

## 7.5 LINKING AIR QUALITY TO AIR-SEA INTERACTION BASED ON SPACE OBSERVATIONS AND A GLOBAL CHEMISTRY-METEOROLOGY MODEL

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### 1. INTRODUCTION

Urbanization and increasing demand of energy needs have resulted in a deterioration of air quality over Asia. North India covering the Indo-Gangatic plain (Figure 1) is the most populated region in south Asia. Observations from satellites have shown that extensive pollution exist in this region because of local anthropogenic emissions where combustion of both bio-fuels and fossil fuels are widespread, also natural emissions such as lightning and biomass burning in summer. During the last decade, the Indian Ocean Experiment (INDOEX) has made both meteorological and atmospheric chemistry measurements with the results indicating that there may be considerable impacts on regional climate (Ramanathan et al., 2005). The analyses of Fishman et al (2003, 2005) have likewise confirmed that regional enhancements of tropospheric O<sub>3</sub> over northern India are readily identified from long term observations using total ozone mapping spectrometer (TOMS)-climatology. The present study is aimed at further extending these conclusions by focusing on the regional dynamics controlling NO<sub>x</sub> induced tropospheric O<sub>3</sub> over north India based on long term proxy satellite observations and a global model simulations.

The chemical composition of the atmosphere over south Asia is driven by the monsoon meteorology. As the Asian monsoon is driven by the differential heating of Ocean and adjacent land masses, its

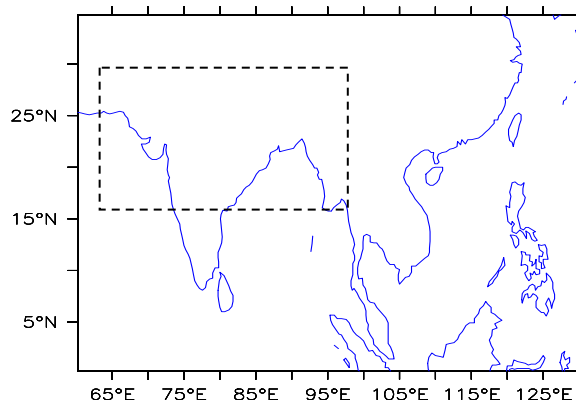


Figure 1 Schematic sketch of north India covering Indo-Gangatic plain of south Asia (enclosed by dashed line, including oceanic region)

variability is mainly linked to land-ocean thermal contrast and changes in the position of convective zones. ENSO phenomenon is the prominent climate variability related to air-sea interaction. The regional chemistry is mostly influenced by synoptic meteorological features such as large scale transport and convective mixing depending on the chemical lifetime of species and the emission strength. There is no distinct dynamic link established between the interannual variation of tropospheric O<sub>3</sub> and ENSO related drought conditions over south Asia. ENSO influences on air pollution are associated with shifts in the convective patterns and precipitation regimes, resulting severe drought over western pacific and reduced summer monsoon rainfall over India. ENSO conditions possibly delays the onset of monsoon as well as increase in the length of non-rainy days in India (Gosawmi and Xavier, 2005). This condition leads to widespread biomass burning and enhanced emissions of hydrocarbons and NO<sub>x</sub>. Studies based on Fishman et al (2005), Thompson et al (2001), Ziemke and Chandra (2001), have related the

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tropospheric O<sub>3</sub> over Asia with ENSO signals through changes in SSTs over western and eastern Pacific. This study examines the impacts of summer ENSO conditions with changes in tropical cellular convection on NO<sub>x</sub> induced O<sub>3</sub> over the Indo-China monsoon regime.

The Indo-Gangatic plain is a vast region close to the foot hills of Himalaya extending an area from East Pakistan in the west of India to the Bay of Bengal in the east. The region includes mega cities in terms of pollution such as New Delhi and Calcutta. Tropospheric O<sub>3</sub> is an important green house gas and is also a powerful oxidizing agent through OH. O<sub>3</sub> in the troposphere is produced by photochemical oxidization of CO, CH<sub>4</sub> and NMHC in the presence of NO<sub>x</sub> (Crutzen, 1974). Partially burned hydrocarbons and NO<sub>x</sub> emissions with maximum hours of sunshine are found to be the major factors controlling tropospheric O<sub>3</sub> over India. The sink of ozone is mainly by photolysis and also by chemical reaction with OH and HO<sub>2</sub> (Crawford et al, 1999). Loss of O<sub>3</sub> generally occurs over southern India close to the equator, having high temperature and humidity as compared to north India. It is also noted that, O<sub>3</sub> production efficiency with respect to precursor emissions vary over north and south India and it is more sensitive to NO<sub>x</sub> as compared to non methane hydrocarbon emissions (Kunhikrishnan et al., 2006). NO<sub>x</sub> can adversely affect the atmospheric chemistry because it catalyzes the production of O<sub>3</sub>. NO<sub>x</sub> is a byproduct of the combustion of biomass and bio-fuel burnings and also from lightning. Tropospheric O<sub>3</sub> and NO<sub>2</sub> over south Asia and the ENSO induced changes on air quality over Indo-Gangatic plain are discussed here, based on observations of tropospheric O<sub>3</sub> from TOMS, Solar Backscatter Ultraviolet (SBUV) instrument (Fishman et al., 2003), and NO<sub>2</sub> from global ozone monitoring experiment (GOME) satellite (Richter and Burrows, 2002) along with output from a global chemistry-meteorology model (Lawrence et al., 1999, von Kuhlmann et al., 2003).

## 2. DATA

Remote sensing of trace gases such as O<sub>3</sub> and NO<sub>2</sub> from space is considered as a new era in atmospheric chemistry and remote sensing (Fishman et al., 1986; Burrows et al., 1999). Observations from different satellite instruments such as TOMS and GOME have provided an opportunity to monitor and analyses the spatial and temporal variability of O<sub>3</sub> and NO<sub>2</sub> column in the troposphere. In this study, we use the two proxy tropospheric data sets from space such as O<sub>3</sub> from TOMS (TOR) and NO<sub>2</sub> column from GOME (TNO<sub>2</sub>) satellites to improve our knowledge on tropospheric O<sub>3</sub> with respect to increasing trends of NO<sub>x</sub> emissions over Asia. The data sets are totally independent with respect to their retrieval methods and satellite missions, but they are chemically reliant species and play key role in the chemical composition of the earth's atmosphere. The TOMS instrument onboard the Nimbus-7 (1978-1993) and the Earth probe (1997-2000) satellites have been used to retrieve the total column of O<sub>3</sub> from space. Tropospheric O<sub>3</sub> is obtained by subtracting the stratospheric column from the total column of O<sub>3</sub>. Details of computing the tropospheric ozone residual (TOR) from TOMS are given in Fishman et al. (2003).

GOME is a scanning spectrometer instrument on ERS-II satellite which was launched by the European space agency in April 1995. The primary measurement of GOME is the backscattered and reflected radiance within the spectral range of 240-790 nm with a spectral resolution of 0.2-0.4 nm. GOME crosses the equator at 10:30 local time in the descending (N-S) node. The Present study uses the tropospheric NO<sub>2</sub> column for the period of 1996-2000, based on the retrieval method as explained in Richter and Burrows (2002). For the retrievals used in this work, the spectral window 423–451 nm, where NO<sub>2</sub> has strong absorption features, has been selected. Generally, GOME uncertainty is smaller for the continental source regions as well as high albedo regions.

MATCH-MPIC is a semi-offline model with a

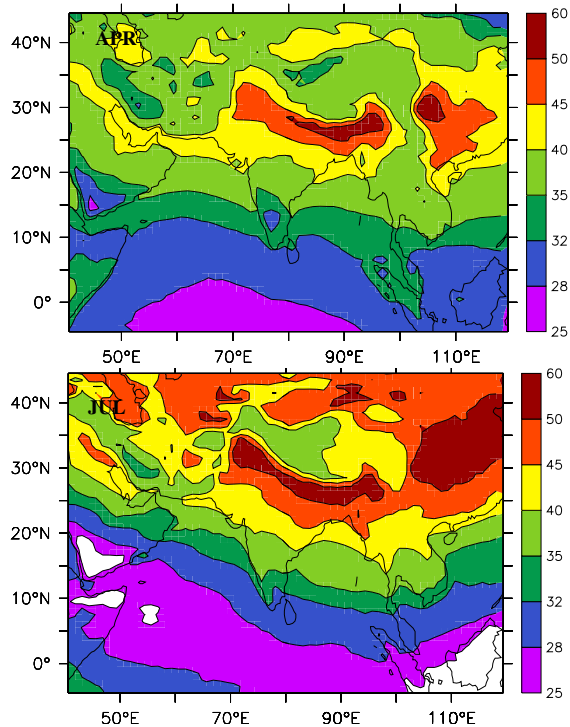


Figure 2 Tropospheric O<sub>3</sub> over south Asia observed from TOMS climatology (1979-2000) in April (top panel) and June (bottom panel).

moderate horizontal resolution of 5.6° (also 1.875°) and a vertical resolution with 28 levels from surface to 2.5 hPa in sigma coordinate system. The details of MATCH-MPIC are given by Lawrence et al., (1999), Rasch et al (1999) and von Kuhlmann et al (2003). The chemical component represents the major sources, transformation and sinks which includes 56 O<sub>3</sub> related species with 140 chemical reactions. The model reads in gridded, time dependent values for most basic meteorological parameters and uses these to derive parameters such as convective transport and cloud microphysics. The meteorology component represents the transport by advection, convection and vertical diffusion and is driven by NCEP reanalysis data. The model transport and physical parameterizations are mainly based on the NCAR climate community model (CCM3). Mass conserving multidimensional flux form scheme (Rasch and Lawrence, 1998) is used to simulate the advection in

the model. MATCH convection schemes are based on the penetrative deep convection (Zhang and McFarlane, 1995) and the shallow moist boundary layer convection (Hack, 2004). The model simulated O<sub>3</sub> related chemistry has been evaluated for different regions over the globe with observations from ground, ship, aircraft and space based platforms (Lawrence et al., 2003, Von Kuhlmann et al., 2003, Kunhikrishnan et al., 2004). The model is able to reproduce some of the observed features such as land-sea contrast, intraseasonal variability of pollution plumes over oceanic regions, vertical profile structures in the tropics and extra tropics especially for some of the key tropospheric species such as NO<sub>x</sub>, O<sub>3</sub> and CO.

### 3. OBSERVATIONS AND MODEL SIMULATIONS

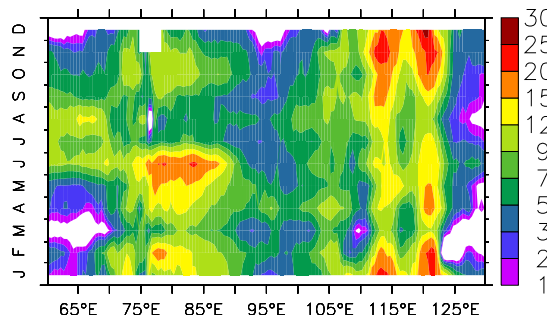


Figure 3 Hovmuller plots of tropospheric NO<sub>2</sub> abundance (in 10<sup>14</sup> molecules/cm<sup>2</sup>) from GOME averaged for north India in 1997

Concurrent observations of O<sub>3</sub> and its precursor NO<sub>2</sub> can provide a better depiction of NO<sub>x</sub> induced O<sub>3</sub> distributions by substantially increasing the spatial and temporal coverage from space. Seasonal TOR over India shows wide range of spatial and temporal variability and its enhancement over north India in summer (Figure 2). TOMS observes an 'O<sub>3</sub> envelope' pollution structure over the Gangatic Plain close to the foot hills of Himalayas throughout the seasons, elongated within the longitudinal domain of 70°E-95°E. It is more pronounced (~35-60 DU) in the dry summer and the initial phase of Indian summer monsoon (June to mid July) over north India (15°N-30°N). Seasonal variation of TOR shows zonal structure with latitudinal gradient in summer, between

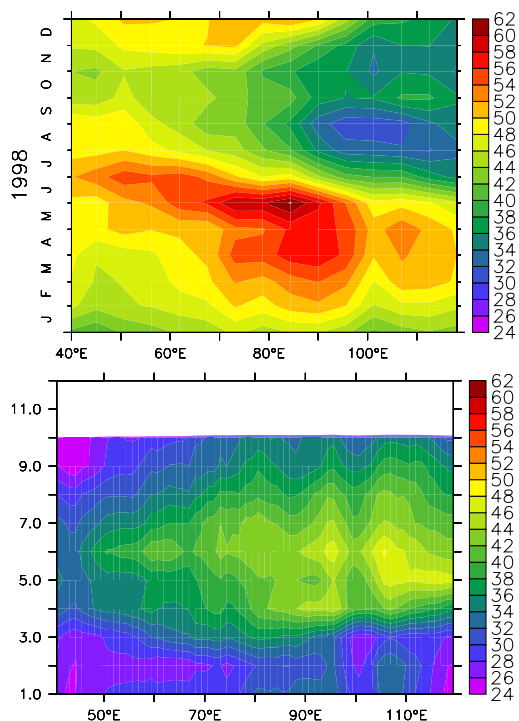


Figure 4 Hovmuller plots of tropospheric O<sub>3</sub> abundance (in DU) from MATCH-MPIC (upper panel) and TOMS (lower panel) averaged for north India in 1997

north and south India, which corresponds to TNO<sub>2</sub> gradient as observed from GOME and has no definite structure in winter or post monsoon. The model derived tropospheric O<sub>3</sub> (TO<sub>3</sub>) is estimated by integrating from surface to tropopause levels from NCEP Reanalysis data. TOR and TO<sub>3</sub> are comparatively less (~25-40 DU) in winter and post-monsoon.

Hovmuller plots (shown in figure 3 and 4) NO<sub>2</sub> and O<sub>3</sub> show that north India is more polluted in summer as compared to south (5°N-15°N) India. The maximum observed NO<sub>2</sub> abundance from GOME (Figure 3) meridionally averaged over north India, accounts with in a range of [15, 25], as compared to [5, 10] (all in 10<sup>14</sup> molecules/cm<sup>2</sup>) over southern India. In proportional to that, tropospheric residual of O<sub>3</sub> from TOMS observes an enhanced abundance of 44-50 DU over north India and is relatively more (52-60 DU) in the model. These features are common in summer (March-July) over the Gangatic plain and the

adjoining central India with considerable interannual variability. More enhancements are commonly seen over north-west and north-east India. NO<sub>x</sub> emission over India-China is high during summer and is found to be ~35% more than the annual mean emissions. The major source regions of biomass/bio-fuel burning in India are the Indo-Gangatic plain, central-east coast and the south (Venkataraman et al., 2005). NASA-MODIS fire counts show that north and central India within 15°-30°N are the region of enhanced biomass burning in summer, varying depending upon the delayed monsoon onset and break conditions. Bio-fuel burnings associated with agricultural operations are common in summer and it would be large and a major fraction as compared to the natural origin over this region. Lightning also contributes considerable upper tropospheric NO<sub>x</sub> emission in summer (Crawford et al., 2000). NO<sub>x</sub> redistribution during ENSO is related to spatial changes in lightning associated with the shifting of convection to the east. Space observations reveal that biomass burning emissions are reported to be significant over the south east (SE) Asia during El-Nino (Wooster and Strub, 2002).

#### 4. ENSO INDUCED WALKER CIRCULATION AND ITS IMPACTS ON REGIONAL O<sub>3</sub> CHEMISTRY

The major tropical circulation which influences the chemical composition and transport are the Walker, monsoon and Hadley circulations. Walker circulation is considered as zonal cells and the centre of convergence is determined by the superposition of the contribution from walker, monsoon and Hadley cells. Walker cells are two times stronger than Hadley and monsoon circulation. The dynamics of this cellular convection is often linked with inter-annual variability of the monsoon. The performance of the Asian summer monsoon is often determined by the relative importance of these cells where weak monsoons are generally associated with strong walker circulations especially seen over south Asia during El-Nino years. Walker cells are closely related to the restoring force, gravity, which acts through

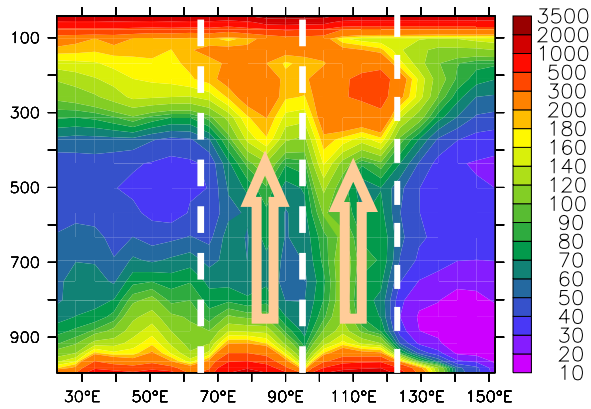


Figure 5 Vertical structure of  $\text{NO}_x$  (in ppt) averaged for north India ( $15^\circ\text{N}$ - $30^\circ\text{N}$ ) for June 1997 from MATCH-MPIC. Arrows indicate the convective updraft over India and SE Asia-China regime.

divergence and can be explained better in terms of velocity potential (VP), which is a surrogate of ENSO. The chemistry over south Asia in summer is also determined by the position of Inter tropical convergence zone (ITCZ) accompanied by the northward movement of convection, which is forced by frictional convergence into low-pressure and ends near the foot of Himalayas. The eastward movement of convection is also seen in the northern hemispheric tropics south to  $30^\circ\text{N}$ . The propagations of convective cells are generally active, prior to the onset of monsoon (spring). These convective cloud clusters along with the tropical circulation play a key role in the regional atmospheric chemistry by uplifting of polluted air from local sources as well as transport from external sources within south Asia are discussed below.

Walker cells have been identified over the south Asia with ascending branch over east India, Bangladesh, and Burma, while its descending limb lies over Pakistan and adjoining northwest Indian desert region. The second oceanic walker cells (Liu et al., 1997) can be seen over the pacific regime with ascending motion over the western equatorial pacific and Indonesia and descending branch over the eastern Pacific. During El-Nino, these cells shift to the east so that the continental convective zones moves to the SE Asia-China regime. This makes major

changes in the redistribution of chemical composition associated with the shift of convective zones to the region of pollution sources with large scale biomass burning and thus by external influences to India via transport and also by subsidence in summer El-Nino. We examine the dynamical impact by convective transport from pollution source regions to nearby downdraft region by descending walker cells. These cells involve secondary circulations associated with convective cloud clusters, which may have large influences on spatial variation of  $\text{O}_3$  production over south Asia. As a case study, the circulation features over south Asia with regional emissions are examined for summer 1997. Seasonal maximum  $\text{NO}_x$  emissions are generally seen over this region in summer: 2.1 Tg (N) [India], 2.0 Tg (N) [South Asia], and 4.68 Tg (N) [China]. The vertical structure of modeled  $\text{NO}_x$  over south Asia averaged for  $15^\circ\text{N}$ - $30^\circ\text{N}$  for June 1997 is shown in figure 5. Most of the  $\text{NO}_x$  in the troposphere is accumulated in the regions close to the surface and also above the 500 hPa level with noted maxima over India, SE Asia and China. The figure depicts the influence of convective updrafts in the UT over the  $\text{NO}_x$  source regions and the compensating subsidence, which accumulates and increases the  $\text{NO}_x$  lifetime in the free troposphere. This takes hundreds of parts per trillion of volume (pptv) of  $\text{NO}_x$  in the UT within the latitudinal frame ( $15^\circ\text{N}$ - $30^\circ\text{N}$ ) over the Indo-China region. These features are common in

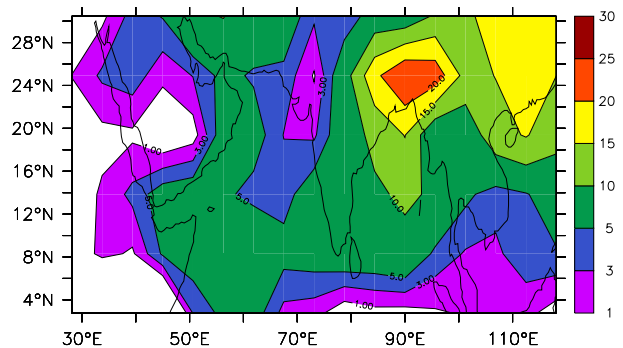


Figure 6 Relative increases in percentage of tropospheric  $\text{O}_3$  from MATCH simulations in 1997 with respect to 1999

summer with interannual variations depending on



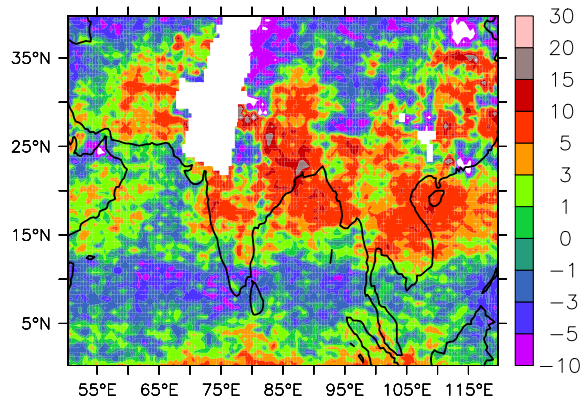


Figure 7 Difference in Tropospheric NO<sub>2</sub> column ( $10^{14}$  molecules/cm<sup>2</sup>) over south Asia from GOME for June 1997 versus June 1999.

regional emission strength. As shown similar to NO<sub>x</sub>, UT enhancement (~50-60%) of O<sub>3</sub> can be seen over central India in 1997 as compared to 1999 (Figure not shown). Tropospheric column in 1997 shows an excess of 5 to 25% of O<sub>3</sub> over most of the regions of India (Figure 6). The meso scale transport time is crucial in determining how fast the tracer like NO<sub>x</sub> is lifting to the UT and thereby increasing its lifetime. Different earlier studies of the effects of deep convection on O<sub>3</sub> (Lelieveld and Crutzen, 1994, Pickering et al., 1996) indicate that convective mixing of O<sub>3</sub> itself reduces the amount of O<sub>3</sub> on the troposphere since the mixing of O<sub>3</sub> rich air down towards the surface when lifetime is shorter. However a study (Lawrence et al., 2003) based on a CTM shows that convective mixing of precursors, especially NO<sub>x</sub>, opposes this reducing effect and leads to a net increase of O<sub>3</sub> in the troposphere, since the photochemical efficiency of NO<sub>x</sub> is greater when it is diluted by convective mixing. This implies that great influence of regional O<sub>3</sub> is from local NO<sub>x</sub> over north India in addition to the external influence from SE Asia and China by transport of O<sub>3</sub> and its precursors. As compared to 1999, ~50 to 120% increase ( $+15 \times 10^{14}$  molecules/cm<sup>2</sup>) of NO<sub>2</sub> column over India and the surrounding region can be seen from GOME in June 1997 (Figure 7). In-situ surface observations over a north Indian station, Delhi report an anomalous increase in O<sub>3</sub> during summer 1997 as compared to

1999 (Figure 8). Generally GOME observes the NO<sub>2</sub> pixels with cloud cover less than 0.1. Retrieval based on TOMS (Fishman et al., 2005, Creilson et al., 2003) indicates that the influence of cloud impacts on O<sub>3</sub> do not produce much differences with the 'TOR residual method' after comparing two different data sets of TOMS with pixels of 'cloud' and 'cloud-free'. We particularly examine the total cloud fraction from the model for two different monsoon years 1997 and 1999; which shows that the overall difference is less than a fraction of 0.17 for India. Further, we do not expect the effects of cloud screening to be large over India in order to produce a significant difference of tropospheric O<sub>3</sub> (~30%) and TNO<sub>2</sub> ( $\geq 50\%$ ) for two different years based on space observations. Moreover the in-situ observations over India also show above 50% more O<sub>3</sub> in 1997 than in 1999 in summer.

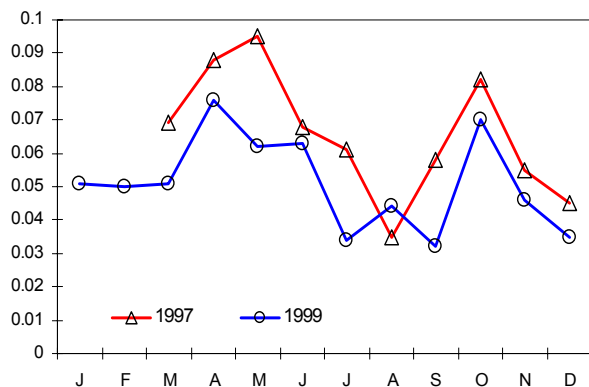


Figure 8 Surface O<sub>3</sub> (in ppm) from in-situ measurements for New Delhi in 1997 and 1999 (The data is based on Jain et al., 2005).

During ENSO years, the centre of convergence (confluence) is shifted to SE Asia and the respective diffluence (center of divergence) aloft eventually shifts toward India with respect to height so that polluted air mass from SE Asia and China is convectively uplifted and transported to the mid to UT over India. Dynamical process related to chemistry can be explained in terms of the anomalies in the VP which is subjected to vary due to the shifting of walker cells. In June 1997, the positive anomalies of VP (0.846 sigma level) for this region shift to SE Asia-China

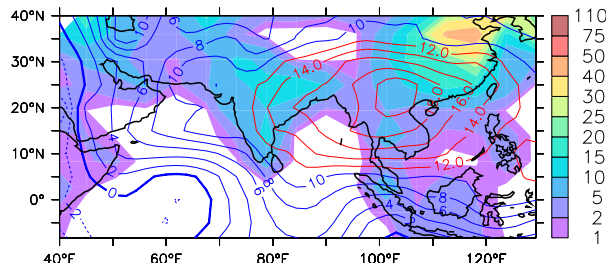


Figure 9 Surface NO<sub>x</sub> emissions (in 10<sup>12</sup> kg/m<sup>2</sup>/s) used in MATCH (in shaded color) and the velocity potential anomaly (in contours, red contours depict the anomalous convection) at 850 hPa for June 1997

and this anomalous convection due to ENSO is centered over the NO<sub>x</sub> source regions (Figure 9). The shaded one represents the NO<sub>x</sub> emissions used in the model and the solid contours correspond to the positive anomalies in the VP at the 0.846 sigma levels, which is a region of strong convergence; that is, the confluence of summer circulation in the LT, normally found over north India shifts to the south Asia-China so that convective updraft to the free troposphere is greater over this region with stronger convection. This enhances the influence of emission from South Asia-China to Indian O<sub>3</sub> by transport in mid to UT with descending branch of walker cells and the tropical easterlies associated with anticyclonic circulations, as shown in figure 10.

Isobaric analysis of velocity wind vectors at different levels shows the northward shifting of weak anti-cyclonic ridge at 400 hPa over the southern peninsular India and it gradually becomes stronger and established a strong anti-cyclonic circulation from 300 to 100 hPa levels centered over central to northwest India. As compared to 1999, the UT ridge normally present at the northeast A1 (98°E, 28°N) close to the Tibet will shift to southwest A2 (90°E, 25°N) in summer 1997 so that El-Nino contributes stronger subsidence due to the influence of divergence over India (Figure 10). Even during El-Nino, the convection can reach up to 500 hPa over central India, which makes more influence of upper

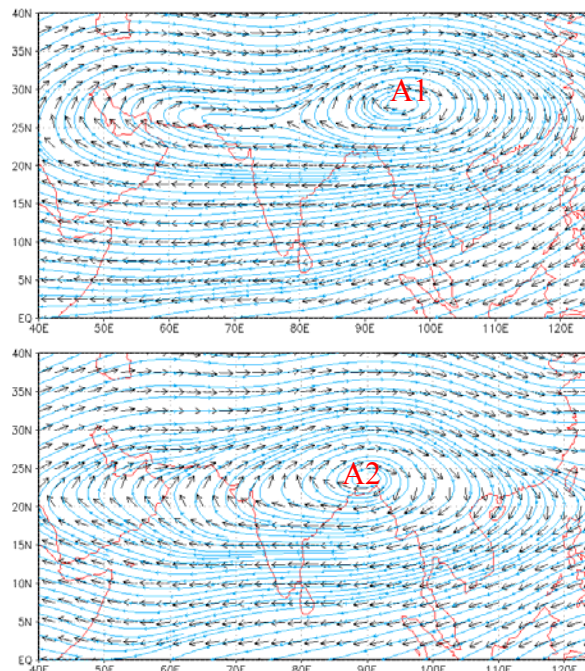


Figure 10 Streamlines with wind velocity vector at 150 hPa in June for 1997 (top panel) and 1999 (bottom panel).

tropospheric O<sub>3</sub> to local emissions. The anti-cyclonic circulations across India are normally active in summer in the vertical plane above MT with subtropical westerly jet in the north and tropical easterlies in the south. It is found that the tropical easterly jet on the lower side of the circulation is relatively weak during ENSO and makes more stagnation of the polluted air mass over India. The horizontal subsidence due to the descending branch of walker cells over India in 1997 appears to be spatially wide and stronger as compared to 1999. The hypothesis is examined with cross sensitivity simulations by increasing 10% of NO<sub>x</sub> emissions in the model base run from India, China, South Asia, Africa and the Middle East based on Kunhikrishnan and Lawrence (2004). Model simulations indicate that 30% of O<sub>3</sub> over India is influenced by the local NO<sub>x</sub> emissions. However NO<sub>x</sub> emission from China and South Asia contributes ~20% of O<sub>3</sub> each to India in summer 1997. It is significant, if we consider the negative feedback of NO<sub>x</sub> through OH. Similarly we note that China-O<sub>3</sub> is also sensitive to NO<sub>x</sub> emissions

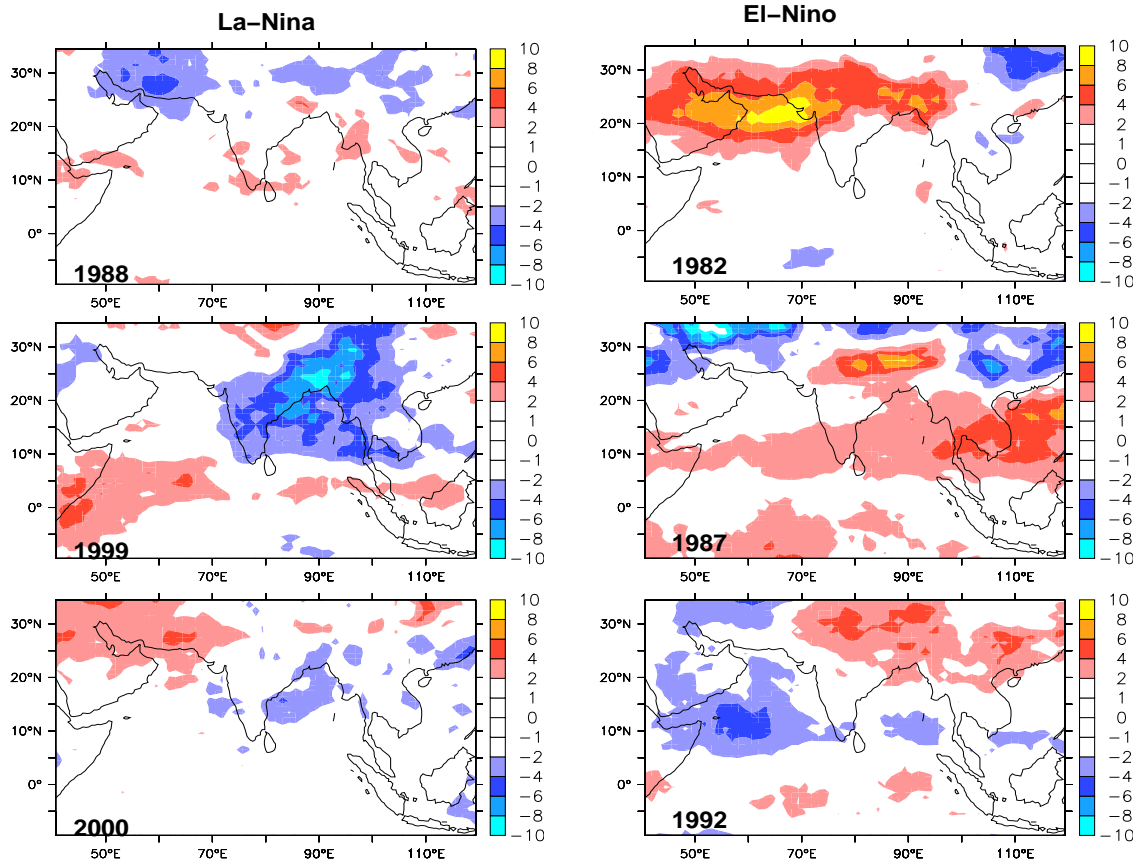


Figure 11 TOR-anomaly from long term TOMS-Climatology (1979-2000) for El-Nino (left hand panel) and La-Nina years (right hand panel) over south Asia for summer (June)

from India and south Asia even though it is relatively small. This shows that the anticyclonic circulation makes the two way sensitivity in the Indo-China regime and traps the pollutants across the region in summer. The sensitivity of Indian  $O_3$  to external emission is found to be 5 to 7% less in normal or La-Nina years as compared to El-Nino years.

##### 5. INTERANNUAL VARIABILITY OF TROPOSPHERIC $O_3$ OVER SOUTH ASIA

The overall feature of TOR anomalies from monthly mean climatology for contrasting ENSO years (Figure 11) in summer (June) depict an additional burden of TOR (2-10 DU) over north India during the negative phase of southern oscillations (1982, 1987, 1988) in contrast to La-Nina years (1988, 1999, 2000) with negligible or negative anomalies. However in the post monsoon (Sep.-October), EIO is more vulnerable to tropospheric  $O_3$  pollution in the El-Nino years with

positive anomalies of  $\sim 2-8$  DU. We note that, the difference in the total cloud fraction over this regime in two contrasting years is generally less than 0.15. This implies that the positive TOR anomalies as observed from TOMS during ENSO years are not an artifact induced by the influence of clouds in the retrieval. The positive TOR anomalies shift from India to the EIO during post monsoon and is followed by the shifting of ITCZ. It is noted that, the influence of ITCZ on TOR over the EIO is apparent only during El-Nino where the TOR anomalies are generally negligible in normal or La-Nina years. The vertical structure of walker circulations can be better depicted in terms of VP in the LT (0.846 sigma) and the UT (0.212 sigma). The VP indicates the divergent (and non-rotational) component of the flow (Krishnamurthy et al., 1972) and, by this, the region with upward or downward motions; that is the local maxima (minima) of the VP indicate upward (downward) motions at a particular



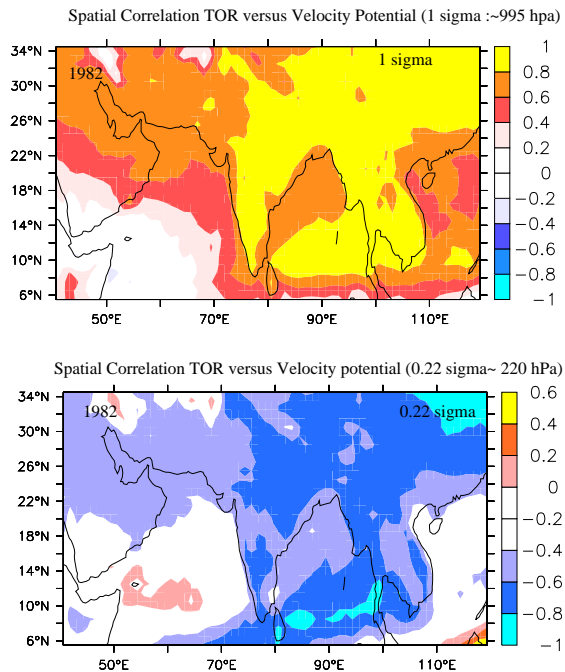


Figure 12 spatial correlation between TOR and velocity potential at 0.846 sigma level (upper panel) and 0.212 sigma level (lower panel) for June 1982

isobaric level. The direct spatial correlation plot of TOR with the respective VP for a strong El-Nino of June 1982 (Figure 12) shows a significant absolute spatial correlation of 0.9 in the LT and 0.6 in the UT, which indicates the influence of local and external influence to the tropospheric O<sub>3</sub> over India. The spatial correlations of TOR-anomaly with respect to changes in the strength of walker circulation (VP-anomaly at 0.22 sigma level) for summer are shown in figure 13. High positive correlation (0.4-0.8) of these two anomalies can be seen over north India, Arabian Sea, East of Pakistan and part of Indian Ocean (Figure 13). In summer El-Nino, the correlation is found to be stronger with more subsidence over north India through stronger walker cells along with more biomass burning as compared to normal or La-Nina years. However the relationship seems to be weaker over south India. Similarly, we are also noted the higher spatial correlation of anomalies of TOR and VP over the EIO during September-October period. In contrast to the stronger link of ENSO on regional O<sub>3</sub>,

we have noted an enhanced model simulated O<sub>3</sub> over India for a weak El-Nino year 2002, because of severe drought conditions and more anomalous UT subsidence over India. This appears to be linked with the weakening trends of Indian monsoon versus ENSO induced changes as recently reported. However the relation with VP and the tropospheric O<sub>3</sub> remains stronger during this time also; which is an indication of the influence of walker cells on tropospheric O<sub>3</sub> over south Asia.

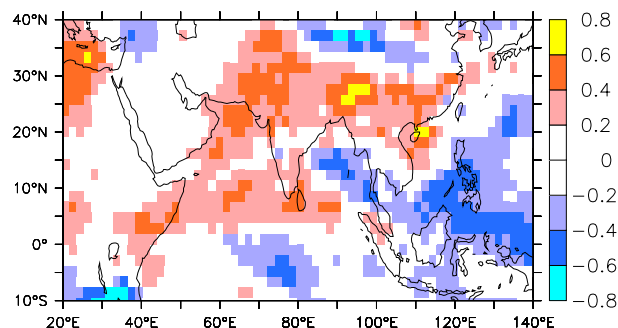


Figure 13 Spatial anomaly correlations between TOR and velocity potential for south Asia during summer (June) from long term data (1980-2000).

## 6. CONCLUDING REMARKS

This study analyzes the long term proxy observations of tropospheric O<sub>3</sub> and NO<sub>2</sub> over south Asia based on TOMS and GOME satellites along with a global chemistry-meteorology model MATCH-MPIC. An enhanced tropospheric O<sub>3</sub> is observed from TOMS over the Indo-Gangatic plain in summer with substantial interannual variability. It is mainly attributed to O<sub>3</sub> precursor emissions from local emissions and transport from SE Asia and China. The model overestimates the tropospheric O<sub>3</sub> (52-64 DU) as compared to that from TOMS (40-54 DU) and are consistent with the relatively large amount of tropospheric NO<sub>2</sub> from GOME (15-25 × 10<sup>14</sup> molecules/cm<sup>2</sup>), observed in summer. Interannual variations of TOR and TNO<sub>2</sub> observations over India show an additional burden of tropospheric O<sub>3</sub> and NO<sub>2</sub> in summer El-Nino years. This study finds a hypothesized link between the NO<sub>x</sub> induced O<sub>3</sub> over the Indo-Gangatic plain and ENSO induced changes in the zonal Walker circulation. Tropical dynamics is

governed by zonal momentum driven by gravity which acts through divergence and convergence. Air quality over this region is examined with velocity potential, which is a surrogate of ENSO. During normal or La-Nina years, the tropospheric O<sub>3</sub> is dominated by the contribution from local emissions. The external influences during these periods are limited by the weaker walker cells with less subsidence and relatively low emissions over SE Asia to transport away the pollutant to the Indian region. In addition to that, the photochemistry is not well active during La-Nina due to high relative humidity and relatively low hours of sunshine hours. Moreover, ENSO impacts the regional O<sub>3</sub> through photochemistry with relatively weak tropical easterly winds and changing the monsoon components such as delay in monsoon onset and increase in the length of non-rainy days.

During summer ENSO, the confluence in the LT is shifted to SE Asia and the respective diffuence eventually shift towards India in the UT with respect to height. These can be seen with an anomalous low over the SE Asia-China NO<sub>x</sub> source regions with the centre of divergence in the UT over India within 200 hPa isobaric levels, which enhances the subsidence and there by trapping the polluted air mass originating from local sources as well as from SE Asia and China. These pollutants are get stagnated by the anticyclonic circulation in the vertical plane extending from MT to UT across north India. This increases the O<sub>3</sub> production efficiency through substantial drying and warming associated with descending branch of walker cells. The findings are supported by the model cross sensitivity simulations. This accounts ~25% O<sub>3</sub> sensitivity of India to local NO<sub>x</sub> emissions, 20% to China and 15% to SE Asia. In turn, O<sub>3</sub> over China indicates relatively low influence (10-15%) of O<sub>3</sub> to SE Asia and India. O<sub>3</sub> Sensitivity over India to external NO<sub>x</sub> emission is comparatively more during El-Nino. The chemical impact of clouds on O<sub>3</sub> through NO<sub>2</sub> photolysis in summer is found to be high and it shows an increase of 20% of O<sub>3</sub> in the UT irrespective of ENSO years.

Interannual variability of TOR from its monthly climatology shows that ENSO induced excess O<sub>3</sub> (~2-8 DU) is seen over north India in summer for all the El-Nino years. The positive anomalies associated with ENSO are further extended over the EIO during post monsoon. This indicates that the role of ITCZ with convection on tropospheric O<sub>3</sub> is significant mostly in ENSO periods. During the normal or La-Nina years, TOR-anomalies are either negligible or negative over the India and the EIO, which indicates that the march of the ITCZ has little impact on the spatial variation of O<sub>3</sub> during this period. Spatial correlation of TOR and velocity potential anomalies at 0.22 sigma level for summer indicates that enhanced O<sub>3</sub> over north India is highly correlated (0.4-0.6) with the subsidence induced by walker cells. This correlation is more significant during summer ENSO years. Similarly we find a considerable correlation of the two anomalies over the EIO during post monsoon. The above findings indicate that walker circulation has considerable influence on tropospheric O<sub>3</sub> distribution over the south Asia. Walker cells are active mainly during summer associated with monsoon circulation. Therefore transport will be much more during June-October. However chemistry will be more active from March to July due to enhanced NO<sub>x</sub> emissions with favorable meteorological conditions. After seeing the recent trends of observations from SCIAMACHY (Richter et al., 2005), we can conclude that anthropogenic emissions are increasing steadily over this region and this may impact the regional air quality more adversely with recent changes in the monsoon dynamics over the Indo-Gangatic plain.

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