ESA Summer School on Data Assimilation, 18- 29th August 2003, Frascati, Italy

Synergetic use of MATCH-MPIC & GOME for the study of Tropospheric NO_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_x over south Asia, especially India and the Indian Ocean, by using MATCH-MPIC* and GOME-satellite observations.

*MATCH-MPIC: Model of Atmospheric Transport and Chemistry- Max Planck Institute for Chemistry version.

2. To analyse the uncertainties in the estimation of regional NO_x emission strength from GOME.

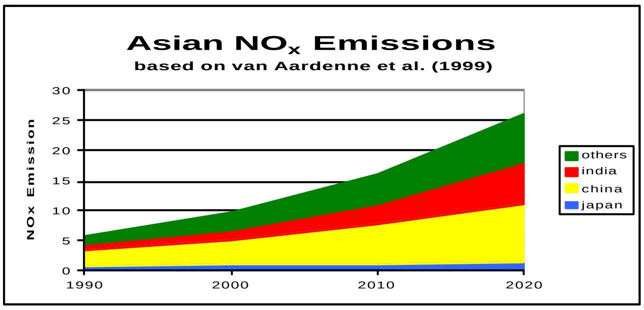
Why Asia?

- 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
- Incresing trend in trace gases/aerosols such as NO_x, CH_4 and Hydrocarbons.

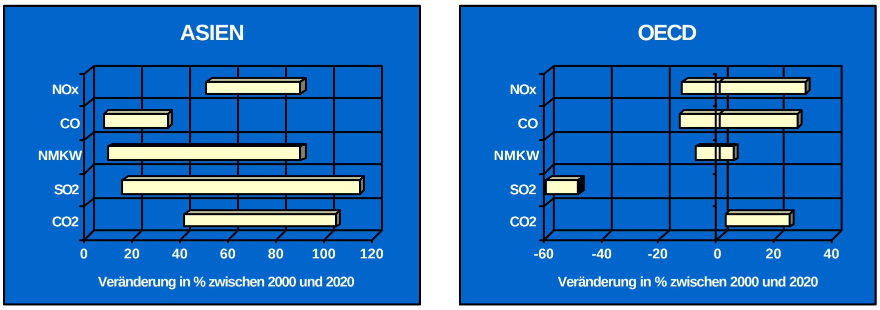
Why NOx ?

$NOx = NO + NO_2$

- 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



IPCC-REPORT



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:
  - CH_4-CO-NO_x-HO_x-O_x
  - 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

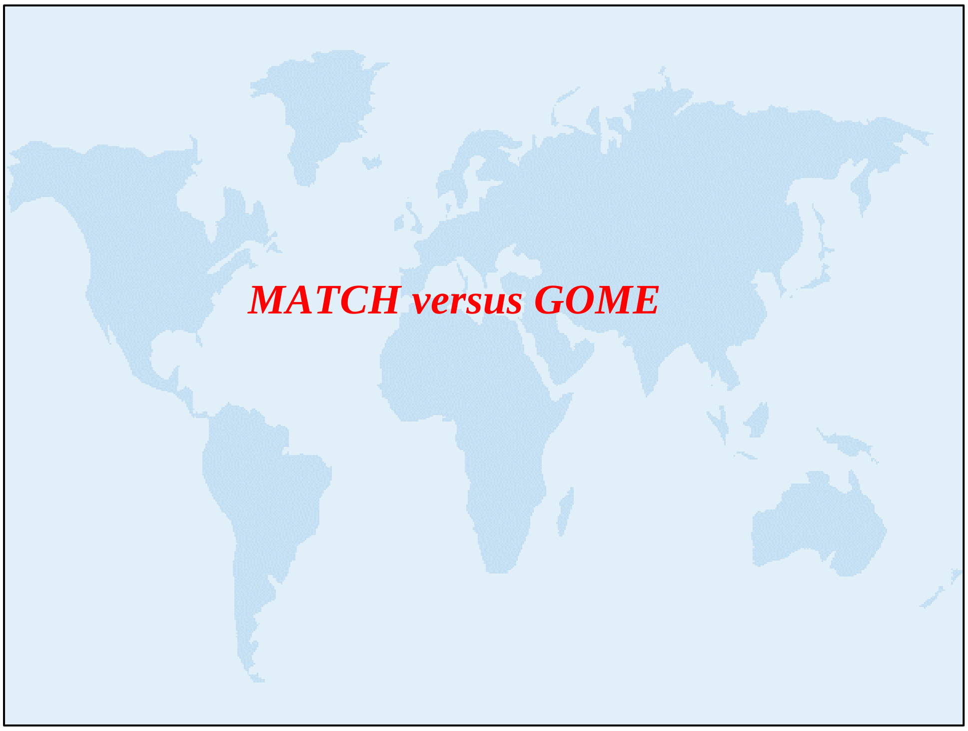
GOME

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_2 absorption : DOAS method
Air mass factors for the NO_2 column : GOMETRAN

Assumptions : Clear sky, a maritime aerosol, a surface albedo- 0.05 , Constant mixing ratio of NO2 < 1.5 km
(Burrows J P et al. (1999) , Richter et al. (2002)

Overall uncertainty in Tropospheric NO_2 col. (Heland et al., 2002).	~ 50% Less in clear situations



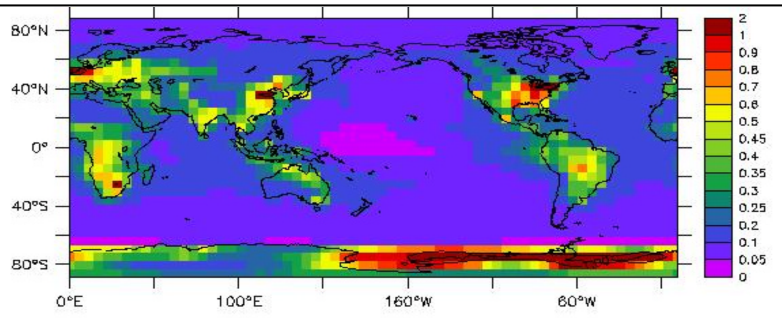
1. GOME Retrieval Assumptions

Basic assumptions are

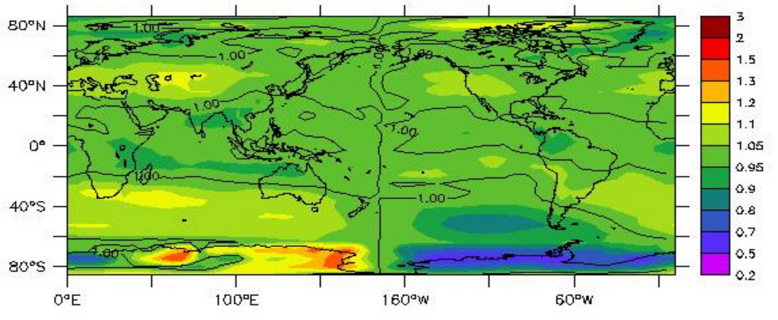
(Richter and Burrows (2002), Burrows et al., 1999)

- **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



Ratio of tropospheric column of NO2 to stratospheric coulmn –July 1997



Ratio of stratospheric column of NO2 to the total NO2 column for the reference sector–July 1997

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)

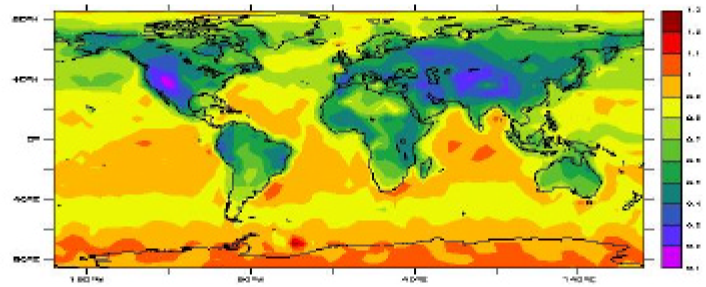
   (MATCH modified to write the output at 10:30 LT)

2. **Could add cloud screening to the model output, similar to the GOME cloud screening.**

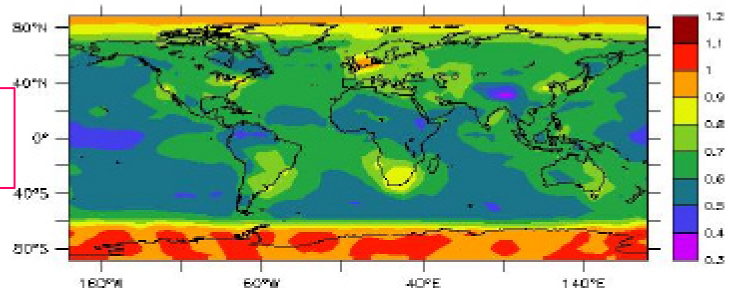
   (Impact of diurnal variation of cloud on NO_2 column)

RATIO – 10:30 to 24 hour Averge NO_2 col. From MATCH, JUL .97

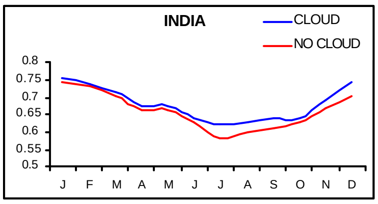
Surface Level

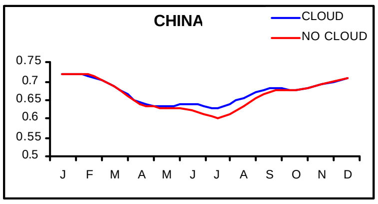


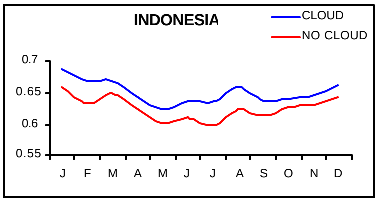
Tropo. NO_2 col. surf-150hPa

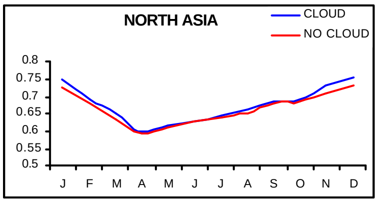


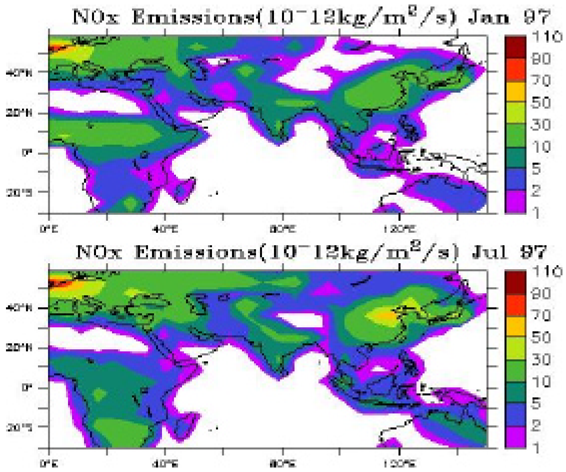
RATIO OF TROPOSPHERIC NO_2 COL. AT 10:30 LT TO 24 H

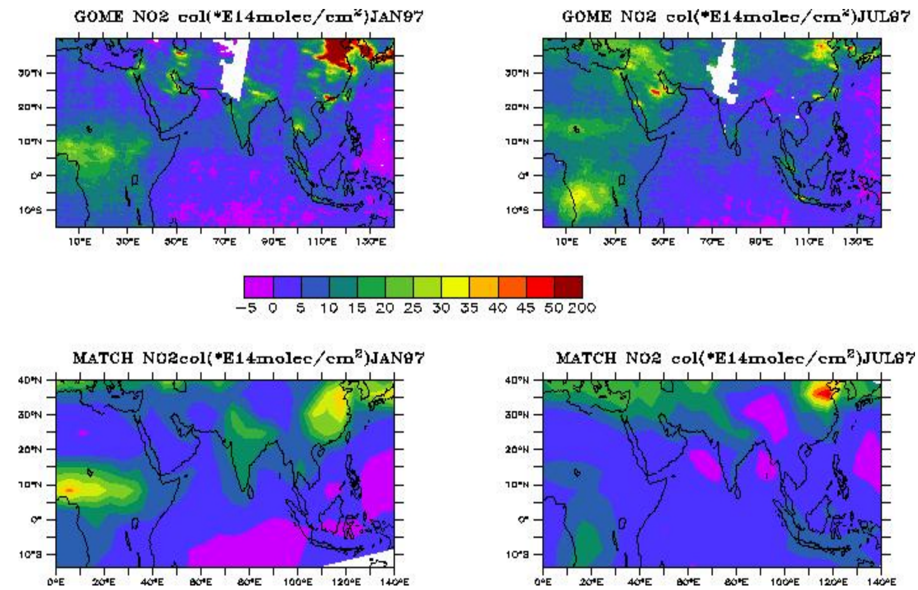


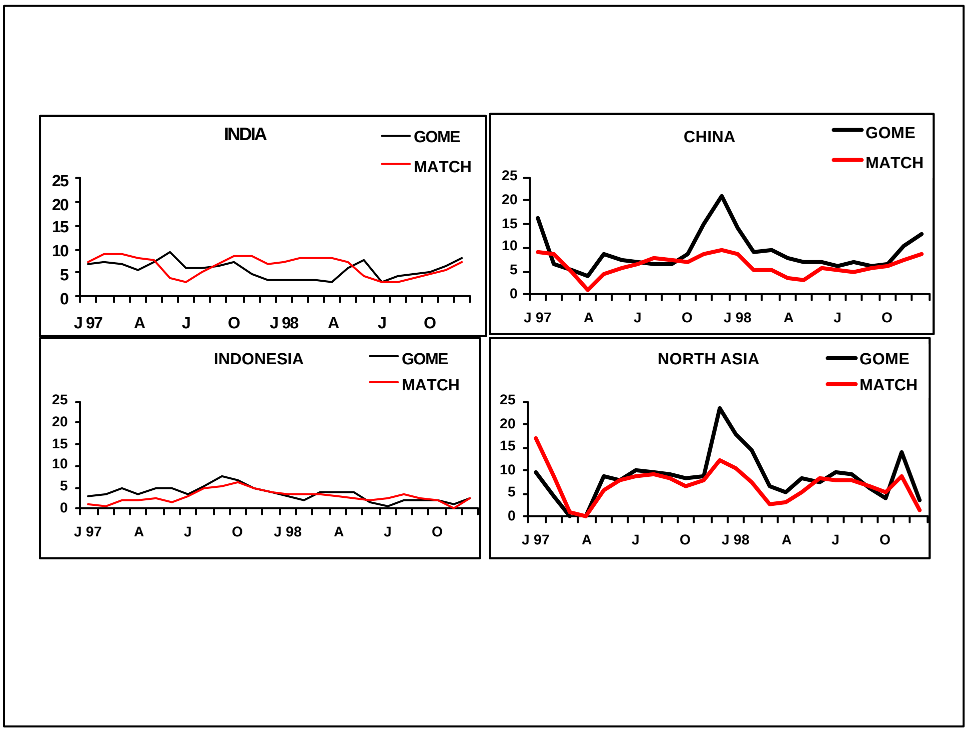


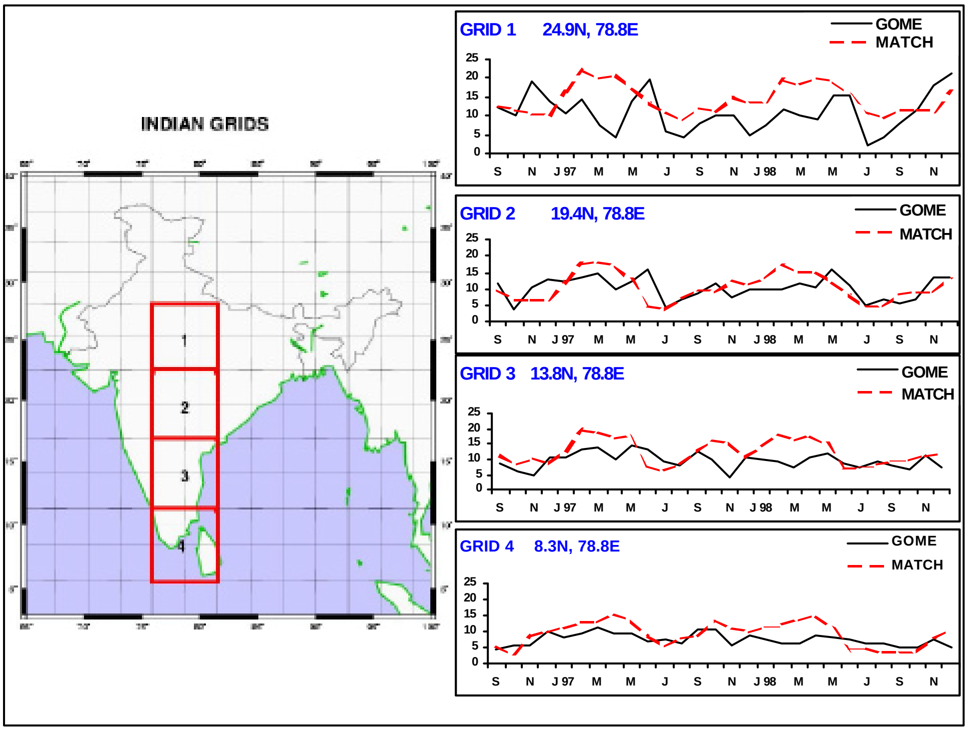





NOx Emissions(10^-12kg/m^2/s) Jan 97
110 90 70 50 30 10 5 2 1
NOx Emissions(10^-12kg/m^2/s) Jul 97
110 90 70 50 30 10 5 2 1


GOME NO2 col(*E14molec/cm^2)JAN97
GOME NO2 col(*E14molec/cm^2)JUL97
-5 0 5 10 15 20 25 30 35 40 45 50 200
MATCH NO2col(*E14molec/cm^2)JAN97
MATCH NO2 col(*E14molec/cm^2)JUL97

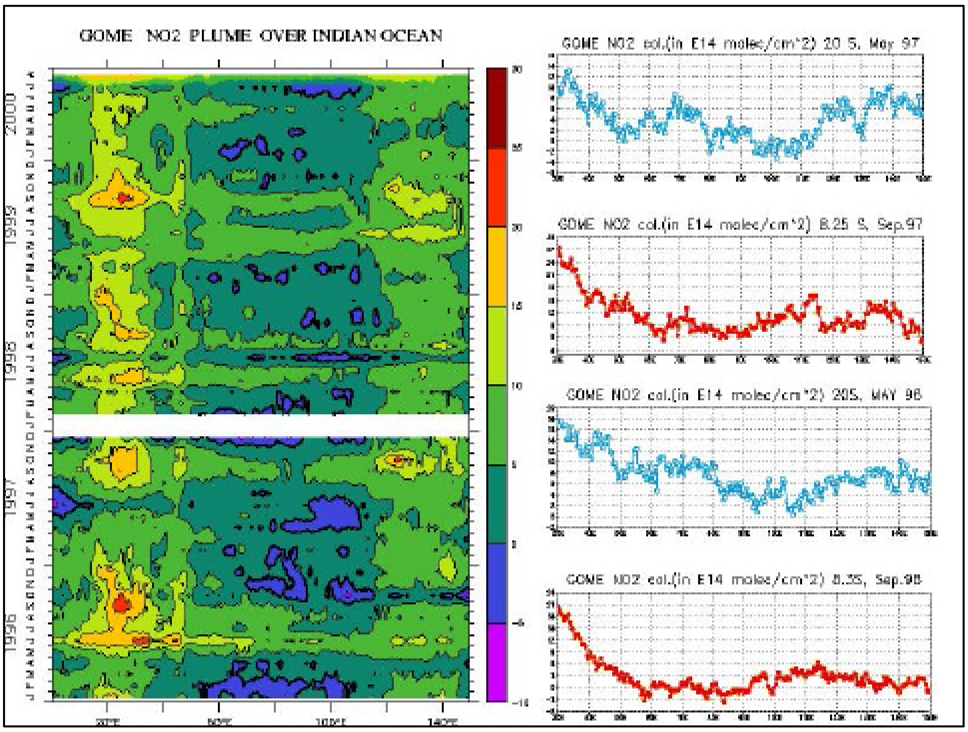

INDIA
GOME
MATCH
CHINA
INDONESIA
NORTH ASIA
J 97
A
J
O
J 98

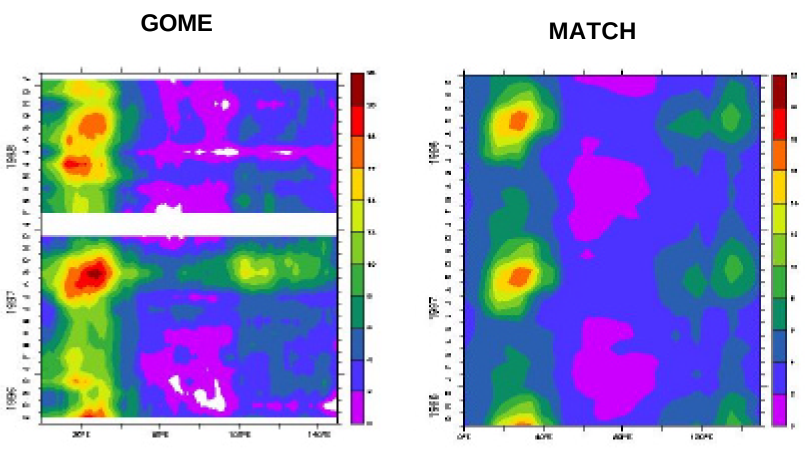

INDIAN GRIDS
GRID 1 24.9N, 78.8E
GRID 2 19.4N, 78.8E
GRID 3 13.8N, 78.8E
GRID 4 8.3N, 78.8E
GOME
MATCH

Region	GOME NO_2 Abundance (10^{14} molec/cm^2)			MATCH NO_2 Abundance (10^{14} molec/cm^2)		
	Mean	Maximum	SD	Mean	Maximum	SD
India (5-35°N, 60-95° E)	5.68	9.6	2.0	6.48	12.6	1.82
China (20-70°N, 90-150°E)	9.05	20.8	4.05	6.21	12.8	2.09
Indonesia (10°S-20°N, 95-140° E)	3.49	7.4	1.65	2.84	8.7	1.33
North Asia (30-70°N, 60-90°E)	8.61	23.7	5.17	7.05	23.1	3.72

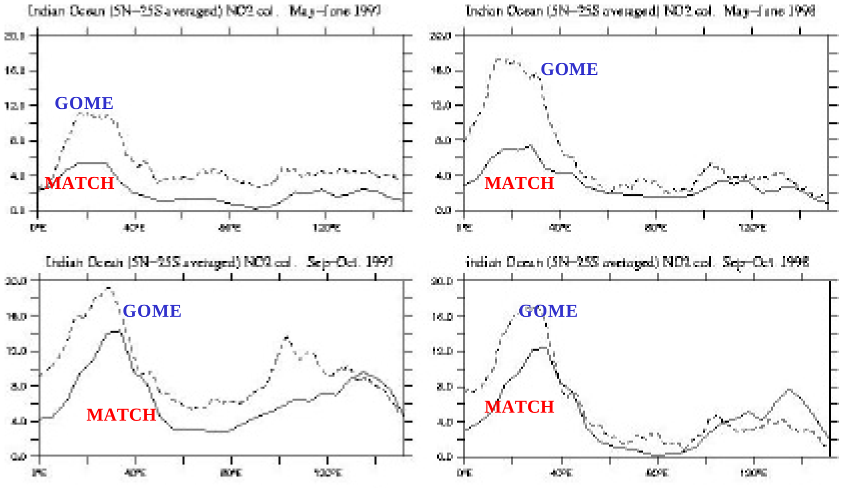
INTERANNUAL VARIABILITY OF NO_2 COLUMN FROM GOME & MATCH

"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……."

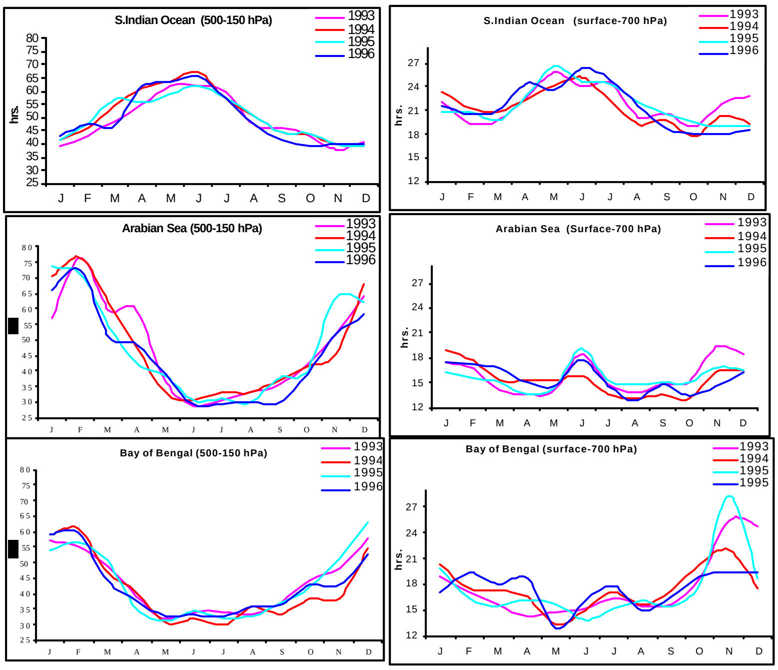




Tropospheric NO2 column (in 1014 molecules/cm^2) from GOME (left) and MATCH(right) over Indian Ocean (5 N–25 S)

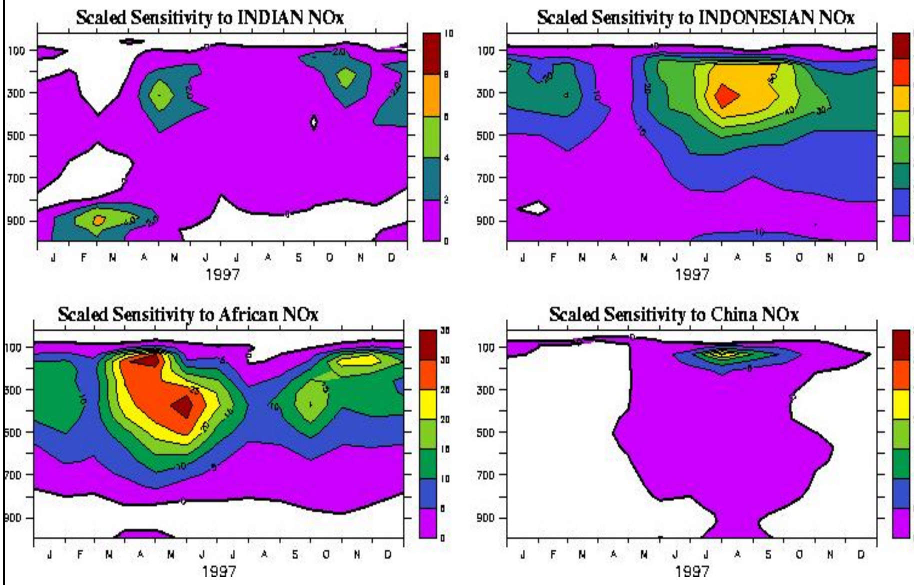


Tropospheric NO2 column (in 10^14 molecules/cm^2) from GOME (dotted line) and MATCH (solid line) over Indian Ocean



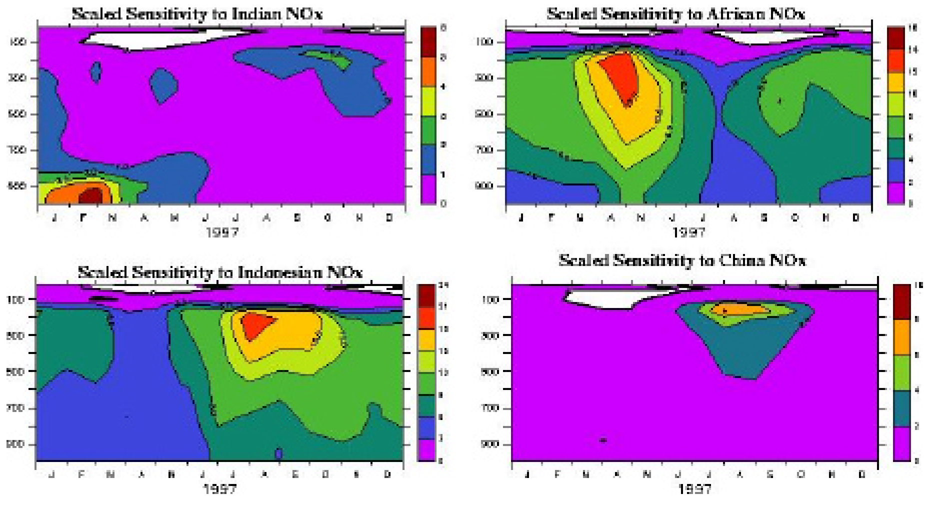
NO2 LIFETIME FROM MATCH OVER INDIAN OCEAN

NO_x Relative changes (%)

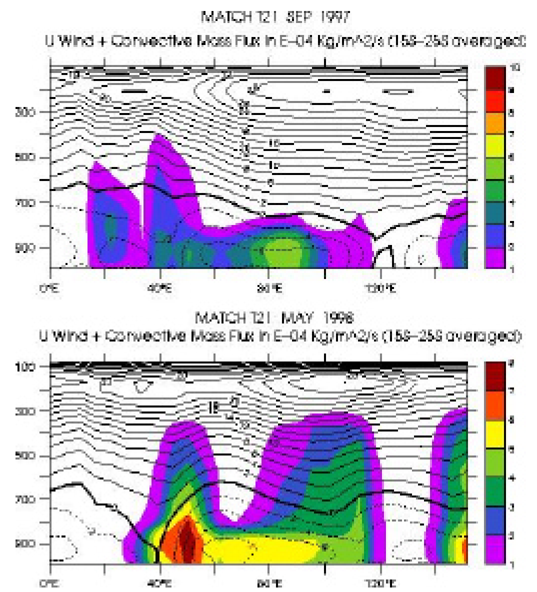


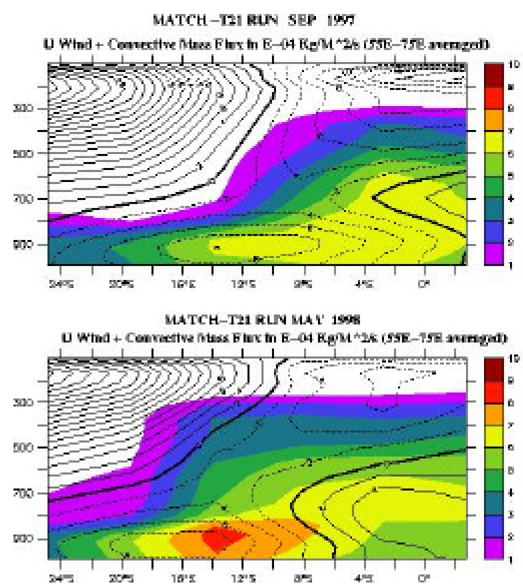
SOUTH INDIAN OCEAN (55ºE-95°E, 5ºN-25°S)

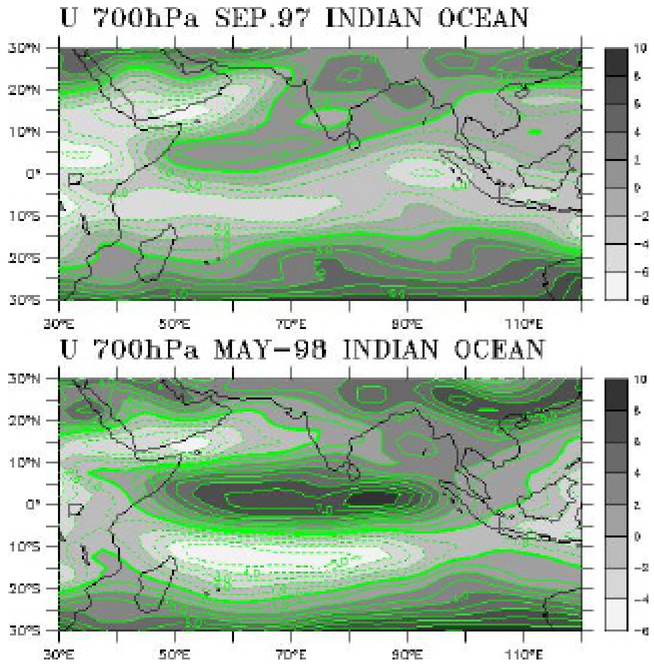
O_3 Relative Changes (%)



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


MATCH T21 SEP 1997
U Wind + Convective Mass Flux in E-04 Kg/m^2/s (15S-25S averaged)
MATCH T21 MAY 1998
U Wind + Convective Mass Flux in E-04 Kg/m^2/s (15S-25S averaged)


MATCH-T21 RUN SEP 1997
U Wind + Convective Mass Flux in E-04 Kg/M^2/s (55E-75E averaged)
MATCH-T21 RUN MAY 1998
U Wind + Convective Mass Flux in E-04 Kg/M^2/s (55E-75E averaged)



Possible Uncertainties in the estimation of Regional NO_x emission strength from GOME!!

Sensitivity of Tropospheric NO_x over India and Indian Ocean

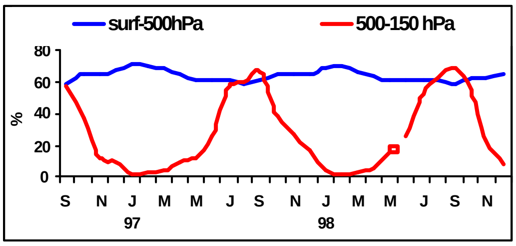
- Sensitivity runs versus Base run of MATCH

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

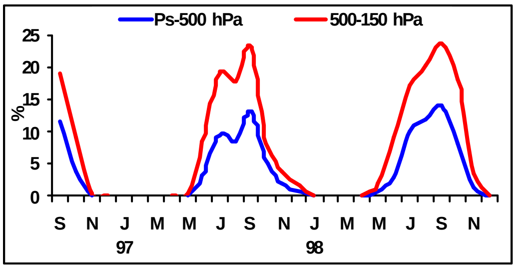
(ii) As (i) except including normal lightning NO_x emission from India.

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

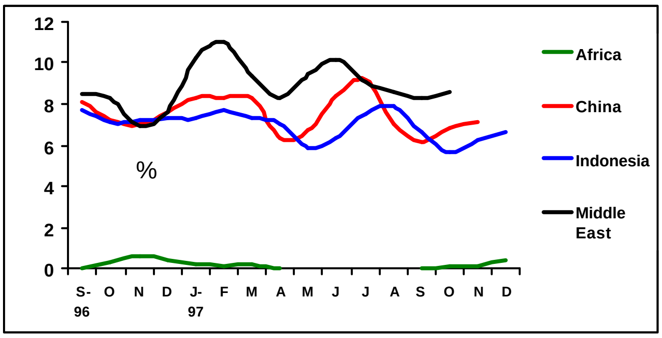
Scaled Sensitivity of NO_x to Local Source-India



Lightning NO_x (%) over India

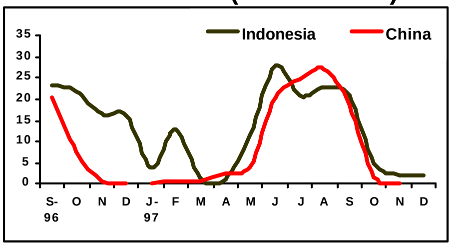


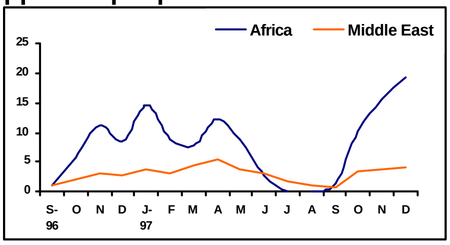
Sensitivity over India to Tropospheric NO_x Sources (%)

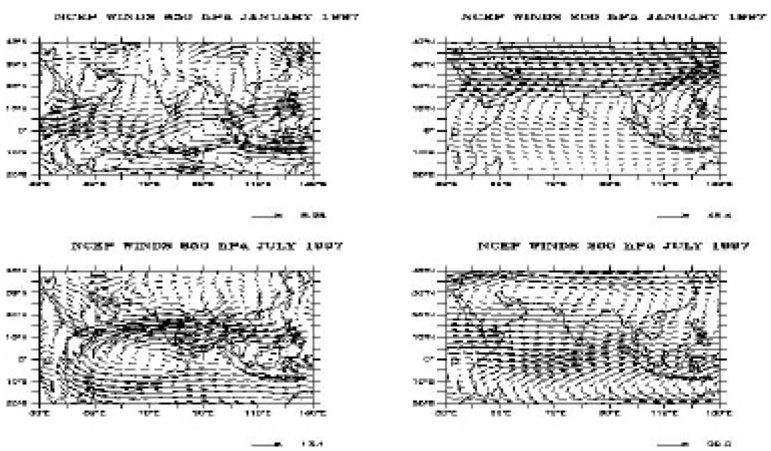


Lower Troposphere (Surface-500 hPa)

(500-150 hPa) Upper Troposphere







LIFETIME OF NO_2 from GOME & MATCH

GOME- Chemical Decay method (850 hPa).

MATCH-Mass-emission method (Asia) &
Main NO_2 loss against HNO_3(Arabian sea, MABL)

Exponential Decay Curve Method -Lifetime of NO_2 from GOME

Chemical decay of NO_2 over Ocean
where there is no emission.

$\partial C/\partial t = L \times C$ i.e. $C = C_0 \cdot e^{-t/\tau}$, where $\tau = 1/L$

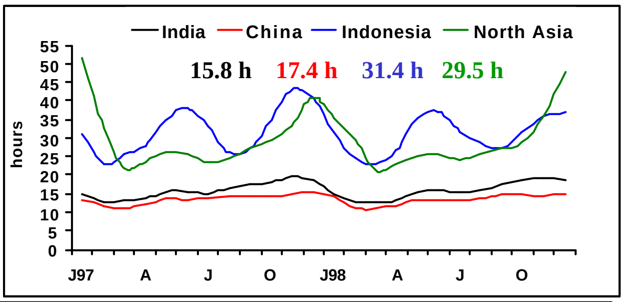
L= constant loss rate, C=concentration after a time t

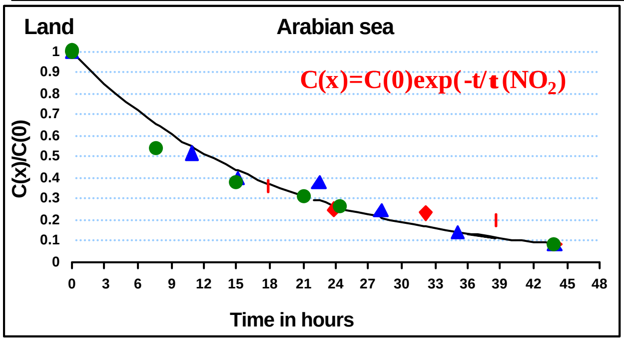
The study site:
Over the Arabian sea along 3 trajectories (31°, 45° and 49°) from Mumbai (72.75° E,19.25° N) - West coast of India.

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

Initial NO_2 maxima from GOME :2.2×10^{15} molecules/cm^2

Source of Wind speed: NCEP monthly mean





MATCH
(MABL-Arabian Sea)

$\tau(NO2)$=16.2 hrs

GOME
(MABL-Arabian Sea)

$\tau(NO2)$=18.2 hrs

Regional NO_x Emission Strength for INDIA

Source	Year	Mean NO_x (Tg(N)/yr) Emission strength	References/ Remarks
MATCH	1997 (met) 1990-emission EDGAR	Mean: 1.72 Max: 2.3 (April)	LT: 15-20 hrs Area: MATCH grids-Indian domain (present study)
GOME	1997	1.87 (an improved one from GOME)	LT: 27 hrs. Area: Extended Indian region including neighbouring nations Wenig (2002)
GOME	1997	2.95	LT: 27 hrs. Area: Extended Indian region including neighbouring nations. Leue et al.(2001)
RAIN-ASIA	1990	1.52	Aardenne et al.(1999).based on anthropogenic sources.
ENERGY STATISTICS-INDIA	1995	3.46	Garg et al.(2001) Based on sector analysis-India

Regional NO_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_x is from remote sources-which introduces a non-negligible uncertainty.**
- **Regionally appropriate lifetime of NO_x is not straightforward from GOME and can be calculated from the model.**

Impact of NO_x emission on other Trace gases over India from MATCH

The findings ......
"moderate increase or decrease of NO_x over India are not expect to lead to large changes in the regional O_3 levels"

Impact of NO_x perturbation on Trace gases over India

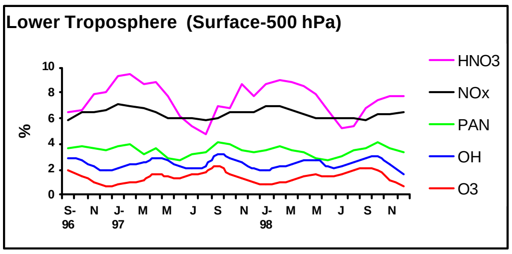
% increase of Trace gases with respect to 10% increase of No_x source over India.

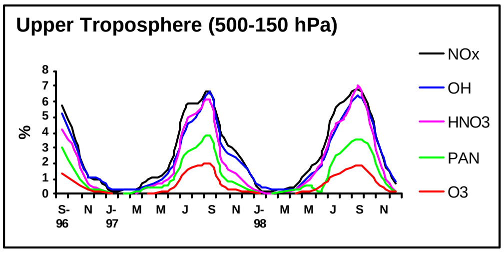
Lower Troposphere (surf-500 hPa)	
(i) NO_x	6-7 %
(ii) O_3	1-2.5 %
(iii) OH	3-5 %
(iv) PAN	5-6 %
(v) HNO_3	5-10 %

Upper Troposphere (500-150 hPa)
Variations are more seasonal with respect to the seasonal variations of NO_x

	Maximum
(i) NO_x	~ 6-7 %
(ii) O_3	~ 1- 2 %
(iii) OH	~ 5-6 %
(iv) PAN	~ 3 %
(v) HNO_3	~ 4-7%

Impact of 10% perturbation of NOx source over India





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

Burrows J.P et al. (1999), J. Atmos. Sci., 56, 151-175

Lawrence M.G et al. (1999), J. Geophy. Res., 104, 26245-26277.

Leue C et al. (2001), J. Geophy. Res., 106, 5493-5505.

Richter A et al. (2002), Adv. Space Res. 29(11), 1673-1683.

von Kuhlmann R et al. (2003a), J. Geophy. Res. 108, D9, 4294.