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:

The Asian summer monsoon (ASM) anticyclone is the most pronounced circulation pattern in the Upper Troposphere and Lower Stratosphere (UTLS) during the Northern Hemisphere summer. Asian summer monsoon convection plays an important role in efficient vertical transport from the surface to the upper-level anticyclone. In this paper we investigate the potential impact of enhanced anthropogenic nitrogen oxides (NO_x) on the distribution of ozone in the UTLS using the fully-coupled aerosol chemistry climate model, ECHAM5-HAMMOZ. Ozone in the UTLS is influenced both by the convective uplift of ozone precursors and by the uplift of enhanced NO_x induced tropospheric ozone anomalies. We performed anthropogenic NO_x emission sensitivity experiments over India and China. In these simulations, covering the years 2000-2010 anthropogenic NO_x emissions have been increased by 38% over India and by 73% over China with respect to the emission base year 2000. These emission increases are comparable to the observed linear trends of 3.8 % per year over India and 7.3% per year over China during the period 2000 to 2010. Enhanced NO_x emissions over India by 38 % and China by 73 % increase the ozone radiative forcing in the ASM Anticyclone (15º-40°N, 60º-120°E) by 16.3 mW m^{-2} and 78.5 mW m^{-2} respectively. These elevated NO_x emissions produce significant warming over the Tibetan Plateau and increase precipitation over India due to a strengthening of the monsoon Hadley

circulation.

However increase in NO_x emissions over India by 73% (similar to the observed increase over China), results in large amount of ozone production over the Indo Gangetic plain and Tibetan Plateau. The higher ozone concentrations, in turn, induce a reversed monsoon Hadley circulation and negative precipitation anomalies over India. The associated subsidence suppresses vertical transport of NO_x and ozone into the ASM anticyclone.

Key words: Asian summer monsoon, Tropospheric ozone, Tropospheric NO_x, NO_x transport, Upper troposphere and lower stratosphere, Ozone radiative forcing.

1. Introduction

Rapid economic development and urbanization in Asia has resulted in an unprecedented growth in anthropogenic emissions of nitrogen oxides (NO_x), carbon monoxide (CO), carbon dioxide (CO_2), methane (CH_4). Many of these species affect concentrations of tropospheric ozone, which is both an important polluting agent and a greenhouse gas (Wild and Akimoto, 2001; Chatani et al 2014; Revell et al., 2015). Ground based and satellite observations show a large amount of these ozone precursors concentrated over India and China (Sinha et al., 2014; Richter et al., 2005; Jacob et al., 1999; Zhao et al., 2013; Gu et al., 2014). Studies show that tropospheric ozone production over Asia is controlled by the abundance of NO_x and VOCs (Sillman, 1995, Lei et al., 2004, Zhang et al., 2004 and Tie et al., 2007), with large regions such as India and China being NO_x limited regions. Therefore, increased NO_x in these regions leads to an increase in ozone concentrations (Yamaji et al., 2006; Sinha et al., 2014; Fadnavis et al., 2014). Recently, positive trends in Asian tropospheric column NO_2 have been reported, i.e. 3.8 % yr^{-1} over India, using SCanning Imaging Absorption SpectroMeter for Atmospheric CHartographY (SCIAMACHY) observations for the period 2003-2011 (Ghude et al., 2013), and 7.3% yr^{-1} over China using Ozone Monitoring Instrument (OMI) observations for the period 2002-2011

(Schneider and van der A., 2012). Lightning contributes to the production of NOx in the middle and upper troposphere (Barret et al, 2016). Over the Asian region, lightning contributes ~40% to NO_x and 20% to ozone production in the middle and upper troposphere during the monsoon season (Tie et al. 2001; Fadnavis et al. 2015). The upper tropospheric ozone concentration is determined by in-situ production from both lightning and ozone precursors which are transported from the boundary layer (Sǿvde et al., 2011; Barret et al, 2016).

Tropospheric ozone has a warming effect on climate, its estimated radiative forcing due to increased concentrations since pre-industrial times being 0.4 W m^{-2}, with a 5 to 95% confidence range of (0.2 to 0.6 W m^{-2}) (Stevenson et al., 2013; Myhre et al., 2013). Previous studies highlighted the importance of the tropical tropopause region for ozone radiative forcing (Lacis et al, 1990; Riese et al., 2012; Rap et al., 2015) and showed that ozone perturbations exert a large influence on the thermal structure of the atmosphere (e.g., Thuburn and Craig, 2002; Foster and Shine 1997). A recent study based on Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) models reported that NO_x and CH_4 are the greatest contributors in determining tropospheric 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, 2016). Satellite observations show the confinement of a number of chemical constituents like water vapor (H_2O), CO, CH_4, ethane, hydrogen cyanide (HCN), PAN and aerosols, within the ASM anticyclone (Park et al., 2004, 2007, 2008; Li et al., 2005; Randel and Park, 2006; Xiong et al., 2009; Randel et al. 2010; Lawrence et al., 2011; Abad et al., 2011; Fadnavis et al., 2013;2014;2015; Barret et al., 2016) which has potential implications on stratospheric chemistry and dynamics. Thus 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 upward transport of ozone poor air masses (Gettelman et al., 2004; Park et al., 2007; Kunze et al., 2010). Observations show that convectively lifted air masses arriving in the anticyclone are ozone poor but rich in ozone precursors. Balloon sonde observations show that ozone variations near the anticyclone are strongly correlated with temperature near the tropopause (Tobo et al., 2008). Thus the linkage of low ozone and high concentrations of ozone precursors with the temperature variation in the anticyclone is an open question.

In this study we ask the question 'how do increasing Asian NO_X emissions and the associated ozone production affect ozone radiative forcing and monsoon circulation?'. We perform sensitivity experiments of increased anthropogenic NO_x emissions 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). We estimate the ozone radiative forcing for the different anthropogenic NO_x emission scenarios, together with associated changes in temperature and the monsoon circulation. The paper is organized as follows: in Section 2 the data and model set up are described;the results are summarized in Section 3 and discussed in Section 4, followed by conclusions given in Section 5.

2. Data description and Model setup

2.1 Satellite measurements

Earth Observing System (EOS) microwave limb sounder (MLS) is one of the four instruments on the NASA's EOS Aura satellite flying 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 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 about 3 km. MLS vertical profiles of ozone show good agreements 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 interpolated to potential temperature levels and gridded horizontally, within latitude bins of equal area (with the equatorial bin of 150km width) and longitude bins of about 8.5 degrees. This data can be accessed from http://mls.jpl.nasa.gov/. For comparison, simulated ozone is convolved with the MLS averaging kernel (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, MOZART2 (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 $O(^1D)$ quenching reaction rates used are taken from Sander et al., (2003) and isoprene nitrates chemistry taken from 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). Interactive calculation of cloud droplet number concentration is according to Lohmann et al (1999) and ice crystal number concentrations are according to Kärcher and Lohmann (2002). The convection scheme is based on the mass flux scheme developed by Tiedke (1989).

The model is run at a T42 spectral resolution corresponding to about 2.8°×2.8° in the horizontal dimension and 31 vertical hybrid σ – p levels from the surface to 10 hPa. In our model simulations, emissions from anthropogenic sources and biomass burning are from the year 2000 RETRO project data set (available at http://eccad.sedoo.fr/) (Schultz et al., 2004; 2005; 2007; 2008). Emissions of SO_2, BC and OC are based on the AEROCOM-II emission inventory, also for the year 2000 (Dentener et al., 2006). The distribution of NO_x emission mass flux (kg m^{-2} s^{-1}) averaged for the Asian summer monsoon season (June–September) is shown in Supplementary Fig. S1. It shows high values over the Indo Gangetic Plains and East China. Other details of model parameterizations, emissions and evaluation are described by Fadnavis et al. (2013; 2014; 2015) and Pozzoli et al. (2008a, b; 2011). Each of our model experiments consists of continuous simulations for eleven years from 2000 to 2010. The base year for emissions is taken as 2000 and emissions were repeated every year throughout the simulation period. Meteorology varied due to varying monthly mean sea surface temperature (SST) and sea ice concentration (SIC). The AMIP2 SSTs and SIC varying for the period 2000 – 2010 were specified as a lower boundary condition.

In order to understand the impact of enhanced anthropogenic NO_x emissions on the distribution of ozone in the UTLS, sensitivity simulations were performed for the period 2000 – 2010. The experimental set up is the same as described by Fadnavis et al., (2015). The four simulations analyzed in this study are: (1) a reference experiment (CTRL) and three sensitivity experiments (referred to as experiments 2 - 4), where the anthropogenic NO_x emissions over India and China are scaled in accordance with the observed trends. In experiment (2), anthropogenic NO_x emissions are increased over India by 38% (Ind38), in experiment (3) increases over China by 73% (Chin73) are prescribed. In order to analyze the effects of similar NO_x percentage increases over India and China, NO_x emissions are increased over India by 73% (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. All four simulations use the same VOC and CO emissions and they all include NO_x production due to lightning (lightning-on) and soil emissions (see Table 1, showing details pertaining to these experiments). Therefore NO_x or ozone anomalies obtained from difference between Ind38, Ind73 and Chin73 with respective to CTRL simulation do not have an impact of lightning or soil emissions as they are same in all the simulations.

In addition, a series of four lightning-off simulations were performed for the same period and boundary conditions as experiments 1-4 (these simulations are the same as the ones documented by Fadnavis et al. (2015))The impact of lightning on NO_x production is estimated by comparing the CTRL (lightning-on) simulation with lightning-off simulations.

The accuracy of the simulation of the monsoon circulation will likely depend on the model resolution and increased vertical resolution may improve the model performance (Druyan et al., 2008; Abhik et al., 2014). While we acknowledge the limitations of our relatively course vertical resolution (dictated by our computational resources), the model is still capable of reasonably simulating the general regional spatial pattern of precipitation and low-level circulation (Rajeevan et al., 2005) (see Supplementary Fig. S2, showing simulated seasonal mean precipitation and circulation at 850 hPa in the CTRL simulation).

The heating rates and radiative forcings associated with the ozone changes in our three sensitivity simulations are calculated using the Edwards and Slingo (1996) radiative transfer model and the fixed dynamical heating approximation for stratospheric temperature adjustment. Similarly to previous studies (Riese et al., 2012; Bekki et al., 2013; Rap et al., 2015), we used the off-line version of the model, with six shortwave and nine longwave bands, and a delta-Eddington 2-stream scattering solver at all wavelengths.

3. Results

3.1 Comparison with MLS satellite measurements in the UTLS

The spatial distributions of ozone mixing ratios from MLS observations at 100 hPa and from the CTRL ECHAM5-HAMMOZ simulation at 90 hPa (the nearest model level) after smoothing with the averaging kernel of MLS are illustrated in Fig. 1a and Fig. 1b, respectively. The climatological horizontal winds plotted in the figure clearly show the anticyclonic upper level monsoon circulation. Recent attempts to characterize the extent of the anticyclone are based either on potential vorticity on isentropic surfaces or geopotential height on pressure surfaces. Here we apply both characterizations of the anticyclone and show the PV contour related to the maximum PV gradient on 380K (calculated from ERA-Interim reanalysis following Ploeger et al., 2015), and the 270m geopotential height anomaly as proposed by Barret et al. (2016). The close agreement of both methods shows that from a climatological point of view the two criteria yield a very similar picture of the anticyclonic circulation and the related trace gas confinement. Locally and at particular dates, however, differences may be larger with potential vorticity correlating better with confined trace gas anomalies than geopotential height (e.g., Garny and Randel, 2013; Ploeger et al., 2015). The spatial pattern of low ozone concentrations in the monsoon anticyclone is well simulated in the model. It is in good agreement with MLS (90-140 ppbv), MIPAS (80-120 ppbv) and SAGE II (<150ppbv) measurements (Kunze et al., 2010; Randel et al., 2001; Randel and Park 2006; Park et al., 2007).

Vertical profiles of ozonesonde measurements (averaged for the monsoon season during 2001-2009) at Indian stations, Delhi (28.61ºN, 77.23ºE), Pune (18.52ºN, 73.85ºE) and Thiruvananthapuram

(8.48ºN, 76.95E) are compared with MLS measurements and ECHAM5-HAMMOZ simulated ozone mixing ratios in Figs. 1(c)-(e). ECHAM5-HAMMOZ simulations show good agreement with MLS data between 200 hPa and 50 hPa at all three stations. Comparison of ozonesonde observations with the ECHAM5-HAMMOZ simulation shows reasonably good agreement at Pune, compared to Delhi and Thiruvananthapuram where there are some discrepancies. The simulated ozone mixing ratios are lower than ozonesonde measurements by 10-40 ppb between 500 – 90 hPa at Pune and by ~70-90 ppb in the upper troposphere (500-150 hPa) at Delhi. At Thiruvananthapuram, while at altitudes below 375 hPa, simulated ozone mixing ratios show good agreement with ozonesonde data, at the altitudes above 375 hPa, simulated values are lower than observations by ~20-70 ppb. The differences between model and ozonesonde data may be due to different grid sizes: the ECHAM5-HAMMOZ model grid size is ~280 km, while balloon observations are within ~30-180 km spatial range (balloon typically drifts ~30–180 km horizontally). In addition, previous work comparing these model simulations with various aircraft observations during the monsoon season, found a reasonable agreement for PAN, NO_x, HNO_3 and O_3 mixing ratios (Fadnavis et al., 2015).

3.2 Transport of enhanced NO_x emissions into the UTLS

Recent satellite observations and model simulations quantified the impact of convective transport of boundary layer pollution into the ASM anticyclone during the Asian summer monsoon season (Gettelman et al., 2004; Randel et al., 2010; Fadnavis et al., 2013, 2014, 2015). These pollutants are further transported across the tropopause as evident in satellite observations of, e.g. water vapour (Bian, 2012), hydrogen cyanide (HCN) (Randel, 2010), CO (Schoeberl et al., 2006), Peroxyacetyl nitrate (PAN) (Fadnavis et al.,2014; 2015), aerosols (Vernier et al., 2015, Fadnavis et al., 2013). To understand the influence of monsoon convection on the vertical distribution of NO_x we show zonal and meridional cross sections over India and China. Vertical distributions of NO_x averaged for the monsoon season over Indian latitudes (8°N-35°N), and Chinese latitudes (20°N-45°N) as obtained from CTRL simulations are shown in the Supplementary Figs. S3(a) and S3(b) respectively. These figures show elevated levels of NO_x extending from the surface to the upper troposphere over India and China. The wind vectors along with the distribution of cloud droplet number concentration (CDNC) and ice crystal number concentration (ICNC), (Supplementary Figs. S4(a), S4(b) and S4(c)) indicate strong convective transport from the Bay of Bengal (BOB), South China Sea and southern slopes of Himalayas which might lift the boundary layer NO_x to the upper troposphere.

During the monsoon season, the NO_x distribution in the UTLS is also influenced by lightning, in addition to transport from anthropogenic sources. Lightning activity during this season was found to be more pronounced in Asia, compared to the other monsoon regions such as North America, South America and Africa (Ranalkar and Chaudhari, 2009; Penki and Kamra, 2013). In our simulations, we find that lightning produces 40-70% of NO_x over north India and Bay of Bengal and 40-60% over the Tibetan Plateau and West China region (Supplementary Fig. S5).

Fig. 2 shows the vertical distribution of anthropogenic NO_x anomalies obtained from the Ind38, Ind73, Chin73 simulations, compared with the CTRL simulation. Ind38 and Chin73 simulations show that the convective winds over the Bay of Bengal (80-90°E) (Figs. 2(a) and 2(c)) and at the southern flank_of the Himalayas (Figs. 2(d) and 2(f)) lift up the enhanced NO_x emissions to the upper troposphere (UT). While most transport is mainly into the UT, parts of it also occur into the lower stratosphere, with cross tropopause transport being particularly evident in the Chin73 simulation (Figs. 2(c) and 2(f)). Randel and Park (2006) and Randel et al. (2010) also reported that pollution transported by Asian monsoon convection enters the stratosphere. Our results are also in good agreement with previous studies indicating significant vertical transport due to strong monsoon convection from the southern slopes of Himalayas (Fu et al., 2006, Fadnavis et al., 2013; 2014) and the South China Sea (Park et al 2009; Chen et al., 2012). In the upper troposphere, NO_x is transported over Iran and Saudi

Arabia along the descending branch of the large scale monsoon circulation (Rodwell and Hoskins, 1995). However, the cross tropopause transport is not present in the Ind73 simulation, where it is inhibited by the wind anomalies that show a descending branch over central India (~20°N, 75°E) (Figs. 2(b) and 2(e)). These descending wind anomalies may also be related to the associated ozone radiative forcing and temperature changes, as discussed in Section 4.

3.3 Impact of enhanced anthropogenic NO_x on the tropospheric ozone distribution

We calculate the change in ozone production over India and China due to enhanced NO_x emissions in the Ind38, Ind73 and Chin73 simulations with respect to the CTRL simulation. Figure 3, showing longitude-pressure cross sections of net ozone production (ppt/day) changes, indicates that the majority of this additional ozone production occurs in the lower troposphere. At altitudes below 300 hPa, the ozone production and loss vary between -15 ppt day^{-1} and 15 ppt day^{-1}. In the upper troposphere (300-150 hPa), the estimated amount of additional net ozone production in Ind38 and Ind73 simulation is 3-7 ppt day^{-1}, while in the Chin73 simulation it is ~3-13 ppt day^{-1}. We also simulate ozone loss near the tropopause in the Ind73 simulation (Figure 3b). We note that these ozone anomalies are not driven by lightning NO_x, as this is included in all simulations. It is interesting to understand ozone production over the highly populated Indo Gangetic Plain and Tibetan Plateau region. A longitude pressure cross section over this region show that ozone production over the Indo Gangetic Plain and Tibetan Plateau in Ind73 is (20-25ppt/day) is much larger than Ind38 (6-20 ppt/day) in the lower troposphere (Supplementary Fig. S6).

Figure 4 shows the vertical distribution of ozone anomalies induced by enhanced anthropogenic NO_x emissions in the three perturbation experiments compared to the CTRL simulation, averaged over India and China. Although the air mass in the monsoon anticyclone is relatively poor in ozone (Fig.1(b)), the elevated amounts of ozone anomalies in response to enhanced anthropogenic NO_x

emissions are clearly seen in Fig. 4. This may be partially due to convective transport of enhanced-NO_x-emission induced ozone anomalies produced in the lower troposphere, and partially due to chemical ozone production from convectively transported boundary layer ozone precursors. Ozone anomalies are enhanced near 300-200 hPa over west Asia (40-60ºE) (Figs. 4a-c), possibly due to the vertical convective transport of ozone anomalies and precursors and also from subsequent horizontal transport in the monsoon anticyclone (Barret et al., 2016).

Latitude-pressure cross sections of enhanced-NO_x-emission induced ozone anomalies plotted in Figs. 4(d) and 4(f) illustrate how convection over the Bay of Bengal, the southern slopes of the Himalayas and the South China Sea lifts the enhanced ozone anomalies from India and China into the upper troposphere. These ozone anomalies are also transported further across the tropopause and into the lower stratosphere, where ozone production is also driven by photolysis and NO_x anomalies.

In the Ind73 simulation, similarly to the NO_x anomaly distribution (Figs. 2(b) and 2(e)), the descending branch of circulation over central India also suppresses the vertical transport of ozone anomalies across the tropopause (Figs. 4(b) and 4(e)). This subsidence may be related to ozone heating rate changes, as there is significant increase in ozone production over the Indo Gangetic plain and Tibetan Plateau in the lower troposphere due to enhanced anthropogenic NO_x emissions (Section 4).

3.4 Distribution of NO_x and ozone in the anticyclone

The distributions of NO_x and ozone anomalies in the monsoon anticyclone region in the Ind38, Ind73 and Chin73 simulations with respect to the CTRL simulation are shown in Figs. 5(a)-(f). A maximum in the NO_x anomalies in the ASM anticyclone (60°E to 120°E) is seen in all the simulations. NO_x anomalies are high at the eastern part of the monsoon anticyclone since convective injection into the anticyclone occurs mainly in that region (Fadnavis et al., 2013). Increase in NO_x anomalies in the Ind38 simulation is higher (Fig. 5(a)) than that in the Ind73 simulation (Figs. 5(b)), mainly due to descending motion over central India in the Ind73 simulation, as seen in the previous sections. In contrast to NO_x anomalies, ozone anomalies in Ind38 are lower than Ind73, especially in the north-eastern part of anticyclone. Satellite observations also show high ozone precursors and low ozone amounts in the anticyclone (Park et al., 2007; Barret et al., 2016). Similarly, the Chin73 simulation shows higher values of NO_x anomalies (>18%) and strong negative ozone anomalies (~-8%) in the north eastern region of the monsoon anticyclone (Figs. 5(c) and 5(f)). Figure 5 also shows that the tropical easterly jet transports NO_x and ozone (from India and China) to Saudi Arabia, Iran and Iraq.

4. Discussion

To estimate the radiative impact of the simulated ozone changes, we use the offline version of the Edwards and Slingo (1996) radiative transfer model. Figure 6 shows the radiative forcing caused by the ozone changes in each of the three sensitivity simulations compared to the CTRL simulation. The overall increase in tropospheric ozone (see Figure 4) has a warming effect on climate, with the regional average radiative forcing in the monsoon anticyclone (15ºN-40ºN, 60-120ºE) estimated at16.3 mW m^{-2}, 69.9 mW m^{-2}, and 78.5 mW m^{-2} in the Ind38, Ind73, and Chin73 simulations, respectively.

We also investigate the impact on the atmospheric heating rates caused by the ozone changes. Figure 7 shows the zonal mean heating rate anomalies for the Ind38, Ind73 and Chin73 simulations, compared to the CTRL simulation. These three simulations show positive and negative heating rates anomalies between 400-200 hPa. However, in the upper troposphere and lower stratosphere (200-50 hPa) ozone heating rates are negative over Indo Gangetic plain (20-30°N) and Tibetan Plateau (30-40ºN) region. In Ind73 simulation, ozone heating rate anomalies are positive in the lower troposphere over the Indo Gangetic plain (1000-750 hPa) and Tibetan plateau (600-400 hPa). This may be due to large amount of ozone production in the lower troposphere over these regions (Fig. S6). This heating may produce changes in the circulation leading to ascending motion over the Tibetan Plateau and a descending branch over central India (~20ºN), i.e. a reversal of monsoon Hadley circulation (Fig. 9(b)).

Figures 8 shows latitude pressure cross-section of temperature anomalies (K) obtained from Ind38, Ind73 and Chin73 simulations. Ind38 and Chin73 simulations show anomalous warming in the upper troposphere over the Tibetan Plateau while it is subdued in the Ind73 simulation. Upper tropospheric warming over the Tibetan plateau is one of the key factors responsible for the ASM circulation (Flohn 1957; Yanai et al., 1992; Meehl, 1994; Li and Yanai, 1996; Wu and Zhang, 1998). Flohn (1957, 1960) suggested that upper tropospheric warming over the Tibetan plateau leads to increased Indian summer monsoon rainfall by enhancing the cross-equatorial circulation that brings rainfall to India (Rajagopalan and Molnar, 2013, Vinoj et al., 2014). Goswami et al., (1999) also reported that there is a strong correlation between Hadley circulation and monsoon precipitation.

Figures 9(a)-(c) depict the change in monsoon Hadley cell circulation (averaged over 70ºE-100ºE) obtained from the difference in the Ind38, Ind73 and Chin73 and CTRL simulations. The Ind38 and Chin 73 simulations show a strengthening of the Hadley circulation; a strong ascending branch of the Hadley cell around 10º-20ºN (Fig. 9(a)), whereas the tilted descending branch of Hadley cell is seen over 20ºN in the Ind73 simulation (Fig. 9(b)). In Ind73 simulation ozone heating rates are positive and negative in the vertical direction near ~20ºN (Fig 7 (b)) which might have attributed tilted descending branch of Hadley cell. Consequently, precipitation anomalies over the Indian region (70º-90º E; 8º-35º N) are positive (0.3 to 0.9 mm day^{-1}) in the Ind38 and Chin73 simulations (Figs. 9(d) and 9(f)), whereas they are negative in the Ind73 simulation (-0.3 to -0.6 mm day^{-1}) (Fig. 9(e)). In the upper troposphere (250 hPa-100 hPa), Ind73 simulation shows subsidence while Chin73 simulation shows ascending motion at these levels over the Indian region. Upper tropospheric subsidence in Ind73 simulation might have contributed to the weak positive and negative precipitation anomalies over the North Indian region (Fig. 9(e)). The Chin73 simulation shows subsidence near 22°N below 200 hPa and ascending motion above it. The Chin73 simulation shows ascending motion near 12ºN rising up to 110 hPa, which leads to positive precipitation anomalies over the Indian peninsula.

Thus, enhanced Indian (Ind38) and Chinese (Chin73) NO_x emissions increase warming over the Tibetan plateau and enhance precipitation over India via a strengthening of the monsoon Hadley circulation. Remarkably, a further increase of NO_x emissions over India (Ind73) leads to high amounts of ozone in the lower troposphere over the Indo Gangetic plain and Tibetan Plateau. The related ozone heating induces a reversal of the monsoon Hadley circulation, thereby resulting in negative precipitation anomalies.

5. Conclusions

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

These simulations show that an increase in anthropogenic NO_x emissions (over India and China) increases ozone production in the lower and mid-troposphere. The monsoon convection at the southern flank of the Himalayas (80-90°E) and over the Bay of Bengal lifts up the NO_x and ozone anomalies from India across the tropopause into the lower stratosphere. Cross tropopause transport also occurs over China due to convection over the South China Sea.

Increase in NO_x emissions in the Ind38, Ind73 and Chin73 simulations leads to increase in ozone radiative forcings, in the anticyclone (15ºN-40ºN, 60ºE-120ºE) of 16.25 mWm^{-2}, 69.88 $mW\ m^{-2}$, and 78.51 $mW\ m^{-2}$ in the Ind38, Ind73, and Chin73 simulations, respectively. Enhanced ozone production (Ind38 and Chin73 simulations) increases ozone heating rates which cause anomalous warming over the Tibetan plateau. Further increase in NO_x emissions over the India region (Ind73 simulation) produces anomalous heating in the lower troposphere over the Indo Gangetic Plain and Tibetan Plateau. This warming elicits the reversal of the monsoon Hadley cell circulation. The descending branch of the monsoon Hadley circulation over the central India impedes vertical transport of ozone and NO_x anomalies.

In the Ind38 and Chin73 simulations, anomalous warming over the Tibetan plateau results in a strengthening of the monsoon Hadley circulation over India and elicits positive precipitation (0.3 to 0.9 mm day^{-1}) anomalies over India. However, in Ind73 simulations the reversal of the Hadley circulation and the concurrent subdued warming in the upper troposphere over the Tibetan plateau results in negative precipitation anomalies (-0.3 to -0.6 mm day^{-1}) over India.

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Table 1: Details of the sensitivity experiments (2000 - 2010).

Name of experiment	**Prescribed SSTs**	**Emissions**
CTRL	AMIP2 SST and SIC varying from 2000 – 2010	RETRO anthropogenic NO_x emissions for the year 2000.
Ind38	AMIP2 SST and SIC varying from 2000 – 2010	RETRO anthropogenic NO_x emissions for the year 2000 are increased by 38% over India for 11 years period 2000-2010
Chin73	AMIP2 SST and SIC varying from 2000 – 2010	RETRO anthropogenic NO_x emissions for the year 2000 are increased by 73% over China for 10 years period 2000-2010.
Ind73	AMIP2 SST and SIC varying from 2000 – 2010	RETRO anthropogenic NO_x emissions for the year 2000 are increased by 73% over India for 10 years period 2000-2010.

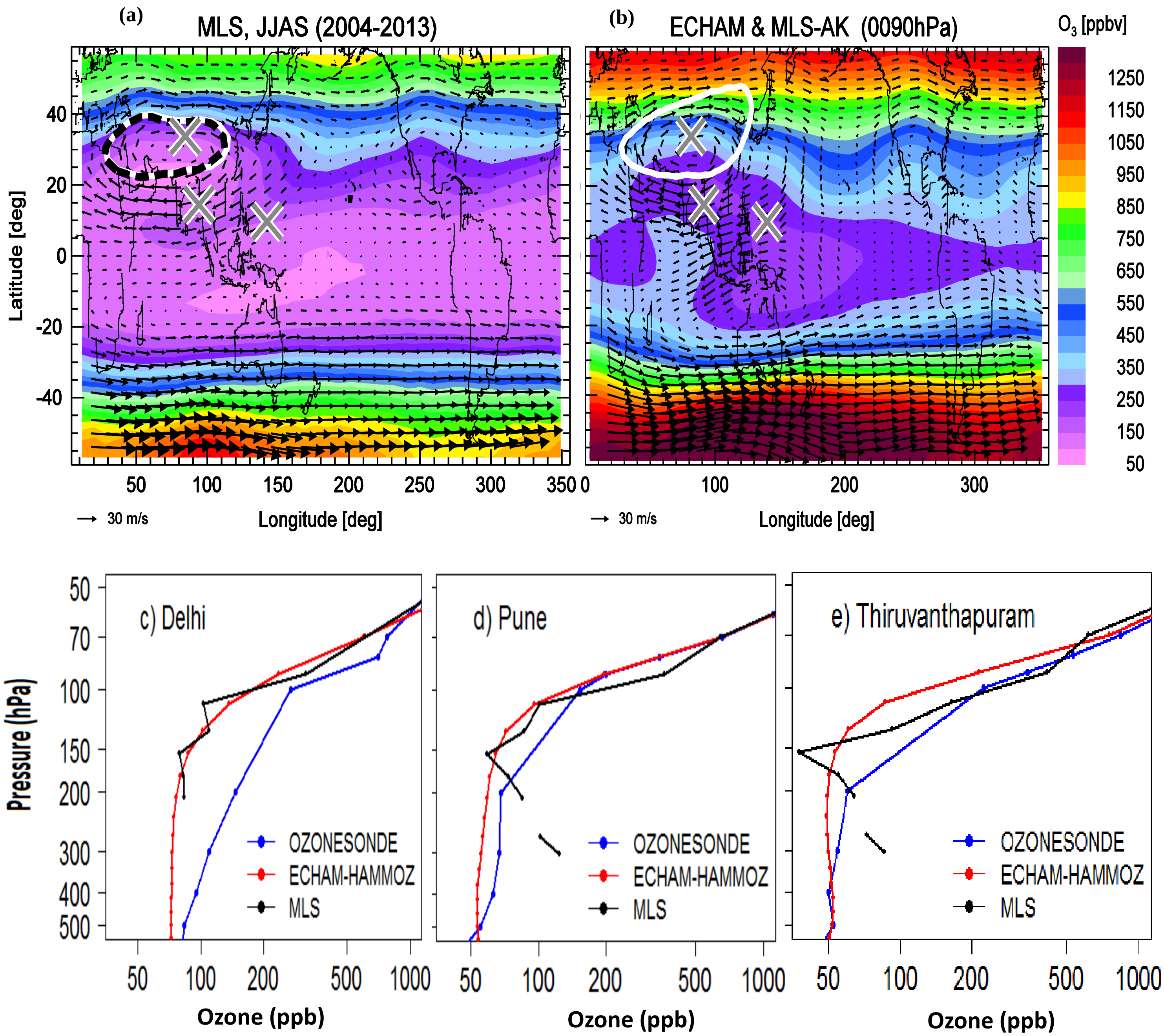

Figure 1: Distribution of ozone mixing ratio (ppb) during the monsoon season (June-September) obtained from (a) MLS observations at 100 hPa, and (b) from ECHAM-HAMMOZ at 90hPa. Black arrows indicate wind vectors, the black dashed contour shows the PV-gradient based transport barrier of the anticyclone (calculated following Ploeger et al., 2015), and the white contour shows the 270m geopotential height anomaly, corresponding to the anticyclone edge definition by Barret et al. (2016) (Meteorological data shows climatological July fields from ERA-Interim reanalysis (a) ERA-Interim

reanalysis and (b) ECHAM5-HAMMOZ. The ECHAM5-HAMMOZ ozone distribution is smoothed using the MLS averaging kernel. Grey crosses highlight the regions of the Tibetan plateau, Bay of Bengal and South China Sea. Bottom panels show the vertical distribution of seasonal (June-September) mean ozone mixing ratios (ppb) from ozonesonde (2001-2009), MLS (2004-2013) and ECHAM5-HAMMOZ CTRL simulation at the (c) Delhi, (d) Pune, and (e) Thiruvananthpuram Indian stations.

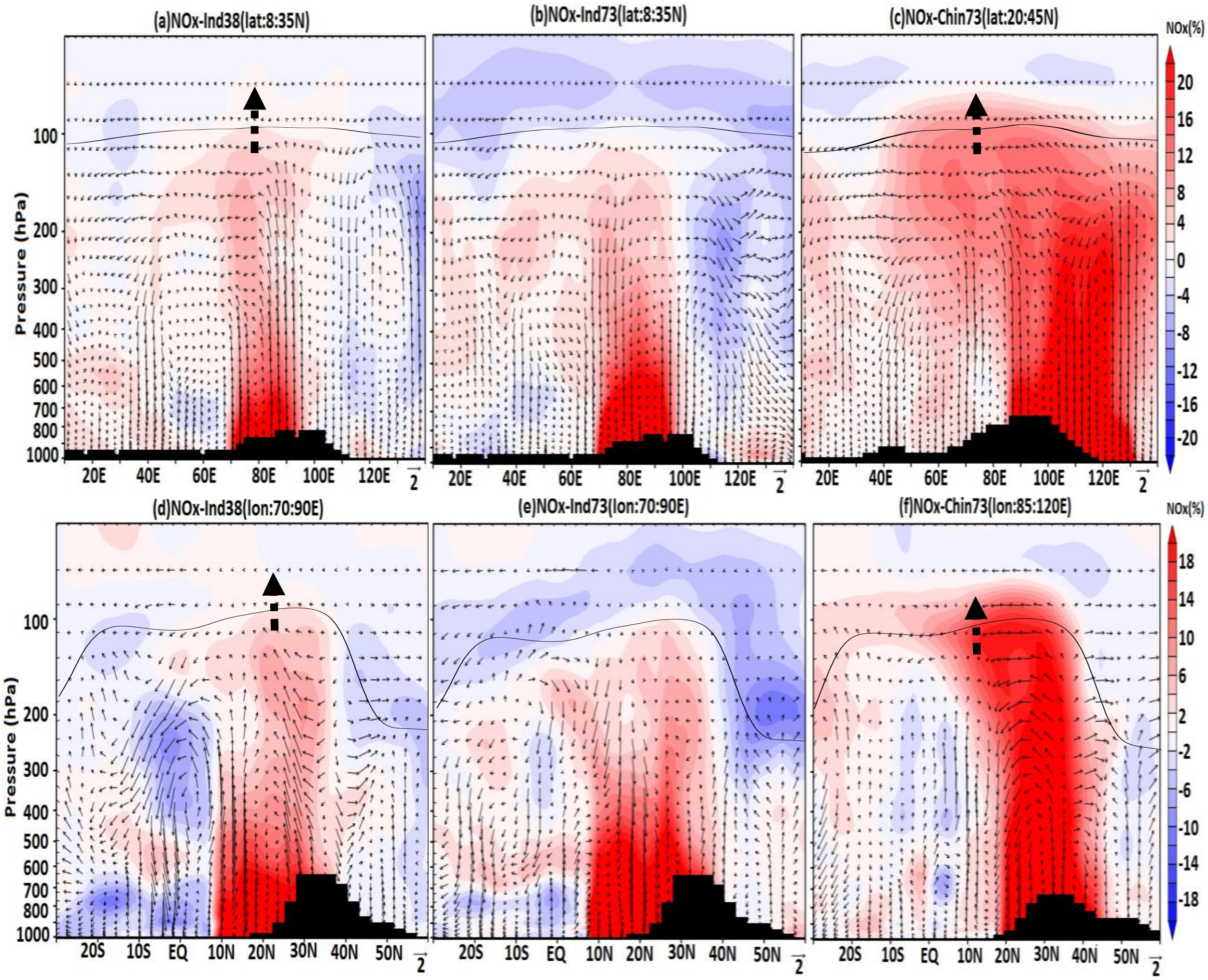

Figure 2: Longitude pressure cross-sections of percentage NO_x anomalies averaged for the monsoon season (June-September) obtained from (a) Ind38 (averaged over 8ºN-35ºN), (b) Ind73 (averaged over 8ºN-35ºN), and (c) Chin73 (averaged over 20ºN-45ºN) simulations. Latitude pressure cross-sections of percentage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

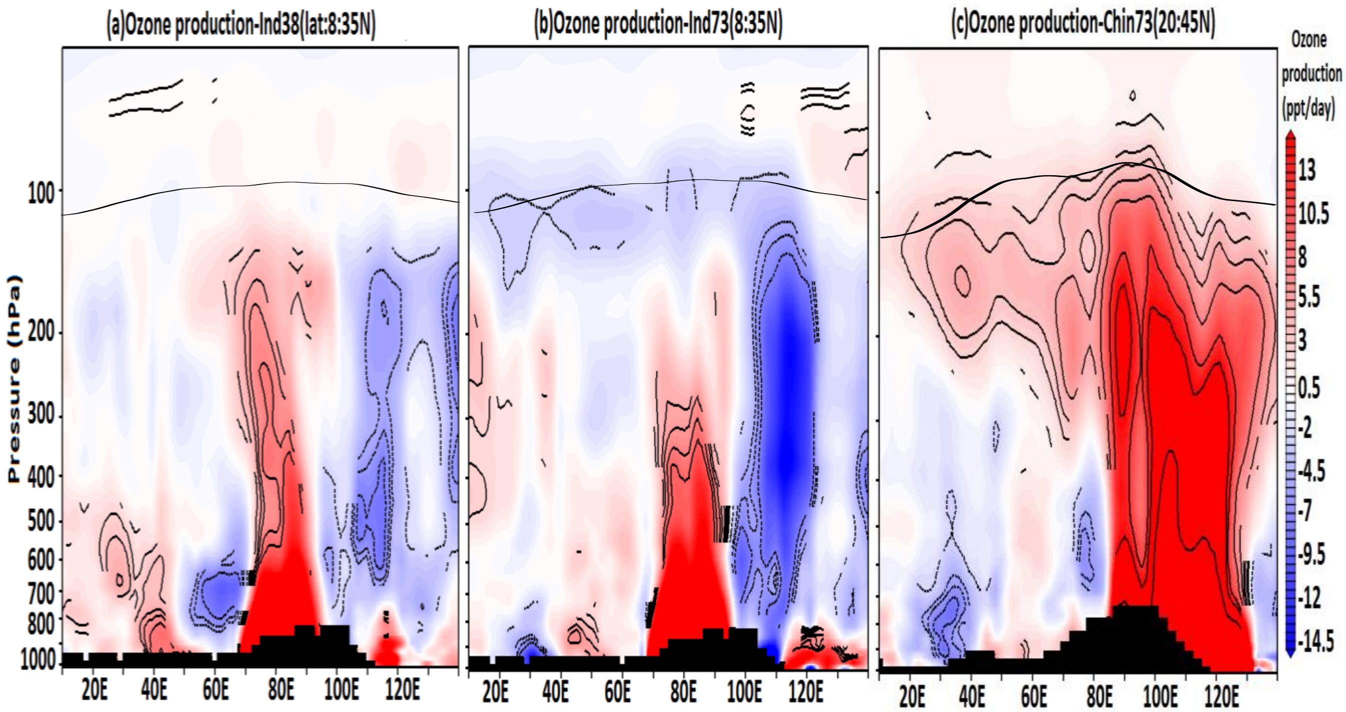

Figure 3: Longitude pressure cross-section of changes in net ozone production (ppt/day) due to enhanced NO_x with respect to the CTRL simulation, averaged for the monsoon season (June-September) obtained from (a) Ind38 (averaged over 8ºN-35ºN), (b) Ind73 (averaged over 8ºN-35ºN), and (c) Chin73 (over 20ºN-45ºN) simulations. The black line shows the tropopause while black contours indicate 95% confidence levels.

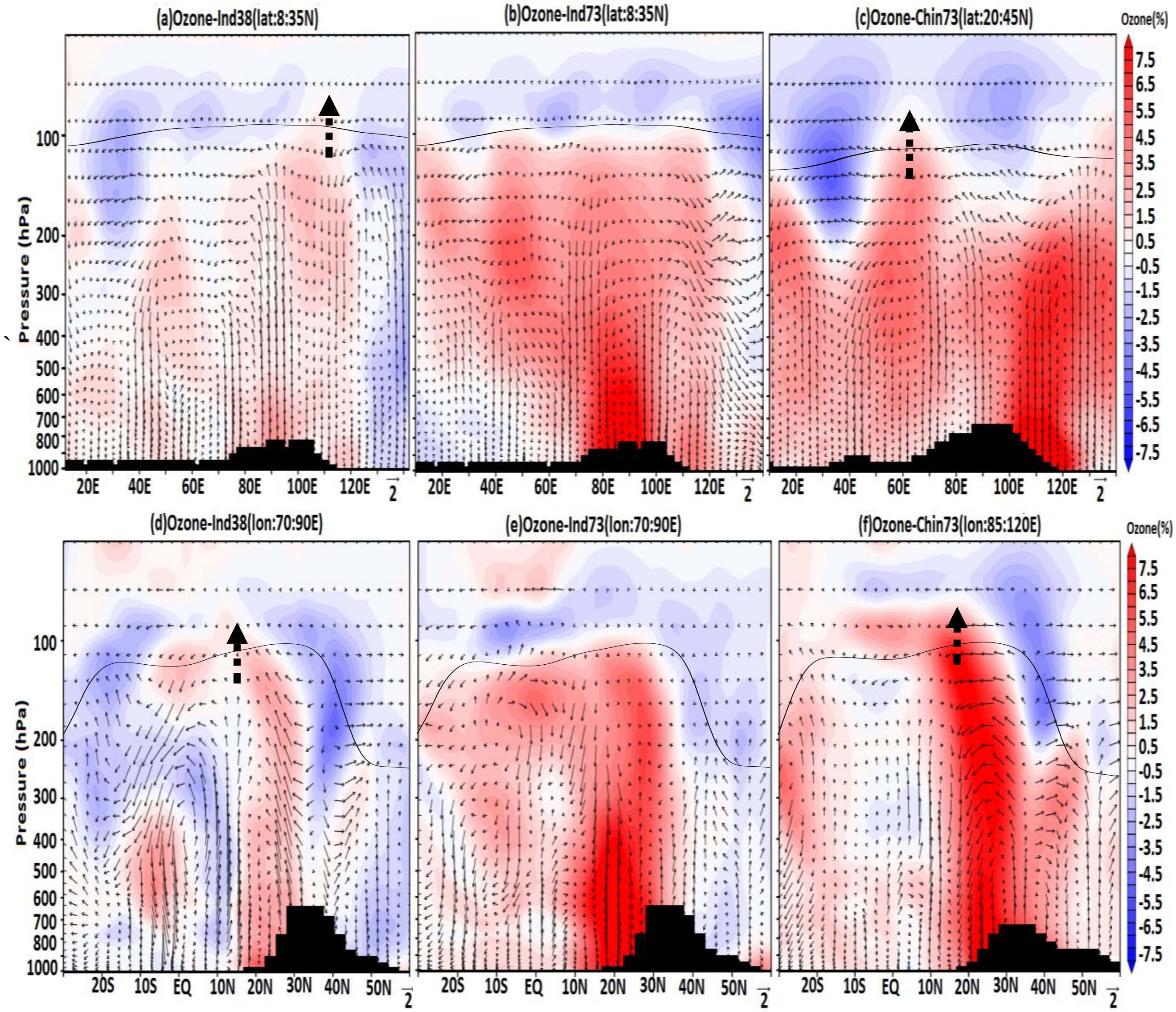

Figure 4: Longitude pressure cross-section of percentage ozone anomalies averaged for the monsoon season (June-September) obtained from (a) Ind38 (averaged over 8ºN-35ºN), (b) Ind73 (averaged over 8ºN-35ºN), and (c) Chin73 (averaged over 20ºN-45ºN) simulations. Latitude pressure cross-section of percentage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

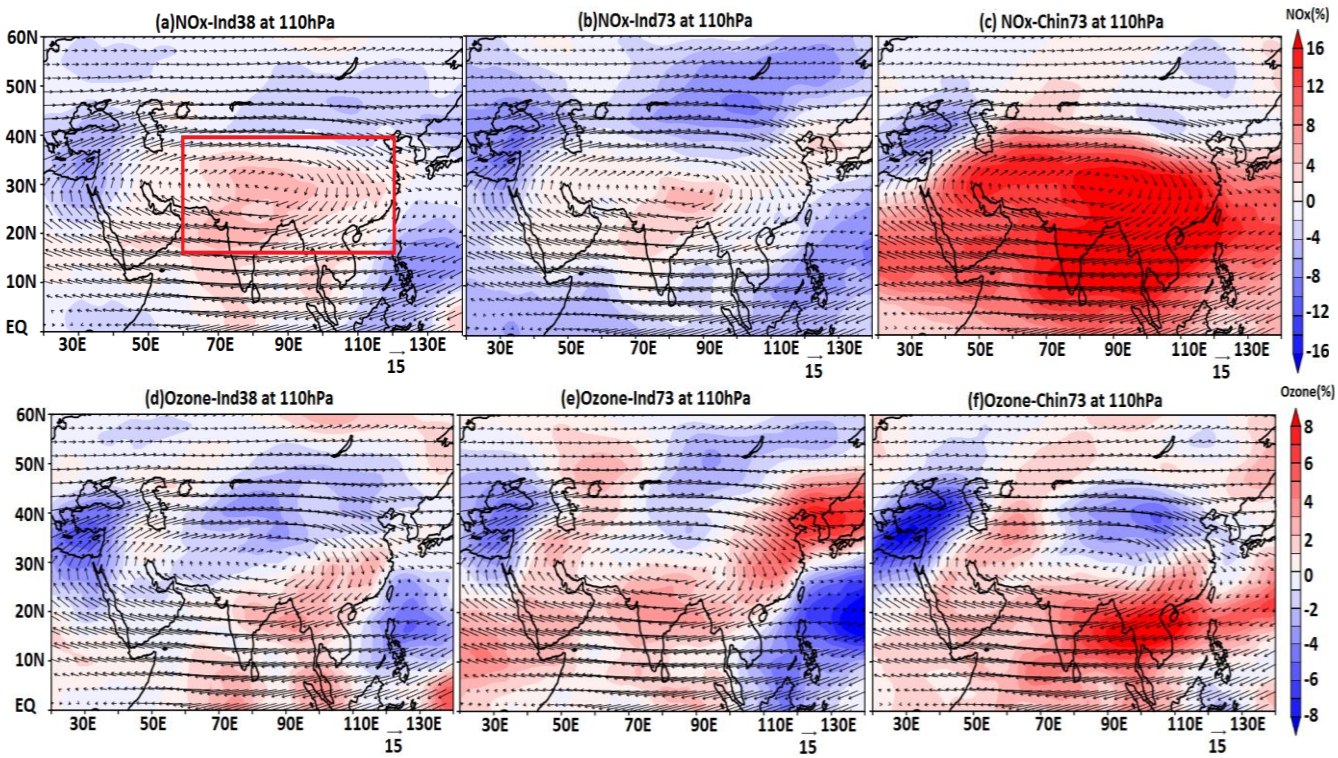

Figure 5: Latitude-longitude cross-section of percentage NO_x anomalies averaged for the monsoon season (June-September) at 110 hPa obtained from (a) Ind38, (b) Ind73, and (c) Chin73 simulations. Panels (d-f) show the same but for percentage ozone anomalies at 110 hPa for the (d) Ind38, (e) Ind73, and (f) Chin73 simulations. Black arrows indicate horizontal winds at 110 hPa. The red box in panel (a) indicates the ASM anticyclone region used to compute the associated radiative forcing regional average.

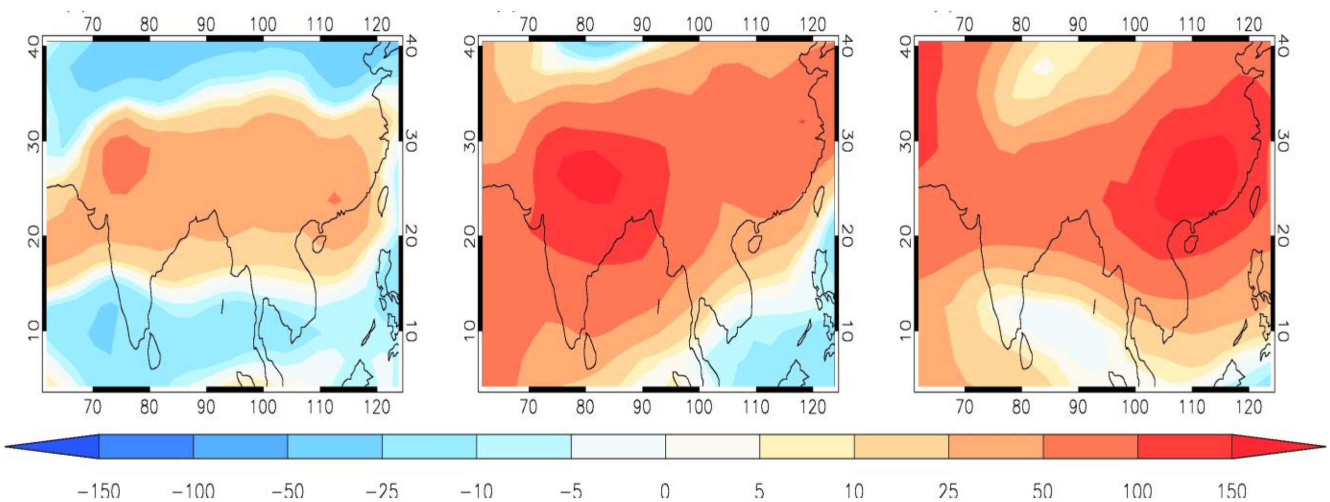

Figure 6: Latitude-longitude distribution of changes in ozone radiative forcing (in mW m^{-2}) for the (a) Ind38, (b) Ind73, and (c) Chin73 perturbed simulations, compared to the CTRL simulation.

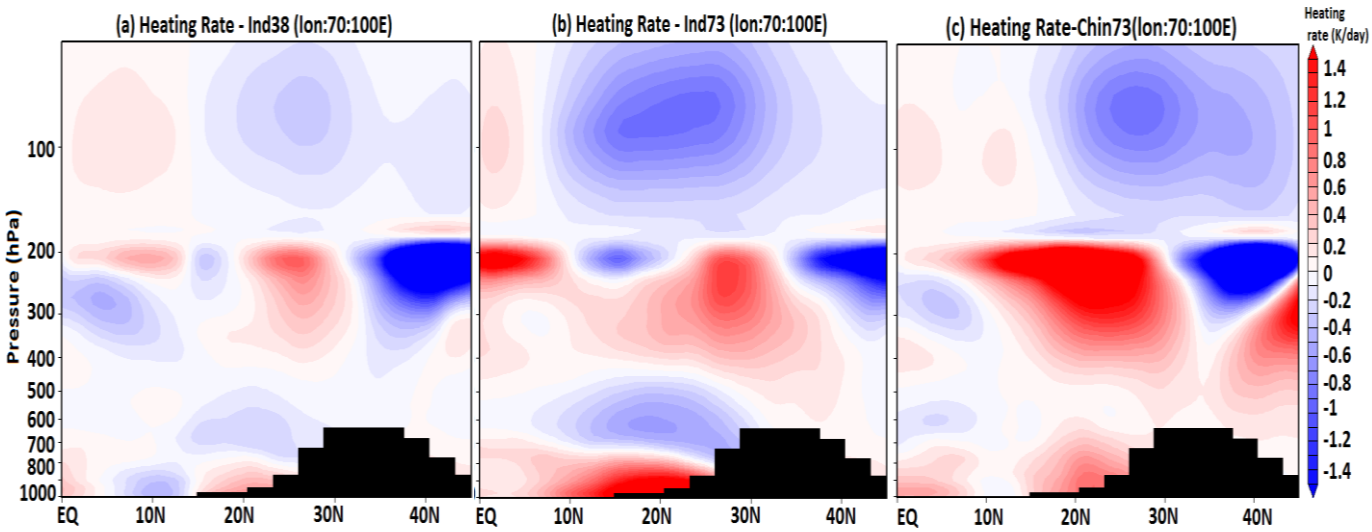

Figure 7: Latitude- pressure distribution of ozone heating rate changes (in K/day) for the (a) Ind38 (averaged over 70º-100ºE), (b) Ind73 (averaged 70º-100ºE), and (c) Chin73 (averaged over 90º -100º E) perturbed simulations, compared to the CTRL simulation.

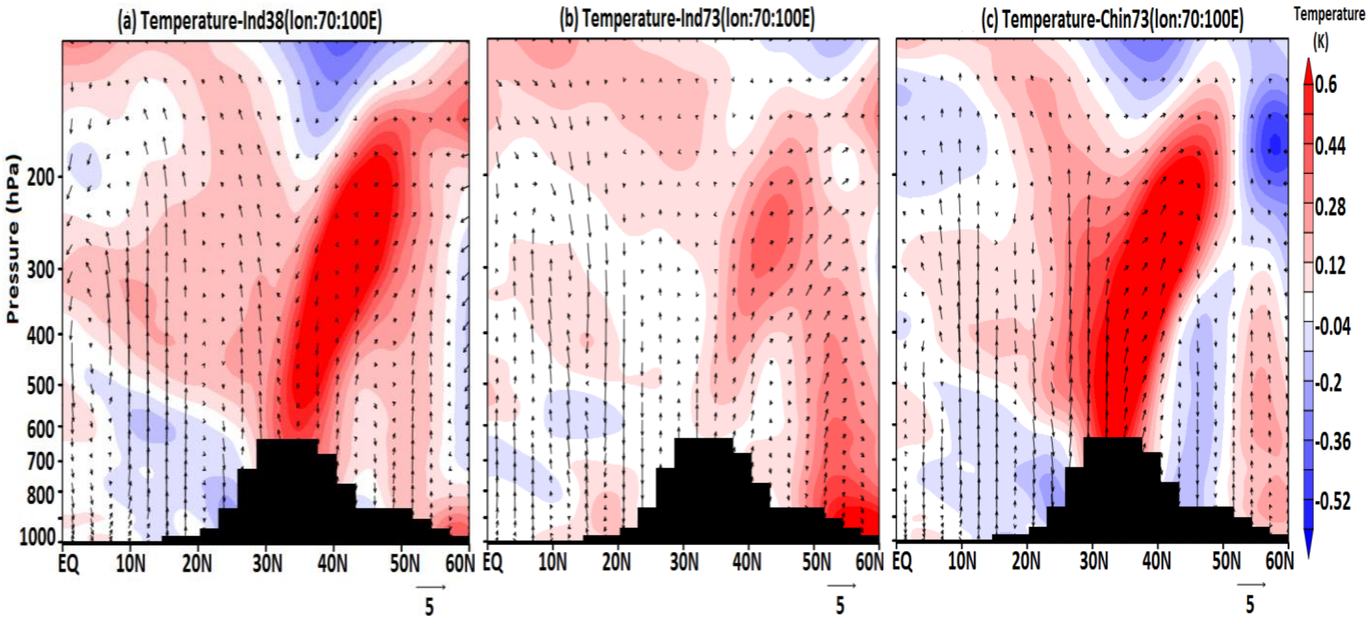

Figure 8: Latitude pressure cross-section of temperature anomalies (K) averaged for the monsoon season (June-September) and over 70ºE-100ºE obtained from (a) Ind38-CTRL, (b) Ind73-CTRL, and (c) Chin73-CTRL simulations. Black arrows indicate wind vectors (the vertical velocity field has been scaled by 300).

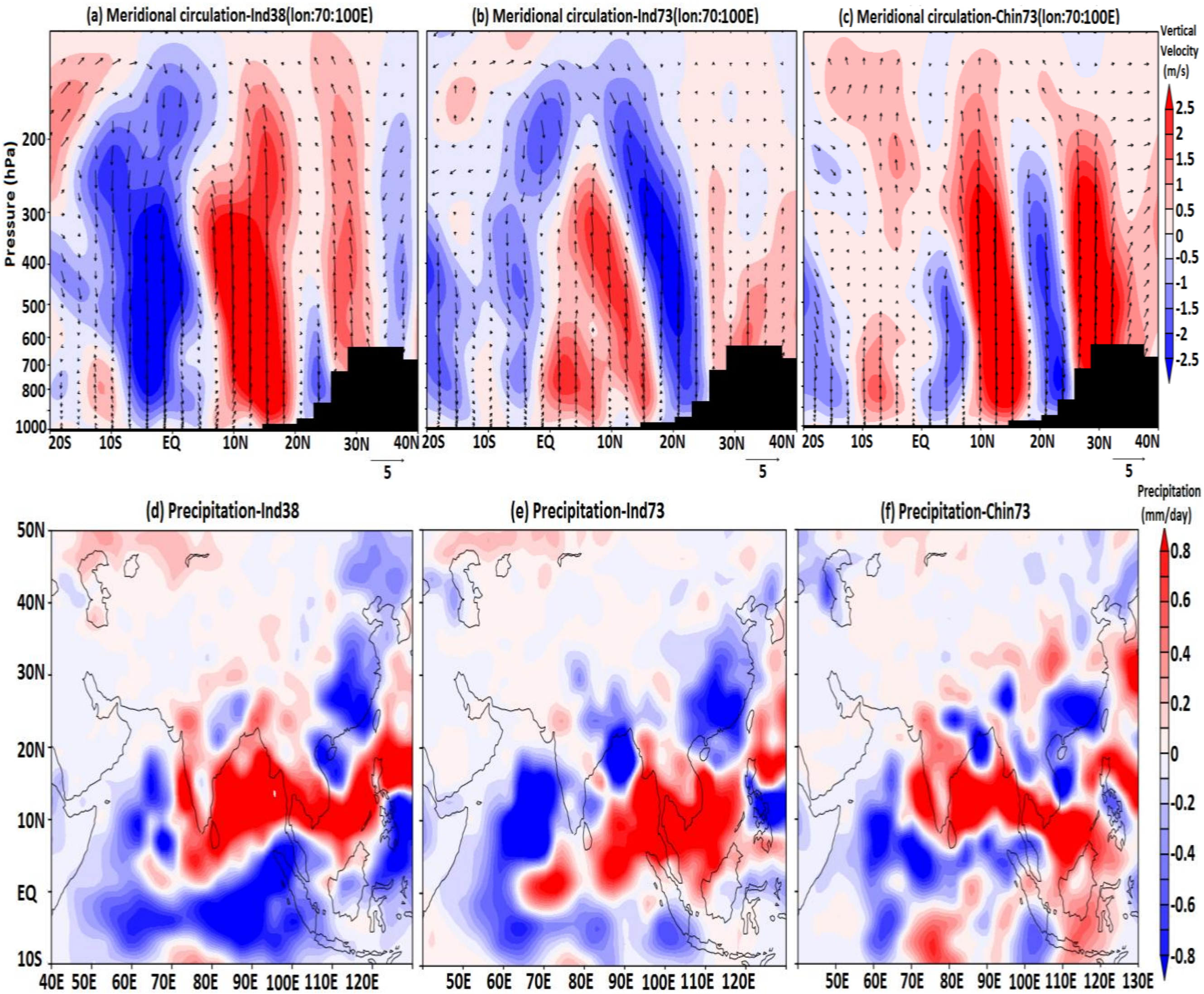

Figure 9: Difference in the meridional circulation due to enhanced NO_X emissions averaged for the monsoon season (June-September) and over 70ºE-110ºE for (a) Ind38-CTRL (b) Ind73-CTRL (c) Chin73-CTRL simulations. Shaded contours indicate the anomalies in vertical velocity (m/s). The vertical velocity field has been scaled by 300. Precipitation anomalies (mm/day) averaged for the monsoon season (June-September) obtained from (d) India38-CTRL (e) Ind73-CTRL, and (f) Chin73-CRTL simulations.

Supplementary Figures

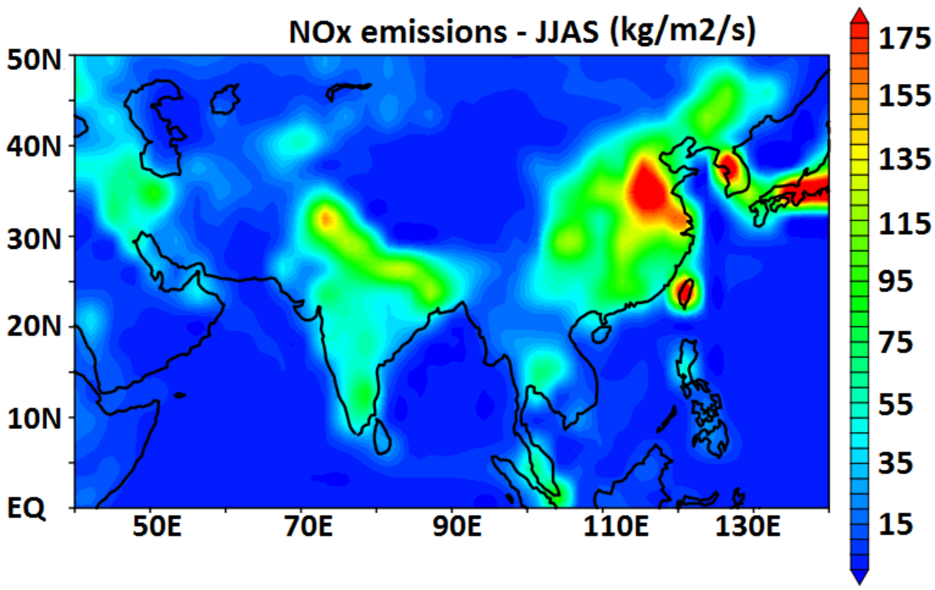

Figure S1: Distribution of NO_x emission mass flux (kg m^{-2} s^{-1}) from RETRO project data set for the year 2000, averaged for the monsoon season (June-September).

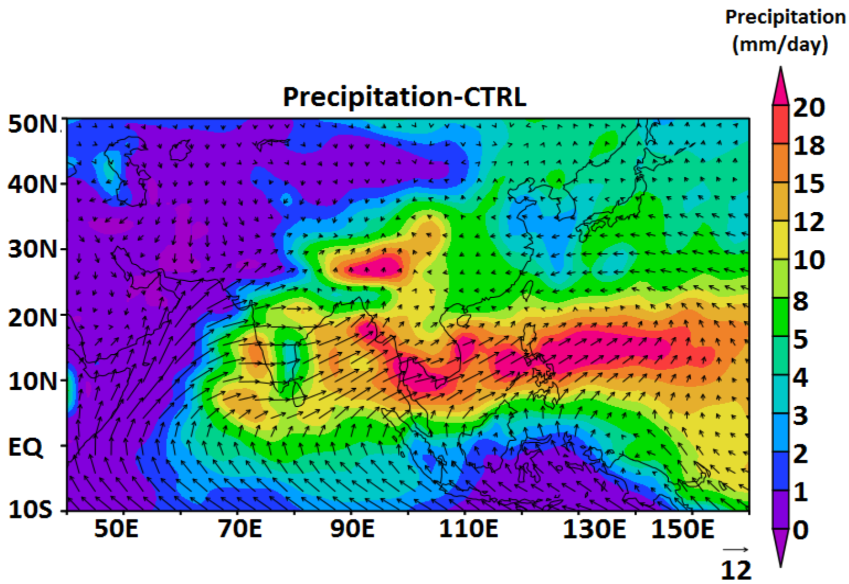

Figure S2: Distribution of seasonal (June-September) mean precipitation (mm/day) as obtained from CTRL simulation. Black arrows indicate winds (m/s).

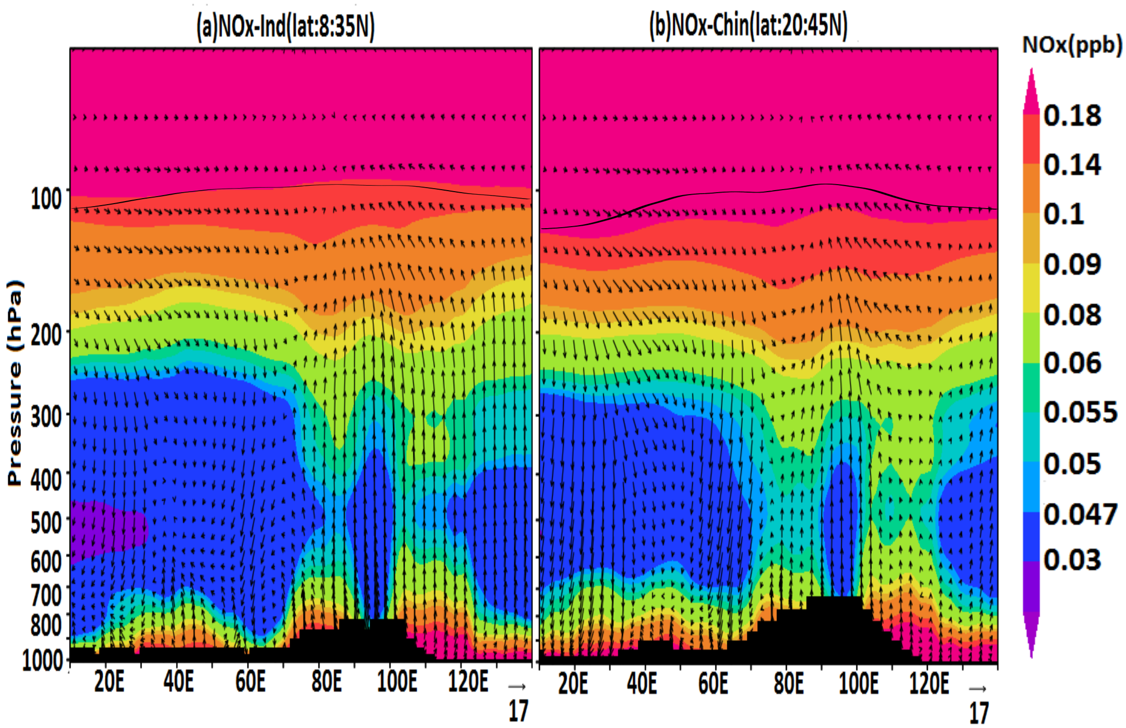

Figure S3: Vertical distribution of NO_x (ppb) averaged for the monsoon season (June-September) over (a) India (8º-35°N) and (b) China (20º-45°N) as obtained from CTRL simulations. Black arrows indicate winds (m/s) (the vertical component has been scaled by 300) and the black line represents the tropopause.

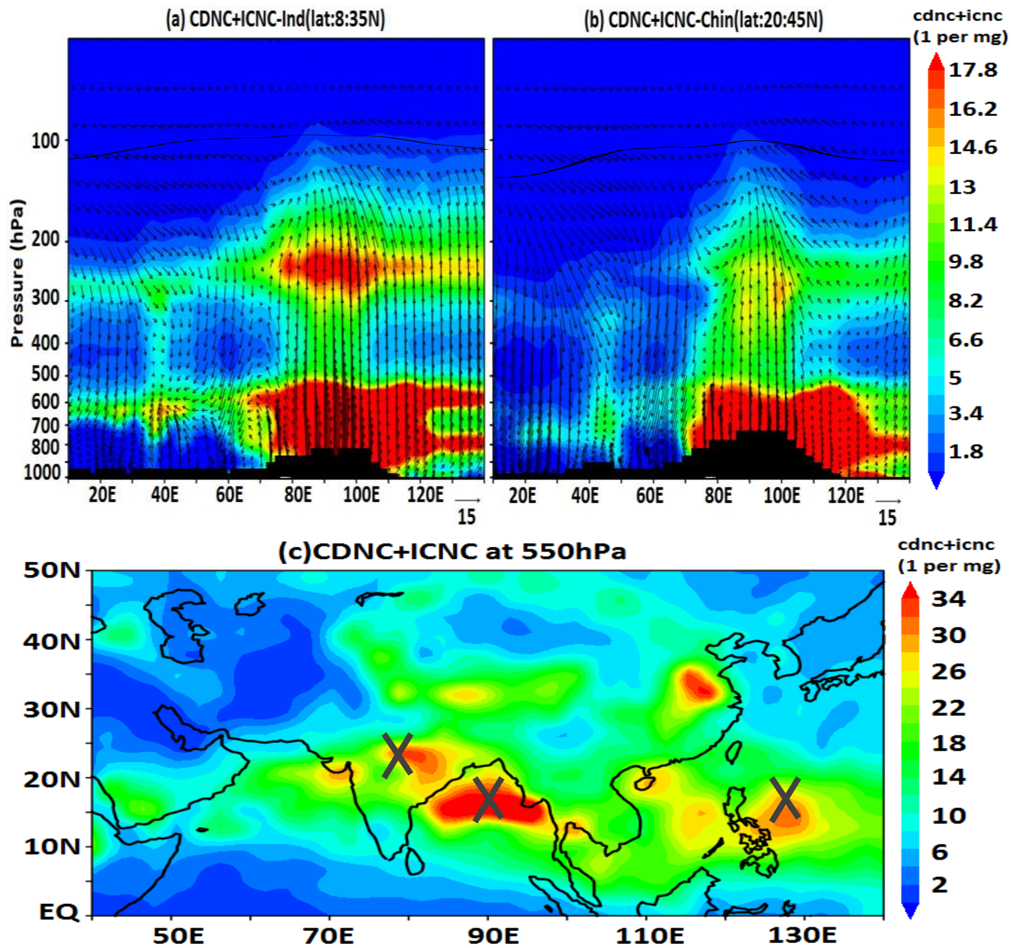

Figure S4: Distribution of combined cloud droplet (CDNC) and ice crystal (ICNC) number concentrations (in mg^{-1}) averaged for the monsoon season (June-September) over (a) India (8º-35°N) and (b) China (20º-45°N) as simulated in the CTRL simulation. Black arrows indicate winds (m/s) (the vertical component has been scaled by 300), and the black line represents the tropopause. (c) Distribution of seasonal (June-September) mean combined CDNC+ICNC (in mg^{-1}) at 550 hPa as simulated in the CTRL simulation. The regions of Bay of Bengal, South China Sea and southern slopes of Himalayas are indicated with a cross symbol.

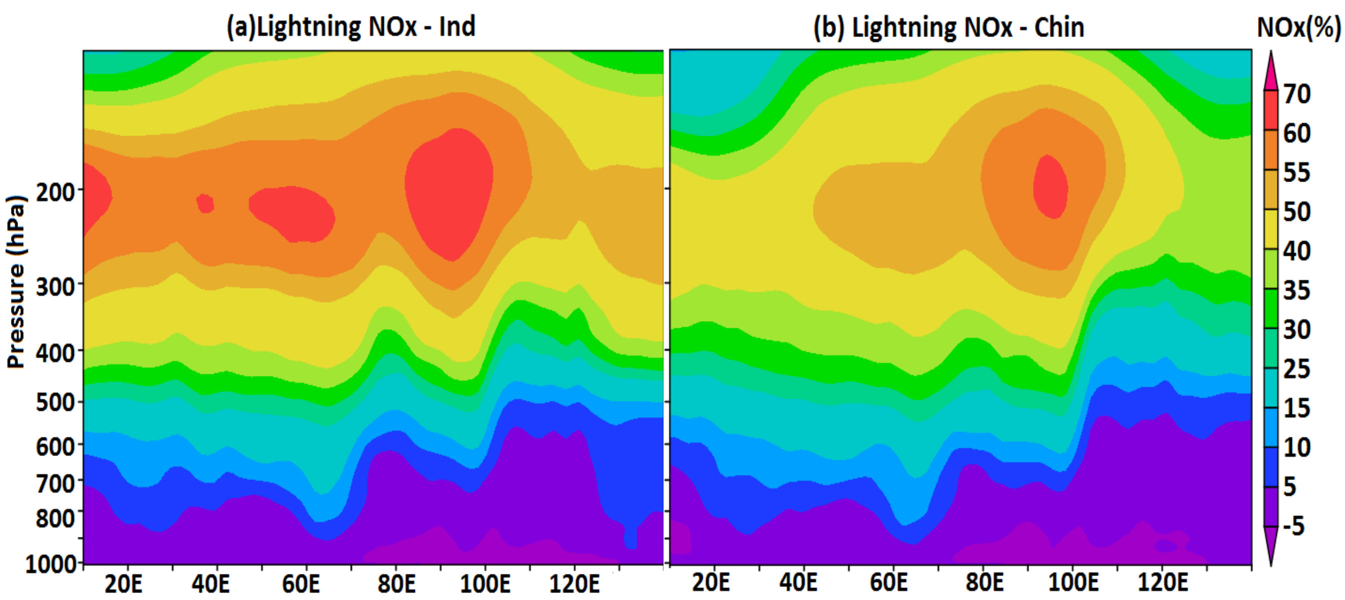

Figure S5: Vertical distribution of percentage NO_x anomalies produced from lightning, averaged for the monsoon season (June-September) over (a) India (8º-35°N) and (b) China (20º-45°N), simulated by comparing the CTRL-Lightning-on and the CTRL-Lightning-off experiments.

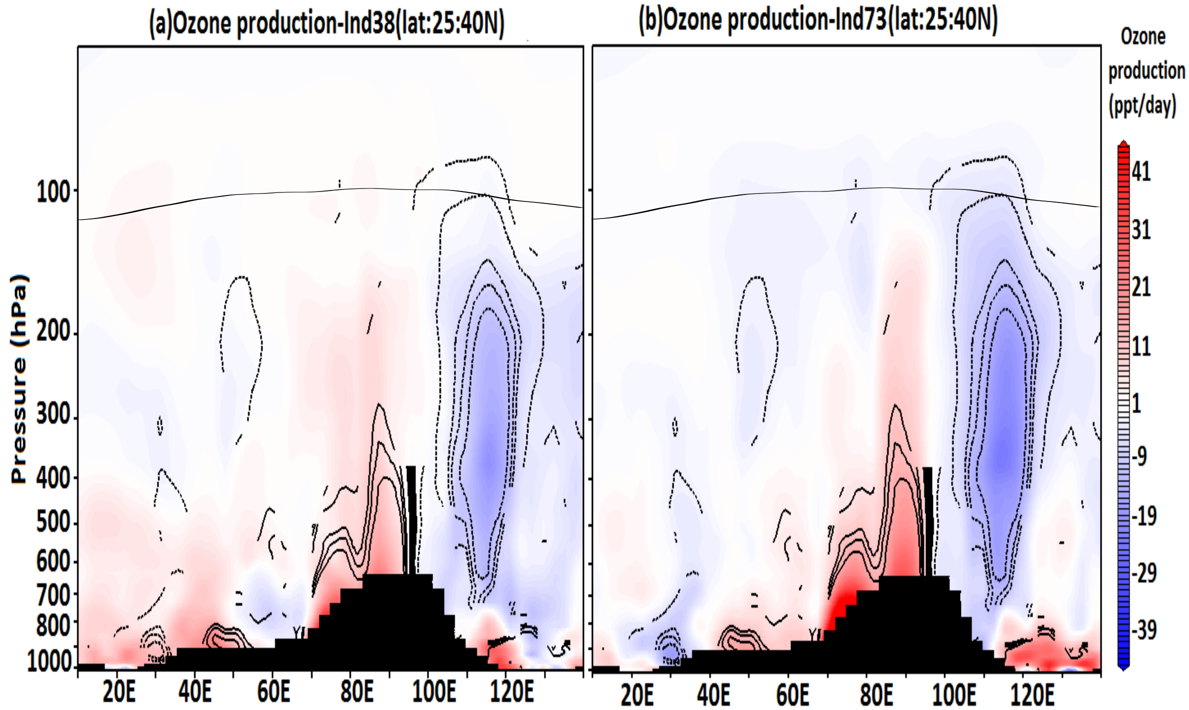

Figure S6: Longitude pressure cross-section of changes in net ozone production (ppt/day) due to enhanced NO_x with respect to the CTRL simulation, averaged for the monsoon season (June-September) and over the Indo-Gangatic plain-Tibetan Plateau region (25ºN-40ºN) for (a) Ind38 (b) Ind73. The black line shows the tropopause while black contours indicate 95% confidence levels.