

Response to Referee #1's Comments

Influence of tropical cyclones on tropospheric ozone: possible implication By Das et al.

This paper presents ozonesonde observations in southern India for two tropical cyclone cases. For both cases, 5-6 ozonesonde profiles during 5-9 days show an ozone enhanced layer that (seems to have) descended from the upper troposphere to lower troposphere during the observation period at the rate of about 1 km/day. As additional sources of information, numerical simulations using the WRF model and microwave satellite tropospheric humidity data are also presented. The authors conclude that the enhanced ozone layer was originated from the stratosphere in association with the tropical cyclone activity. The ozone observation results are very interesting, and the hypothesis that the observed ozone layer was associated with the tropical cyclone activity is also very interesting. However, I think that the authors need more data analysis to confirm that their conclusions are really supported by all the available data and information. I do not think that in the current manuscript, the hypothesis has been proved correct.

Response : We would like to sincerely thank the anonymous referee for very positive evaluation and constructive comments/suggestion for the improvement of the manuscript.

Point-by-point responses on how we have addressed each recommendations/suggestions are given below. Please note that manuscript is also altered in view of reviewer - 2 and 3's comments and suggestions.

In the following, I write the key questions.

*(1) In Introduction, the authors just cite some papers that discuss possible roles of tropical cyclones in the stratosphere-troposphere exchange. Add more specific discussion, based on previous works (including those discussing the dynamical and thermodynamical structure of tropical cyclones), how tropical cyclones could work for transport of lower stratospheric air into the troposphere. **What is a horizontal scale of such a transport?** At the core (or the eye) of the tropical cyclones, there might exist a net downward transport. But, what the ozone observations showed might be of much larger horizontal scale, including the outer region of a tropical cyclone, and thus related to other flow structures of the tropical cyclones. Also, does the descent rate of 1 km/day correspond to, for example, the subsidence in non-convective tropical region by radiative cooling?*

Response : Introduction is elaborated in view of

- (i) dynamical and thermodynamical structure of tropical cyclone with references**
- (ii) Mechanism for transport of lower stratospheric air into the troposphere**
- (iii) We do not have many observations to comments on its horizontal scale. However, using simulation it was found that the horizontal scale is about 50 km X 250 km (Das et al., 2011).**
- (iv) We do not have observations of stratospheric intrusion during non-convective days for estimating descend rate.**

(2) Reanalysis data and numerical simulation data can be used to make trajectory calculations. I think that trajectory calculations are necessary to show the origin of the ozone enhanced layer and to prove that the layers at different altitudes in different soundings are actually an identical layer.

Response : We thank referee for the suggestion. We have tried to do the back trajectories (ARL/NOAA HYSPLIT) analysis but could not able to capture event. This may be due to the poor temporal and spatial resolution of the reanalysis data (input).

(3) It seems to me that the dates of the numerical simulation results shown in Figure 3 and of the satellite humidity data shown in Figure 4 do not correspond well to those for ozonesonde observations shown in Figure 2. For example, for the Nilam case in 2012, ozonesonde data are from 30 October to 7 November, while the

numerical simulation results are on 30 October for a snapshot and from 27 October to 2 November for the time series. The satellite data are shown on 25 October, with the time series for 15 October to 10 November. I am puzzled at the choice of these dates. Therefore, the question about whether the layers at different altitudes in different soundings are actually an identical layer or not cannot be readily answered.

Response : The ozone sonde observation for Nilam is from 30 October to 7 November 2012. The time-series for Nilam showing in (right panel) Fig.3(c) and 3 (d) (RH) is from 27 October to 2 November 2012, which is well within the observations. To minimize the number of similar figures, we have only shown one set of figures (Figure 3(a) and 3(b)) on 30 October 2012 over the ozone sonde observation site (Trivandrum).

Similarly, the ozone sonde observations for Phailin are from 11-15 October 2013. The time-series for Phailin is shown in Figure 4(c) and 4(d) are from 7-12 October 2013, which is well within the observation period. In addition we have shown one example of height-latitude cross-section on 10 October 2013 over Trivandrum.

Similar methodology is adapted for satellite observations. To avoid the confusion, we have split the Fig.3 and 4 (old manuscript) in two different parts.

(4) The surface ozone actually showed a step-like change in the behavior in Figure 2. However, there are several factors that control the surface ozone (as the authors have acknowledged), and I think that more discussion is needed to attribute the elevated night-time ozone and elevated daytime ozone to the ozone transport from the above. For example, after the passage of a tropical cyclone, stronger sunshine and higher human activities might lead to elevated daytime surface ozone, and prevailing oceanic air-mass following the cyclone might lead to weaker destruction of surface ozone at night-time. I think there are several previous publications that discuss diurnal variations of surface ozone around the tropical coastal regions, which would be helpful for the interpretation of the current results.

Response : Thank you for the suggestions.

We have discussed the changes in sunshine in the revised manuscript. Along with the surface-ozone variation, we have shown the time series of ground-reaching total solar radiation (Please see the Figure below). This shows that there was not much change in the radiation among the days 11-13 and 14-17 October 2013. Thus, this indicates that the observed enhancement was not due to change in sunshine. At the Trivandrum station, ozone remains at its daytime value even after evening hours until the onset of land-breeze (David and Nair, 2011) and does not increase in the evening hours. Interestingly, the enhancement observed on 14 October is during evening hours (16-17 hours) where solar radiation is very low or even zero. (Discussion with the reference including the diurnal variability of surface ozone is made in the revised manuscript). Over the 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 dependency on local precursor gas emission/human activity. Change in human activity during 11-17 October would not have happened considerably and these may not be Bio-mass burning due to rain associated with cyclone. However, the possibility of change of human activity cannot be denied fully. The day-to-day variability of surface ozone over Trivandrum is ~ 9.5 ppbv (1-sigma standard deviation). The observed enhancement of about 10 ppbv in the day-time and 10-15 ppbv in the night-time is above the day-to-day variability which is attributed due to the transport from the UTLS region. However, other possibility of this enhancement cannot be fully ruled out.

There are few studies which clearly proven the enhancement of surface ozone due to intrusion of stratospheric into the troposphere associated with severe weather condition (Stohl et al., 2000; Jiang et al., 2015).

Minor comments.

Technical description is also necessary for the IMD's ozonesonde. Also, are there any intercomparison results between the ECC ozonesonde and IMD ozonesonde?

Response : We thank referee for the suggestion. Now we have incorporated in the revised manuscript.

For the WRF simulations, is the domain for 60 km horizontal resolution from 1S to 25N and 60E to 100E? How about the domain for 20 km horizontal resolution? Also, is the 20 km horizontal resolution appropriate for a tropical cyclone simulation? Cite some papers to discuss the ability and limitation with this setting for a tropical cyclone simulation.

Response : The model domain has been configured with two nested domains of 60 and 20 km horizontal resolution. The inner domain is 20 km and thus it is the horizontal resolution for 1°S to 25°N and 60°E to 100°E. To see the gross-features, 20 km horizontal resolution is appropriate. We have earlier carried out with two nested domains of 81 km and 27 km (Das et al., 2011; Pan et al., 2015).

References :

Das, S.S., Sijikumar, S., Uma, K.N., 2011.: Further investigation on stratospheric air intrusion into the troposphere during the episode of tropical cyclone: Numerical simulation and MST radar observations, Atmos. Res., 101, 928-937.

Jiang, Y. C., Zhao, T. L., Liu, J., Xu, X. D., Tan, C. H., Cheng, X. H., Bi, X. Y., Gan, J. B., You, J. F., and Zhao, S. Z., 2015: Why does surface ozone peak before a typhoon landing in southeast China?, Atmos. Chem. Phys., 15, 13331-13338, doi:10.5194/acp-15-13331-2015.

Stohl, A., Wernli, H., Bourqui, M., Forster, C., James, P., Liniger, M. A., Seibert, P., and Sprenger, M., 2003.: A new perspective of stratosphere-troposphere exchange. Bull. Am. Met. Soc. 84, 1565-1573.70, doi: <http://dx.doi.org/10.1175/BAMS-84-11-1565>.

Pan, L.L. et al., 2015.: Thunderstorms enhance tropospheric ozone by wrapping and shedding stratospheric air, Geophys. Res. Lett., 41, 7785-7790, doi : 10.1002/2014gl061921.

Response to Referee #2's Comments

The paper "Influence of tropical cyclones on tropospheric ozone: possible implication" by Das. S. S. et al. discusses the role tropical cyclones may play in controlling tropospheric ozone level. The paper is based on the analysis of ozone and humidity measurements in the vicinity of two tropical cyclones which occurred in the Bay of Bengal in 2012 and 2013. The paper also takes advantage of a series of WRF simulations which describes the dynamical field in the vicinity of the cyclones. Understanding the role of tropical convection in the stratosphere to troposphere exchange is certainly a scientific question which falls well within the scope of ACP. This is made even more interesting by the impact these processes could have on near surface conditions. Although studies of this kind are not new the authors present results of two cyclones which could potential add relevant information on these exchange processes. Das S. et Al. also appear to reach quite substantial conclusions by the end of the paper (pages 19315 lines 4-+9920) but these don't seem to be fully supported by the evidences presented in the paper. In particular I don't think the paper demonstrates to a sufficient level:

Response : We would like to sincerely thank Dr.C.Buontempo for reviewing the manuscript and for his constructive comments/suggestion and accepting the conceptual view.

Point-by-point responses on how we have addressed each recommendations/suggestions are given below. Please note that manuscript is also altered in view of reviewer - 1 and 3's comments and suggestion.

1) that the ozone enhancement is attributable entirely to a stratospheric intrusion and not to local source (e.g. lightning), tropospheric advection, or anthropogenic origin.

Response : During the period of observations, the sky was fully cloudy, raining and the possibility of anthropogenic activities are negligible during rain event. Moreover, the enhancement observed in ozone is at much higher height i.e. middle and upper troposphere.

There were lightning event all around the Indian region not specifically over the cyclone area as observed from satellite measurements. However, we agree with referee that we cannot fully rule out the thunderstorm.

There may be possibility of advection which is already discussed and also attributed due to cyclone. Now we have modified / revised the manuscript accordingly.

2)that the intrusion in the troposphere would add as a significant stratospheric sink as suggested in line 7-10 on page 19307

Response : Yes, we agree with the referee but the amount is too low as that of stratospheric ozone which cannot be traceable.

3) how significant such exchange process is for the overall ozone budget of the troposphere and what its possible impact on the living organisms could be (page 19307 line 27 and page 19308 line 15-19).

Response : Ozone budget and its impact on living organism are now discussed in the revised manuscript (*National Research Council, 1991*).

4)whether the tropical cyclones in the bay of Bengal could be considered representative of the tropical cyclones in other part of the tropics.

Response : Probably yes. We have come across few latest literatures which proved that thunderstorms enhance tropospheric ozone [*Pan et al., 2014*]. More discussion is made in the revised manuscript with references.

There are few studies which clearly proven the enhancement of surface ozone due to intrusion of stratospheric into the troposphere associated with severe weather condition (*Stohl et al., 2000; Jiang et al., 2015*)

5) *what is the extent of the area for which the ozone enhancement area has been recorded.*

Response : It is very difficult to say the extent of the area as there is no much observational (ozonesonde) evidence and satellite passes are very limited in time and space. However, with the numerical simulations it is found that the entire band of the cyclone participated in the exchange process, i.e. 50 km x 250 km.

6) *how significant the enhancement of surface ozone detected during the passage of the cyclones is when compared to the normal level of variability at the station.*

Response : The variability at the station depends on the synoptic meteorological condition in addition to changes in local anthropogenic activity. The variability (i.e. 1-sigma standard deviation) is ~9.5 ppbv during the month of October. Thus, observed enhancement is of the order of normal variability at the station. Thus, it is possible that observed enhancement could be due to surface reaching intrusion effect but possibility of local/anthropogenic activity could not be fully ruled out.

7) *how well the WRF-ARW is able to describe the dynamical field around the cyclone cyclones that exists.*

Response : We have used different Physics schemes using WRF-ARW simulation and found one scheme is compared well with the observation of zonal, meridional and vertical winds (Das et al., 2011). Das et al. (2011) and Pan et al. (2015) have uses of WRF-simulation for Stratosphere to troposphere exchange during the passage of tropical cyclone.

8) *I would think the paper would become significantly stronger if an estimate of the overall ozone mass exchange though cyclones would be attempted by the authors on the basis of the measurements they acquired and the number of tropical*

Response : We agree with the referee but we have only two cases of tropical cyclone where we have few ozonesonde observations and it is not extensive enough to allow us to estimate the ozone flux. In near future we will definitely estimate of the overall ozone mass exchange with more number of cyclone cases and simultaneous observations of ozonesonde at different locations. We thank referee for the suggestion.

Detailed comments:

Page 19306 Line 18 : at least one of the many references should be mentioned here.

Response : Now reference is included.

Page 19306 Line 18 : I would think the properly of Ozone as GHG don't depend so much on its location in the atmosphere

Response : Corrected.

Page 19306 Line 24 : I don't think that categories such as "bad" and "good" are particularly useful for the discussion here especially because it is not clear to whom such change would be good or bad.

Response : Following the reviewer's comments, entire sentence is removed in the revised manuscript.

Page 19308 lines 9-10: at least at the first order the amount of radiation that reach the surface should not depend on the specific vertical profile of ozone as much as on its total column amount.

Response : Sentence is modified in the revised manuscript.

Page 19313 line 14: I don't think it can be safely assumed that tropical cyclones have no lightning for example:
<http://journals.ametsoc.org/doi/abs/10.1175/MWR-D-11-00236.1>

Response : We thank referee for raising this important point. Now we have revised the statements in this version of manuscript.

Page 19314 line 24: I think the provides only once piece of evidence in support of the thesis the authors suggest rather than a demonstration similarly I would be more cautious in the conclusion c.

Response : Now we have elaborated the discussion in this context.

Page 19315 lines 11-14: I don't think this statement is fully supported by the evidences presented by the authors. The enhancements the authors suggest (which I assume can only be local) is probably offset by a slow ascent happening on the large scale. Understanding the balance between these two competing processes would be the only way to the long-term impact tropical cyclones may have on the ozone concentration of the troposphere.

Response : With the limited observations of ozonesonde along with numerical simulation we have tried to established the stratospheric intrusion associated with the passage of tropical cyclone.

References :

Pan, L.L. et al., 2015.: Thunderstorms enhance tropospheric ozone by wrapping and shedding stratospheric air, Geophys. Res. Lett., 41, 7785-7790, doi : 10.1002/2014gl061921.

Jiang, Y. C., Zhao, T. L., Liu, J., Xu, X. D., Tan, C. H., Cheng, X. H., Bi, X. Y., Gan, J. B., You, J. F., and Zhao, S. Z., 2015: Why does surface ozone peak before a typhoon landing in southeast China?, Atmos. Chem. Phys., 15, 13331-13338, doi:10.5194/acp-15-13331-2015.

Stohl, A., Wernli, H., Bourqui, M., Forster, C., James, P., Liniger, M. A., Seibert, P., and Sprenger, M., 2003.: A new perspective of stratosphere-troposphere exchange. Bull. Am. Met. Soc. 84, 1565-1573.70, doi:
<http://dx.doi.org/10.1175/BAMS-84-11-1565>.

National Research Council (1991), Rethinking the Ozone Problem in Urban and Regional Air Pollution, 1051 Committee on Tropospheric ozone formation and measurement, Natl. Acad. Press, Washington, D.C.

Response to Referee #3's Comments

Review for "Influence of tropical cyclones on tropospheric ozone: possible implication" by Das et al. The authors present an interesting study where two cases of tropical cyclones enhance tropospheric ozone levels. The two cyclones Nilam and Phailin are discussed with data from ozone dropsondes, surface ozone measurements and relative humidity derived from satellite scans. Furthermore, numerical simulations are used to get a more complete picture of the dynamics of the events. All in all, the argumentation is clear enough and the data support the main statements. However, there are also some concerns which need some consideration before the manuscript is ready for publication.

Response : We would like to sincerely thank the anonymous referee for very positive evaluation and constructive comments/suggestion for the improvement of the manuscript.

Point-by-point responses on how we have addressed each recommendations/suggestions are given below. Please note that manuscript is also altered in view of reviewer - 1 and 2's comments and suggestion.

Major concerns:

1. In its present state, the introduction is not very well structured and at several places remains rather unspecific.

• For instance, it is written that Appenzeller and Davies (1992) attribute the stratospheric intrusion to "disturbed weather conditions over mid-latitude". This is not specific enough!

Response : We have rewritten the introduction section.

• Furthermore, I would not agree that stratospheric intrusions are generally a slow process, as written in P19307,L10. See for instance recent studies by Bourqui and Trepanier (2010), Skerlak et al (2014) or the review by Stohl et al. (2003) with its particular focus on the synoptic scale of STE (see below). In short, a more careful review of current literature seems to be appropriate.

Response : Now the sentence is revised in view of recent literature survey and more references are added in this version of manuscript.

• The first paragraph starts with some very general statements about ozone in the atmosphere. I wonder whether this could be considerably shortened and the focus brought much faster to the main topic of the study, i.e., how tropical cyclones influence the tropospheric ozone levels.

Response : We feel general introduction is necessary to make the reader comfortable. However, we have revised these introductory sentences taking in account of all the reviewer's comments.

• At L19307,L8-9 it is written that stratospheric intrusions "... also decreases the stratospheric ozone, which in principle enhances the penetration of UV to reach the Earth's surface." In principle yes, but I doubt that it is of practical importance. Is there a reference for this statement?

Response : As per our knowledge there are no such literatures which have shown quantitatively the amount of decreasing stratospheric ozone and its effect in enhancing the penetration of UV radiation to Earth's surface. However, this sentence is well accepted and mentioned in many literatures (e.g. Baray et al., 1999 and Cairo et al., 2008)

• The last sentence of the introduction repeats statements from before. There is no need to 'complete' the introduction with such a summarizing statement.

Response : Now the sentence is omitted in the revised manuscript.

• In P19308,L8-10 the effect of humidity on ozone is discussed. But it remains unclear how, in the context of the paper's research topic, this fits in. The sentence looks a little 'out-of-context'!

Response : This sentence is omitted. Introduction is rewritten in the revised manuscript.

• The aim of the study is only handled in one single sentence near the end of the introduction: "The present study addresses the influence of tropical cyclones quantitatively on enhancement of tropospheric ozone by stratospheric intrusion." First, the sentence structure looks a little strange to me, second, I would appreciate when the aim of the studied is presented in some greater detail.

Response : This sentence is revised accordingly.

2. Some physical arguments remain unclear, or can be critically questioned.

• In Figure 2 the surface ozone measurements for Phailin are shown. There is a nice shift in background ozone levels from 14 to 15 October 2013. The authors attribute this increase to air descending within a stratospheric intrusion. In the same line they mention other processes which influence ozone levels (P19313, photochemical reactions, biomass burning and lightning). However, they state that given the cyclones' characteristics, the impact of any of the three mechanisms will be very low. If this is the case, where does the diurnal cycle in the surface ozone measurement coming from? In short, I am not fully convinced that the other processes are really negligible. Note also that on 16 October the surface ozone measurement reaches very low ozone mixing ratios, although the stratospheric intrusion already 'took place'.

Response : By the statement "given the cyclones' characteristics, the impact of any of the three mechanisms (photochemical reactions, biomass burning and lightning) will be very low", we wanted to say that changes in these mechanisms would be low. We agree that the diurnal variability in surface ozone is influenced by photochemical processes under the presence of precursor gases like NO_x, CO, etc (David and Nair, JGR, 2011). But the enhancement in ozone is observed in the middle and upper troposphere and as the day progresses the enhancement height of ozone decreases. If the enhancement is due to local activities e.g. biomass burning then initially we might have observed ozone enhancement in the surface or lower troposphere which is in contrast to our observations. Considering the variability in the surface ozone at which is ~9.5 ppbv (i.e standard deviation during month of October), the observed enhancement (~10 ppbv) fall under the day to day variations. Thus, in the view of this, it is possible that observed enhancement could be due to surface reaching intrusion effect but possibility of local/anthropogenic activity could not be fully ruled out.

There are few studies which clearly proven the enhancement of surface ozone due to intrusion of stratospheric into the troposphere associated with severe weather condition (Stohl *et al.*, 2000; Jiang *et al.*, 2015).

• As a 'proof' that stratospheric air is really coming down, it is written that "Enhanced potential vorticity 0.5–1.5 PVU is also observed vertically down from the stratosphere to the surface, overlapping the downdraft regions." There is indeed a clear PV maximum discernible in Figure 3, at about mid-tropospheric levels. But one might argue that this is diabatically produced PV, due to condensational heating, which therefore is not of stratospheric origin. This option should clearly be discussed in the manuscript.

Response : We thank referee for the suggestion. Now we have incorporated in the revised manuscript.

• Some of the formulations are not careful enough. For instance, (P19313,L25) "The potential temperature contours indicate the presence of unstable atmosphere at this location". I do not see any sign of an unstable air column. Possibly, what is meant is that the stability is reduced?!

Response : Corrected in the revised manuscript.

Minor concerns:

- P19309,L3: There is no need to repeat at several places that it is a very severe cyclonic storm'. The text seem here, and other places, a little repetitive.

Response : Corrected in the revised manuscript.

- P19309: I am not familiar with ozone analysis. What does "1% linearity" mean?

Response : The surface ozone analyser was calibrated by applying known mixing ratios of ozone in the measurement range (i.e. 0 to 70 ppbv). The calibration constant is fixed based multipoint calibration (i.e. based on linear fit between know values of ozone and those measured by the analyser). The analyser

shows linear behaviour, however it deviates from linearity by $\pm 1\%$. Thus, ozone mixing ratios may have deviation of $\pm 1\%$ of its measured value. Note that our ozone measurement range is from few ppbv to 30 ppbv, $\pm 1\%$ correspond to 0.3 ppbv which is very small in the context of observed enhancement (i. e. ~ 10 ppbv).

- P19309,L23: "This data is also used to do the qualitative analysis of stratospheric air." What do you mean with 'qualitative analysis of stratospheric air'?

Response : Sentence is removed from the revised manuscript.

- Section 3. The last paragraph of section 3.2 would better be placed in the introduction to the whole section 3. It applies to both cases and already refers to the images in Figure 1.

Response : Following the reviewer's suggestion, we have moved this sentence to the whole section 3.

- Section 4: I strongly suggest to rename section 2 into "campaign details and data" (or something in this direction). Then, the description of section 4 could be added to this new section. At the moment, section 4 with all the technical details of the NWP simulation looks rather out of place. It distracts the reader from the physical discussion.

Response : Following the reviewer's suggestion, we have moved the section under 'Numerical Simulation' to the section 2.

- Figure 1: Mention in the caption what the blue star refers to!

Response : Added in the figure caption.

- Figure 2: Describe in caption, what the mean ozone profile refers to. It's discussed in the text, but not in the figure caption.

Response : Added in the figure caption.

- Figure 3: The position of the height-latitude cross-section could be shown in Figure 1.

Response : It is very difficult to accommodate this information in Fig.1 (a) as it is already very crowded. We hope reviewer will kindly accept not to incorporate this particular suggestion.

Reference

David, L. M., and Nair, P.R. 2011.: Diurnal and seasonal variability of surface ozone and NO_x at a tropical coastal site: Association with mesoscale and synoptic meteorological conditions, 116, D10303, doi : 10.1029/2010JD015076.

Jiang, Y. C., Zhao, T. L., Liu, J., Xu, X. D., Tan, C. H., Cheng, X. H., Bi, X. Y., Gan, J. B., You, J. F., and Zhao, S. Z., 2015: Why does surface ozone peak before a typhoon landing in southeast China?, Atmos. Chem. Phys., 15, 13331-13338, doi:10.5194/acp-15-13331-2015.

Stohl, A., Wernli, H., Bourqui, M., Forster, C., James, P., Liniger, M. A., Seibert, P., and Sprenger, M., 2003.: A new perspective of stratosphere-troposphere exchange. Bull. Am. Met. Soc. 84, 1565-1573.70, doi: <http://dx.doi.org/10.1175/BAMS-84-11-1565>.

1 | **Influence of the Tropical Cyclones on Tropospheric Ozone: Possible Implication**

2
3 | *Siddarth Shankar Das^{1*}, M. V. Ratnam², K. N. Uma¹, K. V. Subrahmanyam¹, I.A.Girach¹,*
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12 | **Abstract.** The present study examines the role of tropical cyclones in the enhancement of
13 | tropospheric ozone. The most significant and new observation is the increase in the upper
14 | tropospheric (10-16 km) ozone by 20-50 ppbv, which has extended down to the middle (6-10
15 | km) and lower troposphere (< 6 km). The descending rate of enhanced ozone layer is found to
16 | be ~ 1 km/day. **Enhancement of surface ozone concentration by ~ 10 ppbv in day-time and**
17 | **10-15 ppbv in the night-time is observed during cyclone.** Numerical simulation of potential
18 | vorticity, vertical velocity and potential temperature, **reproduced the key feature of**
19 | **observations. Simulation study indicates the downward transport of stratospheric air in to the**
20 | **troposphere.** Space borne observations of relative humidity indicate the presence of sporadic
21 | dry air in the upper and middle troposphere over the cyclonic region. These observations
22 | constitute quantitatively an experimental evidence of enhanced **and redistribution of**
23 | tropospheric ozone during cyclonic storms.

24
25 | **[Key words:** Stratosphere-troposphere exchange processes, tropopause, ozone, water vapour]

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34 **1. Introduction**

35 ~~Stratospheric ozone (O₃) maximum around 25-30 km altitude~~, regulates the amount of
36 ultraviolet radiation coming from the Sun to the Earth's surface. There are ample studies
37 indicating depletion of stratospheric ozone since 1890's and its consequences of impairing
38 the entire ecological system (e.g. *Forster et al., 2007*). Ozone is an important greenhouse gas,
39 which acts as an oxidant in the troposphere and have an important role in climate forcing
40 (*Forster et al., 2007; Pan et al., 2015*). The tropopause is a layer that separates the
41 troposphere and the stratosphere, and plays a key role in controlling the mixing of minor
42 constituents, viz., ozone and water vapour between these two layers. One of the major
43 consequences of the tropospheric ozone enhancement is on the living organism, as it acts as a
44 toxic agent among the air pollutants (*National Research Council, 1991*).

45 Owing to the importance of ozone chemistry and its implication for air quality and
46 climate change, studies focusing on the origin of ozone enhancement in the troposphere, its
47 trends and distributions need immediate consideration. Increase in the tropospheric ozone is
48 considered to be due to (1) in-situ photochemical formation associated with lightning,
49 advection, anthropogenic (e.g., *Jacobson, 2002* and references therein), and (2) stratospheric
50 flux (*Wild, 2007* and reference therein; *Skerlak et al., 2014*). Increase of the ozone downward
51 flux from the stratosphere to the troposphere not only increases the tropospheric ozone, but
52 also decreases the stratospheric ozone, which in principle enhances the penetration of UV
53 radiation to reach the Earth's surface.

54 In general, downward flow of air masses from the stratosphere to the troposphere, i.e.
55 stratospheric intrusion is a middle and higher latitude phenomenon linked with synoptic scale
56 disturbances (*Holton et al., 1995*). This downward flow is attributed to the dissipation of
57 extra-tropical planetary and gravity waves in the stratosphere (*Stohl et al., 2003* and
58 references therein). *Liang et al.* (2009) have described the time scale of stratospheric ozone

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81 intrusion that occurs in 3 steps, and takes about three months to reach from stratosphere to
82 lower troposphere. In extra-tropic there is a continuous downward flow from the stratosphere
83 to the troposphere (Stohl et al., 2003; Bourqui and Trepanier, 2010). In the global ozone
84 budget, 25-50 % of tropospheric ozone source is from middle latitude stratospheric intrusion
85 (Bourqui and Trepanier, 2010). Appenseller and Davies (1992) have also discussed that
86 exchange between the stratosphere and the troposphere (both directions) is highly episodic
87 due to the perturbation of tropopause associated with strong mesoscale convective systems.
88 A global climatology of STE was established by Skerlak et al. (2014) using ERA-Interim
89 reanalysis data. The authors have estimated fluxes of mass and ozone across the tropopause.
90 Maximum ozone forms over the tropics, moves upward and then pole-ward by Brewer-
91 Dobson circulation, which further descends to the lower stratosphere and troposphere at high
92 latitudes. There is much observational evidence supporting the slow intrusion of
93 stratospheric air into the troposphere during cut-off lows (Vaughan and Price, 1989),
94 high/low pressure systems (Davies and Schuepbach, 1994), the tropopause folds (Sprenger
95 and Wernli, 2003) and in a rapid episodic manner which generally triggered by overshooting
96 convection, like a tropical cyclones (Loring et al., 1996; Baray et al., 1999; Cairo et al.,
97 2008; Das, 2009, Das et al., 2011, Zhan and Wang, 2012; Jiang et al., 2015).The tropical
98 cyclones are the synoptic-scale disturbances of organised convective systems which weaken
99 the tropopause by overshooting convection and enhancing the stratosphere-troposphere
100 exchange (STE) in a spontaneous manner. In addition, turbulence caused due to wind shear
101 (Shapiro, 1976) and breaking of gravity wave (Langford et al., 1996) can also be the
102 causative mechanisms for the occurrence of stratospheric intrusion. Recent study by Pan et
103 al. (2015) have shown the enhancement of tropospheric ozone is associated with
104 thunderstorm event (Pan et al., 2014). Slow stratospheric intrusion is reasonably well

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106 understood and is a regular phenomenon, whereas the rapid intrusion needs to be understood
107 in detail.

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108 The intrusion of the stratospheric air into the troposphere has been demonstrated by
109 *Hocking et al.* (2007) for mid-latitude condition and *Das* (2009) for low latitude during
110 cyclone using high power radar system (VHF radar). Subsidence of stratospheric air is
111 generally observed in the vicinity of cyclone (*Baray et al.*, 1999; *Cairo et al.*, 2008; *Leclair*
112 *De Bellevue et al.*, 2006, 2007; *Das*, 2009, *Das et al.*, 2011). *Appenzeller and Davies* (1992)
113 have shown the intrusion of the stratospheric air into the troposphere due to disturbed weather
114 condition over mid-latitude, combining weather prediction model and satellite observations.

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115 Stratospheric intrusion can bring ozone rich air from stratosphere to the troposphere
116 which further affects the surface ozone concentration (*Bourqui and Trepanier*, 2010). Earlier
117 studies using aircraft measurement shows that the perturbation of the tropopause associated
118 with deep convective thunderstorm can transport ozone from the stratosphere to the
119 troposphere (*Dickerson et al.*, 1987; *Poulida et al.*, 1996; *Stenchikov et al.*, 1996; *Pan et al.*,
120 2015). *Stohl et al.* (2000) have shown that episodic stratospheric intrusion is associated with
121 severe weather condition which enhanced the surface ozone concentration. A recent study by
122 *Jiang et al.* (2015) have shown that high surface ozone by 21-42 ppbv and the nocturnal
123 surface ozone level exceeding 70 ppbv over the southeastern coast of China during the
124 passage of tropical cyclone (before land fall).

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125 A tropical cyclone is a heat engine of hot cumulus towers, areas of low pressure and is
126 characterized by cyclonic tangential and inflowing radial winds. The tropical cyclone
127 circulation consists of (a) the rotational air flow in the horizontal direction, (b) the in-up-out-
128 down overturning flow in the vertical direction, (c) out-flow at in the vicinity of UTLS region
129 and (d) subsiding nature in the periphery (*Jiang et al.*, 2015). The eye (centre of the tropical
130 cyclone) is about tens of kilometre and it is the core of downdraft with relatively no cloud

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141 with calm wind structure. The bands of the tropical cyclone have intense vertical extended
142 cumulus cloud up to UTLS region. These regions are accompanied with updrafts, whereas
143 downdrafts are encounter between these bands. The eyewall region is characterised by local
144 maximum equivalent potential temperature, whereas minimum is found in the middle to
145 upper troposphere. The eyewall and radius of maximum winds increases with height. The low
146 pressure core extended to UTLS region and the horizontal pressure gradient decreases with
147 height (*Koteswaram, 1967*). *Mitra (1996)* and *Das (2009)* reported the weakening of the
148 tropopause due to a tropical cyclone. Detail study on the dynamical and thermo dynamical
149 structure of tropical cyclone can be found in *Hence and Houze (2012)* and review article on
150 clouds in the tropical cyclone can be found in *Houze (2010)*. Thus, the tropical cyclones have
151 influence on stratosphere-troposphere exchange process which causes air mass and energy
152 transports in the troposphere and redistribution of tropospheric ozone (e.g. *Jiang et al., 2015*).
153 A complete review on the effect of the tropical cyclones on the upper troposphere and lower
154 stratosphere can be found in *Cairo et al. (2008)*. In spite of many observational and modelling
155 studies, the exchange of air mass from the stratosphere to the lower troposphere in short-time
156 scale associated with tropical cyclones is still unclear and further studies are needed.

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

161 2. Campaign details and data analysis

162 An intense campaign, named as 'Troposphere-Stratosphere Exchange-Cyclone (TSE-C)'
163 under the Climate And Weather of Sun-Earth System (CAWSES)-India phase-II programme
164 was conducted during two cyclone events. Under this campaign, a series of ozonesondes
165 were launched from Trivandrum (8.5°N, 76.5°E) during the intense period of cyclonic storm

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167 Nilam from 30 October to 7 November 2012 and a very-severe cyclonic storm Phailin from
168 11 to 15 October 2013. The ozonesondes used are EN-SCI (USA) make, which were
169 integrated with the GPS based radiosondes of i-met make. These standard ozonesonde are
170 made up of the Electrochemical Concentration Cell (ECC) (*Komhyr et al.*, 1995). The
171 uncertainty in the ozone measurements is 5-10 %. Table 1 also provides the details of
172 ozonesonde measurements conducted during the passage of cyclonic storm Nilam and
173 Phailin. Ozonesonde data was obtained at a fixed height resolution by down sampling at 100
174 m height resolution by linear interpolation method. The India Meteorological Department
175 (IMD) also conduct 1-2 **IMD-ozonesonde** launches per month. The background profiles (non-
176 convective day at least for 3 days) is constructed by averaging the ozone data (23 profiles)
177 obtained from **the** IMD combined with our observations from 1995-2013 for the month
178 October over Trivandrum. **The IMD-ozonesonde used Brewer bubbler electrochemical sonde**
179 **developed in the Ozone Research Laboratory of the IMD. These IMD ozone sonde were**
180 **compared with ECC sondes and found that it is underestimated by 5-10 % in the troposphere**
181 **(*Kerr et al.*, 1994; *Deshler et al.*, 2008), which is about <2 ppbv of the observed mean value.**
182 **Detail system description of IMD-Ozonesonde can be found elsewhere (*Sreedharan*, 1968;**
183 ***Alexander and Chatterjee*, 1980). There is no ozonesonde launch by IMD in campaign mode**
184 (daily one launch) during the low pressure system. The measurements of near-surface ozone
185 are carried out using the online UV photometric ozone analyser (Model AC32M) of
186 Environment S.A, France. This ozone analyser works on the principle of UV absorption of
187 ozone at the wavelength 253.7 nm. The instrument has a lower detection limit of 1 ppbv and
188 1% linearity. The data is sampled with an interval of 5 min.

189 The SAPHIR (Sondeur Atmospherique du Profild' Humidite Intertropical par
190 Radiometrie) onboard Megha-Tropiques satellite is a multichannel passive microwave
191 humidity sounder, measuring brightness temperatures in six channels located close to the

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193 183.31GHz water vapor absorption line ($\pm 0.15, \pm 1.20, \pm 2.80, \pm 4.30, \pm 6.60$ and ± 11.0 , GHz).
194 These channels allow retrieving the integrated relative humidity respectively between the
195 levels of 1000–850 hPa, 850–700 hPa, 700–550 hPa, 550–400 hPa, 400–250 hPa, and 250–
196 100 hPa. The radiometer has a cross-track scan of $\pm 43^\circ$, providing a swath of 1705 km and a
197 10 km resolution at nadir. This data is also used to do the qualitative analysis of stratospheric
198 air. The detail instrumentation can be found in *Raju* (2013), and retrieval algorithm and
199 validation can be found in *Gohil et al.* (2012); *Mathur et al.* (2013) and *Venkat Ratnam et al.*
200 (2013); *Subrahmanyam and Kumar* (2013), respectively.

201 Apart from the ozonesonde observations, a high resolution numerical simulation using the
202 Advanced Research Weather Research and Forecast (WRF-ARW) model version 3.6 has also
203 been carried out for both cases of cyclones. The model domain has been configured with two
204 nested domains of 60 km and 20 km horizontal resolution, and covers an area extending from
205 1°S to 25°N and 60°E to 100°E. The innermost domain has been used for the present study.
206 The initial and lateral boundary conditions have been taken from ERA-Interim reanalysis on
207 0.75° x 0.75° continuously at every 6 hours. The present simulation was carried out with the
208 model Physics options :(i) New Simplified Arakawa-Schubert (NSAS) (*Han and Pan, 2011*),
209 (ii) Yonsei University (YSU) boundary layer scheme (*Hong et al., 2006*), (iii) Rapid
210 Radiative Transfer Model (RRTM) long wave radiation scheme (*Mlawer et al., 1997*), (iv)
211 WRF Single Moment (WSM) 5 class microphysical scheme (*Hong et al., 2004*), and (v)
212 NOAA land-surface scheme (*Smirnova et al., 2000*).

213 3. Meteorological background

214 The present experiments were conducted during the passage of the (1) cyclonic storm
215 ‘Nilam’ from 28 October to 1 November 2012 and (2) very severe cyclonic storm ‘Phailin’
216 from 4-14 October 2013 over the Bay of Bengal (BOB). The track of each tropical cyclones
217 and outgoing long wave radiation (OLR) images (date and time are stamped) are shown in

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221 | Figures 1a and 1b, respectively. The detailed bulletin can be found in www.imd.gov.in.
222 | During these campaigns, several ozonesondes were launched from Trivandrum, whenever the
223 | intensity of cyclones is maximum and the path/eye was close to the launching site. The
224 | details of each of the tropical cyclone used for present analysis are as follows:

225 | 3.1 Case-1 (Nilam)

226 | A depression formed over the southeast of BOB ($\sim 9.5^{\circ}\text{N}$, 86.0°E) at 11:30 IST of 28
227 | October 2012. It moved westwards and intensified into a deep-depression on the morning of
228 | 29 October 2012 over southwest BOB, about ~ 550 km south-southeast of Chennai. It
229 | continued to move westwards and intensified into a Cyclonic Storm, 'Nilam' in the morning
230 | of 30 October 2012 over southwest BOB. Then it moved north-northwest, crossed the north
231 | Tamilnadu coast near Mahabalipuram (12.6°N , 80.2°E), south of Chennai in the evening
232 | hours of 31 October 2012. After the landfall the cyclonic storm, Nilam moved west-northwest
233 | and weakened gradually into a deep depression and then into a depression in the morning
234 | hours of 1 November 2012.

235 | 3.2 Case-2 (Phailin)

236 | A low pressure system was formed over Tenasserim coast ($\sim 12.0^{\circ}\text{N}$, 96°E), on early
237 | morning of 6 October 2013. It intensified into a depression over the same region on 8 October
238 | and then moved towards the west-northwestwards. It further intensified into a deep
239 | depression on early morning of 9 October 2013 and then into a cyclonic storm, 'Phailin' in
240 | the evening hours. Moving northwestwards, it finally converted into a severe cyclonic storm
241 | in the morning hours of 10 October 2013 over east central BOB. The very severe cyclonic
242 | storm continued to move northwestwards and crossed Andhra Pradesh and Odisha coast near
243 | Gopalpur (19.2°N , 84.9°E) in the late evening of 12 October 2013. It further continued to
244 | move north-northwestwards after the landfall for some time and then northward and finally
245 | north-northeastwards up to southwest Bihar. The system weakened gradually into a cyclonic

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246 storm from 13 October 2013 and finally the intensity decreased to a low pressure system on
247 14 October 2013.

249 4. Results and Discussion

250 Figure 2 (a-b) shows the profiles of ozone mixing ratio (OMR) and relative humidity (RH)
251 from ozonesonde measurements during the passage of the tropical cyclones Nilam (top
252 panels) and Phailin (bottom panels). The background ozone profile is obtained by averaging
253 individual profile (23 profiles) over Trivandrum of October from 1995-2013 and shown by
254 dotted lines in Figure 2. During the passage of Nilam on 30 October 2012, enhancement in
255 tropospheric ozone (marked by horizontal arrows) from background by 40-50 ppbv was
256 observed in the height region between 8-9 km (~1 km width) and 11-14 km (~3 km
257 width). These enhancements persisted till 31 October 2012 but at the height region reduced 6-
258 7 km with a reduced width. However, the enhancement of about ~40 ppbv was still observed
259 on 2 November 2012 but the height region decreased to 5-6 km. After two days, we had again
260 observations from 5-7 November 2012. The height of enhancement in the ozone profiles were
261 reduced to ~4, ~3 and ~1.5 km by 40, 30, and 20 ppbv on 5, 6 and 7 November 2012,
262 respectively. The present observation reveals that the downward propagation of the enhanced
263 upper tropospheric ozone into the lower troposphere occurs in episodic manner. The
264 descending rate of the ozone rich layer from the upper troposphere to the boundary layer
265 during Nilam is approximately estimated to be ~875 m/day. It is also noted that the
266 corresponding relative humidity profiles during Nilam did not decrease with increasing ozone
267 mixing ratio except on 2 November 2012. A significant sudden decrease in relative humidity
268 is observed on 2 November 2012 at ~6 km, where the maximum enhancement (~ 70 ppbv) of
269 tropospheric ozone is observed. This is due to the presence or accumulation of dry air, which
270 is believed to have been originated from the stratosphere. A similar phenomenon is also

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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 (8.5°N, 76.5°E), whenever the intensity of cyclones were maximum and the path/eye was close to the launching site. ¶

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Apart from the radiosonde/ozonesonde observations, a high resolution numerical simulation using the Advanced Research Weather Research and Forecasting (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 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*). ¶

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311 observed during the passage of Phailin. Intrusion from ~14 km to 6 km (marked by horizontal
312 arrows) is clearly observed in the ozone profiles from 11-15 October 2013. During Phailin,
313 tropospheric ozone enhancement by 20-30 ppbv is observed and the width of the enhanced
314 ozone layer is larger than that during Nilam. During Phailin, descending rate of enhanced
315 ozone layer from the upper troposphere to the boundary layer is estimated to be ~1000 m/day.
316 Relative humidity profiles also show the sudden decrease between 2-6 km on 14 and 15
317 October 2013, indicating the presence of dry air similar to that observed during the Nilam.

318 As discussed in the introductory section, significant perturbation in the tropopause
319 due to deep convection will lead to the transport of ozone rich stratospheric air in to the
320 troposphere. Figure 3 shows variation in the cold point tropopause height (CPT-H) and cold
321 point tropopause temperature (CPT-T) derived from the temperature measurement by
322 ozonesonde launched during passing of the tropical cyclones (a) Nilam and (b) Phailin over
323 Trivandrum. Significant perturbation in the tropopause is observed for both the cyclone cases.
324 The CPT-H gradually decreased from 17.8 km on 30 October to 16.7 km on 2 November
325 2012 for Nilam. Afterwards, the CPT-H gradually increased and reached to 17.5 km.
326 Similarly for Phailin, the CPT-H decreases from 16.5 km on 11 October 2013 to 15.8 km on
327 12 October 2013 and then gradually increases. In both the cyclone events, CPT-T show anti-
328 correlation with CPT-H as expected. The height above the tropopause (i.e. stratosphere) is in
329 radiative equilibrium, whereas the height below the tropopause (i.e. troposphere) is in
330 convective equilibrium. Thus, turbulent mixing is taking place below the tropopause and
331 above it (i.e. stratosphere), the air is highly stable and of subsidence in nature. On 30 October
332 2012, when the tropopause height is at 17.8 km the turbulent mixing is taking place below
333 17.8 km and the mixing height decreased following the tropopause height and reached as low
334 as 16.7 km on 2 November 2012. When the tropopause height increased after 1 November
335 2012, the turbulent mixing height also increased but the stratospheric air still remained in the

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336 troposphere as it cannot follow the spontaneous increase in the tropopause height. During this
337 process (i.e. increasing height of the tropopause), the stratospheric air get mixed with
338 tropospheric air by the mean of turbulent mixing and it further intrude down to the lower
339 troposphere due to the presence of downdrafts as observed in the simulation (discussed later).

340 In addition to the profiling of ozone, we have surface measurement of ozone and solar
341 flux during the Phailin. Figure 4 shows the time series of near-surface ozone mixing ratio
342 along with solar irradiation from 11 to 19 October 2013. As expected, clear diurnal variability
343 is observed in the time-series of ozone. In general there are three main mechanisms for the
344 production of ozone in the atmospheric boundary layer: (1) photochemical reaction via NO_x
345 and CO channel, (2) Bio-mass burning /fossils fuel, and (3) lightning. However, *David and*
346 *Nair (2011)* have shown the diurnal pattern of surface ozone observed over Trivandrum is
347 due to the mesoscale circulation, i.e., local sea and land breeze and the availability of NO_x.

348 From 11 to 14 October the maximum and minimum average peak of ozone are observed to be
349 24 and 1 ppbv, respectively, whereas from 14 to 18 October 2013, the maxima and minima is
350 observed to be 35 and 10 ppbv, respectively. Even though there was no solar radiation in the
351 evening hours, there are enhancements in surface ozone concentration (indicated by vertical
352 arrows) on 14-15, 16-17, 18-19 October 2013. The upper and lower average is indicated by
353 horizontal solid and dash lines respectively. The ozone profiles obtained from ozonesonde
354 measurements also show that enhanced ozone layer propagates downward from the upper
355 troposphere starting during 11-15 October 2013. It is well expected the enhanced ozone layer
356 further propagates downward to near-surface which is observed as enhanced as surface ozone
357 even there is a cut-off in solar radiation. Time-series of solar irradiation shows that there was
358 not much change in the radiation among the day 11-13 and 14-17 October 2013. This indicates
359 that the observed enhancement was not due to change in sunshine. At Trivandrum, ozone
360 remains to its daytime value even after evening hours. Interestingly, the enhancement

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365 observed on 14 October 2013 is during evening hours (16-17 hours) where solar radiation is
366 very low or even zero. Over the observation site, land-breeze prevails during night-time. The
367 change in night-time ozone depends on the precursor gas (e.g. NO) concentration in land-
368 breeze, which has dependency on local precursor gas emission/human activity. Due to the
369 cyclonic condition over Trivandrum, change in human activity during 11-17 October 2013
370 would not have happened considerably and Bio-mass burning may not be possible due to
371 rain. The day-to-day variability of surface ozone over Trivandrum is ~ 9.5 ppbv (1-sigma
372 standard deviation). The observed enhancement of about 10 ppbv in the day-time and 10-15
373 ppbv in the night-time is above the day-to-day variability which is attributed due to the
374 transport from the UTLS region. In a recent study by *Jiang et al. (2015)*, increase of surface
375 ozone by 21-42 ppbv and surface nocturnal surface ozone levels exceeding 70 ppbv is
376 observed in the region Xiamen and Quanzhou over the southeasteastern coast of China before
377 the Typhoon Hagibis landing. However, there are possible of influence of lightening
378 associated with cyclone and thus other possibility of this surface ozone cannot be fully ruled
379 out.

380 Further, to support the present observations of stratospheric intrusion into the troposphere
381 and further to surface, dynamical analysis is carried out using WRF-ARW simulation. *Das et*
382 *al. (2011)* and *Pan et al. (2015)* have shown ability of WRF simulation during the tropical
383 cyclone. Figure 5 shows the height-time cross-section of (a) vertical velocity along with
384 potential vorticity (magenta line) and potential temperature (black line) contours, and (b)
385 relative humidity along with equivalent potential temperature (black line) and zonal wind
386 (grey line) for Nilam (left panels) and Phailin (right panels) over Trivandrum using WRF
387 simulation. Figure 5 (a) shows the presence of strong updrafts (red) and downdrafts (blue)
388 marked with rectangle box at UTLS regions. Enhanced potential vorticity 0.5-1.5 PVU is
389 also observed vertically down from the stratosphere to the troposphere overlapping the

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Deleted: 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 NOx 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. The increase of 10-15 ppbv in near-surface ozone is mainly attributed due to the intrusion of air from the higher heights. ¶

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406 downdraft regions. The potential temperature contours indicate (Fig.3 (a)) the presence of
407 reduced stability during 29-31 October 2012 (Nilam) and 9-11 October 2013 (Phailin).

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

417 Similarly, Figure 6 shows the height-latitude cross-section of (a) vertical velocity along
418 with potential vorticity (magenta line) and potential temperature (black line) contours, and (b)
419 relative humidity cross-section along with equivalent potential temperature (black line) and
420 zonal wind (grey line) at 79°E at 18 GMT on 30 October 2012 for Nilam (left panels) and 18
421 GMT on 10 October 2013 for Phailin (right panels) using WRF simulation. The vertical
422 velocity profiles shows the presence of downdraft (blue) followed by updraft (red) between
423 8-17°N at the UTLS region in both the cyclone cases. Enhanced potential vorticity 0.5-1.5
424 PVU is also observed vertically down from the stratosphere to the lower troposphere,
425 overlapping the downdraft regions. It is also true that enhanced potential vorticity may be
426 also associated with diabatically by condensational heating but the enhancement is only
427 observed with the presence of downdraft at UTLS region. Thus, in the present study,
428 enhancement in the potential vorticity indicates the presence of stratospheric air in the
429 troposphere. The potential temperature contours indicate the presence of reduced stability of
430 the atmosphere at this location and noticed that stable stratospheric air penetrated downward

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442 at 12-14°N for Nilam and 16-18 °N for Phailin. Relative humidity profiles indicate the
443 presence of dry air at ~ 8°N which is in the vicinity of ozonesonde observational site. The
444 equivalent potential temperature contours in Figure 6 (b) indicate that from surface to 10 km
445 it is highly unstable for vertical motion and favourable condition for the convection to take
446 place at 6-12°N for Nilam and 12-18 °N for Phailin. In the same latitude regions, from 10 km
447 to the tropopause level, the vertical motion is suppressed and the atmosphere is found to be
448 statically stable to the unstratified atmosphere for both Nilam and Phailin. The present
449 condition indicates the presence of statically stable stratospheric air in the upper and middle
450 troposphere in the latitudinal cross-section at 79°E at 18 GMT on 30 October 2012 and 10
451 October 2013. Numerical simulation reproduced the key features and confirms the presence
452 of stratospheric intrusion in to the troposphere.

453 To get further insight, relative humidity derived from SAPHIR onboard the Megha-
454 Tropiques satellite is used. The relative humidity (daily mean) shown is an average over 12-
455 14 passes per day. Figure 7, shows the height-time intensity plot of daily mean relative
456 humidity during the passage of the cyclones: Nilam (left panel) and Phailin (right panel). The
457 grid is averaged from 4-8°N and 83-88°E. Strong dry air intrusion originated from lower
458 stratosphere is observed between 23-27 October 2012 (Nilam) and 12-18 October 2013
459 (Phailin). In both cyclones, dry air (low humidity region) reached down to the height of 8 km.
460 For the perception of spatial distribution of relative humidity, latitude-longitude plot of
461 relative humidity averaged over different height level is shown in Figure 8. The low value of
462 relative humidity i.e., the presence of dry air on the same day of enhanced ozone mixing ratio
463 in between 5 and 10 km prove that the dry air present in the upper and middle troposphere is
464 of stratospheric origin. The present observations show the influence of the tropical cyclone on
465 the air mass exchange from the stratosphere to the lower troposphere and redistribution of

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Deleted: The present observations show the influence of tropical storms on atmospheric composition especially ozone and water vapour and from its effect on weather system.

487 tropospheric ozone. Further analysis is required to quantify the amount of mass exchange
488 taking place between the stratosphere and the troposphere.

489 5. Summary and Conclusions

490 Important results brought out in the present analysis during the passage of a cyclonic

491 storms Nilam (2012) and Phailin (2013):

- 492 a) enhancement of upper tropospheric ozone by 20-50 ppbv from its climatological
493 mean is observed
- 494 b) The upper tropospheric ozone propagates downwards to the lower troposphere with a
495 rate of 0.8-1 K/day
- 496 c) About 10 ppbv in the day-time and 10-15 ppbv in the night-time enhancement of
497 surface ozone is noticed
- 498 d) Perturbation of the tropopause is also noticed
- 499 e) Numerical simulation shows the presence of stable dry ozone rich stratospheric air in
500 the upper and middle troposphere over the cyclone prone area

501 The present observation emphasizes the influence of the tropical cyclones in redistribution of
502 the tropospheric ozone and enriched surface ozone. The study clearly reveals that cyclone
503 plays a vital role in changing the atmospheric composition apart from general weather
504 phenomena.

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511 the Space Physics Laboratory (SPL) who participated in this STE-C campaign. The India
512 Meteorological Department (IMD) is highly acknowledged for providing the climatological

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Deleted: <#>Enhancement of the ozone by 20-50 ppbv is observed between 11 and 14 km compared to that of the background average profile.¶

<#>Enhanced ozone in the upper troposphere descend down to surface and the descent rate is found to be 0.87-1 km/day¶

Presence of dry air in the upper and middle troposphere during the period of enhancement of ozone in the middle/lower troposphere, which is of stratospheric origin is also been reported in both the cases.

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531 ozonesonde data. K.V. Suneeth and S. Aneesh are thankful to ISRO for providing doctoral
532 fellowship during the study period.

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712 **Figure Captions**

713 **Figure 1.** (a) Track of cyclones Nilam and Phailin (top panels) and (b) its Outgoing Long-
714 wave Radiation (OLR) wave radiation at 14:30 GMT on 30 Oct. 2012 (Nilam) and 9:00 GMT
715 on 10 Oct. 2013 (Phailin). In each panels, date and time is mentioned along the track. In first
716 panel, 18-1/11 indicates 18 GMT of 1 November 2012 and similarly followed for others.

717 Blue star in Fig.1(a) indicates the Ozonesonde launching site Trivandrum.
718 **Figure 2.** (a) Profiles of ozone mixing ratio (OMR) (dark black line) and relative humidity
719 (grey line) for individual days during passing of tropical cyclones (a) Nilam and (b) Phailin.
720 The mean ozone mixing ratio profile for non-convective days (as control day) is shown in
721 dotted line. Mean profile is obtained by averaging ozone data over Trivandrum for the month
722 of October from 1995-2013. Horizontal arrows indicate the height of enhanced ozone.

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

726 **Figure 4.** Time series of surface ozone mixing ratio along with solar radiation from 00 IST on
727 11 October 2013 to 23:55 IST on 19 October 2013. Solid and dotted horizontal lines indicate
728 the mean maximum and minimum surface ozone. The vertical arrows indicate the nocturnal
729 enhancement of surface ozone. The data is collected every 5 min.

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

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

749 **Figure 7,** Pressure-time cross-section of relative humidity obtained from SAPHIR onboard
750 Megha-Tropiques satellite during the cyclones Nilam (left panel) from 15 October to 10
751 November 2012 and Phailin (right panel) from 2 to 22 October 2013. The data is averaged
752 over from 40°N to 80°N and 83°E to 88°E.

753 **Figure 8,** Latitude-longitude distribution of relative humidity derived from SAPHIR onboard
754 Megha-Tropiques at different pressure levels (stamped on each panel) for Nilam (25 October
755 2012) and Phailin (14 October 2013). The data is averaged for one day which is about 12-14
756 passes at different timings and arrows indicate the presence of dry air.

757 **Table Captions**

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

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Deleted: 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 Oct. 2012 (Nilam) and 9:00 GMT on 10 Oct. 2013 (Phailin). In each panels, date and time is mentioned along the track. In first panel, 18-1/11 indicate 18 GMT of 1 November 2012 and similarly followed others. ¶

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

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 GMT on 30 October 2012 for Nilam (left panels) and 20 GMT on 10 October 2013 for Phailin (right panels). (c) and (d) Same as Fig. 4a and 4b, respectively but for height-time cross-section over Thumba (8.5°N, 76.9°E). The above parameters are obtained from WRF simulation. ¶

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 Oct. 2012).

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