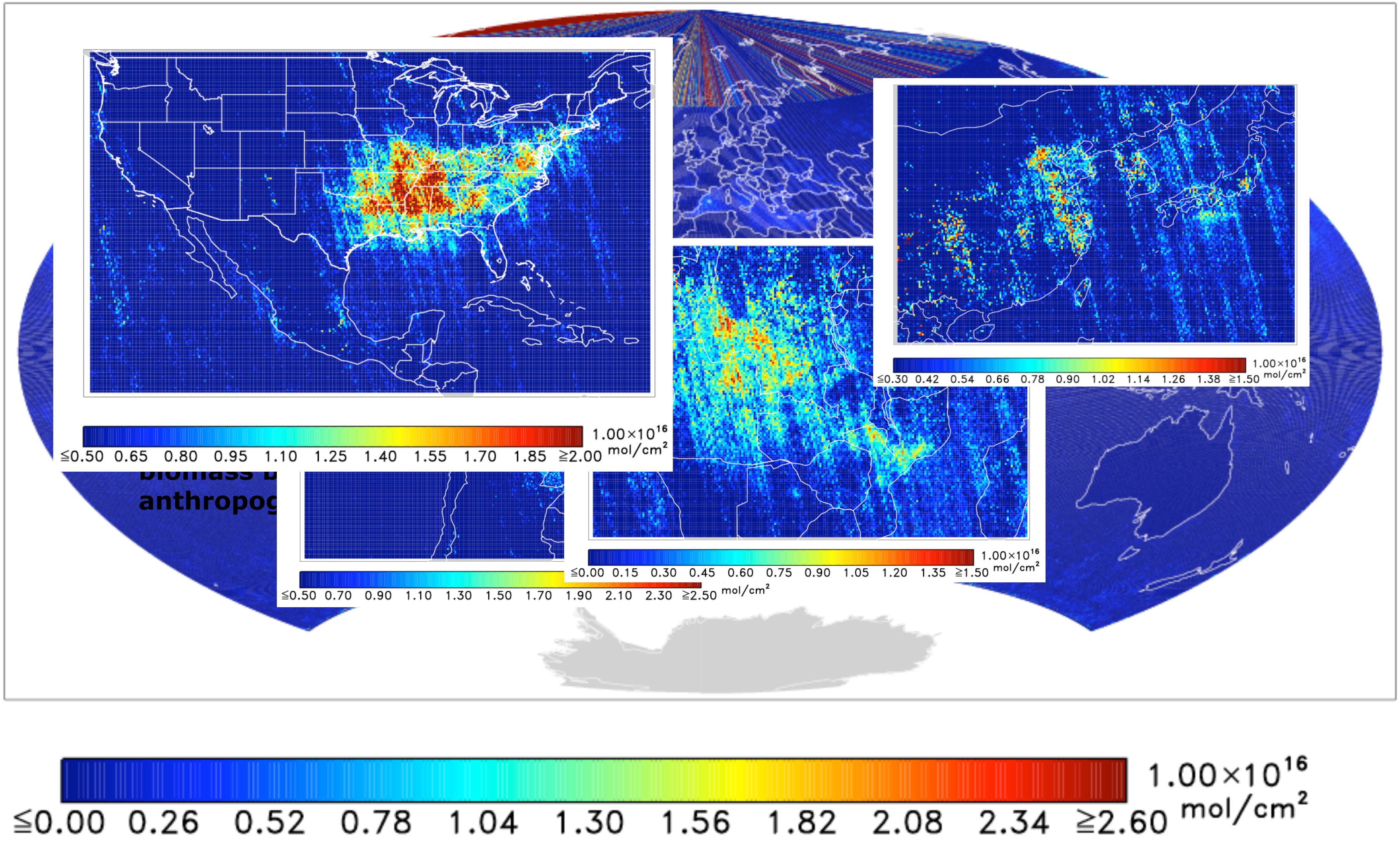


Monthly mean afternoon (13:00 to 17:00 local time) surface ozone concentrations simulated by GEOS-Chem using bottom-up inventories for NMVOCs in (a) March, (b) June, (c) September, and (d) December, 2001. (Courtesy T.-M. Fu)

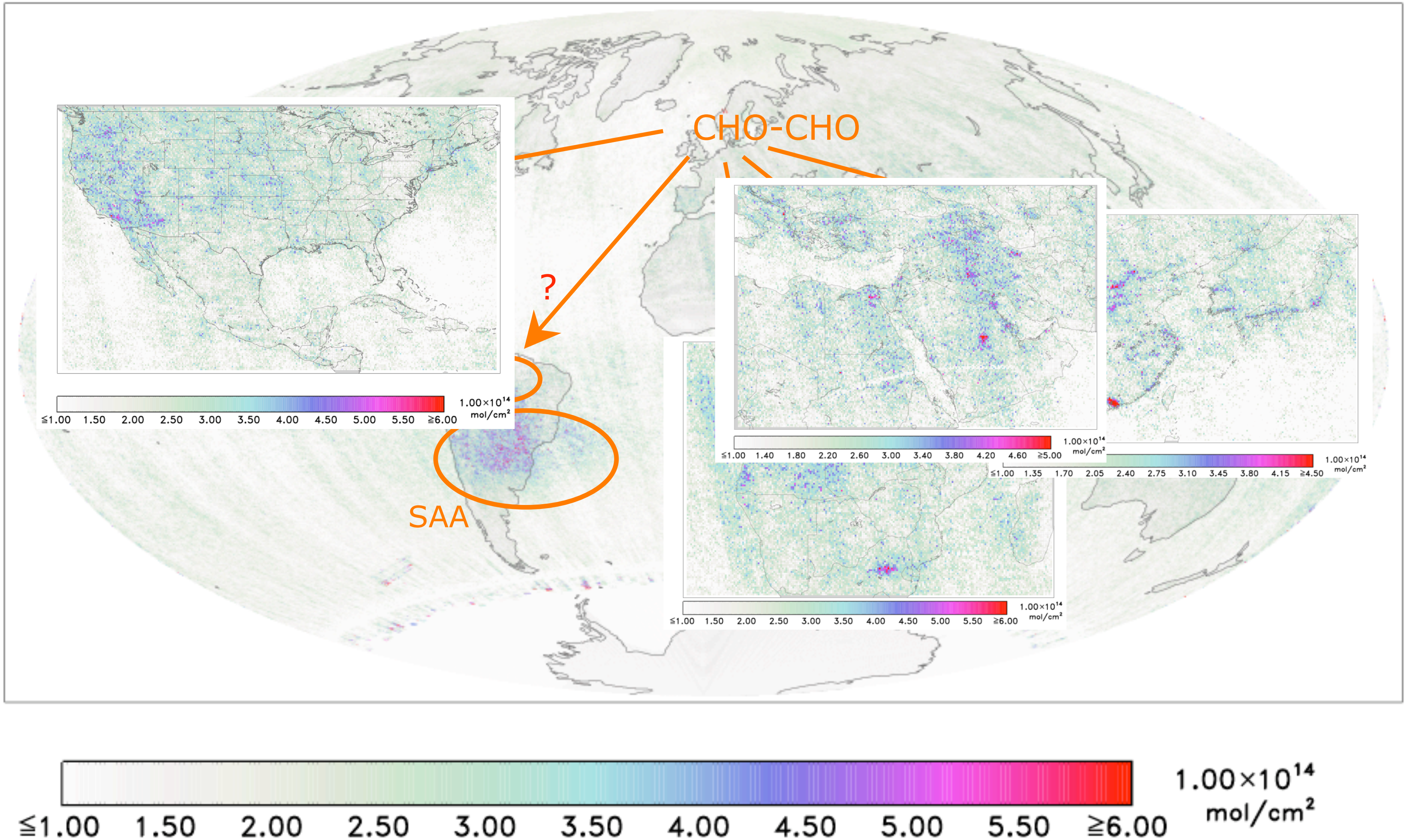


Difference in modeled monthly mean afternoon (13:00 to 17:00 local time) surface ozone concentrations using GOME-inferred reactive NMVOC emission versus the bottom-up inventories for (a) March, (b) June, (c) September, and (d) December, 2001. (Courtesy T.-M. Fu)

HCHO — monthly average 08/2005



OMI CHO-CHO Geometric Vertical Column for July 2005



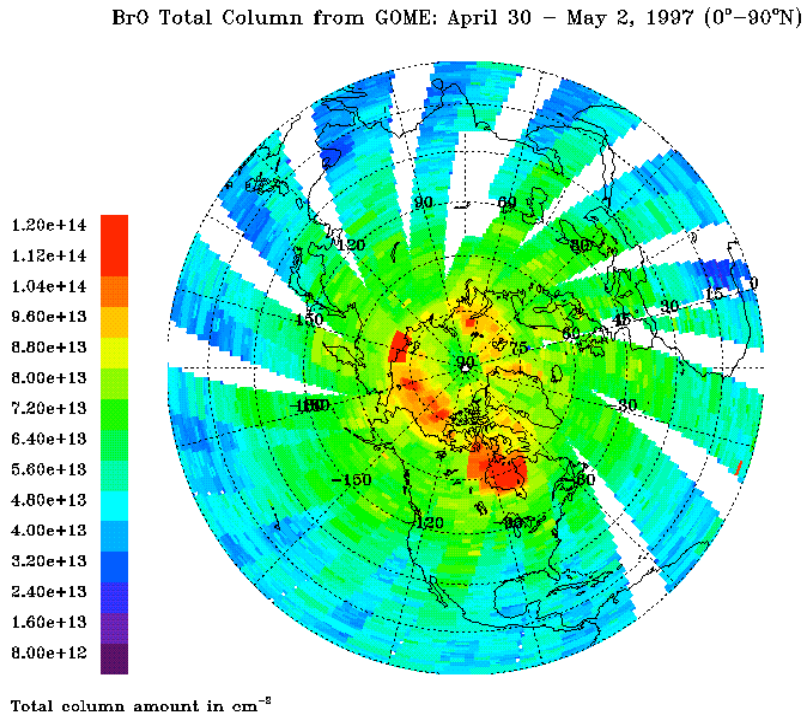
HCHO / VOCs: The Future

- Continually improved inventories and partitioning
  - Higher spatial resolution (measurements and models)
- Trends in VOC emissions
- GOME-2 CHOCHO? (0930 Xing)

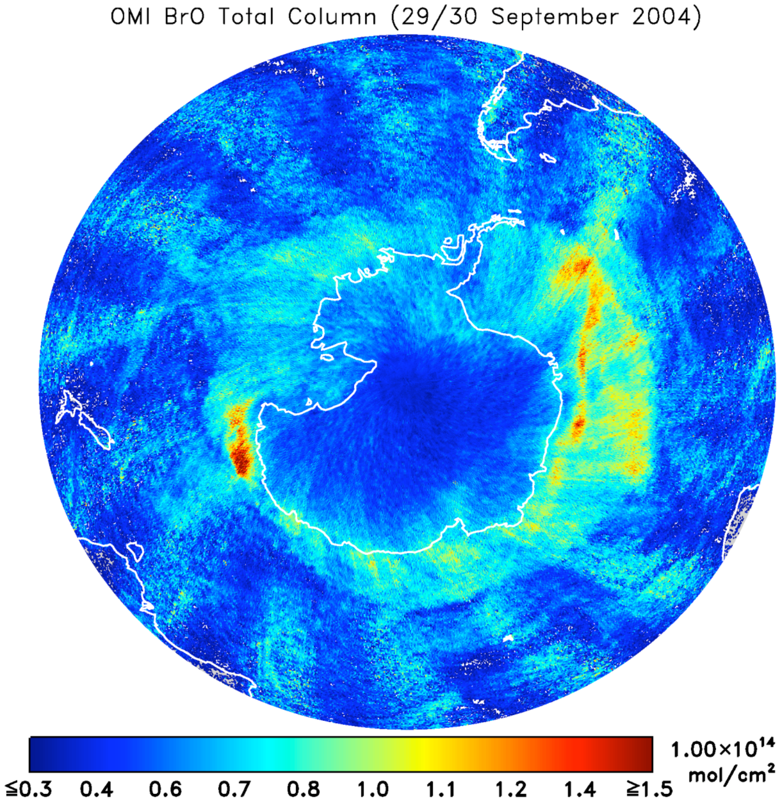
→Air quality forecasting

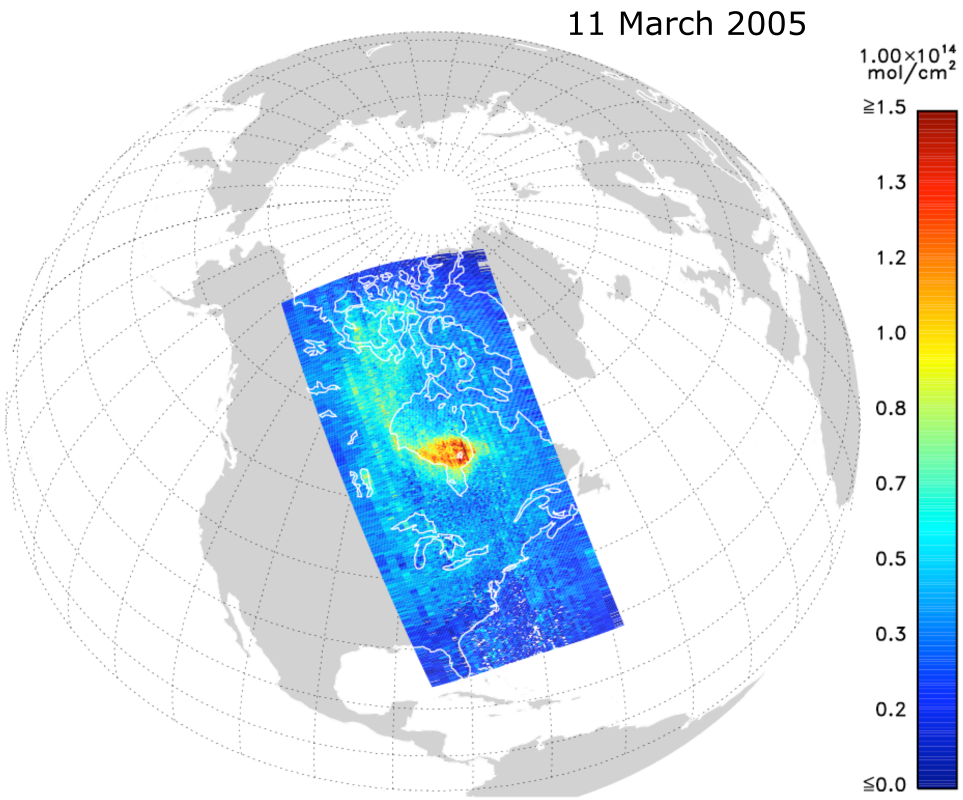
Widespread persistent near-surface O_3 depletion at northern high latitudes in spring

- BrO is a strong source of O_3 destruction in the stratosphere.
- BrO is measured globally by GOME and OMI
- Enhanced tropospheric BrO has been observed over the Arctic and Antarctic ice pack in the polar spring.

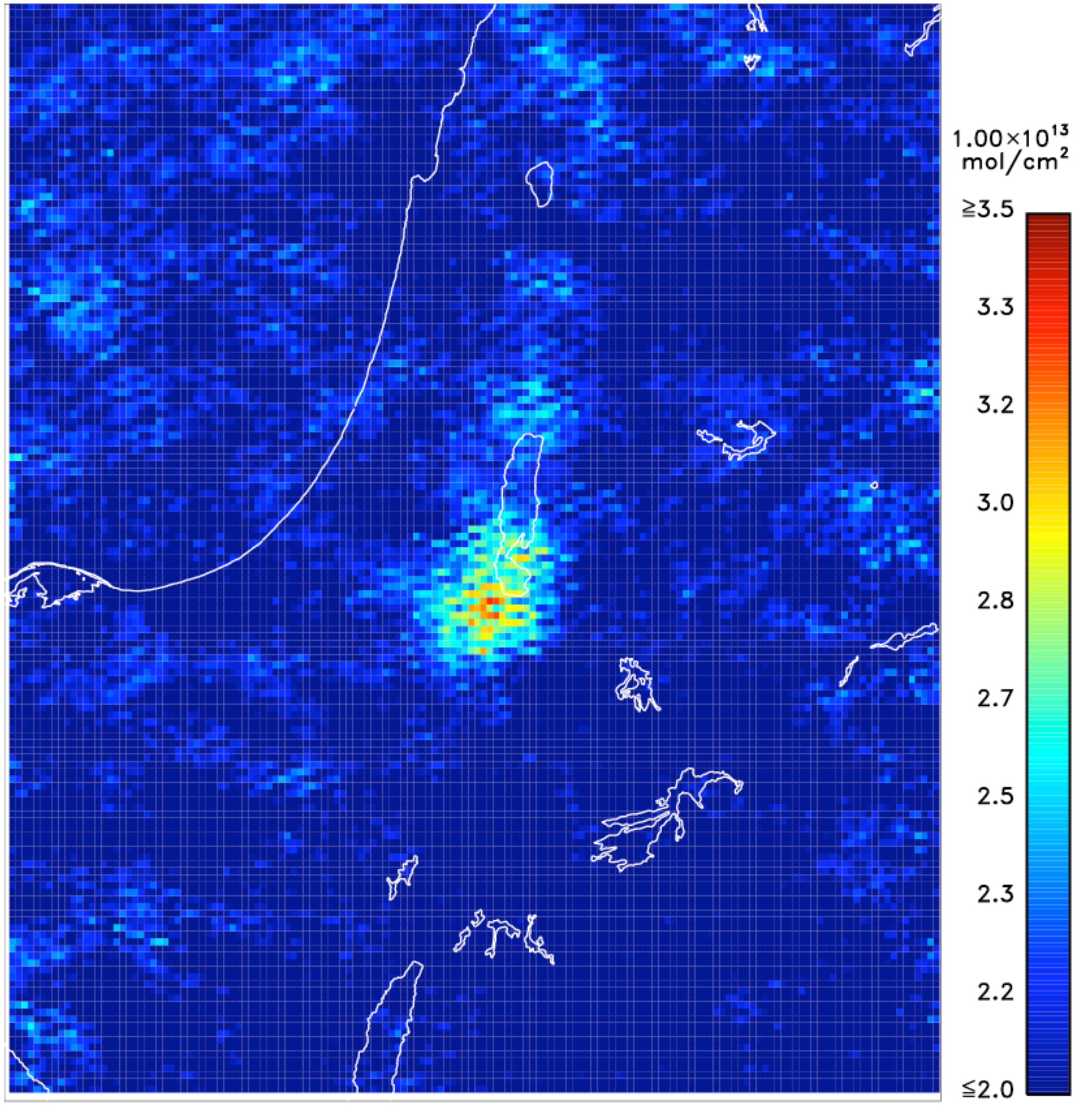


OMI BrO Tropospheric Shelf Ice



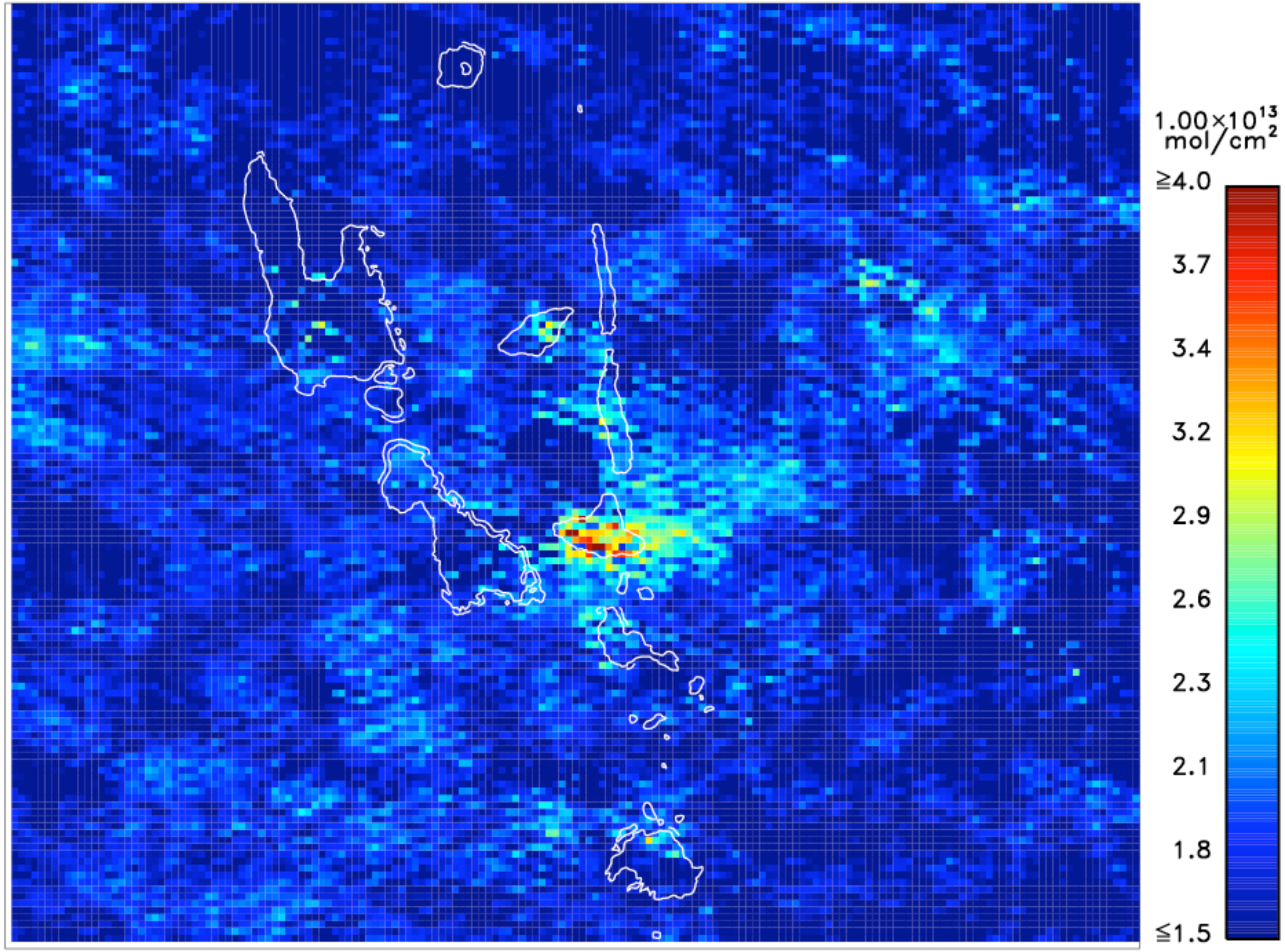


OMI BrO Tropospheric Salt Lakes 1st Observation from Satellite



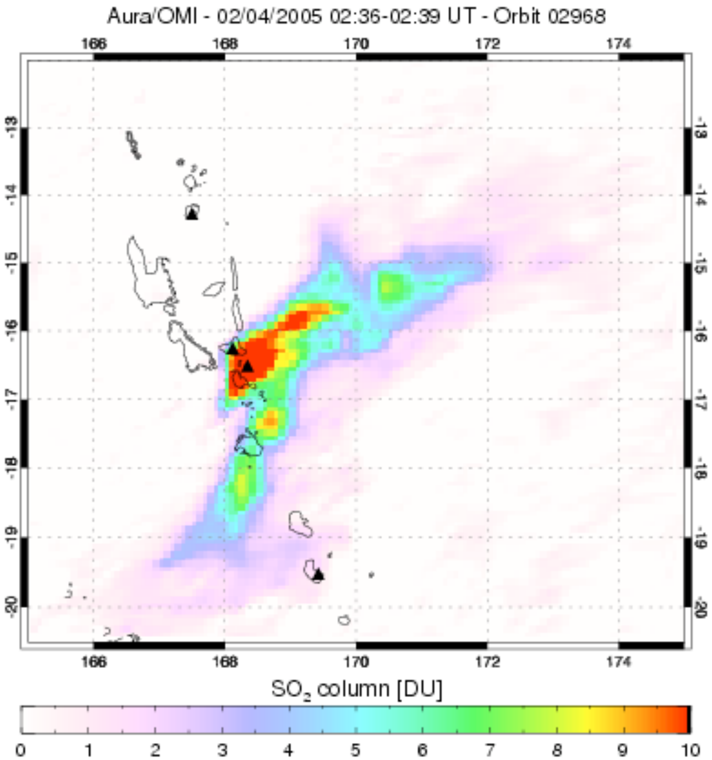
OMI BrO Volcanoes ... 1st Observation from Satellite

Ambrym: First satellite-based BrO observation in volcanic plumes!

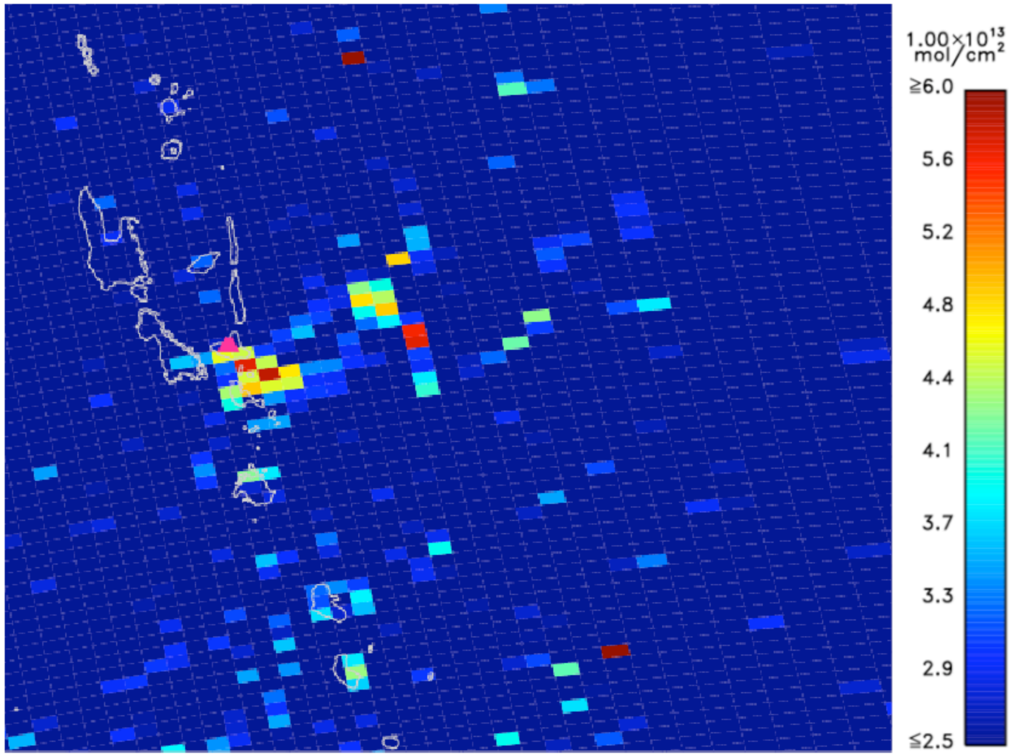


BrO Tropospheric Volcanoes … 1st Observation from Satellite

Ambrym Eruption: 4th February 2005, OMI Granule 02968



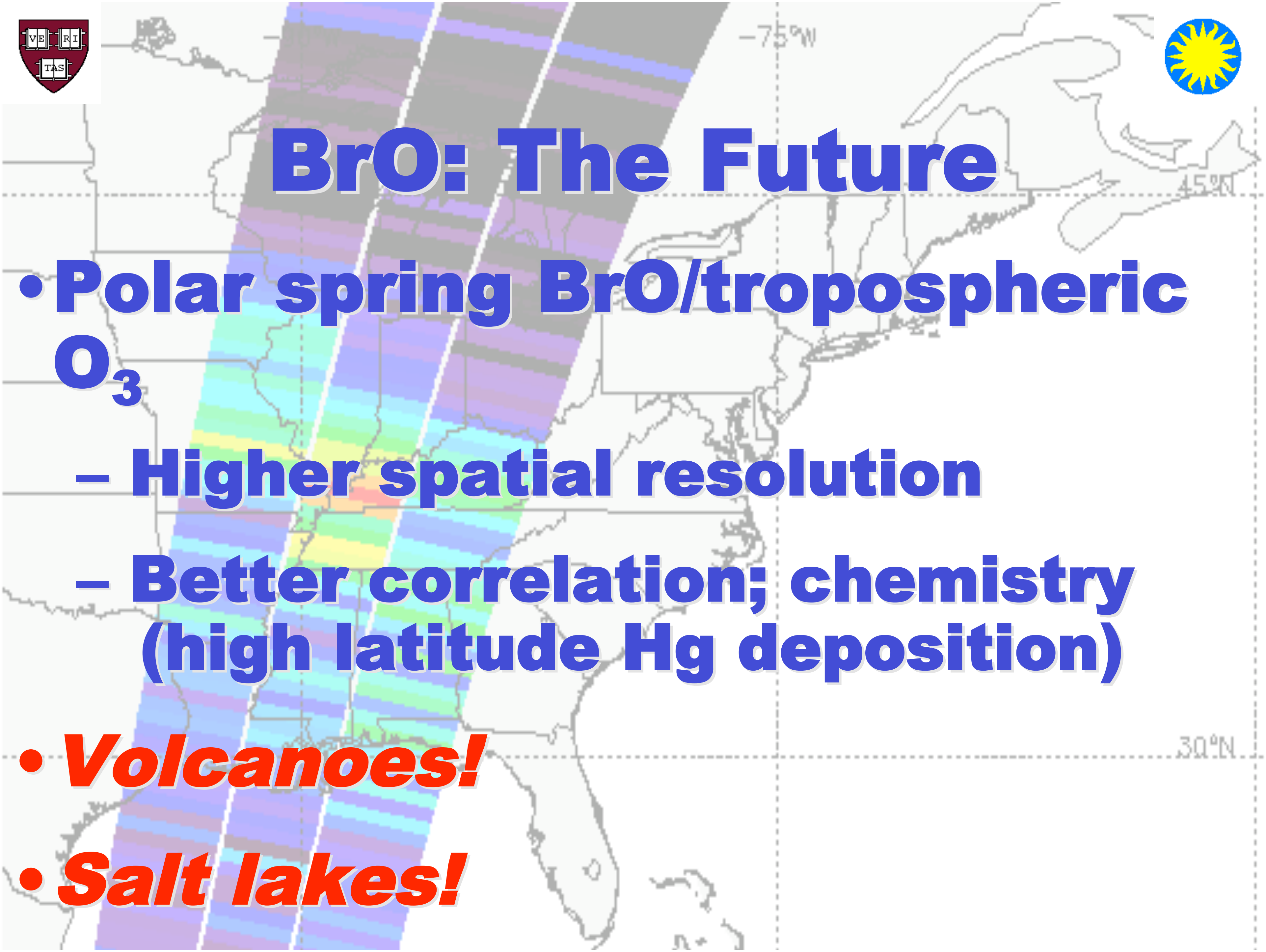
SO_2 courtesy of Simon Carn, UMBC



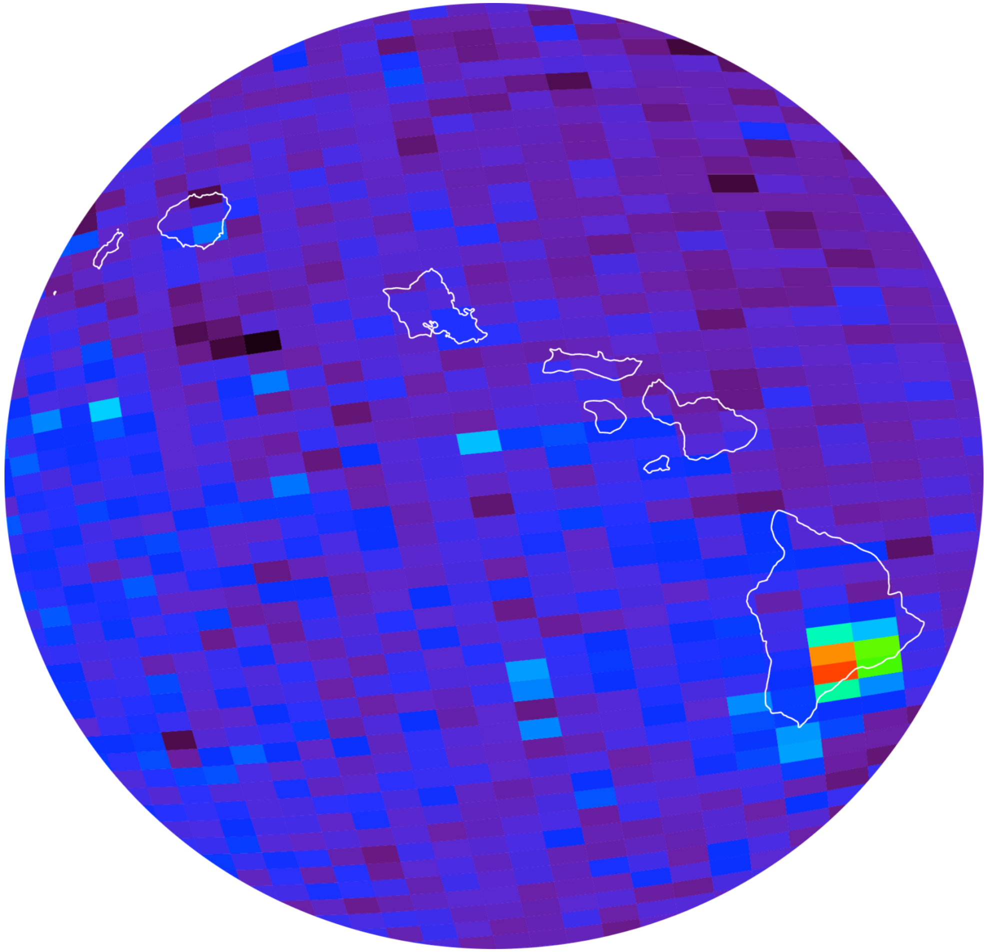
BrO

BrO: The Future

- Polar spring BrO/tropospheric O_3
  - – Higher spatial resolution
  - – Better correlation; chemistry (high latitude Hg deposition)
- ***Volcanoes!***
- ***Salt lakes!***



SO_2 Tropospheric Volcanoes



Kilauea activity, source of the VOG event in Honolulu on 9 November 2004

→Air quality forecasting

Conclusions and Future Directions

- **Improved laboratory (and solar) measurements!**
- **Additional species measurements**
  - **CHOCHO in progress**
- **Process studies for US (seasonal variation), Amazon and African biomass burning, soil NO_x emissions, Asian pollution, intercontinental pollution transport, polar springtime chemistry, air quality forecasts, ….**
- **Future instruments**
  - **High spatial resolution (GOME-2, OMPS)**
  - **Geostationary pollution monitoring?**

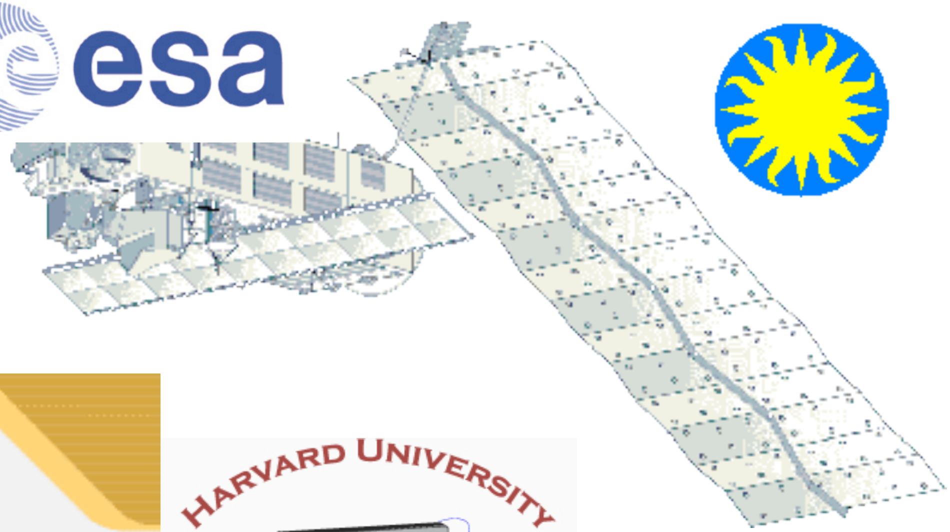
DALHOUSIE University

VE RI TAS

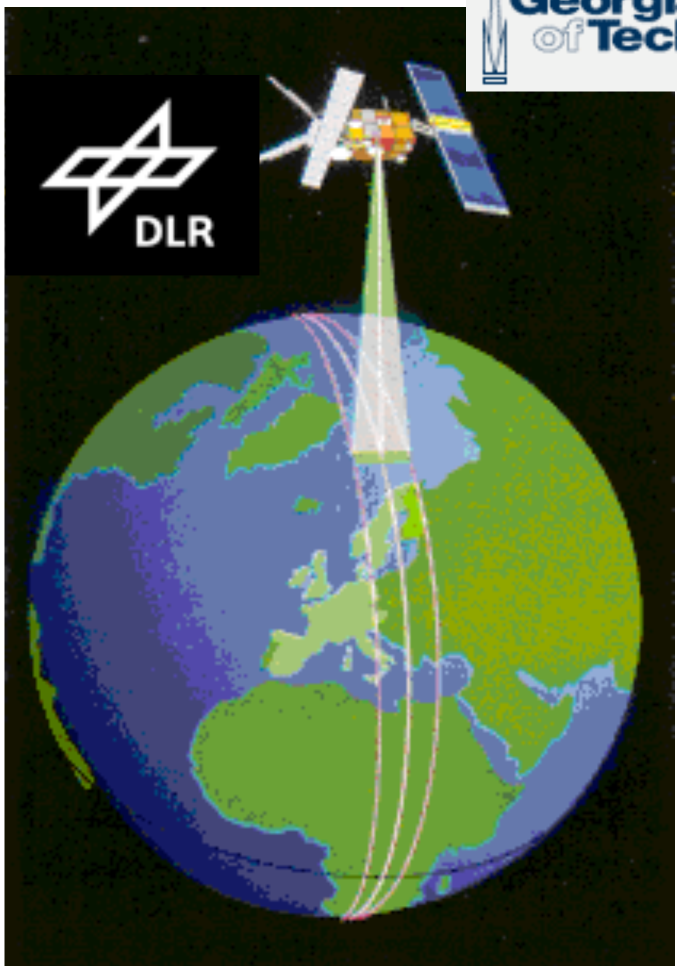
SCIAMACHY

esa

The End!



Georgia Institute of Technology



UNIVERSITY OF WASHINGTON

HARVARD UNIVERSITY

ATMOSPHERIC CHEMISTRY MODELING GROUP

NCAR

OMI

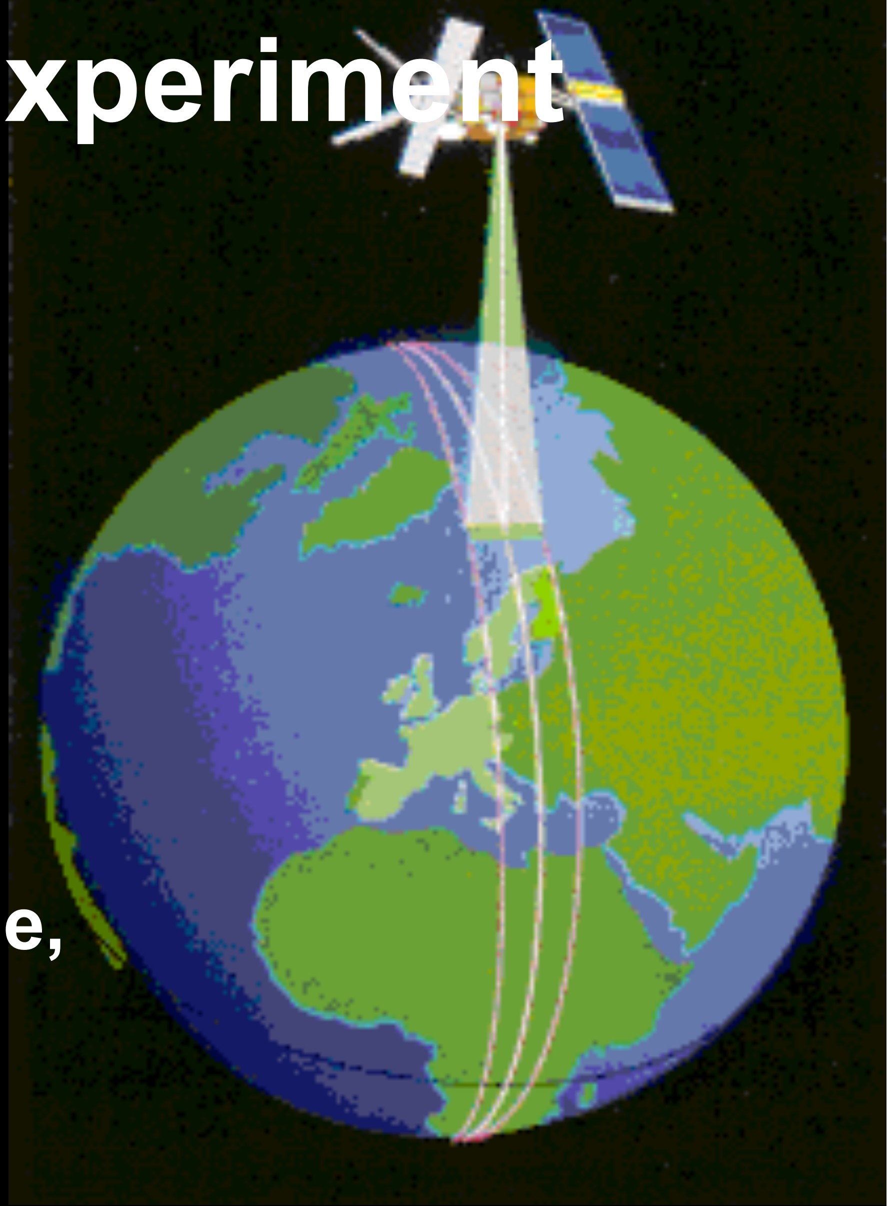
NASA

EOS AURA

Chemistry & Climate

ESA Global Ozone Monitoring Experiment

- Nadir-viewing UV/vis/NIR
  - 240-400 nm @ 0.2 nm
  - 400-790 nm @ 0.4 nm
- Launched April 1995
- Footprint 320 x 40 km^2
- 10:30 am cross-equator time, descending node
- Global coverage in 3 days



SCIAMACHY

- German/Dutch/Belgian Atmospheric Spectrometer
- 2002 launch on ESA Envisat
- Adds (to GOME) continuous coverage to 1700 nm, plus IR bands at 2.0 μm (CO_2) and 2.4 μm (CO, N_2O)
- Higher spatial resolution footprint than GOME (as good as 30 × 60 km^2)
- Adds limb scattering and limited solar occultation measurements
  - Nadir-limb subtraction improves tropospheric measurements
- Data and validation are still in a preliminary stage