Measuring Tropospheric NO₂ from SCIAMACHY During INTEX and Improving NOx Emission Inventories

Randall Martin
Aaron Van Donkelaar

Chris Sioris
Kelly Chance

Yongtao Hu
Armistead Russell

Tom Ryerson

Ron Cohen

Bill Brune

UC Berkeley

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
\( \frac{I_{B,c}}{I_{B,o} + I_{B,c}} \)

- FRESCO Clouds Fields [Koelemeijer et al., 2002]
- Surface Reflectivity [Koelemeijer et al., 2003]
- LIDORT Radiative Transfer Model [Spurr et al., 2002]
- GEOS-CHEM NO₂ & aerosol profiles

Based on Martin et al., 2002, 2003
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

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 NO\(_2\)

Surface Reflectance
- Clouds
- Aerosols
- Assumed NO\(_2\) 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 17$^{th}$ and 83$^{rd}$ 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

$r^2 = 0.69$

1:1 line

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

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

Chris Sioris
Cloud-filtered Tropospheric NO$_2$ Columns Observed from the SCIAMACHY Satellite Instrument

±(5x10$^{14}$ molec cm$^{-2}$ + 30%)

SCIAMACHY Tropospheric NO$_2$ (10$^{15}$ molec cm$^{-2}$)
SCIAMACHY Shows Elevated NOx Export from North America

Jul-Aug 2004

SCIAMACHY NO\textsubscript{2} (10\textsuperscript{15} molec cm\textsuperscript{-2})

GEOS-CHEM NO\textsubscript{2} (10\textsuperscript{15} molec cm\textsuperscript{-2})
SCIAMACHY Shows Elevated NOx Export from North America

May-Oct 2004
EMIS: Emissions Mapping Integration Science
Optimize North American NO\textsubscript{x} Emissions

SCIAMACHY NO\textsubscript{2} Columns

NO\textsubscript{x} Emissions (SMOKE/G.Tech)

10\textsuperscript{15} molecules cm\textsuperscript{-2}

Models-3 \rightarrow GEOS-CHEM

Top-Down Emissions

10\textsuperscript{11} molec N cm\textsuperscript{-2} s\textsuperscript{-1}

Error weighting

A posteriori emissions
North American NOx Emissions (May – October)

Largest Change in Northeastern US Coast

GEOS-CHEM (NAPAP Scaled to 1998)

SCIAMACHY (2004)

SCIAMACHY - NAPAP

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

$7.6$ Tg N yr$^{-1}$

$r^2 = 0.85$

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

$8.4$ Tg N yr$^{-1}$

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

$0.8$ Tg N yr$^{-1}$
Evaluate Top-Down and Bottom-Up NOx Inventories

Conduct GEOS-CHEM Simulation For Each Inventory

Sampled GEOS-CHEM Along Flight Tracks

Simulation with SCIAMACHY – Original NOx Emission Inventory

$\Delta \text{NO}_x$ (ppbv)  

$\Delta \text{HNO}_3$ (ppbv)
P3-B Measurements Support Top-Down Inventory

DC-8 Measurements Inconclusive

New England

New England + Gulf

Remote

In Situ

GEOS-CHEM (Bottom-up)

GEOS-CHEM (Top-Down)
Major Discrepancy in NOx Emissions from Megacities

48 Tg N
May-Oct 2004

48 - 38 Tg N
GEIA 1998
INTEX Workshop
Virginia, USA, March 29, 2005

Intercontinental Transport of NO$_2$
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
  – Separation of transport and lightning NO₂?
Approach

- 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/cm$^2$
- Area of enhanced values larger than $1 \cdot 10^5$ km$^2$
First results - Histograms

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