

Dear Sir,

On behalf of my co-authors, I would like to sincerely thank the editor for processing and reviewing the revised manuscript. I also thank the editor and all the three referees for their constructive comments and suggestions which improved the manuscript significantly. As suggested by the editor, we will definitely take-up a separate study on air trajectory calculation during the passage of tropical cyclone.

Following the editor's and the reviewer's suggestion and comments, we have revised the manuscript accordingly. Necessary English grammar corrections are also made in the revised manuscript.

Point-by-point responses on how we have addressed each recommendation/suggestions are attached herewith.

We are herewith submitting the revised manuscript (track change) with figures and table for the consideration of publication in your esteemed journal 'Atmospheric Physics and Chemistry'. I request you to kindly process the manuscript and do the needful. Please acknowledge the receipt of the same.

Thanking you
With best regards
Siddarth Shankar Das

Response to Editor's Comments

We would like to sincerely thank the Editor for a very positive evaluation and constructive comments and suggestion which improved the manuscript significantly. Necessary English grammar corrections are also made in the revised manuscript Point-by-point response on how we have addressed each recommendations and suggestion are given below.

**I16: '0.8-1 km/day, which is thrice than that of non-convective day descending rate.' — unclear. First 'thrice than that' would be much clearer as 'three times that'. ('thrice' is old-fashioned and not in common use.) Second I'm not clear what you mean by 'non-convective day descending rate'. Do you mean typical descent rate on a non-convective day? Do you mean 'day-time' (if so why)? Or what? 'descending' should be replace by 'descent' in the two occurrences in the sentence as a whole. The text on this topic later in the paper (I215-217) should also be clarified.*

In the present manuscript, non-convective days mean cloud free or clear-sky days. Non-convective days decent rate was estimated by Gettelman et al. (2004) and it is an average value. Non-convective days is the average of day and night time (diurnal mean). The sentence is rephrased in the Abstract. The decent rate estimated by Gettelman et al. (2004) during clear-sky days is 0.1-0.3 km/day. Our estimation on decent rate during the passage of tropical cyclone is 0.8-1 km/day which is three-times that of clear sky days. This estimation was previously suggested by Referee #1.

**I61: '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.' — I suppose that this is a description that starts from a level significantly above the tropopause. It is a little confusing bearing in mind your 'slow' vs 'fast' characterisation later. Surely the processes that you identify as 'slow' act on time scales much less than three months? I suggest that you are clearer about what 'slow' means. [Note that this was a point highlighted previously by Referee 3 — you say in your reply 'Thanks for pointing out this aspect and appropriate modifications are done in the revised manuscript' — but in my view you have not provided the appropriate modification.]*

This sentence is omitted in the revised manuscript to avoid the confusion between slow and fast processes. Subsequent sentence is also modified accordingly.

I115: 'The tropical cyclones are the synoptic-scale disturbances of organised convective systems which weaken the tropopause by overshooting convection.' — this seems a strange characterisation of tropical cyclones. But perhaps you are simply trying to say that one effect of tropical cyclones is to organise convection?

This sentence is revised in this version of manuscript.

I369: 'descend' > 'descent'

Corrected

**I341: 'Numerical simulation shows the presence of stable dry ozone rich stratospheric air in the upper and middle troposphere over the cyclone prone area.' — this seems very misleading to me — as far as I can tell you have no numerical model results including ozone — so the 'ozone-rich' is a guess (based on the fact that the numerical simulations seem to show air that is 'meteorologically' stratospheric. Referee 3 has previously suggested trajectory studies to resolve some of this ambiguity — I accept that carrying out trajectory studies is outside the scope of this paper, but I do strongly recommend some kind of statement at this point of the paper saying something like 'The descent of stratospheric air has been deduced indirectly here from a combination of ozone observations and meteorological observations and modelling. A natural next step to confirm this descent would be to carry out trajectory (or chemical tracer) studies in the WRF model.' — This sort of statement would, in my view, add to the value of your paper rather than diminish it.*

Following the editor's suggestion, we have revised the sentence accordingly in the revised manuscript.

I470: 'planed' > 'planned'

Corrected.

I715: 'Arctic' > Arctic'

Corrected

Reference : Gettelman, A., Forster, P. M. de F., Fujiwara, M., Fu, Q., Vömel, H., Gohar, L. K., Johanson, C., and Ammerman, M., 2004.: Radiation balance of the tropical tropopause layer. Journal of Geophysical Research, 109, D07103, doi: 10.1029/2003JD004190.

Influence of the Tropical Cyclones on Tropospheric Ozone: Possible Implication

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Abstract. The present study examines the role of tropical cyclones in the enhancement of tropospheric ozone. The most significant and new observation reported is the increase in the upper tropospheric (10-16 km) ozone by 20-50 ppbv, which has extended down to the middle (6-10 km) and lower troposphere (< 6 km). The decent rate of enhanced ozone layer during the passage of tropical cyclone is 0.8-1 km/day, which is three times that of a clear-sky day (non-convective). Enhancement of surface ozone concentration by ~ 10 ppbv in day-time and 10-15 ppbv in the night-time is observed during a cyclone. Potential vorticity, vertical velocity and potential temperature obtained from numerical simulation, reproduces the key feature of the observations. A simulation study indicates the downward transport of stratospheric air into the troposphere. Space-borne observations of relative humidity indicate the presence of sporadic dry air in the upper and middle troposphere over the cyclonic region. These observations constitute quantitatively an experimental evidence of redistribution of stratospheric ozone during cyclonic storms.

[Key words: Stratosphere-troposphere exchange processes, tropopause, ozone, water vapour]

1. Introduction

Stratospheric ozone (O_3) layer found around 25-30 km altitude regulates the amount of ultraviolet radiation coming from the Sun to the Earth's surface. Ozone is an important greenhouse gas, which acts as an oxidant in the troposphere and has an important role in the climate forcing (Forster *et al.*, 2007; Pan *et al.*, 2015). One of the major consequences of the tropospheric ozone enhancement is on the living organisms, as it acts as a toxic agent among the air pollutants (National Research Council, 1991). Increase in the tropospheric ozone is considered to be due to (1) in-situ photochemical formation associated with lightning, advection, anthropogenic activities (e.g., Jacobson, 2002 and references therein), and (2) stratospheric flux (Wild, 2007 and reference therein; Skerlak *et al.*, 2014). The tropopause, which acts a barrier between the troposphere and the stratosphere, plays a key role in controlling the flow of minor constituents from one layer to other. The increase of the ozone downward flux from the stratosphere to the troposphere not only increases the tropospheric ozone, but also decreases the stratospheric ozone. The ozone presence in the troposphere (intruded from the stratosphere) will further react with tropospheric water vapour and the tropospheric ozone gets destroyed. In principle, the total columnar ozone decreases and thus there will be an enhancement in the penetration of UV radiation to the Earth's surface.

In general, stratospheric air intrusion into the troposphere is observed over the middle and higher latitudes, which are linked with synoptic scale disturbances (e.g. Stohl *et al.*, 2003). This downward flow is attributed to the dissipation of extra-tropical planetary and gravity waves in the stratosphere (Holton *et al.*, 1995). Stohl *et al.* (2003), and Bourqui and Trepanier (2010) have reported the continuous downward flows from the stratosphere to the troposphere in much small time-scale over extra-tropics. In the global ozone budget, 25-50 % of tropospheric ozone source is from middle latitude stratospheric intrusion (Bourqui and Trepanier, 2010). Appenseller and Davies (1992) have also discussed that exchange between

the stratosphere and the troposphere (both directions) is highly episodic. 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*), the 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; Das, 2009; Das et al., 2011; Zhan and Wang, 2012; Jiang et al., 2015; Venkat Ratnam et al., 2016*). The overshooting convection associated with tropical cyclone can weaken the tropopause stability which plays a key role in the stratosphere-troposphere exchange. In addition, turbulence caused due to wind shear (*Shapiro, 1976*) and breaking of gravity wave (*Langford et al., 1996*) can also be the causative mechanisms for the occurrence of stratospheric intrusion. A recent study by *Pan et al. (2015)* has shown the enhancement of tropospheric ozone associated with the thunderstorm event. Subsidence of stratospheric air is generally observed in the vicinity of cyclone (*Appenzeller and Davies, 1992; Baray et al., 1999; Cairo et al., 2008; Leclair De Bellevue et al., 2006, 2007; Das, 2009; Das et al., 2011; Venkat Ratnam et al., 2016*). Slow stratospheric intrusion is reasonably well understood and is a regular phenomenon, whereas the rapid intrusion needs to be understood in detail.

The increase in surface ozone is also linked with stratospheric intrusion (e.g. *Bourqui and Trepanier, 2010*). Earlier studies have also shown the stratospheric air intrusion into the troposphere is associated with the deep convection by tropopause perturbation using aircraft measurement (*Dickerson et al., 1987; Poulida et al., 1996; Stenchikov et al., 1996; Pan et al., 2015*). *Stohl et al. (2000)* have shown that episodic stratospheric intrusion is associated with severe weather condition which enhanced the surface ozone concentration.

The bands of the tropical cyclone have intense vertical extended cumulus cloud up to UTLS region. These bands of the cloud are accompanied with updrafts, whereas downdrafts

are encounter between these bands. The eyewall region is characterised by local maximum equivalent potential temperature, whereas minimum is found in the middle to upper troposphere. The eyewall and radius of maximum winds increase with height. The low-pressure core extended to UTLS region and the horizontal pressure gradient decreases with height (*Koteswaram*, 1967). *Mitra* (1996) and *Das* (2009) reported the weakening of the tropopause during the passage of tropical cyclone. Detail study on the dynamical and thermodynamical structure of tropical cyclone can be found in *Hence and Houze* (2012) and the review article on clouds in the tropical cyclone can be found in *Houze* (2010). Thus, the tropical cyclones have an influence on stratosphere-troposphere exchange process which causes air mass and energy transports in the troposphere and redistribution of stratospheric ozone (e.g. *Jiang et al.*, 2015). A complete review on the effect of the 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 exchange of air mass from the stratosphere to the lower troposphere in short-time scale associated with tropical cyclones is still unclear and further studies are needed. The present study addresses the influence of the tropical cyclones quantitatively on the enhancement of tropospheric ozone by the stratospheric intrusion.

2. Campaign details and the data analysis

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 (*Pallamraju et al.*, 2014) 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 7 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 Concentration Cell (ECC) (*Komhyr et al.*,

1995). The uncertainty in the ozone measurements is 5-10 %. Table 1 also provides the details of ozonesonde measurements conducted during the passage of these cyclonic storms. Ozonesonde data was obtained at a fixed height resolution by down sampling at 100 m height resolution by the linear interpolation method. The India Meteorological Department (IMD) also launches ozonesonde launches every fortnight. The background profiles (non-convective day at least for 3 days) is constructed by averaging the ozonesonde data (23 profiles) obtained from the IMD combined with our observations from 1995-2013 for the month October over Trivandrum. The IMD-ozonesonde used Brewer bubbler electrochemical sonde developed in the Ozone Research Laboratory of the IMD. These IMD ozone sonde were compared with ECC sondes and found that it is underestimated by 5-10 % in the troposphere (*Kerr et al.*, 1994; *Deshler et al.*, 2008), which is about <2 ppbv of the observed mean value. Detail system description of IMD-Ozonesonde can be found elsewhere (*Sreedharan*, 1968; *Alexander and Chatterjee*, 1980). There is no ozonesonde launch by IMD in this campaign. The measurements of near-surface ozone are carried out using the online UV photometric ozone analyser (Model AC32M) of Environment S.A, France. This ozone analyser works on the principle of UV absorption of ozone at the wavelength 253.7 nm. The instrument has a lower detection limit of 1 ppbv and 1% linearity. The data is sampled at an interval of 5 minutes.

The SAPHIR (Sondeur Atmospherique du Profild' Humidite Intertropical par Radiometrie) on-board Megha-Tropiques satellite is a multichannel passive microwave humidity sounder, measuring brightness temperatures in six channels located close to the 183.31GHz water vapor absorption line (± 0.15 , ± 1.20 , ± 2.80 , ± 4.30 , ± 6.60 and ± 11.0 GHz). These channels allow retrieving the integrated relative humidity respectively between the levels of 1000–850 hPa, 850–700 hPa, 700–550 hPa, 550–400 hPa, 400–250 hPa, and 250–100 hPa. The radiometer has a cross-track scan of $\pm 43^\circ$, providing a swath of 1705 km and a

10 km resolution at nadir. This data is also used for the qualitative analysis of the stratospheric air. The detail instrumentation can be found in *Raju* (2013), and retrieval algorithm and validation can be found in *Gohil et al.* (2012); *Mathur et al.* (2013) and *Venkat Ratnam et al.* (2013); *Subrahmanyam and Kumar* (2013), respectively.

Apart from the 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 the cases of the cyclones. The model domain has been configured with two nested domains of 60 km and 20 km horizontal resolution, and covers an area extending from 1°S to 25°N and 60°E 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° x 0.75° continuously at every 6 hours. 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).

3. Meteorological background

The present experiments were conducted during the passage of the (1) cyclonic storm ‘Nilam’ from 28 October to 1 November 2012 and (2) very severe cyclonic storm ‘Phailin’ from 4-14 October 2013 over the Bay of Bengal (BOB). The track of each tropical cyclones and outgoing long wave radiation (OLR) images (date and time are stamped) are shown in Figures 1a and 1b, respectively. The detailed bulletin can be found in www.imd.gov.in. During these campaigns, several ozonesondes were launched from Trivandrum, whenever the intensity of cyclones is maximum and the path/eye was close to the launching site. The details of each of the tropical cyclone used for present analysis are as follows:

3.1 Case-1 (Nilam)

A depression formed over the southeast of BOB ($\sim 9.5^{\circ}\text{N}$, 86.0°E) at 11:30 IST (IST=UT+5.5h) of 28 October 2012. It moved westwards and intensified into a deep depression on the morning of 29 October 2012 over southwest BOB, about ~ 550 km south-southeast of Chennai. It continued to move westwards and intensified into a Cyclonic Storm, 'Nilam' in the morning of 30 October 2012 over southwest BOB. Then it moved north-northwest, crossed the north Tamilnadu coast near Mahabalipuram (12.6°N , 80.2°E), south of Chennai in the evening hours of 31 October 2012. After the landfall the cyclonic storm, Nilam moved west-northwest and weakened gradually into a deep depression and then into a depression in the morning hours of 1 November 2012.

3.2 Case-2 (Phailin)

A low-pressure system was formed over Tenasserim coast ($\sim 12^{\circ}\text{N}$, 96°E), on the early morning of 6 October 2013. It intensified into a depression over the same region on 8 October and then moved towards the west-north-westwards. It further intensified into a deep depression on the early morning of 9 October 2013 and then into a cyclonic storm, 'Phailin' in the evening hours. Moving north-westwards, it finally converted into a severe cyclonic storm in the morning hours of 10 October 2013 over east central BOB. The very severe cyclonic storm continued to move north-westwards and crossed Andhra Pradesh and Orissa coast near Gopalpur (19.2°N , 84.9°E) in the late evening of 12 October 2013. It further continued to move north-north-westwards after the landfall for some time and then northward and finally north-north-eastwards 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.

4. Results and Discussion

Figure 2 (a-b) shows the profiles of ozone mixing ratio (OMR) and relative humidity (RH) from ozonesonde measurements during the passage of the tropical cyclones Nilam (top panels) and Phailin (bottom panels). The background ozone profile is obtained by averaging individual profile (23 profiles) over Trivandrum of October from 1995-2013 and is shown by dotted lines in Figure 2. During the passage of Nilam on 30 October 2012, enhancement in tropospheric ozone (marked by horizontal arrows) from the 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). These enhancements persisted till 31 October 2012 but at the height region reduced 6-7 km with a reduced width. However, the enhancement of about ~40 ppbv was still observed on 2 November 2012 but the height region decreased to 5-6 km. After two days, we had again observations from 5-7 November 2012. The height of enhanced ozone layer in the troposphere reduced to ~4 km (40 ppbv), ~3 km (30 ppbv) and ~1.5 km (20 ppbv) on 5, 6 and 7 November 2012, respectively. The present observation reveals that the downward propagation of the enhanced upper tropospheric ozone layer into the lower troposphere occurring in an episodic manner. The descent rate of the ozone rich layer from the upper troposphere to the boundary layer during Nilam is approximately estimated to be ~875 m/day. It is also noted that the corresponding RH profiles during Nilam did not decrease with increasing ozone mixing ratio except on 2 November 2012. A significant sudden decrease in RH is observed on 2 November 2012 at ~6 km, where the maximum enhancement (~ 70 ppbv) of the tropospheric ozone layer is observed. This indicates the presence of accumulated dry air at 6 km. As the stratospheric air is dry and ozone rich and thus there may be a possibility that on 2 November 2012 the accumulated dry ozone rich air at 6 km may be of stratospheric origin.

A similar phenomenon is also observed during the passage of Phailin. Intrusion from ~14 km to 6 km (marked by horizontal arrows) is clearly observed in the ozone profiles from 11-

15 October 2013. During Phailin, tropospheric ozone increases by 20-30 ppbv and the width of the enhanced ozone layer is larger than that observed during the Nilam. During Phailin, the descent rate of enhanced ozone layer from the upper troposphere to the boundary layer is estimated to be ~1000 m/day. The descent rate in the tropical non-convective region, under the assumption of no vertical winds, may be inferred from the radiative heating rate in the tropical clear-sky regions. *Gettelman et al.* (2004) estimated tropical clear-sky radiative heating rates by using ozone and water vapour sounding data together with the radiative transfer models and found -1 to -2 K/day in the troposphere. If the temperature lapse rate is 6-10 K/km in the upper troposphere, the descent rate is estimated to be 0.1-0.3 km/day. In the present observations, 0.8-1 km/day descent rate is estimated during the passage of tropical cyclones which is three times that of clear-sky (non-convective) days radiative subsidence. This may indicate that downward flow in association with the tropical cyclones (in their outer regions) enhanced the transport of the ozone from the stratosphere to the lower troposphere.

As discussed in the introductory section, significant perturbation in the tropopause due to deep convection will lead to the transport of ozone-rich stratospheric air into the troposphere. Figure 3 shows variation in the cold point tropopause height (CPT-H) and cold point tropopause temperature (CPT-T) derived from radiosonde measurements during (a) Nilam and (b) Phailin over Trivandrum. Significant perturbation in the tropopause height and the temperature are observed for both the cyclone cases. The climatological mean tropopause height and temperature over southern India (peninsular) are observed to be ~16.5 km and ~191 K (*Sunilkumar et al.*, 2013). The CPT-H gradually decreased from 17.8 km on 30 October to 16.7 km on 2 November 2012 for Nilam. Afterwards, the CPT-H gradually increased and reached to 17.5 km. Similarly for Phailin, the CPT-H decreases from 16.5 km on 11 October 2013 to 15.8 km on 12 October 2013 and then gradually increases. The height

above the tropopause (i.e. stratosphere) is in radiative equilibrium, whereas the height below the tropopause (i.e. troposphere) is in radiative-convective equilibrium.

In addition to the profiling of ozone, we have the surface measurement of ozone and solar flux during the Phailin. Figure 4 shows the time series of near-surface ozone mixing ratio along with solar irradiation from 11 to 19 October 2013. As expected, clear diurnal variability is observed in the time-series of surface ozone. In general, there are three main mechanisms for the production of ozone in the atmospheric boundary layer: (1) photochemical reaction via NO_x and CO channel, (2) Bio-mass burning /fossils fuel, and (3) lightning. However, *David and Nair* (2011) have shown the diurnal pattern of surface ozone observed over Trivandrum is due to the mesoscale circulation, i.e., local sea and land breeze and the availability of NO_x. From 11 to 14 October the maximum and minimum average peak of surface ozone are observed to be 24 and 1 ppbv, respectively, whereas from 14 to 18 October 2013, the maxima and minima are observed to be 35 and 10 ppbv, respectively. Even though there was no solar radiation in the evening hours, there are enhancements in surface ozone concentration (indicated by vertical arrows) on 14-15, 16-17, 18-19 October 2013. The upper and lower average is indicated by horizontal solid and dash lines, respectively. The ozone profiles obtained from ozonesonde measurements also show that enhanced ozone layer propagates downward from the upper troposphere during 11-15 October 2013. There is a possibility that the enhanced tropospheric ozone can further propagate downward to near-surface in the presence of downdrafts. The enhancement in the surface ozone even after the cut-off in solar radiation can be linked to the downward flow of upper tropospheric ozone in the presence of downdrafts. Time-series of solar irradiation shows that there was not much change in the radiation among the days 11-13 and 14-17 October 2013. This indicates that the observed enhancement in the surface ozone is not due to change in the sunshine. Over the observation site, land-breeze prevails during night-time. The change in night-time ozone

depends on the precursor gas (e.g. NO) concentration in land-breeze, which has a dependency on local precursor gas emission/human activity. Due to the cyclonic condition over Trivandrum, change in human activity during 11-17 October 2013 would not have happened considerably and Bio-mass burning may not be possible due to associated rains. The day-to-day variability of surface ozone over Trivandrum is ~ 9.5 ppbv (1-sigma standard deviation). The observed enhancement in surface ozone is found to be ~ 10 ppbv in the day-time and 10-15 ppbv in the night time. In a recent study by *Jiang et al.* (2015), an increase of surface ozone by 21-42 ppbv and surface nocturnal surface ozone levels exceeding 70 ppbv is observed in the region Xiamen and Quanzhou over the south-eastern coast of China before the Typhoon Hagibis landing. However, there are possible of an influence of lightening associated with cyclone and thus, other possibility of this surface ozone cannot be fully ruled out. A planned experiment by setting-up various ground-based instruments is required to rule out the enhancement of surface ozone.

Further, to support the present observations of stratospheric intrusion into the troposphere and further to surface, a dynamical analysis is carried out using WRF-ARW simulations. *Das et al.* (2011) and *Pan et al.* (2015) have shown the ability of WRF simulations during the tropical cyclone. Figure 5 shows the height-time cross-section of (a) vertical velocity along with potential vorticity (magenta line) and potential temperature (black line) contours, and (b) relative humidity along with equivalent potential temperature (black line) and zonal wind (grey line) for Nilam (left panels) and Phailin (right panels) over Trivandrum using WRF simulations. Figure 5 (a) shows the presence of strong updrafts (red) and downdrafts (blue) marked with rectangle box in the UTLS regions. Enhanced potential vorticity of 0.5-1.5 PVU is also observed vertically down from the stratosphere to the troposphere overlapping the downdraft regions. The potential temperature contours indicate (Fig.5 (a)) the presence of reduced stability during 29-31 October 2012 (Nilam) and 9-11 October 2013 (Phailin).

Height-time cross-section of relative humidity shown in Figure 5 (b) indicates presence of dry air from UTLS region to the 2-4 km. The equivalent potential temperature contours in Figure 5 (b) indicate that from the surface to ~8 km it is highly unstable for vertical motion and favourable condition for the convection to take place during 29-31 October 2012 (Nilam) and 9-11 October 2013 (Phailin). During the same periods, from 10 km to the tropopause level, the vertical motion is suppressed and the atmosphere is found to be statically stable to the un-saturated atmosphere. The present condition indicates the presence of statically stable stratospheric air in the upper and middle troposphere. In addition, strong wind shear is also observed in the UTLS region.

Similarly, Figure 6 shows the height-latitude cross-section of (a) vertical velocity along with potential vorticity (magenta line) and potential temperature (black line) contours, and (b) relative humidity cross-section along with equivalent potential temperature (black line) and zonal wind (grey line) at 79°E at 18 GMT on 30 October 2012 for Nilam (left panels) and 18 GMT on 10 October 2013 for Phailin (right panels) using WRF simulations. The vertical velocity profiles shows the presence of downdraft (blue) followed by updraft (red) between 8-17°N in the UTLS region in both the cyclone cases. Enhanced potential vorticity of 0.5-1.5 PVU is also observed vertically down from the stratosphere to the lower troposphere, overlapping the downdraft regions. High potential vorticity in the troposphere is also a signature of stratospheric air in the troposphere. It is also true that enhanced potential vorticity may be also associated with diabatically by condensational heating but the enhancement is only observed with the presence of downdraft in the UTLS region. The potential temperature contours indicate the presence of reduced stability of the atmosphere at this location and noticed that stable stratospheric air penetrated downward at 12-14°N for Nilam and 16-18°N for Phailin. Relative humidity profiles indicate the presence of dry air at ~ 8°N which is in the vicinity of ozonesonde observational site. The equivalent potential

temperature contours in Figure 6 (b) indicate that from the surface to 10 km it is highly unstable for vertical motion and favourable condition for the convection to take place at 6-12°N for Nilam and 12-18°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 un-saturated atmosphere for both Nilam and Phailin. The present condition indicates the presence of statically stable stratospheric air in the upper and middle troposphere in the latitudinal cross-section at 79°E at 18 GMT on 30 October 2012 and 10 October 2013. Numerical simulation reproduced the key features supports the possibility of stratospheric air intrusion into the troposphere during the passage of tropical cyclone.

To get further insight, relative humidity derived from SAPHIR on-board the Megha-Tropiques satellite is used. The relative humidity (daily mean) shown is an average over 12-14 passes per day. Figure 7 shows the height-time intensity plot of daily mean relative humidity during the passage of the cyclones: Nilam (left panel) and Phailin (right panel). The grid is averaged from 4-8°N and 83-88°E. Strong dry air intrusion originated in the lower stratosphere is observed between 23-27 October 2012 (Nilam) and 12-18 October 2013 (Phailin). In both the cyclones, dry air (low humidity region) reached down to the altitude of 8 km. For the perception of the spatial distribution of relative humidity, a latitude-longitude plot of relative humidity averaged over different pressure level is shown in Figure 8. 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 indicate the possibility of dry air present in the troposphere is of stratospheric origin. The present observations show the influence of the tropical cyclone on the air mass exchange from the stratosphere to the lower troposphere and redistribution of stratospheric ozone. Further trajectory and chemical analysis are required to quantify the amount of mass exchange taking place between the stratosphere and the troposphere.

5. Summary and Conclusions

Important results brought out in the present analysis during the passage of a cyclonic storms Nilam (2012) and Phailin (2013) are summarized below:

- a) Increase in the upper tropospheric ozone by 20-50 ppbv from its climatological mean is observed.
- b) The upper tropospheric ozone propagates downwards to the lower troposphere at a rate of 0.8-1 km/day.
- c) About 10 ppbv in the day-time and 10-15 ppbv in the night-time increase in the surface ozone is noticed
- d) Significant variation in the cold-point tropopause altitude and temperature associated with tropical cyclone as that of the climatological mean are noticed.

In the present study, the descent of stratospheric air into the troposphere has been deduced indirectly from a combination of ozone and meteorological observations, and modelling. The study clearly reveals that the cyclones play a vital role in changing the atmospheric composition apart from general weather phenomena.

Acknowledgments

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355 of the manuscript.

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Figure Captions

Figure 1. (a)Track of cyclones Nilam and Phailin (top panels) and (b) its Outgoing Long wave Radiation (OLR) wave radiation at 14:30 GMT on 30 Oct. 2012 (Nilam) and 9:00 GMT on 10 Oct. 2013 (Phailin). In each panel, date and time are mentioned along the track. In the first panel, 18-1/11 indicates 18 GMT of 1 November 2012 and similarly followed for others. The blue star in Fig.1(a) indicates the Ozonesonde launching site Trivandrum.

Figure 2. (a) Profiles of ozone mixing ratio (OMR) (dark black line) and relative humidity (grey line) for individual days during the passage of tropical cyclones (a) Nilam and (b) Phailin. The mean ozone mixing ratio profile for non-convective days (as control day) is shown in dotted line. The mean profile is obtained by averaging ozone data over Trivandrum for the month of October from 1995-2013. Horizontal arrows indicate the height of enhanced ozone.

Figure 3. Variation of cold point tropopause height (CPT-H) and cold point tropopause temperature (CPT-T) derived from temperature measurement by ozonesonde launched during the passage of tropical cyclones (a) Nilam and (b) Phailin over Trivandrum.

Figure 4. Time series of surface ozone mixing ratio (thick line) along with solar radiation (dotted line) from 00 IST on 11 October 2013 to 23:55 IST on 19 October 2013. Solid and dotted horizontal lines indicate the mean maximum and minimum surface ozone. The vertical arrows indicate the nocturnal enhancement of surface ozone. The data is collected with 5 minutes resolution.

Figure 5. Height-time cross-section of (a) vertical velocity along with potential vorticity (magenta line) and potential temperature (black line) contours, and (b) relative humidity along with equivalent potential temperature (black line) and zonal wind (grey line) for Nilam (left panels) over Trivandrum (8.5°N,76.9°E) from 27 October to 2 November 2012 and Phailin (right panels) from 7 to 12 October 2013. Rectangle boxes indicate the presence

of strong updrafts and downdrafts and the dry air between stratosphere and troposphere.

The above parameters are obtained from WRF simulation.

Figure 6. Same as Figure 5 but at 79°E at 18 GMT on 30 October 2012 for Nilam (left panels) and 18 GMT on 10 October 2013 for Phailin (right panels). **Figure 7.** Pressure-time cross-section of relative humidity obtained from SAPHIR onboard Megha-Tropiques satellite during the cyclones Nilam (left panel) from 15 October to 10 November 2012 and Phailin (right panel) from 2 to 22 October 2013. The data is averaged over from 4°N to 8°N and 83°E to 88°E.

Figure 8. Latitude-longitude distribution of relative humidity derived from SAPHIR onboard Megha-Tropiques 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 about 12-14 passes at different timings and arrows indicate the presence of dry air.

Table Caption

Table 1. Details of ozonesonde launched from Trivandrum including the historical data for control day analysis.

Figure 1. (a)Track of cyclones Nilam and Phailin (top panels) and (b) its Outgoing Long wave Radiation (OLR) wave radiation at 14:30 GMT on 30 Oct. 2012 (Nilam) and 9:00 GMT on 10 Oct. 2013 (Phailin). In each panel, date and time are mentioned along the track. In the first panel, 18-1/11 indicates 18 GMT of 1 November 2012 and similarly followed for others. The blue star in Fig.1(a) indicates the Ozonesonde launching site Trivandrum.

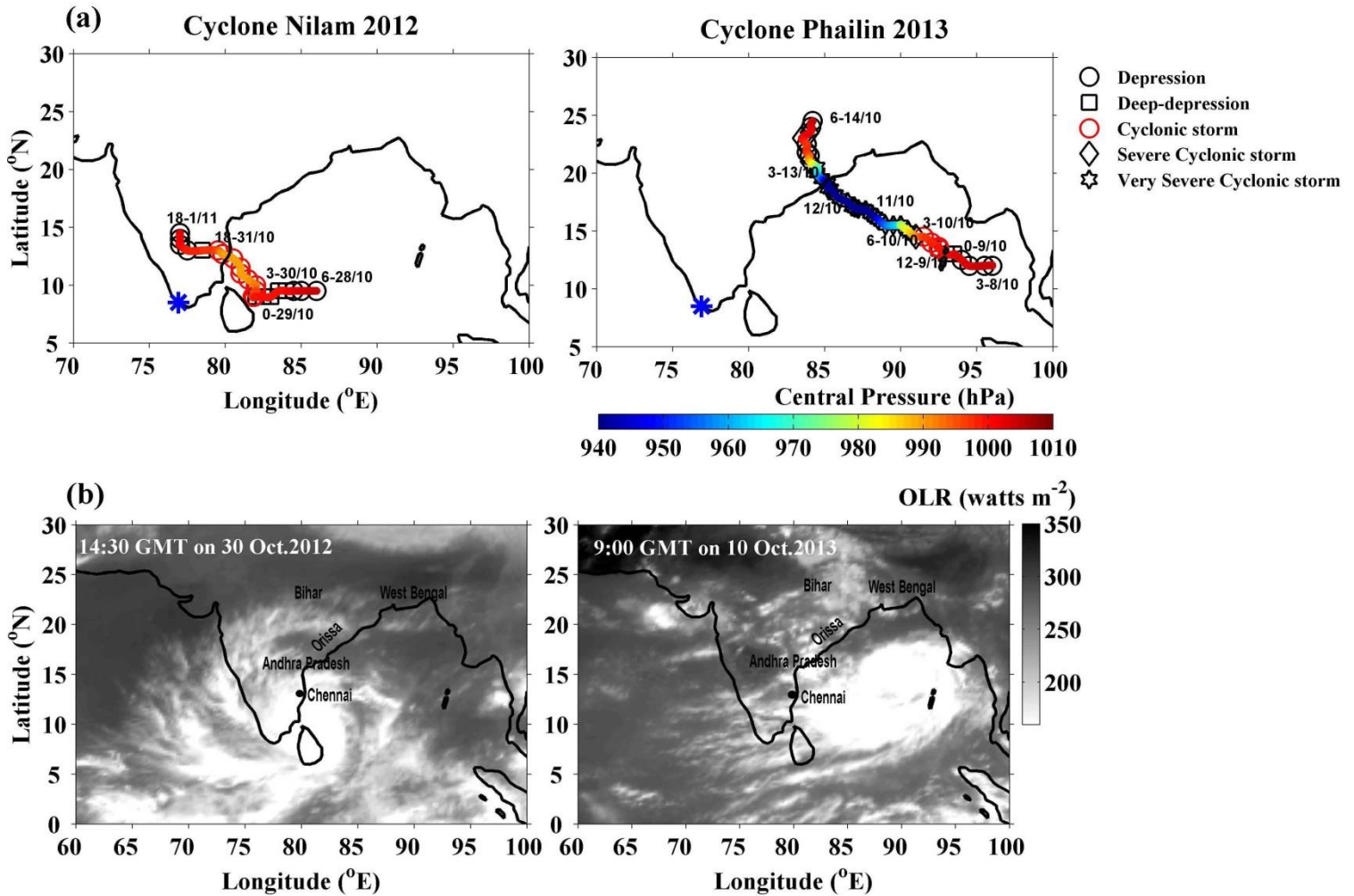


Figure 2. (a) Profiles of ozone mixing ratio (OMR) (dark black line) and relative humidity (grey line) for individual days during the passage of tropical cyclones (a) Nilam and (b) Phailin. The mean ozone mixing ratio profile for non-convective days (as control day) is shown in dotted line. The mean profile is obtained by averaging ozone data over Trivandrum for the month of October from 1995-2013. Horizontal arrows indicate the height of enhanced ozone.

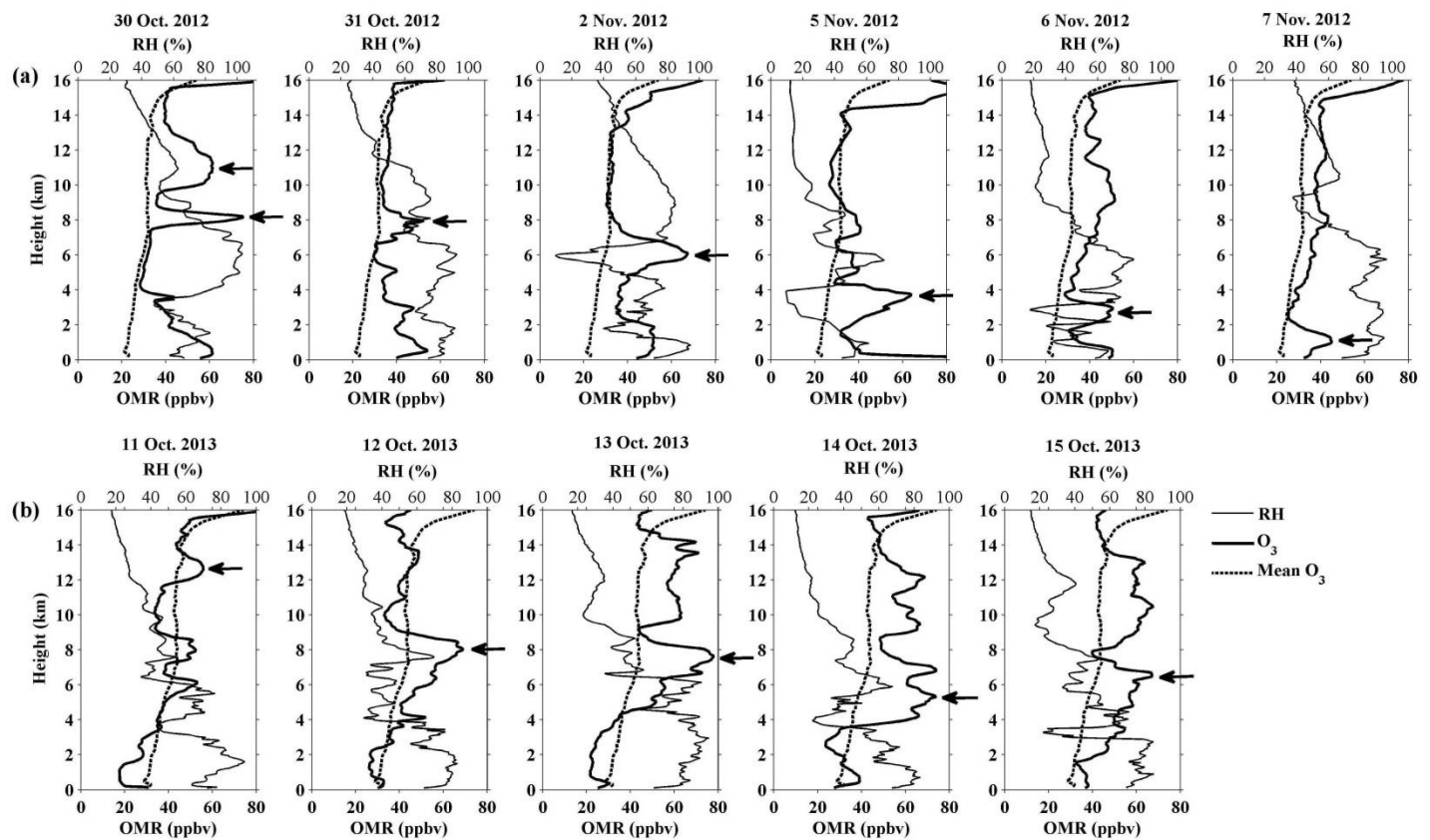


Figure 3. Variation of cold point tropopause height (CPT-H) and cold point tropopause temperature (CPT-T) derived from temperature measurement by ozonesonde launched during the passage of tropical cyclones (a) Nilam and (b) Phailin over Trivandrum.

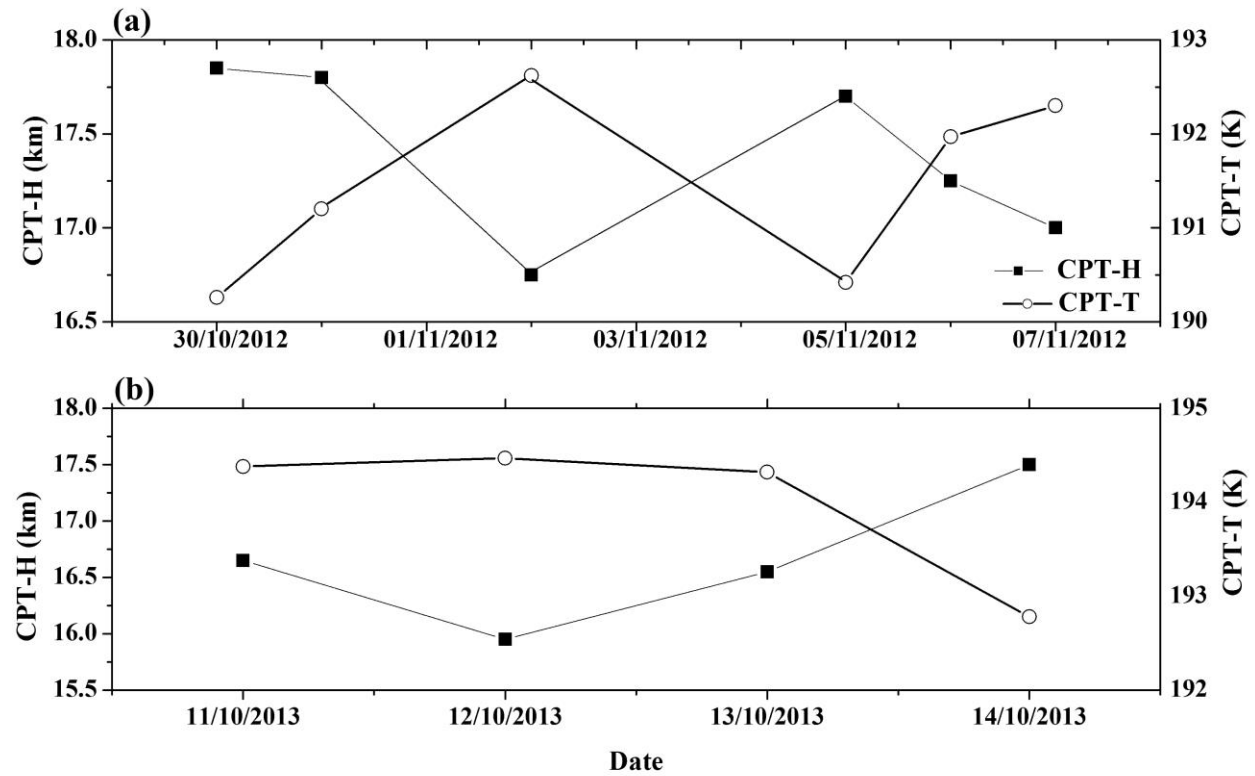


Figure 4. Time series of surface ozone mixing ratio along with solar radiation from 00 IST on 11 October 2013 to 23:55 IST on 19 October 2013. Solid and dotted horizontal lines indicate the mean maximum and minimum surface ozone. The vertical arrows indicate the nocturnal enhancement of surface ozone. The data is collected every 5 min.

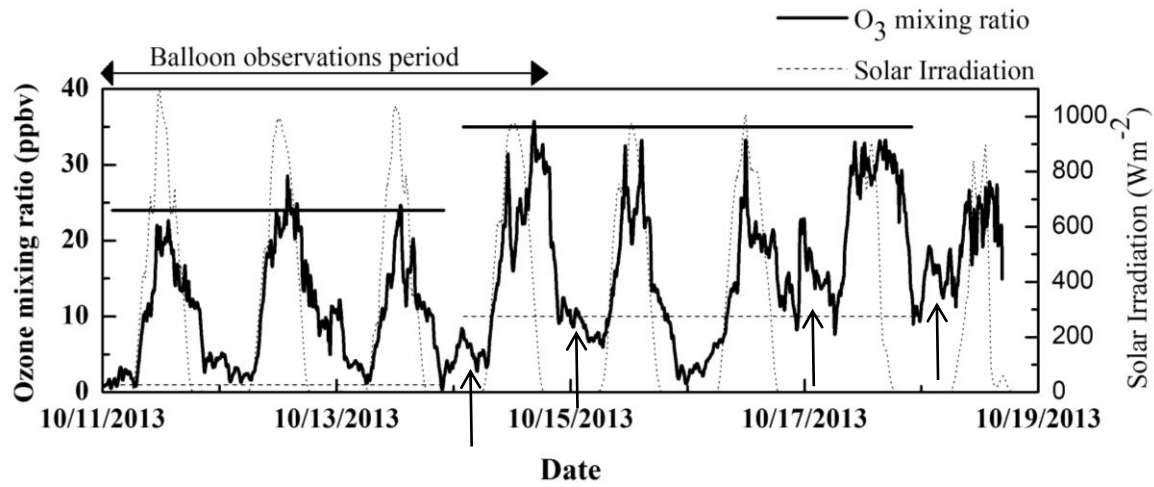


Figure 5. Height-time cross-section of (a) vertical velocity along with potential vorticity (magenta line) and potential temperature (black line) contours, and (b) relative humidity along with equivalent potential temperature (black line) and zonal wind (grey line) for Nilam (left panels) over Trivandrum (8.5°N,76.9°E) from 27 October to 2 November 2012 and Phailin (right panels) from 7 to 12 October 2013. Rectangle boxes indicate the presence of strong updrafts and downdrafts and the dry air between stratosphere and troposphere. The above parameters are obtained from WRF simulation.

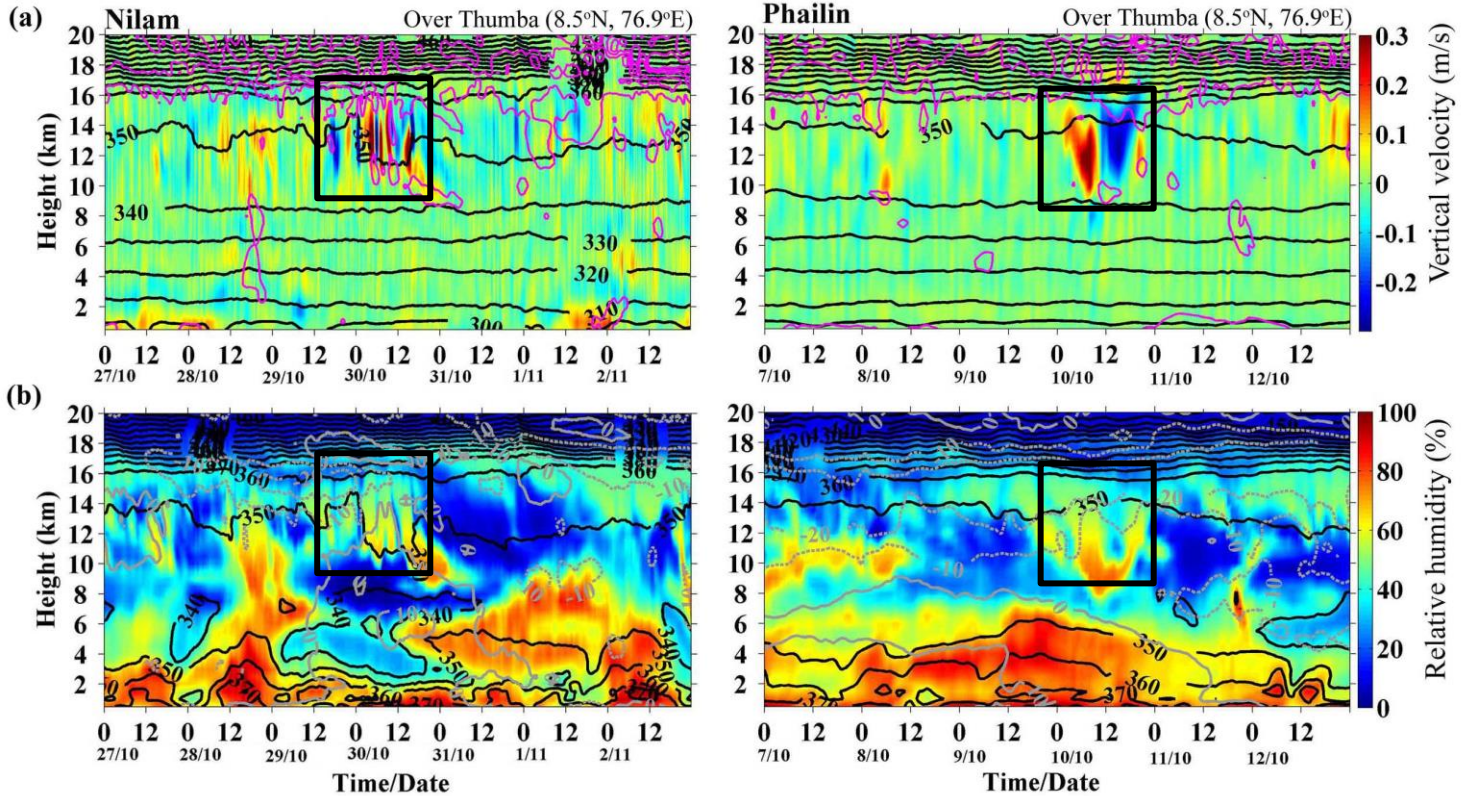


Figure 6. Height-latitude cross-section of (a) vertical velocity along with potential vorticity (magenta line) and potential temperature (black line) contours, and (b) relative humidity cross-section along with equivalent potential temperature (black line) and zonal wind (grey line) at 79°E at 18 GMT on 30 October 2012 for Nilam (left panels) and 18 GMT on 10 October 2013 for Phailin (right panels). The above parameters are obtained from WRF simulation.

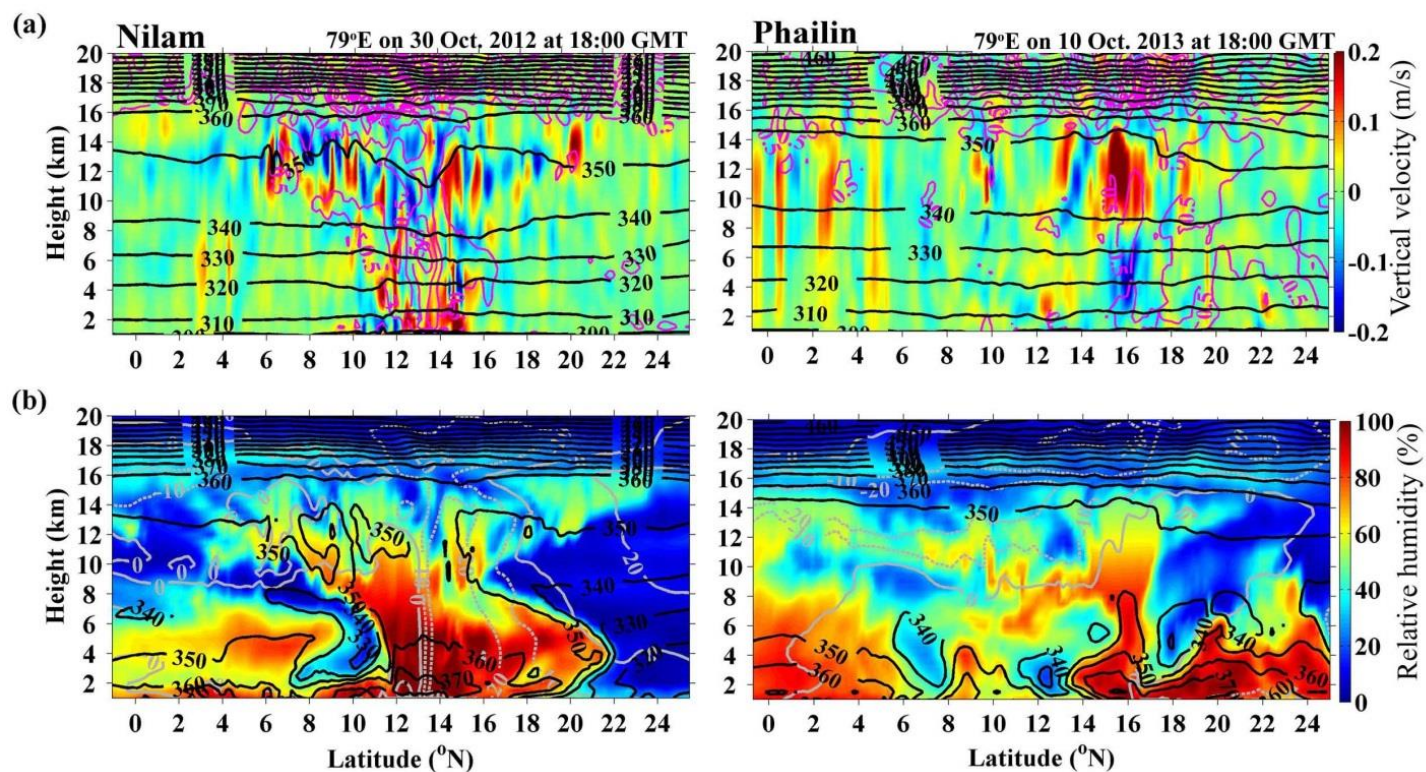


Figure 7. Pressure-time cross-section of relative humidity obtained from SAPHIR onboard Megha-Tropiques satellite during the cyclones Nilam (left panel) from 15 October to 10 November 2012 and Phailin (right panel) from 2 to 22 October 2013. The data is averaged over from 4°N to 8°N and 83°E to 88°E.

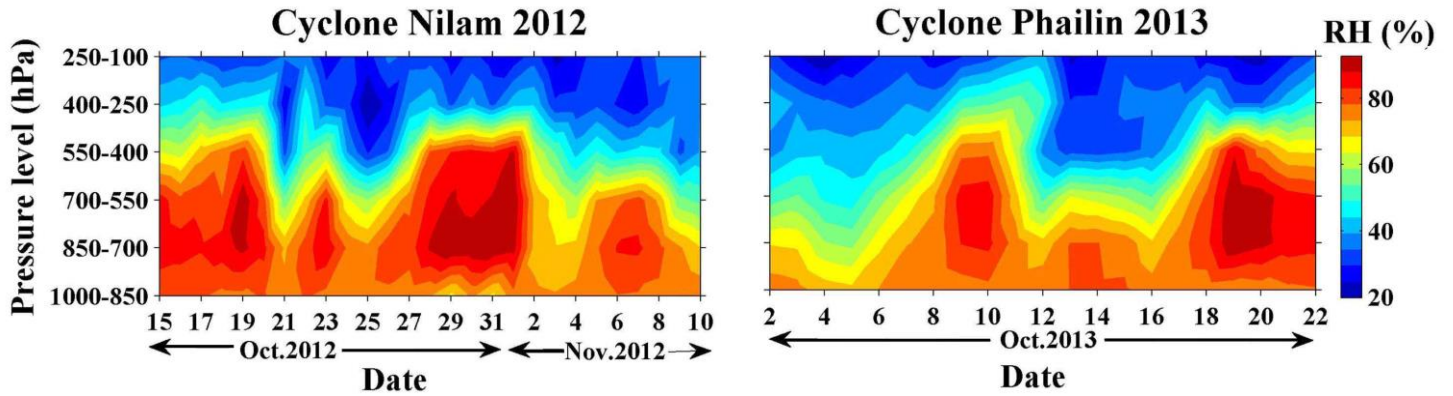


Figure 8. Latitude-longitude distribution of relative humidity derived from SAPHIR onboard Megha-Tropiques at different pressure levels (stamped on each panel) for Nilam (25 October 2012) and Phailin (14 October 2013). The data is averaged for one day which is about 12-14 passes at different timings and arrows indicate the presence of dry air.

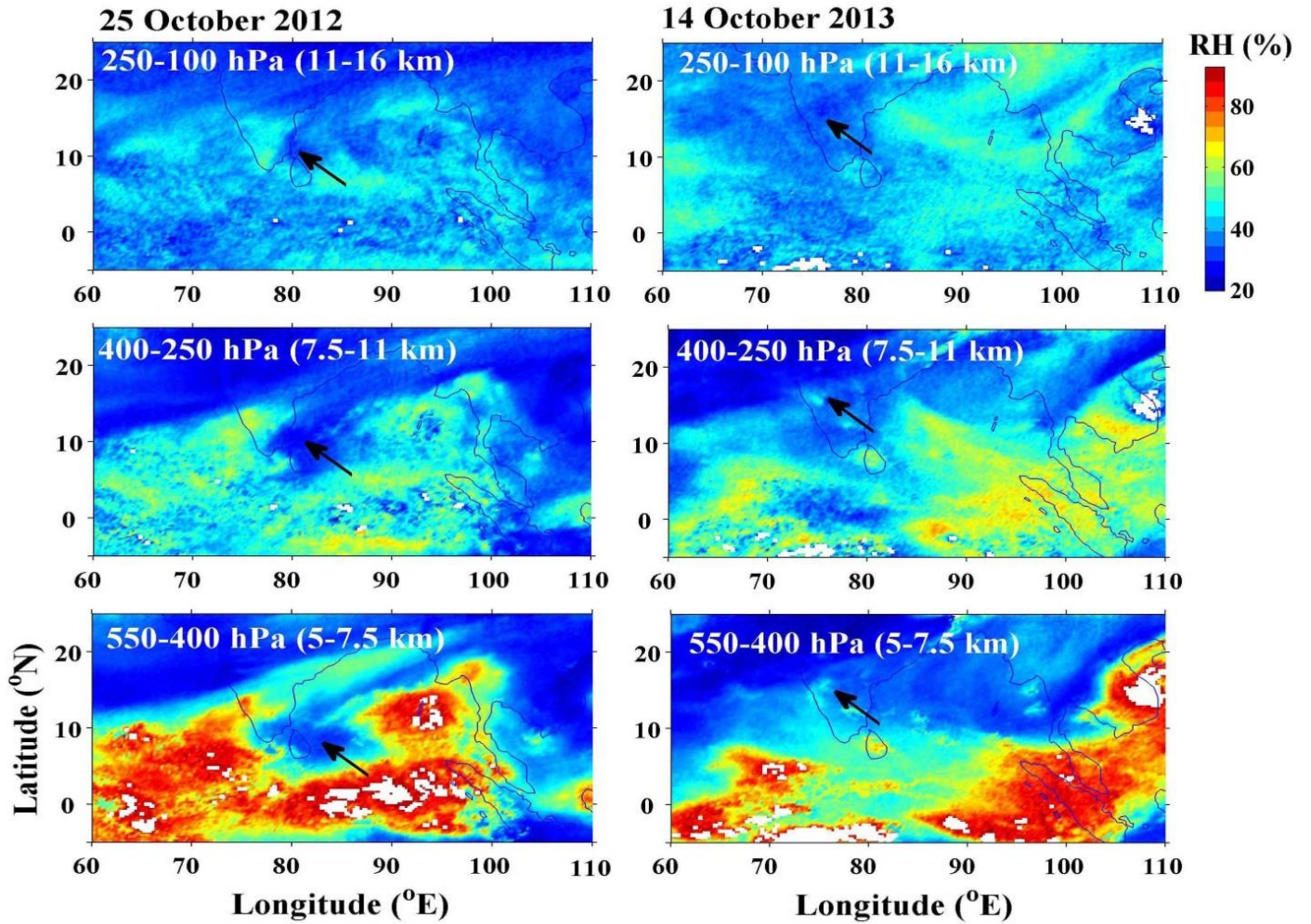


Table 1. Details of ozonesonde launched from Trivandrum including the historical data for control day analysis.

Description	Date
Cyclone Nilam	30 Oct. 2012
	31 Oct. 2012
	2 Nov. 2012
	5 Nov. 2012
	6 Nov. 2012
	7 Nov. 2012
	11 Oct. 2013
Cyclone Phailin	12 Oct. 2013
	13 Oct. 2013
	14 Oct. 2013
	15 Oct. 2013
	24 Oct. 1995
Control Days	25 Oct. 1995
	7 Oct. 1998
	21 Oct. 1998
	4 Oct. 2000
	4 Oct. 2002
	1 Oct. 2003
	15 Oct. 2003
	30 Oct. 2003
	27 Oct. 2004
	28 Sep. 2005
	25 Oct. 2006
	7 Oct. 2009
	12 Oct. 2011
	13 Oct. 2011
	14 Oct. 2011
	19 Oct. 2011
	27 Oct. 2011
	3 Oct. 2012
	14 Oct. 2012
	28 Oct. 2013
	29 Oct. 2013

Influence of the Tropical Cyclones on Tropospheric Ozone: Possible Implication

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Abstract. The present study examines the role of tropical cyclones in the enhancement of tropospheric ozone. The most significant and new observation reported is the increase in the upper tropospheric (10-16 km) ozone by 20-50 ppbv, which has extended down to the middle (6-10 km) and lower troposphere (< 6 km). The decent rate of enhanced ozone layer during the passage of tropical cyclone is 0.8-1 km/day, which is three times that of a clear-sky day (non-convective). Enhancement of surface ozone concentration by ~ 10 ppbv in day-time and 10-15 ppbv in the night-time is observed during a cyclone. Potential vorticity, vertical velocity and potential temperature obtained from numerical simulation, reproduces the key feature of the observations. A simulation study indicates the downward transport of stratospheric air into the troposphere. Space-borne observations of relative humidity indicate the presence of sporadic dry air in the upper and middle troposphere over the cyclonic region. These observations constitute quantitatively an experimental evidence of redistribution of stratospheric ozone during cyclonic storms.

[Key words: Stratosphere-troposphere exchange processes, tropopause, ozone, water vapour]

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39 1. Introduction

40 Stratospheric ozone (O₃) layer found around 25-30 km altitude regulates the amount
41 of ultraviolet radiation coming from the Sun to the Earth's surface. Ozone is an important
42 greenhouse gas, which acts as an oxidant in the troposphere and has an important role in the
43 climate forcing (*Forster et al.*, 2007; *Pan et al.*, 2015). One of the major consequences of the
44 tropospheric ozone enhancement is on the living organisms, as it acts as a toxic agent among
45 the air pollutants (*National Research Council*, 1991). Increase in the tropospheric ozone is
46 considered to be due to (1) in-situ photochemical formation associated with lightning,
47 advection, anthropogenic activities (e.g., *Jacobson*, 2002 and references therein), and (2)
48 stratospheric flux (*Wild*, 2007 and reference therein; *Skerlak et al.*, 2014). The tropopause,
49 which acts a barrier between the troposphere and the stratosphere, plays a key role in
50 controlling the flow of minor constituents from one layer to other. The increase of the ozone
51 downward flux from the stratosphere to the troposphere not only increases the tropospheric
52 ozone, but also decreases the stratospheric ozone. The ozone presence in the troposphere
53 (intruded from the stratosphere) will further react with tropospheric water vapour and the
54 tropospheric ozone gets destroyed. In principle, the total columnar ozone decreases and thus
55 there will be an enhancement in the penetration of UV radiation to the Earth's surface.

56 In general, stratospheric air intrusion into the troposphere is observed over the middle
57 and higher latitudes, which are linked with synoptic scale disturbances (e.g. *Stohl et al.*,
58 2003). This downward flow is attributed to the dissipation of extra-tropical planetary and
59 gravity waves in the stratosphere (*Holton et al.*, 1995). *Stohl et al. (2003), and Bourqui and*
60 *Trepanier (2010) have reported the* continuous downward flows from the stratosphere to the
61 troposphere in much small time-scale over extra-tropics. In the global ozone budget, 25-50 %
62 of tropospheric ozone source is from middle latitude stratospheric intrusion (*Bourqui and*
63 *Trepanier*, 2010). *Appenseller and Davies* (1992) have also discussed that exchange between

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Deleted: (*Stohl et al.*, 2003; *Bourqui and Trepanier*, 2010)

the stratosphere and the troposphere (both directions) is highly episodic. 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), the 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; Das, 2009; Das et al., 2011; Zhan and Wang, 2012; Jiang et al., 2015; Venkat Ratnam et al., 2016). The overshooting convection associated with tropical cyclone can weaken the tropopause stability which plays a key role in the stratosphere-troposphere exchange. In addition, turbulence caused due to wind shear (Shapiro, 1976) and breaking of gravity wave (Langford et al., 1996) can also be the causative mechanisms for the occurrence of stratospheric intrusion. A recent study by Pan et al. (2015) has shown the enhancement of tropospheric ozone associated with the thunderstorm event. Subsidence of stratospheric air is generally observed in the vicinity of cyclone (Appenzeller and Davies, 1992; Baray et al., 1999; Cairo et al., 2008; Leclair De Bellevue et al., 2006, 2007; Das, 2009; Das et al., 2011; Venkat Ratnam et al., 2016). Slow stratospheric intrusion is reasonably well understood and is a regular phenomenon, whereas the rapid intrusion needs to be understood in detail.

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The increase in surface ozone is also linked with stratospheric intrusion (e.g. Bourqui and Trepanier, 2010). Earlier studies have also shown the stratospheric air intrusion into the troposphere is associated with the deep convection by tropopause perturbation using aircraft measurement (Dickerson et al., 1987; Poulida et al., 1996; Stenchikov et al., 1996; Pan et al., 2015). Stohl et al. (2000) have shown that episodic stratospheric intrusion is associated with severe weather condition which enhanced the surface ozone concentration.

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The bands of the tropical cyclone have intense vertical extended cumulus cloud up to UTLS region. These bands of the cloud are accompanied with updrafts, whereas downdrafts

are encounter between these bands. The eyewall region is characterised by local maximum equivalent potential temperature, whereas minimum is found in the middle to upper troposphere. The eyewall and radius of maximum winds increase with height. The low-pressure core extended to UTLS region and the horizontal pressure gradient decreases with height (*Koteswaram*, 1967). *Mitra* (1996) and *Das* (2009) reported the weakening of the tropopause during the passage of tropical cyclone. Detail study on the dynamical and thermodynamical structure of tropical cyclone can be found in *Hence and Houze* (2012) and the review article on clouds in the tropical cyclone can be found in *Houze* (2010). Thus, the tropical cyclones have an influence on stratosphere-troposphere exchange process which causes air mass and energy transports in the troposphere and redistribution of stratospheric ozone (e.g. *Jiang et al.*, 2015). A complete review on the effect of the 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 exchange of air mass from the stratosphere to the lower troposphere in short-time scale associated with tropical cyclones is still unclear and further studies are needed. The present study addresses the influence of the tropical cyclones quantitatively on the enhancement of tropospheric ozone by the stratospheric intrusion.

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2. Campaign details and the data analysis

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 (*Pallamraju et al.*, 2014) 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 7 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 Concentration Cell (ECC) (*Komhyr et al.*,

133 1995). The uncertainty in the ozone measurements is 5-10 %. Table 1 also provides the
134 details of ozonesonde measurements conducted during the passage of these cyclonic storms.
135 Ozonesonde data was obtained at a fixed height resolution by down sampling at 100 m height
136 resolution by the linear interpolation method. The India Meteorological Department (IMD)
137 also launches ozonesonde launches every fortnight. The background profiles (non-convective
138 day at least for 3 days) is constructed by averaging the ozonesonde data (23 profiles) obtained
139 from the IMD combined with our observations from 1995-2013 for the month October over
140 Trivandrum. The IMD-ozonesonde used Brewer bubbler electrochemical sonde developed in
141 the Ozone Research Laboratory of the IMD. These IMD ozone sonde were compared with
142 ECC sondes and found that it is underestimated by 5-10 % in the troposphere (*Kerr et al.*,
143 1994; *Deshler et al.*, 2008), which is about <2 ppbv of the observed mean value. Detail
144 system description of IMD-Ozonesonde can be found elsewhere (*Sreedharan*, 1968;
145 *Alexander and Chatterjee*, 1980). There is no ozonesonde launch by IMD in this campaign.
146 The measurements of near-surface ozone are carried out using the online UV photometric
147 ozone analyser (Model AC32M) of Environment S.A, France. This ozone analyser works on
148 the principle of UV absorption of ozone at the wavelength 253.7 nm. The instrument has a
149 lower detection limit of 1 ppbv and 1% linearity. The data is sampled at an interval of 5
150 minutes.

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151 The SAPHIR (Sondeur Atmospherique du Profild' Humidite Intertropical par
152 Radiometrie) on-board Megha-Tropiques satellite is a multichannel passive microwave
153 humidity sounder, measuring brightness temperatures in six channels located close to the
154 183.31GHz water vapor absorption line (± 0.15 , ± 1.20 , ± 2.80 , ± 4.30 , ± 6.60 and ± 11.0 GHz).
155 These channels allow retrieving the integrated relative humidity respectively between the
156 levels of 1000–850 hPa, 850–700 hPa, 700–550 hPa, 550–400 hPa, 400–250 hPa, and 250–
157 100 hPa. The radiometer has a cross-track scan of $\pm 43^\circ$, providing a swath of 1705 km and a

159 10 km resolution at nadir. This data is also used for the qualitative analysis of the
160 stratospheric air. The detail instrumentation can be found in *Raju* (2013), and retrieval
161 algorithm and validation can be found in *Gohil et al.* (2012); *Mathur et al.* (2013) and *Venkat*
162 *Ratnam et al.* (2013); *Subrahmanyam and Kumar* (2013), respectively.

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

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175 3. Meteorological background

176 The present experiments were conducted during the passage of the (1) cyclonic storm
177 ‘Nilam’ from 28 October to 1 November 2012 and (2) very severe cyclonic storm ‘Phailin’
178 from 4-14 October 2013 over the Bay of Bengal (BOB). The track of each tropical cyclones
179 and outgoing long wave radiation (OLR) images (date and time are stamped) are shown in
180 Figures 1a and 1b, respectively. The detailed bulletin can be found in www.imd.gov.in.
181 During these campaigns, several ozonesondes were launched from Trivandrum, whenever the
182 intensity of cyclones is maximum and the path/eye was close to the launching site. The
183 details of each of the tropical cyclone used for present analysis are as follows:

186 3.1 Case-1 (Nilam)

187 A depression formed over the southeast of BOB ($\sim 9.5^{\circ}\text{N}$, 86.0°E) at 11:30 IST
188 (IST=UT+5.5h) of 28 October 2012. It moved westwards and intensified into a deep-
189 depression on the morning of 29 October 2012 over southwest BOB, about ~ 550 km south-
190 southeast of Chennai. It continued to move westwards and intensified into a Cyclonic Storm,
191 'Nilam' in the morning of 30 October 2012 over southwest BOB. Then it moved north-
192 northwest, crossed the north Tamilnadu coast near Mahabalipuram (12.6°N , 80.2°E), south of
193 Chennai in the evening hours of 31 October 2012. After the landfall the cyclonic storm, Nilam
194 moved west-northwest and weakened gradually into a deep depression and then into a
195 depression in the morning hours of 1 November 2012.

196 3.2 Case-2 (Phailin)

197 A low-pressure system was formed over Tenasserim coast ($\sim 12.0^{\circ}\text{N}$, 96°E), on the early
198 morning of 6 October 2013. It intensified into a depression over the same region on 8
199 October and then moved towards the west-north-westwards. It further intensified into a deep
200 depression on the early morning of 9 October 2013 and then into a cyclonic storm, 'Phailin'
201 in the evening hours. Moving north-westwards, it finally converted into a severe cyclonic
202 storm in the morning hours of 10 October 2013 over east central BOB. The very severe
203 cyclonic storm continued to move north-westwards and crossed Andhra Pradesh and Orissa
204 coast near Gopalpur (19.2°N , 84.9°E) in the late evening of 12 October 2013. It further
205 continued to move north-north-westwards after the landfall for some time and then northward
206 and finally north-north-eastwards up to southwest Bihar. The system weakened gradually into
207 a cyclonic storm from 13 October 2013 and finally the intensity decreased to a low-pressure
208 system on 14 October 2013.

209 4. Results and Discussion

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Figure 2 (a-b) shows the profiles of ozone mixing ratio (OMR) and relative humidity (RH) from ozonesonde measurements during the passage of the tropical cyclones Nilam (top panels) and Phailin (bottom panels). The background ozone profile is obtained by averaging individual profile (23 profiles) over Trivandrum of October from 1995-2013 and is shown by dotted lines in Figure 2. During the passage of Nilam on 30 October 2012, enhancement in tropospheric ozone (marked by horizontal arrows) from the 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). These enhancements persisted till 31 October 2012 but at the height region reduced 6-7 km with a reduced width. However, the enhancement of about ~40 ppbv was still observed on 2 November 2012 but the height region decreased to 5-6 km. After two days, we had again observations from 5-7 November 2012. The height of enhanced ozone layer in the troposphere reduced to ~4 km (40 ppbv), ~3 km (30 ppbv) and ~1.5 km (20 ppbv) on 5, 6 and 7 November 2012, respectively. The present observation reveals that the downward propagation of the enhanced upper tropospheric ozone layer into the lower troposphere occurring in an episodic manner. The descent rate of the ozone rich layer from the upper troposphere to the boundary layer during Nilam is approximately estimated to be ~875 m/day. It is also noted that the corresponding RH profiles during Nilam did not decrease with increasing ozone mixing ratio except on 2 November 2012. A significant sudden decrease in RH is observed on 2 November 2012 at ~6 km, where the maximum enhancement (~ 70 ppbv) of the tropospheric ozone layer is observed. This indicates the presence of accumulated dry air at 6 km. As the stratospheric air is dry and ozone rich and thus there may be a possibility that on 2 November 2012 the accumulated dry ozone rich air at 6 km may be of stratospheric origin.

A similar phenomenon is also observed during the passage of Phailin. Intrusion from ~14 km to 6 km (marked by horizontal arrows) is clearly observed in the ozone profiles from 11-

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239 15 October 2013. During Phailin, tropospheric ozone increases by 20-30 ppbv and the width
 240 of the enhanced ozone layer is larger than that observed during the Nilam. During Phailin, the
 241 descent rate of enhanced ozone layer from the upper troposphere to the boundary layer is
 242 estimated to be ~1000 m/day. The descent rate in the tropical non-convective region, under
 243 the assumption of no vertical winds, may be inferred from the radiative heating rate in the
 244 tropical clear-sky regions. Gettelman *et al.* (2004) estimated tropical clear-sky radiative
 245 heating rates by using ozone and water vapour sounding data together with the radiative
 246 transfer models and found -1 to -2 K/day in the troposphere. If the temperature lapse rate is 6-
 247 10 K/km in the upper troposphere, the descent rate is estimated to be 0.1-0.3 km/day. In the
 248 present observations, 0.8-1 km/day descent rate is estimated during the passage of tropical
 249 cyclones which is three times that of clear-sky (non-convective) days radiative subsidence.
 250 This may indicate that downward flow in association with the tropical cyclones (in their outer
 251 regions) enhanced the transport of the ozone from the stratosphere to the lower troposphere.

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252 As discussed in the introductory section, significant perturbation in the tropopause
 253 due to deep convection will lead to the transport of ozone-rich stratospheric air into the
 254 troposphere. Figure 3 shows variation in the cold point tropopause height (CPT-H) and cold
 255 point tropopause temperature (CPT-T) derived from radiosonde measurements during (a)
 256 Nilam and (b) Phailin over Trivandrum. Significant perturbation in the tropopause height and
 257 the temperature are observed for both the cyclone cases. The climatological mean tropopause
 258 height and temperature over southern India (peninsular) are observed to be ~16.5 km and ~
 259 191 K (Sunilkumar *et al.*, 2013). The CPT-H gradually decreased from 17.8 km on 30
 260 October to 16.7 km on 2 November 2012 for Nilam. Afterwards, the CPT-H gradually
 261 increased and reached to 17.5 km. Similarly for Phailin, the CPT-H decreases from 16.5 km
 262 on 11 October 2013 to 15.8 km on 12 October 2013 and then gradually increases. The height

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270 above the tropopause (i.e. stratosphere) is in radiative equilibrium, whereas the height below
271 the tropopause (i.e. troposphere) is in radiative-convective equilibrium.

272 | In addition to the profiling of ozone, we have the surface measurement of ozone and
273 solar flux during the Phailin. Figure 4 shows the time series of near-surface ozone mixing
274 ratio along with solar irradiation from 11 to 19 October 2013. As expected, clear diurnal
275 | variability is observed in the time-series of surface ozone. In general, there are three main
276 mechanisms for the production of ozone in the atmospheric boundary layer: (1)
277 photochemical reaction via NO_x and CO channel, (2) Bio-mass burning /fossils fuel, and (3)
278 lightning. However, *David and Nair* (2011) have shown the diurnal pattern of surface ozone
279 observed over Trivandrum is due to the mesoscale circulation, i.e., local sea and land breeze
280 and the availability of NO_x. From 11 to 14 October the maximum and minimum average
281 peak of surface ozone are observed to be 24 and 1 ppbv, respectively, whereas from 14 to 18
282 | October 2013, the maxima and minima are observed to be 35 and 10 ppbv, respectively. Even
283 though there was no solar radiation in the evening hours, there are enhancements in surface
284 ozone concentration (indicated by vertical arrows) on 14-15, 16-17, 18-19 October 2013. The
285 upper and lower average is indicated by horizontal solid and dash lines, respectively. The
286 ozone profiles obtained from ozonesonde measurements also show that enhanced ozone layer
287 propagates downward from the upper troposphere during 11-15 October 2013. There is a
288 | possibility that the enhanced tropospheric ozone can further propagate downward to near-
289 surface in the presence of downdrafts. The enhancement in the surface ozone even after the
290 cut-off in solar radiation can be linked to the downward flow of upper tropospheric ozone in
291 | the presence of downdrafts. Time-series of solar irradiation shows that there was not much
292 change in the radiation among the days 11-13 and 14-17 October 2013. This indicates that the
293 observed enhancement in the surface ozone is not due to change in the sunshine. Over the
294 observation site, land-breeze prevails during night-time. The change in night-time ozone

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298 depends on the precursor gas (e.g. NO) concentration in land-breeze, which has a dependency
 299 on local precursor gas emission/human activity. Due to the cyclonic condition over
 300 Trivandrum, change in human activity during 11-17 October 2013 would not have happened
 301 considerably and Bio-mass burning may not be possible due to associated rains. The day-to-
 302 day variability of surface ozone over Trivandrum is ~ 9.5 ppbv (1-sigma standard deviation).
 303 The observed enhancement ~~in surface~~ ozone is found to be ~10 ppbv in the day-time and 10-
 304 15 ppbv in the night time. ~~In a recent study by Jiang et al. (2015), an~~ increase of surface
 305 ozone by 21-42 ppbv and surface nocturnal surface ozone levels exceeding 70 ppbv is
 306 observed in the region Xiamen and Quanzhou over the south-eastern coast of China before
 307 the Typhoon Hagibis landing. However, there are possible of an influence of lightening
 308 associated with cyclone and thus, other possibility of this surface ozone cannot be fully ruled
 309 out. A ~~planned~~ experiment by setting-up various ground-based instruments is required to rule
 310 out the enhancement of surface ozone.

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311 Further, to support the present observations of stratospheric intrusion into the troposphere
 312 and further to surface, a dynamical analysis is carried out using WRF-ARW simulations. Das
 313 et al. (2011) and Pan et al. (2015) have shown the ability of WRF simulations during the
 314 tropical cyclone. Figure 5 shows the height-time cross-section of (a) vertical velocity along
 315 with potential vorticity (magenta line) and potential temperature (black line) contours, and (b)
 316 relative humidity along with equivalent potential temperature (black line) and zonal wind
 317 (grey line) for Nilam (left panels) and Phailin (right panels) over Trivandrum using WRF
 318 simulations. Figure 5 (a) shows the presence of strong updrafts (red) and downdrafts (blue)
 319 marked with rectangle box in the UTLS regions. Enhanced potential vorticity of 0.5-1.5
 320 PVU is also observed vertically down from the stratosphere to the troposphere overlapping
 321 the downdraft regions. The potential temperature contours indicate (Fig.5 (a)) the presence of
 322 reduced stability during 29-31 October 2012 (Nilam) and 9-11 October 2013 (Phailin).

Height-time cross-section of relative humidity shown in Figure 5 (b) indicates presence of dry air from UTLS region to the 2-4 km. The equivalent potential temperature contours in Figure 5 (b) indicate that from the surface to ~8 km it is highly unstable for vertical motion and favourable condition for the convection to take place during 29-31 October 2012 (Nilam) and 9-11 October 2013 (Phailin). During the same periods, from 10 km to the tropopause level, the vertical motion is suppressed and the atmosphere is found to be statically stable to the un-saturated atmosphere. The present condition indicates the presence of statically stable stratospheric air in the upper and middle troposphere. In addition, strong wind shear is also observed in the UTLS region.

Similarly, Figure 6 shows the height-latitude cross-section of (a) vertical velocity along with potential vorticity (magenta line) and potential temperature (black line) contours, and (b) relative humidity cross-section along with equivalent potential temperature (black line) and zonal wind (grey line) at 79°E at 18 GMT on 30 October 2012 for Nilam (left panels) and 18 GMT on 10 October 2013 for Phailin (right panels) using WRF simulations. The vertical velocity profiles shows the presence of downdraft (blue) followed by updraft (red) between 8-17°N in the UTLS region in both the cyclone cases. Enhanced potential vorticity of 0.5-1.5 PVU is also observed vertically down from the stratosphere to the lower troposphere, overlapping the downdraft regions. High potential vorticity in the troposphere is also a signature of stratospheric air in the troposphere. It is also true that enhanced potential vorticity may be also associated with diabatically by condensational heating but the enhancement is only observed with the presence of downdraft in the UTLS region. The potential temperature contours indicate the presence of reduced stability of the atmosphere at this location and noticed that stable stratospheric air penetrated downward at 12-14°N for Nilam and 16-18°N for Phailin. Relative humidity profiles indicate the presence of dry air at ~ 8°N which is in the vicinity of ozonesonde observational site. The equivalent potential

352 | temperature contours in Figure 6 (b) indicate that from the surface to 10 km it is highly
353 | unstable for vertical motion and favourable condition for the convection to take place at 6-
354 | 12°N for Nilam and 12-18°N for Phailin. In the same latitude regions from 10 km to the
355 | tropopause level, the vertical motion is suppressed and the atmosphere is found to be
356 | statically stable to the un-saturated atmosphere for both Nilam and Phailin. The present
357 | condition indicates the presence of statically stable stratospheric air in the upper and middle
358 | troposphere in the latitudinal cross-section at 79°E at 18 GMT on 30 October 2012 and 10
359 | October 2013. Numerical simulation reproduced the key features supports the possibility of
360 | stratospheric air intrusion into the troposphere during the passage of tropical cyclone.

361 | To get further insight, relative humidity derived from SAPHIR on-board the Megha-
362 | Tropiques satellite is used. The relative humidity (daily mean) shown is an average over 12-
363 | 14 passes per day. Figure 7 shows the height-time intensity plot of daily mean relative
364 | humidity during the passage of the cyclones: Nilam (left panel) and Phailin (right panel). The
365 | grid is averaged from 4-8°N and 83-88°E. Strong dry air intrusion originated in the lower
366 | stratosphere is observed between 23-27 October 2012 (Nilam) and 12-18 October 2013
367 | (Phailin). In both the cyclones, dry air (low humidity region) reached down to the altitude of
368 | 8 km. For the perception of the spatial distribution of relative humidity, a latitude-longitude
369 | plot of relative humidity averaged over different pressure level is shown in Figure 8. The low
370 | value of relative humidity i.e., the presence of dry air on the same day of enhanced ozone
371 | mixing ratio in between 5 and 10 km indicate the possibility of dry air present in the
372 | troposphere is of stratospheric origin. The present observations show the influence of the
373 | tropical cyclone on the air mass exchange from the stratosphere to the lower troposphere and
374 | redistribution of stratospheric ozone. Further trajectory and chemical analysis are required to
375 | quantify the amount of mass exchange taking place between the stratosphere and the
376 | troposphere.

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379 5. Summary and Conclusions

380 Important results brought out in the present analysis during the passage of a cyclonic
381 storms Nilam (2012) and Phailin (2013) are summarized below:

- 382 a) Increase in the upper tropospheric ozone by 20-50 ppbv from its climatological mean
383 is observed.
- 384 b) The upper tropospheric ozone propagates downwards to the lower troposphere at a
385 rate of 0.8-1 km/day.
- 386 c) About 10 ppbv in the day-time and 10-15 ppbv in the night-time increase in the
387 surface ozone is noticed
- 388 d) Significant variation in the cold-point tropopause altitude and temperature associated
389 with tropical cyclone as that of the climatological mean are noticed.

390 In the present study, the descent of stratospheric air into the troposphere has been deduced
391 indirectly from a combination of ozone and meteorological observations, and modelling. The
392 study clearly reveals that the cyclones play a vital role in changing the atmospheric
393 composition apart from general weather phenomena.

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The present observation emphasizes the influence of the tropical cyclones in redistribution of the stratospheric ozone and enriched surface ozone.

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596 **Figure Captions**

597 **Figure 1.** (a)Track of cyclones Nilam and Phailin (top panels) and (b) its Outgoing Long
598 wave Radiation (OLR) wave radiation at 14:30 GMT on 30 Oct. 2012 (Nilam) and 9:00
599 GMT on 10 Oct. 2013 (Phailin). In each panel, date and time are mentioned along the track.
600 In the first panel, 18-1/11 indicates 18 GMT of 1 November 2012 and similarly followed
601 for others. The blue star in Fig.1(a) indicates the Ozonesonde launching site Trivandrum.

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602 **Figure 2.** (a) Profiles of ozone mixing ratio (OMR) (dark black line) and relative humidity
603 (grey line) for individual days during the passage of tropical cyclones (a) Nilam and (b)
604 Phailin. The mean ozone mixing ratio profile for non-convective days (as control day) is
605 shown in dotted line. The mean profile is obtained by averaging ozone data over
606 Trivandrum for the month of October from 1995-2013. Horizontal arrows indicate the
607 height of enhanced ozone.

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608 **Figure 3.** Variation of cold point tropopause height (CPT-H) and cold point tropopause
609 temperature (CPT-T) derived from temperature measurement by ozonesonde launched
610 during the passage of tropical cyclones (a) Nilam and (b) Phailin over Trivandrum.

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611 **Figure 4.** Time series of surface ozone mixing ratio (thick line) along with solar radiation
612 (dotted line) from 00 IST on 11 October 2013 to 23:55 IST on 19 October 2013. Solid and
613 dotted horizontal lines indicate the mean maximum and minimum surface ozone. The
614 vertical arrows indicate the nocturnal enhancement of surface ozone. The data is collected
615 with 5 minutes resolution.

616 **Figure 5.** Height-time cross-section of (a) vertical velocity along with potential vorticity
617 (magenta line) and potential temperature (black line) contours, and (b) relative humidity
618 along with equivalent potential temperature (black line) and zonal wind (grey line) for
619 Nilam (left panels) over Trivandrum (8.5°N,76.9°E) from 27 October to 2 November 2012
620 and Phailin (right panels) from 7 to 12 October 2013. Rectangle boxes indicate the presence

of strong updrafts and downdrafts and the dry air between stratosphere and troposphere.

The above parameters are obtained from WRF simulation.

Figure 6. Same as Figure 5 but at 79°E at 18 GMT on 30 October 2012 for Nilam (left panels) and 18 GMT on 10 October 2013 for Phailin (right panels). **Figure 7.** Pressure-time cross-section of relative humidity obtained from SAPHIR onboard Megha-Tropiques satellite during the cyclones Nilam (left panel) from 15 October to 10 November 2012 and Phailin (right panel) from 2 to 22 October 2013. The data is averaged over from 4°N to 8°N and 83°E to 88°E.

Figure 8. Latitude-longitude distribution of relative humidity derived from SAPHIR onboard Megha-Tropiques 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 about 12-14 passes at different timings and arrows indicate the presence of dry air.

Table Caption

Table 1. Details of ozonesonde launched from Trivandrum including the historical data for control day analysis.