



Influence of enhanced Asian NO_x emissions on ozone in the Upper Troposphere and Lower Stratosphere (UTLS) in chemistry climate model simulations

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Abstract:

Asian summer monsoon convection plays an important role in efficient vertical transport from the surface to the anticyclone. In this paper we investigate the potential impact of convectively transported anthropogenic nitrogen oxides (NO_x) on the distribution of ozone in the Upper Troposphere and Lower Stratosphere (UTLS) from simulations with the fully-coupled aerosol chemistry climate model, ECHAM5-HAMMOZ. We performed anthropogenic NO_x emission sensitivity experiments over India and China. In these simulations, anthropogenic NO_x emissions for the period 2000-2010 have been increased by 38% over India and by 73% over China in accordance with satellite observed trends over India of 3.8 % per year and China of 7.3% per year. These NO_x emission sensitivity simulations show that strong convection over the Bay of Bengal and the Southern slopes of the Himalayas transports Indian emissions into the UTLS. Convective transport from the South China Sea injects Chinese emissions into the lower stratosphere. Indian and Chinese emissions are partially transported over the Arabian Sea and west Asia by the tropical easterly jet. Enhanced NO_x emissions over India and China increase the ozone radiative forcing over India by 0.112 W/m² and 0.121 W/m² respectively. These elevated emissions produces significant warming over the Tibetan Plateau and increase precipitation over India due to a strengthening of the monsoon Hadley circulation. However doubling of NO_x emissions over India (73%); equal to China, produced high ozone in the



lower troposphere. It induced a reverse monsoon Hadley circulation and negative precipitation anomalies over India. The associated subsidence suppressed vertical transport of NO_x and ozone into the anticyclone.

Key words: Asian summer monsoon, Ozone, increase in tropospheric NO_x , NO_x transport, Upper troposphere and lower stratosphere.

1. Introduction

Rapid economic development and urbanization in Asia has resulted in an unprecedented growth in anthropogenic emissions like nitrogen oxides (NO_x), carbon monoxide (CO), carbon dioxide (CO_2), methane (CH_4), etc. Many of these species have implications on tropospheric ozone (Wild and Akimoto, 2001; IPCC, 2001). Ground based and satellite observations show a high amount of these ozone precursors concentrated over the India and China (Sinha et al., 2014; Richter et al., 2005; Jacob et al., 1999; Zhao et al., 2013; Gu et al., 2014). Recent studies show that tropospheric ozone production over Asia is controlled by the abundance of NO_x and VOCs and the ratios of these species (Sillman, 1995, Lei et al., 2004, Zhang et al., 2004 and Tie et al., 2007). These studies reveal that in Asia, particularly India and China are NO_x limited regions, i.e., controlling NO_x in these regions would reduce ozone concentrations (Yamaji et al., 2006; Sinha et al., 2014; Fadnavis et al., 2014). Thus levels of tropospheric ozone, a key polluting agent and greenhouse gas, are controlled by NO_x concentration over these regions. Recently, a positive trend of $3.8 \text{ \% yr}^{-1}$ (for the period 2003-2011) in tropospheric column NO_2 over India has been reported using SCanning Imaging Absorption SpectroMeter for AtmosphericCHartographY (SCIAMACHY) observations (Ghude et al., 2013) and of $7.3 \text{ \% yr}^{-1}$ (for the period 2002-2011) over China from Ozone Monitoring Instrument (OMI) observations (Schneider and van der A, 2012). Apart from anthropogenic sources, lightning also contributes to the production of NO_x . Over the Asian region, in the middle and upper troposphere, lightning contributes $\sim 40 \text{ \%}$ to NO_x



and 20% to ozone production during monsoon season (Tie et al 2001; Fadnavis et al 2015). The upper tropospheric ozone concentration is determined both by in-situ production as well as from convective transport from the boundary layer (Sóvde et al., 2011).

Ozone in the troposphere absorbs relatively little UV radiation but it has a huge climatic impact (Lacis et al., 1990). The estimated global average radiative forcing increasing tropospheric ozone since pre-industrial times is estimated to be $+0.35 \pm 0.15 \text{ Wm}^{-2}$ (IPCC, Ramaswamy et al., 2001). Previous studies revealed that ozone perturbations exert a large influence on temperature near the surface and tropopause (e.g., Thuburn and Craig, 2002; Foster and Shine 1997). Thus the tropospheric ozone distribution and its variability at different altitudes influence the thermal structure of the atmosphere. Climate change due to ozone variations is highest near the tropopause because the greenhouse efficiency per molecule is highest there (Lacis et al, 1990; Forster and Shine, 1997; Riese et al., 2012). A study based on ACCMIP models reports that NO_x and CH_4 are the greatest contributors in determining ozone radiative forcing (Stevenson et al., 2013).

Asian Summer Monsoon (ASM) convection efficiently transports Asian pollutants from the boundary layer into the Upper Troposphere and Lower Stratosphere (UTLS) (Randel and Park, 2006; Randel et al. 2010; Fadnavis et al., 2013, 2015). Studies pertaining to modeling and trajectory analysis confirm this finding (Li et al., 2005; Park et al., 2007; Randel et al., 2010; Chen et al., 2012; Vogel et al., 2015). Satellite observations show the confinement of a number of chemical constituents like water vapor (H_2O), CO, ethane (C_2H_6), CH_4 , nitrous oxide (N_2O), hydrogen cyanide (HCN), and aerosols, within the ASM anticyclone (Randel et al. 2010; Li et al., 2005; Randel and Park, 2006; Lawrence et al., 2011) which has potential implication on stratospheric chemistry and dynamics. These studies indicate that the rise in anthropogenic emissions over the ASM region alters the chemical composition of the UTLS (Lawrence et al., 2011; Fadnavis et al, 2014, 2015) during the monsoon season. Another prominent feature of the satellite observations is an ozone minimum in the ASM anticyclone (near 100



hPa) (Gettelman et al., 2004; Konopka et al., 2010; Braesicke et al., 2011). This ozone minimum is linked to ozone poor air masses arriving from the Bay of Bengal and Arabian Sea. Balloonsonde observations show that ozone variations near the anticyclone are strongly correlated with temperature near the tropopause (Tobo et al., 2008). Thus observations show that convectively lifted air masses arriving in the anticyclone are ozone poor but rich in ozone precursors. Thus the linkage of low ozone and high ozone precursors with temperature variation in the anticyclone is an open question. In this study we ask the question ‘how do rising Asian NO_x (an important ozone precursor) emissions and associated ozone production affect the ozone radiative forcing and monsoon circulation?’ For this we perform anthropogenic NO_x emission sensitivity experiments using the state-of-the-art ECHAM5-HAMMOZ (European Centre General Circulation Model version5) chemistry climate model (Roeckner et al., 2003; Horowitz et al., 2003; Stier et al., 2005). From these simulations ozone radiative forcing for different anthropogenic NO_x emission scenarios is estimated. Associated temperature and monsoon circulation changes are also reported. The paper is organized as follows: data description and model setup are discussed in section 2; results are discussed in section 3. Section 4 comprises of discussion followed by conclusion in section 5.

2. Data description and Model setup

2.1 Satellite measurement

Earth Observing System (EOS) microwave limb sounder (MLS) is a NASA Aura satellite which flies in the polar sun-synchronous orbit. It measures the thermal emissions at millimeter and sub-millimeter wavelengths (Waters et al., 2006). It performs 240 limb scans per orbit with a footprint of ~ 6 km across-track and ~ 200 km along-track, providing ~ 3500 profiles per day. MLS also measures the vertical profiles of temperature, ozone, CO , H_2O , and many other constituents in the mesosphere, stratosphere and upper troposphere (Waters et al., 2006). In the UTLS, MLS has a vertical resolution of



3 km. MLS vertical profiles of ozone show good agreement with the Stratospheric Aerosol and Gas Experiment II (SAGE-II), Halogen Occultation Experiment (HALOE), Atmospheric Chemistry Experiment (ACE) and ozonesonde measurements (Froidevaux et al., 2006). The MLS ozone profiles are considered to be useful in the range of 215 – 0.46 hPa (Livesey et al., 2005). In this study we analyze the MLS level 2 (version 4) ozone mixing ratios data for the period 2004 – 2013. The data has been gridded at $8.3^{\circ} \times 3^{\circ}$. This data can be accessed from <http://mls.jpl.nasa.gov/>. For comparison simulated ozone is convolved with the MLS averaging kernel. Details of MLS averaging kernels are documented by Livesey et al. (2011).

2.2 Model simulation and experimental setup

We employ the aerosol-chemistry-climate model ECHAM5-HAMMOZ which comprises the general circulation model ECHAM5 (Roeckner et al., 2003), the tropospheric chemistry module, MOZ (Horowitz et al 2003) and the aerosol module, Hamburg aerosol model (HAM) (Stier et al., 2005). It includes NO_x , VOC and aerosol chemistry. The gas phase chemistry is based on the chemical scheme provided by the MOZART-2 model (Horowitz et al., 2003) which includes detailed chemistry of the O_x - NO_x hydrocarbon system with 63 tracers and 168 reactions. The $\text{O}(^1\text{D})$ quenching reaction rates were updated according to Sander et al., (2003) and isoprene nitrates chemistry according to Fiore et al., (2005). The dry deposition in ECHAM5-HAMMOZ follows the scheme given by Ganzeveld and Lelieveld (1995). Soluble trace gases like HNO_3 and SO_2 are also subject to wet deposition. In-cloud and below-cloud scavenging follows the scheme given by Stier et al. (2005).

The model is run at a T42 spectral resolution corresponding to about $2.8^{\circ} \times 2.8^{\circ}$ in the horizontal dimension and 31 vertical hybrid $\sigma - p$ levels from the surface to 10 hPa. The details of model parameterizations, emissions and validation are described by Fadnavis et al. (2013; 2014; 2015) and Pozzoli et al. (2008a, b; 2011). The base year for trace gas emissions is taken as 2000. Each member of



our sensitivity study consists of continuous simulations for eleven years from 2000 to 2010. For each simulation emissions were the same. Meteorology varied due to varying sea surface temperature (SST) and sea ice (SIC). The AMIP2 SSTs and SIC representative of the period 2000 – 2010 were specified as a lower boundary condition.

In order to understand the impact of increasing NO_x emissions on the distribution of ozone in the UTLS, simulations were performed for the period 2000 – 2010. The experimental set up is the same as described by Fadnavis et al., (2015). The four experiments analyzed in this study are (1) A reference experiment (CTRL) and three sensitivity experiments (referred to as experiments 2 - 4), where the NO_x emissions over India and China are scaled in accordance with the observed trends. In experiment (2), NO_x emissions are increased over India by 38% (Ind38), in experiment (3) increases that over China by 73% (Chin73) are prescribed. In order to analyze the effects of equal NO_x emissions over India and China, NO_x emissions are increased over India by 73% over India (Ind73) in experiment (4). The emission perturbations were obtained from observed NO_2 trends of 3.8% per year over India (Ghude et al., 2013) and 7.3% per year over China (Schneider and van der A, 2012). Hiboll et al., (2013) also reported similar increasing NO_x values over megacities in India and China.

To calculate the heating associated with ozone changes over India and China, we use the radiative transfer model Santa Barbara DISORT Atmospheric Radiative Transfer (SBDART). This model is designed and developed by the University of California, Santa Barbara (Ricchiazzi et al., 1998) and is based on a collection of well-tested and reliable physical models. We used eight streams in the radiative transfer calculation and computations were made for solar zenith angles at every 5° .

3. Results

3.1 Comparison with AIRS satellite measurements in the UTLS

Figure 1(a) shows the spatial distribution of MLS ozone mixing ratios at 100 hPa. The



145 distribution of simulated ozone mixing ratios at the nearest model level, 90 hPa is plotted in figure 1(b).
146 ECHAM5-HAMMOZ simulation is smoothed with averaging kernel of MLS. The winds plotted in
147 figure 1(a) and 1 (b) show the extent of the monsoon anticyclone (20°E-120°E). Winds plotted in figure
148 1(b) depict the extent of the monsoon anticyclone (20°E-120°E). The spatial pattern of low ozone
149 concentrations in the monsoon anticyclone is well simulated in the model. In the recent past, low
150 ozone mixing ratios (~40-160 ppbv) in the monsoon anticyclone have been reported from MLS (90-140
151 ppbv), MIPAS (80-120 ppbv), SAGE II (<150ppbv) measurements (Kunze et al., 2010; Randel et al.,
152 2001; Randel and Park 2006; Park et al., 2007). Vertical profiles of ozonesonde (averaged for the
153 monsoon season during 2001-2009) at Indian stations, Delhi (28.61°N, 77.23°E), Pune (18.52°N,
154 73.85°E) and Thiruvananthapuram (8.48°N, 76.95°E) are compared with MLS measurements and
155 ECHAM5-HAMMOZ simulated ozone mixing ratios in figures 1(c)-(e). These figures depict that
156 vertical variation of simulated ozone show good agreement with ozonesonde and MLS observations.
157 Fadnavis et al., (2015) compared the model simulation with aircraft observations over the various
158 regions all over the globe during the monsoon season. This study reported a reasonable agreement for
159 PAN, NO_x, HNO₃ and ozone mixing ratio.

160

161 **3.2 Transport of enhanced NO_x emissions into the UTLS**

162 Recent satellite observations and model simulations established the impact of convective
163 transport of boundary layer pollution into the ASM anticyclone during the Asian monsoon season
164 (Gettelman et al., 2004; Randel et al., 2010; Fadnavis et al., 2013, 2014, 2015). These pollutants are
165 further transported across the tropopause as evident in satellite observations of, e.g. water vapour (Bian,
166 2012), hydrogen cyanide (HCN) (Randel, 2010), CO (Schoeberl et al., 2006), Peroxyacetyl nitrate
167 (PAN), aerosols (Vernier et al., 2015, Fadnavis et al 2013) etc. To understand the influence of monsoon
168 convection on the vertical distribution of NO_x we show zonal and meridional cross sections over India



169 and China. The vertical distribution of NO_x averaged for the monsoon season over the Indian latitudes
170 (8° - 35°N), and Chinese latitudes (20° - 40°N) as obtained from CTRL simulations is shown in the
171 supplementary figures S1(a) and S1(b) respectively. These figures show elevated levels of NO_x
172 extending from the surface to the upper troposphere (UT) over India and China. The wind vectors
173 along with the distribution of cloud droplet number concentration and ice crystal number concentration
174 together, (Figure S2(a) and S2(b)) indicate strong convective transport from the Bay of Bengal (BOB),
175 southern slopes of Himalayas, and south China Sea which might lift the boundary layer NO_x to the
176 upper troposphere.

177 During the monsoon season, the NO_x distribution in the UTLS is influenced by lightning apart
178 from transport from anthropogenic sources. In this season, lightning activity is highest in Asia
179 (Ranalkar and Chaudhari, 2009; Penki and Kamra, 2013) compared to the other monsoon regions
180 (North America, South America and Africa). However, in our simulations there is no contribution from
181 lightning.

182 We show longitude-pressure (Figures 2(a)-(c)) and latitude-pressure sections (Figures 2(d)-(f))
183 of anomalies of anthropogenic NO_x obtained from Ind38, Ind73, Chin73 with respect to CTRL
184 simulations. The black arrows indicate the anomalies of wind vectors. Ind38 and Chin73 simulations
185 (Figures 2(a) and 2(d)) show that the convective winds at the southern flank of the Himalayas (80° -
186 90°E) and over the Bay of Bengal lift up the enhanced NO_x emissions to the upper troposphere (UT).
187 The majority of the outflow is into the UT and only part of it is transported into the lower stratosphere.
188 Cross tropopause transport due to convection over South China Sea is evident in the Chin73 simulation
189 (Figure 2(c) and 2(f)). Previous studies also indicate significant vertical transport due to strong
190 monsoon convection from the southern slopes of Himalayas (Fu et al., 2006, Fadnavis et al., 2013;
191 2014) and the South China sea (Park et al 2009; Chen et al., 2012), which is in agreement with our
192 results. In the upper troposphere, NO_x is transported over Iran and Saudi Arabia along the descending



193 branch of circulation.

194 However the simulation with doubled NO_x emissions over India (Ind73) does not show cross
 195 tropopause transport. The wind anomalies show a descending branch over central India ($\sim 20^\circ\text{N}$, 75°E)
 196 (Figures 2(b) and 2(e)) which might have inhibited the cross-tropopause transport in the Ind73
 197 simulation. The descending wind anomalies may be related to ozone radiative forcing and associated
 198 temperature changes (discussed later in section 4).

199 Figures 2(d) and 2(f) also indicate transport of NO_x from India and China across the tropopause
 200 and entrainment into the tropical pipe. Garny and Randel (2013) and Randel et al. (2010) also reported
 201 that pollution transported by Asian monsoon convection enters the deep stratosphere via the tropical
 202 pipe.

203

204 **3.3 Impact of enhanced anthropogenic NO_x on the tropospheric ozone distribution**

205 Tropospheric ozone is a radiatively active trace gas (Ramanathan et al., 1987) therefore it is
 206 important to know the impact of enhanced NO_x emissions on tropospheric ozone production (Derwent,
 207 1990). We estimate the change in ozone production due to enhanced NO_x emissions in the Ind38, Ind73
 208 and Chin73 simulations with respect to CTRL. The longitude-pressure cross sections of changes in
 209 amount of net ozone production (%) over India and China are shown in figures 3(a)-(c). It can be seen
 210 that at the altitudes below 300 hPa there is ozone production and loss varying in between -60% to
 211 +60%. In the upper troposphere (300-150hPa), the estimated amount of net ozone production in Ind38
 212 and Ind73 simulation is $\sim 5\text{-}10\%$ and that from Chin73 simulation is $\sim 10\text{-}30\%$. Comparison of figures 2
 213 and 3 show that ozone production by NO_x photolysis occur in the regions where there is a transport of
 214 NO_x . In Ind73 simulation there is no change in ozone production near the tropopause as the NO_x
 215 emissions do not reach up to these altitudes (Figures 2(b) and 2(e)). In the Ind73 simulation there is
 216 significant increase in net ozone production ($\sim 40\text{-}90\%$ between 1000-500 hPa) over the Indo Gangetic



217 Plain (figure not included).

218 Figures 4(a)-(c) show the longitude-pressure distribution of ozone mixing ratio anomalies
219 obtained from Ind38 and Ind73 averaged over 8°-35°N, and Chin73 over 20°-45°N. Although the air
220 mass in the monsoon anticyclone is poor in ozone (Figure 1(b)), the elevated ozone anomalies in
221 response to increased NO_x emissions are seen in figure 4 (in the Ind38 and Chin73 simulations). This
222 may be partially due to transported ozone via ASM convection and partially due to production of ozone
223 (Figures 3 and 4); there is no contribution from lightning in our simulations. It can be seen from figures
224 4(a)-(c) that ozone levels are enhanced near 300-200 hPa over Arabia. This may be due to the ASM
225 vertical convective transport and subsequent horizontal transport in the monsoon anticyclone (Randel et
226 al., 2010, Garny and Randel, 2013; Vogel et al., 2015).

227 Latitude-pressure cross section of ozone anomalies plotted in figures 4(d) and 4(f) show that
228 convection over the Bay of Bengal, southern slopes of the Himalayas and the South China Sea lifts
229 ozone from India and China into the upper troposphere and it is then transported across the tropopause
230 into the tropical pipe. Ozone and NO_x transported across the tropopause will enhance their amount in
231 the surf mixing zone above the tropopause (Vogel et al., 2015). In the lower stratosphere there will be
232 ozone production by photolysis from transported NO_x.

233 Doubling of NO_x emissions over India (Ind73) results in a descending branch of circulation
234 over central India (Figures 4(b) and 4(e)). This subsidence suppressed the vertical transport of ozone
235 across the tropopause. It is also evident in NO_x distribution (Figures 2(b) and 2(e)). As discussed
236 earlier, this subsidence may be related to temperature changes due to ozone radiative forcing and
237 heating rates as there is significant increase in ozone production over the Indo Gangetic plain (1000 –
238 500 hPa) (discussed in section 4).

239

240 **3.4 Ozone distribution in the anticyclone**



We estimate changes in NO_x and ozone in the monsoon anticyclone from Ind38, Ind73 and Chin73 simulations with respect to the CTRL simulation. The distribution of these species is shown at 110 hPa as representative for the anticyclone. Figures 5(a)-(c) depict percentage changes in NO_x mixing ratios for Ind38, Ind73 and Chin73 simulations. A maximum in the NO_x anomalies in the ASM anticyclone (20°E to 120°E) is seen in all the simulations. Ozone anomalies are high at the eastern part of monsoon anticyclone since convective injection into the anticyclone occurs at the eastern part of anticyclone (Fadnavis et al., 2013). Percentage increase in NO_x amounts in the Ind38 simulation is higher (Figure 5(a)) than that in the Ind73 simulation (Figure 5(b)). This is mainly due to descending motion over the central India, as seen in the previous sections. However, the Chin73 simulation shows higher values of increasing NO_x ($>18\%$) in the monsoon anticyclone (Figure 5(c)). In the anticyclone, the increase in ozone mixing ratios in Ind38 simulation is 1-3%, Ind73 is 1-5% and Chin73 is 1-8%. These figures show that easterly jet transport NO_x and ozone (from India and China) to Saudi Arabia, Iran and Iraq.

4. Discussion

Increase in ozone mixing ratios may enhance ozone radiative forcing and exert warming (Brasseur, 1997). We estimate anomalies in ozone radiative forcing at the top of the atmosphere for Ind38, Chin73, Ind73 with respect to CTRL simulation over central India ($20^\circ\text{-}30^\circ\text{N}$, $75^\circ\text{-}85^\circ\text{E}$) and China ($30^\circ\text{-}45^\circ\text{N}$, $100^\circ\text{-}135^\circ\text{E}$). The increase in ozone radiative forcing over India, in the Ind38 simulation is 0.112 W/m^2 , and in the Chin73 simulation is 0.121 W/m^2 . In comparison, the estimated global average radiative forcing due to increases in tropospheric ozone since pre-industrial times is estimated to be $+0.35 \pm 0.15 \text{ Wm}^{-2}$ (Brasseur et al., 1997; IPCC, Ramaswamy et al., 2001).

Further, we analyze the temperature anomalies for the Ind38, Ind73 and Chin73 simulations. Figures 6(a)-(c) show the zonal distribution of temperature anomalies. These simulations show surface



265 warming over the 30°-40°N, regions covering North India and North China. The striking feature evident
 266 in Ind38, and Chin73 simulations is anomalous warming over the Tibetan Plateau while it is subdued in
 267 Ind73 simulation.

268 In order to understand the atmospheric heating caused due to ozone, we calculate the ozone
 269 heating rate using the radiative transfer model, SBDART. ECHAM5-HAMMOZ model simulated
 270 temperature, pressure, as well as water vapour concentration and ozone concentrations at various levels
 271 are incorporated into the SBDART model in order to estimate the ozone heating rate in the vertical.
 272 Figure 7(a) shows the vertical profile of ozone radiative heating rate as obtained from CTRL simulation
 273 averaged over Indian (70°-94°E, 20°-30°N) and Chinese (70°-100°E, 30°-40°N) regions. It shows a high
 274 value of ~ 0.003- 0.009 K/day at ~1-4 km. A net radiative heating rate of 0.004 – 0.008 K/day in the
 275 troposphere due to changes in ozone was earlier reported by Ramaswamy and Bowen (1994).

276 Anomalous warming over the Tibetan plateau seen in Figure 6 may be due to heating caused by
 277 ozone. Figure 7(b) shows the vertical distribution of anomalies in ozone heating rates averaged over
 278 Tibetan plateau (70-100°E, 20-40°N). Positive ozone heating rate anomaly (0.0005-0.0001 K/day) is
 279 seen below 14 km in Ind38 and Chin73, on the contrary, Ind73 shows negative anomalies in ozone
 280 heating rate below 8 km. In the upper troposphere (8 – 14 km) ozone heating rates are positive in all the
 281 simulations. They are less in Ind73 ($2.09 \times 10^{-6} - 4.5 \times 10^{-6}$ K/day) as compared to Ind38 ($1.91 \times 10^{-5} -$
 282 2.74×10^{-5} K/day) and Chin73 ($2.17 \times 10^{-5} - 2.99 \times 10^{-5}$ K/day). The estimated heating rates over the
 283 Indo Gangetic plain are $\sim 3 \times 10^{-4}$ K/day (averaged over 1000-800 hPa) this may be due to high amount
 284 of ozone production. Ozone heating rate anomaly of $\pm 2 \times 10^{-4}$ K/day was previously reported by Guo
 285 et al., 2008, over East Asia and the Tibetan plateau region.

286 From figures 3, 6 and 7 it can be said that doubling of NO_x emissions (Ind73 simulation) over
 287 India increases net ozone production over Indo Gangetic plain. Enhanced ozone production leads to
 288 anomalous ozone heating in the lower troposphere over the Indo Gangetic plain ($\sim 3 \times 10^{-4}$ K/day). This



289 elicited reversal of monsoon Hadley cell circulation. The descending branch of monsoon Hadley cell
290 circulation over central India (figures 2 and 4) resisted vertical transport of ozone which resulted in
291 subdued Tibetan plateau warming.

292 The meridional temperature gradient in the upper troposphere over the Tibetan plateau during
293 the monsoon is one of the key factors responsible for the ASM circulation (Flohn 1957; Yanai et al.,
294 1992; Meehl, 1994; Li and Yanai, 1996; Wu and Zhang, 1998). Flohn (1957, 1960) suggested that
295 warming over the Tibetan plateau leads to a reversal of the meridional temperature gradient triggering
296 large-scale change in the general circulation over Asia. The warm ascending air above the Tibetan
297 plateau gradually spreads southward and descends over the northern Indian Ocean. The south-westerly
298 winds at the surface on the other hand complete the Hadley cell. This local circulation system releases
299 latent heat and further maintains the Tibetan high warm core. Thus heating over the Tibetan plateau
300 leads to increased Indian summer monsoon rainfall by enhancing the cross-equatorial circulation and
301 concurrently strengthening both the Somali Jet and the westerly winds that bring rainfall to India
302 (Rajagopalan and Molnar, 2013, Vinoj et al., 2014). Goswami et al., (1999) also reported that there is a
303 strong correlation between Hadley circulation and monsoon precipitation.

304 Figures 8(a)-(c) depict the monsoon Hadley cell circulation (averaged over 70°E-110°E)
305 obtained from the Ind38 Ind73 and Chin73 simulations. The Ind38 and Chin 73 simulations show
306 strengthening of the Hadley circulation; a strong ascending branch of the Hadley cell around 20°N
307 (Figure 8(a)), whereas it is reversed in Ind73 simulation (Figure 8(b)). Consequently, precipitation
308 anomalies over the Indian region are positive (0.3 – 0.9 mm/day) in Ind38 and Chin73 simulations
309 (figure 8(d)) whereas they are negative in Ind73 simulation (-0.3 – -0.6 mm/day) (Figure 9(e)). Thus
310 enhanced Indian and Chinese NO_x emissions increases warming over the Tibetan plateau by ozone
311 heating and enhances precipitation over India via strengthening Hadley circulation. Whereas doubling



312 of NO_x emissions over India produced high amount of ozone over the Indo Gangetic plain. Related
313 ozone heating in the lower troposphere reverses the monsoon Hadley circulation and thereby resulted in
314 negative precipitation anomalies.

315

316 5. Conclusions

317 In this paper we investigate the potential impacts of enhanced anthropogenic NO_x emissions on
318 ozone production and distribution during the monsoon season using the state-of-the-art ECHAM5-
319 HAMMOZ model simulations. We performed sensitivity experiments for anthropogenic NO_x
320 enhancement, 38% over India (Ind38 simulation) and 73% over China (Chin73 simulation) in
321 accordance with observed trends 3.8% per year over India and 7.3% per year over China (Ghude et al.,
322 2013; Schneider and van der A, 2012). In another experiment, anthropogenic NO_x emissions over India
323 are doubled (73%), equal to China (Ind73 simulation).

324 These simulations show that increase in anthropogenic NO_x emissions (over India and China)
325 increases ozone production in the lower and mid-troposphere. The monsoon convection at the southern
326 flank of the Himalayas (80-90°E) and over the Bay of Bengal lifts up the NO_x and ozone from India
327 across tropopause into the lower stratosphere. Cross tropopause transport also occurs over China due to
328 convection over the South China Sea. These air masses are then transported deeper into the stratosphere
329 via the tropical pipe.

330 Increase in NO_x emissions in Ind38 and Chin73 simulations increases ozone radiative forcing
331 over India by 0.112 W/m² and 0.121 W/m² respectively. Enhanced ozone production (Ind38 and
332 Chin73 simulations) increases ozone heating rates which cause anomalous warming over the Tibetan
333 plateau. Doubling of NO_x emissions over the India region (Ind73 simulation) produces anomalous
334 heating near surface over the Indo Gangetic plain ($\sim 3 \times 10^{-4}$ K/day) due to high amount of ozone



335 production (~40-90%). This elicited subsidence over central India (via reversal of monsoon Hadley
336 circulation) and resisted vertical transport of ozone which resulted in subdued Tibetan plateau warming.

337 In Ind38 and Chin73 simulations, anomalous warming over the Tibetan plateau resulted in
338 strengthening of the monsoon Hadley circulation over India and elicited positive precipitation (0.3 –
339 0.9 mm/day) anomalies over India. However, doubling of NO_x emissions over India (Ind73) induced
340 reverse Hadley circulation due to anomalous heating, in the lower troposphere over the Indo Gangetic
341 plain. The descending branch of the Hadley circulation resisted the cross tropopause transport of ozone
342 and NO_x. The reversal of the Hadley circulation and concurrent subdued warming over the Tibetan
343 plateau resulted in negative precipitation anomalies (-0.3 – -0.6 mm/day) over India.

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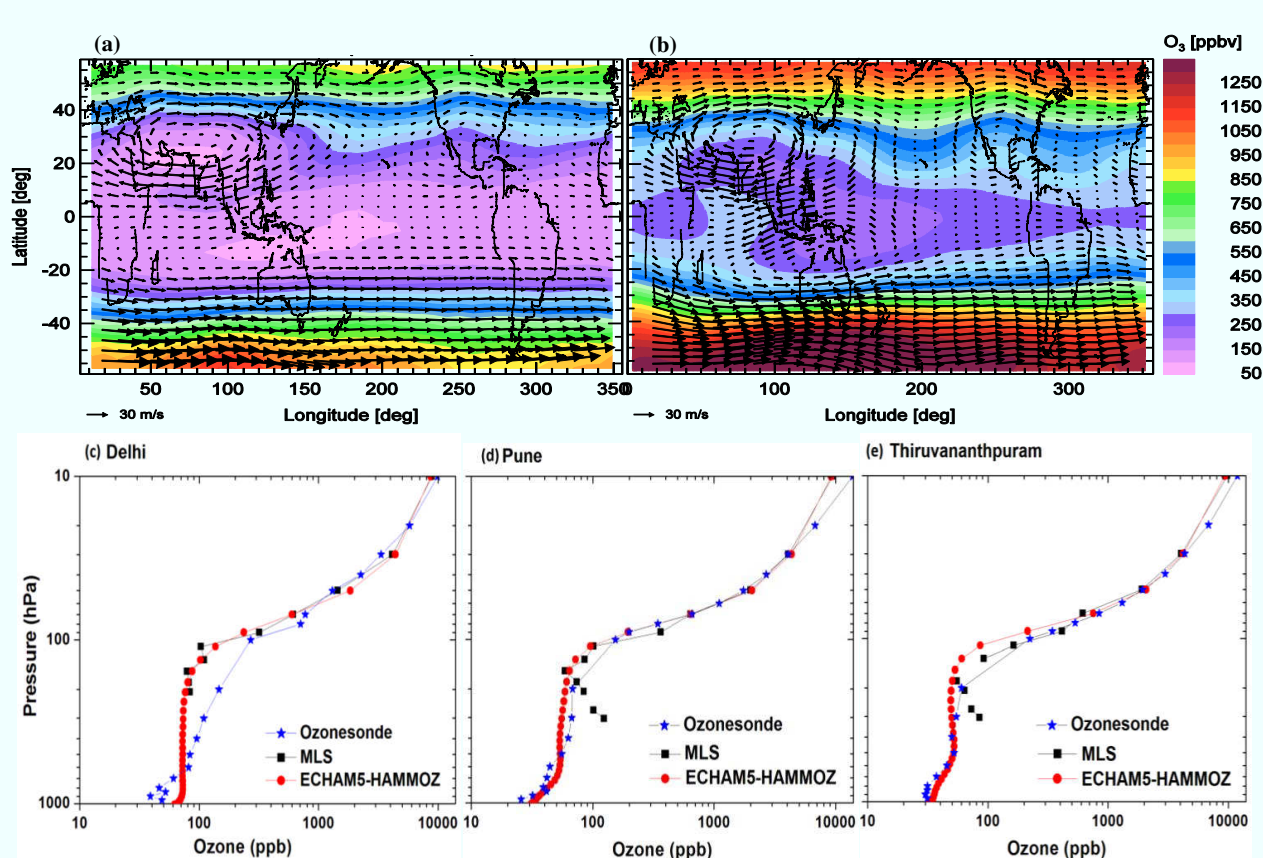


Figure 1: Distribution of ozone mixing ratio (ppbv) during the monsoon season (June-September) obtained from (a) MLS observations at 100 hPa. Black arrows indicate wind vectors from ERA-Interim. Show the same but obtained from (b) ECHAM5-HAMMOZ CTRL simulation at 90 hPa. Black arrows indicate simulated wind vectors. ECHAM5-HAMMOZ simulation is smoothed with averaging kernel of MLS. Vertical distribution of seasonal mean (June-September) ozone mixing ratios (ppb) as obtained from ozonesonde (2001-2009), MLS (2004-2013) and ECHAM5-HAMMOZ CTRL simulation at Indian stations, (c) Delhi (d) Pune (e) Thiruvananthapuram.

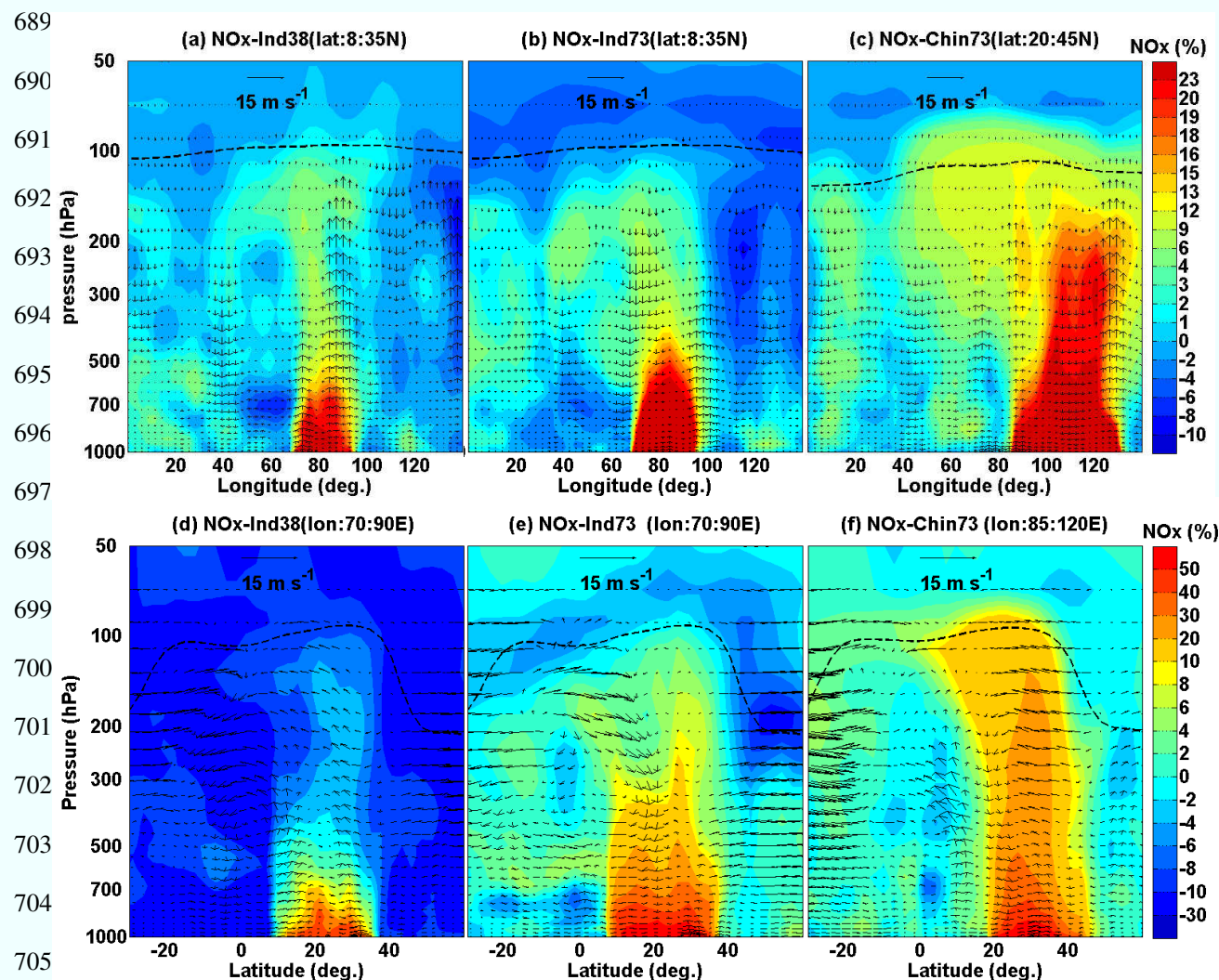


Figure 2: Longitude pressure cross-section of NO_x anomalies (%) averaged for the monsoon season (June-September) obtained from (a) Ind38 (averaged over 8°N-35°N), (b) Ind73 (averaged over 8°N-35°N), (c) Chin73 (averaged over 20°N-45°N) simulations. Latitude pressure cross-section of NO_x anomalies (%) averaged for the monsoon season (June-September) obtained from (d) Ind38 (averaged over 70°E-90°E), (e) Ind73 (averaged over 70°E-90°E), (f) Chin73 (averaged over 85°E-120°E) simulations. Black arrows indicate wind vectors. The vertical velocity field has been scaled by 300. The dashed line represents the tropopause.

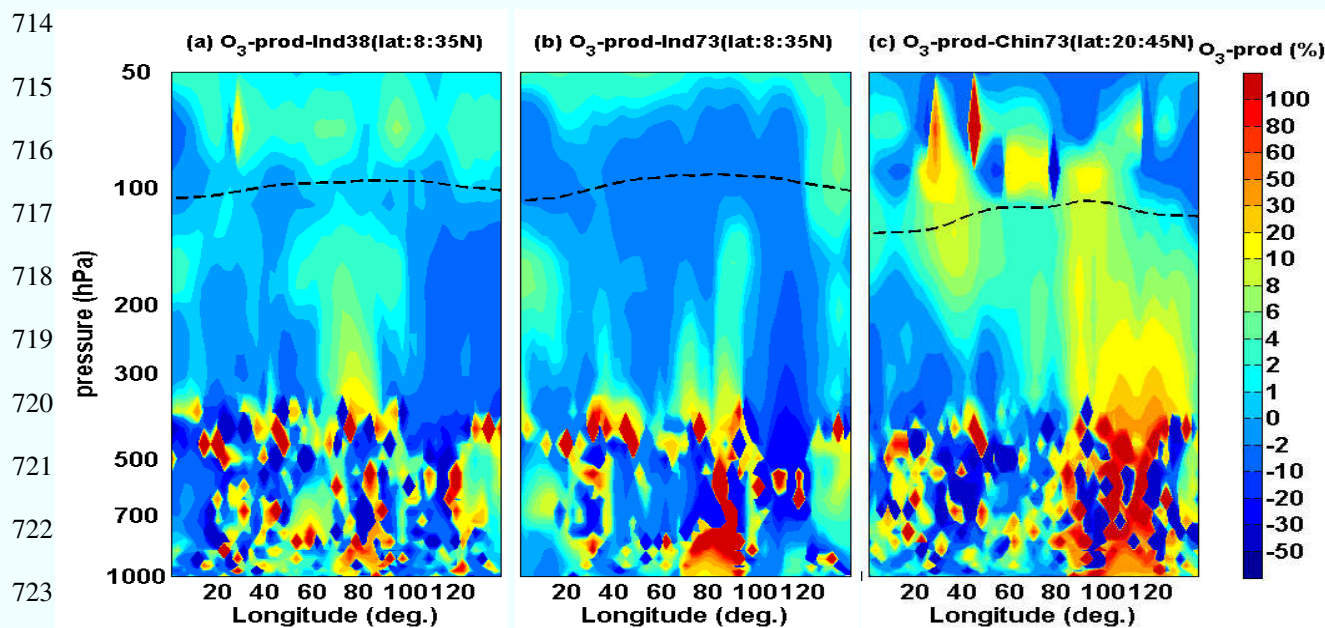


Figure 3: Longitude pressure cross-section of changes in net ozone production (%) (due to enhanced NO_x with respect to CTRL simulation) averaged for the monsoon season (June-September) obtained from (a) Ind38 (averaged over 8°N-35°N), (b) Ind73 (averaged over 8°N-35°N), (c) Chin73 (over 20°N-45°N) simulations. The dashed line represents the tropopause.

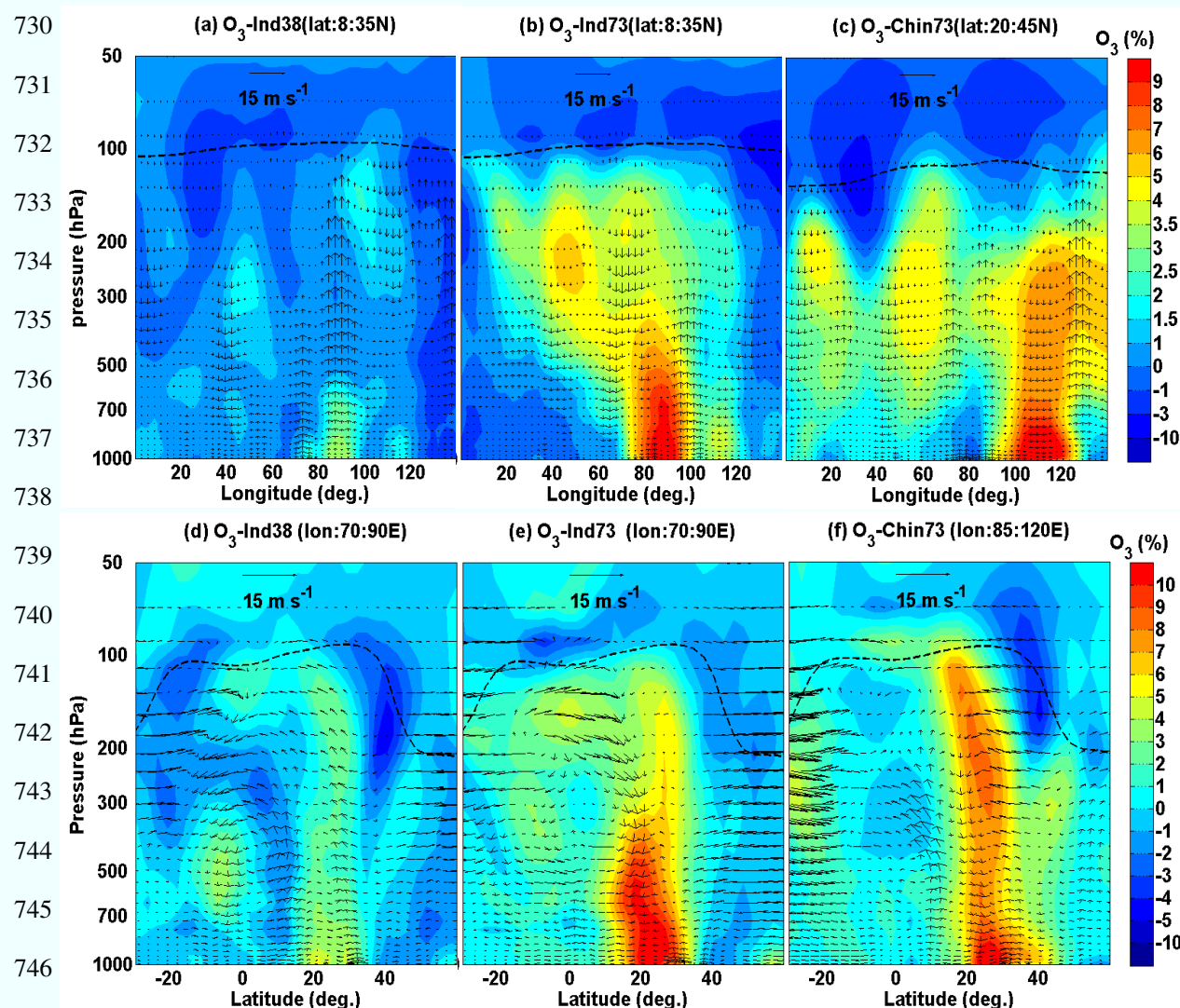


Figure 4: Longitude pressure cross-section of ozone anomalies (%) averaged for the monsoon season (June-September) obtained from (a) Ind38 (averaged over 8°N-35°N), (b) Ind73 (averaged over 8°N-35°N), (c) Chin73 (averaged over 20°N-45°N) simulations. Latitude pressure cross-section of ozone anomalies (%) averaged for the monsoon season (June-September) obtained from (d) Ind38 (averaged over 70°E-90°E), (e) Ind73 (averaged over 70°E-90°E), (f) Chin73 (averaged over 85°E-120°E) simulations. Black arrows indicate wind vectors. The vertical velocity field has been scaled by 300. The dashed line represents the tropopause.

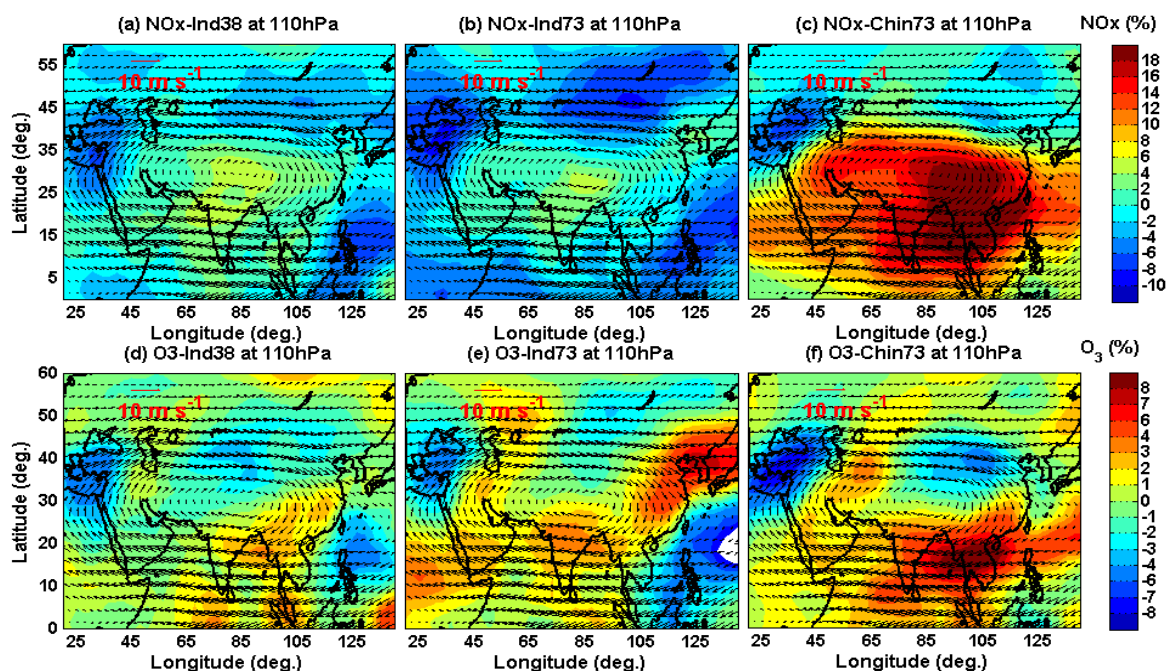


Figure 5: Latitude-longitude cross-section of NO_x anomalies (%) averaged for the monsoon season (June-September) at 110 hPa obtained from (a) Ind38 (b) Ind73 (c) Chin73 simulations. (d-f) Show the same but for ozone (%) at 110 hPa for (d) Ind38 (e) Ind73 (f) Chin73 simulations. Black arrows indicate horizontal winds at 110 hPa.

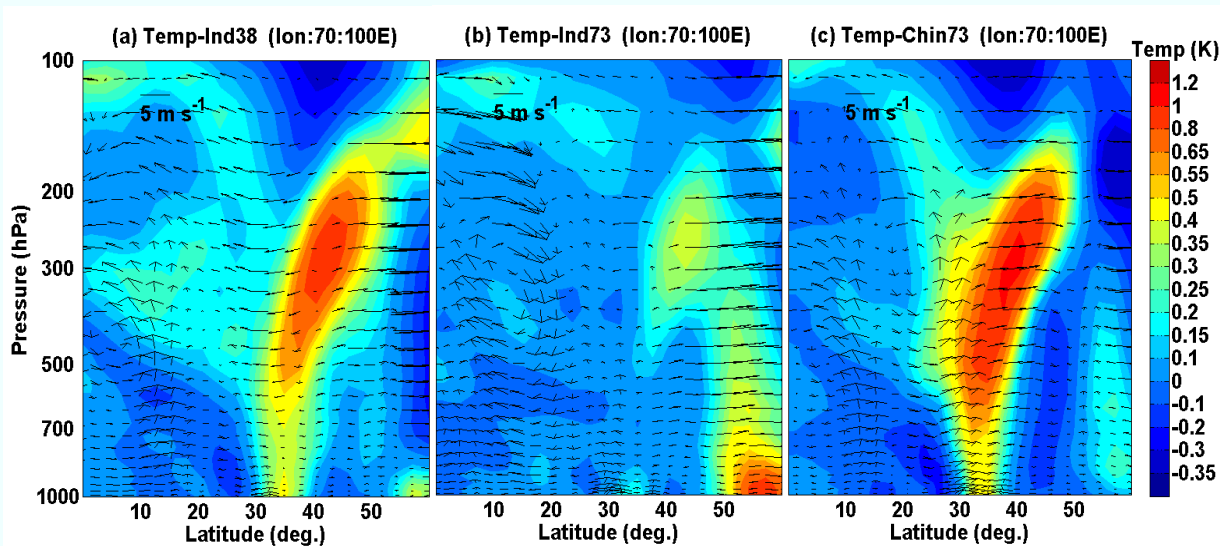


Figure 6: Latitude pressure cross-section of temperature anomalies (K) averaged for the monsoon season (June-September) obtained from (a) Ind38 (averaged over 70°E-94°E) (b) Ind73 (averaged over 70°E-94°E) and (c) Chin73 (averaged over 70°E-100°E) simulations. Black arrows indicate wind vectors. The vertical velocity field has been scaled by 300.

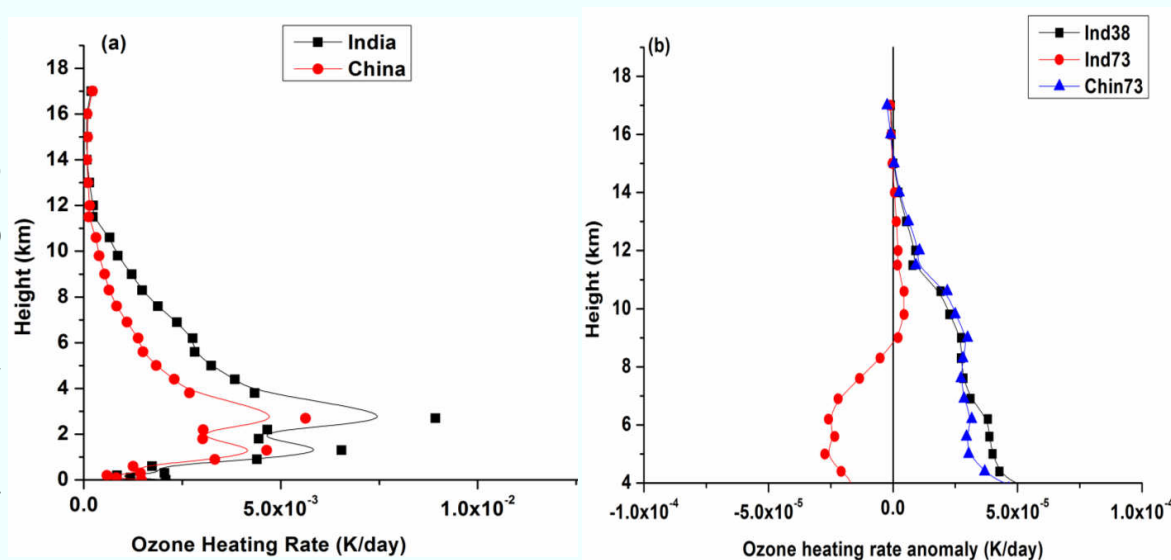


Figure 7: Vertical profile of (a) ozone heating rate (K/day) obtained from CTRL simulations averaged over India (70° - 94° E, 20° - 30° N) and China (70° - 100° E, 30° - 40° N), (b) ozone heating rate anomaly(K/day) averaged over Tibetan Plateau (70° - 100° E, 20° - 30° N) obtained from Ind38, Ind73 and Chin73 simulations.

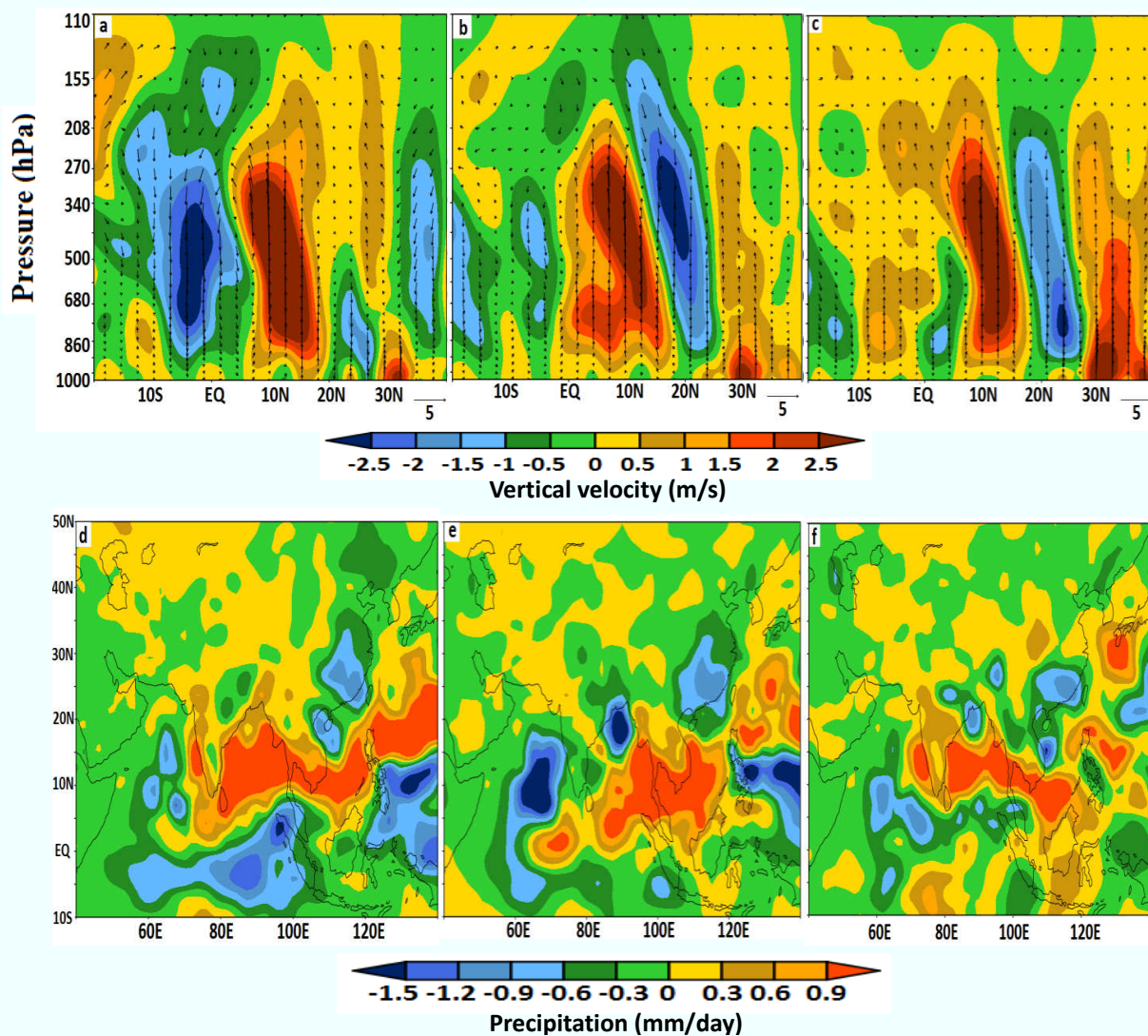


Figure 8: Change in the meridional circulation due to enhanced NO_x emissions averaged for the monsoon season (June-September) and over 70°E - 110°E for (a) Ind38 (b) Ind73 (c) Chin73 simulations. Shaded contours indicate the anomalies in vertical velocity (m/s). The vertical velocity field has been scaled by 300. Precipitation anomalies (mm/day) averaged for the monsoon season (June-September) obtained from (d) India38 (e) Ind73 (f) Chin73 simulations with respect to CTRL.