

Owing to the importance of ozone chemistry and its implication for air quality and climate change, studies focusing on the origin of ozone enhancement in the troposphere, its trends and distributions need immediate consideration. Increase in the tropospheric ozone is considered to be due to (1) in-situ photochemical formation associated with lightning, advection, anthropogenic (e.g., Jacobson, 2002 and references therein), and (2) stratospheric flux (Wild, 2007 and reference therein). Increase of the ozone flux from the stratosphere to the troposphere (stratospheric intrusion) not only increases the tropospheric ozone, but also decreases the stratospheric ozone, which in principle enhances the penetration of UV to reach the Earth's surface.

In general, stratospheric intrusion is a slow process and is a mid- and high-latitude phenomenon linked with synoptic scale disturbances (Holton et al., 1995). Liang et al. (2009) have described the time scale of stratospheric ozone intrusion that occurs in 3 steps, and takes about three months to reach from stratosphere to lower troposphere. Maximum ozone forms over the tropics, moves upward and then pole-ward by Brewer–Dobson circulation, which further descends to the lower stratosphere and troposphere at high latitudes. There is much observational evidence supporting the slow intrusion of stratospheric air into the troposphere during cut-off lows (Vaughan and Price, 1989), high/low pressure systems (Davies and Schuepbach, 1994), tropopause folds (Sprenger and Wernli, 2003) and in a rapid episodic manner which generally triggered by overshooting convection, like a tropical cyclones (Loring et al., 1996; Baray et al., 1999; Cairo et al., 2008 and references therein; Das, 2009; Das et al., 2011, Zhan and Wang, 2012). The tropical cyclones are the synoptic-scale disturbances of organised convective systems which weaken the tropopause by overshooting convection and enhancing the stratosphere–troposphere exchange in a spontaneous manner. Slow stratospheric intrusion is reasonably well understood and is a regular phenomenon, whereas the rapid intrusion needs to be understood in detail and possibly its ill effect on living organisms.

The intrusion of the stratospheric air into the troposphere has been demonstrated by Hocking et al. (2007) for mid-latitude condition and Das (2009) for low latitude during

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cyclone using high power radar system (VHF radar). Subsidence of stratospheric air is generally observed in the vicinity of cyclone (Baray et al., 1999; Cairo et al., 2008; Leclair De Bellevue et al., 2006, 2007; Das, 2009, 2011). Appenzeller and Davies (1992) have shown the intrusion of the stratospheric air into the troposphere due to disturbed weather condition over mid-latitude, combining weather prediction model and satellite observations. In addition to the stratospheric intrusion associated with organised convections, there are ample observations, which reveal that humid air from the troposphere could penetrate into the stratosphere. Further, stratospheric ozone will get destroyed by the humid air, which penetrates beyond the tropopause (Rossow and Pearl, 2007; Cairo et al., 2008). A complete review on the effect of tropical cyclones on the upper troposphere and lower stratosphere can be found in Cairo et al. (2008). In spite of many observational and modelling studies, the cross-tropopause transport in short-time scale associated with tropical cyclones is still unclear.

The present study addresses the influence of tropical cyclones quantitatively on enhancement of tropospheric ozone by stratospheric intrusion. This ozone intrusion observed in the lower troposphere not only acts as a pollutant but also cool the stratosphere and warm the troposphere, hence plays a vital role in the Earth's radiation budget.

2 Campaign details

An intense campaign, named as “Troposphere–Stratosphere Exchange-Cyclone (TSE-C)” under the Climate And Weather of Sun-Earth System (CAWSES)-India phase-II programme was conducted during two cyclone events. Under this campaign, a series of ozonesondes were launched from Trivandrum (8.5° N, 76.5° E) during the intense period of cyclonic storm Nilam from 30 October to 07 November 2012 and a very-severe cyclonic storm Phailin from 11 to 15 October 2013. The ozonesondes used are EN-SCI (USA) make, which were integrated with the GPS based radiosondes of i-met make. These standard ozonesonde are made up of the Electrochemical Concentra-

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for some time and then northward and finally north-northeastwards up to southwest Bihar. The system weakened gradually into a cyclonic storm from 13 October 2013 and finally the intensity decreased to a low pressure system on 14 October 2013.

The track of each tropical cyclones and outgoing long wave radiation (OLR) images (date and time are stamped) are shown in Fig. 1a and b, respectively. The detailed bulletin can be found in www.imd.gov.in. During these campaigns, several ozonesondes were launched from Trivandrum (8.5° N, 76.5° E), whenever the intensity of cyclones were maximum and the path/eye was close to the launching site.

4 Numerical simulation

Apart from the radiosonde/ozonesonde observations, a high resolution numerical simulation using the Advanced Research Weather Research and Forecast (WRF-ARW) model version 3.6 has also been carried out for both cases of cyclones. The model domain has been configured with two nested domains of 60 and 20 km horizontal resolution, and covers an area extending from 1° S to 25° N and 60 to 100° E. The innermost domain has been used for the present study. The initial and lateral boundary conditions have been taken from ERA-Interim reanalysis on 0.75° × 0.75° continuously at every 6 h. The present simulation was carried out with the model Physics options: (i) New Simplified Arakawa–Schubert (NSAS) (Han and Pan, 2011), (ii) Yonsei University (YSU) boundary layer scheme (Hong et al., 2006), (iii) Rapid Radiative Transfer Model (RRTM) long wave radiation scheme (Mlawer et al., 1997), (iv) WRF Single Moment (WSM) 5 class microphysical scheme (Hong et al., 2004), and (v) NOAA land-surface scheme (Smirnova et al., 2000).

5 Results and discussion

Figure 2a and b show the profiles of ozone mixing ratio (OMR) and relative humidity (RH) from ozonesonde measurements during the passage of tropical cyclones Nilam 19311

(top panels) and Phailin (bottom panels). The background ozone profile is obtained by averaging individual profile (23 profiles) of October from 1995–2013 and shown by dotted lines in Fig. 2. During the passage of Nilam on 30 October 2012, enhancement in tropospheric ozone (marked by horizontal arrows) from background by 40–50 ppbv was observed in the height region between 8–9 km (~ 1 km width) and 11–14 km (~ 3 km width). This enhancement persisted till 31 October 2012 but at the height region between 6–7 km with a reduced width. However, the enhancement of about ~ 40 ppbv was still observed on 02 November 2012 but the height region decreased to 5–6 km. After two days, we had again observations from 05–07 November 2012. The height of enhancement in the ozone profiles were reduced to ~ 4, ~ 3 and ~ 1.5 km by 40, 30, and 20 ppbv on 05, 06 and 07 November 2012, respectively. The present observation reveals that the intrusion of the enhanced upper tropospheric ozone into the lower troposphere occurs in episodic manner, sloping down from the upper troposphere to near surface. The descending rate of the ozone rich layer from the upper troposphere to the boundary layer during Nilam is approximately estimated to be ~ 875 m day⁻¹. It is also noted that the corresponding relative humidity profiles during Nilam did not decrease with increasing ozone mixing ratio except on 02 November 2012. A significant sudden decrease in relative humidity is observed on 02 November 2012 at ~ 6 km, where the maximum enhancement of tropospheric ozone is observed. This is due to the presence or accumulation of dry air, which is believed to have been originated from the stratosphere. A similar phenomenon is also observed during the passage of Phailin. Intrusion from ~ 14 to 6 km (marked by horizontal arrows) is clearly observed in the ozone profiles from 11–15 October 2013. During Phailin, tropospheric ozone enhancement by 20–30 ppbv is observed and the width of the enhanced ozone layer is larger than that during Nilam. During Phailin, descending rate of enhanced ozone layer from the upper troposphere to the boundary layer is estimated to be ~ 1000 m day⁻¹. Relative humidity profiles also show the sudden decrease between 2–6 km on 14 and 15 October 2013, indicating the presence of dry air similar to that observed during the Nilam. In addition to the profiling of ozone, we have surface measurement of ozone

during the Phailin. Figure 2c shows the time series of near-surface ozone mixing ratio from 11 to 19 October 2013. As expected, clear diurnal variability is observed in the time-series of ozone. From 11 to 14 October the maximum and minimum average peak of ozone are observed to be 24 and 1 ppbv, respectively, whereas after 14 up to 18 October 2013, the maxima and minima is observed to be 35 and 10 ppbv, respectively. The upper and lower average is indicated by horizontal solid and dash lines respectively. The ozone profiles also show that enhanced ozone layer propagates downward from the upper troposphere starting on 11 October, reaches to the lower troposphere and near-surface on 15 October 2013. In addition to the intrusion of ozone from the upper troposphere to the surface, there are three main mechanisms for the production of ozone in the atmospheric boundary layer: (1) photochemical reaction via NO_x channel, (2) bio-mass burning/fossils fuel, and (3) lightning. During the passage of tropical cyclone, there is a substantial reduction in the solar radiation, no bio-mass burning and no lightning and thus, the formation of ozone by any of the above three mechanisms will be very low. Thus, increase of 10–15 ppbv in near-surface ozone is mainly attributed due to the intrusion of air from the higher heights.

Further, dynamical analysis is carried out using WRF-ARW simulation. Figure 3 shows the height-latitude cross-section of (a) vertical velocity along with potential vorticity and potential temperature contours, and (b) relative humidity cross-section along with equivalent potential temperature and zonal wind at 79°E at 18:00 GMT on 30 October 2012 for Nilam (left panels) and 20:00 GMT on 10 October 2013 for Phailin (right panels). The vertical velocity profiles shows the presence of downdraft (blue) followed by updraft (red) between $8\text{--}17^\circ \text{N}$ at the UTLS region in both the cyclone cases. Enhanced potential vorticity 0.5–1.5 PVU is also observed vertically down from the stratosphere to the surface, overlapping the downdraft regions. The potential temperature contours indicate the presence of unstable atmosphere at this location and noticed that stable stratospheric air penetrated downward at $12\text{--}14^\circ \text{N}$ for Nilam and $16\text{--}18^\circ \text{N}$ for Phailin. Relative humidity profiles indicate the presence of dry air at $\sim 8^\circ \text{N}$ which is in the vicinity of ozonesonde observational site. The equivalent potential temperature

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contours in Fig. 3b indicate that from surface to 10 km it is highly unstable for vertical motion and favourable condition for the convection to take place at $6\text{--}12^\circ \text{N}$ for Nilam and $12\text{--}18^\circ \text{N}$ for Phailin. In the same latitude regions, from 10 km to the tropopause level, the vertical motion is suppressed and the atmosphere is found to be statically stable to the unstratified atmosphere. The present condition indicates the presence of statically stable stratospheric air in the upper and middle troposphere. Similarly, Fig. 3 shows the height-time cross-section of (a) vertical velocity along with potential vorticity and potential temperature contours, and (b) relative humidity cross-section along with equivalent potential temperature and zonal wind at Trivandrum at 18:00 GMT on 30 October 2012 for Nilam (left panels) and 20:00 GMT on 10 October 2013 for Phailin (right panels). From these figures, presence of strong downdrafts of dry air is clearly seen during 29–31 October 2012 (10–12 October 2013) for Nilam (Phailin). Numerical simulation confirms the presence of stratospheric intrusion in to the troposphere.

To get further insight, relative humidity derived from SAPHIR onboard the Megha-Tropiques satellite is used. The relative humidity shown is an average over 12–14 passes day^{-1} . Figure 4a shows the height-time intensity plot of relative humidity during the passage of the cyclones: Nilam (left panel) and Phailin (right panel). The grid is averaged from $4\text{--}8^\circ \text{N}$ and $83\text{--}88^\circ \text{E}$. Strong dry air intrusion originated from lower stratosphere is observed between 23–27 October 2012 (Nilam) and 12–18 October 2013 (Phailin). In both cyclones, dry air (low humidity region) reached down to the height of 8 km. For the perception of spatial distribution of relative humidity, latitude–longitude plot of relative humidity averaged over different height level is shown in Fig. 4b. The low value of relative humidity i.e., the presence of dry air on the same day of enhanced ozone mixing ratio in between 5 and 10 km prove that the dry air present in the upper and middle troposphere is of stratospheric origin. The present observations show the influence of tropical storms on atmospheric composition especially ozone and water vapour apart from its effect on weather system.

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- Hong, S. Y., Noh, Y., and Dudhia, J.: A new vertical diffusion package with an explicit treatment of entrainment processes, *Mon. Weather Rev.*, 134, 2318–2341, 2006.
- Jacobson, M. Z.: Control of fossil-fuel particulate black carbon and organic matter, possibly the most effective method of slowing global warming, *J. Geophys. Res.*, 107, 16–22, 2002.
- 5 Komhyr, W. D., Barnes, R. A., Brothers, G. B., Lathrop, J. A., and Opperman, D. P.: Electrochemical concentration cell ozonesonde performance evaluation during STOIC 1989, *J. Geophys. Res.*, 100, 9231–9244, 1995.
- Leclair, De Bellevue, J., R'echou, A., Baray, J. L., Ancellet, G., and Diab, R. D.: Signatures of stratosphere to troposphere transport near deep convective events in the southern subtropics, *J. Geophys. Res.*, 111, D24107, doi:10.1029/2005JD006947, 2006.
- 10 Leclair, De Bellevue, J., Baray, J. L., Baldy, S., Ancellet, G., Diab, R. D., and Ravetta, F.: Simulations of stratospheric to tropospheric transport during the tropical cyclone Marlene event, *Atmos. Environ.* 41, 6510–6526, 2007.
- Liang, Q., Douglass, A. R., Duncan, B. N., Stolarski, R. S., and Witte, J. C.: The governing processes and timescales of stratosphere-to-troposphere transport and its contribution to ozone in the Arctic troposphere, *Atmos. Chem. Phys.*, 9, 3011–3025, doi:10.5194/acp-9-3011-2009, 2009.
- 15 Loring Jr., R. O., Fuelberg, H. E., Fishman, J., Watson, M. V., and Browell, E. V.: Influence of a middle-latitude cyclone on tropospheric ozone distributions during a period TRACEA, *J. Geophys. Res.*, 101, 23941–23956, 1996.
- Mathur, A. K., Gangwar, R. K., Gohil, B. S., Deb, S. K., Kumar, P., Shukla, M. V., Simon, B., and Pal, P. K.: Humidity profile retrieval from SAPHIR on-board the Megha-Tropiques, *Curr. Sci.*, 104, 1650–1655, 2013.
- 25 Mlawer, E. J., Taubman, S. J., Brown, P. D., Iacono, M. J., and Clough, S. A.: Radiative transfer for inhomogeneous atmosphere: RRTM, a validated correlated-k model for the long wave, *J. Geophys. Res.*, 102, 16663–16682, 1997.
- Raju, G.: Engineering challenges in the Megha-Tropiques, *Curr. Sci.*, 104, 1662–1670, 2013.
- Rossow, W. B. and Pearl, C.: 22-year survey of tropical convection penetrating into the lower stratosphere, *Geophys. Res. Lett.*, 34, L04803, doi:10.1029/2006GL028635, 2007.
- 30 Sprenger, M. and Wernli, H.: A northern hemispheric climatology of cross-tropopause exchange for the ERA15 time period (1979–1993), *J. Geophys. Res.*, 108, 8521, doi:10.1029/2002JD002636, 2003.

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- Smirnova, T. G., Brown, J. M., Benjamin, S. G., and Kim, D.: Parameterization of cold season processes in the MAPS land-surface scheme, *J. Geophys. Res.*, 105, 4077–4086, 2000.
- Subrahmanyam, K. V. and Kumar, K. K.: Megha-Tropiques/SAPHIR measurements of humidity profiles: validation with AIRS and global radiosonde network, *Atmos. Meas. Tech. Discuss.*, 6, 11405–11437, doi:10.5194/amtd-6-11405-2013, 2013.
- 5 Vaughan, G. and Price, J.: Ozone transport into the troposphere in a cut-off low event, in: *Ozone in the Atmosphere*, edited by: Bojkov, R. and Fabian, P., A. Deepak Publishing, Hampton (USA), 415–418, 1989.
- Venkat Ratnam, M., Basha, G., Murthy, B. V. K., and Jayaraman, A.: Relative humidity distribution from SAPHIR experiment on board Megha-Tropiques satellite mission: comparison with global radiosonde and other satellite and reanalysis data sets, *J. Atmos. Res.*, 118, 9622–9630, 2013.
- 10 Wild, O.: Modelling the global tropospheric ozone budget: exploring the variability in current models, *Atmos. Chem. Phys.*, 7, 2643–2660, doi:10.5194/acp-7-2643-2007, 2007.
- 15 Zhan, R. and Wang, Y.: Contribution of tropical cyclones to stratosphere–troposphere exchange over the northwest Pacific: estimation based on AIRS satellite retrievals and ERA-Interim data, *J. Atmos. Res.*, 117, D12112, doi:10.1029/2012JD017494, 2012.

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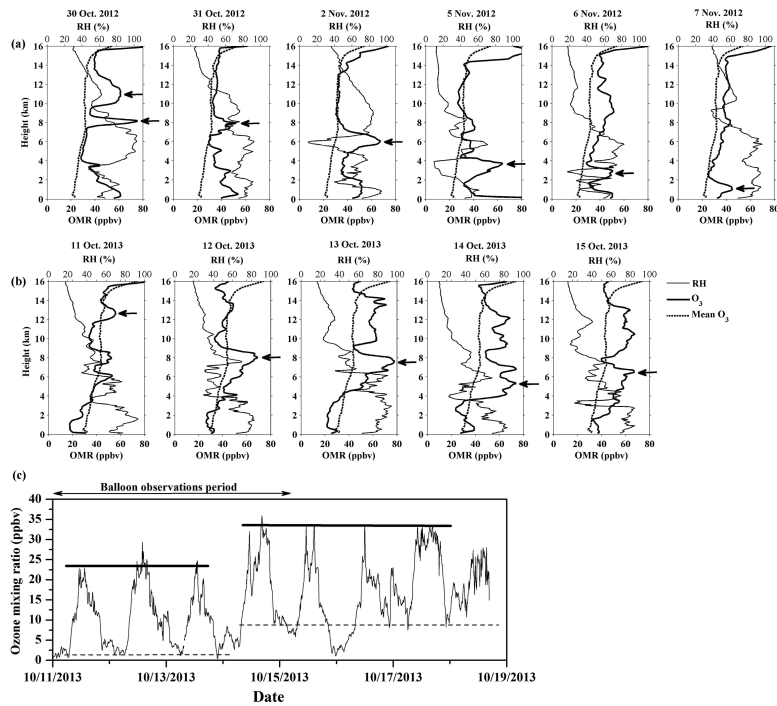


Figure 2. (a) Profiles of ozone mixing ratio (OMR) (dark black line) and relative humidity (gray line) for individual days during passing of tropical cyclones (a) Nilam and (b) Phailin. The long-term OMR mean for non-convective days (as control day) is shown in dotted line. (c) Time series of surface ozone mixing ratio from 11 October 2013 at 00:00 LT to 19 October 2013 at 23:55 LT. The data is collected every 5 min.

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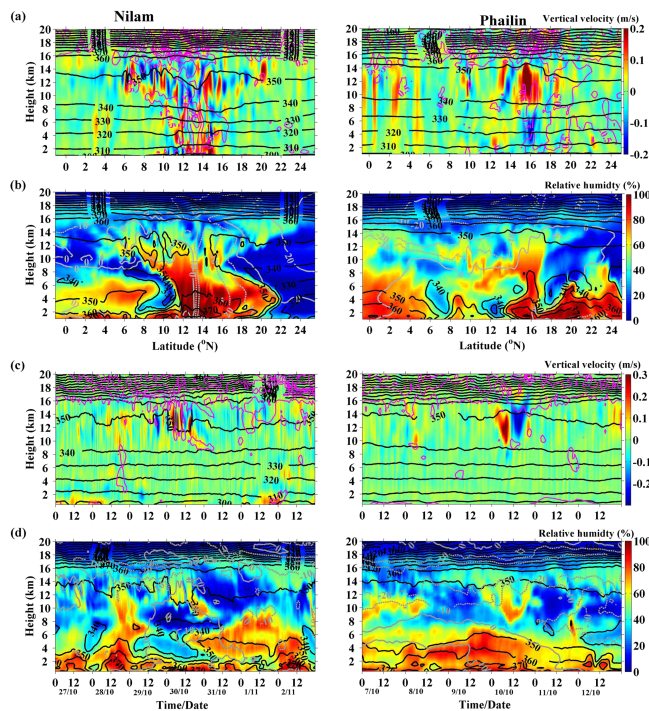


Figure 3. Height-latitude cross-section of (a) vertical velocity along with potential vorticity (magenta) and potential temperature (black) contours, and (b) relative humidity cross-section along with equivalent potential temperature (black) and zonal wind (grey) at 79° E at 18:00 GMT on 30 October 2012 for Nilam (left panels) and 20:00 GMT on 10 October 2013 for Phailin (right panels). (c and d) Same as Fig. 4a and b, respectively but for height-time cross-section over Thumba (8.5° N, 76.9° E). The above parameters are obtained from WRF simulation.

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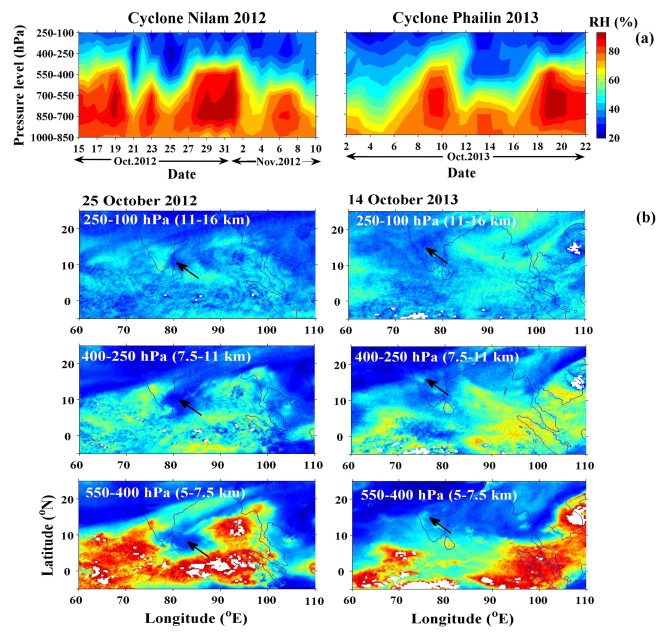


Figure 4. (a) Pressure-time variation of relative humidity obtained from SAPHIR onboard Megha-Tropiques satellite during the cyclones Nilam (left panel) and Phailin (right panel). (b) Latitude-longitude distribution of relative humidity derived from same satellite at different pressure levels (stamped on each panel) for Nilam (25 October 2012) and Phailin (14 October 2013). The data is averaged for one day which is 12–14 passes at different timings and arrows in (b) indicates the presence of dry air.