Synergetic use of MATCH-MPIC & GOME for the study of Tropospheric NO\textsubscript{x} over Asia

T. Kunhikrishnan*
Department of Atmospheric Chemistry/NWG
Max Planck Institute, Mainz.
Germany.
(kunhi@mpch-mainz.mpg.de)

(Permanent affiliation: India Meteorological Department)
Pune, 411005, INDIA

Objectives

1. To understand Tropospheric NO\textsubscript{x} over south Asia, especially India and the Indian Ocean, by using MATCH-MPIC* and GOME-satellite observations.


2. To analyse the uncertainties in the estimation of regional NO\textsubscript{x} emission strength from GOME.
Why Asia?

- Tropical regions-High insolation & Humidity-modify the oxidising efficiency
- Key role in Global Atmospheric Chemistry and Climate
- Data sparse region- Little knowledge
- Rapidly growing Anthropogenic Emissions
- Increasing trend in trace gases/aerosols such as NO\(_x\), CH\(_4\) and Hydrocarbons.

Why NO\(_x\) ?

NO\(_x\) = NO + NO\(_2\)

- Sources & distribution, losses and other properties of NO\(_x\) are poorly understood over Asia
- Increasing trends of NO\(_x\) and acid deposition
- Its importance as O\(_3\) & OH precursor
- Importance in radiative budget (IPCC-2001)
- It is toxic for humans and crops
Asian NOx Emissions based on van Aardenne et al. (1999)

**IPCC-REPORT**

**ASIEN**

<table>
<thead>
<tr>
<th></th>
<th>2000</th>
<th>2020</th>
</tr>
</thead>
<tbody>
<tr>
<td>NOx</td>
<td>10</td>
<td>20</td>
</tr>
<tr>
<td>CO</td>
<td>5</td>
<td>7</td>
</tr>
<tr>
<td>NMKW</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>SO2</td>
<td>2</td>
<td>3</td>
</tr>
<tr>
<td>CO2</td>
<td>1</td>
<td>2</td>
</tr>
</tbody>
</table>

**OECD**

<table>
<thead>
<tr>
<th></th>
<th>2000</th>
<th>2020</th>
</tr>
</thead>
<tbody>
<tr>
<td>NOx</td>
<td>15</td>
<td>25</td>
</tr>
<tr>
<td>CO</td>
<td>10</td>
<td>12</td>
</tr>
<tr>
<td>NMKW</td>
<td>5</td>
<td>7</td>
</tr>
<tr>
<td>SO2</td>
<td>3</td>
<td>4</td>
</tr>
<tr>
<td>CO2</td>
<td>2</td>
<td>3</td>
</tr>
</tbody>
</table>

Veränderung in % zwischen 2000 und 2020

**The tools**

GOME Observations & MATCH Model simulations
MATCH MPIC - Global Chemical Transport Model

2 components: Meteorological & Chemical

Hori. Resolution: T63 (1.875°) & T21 (5.625°)

Vertical resolution: 28 levels, surface-2.7hPa
in sigma coordinates

Model time step: 30 minutes.

MATCH Dynamics

(Model of Atmospheric Transport and Chemistry)

(Lawrence et al. (1999,1996), Rasch et al.(1997),

- Offline, Driving Meteorology from NCEP
- Advection: SPITFIRE
- Convection: Zhang/McFarlane/Hack
- Vertical Diffusion: Holtslag and Boville
- Clouds:
  - FRACTION: Slingo
  - Microphysics: Rasch and Kristjansson
- Full Tropospheric Hydrological Cycle.
MATCH-MPIC Chemistry
(von Kuhlmann, Lawrence, Crutzen)

- Chemical Species/Reactions:
  - CH4-CO-NOx-HOx-Ox
  - Isoprene, Ethane, Propane(Acetone),
  - Ethene, Propene, higher Alkanes
  - 56 Species with 140 Reactions
  - MIM-Mainz Isoprene Mechanism
    (Poeschl et al.)
- Online Photolysis Rates
  (Landgraf/Crutzen)
- Flexible Integration Scheme (KPP)

MATCH-MPIC Chemistry
( von Kuhlmann, Lawrence, Crutzen)

- EMISSIONS:
  - Industrial (except Ships): EDGAR
  - Ships: Corbett et al.
  - Biomass Burning: Galanter et al.
  - Biogenic (land): Guenther et al.
  - Oceanic: Bates et al.
  - Lightning NOx: Price and Rind
- Dry Deposition:
  - Resistance Model (Ganzeveld et al.)
- Wet deposition and cloud settling:
  - Based on Solubility and Model Precipitation
- Solubility Couples wet dep, dry dep,
  cloud settling and convective transport
**GOME**

Spatial resolution: 40 km lat. × 320 km lon.
Wavelength region: 240-790 nm
Spectral resolution: 0.2-0.4 nm
The data: Pixels with a cloud cover < 0.1

Column densities of NO\textsubscript{2} absorption: DOAS method
Air mass factors for the NO\textsubscript{2} column: GOMETRAN

Assumptions: Clear sky, a maritime aerosol, a surface albedo 0.05, Constant mixing ratio of NO\textsubscript{2} < 1.5 km

(Burrows J P et al. (1999), Richter et al. (2002))

Overall uncertainty in Tropospheric NO\textsubscript{2} col. ~ 50%
Less in clear situations (Heland et al., 2002).

**MATCH versus GOME**
1. GOME Retrieval Assumptions

Basic assumptions are
(Richter and Burrows (2002), Burrows et al., 1999)

- Zonal symmetry of stratospheric NO$_2$ column
- Tropospheric NO$_2$ col. is negligible within the oceanic reference sector 180-170W

Trop. NO$_2$ col. = Total NO$_2$ col. NO$_2$ col. for the ref. sector
**GOME Retrieval Assumptions from MATCH !!**

- **Ratio of tropospheric to Stratospheric NO\(_2\) column**
  - Ref. sector- very low (~zero)
  - India: 0.28-0.46
  - Indonesia: 0.19-0.23
  - China: 0.22-0.69
  - North Asia: 0.17-0.46.

- **Mean deviation from Zonal symmetry**
  - India: 12.1 %
  - Indonesia: 8.9 %
  - China: 3.7 %
  - North Asia: 8.2 %

---

**2. SAMPLING ISSUES**

1. **Sampling Time Correction: (STC)**
   - Ratio: MATCH 24-hour average to 10:30 LT (GOME-time)
   - (MATCH modified to write the output at 10:30 LT)

2. Could add cloud screening to the model output, similar to the GOME cloud screening.
   - (Impact of diurnal variation of cloud on NO\(_2\) column)
RATIO – 10:30 to 24 hour Average NO$_2$ col. From MATCH, JUL 97

Surface Level

Tropo. NO$_2$ col. surf-150hPa

RATIO OF TROPOSPHERIC NO$_2$ COL. AT 10:30 LT TO 24 H

INDIA

CHINA

INDONESIA

NORTH ASIA
### INTERANNUAL VARIABILITY OF NO$_2$ COLUMN FROM GOME & MATCH

“The primary objective of the GOME mission is to provide reliable and most frequent space observations of trace gases to estimate the long term changes in the troposphere......”

<table>
<thead>
<tr>
<th>Region</th>
<th>GOME NO$_2$ Abundance (10$^{15}$ molec/cm$^2$)</th>
<th>MATCH NO$_2$ Abundance (10$^{15}$ molec/cm$^2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Mean</td>
<td>Maximum</td>
</tr>
<tr>
<td>India (5.35°N, 60-95°E)</td>
<td>5.68</td>
<td>9.6</td>
</tr>
<tr>
<td>China (20-70°N, 90-150°E)</td>
<td>9.05</td>
<td>20.8</td>
</tr>
<tr>
<td>Indonesia (10°S-20°N, 95-140°E)</td>
<td>3.49</td>
<td>7.4</td>
</tr>
<tr>
<td>North Asia (30-70°N, 60-90°E)</td>
<td>8.61</td>
<td>23.7</td>
</tr>
</tbody>
</table>
NO2 LIFETIME FROM MATCH OVER INDIAN OCEAN
NO\textsubscript{x} Relative changes (%)

SOUTH INDIAN OCEAN (55°E-95°E, 5°N-25°S)

O\textsubscript{3} Relative Changes (%)

SOUTH INDIAN OCEAN (55°E-95°E, 5°N-25°S)
Possible Uncertainties in the estimation of Regional NO\textsubscript{x} emission strength from GOME!!
Sensitivity of Tropospheric NO\textsubscript{x} over India and Indian Ocean

- **Sensitivity runs versus Base run of MATCH**

(i) Setting emission to 90% (a reduction of 10%) of its base source over India and unchanged for the rest of the world.

(ii) As (i) except including normal lightning NO\textsubscript{x} emission from India.

(iii) As (i) for Indonesia, China, Africa and Middle East and see the relative changes over India/Indian Ocean.

Scaled Sensitivity of NO\textsubscript{x} to Local Source-India

Lightning NO\textsubscript{x} (%) over India
Sensitivity over India to Tropospheric NO$_x$ Sources (%)

Lower Troposphere
(Surface-500 hPa)

Upper Troposphere
(500-150 hPa)
LIFETIME OF NO₂ from GOME & MATCH

GOME- Chemical Decay method (850 hPa).

MATCH-Mass-emission method (Asia) & Main NO₂ loss against HNO₃ (Arabian sea, MABL)

Exponential Decay Curve Method -Lifetime of NO₂ from GOME

Chemical decay of NO₂ over Ocean where there is no emission.
\[ \frac{\partial C}{\partial t} = L \times C \]
i.e. \[ C = C₀ \times e^{t/L} \]
where \( \tau = \frac{1}{L} \)
\( L \) = constant loss rate, \( C \) = concentration after a time \( t \)

The study site:
Over the Arabian sea along 3 trajectories (31°, 45° and 49°) from Mumbai (72.75° E, 19.25° N) - West coast of India.

Period:
During January (1997) when the winds are steadily offshore.

Initial NO₂ maxima from GOME: \( 2.2 \times 10^{15} \) molecules/cm²

Source of Wind speed: NCEP monthly mean
Regional NO\textsubscript{x} Emission Strength for INDIA

<table>
<thead>
<tr>
<th>Source</th>
<th>Year</th>
<th>Mean NO\textsubscript{x} (TgN/yr)</th>
<th>References/Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>MATCH</td>
<td>1997 (met)</td>
<td>Mean: 1.72</td>
<td>LT: 15-20 hrs</td>
</tr>
<tr>
<td></td>
<td>1990-emission</td>
<td>Max: 2.3 (April)</td>
<td>Area: MATCH grids-Indian domain</td>
</tr>
<tr>
<td></td>
<td>EDGAR</td>
<td></td>
<td>(present study)</td>
</tr>
<tr>
<td>GOME</td>
<td>1997</td>
<td>1.87 (an improved one from GOME)</td>
<td>LT: 27 hrs. Area: Extended</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Indian region including</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>neighbouring nations.</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Wenig (2002)</td>
</tr>
<tr>
<td>GOME</td>
<td>1997</td>
<td>2.95</td>
<td>LT: 27 hrs. Area: Extended</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Indian region including</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>neighbouring nations.</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Leue et al.(2001)</td>
</tr>
<tr>
<td>RAIN-ASIA</td>
<td>1990</td>
<td>1.52</td>
<td>Aardenne et al.(1999), based</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>on anthropogenic sources.</td>
</tr>
<tr>
<td>ENERGY STATISTICS-INDIA</td>
<td>1995</td>
<td>3.46</td>
<td>Garg et al.(2001)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Based on sector analysis-India</td>
</tr>
</tbody>
</table>
Regional NO\textsubscript{x} emission strength from GOME? How can we improve the method with Model informations? (Findings from the case study for India)

- A significant fraction of Tropospheric NO\textsubscript{x} is from remote sources which introduces a non-negligible uncertainty.

- Regionally appropriate lifetime of NO\textsubscript{x} is not straightforward from GOME and can be calculated from the model.

Impact of NO\textsubscript{x} emission on other Trace gases over India from MATCH

The findings......

“moderate increase or decrease of NO\textsubscript{x} over India are not expect to lead to large changes in the regional O\textsubscript{3} levels”
Impact of NO$_x$ perturbation on Trace gases over India

% increase of Trace gases with respect to 10% increase of NO$_x$ source over India.

<table>
<thead>
<tr>
<th>Lower Troposphere (surf-500 hPa)</th>
<th>Upper Troposphere (500-150 hPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(i) NO$_x$ 6-7 %</td>
<td>Variations are more seasonal with respect to the seasonal variations of NO$_x$</td>
</tr>
<tr>
<td>(ii) O$_3$ 1-2.5 %</td>
<td>Maximum</td>
</tr>
<tr>
<td>(iii) OH 3-5 %</td>
<td>(i) NO$_x$ ~ 6-7 %</td>
</tr>
<tr>
<td>(iv) PAN 5-6 %</td>
<td>(ii) O$_3$ ~ 1-2 %</td>
</tr>
<tr>
<td>(v) HNO$_3$ 5-10 %</td>
<td>(iii) OH ~ 5-6 %</td>
</tr>
<tr>
<td></td>
<td>(iv) PAN ~ 3 %</td>
</tr>
<tr>
<td></td>
<td>(v) HNO$_3$ ~ 4-7%</td>
</tr>
</tbody>
</table>

Impact of 10% perturbation of NOx source over India

![Lower Troposphere (Surface-500 hPa) graph]

![Upper Troposphere (500-150 hPa) graph]
Thanks are due to

* Dr. Mark G Lawrence  Prof. John P. Burrows
* Dr. Rolf von Kuhlmann  Dr. Andreas Richter
  Dr. Annette Ladstätter-Weißenmayer
  Dr. Mark Weber

Institute of Environmental Physics &
Remote Sensing, University of Bremen

* Max Planck Institute, Mainz

References