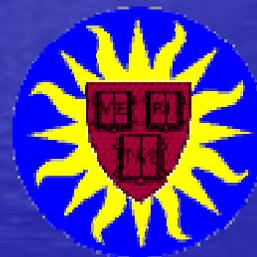


Measuring Tropospheric NO₂ from **SCIAMACHY** During **INTEX** and Improving NO_x Emission Inventories

Randall Martin
Aaron Van Donkelaar



Chris Sioris
Kelly Chance



Yongtao Hu
Armistead Russell



Tom Ryerson



Ron Cohen

UC Berkeley

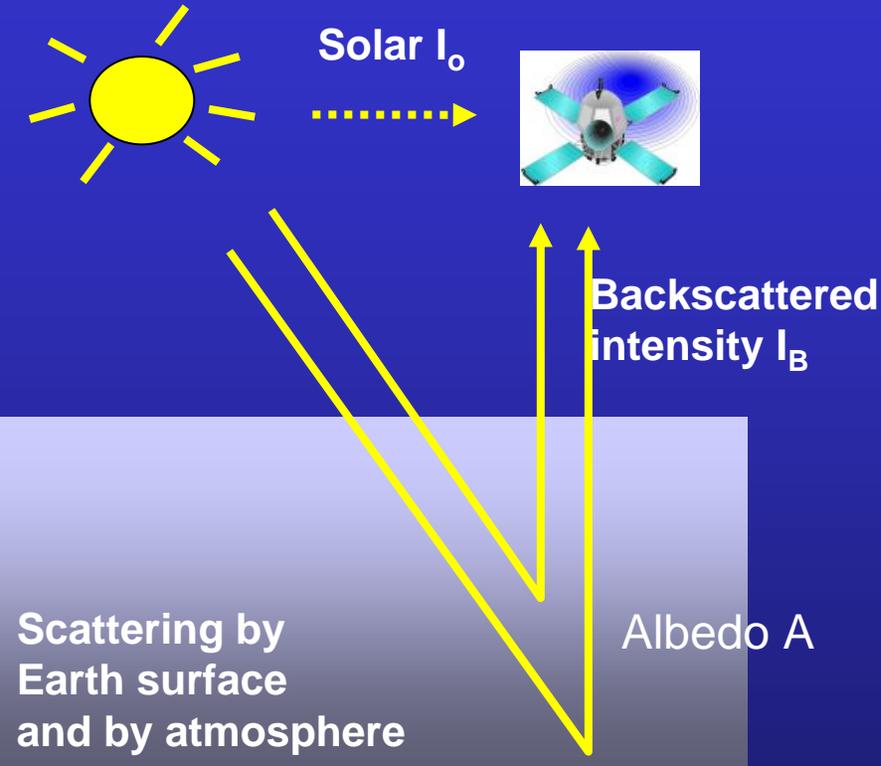
Bill Brune



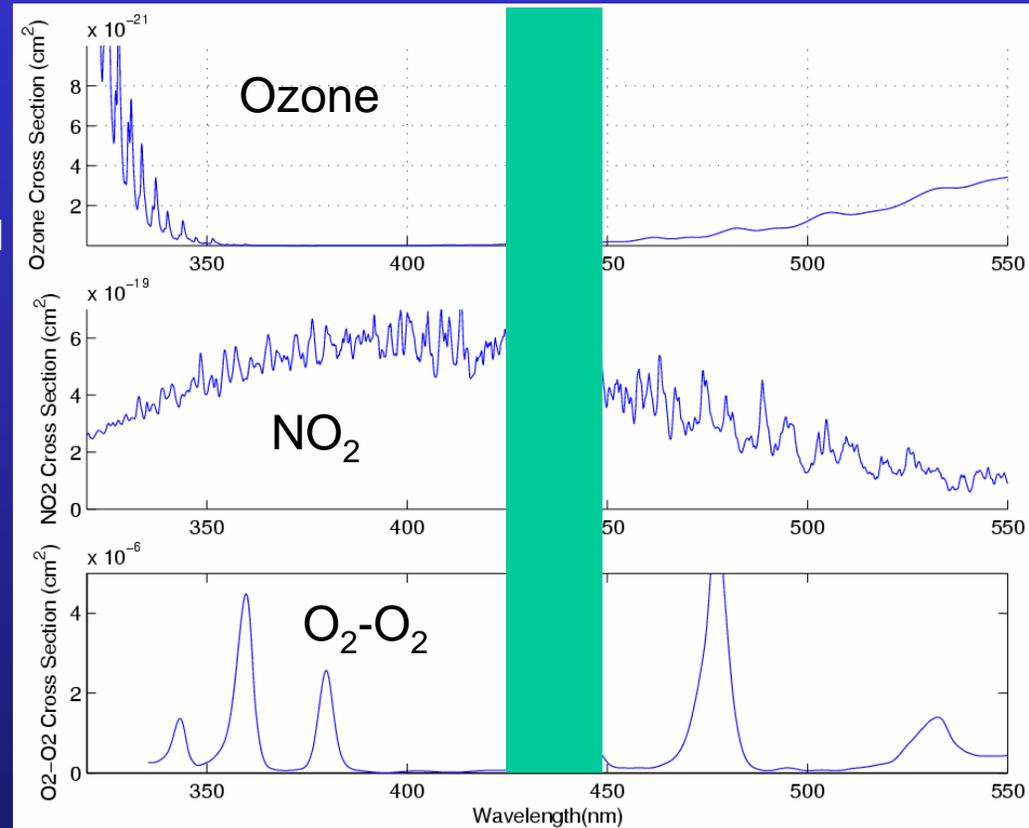
Jack Dibb



Spectral Fit of NO₂



Distinct NO₂ Spectrum



Nonlinear least-squares fitting

$$I_B(\lambda) = A(\lambda)I_0(\lambda)e^{-\sum \tau_s} + Ring$$

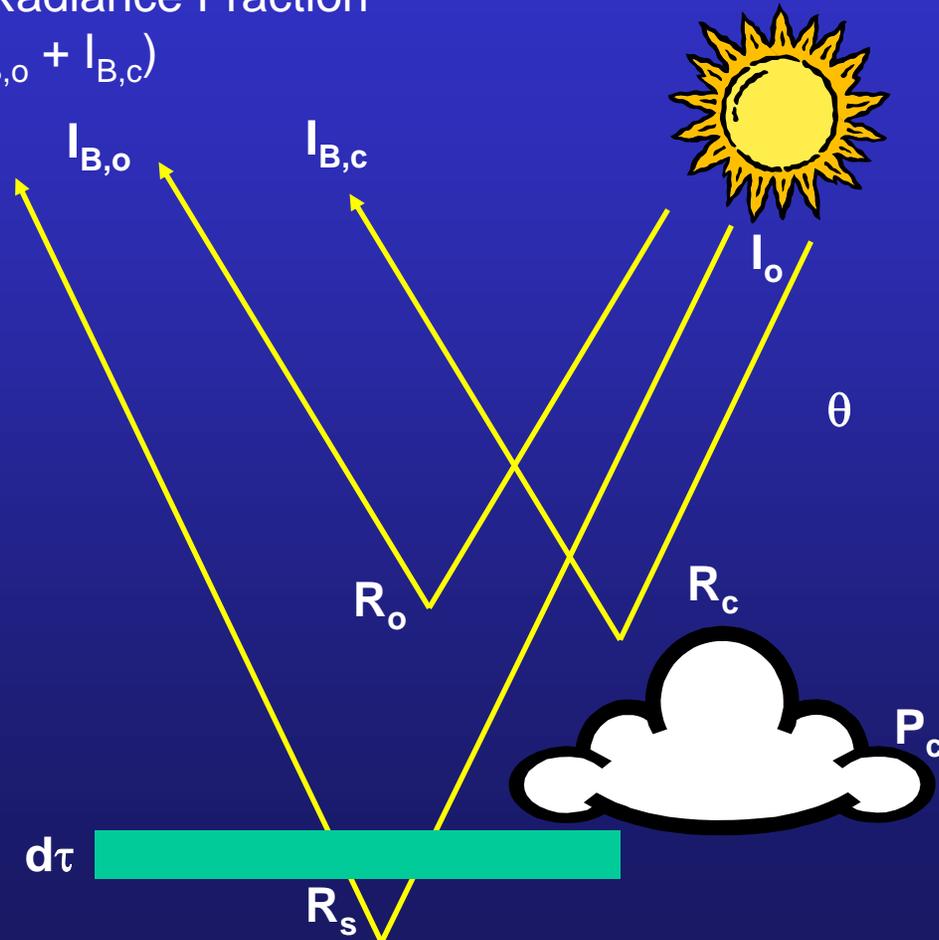
Also Weak H₂O line

Based on Martin et al., 2002

Perform a Radiative Transfer Calculation to Account for Viewing Geometry and Scattering

Cloud Radiance Fraction

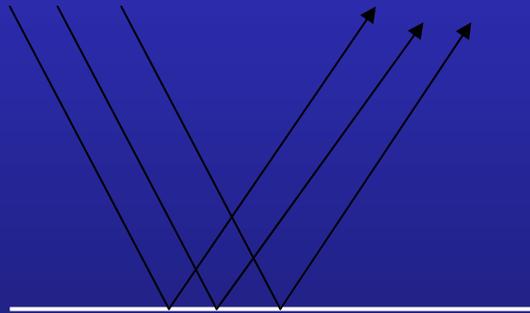
$$I_{B,c} / (I_{B,o} + I_{B,c})$$



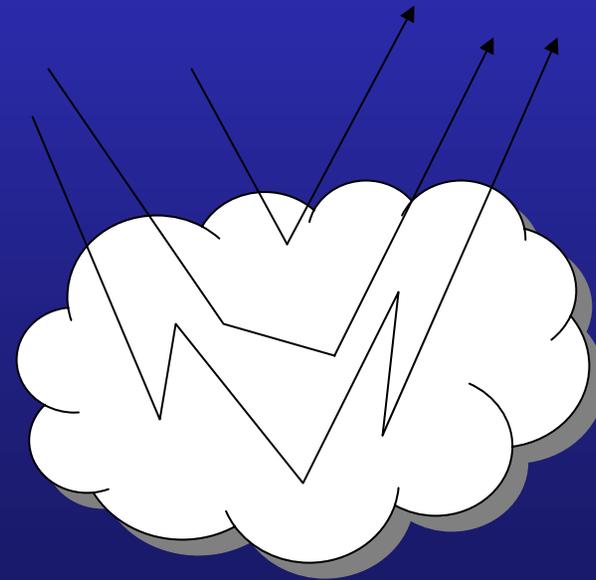
- FRESCO Clouds Fields [Koelemeijer et al., 2002]
- Surface Reflectivity [Koelemeijer et al., 2003]
- LIDORT Radiative Transfer Model [Spurr et al., 2002]
- GEOS-CHEM NO_2 & aerosol profiles

**Data Provided for all Cloud Fractions However
Use of High Cloud Fraction Data Is Discouraged!
Cloud Radiance Fraction < 0.4 Recommended in Header**

FRESCO Cloud Algorithm



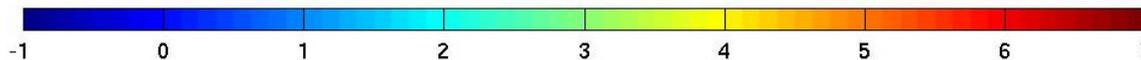
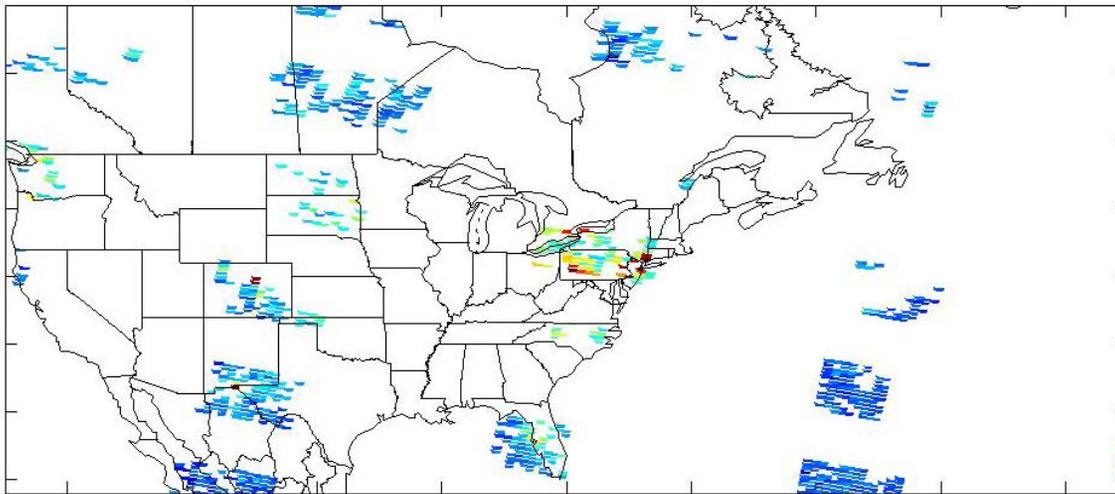
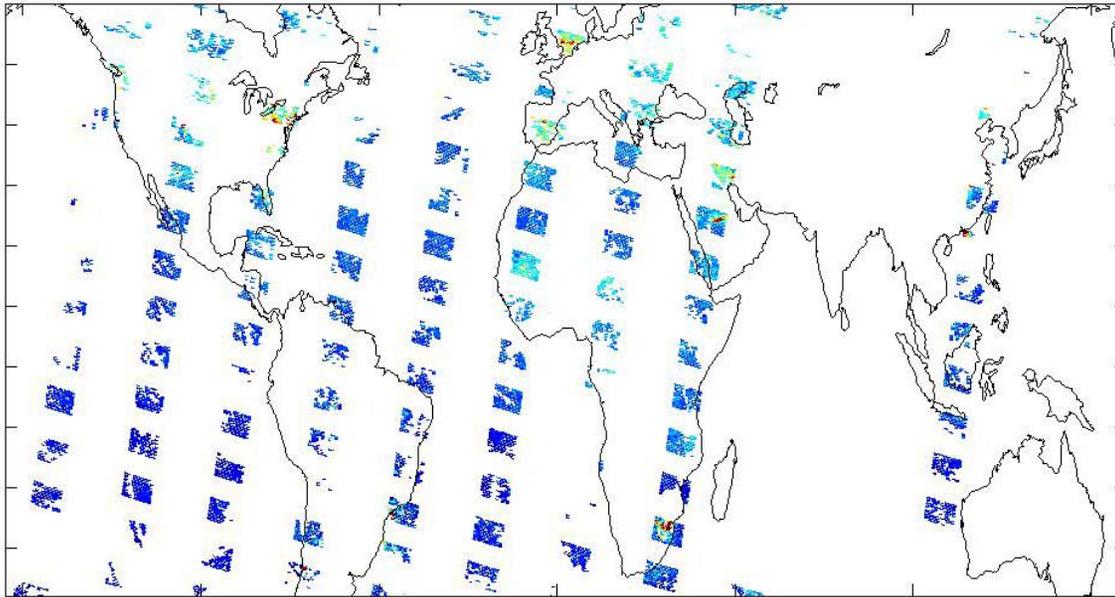
Reality



Surface

Sample JPGs Provided For Each Day

SCIAMACHY cloud-filtered measurements for 20040701



Tropospheric NO₂ (10^{15} molec cm⁻²)

Missing Data:

Cloudy

Missing Cloud Fields

Satellite Downlink Issues

Typical Individual
Measurement Uncertainty

$\pm(1 \times 10^{15} \text{ molec cm}^{-2} + 40\%)$

Spectral Fit

Stratospheric NO₂

Surface Reflectance

Clouds

Aerosols

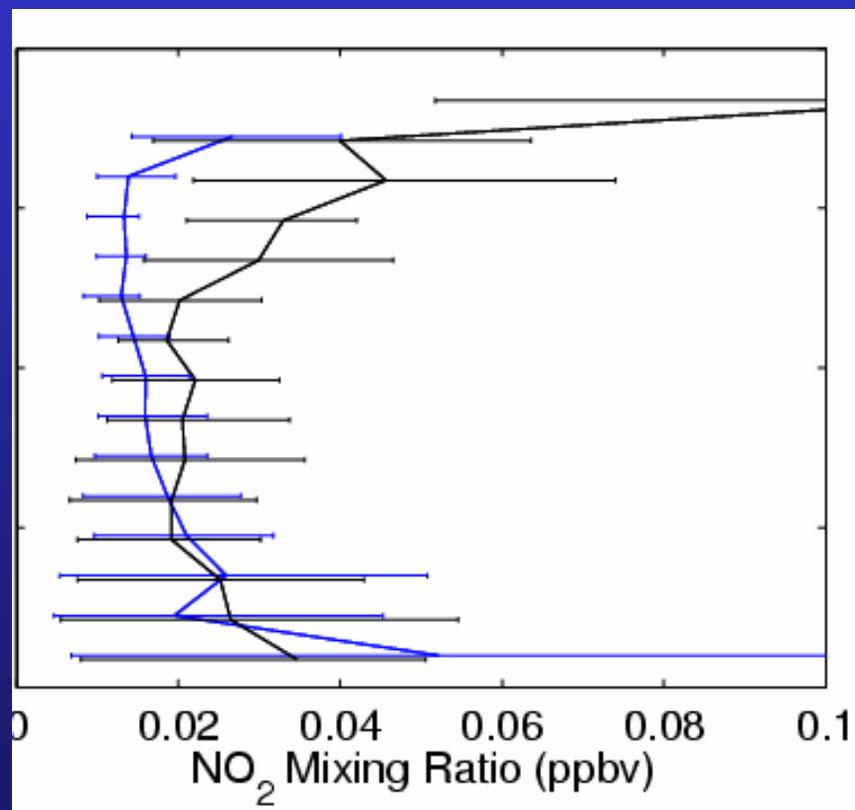
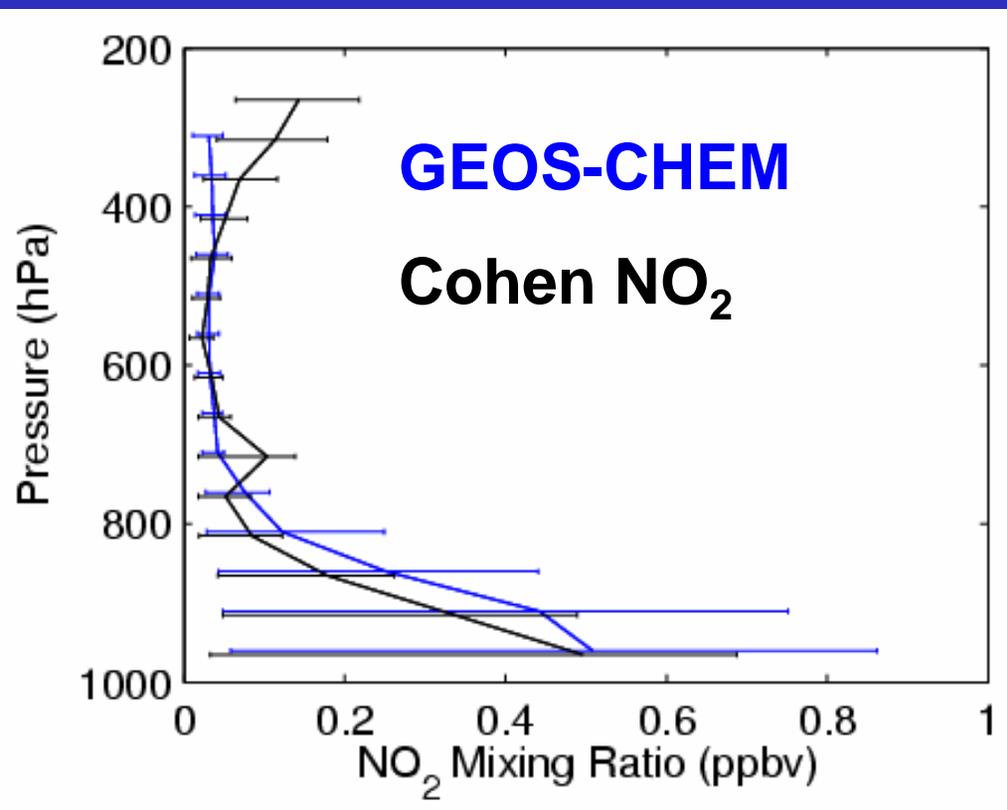
Assumed NO₂ Profile

Preliminary Comparison Between Average Assumed and Measured NO_2 Profiles

Need to Continue Analysis for Individual Flights

West of -60 degrees lon, "land"

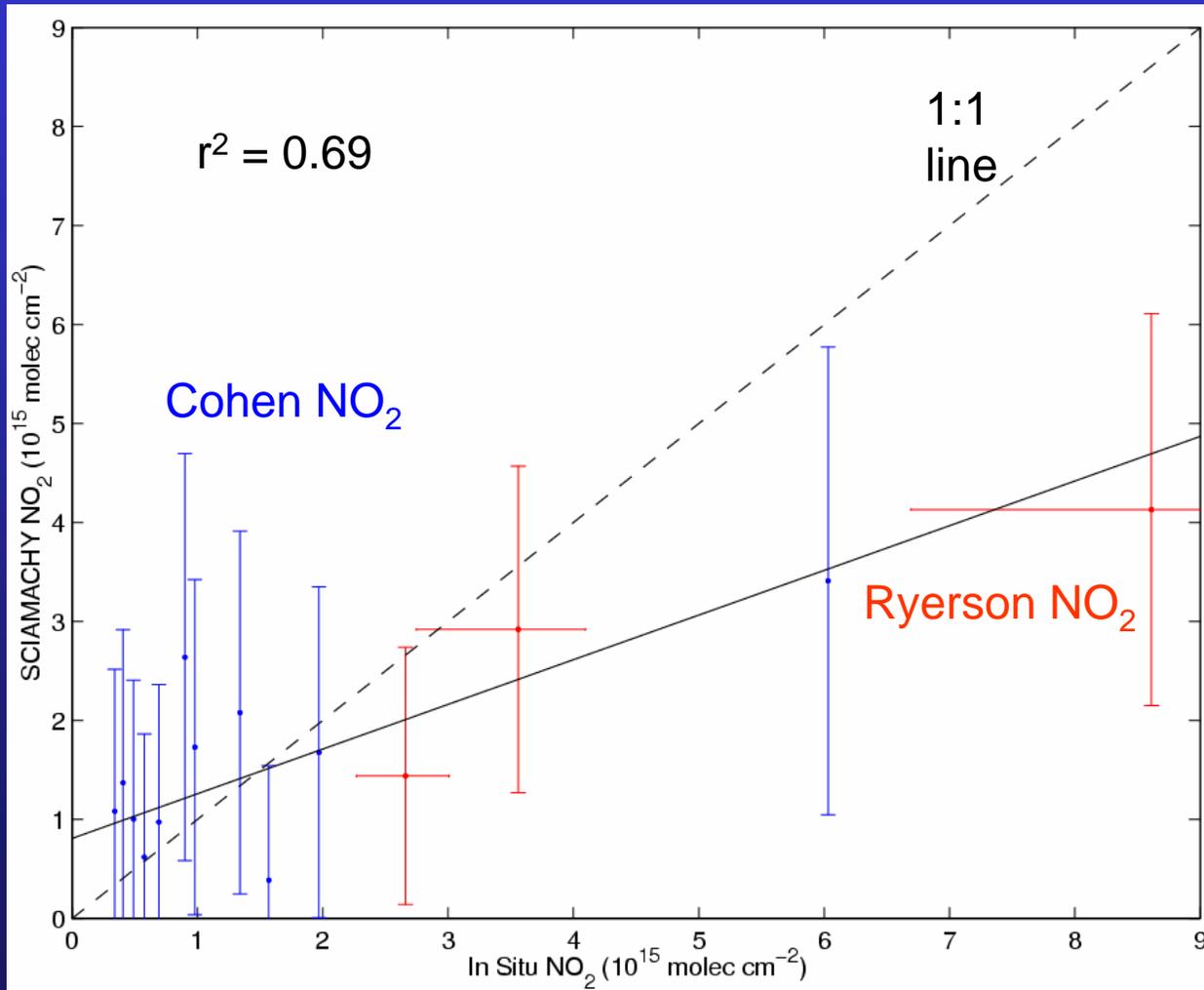
East of -60 degrees lon, "ocean"



Errorbars Show 17th and 83rd percentiles

Reasonable Agreement Between Coincident SCIAMACHY and In-Situ Cloud-Free Measurements

Difficult Comparison over Source Regions Due to Ambiguous Column Below Aircraft and Spatial Heterogeneity

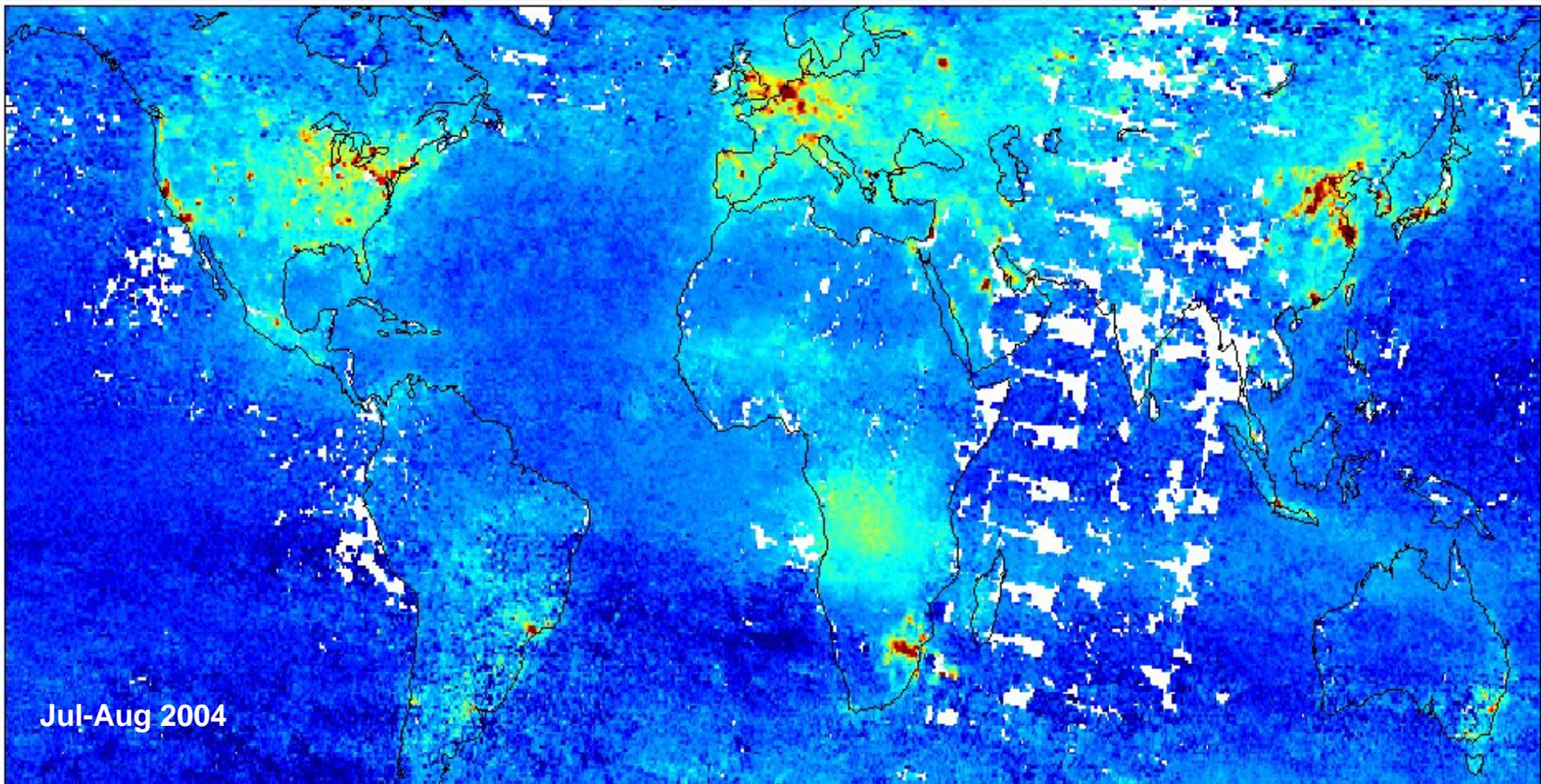


- Coincident measurements
 - Cloud-radiance fraction < 0.4
 - In-situ measurements below 1 km
- Assume constant mixing ratio below lowest measurement

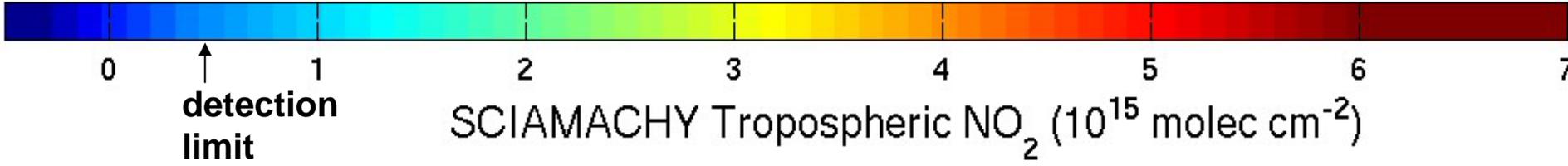
Chris Sioris

In situ errorbars show 17th & 83rd percentiles – not completed for DC8

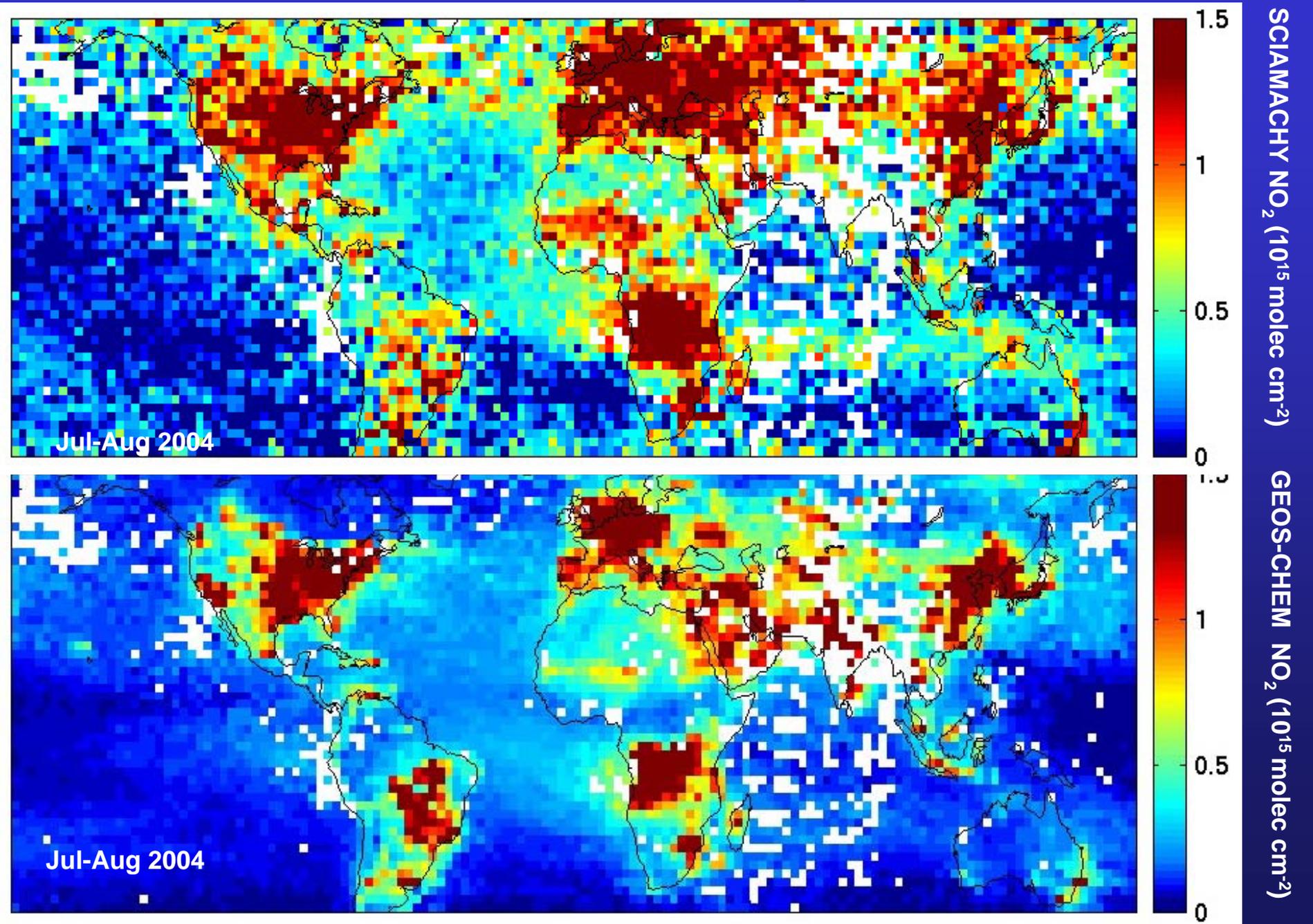
Cloud-filtered Tropospheric NO₂ Columns Observed from the SCIAMACHY Satellite Instrument



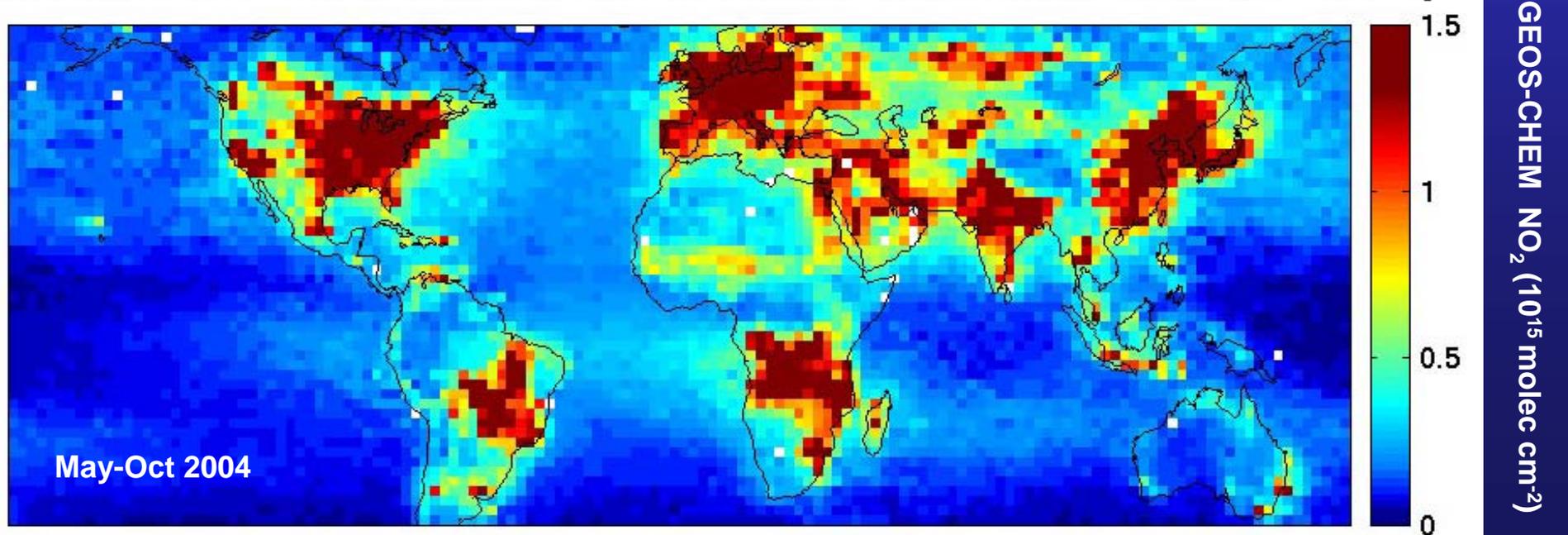
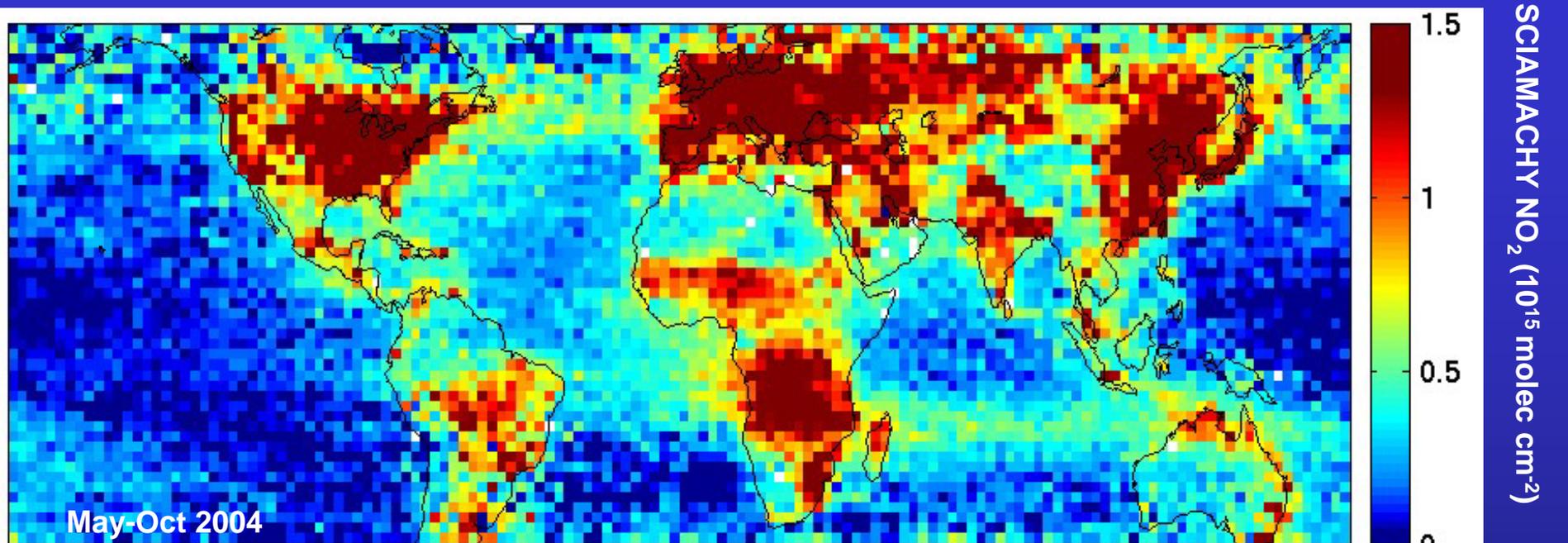
$\pm(5 \times 10^{14} \text{ molec cm}^{-2} + 30\%)$



SCIAMACHY Shows Elevated NO_x Export from North America



SCIAMACHY Shows Elevated NO_x Export from North America

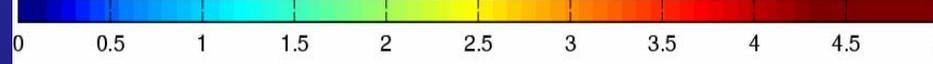
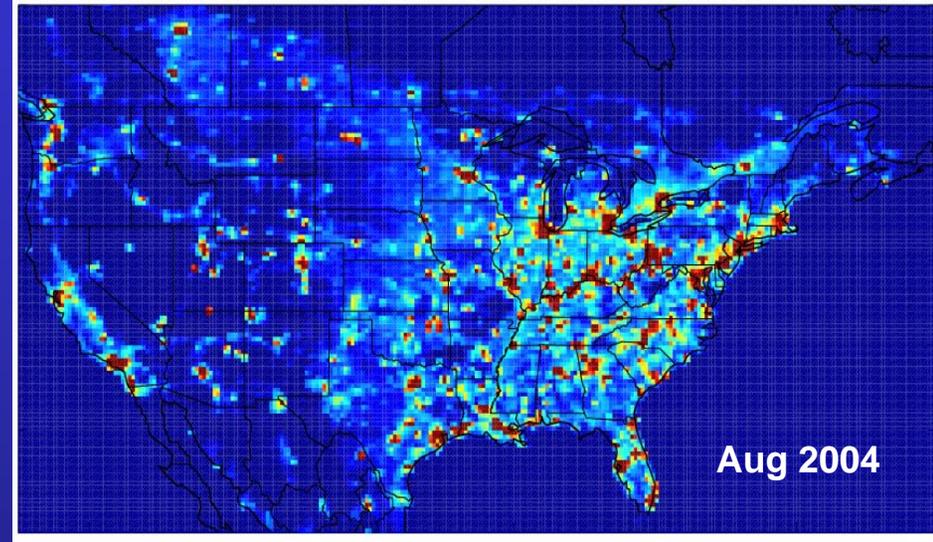
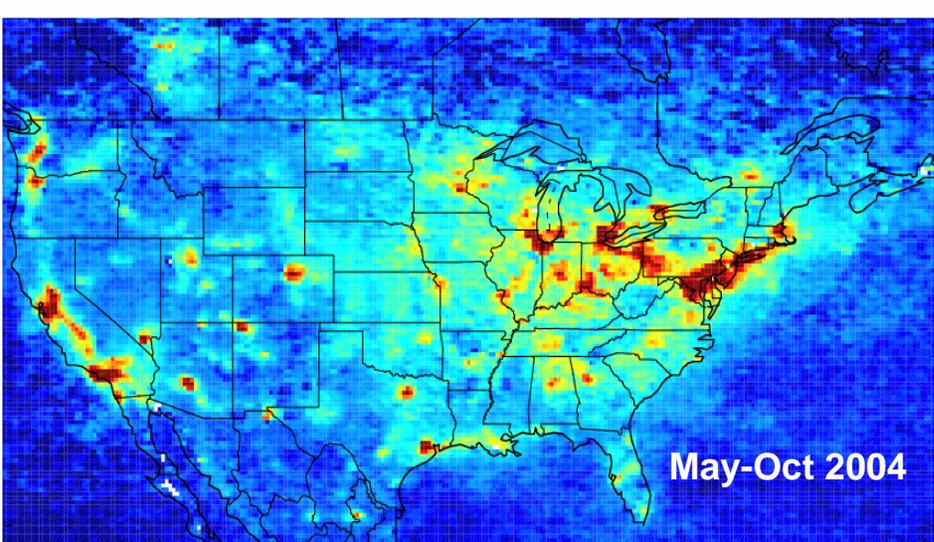


EMIS: Emissions Mapping Integration Science

Optimize North American NO_x Emissions

SCIAMACHY NO₂ Columns

NO_x Emissions (SMOKE/G.Tech)



10^{15} molecules cm⁻²

10^{11} molec N cm⁻² s⁻¹

Models-3 ↓ GEOS-CHEM

Top-Down Emissions

Error weighting

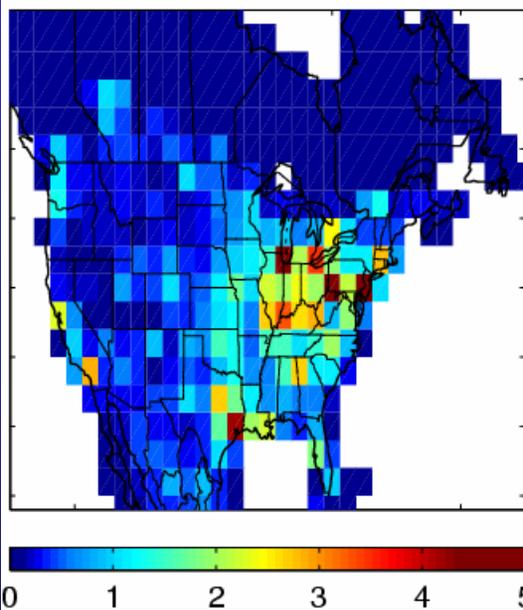
A posteriori emissions

North American NOx Emissions (May – October)

Largest Change in Northeastern US Coast

GEOS-CHEM

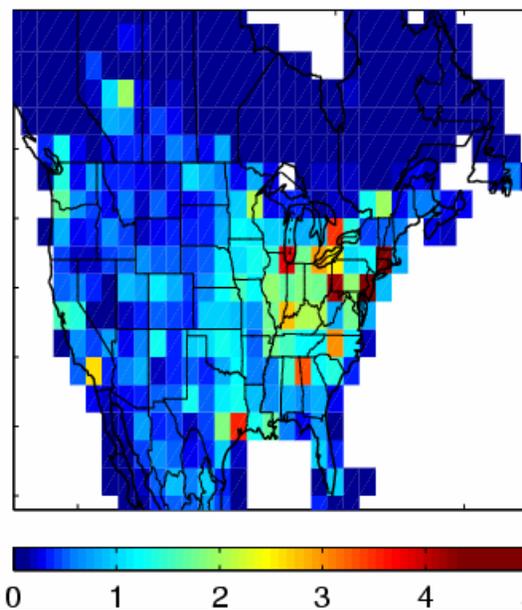
(NAPAP Scaled to 1998)



10^{11} atoms N cm^{-2} s^{-1}

7.6 Tg N yr^{-1} $r^2 = 0.85$

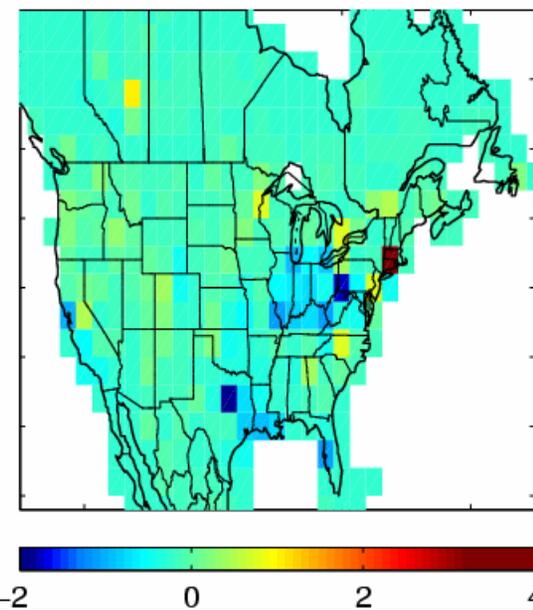
SCIAMACHY (2004)



10^{11} atoms N cm^{-2} s^{-1}

8.4 Tg N yr^{-1}

SCIAMACHY - NAPAP



10^{11} atoms N cm^{-2} s^{-1}

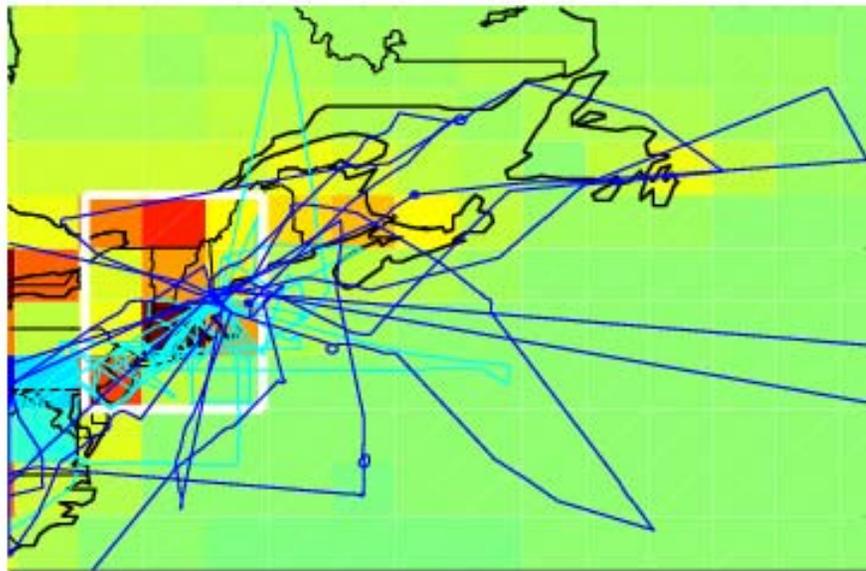
0.8 Tg N yr^{-1}

Evaluate Top-Down and Bottom-Up NO_x Inventories

Conduct GEOS-CHEM Simulation For Each Inventory

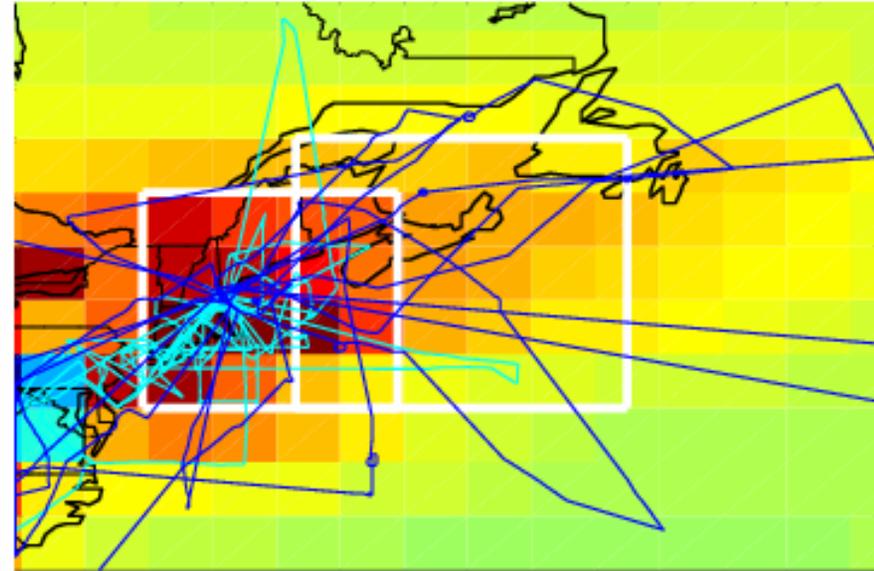
Sampled GEOS-CHEM Along Flight Tracks

Simulation with SCIAMACHY – Original NO_x Emission Inventory



-0.5 0 0.5

ΔNO_x (ppbv)



-0.5 0 0.5

ΔHNO_3 (ppbv)

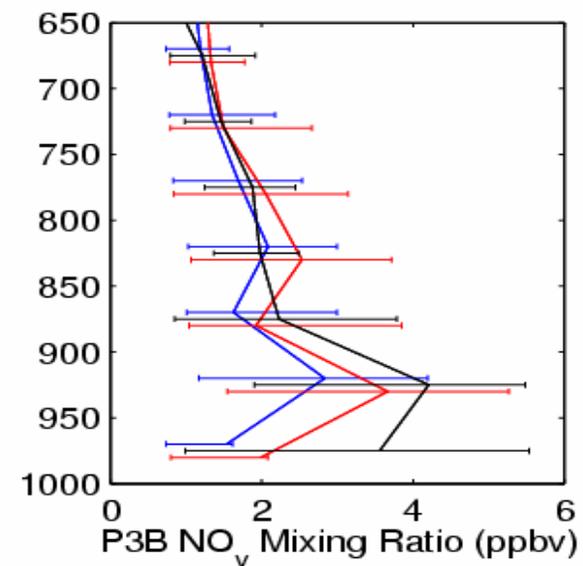
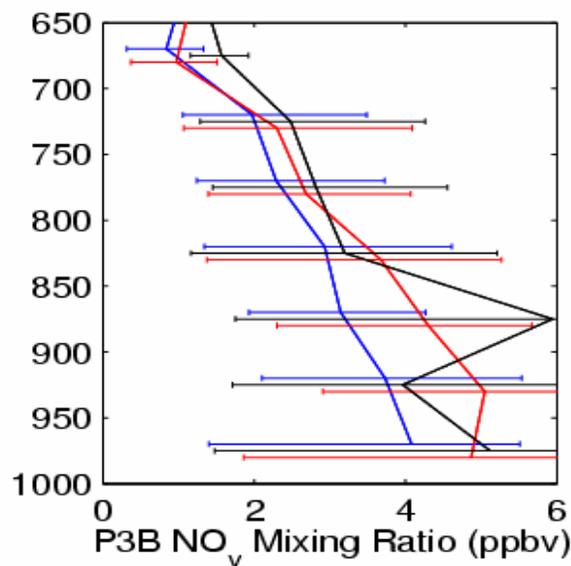
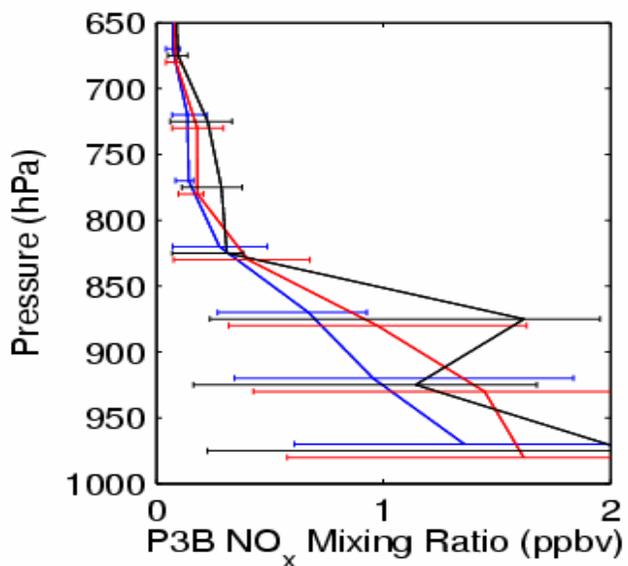
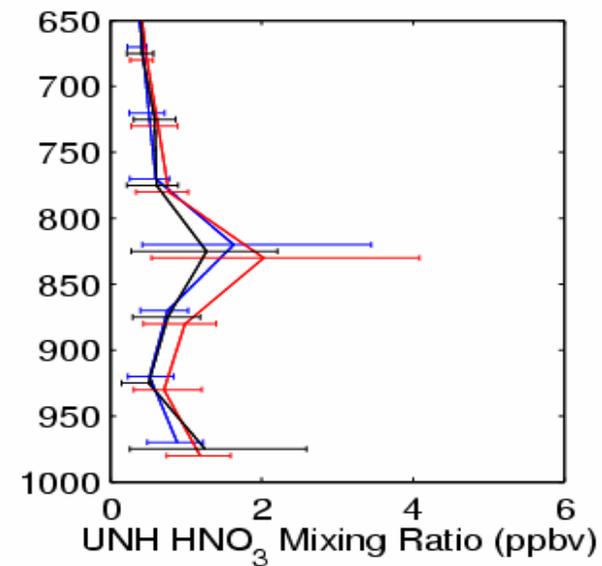
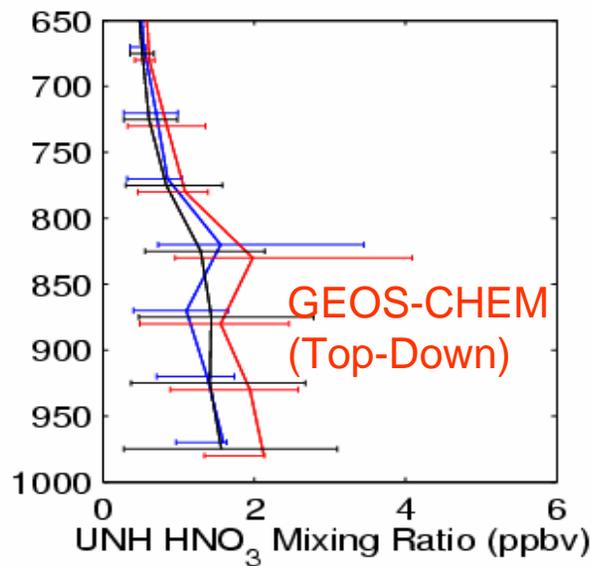
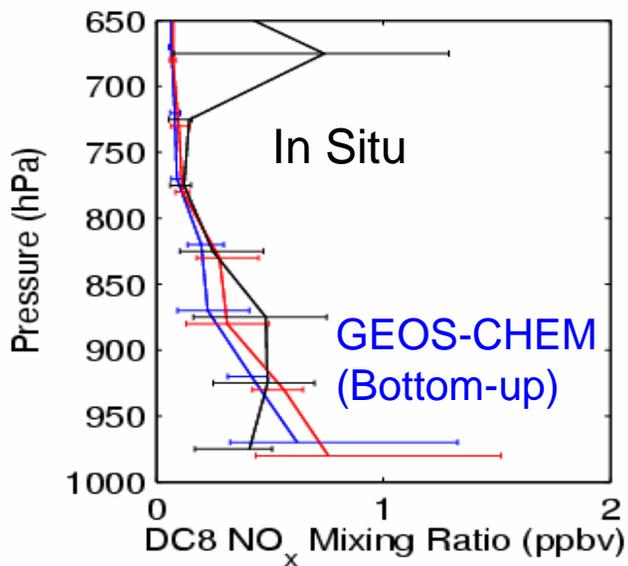
P3-B Measurements Support Top-Down Inventory

DC-8 Measurements Inconclusive

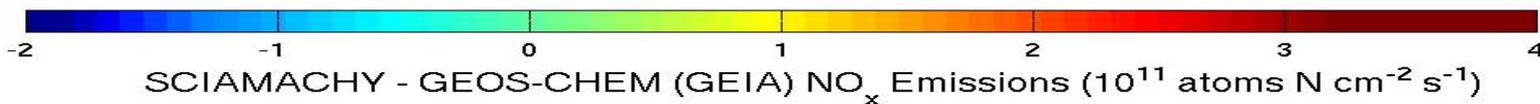
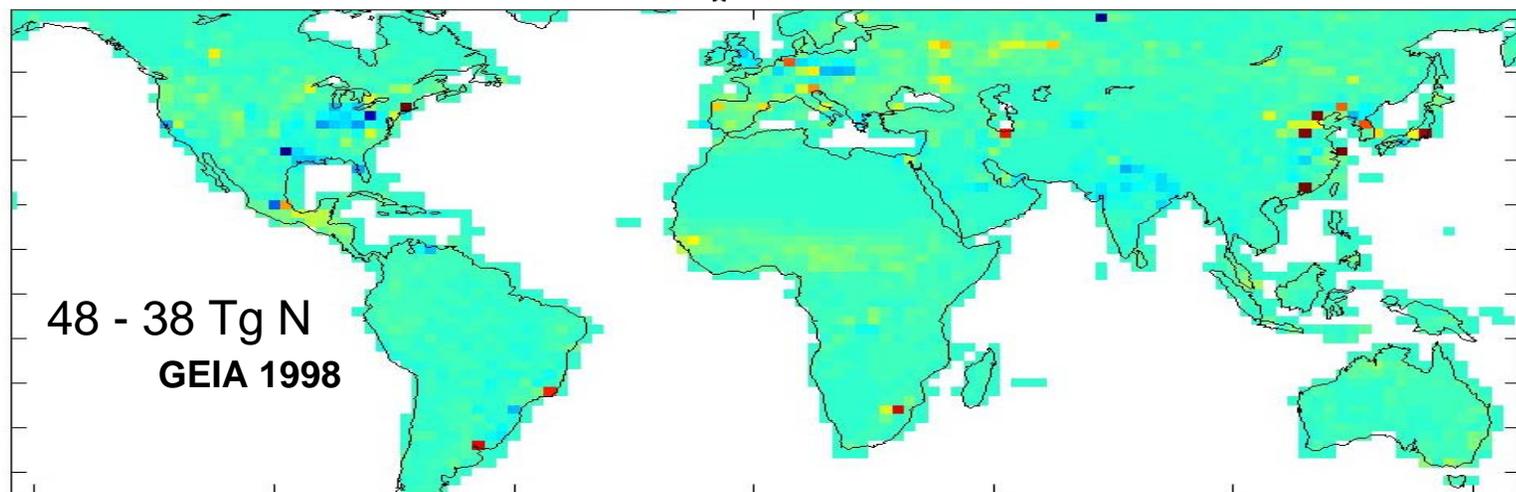
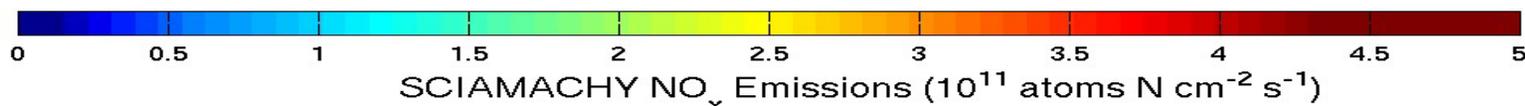
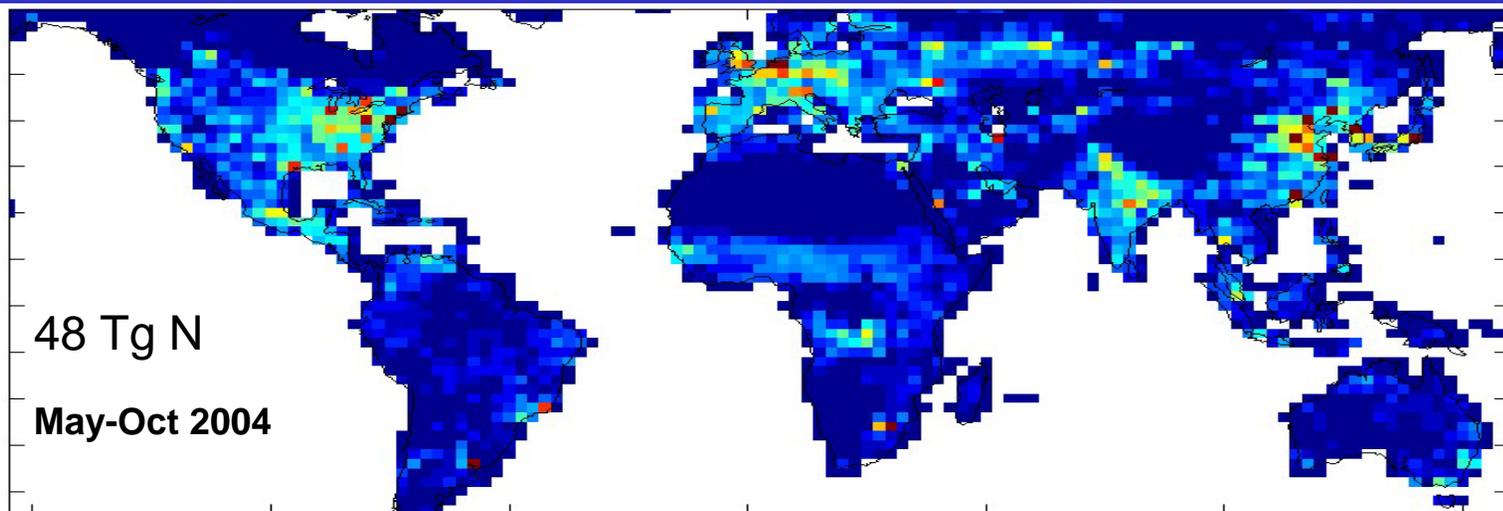
New England

New England + Gulf

Remote



Major Discrepancy in NO_x Emissions from Megacities



INTEX Workshop

Virginia, USA, March 29, 2005

Intercontinental Transport of NO₂ Observations from GOME and SCIAMACHY

A. Heckel, A. Richter, J. P. Burrows

Institute of Environmental Physics and
Institute of Remote Sensing
University of Bremen

North American Export – Climatology

- **Aim:**

- Identification of the typical export pathways in the satellite NO₂ data set
- Quantification of the export amount and range
- Impact on European air quality?

- **Approach:**

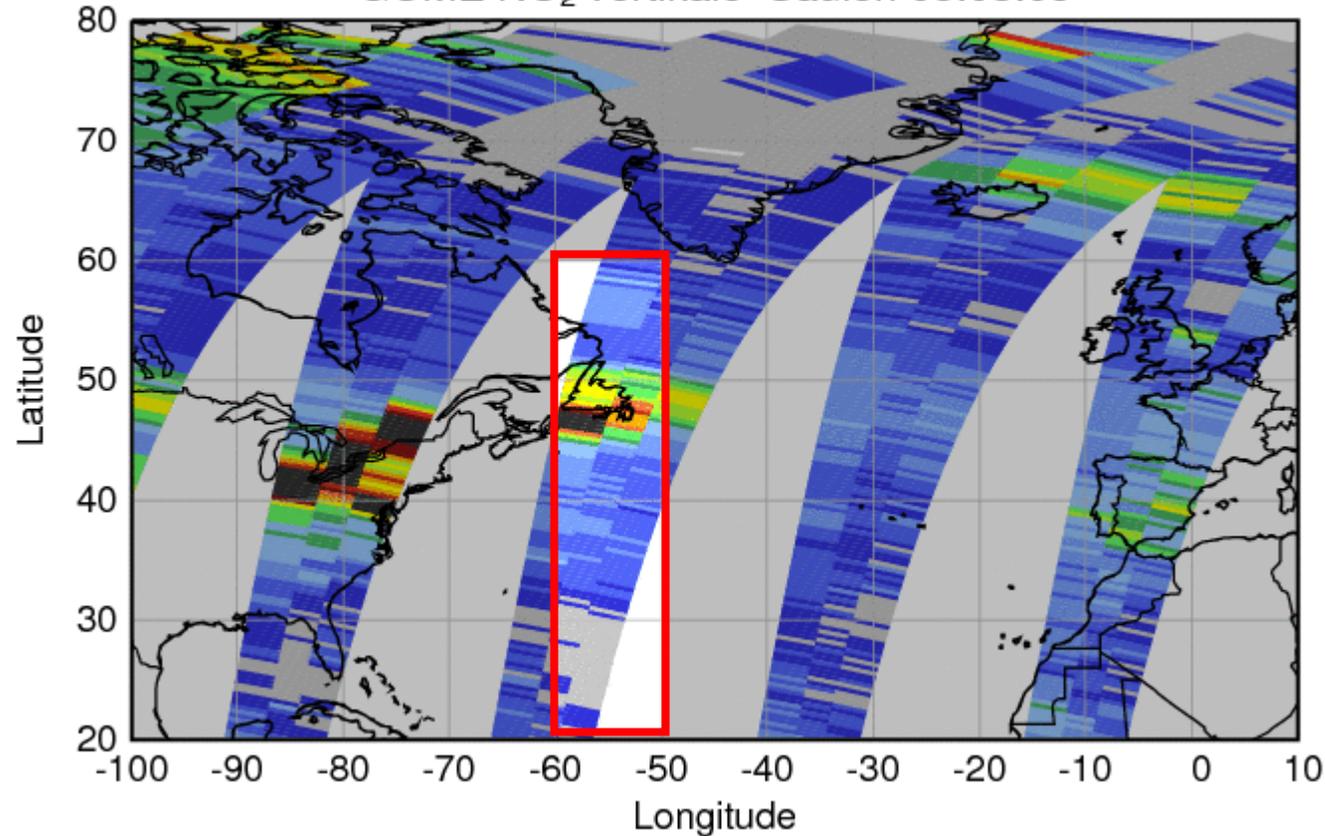
- Using the 10 year GOME and SCIAMACHY time series
- Detection of outflow events by applying thresholds to the NO₂ tropospheric columns over North Atlantic
- Counting all events between 01.Jan 1996 and 01.Jul 2003
- Selection for cloudy / clear scenes
- Backwards trajectory analysis for selected cases

- **Limitations / Problems:**

- Short lifetime of NO₂ => only “fast” events can be observed
- Air Mass Factors do not yet account actual profile shape and clouds

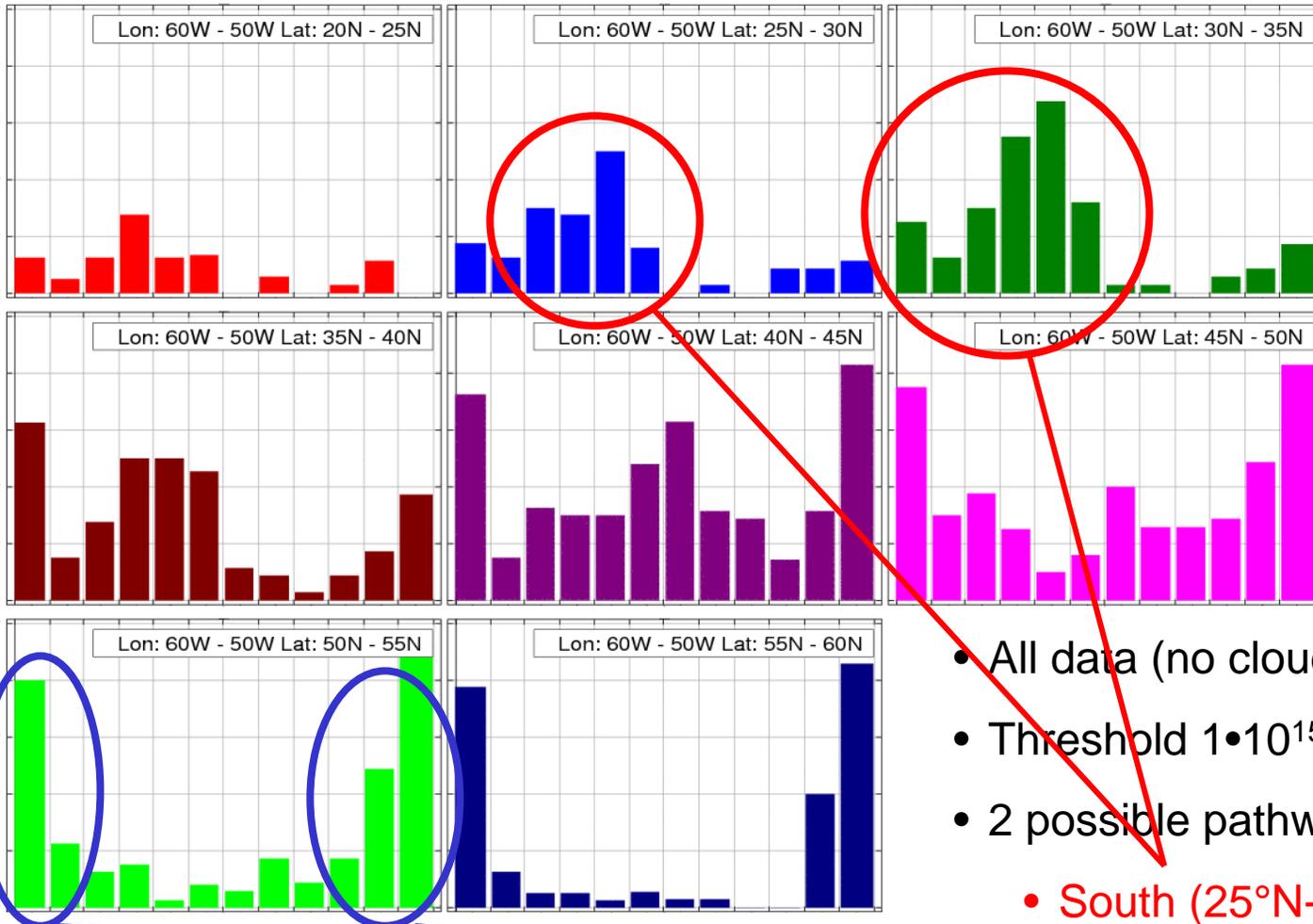
Approach

GOME NO₂ vertikale Säulen 05.03.03



- Longitudinal section between 60°W and 50°W
- Divided into boxes of 5° latitude from 20°N to 60°N
- Columns larger than $1 \cdot 10^{15}$ molec/cm²
- Area of enhanced values larger than $1 \cdot 10^5$ km²

First results - Histograms



- All data (no cloud screening)
- Threshold $1 \cdot 10^{15}$ molec/cm²
- 2 possible pathways:
 - South (25°N-35°N) in spring
 - North (45°N-60°N) in winter

Acknowledgements

- NO₂ in situ data were provided by Ronald Cohen, University of California, Berkeley
- GOES and Meteosat IR imagery were provided by UNIDATA and Space Science and Engineering Center (SSEC), University of Wisconsin-Madison
- Owen Cooper from CIRES institute at University Colorado, Boulder
- Trajectories computed by NOAA HYSPLIT Web Interface
- SCIAMACHY lv0 and lv1 data were provided by ESA through DLR/DFD
- Financial support by NOAA, NASA, and the University of Bremen

