



Trend analysis of atmospheric temperature, water vapour, ozone, methane and carbon-monoxide over few major cities of India using satellite data

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In this study, decadal trend analysis of atmospheric temperature, water vapour, ozone, methane and carbon-monoxide has been presented over few major cities of India using Aqua-AIRS products from 2003 to 2012. The atmospheric column is studied in few atmospheric layers, viz., surface-850, 850–500, 500–100, 100–50 and 50–1 hPa for temperature, water vapour and ozone. However, CH₄ and CO results are presented in total column amounts. Non-parametric Mann–Kendall test has been applied to investigate the trends of annual means of parameters and Sen's slope estimate has been used to find the rate of the change, if there is a trend. The layer average temperature (LAT) has been found to be increasing in lower troposphere (surface-850 hPa) and decreasing in lower stratosphere (100–50 hPa). The warming trend over Chennai is found to be not limited in lower tropospheric region, but extended in 850–500 hPa layer also. However, LAT (850–500 hPa) has decreasing trend over Thiruvananthapuram. LAT in 500–100 hPa has significant decreasing trend only over Ahmedabad. The decreasing LAT trend in 100–50 hPa is quite prominent with significant decreasing trends over Mumbai, Ahmedabad, Kolkata and Hyderabad. The layer integrated water vapour (LIWV) is found to be increasing mainly in surface-850 hPa and 850–500 hPa layers. The decreasing trend of LIWV has been observed only over Ahmedabad in 500–100 hPa layer. For total column water vapour, the trends are mostly increasing, however, it is statistically significant only over Hyderabad. The layer integrated ozone has been found to be increasing in troposphere and decreasing in lower stratosphere. The increasing trend of ozone in troposphere is most prominent in lower-mid tropospheric region (850–500 hPa layer). No significant trend has been observed for total column ozone. Total column methane has shown significant increasing trend over all cities with very good significance level. However, for total column carbon-monoxide, the trends are decreasing and the decreasing trends are significant over Delhi and Mumbai.

Keywords. Trend analysis; Mann–Kendall test; AIRS.

1. Introduction

These days, dealing with climate change and climate variability is one of the greatest challenges. The various studies have been conducted for

estimating the trends in various parameters including atmospheric parameters like temperature, water vapour and trace gases over regional, national and continental levels. The present study is aimed at evaluating the trends of atmospheric

parameters over few major cities of India using satellite data. The atmospheric parameters considered in the study are temperature, water vapour, ozone, methane and carbon-monoxide.

According to IPCC (2007), the Earth's temperature has been found to be increasing since 1975 at local, regional and global scales. Hertig *et al.* (2010) have analysed the trend of extreme temperature of Mediterranean during the time period from 1961 to 1990 using daily station data. They saw the increasing trend over western Mediterranean area; however, an opposite trend was identified over eastern Mediterranean region. Unkasevic *et al.* (2005) have shown an increasing trend in summer mean temperature at Belgrade. For Korean region, using the temperature data for time period 1974–1997, Chung and Yoon (2000) found that the annual mean temperature was increased by 0.96 K during the period. They also showed that the increase in temperature was 0.58 K for the rural and coastal areas and 1.5 K for cities. Daniel *et al.* (2014) have shown that there was a statistically significant increasing trend of temperature over the upper Blue Nile River basin of Ethiopia. Also for southwestern region of Ethiopia, Jury and Funk (2012) revealed increasing temperature trend during the time period from 1948 to 2006. For Indian region, India Meteorological Department (IMD) has maintained a well distributed network of more than 500 stations for more than a century. It has been shown that the annual mean temperature of the country has increased by 0.56 K in the time period from 1901 to 2009 (Attri and Tyagi 2010). However, the global increase in temperature is about 0.74 K (IPCC 2007). Except over parts of Gujarat, Bihar and Rajasthan, where the temperature trends are significantly decreasing, the mean annual temperature has shown the statistically significant increasing over most parts of India.

Water vapour is one of the most important constituent of the atmosphere and carries a large amount of latent heat. The variations in atmospheric water vapour concentrations is strongly related to variations in global radiation budget, which further influences the atmospheric temperature, rainfall and droughts (Wang *et al.* 2017). Various studies have been done over the years to quantify the changes in the atmospheric water vapour. Ross and Elliott (2001) have shown that there was an increasing trend of water vapour over Northern Hemisphere except Europe and the increasing rate was more than 3%/decade over some islands in the western tropical Pacific and

most of the areas of East China and North America during 1973–1995. Mieruch *et al.* (2008) used the satellite data from Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCHIMACHY) and Global Ozone Monitoring Experiment (GOME) from 1996 to 2006 to examine the water vapour over the global scale. Their results indicated significant decreasing trends of water vapour over northwest USA, central America, Amazon, Arabian peninsula and central Africa, but the water vapour trends were found to be significantly increasing over Oceania, Greenland, Siberia and East Europe. Sun *et al.* (2000) demonstrated the increasing trend of surface water vapour over eastern China, tropical western Pacific islands and former Soviet Union during second half of the 20th century. Trenberth *et al.* (2005) used Special Sensor Microwave Imager (SSM/I) data over ocean for time period 1988–2003 and concluded that the atmospheric precipitable water (PWV) has increasing trend over whole ocean and the rate of increase was $1.3 \pm 0.3\%$ /decade.

Ozone distribution in the atmosphere is such that the ozone concentration peaks in the stratosphere and about 90% of ozone is lying in stratosphere. Although quantitatively tropospheric ozone is very less as compared to stratospheric ozone, it has a significant role in various physical and chemical process of the troposphere. Good number of studies has been conducted to understand the tropospheric and stratospheric ozone trends along with total column ozone trends. In addition to discovery of Antarctic ozone hole, a significant falling trend in global total column ozone has been observed during 1980s (Solomon *et al.* 1986). In the recent past, the recovery of Antarctic ozone hole has been reported by Solomon *et al.* (2016); however, the total ozone over non-polar regions has been found be stable since 2000 (WMO 2014). The tropospheric and stratospheric ozone trends are found to have complex pattern and are also under regional influences (Staehelin *et al.* 2001). Bojkov and Reinsel (1984) used ozonesonde data and showed that the ozone in 850–400 hPa layer has increased at a rate of $1.12 \pm 0.54\%$ /year during 1970–1981 over Northern Hemisphere. The increasing trends of tropospheric ozone over Indian cities has been shown by Kulkarni *et al.* (2010). Using Ozone Mapping Instrument (OMI) data, Nishanth and Satheesh (2011) reported that the rate of increase of tropospheric ozone is about 8.8%/year over the Indian coastal cities. Saraf and Beig (2004) showed that tropospheric ozone had no

statistically significant trends over Trivandrum, but significant rising trend over Pune and Delhi was observed using ozonesonde data during 1972–2001. Many studies have found that the ozone has decreasing trends in lower stratosphere (Gebhardt *et al.* 2014; Sioris *et al.* 2014; Vigouroux *et al.* 2015). The reduction in stratospheric ozone by chlorofluorocarbons was first demonstrated by Rowland and Molina (1975). Also, as per the report of WMO (2014), the global ozone concentration in stratosphere has decreased from 1980s to mid-1990s. However, since 1990s, decline in decreasing trend of ozone concentration in stratosphere has been observed (Newchurch *et al.* 2003).

Methane with global warming potential of 28 times more than carbon-dioxide over 100 yrs (IPCC 2014), is one of most important anthropogenic greenhouse gas. Methane has increased at a sharp rate in last few centuries with a concentration of 700 ppb in 1750 to 1808 ppb in 2010 (IPCC 2014). The natural sources of CH₄ are wetlands, termites and oceans which contribute 36% of total amount of CH₄ emissions. However, anthropogenic activities including landfills, livestock farming, fossil fuels burning and agriculture accounts for 64% of total CH₄ emissions (Bousquet *et al.* 2006; Kirschke *et al.* 2013). The rate of growth of CH₄ concentration has been observed to be about 0.9%/annum since 1800s (Bubier and Moore 1994). However, the long term data analysis of methane has shown that in recent past, the rate of growth of methane has been reduced (Dlugokencky *et al.* 1998). Although the exact cause of decline in methane growth rate is not well understood, reduction in anthropogenic emissions and drought in wetlands are thought of possible reasons for observed variation in CH₄ increase rate.

Atmospheric carbon-monoxide is an important trace gas having significant role in atmospheric chemistry and climate change with direct radiative forcing of 0.024 Wm⁻² and indirect radiative forcing of 0.23 Wm⁻² (IPCC 2014). The two main source of CO includes biomass burning and fossil fuels combustion, and oxidation of hydrocarbons in the troposphere; and the main sink of CO is oxidation by OH radicals (Wallace and Hobbs 2006). Li and Liu (2011) have shown the decreasing trend of CO over some Chinese cities using surface observations for more than 10 yrs. Novelli *et al.* (2003) and Angelbratt *et al.* (2011) have also reported declining CO concentration over USA and Europe, respectively, again using *in-situ* observations. Satellite data have also proved the

decreasing trends of CO over most parts of the globe (Warner *et al.* 2013; Worden *et al.* 2013).

In the present study, satellite data has been used to analyse the decade trends of atmospheric temperature profile, water vapour profile, ozone profile, total column methane and total column carbon-monoxide over few major cities of India. The attempt has been done using Atmospheric Infrared Sounder (AIRS) sounding products. The atmospheric profiles from AIRS have been analysed by taking the averages in different atmospheric layers. Also, the trends are investigated by taking annual means and using non-parametric Mann–Kendell test. The rate of the change of the parameter has also been estimated using Sen's slope estimation method, if the trend is found.

2. Study area and data used

In the present study, the trend analysis of atmospheric temperature profile, water vapour profile, ozone profile, total column methane and total column carbon-monoxide has been done over few cities in Indian region. The atmospheric profiles were divided into various atmospheric layers, viz., surface-850, 850–500, 500–100, 100–50 and 50–1 hPa layers. The temperature was studied as layer average temperature in first four layers. The water vapour was analysed as layer integrated water vapour in first three layers and ozone as layer integrated ozone in all five layers in addition to total column water vapour and total column ozone. Cities considered in the study are Delhi, Mumbai, Ahmedabad, Kolkata, Chennai, Thiruvananthapuram and Hyderabad. Locations of these cities are shown in figure 1.

AIRS sounding products have been utilized for the present study on trend analysis of atmospheric parameters. AIRS is first of the new generation of hyperspectral infra-red sounders. It was launched on-board Aqua satellite in 2002. It is orbiting in a polar sun-synchronous orbit and its local equatorial crossing times are 1:30 am and 1:30 pm. AIRS has 2378 channels with a spectral resolution of $\lambda/d\lambda = 1200$ and each channel is sensitive to a particular layer in the atmosphere. As the AIRS channels are sensitive at different heights in the atmosphere, the vertical profiles in the atmosphere can be retrieved. So, the AIRS data products include good vertically resolved temperature and water vapour profiles in addition to ozone, methane, carbon-monoxide and other trace gases. These products are being used in

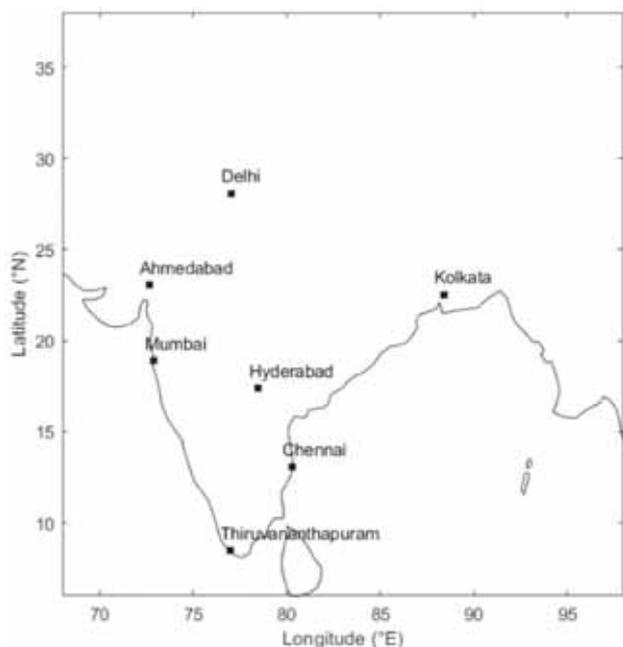


Figure. 1. Location of major cities considered over India.

various applications like climate change studies, numerical weather prediction models, etc., AIRS being working in IR region is not providing data below clouds. Therefore, to cover cloudy areas, Advanced Microwave Sounding Unit-A (AMSU-A) was also launched along with AIRS onboard Aqua Satellite. In the study, the products used were retrieved using both AIRS and AMSU-A data to provide full coverage. AMSU-A footprint size is approximately 45 km in which there are nine footprints of AIRS of size about 15 km. So, the data used is of 45 km horizontal resolution. AIRS level 2 standard physically retrieved products used in the study was downloaded from <http://mirador.gsfc.nasa.gov/>. Daily descending pass, i.e., night-time data was used for the 10 yrs period from January 2003 to December 2012. The data available at Mirador–Goddard Earth Science Data Information and Services Centre has quality screening option. Three quality options, viz., 0, 1 and 2 are available. The quality = 0 means that the data is of best quality; quality = 1 signify good quality and it was recommended to use when temporally and/or spatially averaged; and the datasets with quality = 2 are not recommended to use (Fishbein *et al.* 2007). The parameters acquired for the present study are: surface air temperature, atmospheric temperature profile, water vapour profile, ozone profile, total column ozone (TOZ), total column CH₄ (TCH₄), total column water vapour (TWW)

and total column CO (TCO). The focus of the present study is on trend analysis and therefore, the data is temporally averaged. Hence, the AIRS+AMSU-A retrieved level 2 data product with quality 0 and 1 is used in the present study. The atmospheric temperature and ozone profiles used in the study are available at 28 standard pressure levels (in hPa): 1100, 1000, 925, 850, 700, 600, 500, 400, 300, 250, 200, 150, 100, 70, 50, 30, 20, 15, 10, 7, 5, 3, 2, 1.5, 1.0, 0.5, 0.2 and 0.1. And water vapour profile is available at first 15 levels of the standard pressure levels. The temperature (in K) is a level quantity and hence reported at various levels above the surface. The water vapour is available in mass mixing ratio (g/kg). It is layer average quantity of the layer between two standard pressure levels and is given at lower level bounding the layer. The water vapour at the lowest level above the surface is the average values of the layer bounded by surface and the next higher level. TWV (in kg/m²) is the integrated water vapour in atmospheric column from top of atmosphere (TOA) to the surface. Here, the TOA is considered at 0.005 hPa. The ozone is also a layer average quantity provided in volume mixing ratio (ppmv). It is also reported at lower level of the layer bounded by two levels and the values at the lowest level corresponds to average volume mixing ratio of the layer bounded by surface and the next higher level. TOZ in DU (dobson unit) is integrated column amount of ozone from TOA to the surface. TCH₄ and TCO available in molecules/cm² are integrated column amounts of methane and carbon-monoxide, respectively, from TOA to the surface (Fishbein *et al.* 2007).

3. Methodology

AIRS level 2 standard physically retrieved quality control products were acquired over the cities considered (Delhi, Mumbai, Ahmedabad, Kolkata, Chennai, Thiruvananthapuram and Hyderabad) in the study for 10 years from January 2003 to December 2012 from <http://mirador.gsfc.nasa.gov/>. The products acquired were the temperature profile, water vapour profile, ozone profile, TWV, TOZ, TCO, TCH₄ and few other necessary parameters like standard pressure levels, surface pressure, surface air temperature, etc. Then layer average temperature (LAT) was calculated for four atmospheric layers: surface–850, 850–500, 500–100 and 100–50 hPa using the temperature profiles.

Water vapour profiles were used to calculate layer integrated water vapour (LIWV) for three atmospheric layers: surface-850, 850–500 and 500–100 hPa as follows:

$$\text{LIWV} = \frac{1}{g} \left(\sum_i wv_i(p_i - p_{i-1}) \right) \times 0.1,$$

where p_i and p_{i-1} are pressure in hPa at i^{th} and $(i-1)^{\text{th}}$ levels; wv_i is water vapour in g/kg in the layer bounded by pressure levels p_i and p_{i-1} ; $g = 9.8 \text{ m s}^{-2}$ is acceleration due to gravity. Here LIWV is in kg/m^2 . Layer integrated ozone was calculated for five atmospheric layers: surface-850, 850–500, 500–100, 100–50 and 50–1 hPa from ozone profiles using the following formula

$$\text{LIOZ} = 0.78961 \times \left(\sum_i oz_i(p_i - p_{i-1}) \right),$$

where p_i and p_{i-1} are pressure in hPa at i^{th} and $(i-1)^{\text{th}}$ levels; oz_i is ozone in ppmv in the layer bounded by pressure levels p_i and p_{i-1} . Here LIOZ is in DU. Then the annual averages of these parameters were calculated over various cities of the country. Now, the data is ready for trend analysis.

Trend of annual means of these parameters was analysed along with significance level using statistical tests. To choose the statistical test for trend study, the data distribution should be known. In this study, the box plots were used to see if the data was normally distributed (Jindal *et al.* 2019). Box plots of annual means of various parameters considered in the study were plotted (not shown here) over different major cities considered. The box plots showed that the data was not normally distributed, therefore, the non-parametric statistical test is expected to give better results as compared to parametric tests. Hence, as described by Jindal *et al.* (2019), non-parametric Mann–Kendall statistical test was implemented in the present study to see if there was a trend in the dataset.

If a trend was found, then a non-parametric Sen’s slope estimator was used to compute the magnitude of the trend. In this method, first the slopes were estimated for all data pairs as:

$$Q_i = \frac{Y_m - Y_l}{T_m - T_l},$$

where Y_m and Y_l are data values at times T_m and T_l , respectively, and $m > l$. Then all these slopes Q_i

were arranged in ascending order of their values and the median of these values gave estimate of Sen’s slope Q .

4. Results and discussion

In this section, the annual means of LAT, LIWV and LIOZ in various atmospheric layers and TWV, TOZ, TCH₄ and TCO over few major cities (Delhi, Mumbai, Ahmedabad, Kolkata, Chennai, Thiruvananthapuram and Hyderabad) of India have been analysed to find trends using non-parametric Mann–Kendall test. The non-parametric statistical test is chosen because the data is found to be not normally distributed using box plots (not shown here). If a statistically significant trend is found for any parameter, the magnitudes of trends have been estimated using Sen’s slope estimator. A significance level of 0.1 (10%) is chosen in the study for Mann–Kendall test. 10% significance level gives the strong evidence against the null hypothesis H_0 with a 1 in 10 chances of the conclusion made being incorrect. Figures 2, 4, 6 and 8 show the time series plot of various parameters over all the cities considered in the study. The circles in the figures represent the annual mean values and thick line represent the annual trend line. The annual trend line has been plotted only for the cities for which the statistical significant trend is found and the slope estimated in the study is used to draw the annual trend line. The results of Mann–Kendall test and Sen’s slope estimate for statistically significant trend are presented in figures 3, 5, 7 and 9 for all the parameters considered in the study over different cities. The Mann–Kendall test results have been shown by bar graph of P -values corresponding to different Z statistic values. The positive Z values have been plotted as grey bar, zero Z values as black bar and negative Z values as white bar. As the significance level chosen in the study is 0.1, a dashed line has been plotted at P -value = 0.1 to have clear view of statistically significant trends. For P -value less than 0.1, i.e., statistically significant trend, the estimated slope using Sen’s slope estimator is written on the top of the bar. Following subsections present the trend results for LAT, LIWV, TWV, LIOZ, TOZ, TCH₄ and TCO.

4.1 Layer average temperature

Figure 2 presents the time series plot of annual means of LAT in surface-850, 850–500, 500–100

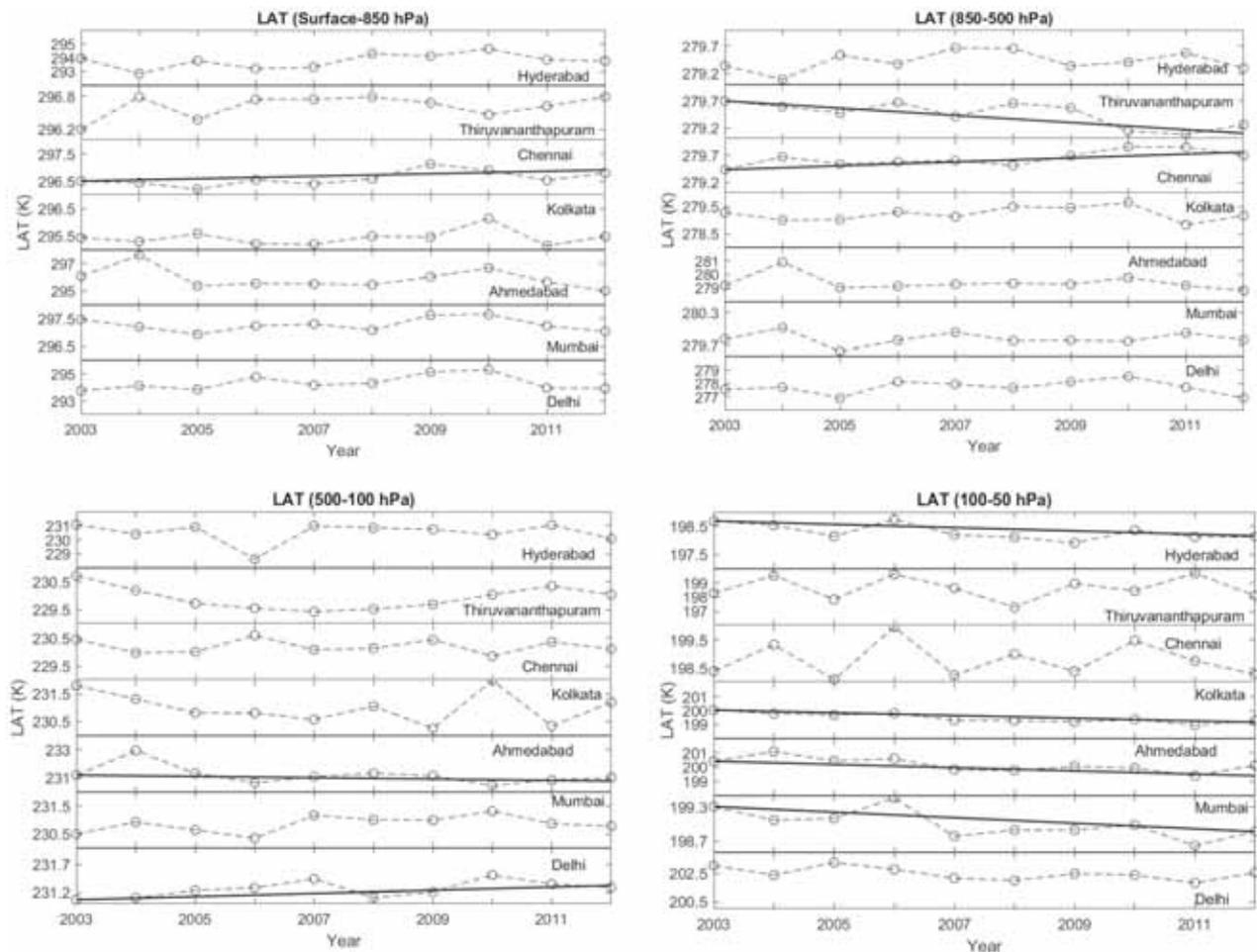


Figure 2. Time series plot of annual means (circles) of layer average temperature in different atmospheric layers over various cities of India. Annual trend line (thick line) is also plotted for the cities where statistically significant trend was observed.

and 100–50 hPa atmospheric layers for few major cities of the country. The annual trend line of LAT has also been plotted over the cities where statistical significant trend is seen in the study. The results of Mann–Kendall test for LAT (surface-850 hPa), LAT (850–500 hPa), LAT (500–100 hPa) and LAT (100–50 hPa) along with slope estimates for statistically significant trend have been presented in figure 3. As the Z value for LAT (surface-850 hPa) is positive for most of cities, the temperature may be increasing in surface-850 hPa atmospheric layer. However, the P -value for Z is less than significance level of 0.1 only over Chennai. Therefore, the null hypothesis of no significant trend is rejected and the increasing trend is found to be statistically significant only over Chennai. For other cities, where P -value is greater than 0.1, the null hypothesis is not rejected and hence, the trends are non-significant. Again, as the trend is significant only over Chennai, the Sen's slope estimator has been applied only for Chennai and

the rate of the change of LAT in surface-850 hPa layer is found to be 0.045 K/year. For LAT (850–500 hPa), the trends are statistically significant only over Chennai and Thiruvananthapuram as the P -value is less than significance level only for these two cities. As the Z value is positive for Chennai, the significant trend is increasing over Chennai, and the trend is significantly decreasing over Thiruvananthapuram as the Z value is negative. Also, the Sen's slope estimator shows that the temperature is increasing at a rate of 0.036 K/year over Chennai and decreasing at a rate of 0.066 K/year over Thiruvananthapuram. For LAT (500–100 hPa), again the trend is not significant except over Delhi where it is increasing and Ahmedabad where it is decreasing. The increasing rate for LAT in 500–100 hPa layer is 0.028 K/year over Delhi and decreasing rate is 0.047 K/year over Ahmedabad. The LAT trends are decreasing over most of the cities in 100–50 hPa layer with statistically significant decreasing trend over Mumbai,

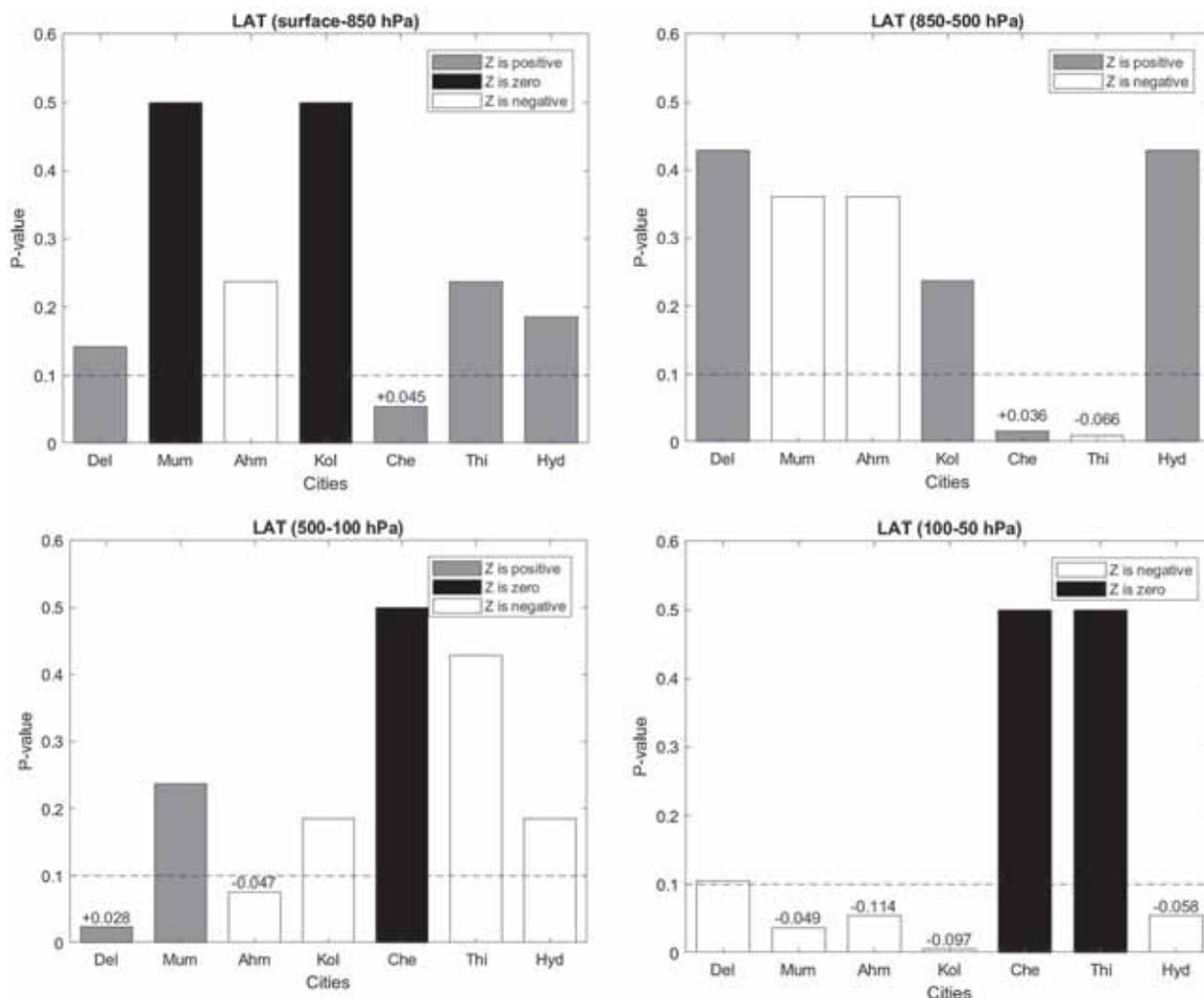


Figure 3. Mann–Kendall test results and Sen’s slope estimate for LAT in various atmospheric layers. *P*-value corresponding to *Z* statistics is plotted in Y-axis for various cities on X-axis. Grey bar corresponds to positive *Z* value, black bar corresponds to zero *Z* value and white bar corresponds to negative *Z* value. The values written on top of few bars are slopes (in K/year) of statistically significant trends.

Ahmedabad, Kolkata and Hyderabad. The slope of the decreasing trend in 100–50 hPa is 0.049, 0.114, 0.097 and 0.058 K/year, respectively, over Mumbai, Ahmedabad, Kolkata and Hyderabad. So in general it can be concluded that the temperature is increasing near the surface and decreasing in lower stratosphere. Many earlier studies have reported such trends of increasing temperature near the surface and decreasing temperature in lower stratosphere (Kothawale and Rupa Kumar 2005; Attri and Tyagi 2010; Kothawale and Singh 2017; Raj *et al.* 2018). The greenhouse effect of increasing trace gases and water vapour near the surface is major reason for increase in near surface temperature. The increased water vapour have significant positive feedback on surface temperature (Hansen *et al.* 1984). The increasing tropospheric water

vapour and tropospheric ozone trends have also been shown in the subsequent subsections. The increased surface temperature further increase the water holding capacity of the air, due to which there is further increase in water vapour, which again increases the temperature near the surface by acting as greenhouse gas. The decrease in temperature in lower stratosphere may be due to decreasing ozone concentration in that region. Ozone present in stratosphere absorbs ultraviolet rays from Sun and keep the stratosphere warm. So if there is decrease in ozone concentration in stratosphere, there may be temperature decrease also (Forster *et al.* 2007; Braesicke *et al.* 2011). Peethani *et al.* (2014) have also showed a good correlation between ozone concentration and temperature in stratosphere. The decreased ozone

concentration in lower stratosphere is also discussed in subsequent subsection. Another reason for decrease in stratospheric temperature is the increased concentration of greenhouse gases in troposphere. This is because with increase in greenhouse gases in troposphere, the thermal radiation passing through it decreases and hence lower amount of radiations reach the stratosphere.

From figure 3, it can also be noticed that the positive temperature trend in surface-850 hPa layer is statistically significant only over Chennai. Ray *et al.* (2019) have suggested that increase in urbanization is an important reason for increase in lower tropospheric temperature. It has also been reported that Chennai is 100% urbanized with population density of 26,903 per km² (Jeganatham and Andimuthu 2013) and Chennai is largest contributor of CO₂ emission in India among major metropolitan cities (Ramachandra and Shwetmala 2009). Over Chennai, the temperature has also statistically significant increasing trend in 850–500 hPa layer which is mainly due to increase in ozone in this layer (shown in subsequent subsections) over Chennai, although some effect of larger CO₂ emission is also there. However, the temperature trend is decreasing over Thiruvananthapuram in 850–500 hPa layer. This may be due to reason that there is no increasing trend of water vapour and ozone (figures 5 and 7), and there is decreasing trend of ozone with *P*-value of 0.186 (close to 0.1, figure 7) in 850–500 hPa layer over Thiruvananthapuram. In 500–100 hPa layer, temperature has significant decreasing trend over Ahmedabad which may be due to decreasing trend of water vapour in 500–100 hPa layer over Ahmedabad (figure 3).

4.2 Layer integrated water vapour and total water vapour

In this subsection, the results of non-parametric Mann–Kendall test have presented for LIWV in surface-850, 850–500 and 500–100 hPa atmospheric layers alongwith TWV over different cities of India. The Sen's slope has been estimated for statistically significant trend and the slopes are mentioned as percentage change per year. Figure 4 presents the time series plots of LIWV in various atmospheric layers and TWV along with statistically significant annual trend lines. The increasing trend line over Delhi and Hyderabad in surface-850 hPa, increasing trend line over Hyderabad in 850–500 hPa, decreasing trend line over

Ahmedabad in 500–100 hPa and increasing trend line over Hyderabad for TWV can be seen from the figure. Mann–Kendall test results and Sen's Slope estimate for LIWV in various atmospheric layers and TWV are shown in figure 5. LIWV in surface-850 hPa layer shows increasing trend almost over all cities except Ahmedabad and Chennai, but the trend is statistically significant over Delhi and Hyderabad as the *P*-value for *Z* is less than significance level (0.1) only over Delhi and Hyderabad. The LIWV (surface-850 hPa) is increasing at a rate of 0.580%/year and 1.663%/year over Delhi and Hyderabad, respectively. For LIWV in 850–500 hPa atmospheric layer, statistically significant trend is increasing only over Hyderabad with a slope of 2.266%/year. LIWV (500–100 hPa) has significant decreasing trend only over Ahmedabad with decrease of 2.718%/year. It can be seen that the water vapour is increasing in lower and lower-mid part of the troposphere. As already seen in previous section, there is increase in near surface temperature which may increase the water holding capacity of the air and hence increase in near surface water vapour. Moreover, this increase in water vapour further provides positive feedback for temperature to increase near the surface by acting as a greenhouse gas. TWV has also been found to have significant increasing trend over Hyderabad at a rate of 2.274%/year, which is due to increase in water vapour in various atmospheric layers.

Also from figures 3 and 5, it can be seen that the LAT is decreasing over Ahmedabad in all atmospheric layers considered, but the trend is significant in 500–100 and 100–50 hPa layers and, LIWV is decreasing over Ahmedabad in all atmospheric layers considered, but the trend is significant in 500–100 hPa. However, both LIWV and LAT in 500–100 hPa layer are decreasing over Ahmedabad. So, here again the positive feedback of water vapour (or temperature) on temperature (or water vapour) may be the reason for simultaneous decrease in temperature and water vapour over Ahmedabad.

4.3 Layer integrated ozone and total column ozone

As done in previous subsections, here also non-parametric Mann–Kendall test results have been presented for LIOZ in various atmospheric layers and TOZ. Figure 6 shows the time series plots and figure 7 shows Mann–Kendall test

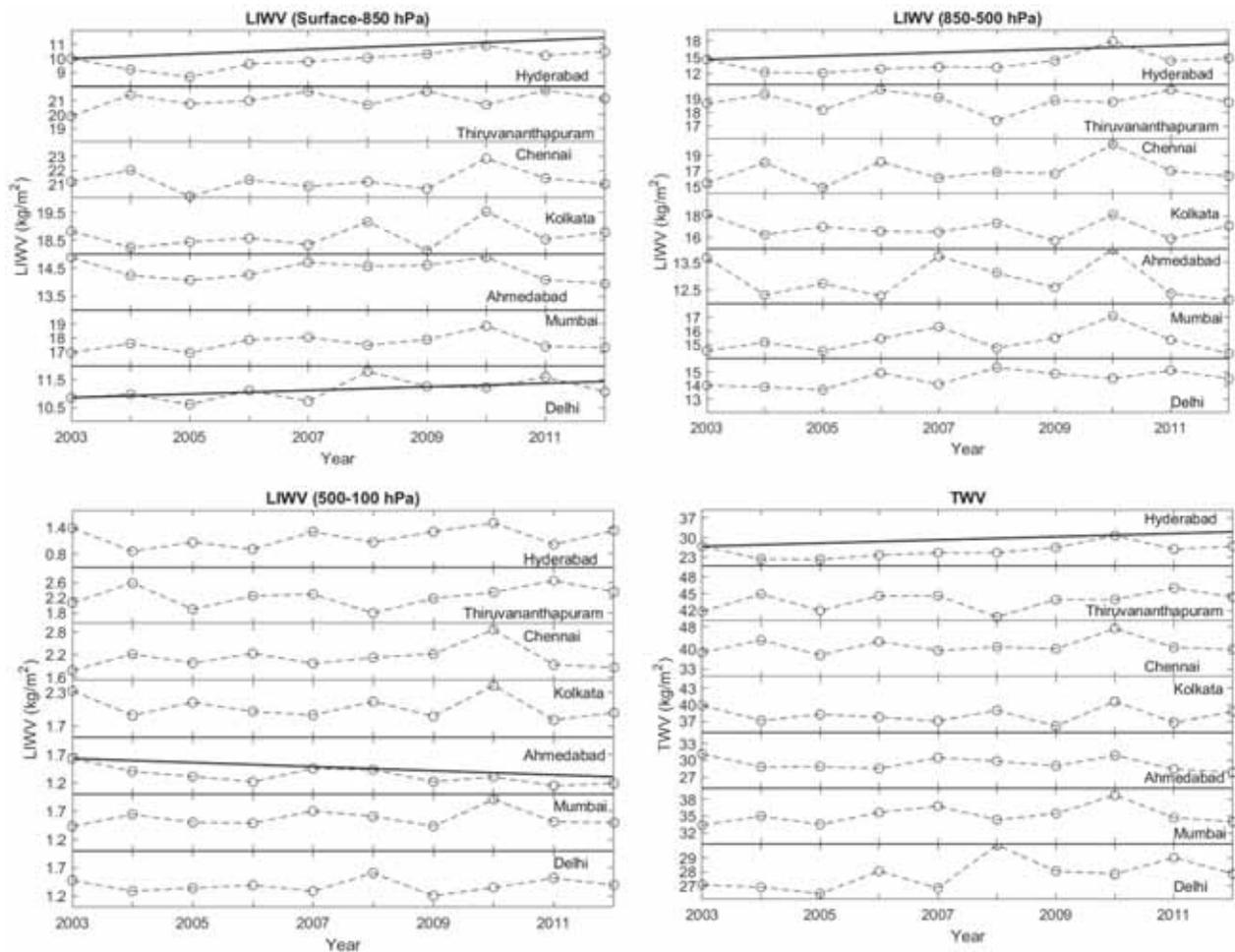


Figure 4. Time series plot of annual means (circles) of layer integrated water vapour in different atmospheric layers and total water vapour over various cities of India. Annual trend line (thick line) is also plotted for the cities where statistically significant trend was observed.

statistics and Sen’s slope estimates for LIOZ (surface-850 hPa), LIOZ (850–500 hPa), LIOZ (500–100 hPa), LIOZ (100–50 hPa), LIOZ (50–1 hPa) and TOZ over different cities of India. Here again, the significant slopes have been reported in percentage change per year. LIOZ (surface-850 hPa) has been found to have statistically significant increasing trend only over Ahmedabad with the slope of 0.142%/year. For LIOZ (850–500 hPa) also, the trend is increasing over most of the cities, however, the trend is significant over Mumbai, Ahmedabad and Chennai with the increase rate of 0.238%/year, 0.269%/year and 0.348%/year, respectively. The results for LIOZ in surface-850 and 850–500 hPa layers showed that the increase in ozone is more prominent in 850–500 hPa layer as compared to surface-850 hPa layer. Even the percentage increase in ozone is more in 850–500 hPa layer (0.269%/year for Ahmedabad) than surface-850 hPa layer (0.142%/year for Ahmedabad). Even

in 500–100 hPa layer, the trends have been found to be increasing, and the increasing trends are statistically significant over Mumbai and Chennai with the rate of increase of 0.229%/year and 0.415%/year, respectively. It can be concluded that the ozone has increasing trend in whole troposphere with most prominent increase in 850–500 hPa (lower-mid troposphere) layer. The higher increase of ozone in 850–500 hPa layer as compared to surface-850 hPa has also been reported in previous studies, e.g., Oltmans *et al.* (2013) used the ozonesonde data for the time 1991–2010 and showed that the ozone was increased by $0.33 \pm 0.14\%$ /year in 850–700 hPa layer as compared to the increase by $0.27 \pm 0.22\%$ /year in surface-850 hPa layer over Wallop’s Island. For Goose Bay, Resolute and Edmonton in Canada also, they found that the increase in ozone was more in 850–700 and 700–500 hPa layers than surface-850 hPa layer. For LIOZ (100–50 hPa), the trends are

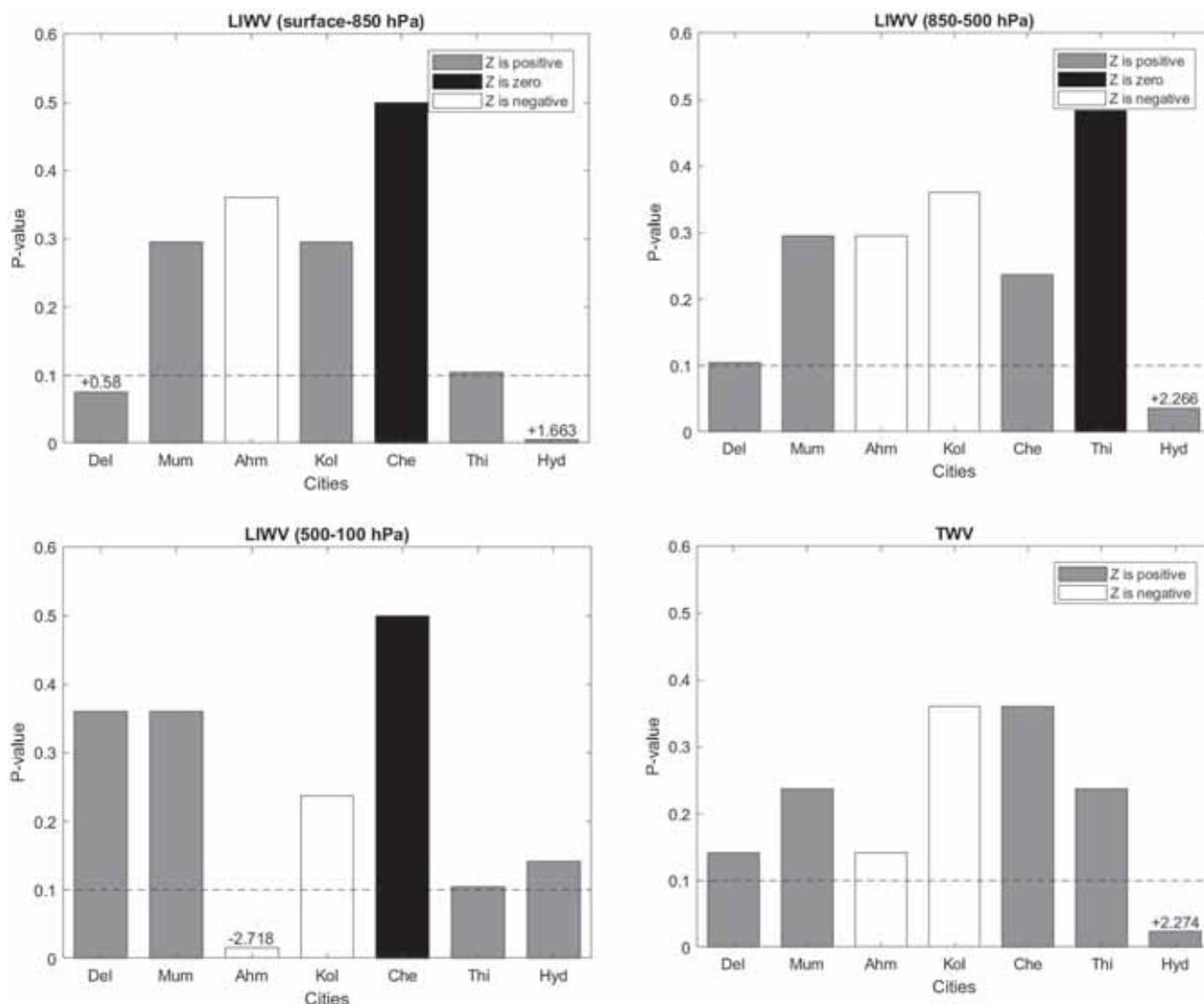


Figure 5. Mann-Kendall test results and Sen's slope estimate for LIWV in various atmospheric layers and TWV. P -value corresponding to Z statistics is plotted in Y-axis for various cities on X-axis. Grey bar corresponds to positive Z value, black bar corresponds to zero Z value and white bar corresponds to negative Z value. The values written on top of few bars are slopes (in %/year) of statistically significant trends.

mostly decreasing and decreasing trend is significant only over Ahmedabad. The slope of the decreasing trend of LIOZ in 100–50 hPa layer is 0.236%/year for Ahmedabad. The LIOZ trends are not significant over any city in 50–1 hPa atmospheric layer. So, in general, the ozone concentration is increasing in troposphere with maximum increase in lower-mid troposphere; however, the ozone is decreasing in lower stratosphere. The increasing concentration of ozone in troposphere is due to increase in ozone precursor pollutants (IPCC 2001). Nair *et al.* (2018) have also reported a positive correlation of surface ozone with NO_x and CH₄. The increasing concentration of CH₄ has also been shown in next subsection. The decreasing concentration of ozone in 100–50 hPa atmospheric

layer (lower stratosphere) may be due to tropical upwelling and stratospheric meridional circulation (Fadnavis *et al.* 2010). Moreover, the ozone in stratosphere is also affected by methane when it is oxidised by ozone to form water vapour in stratosphere. So, increasing concentration of methane may be another possible reason for decreasing ozone in stratosphere. TOZ has not shown any significant trend over any city. The contrasting increase and decrease of ozone in different layers may be partially responsible for no net trend in total column ozone. Tandon and Attri (2011) have also shown that there is no significant trend for TOZ over 60% of the Indian geographical region; however, 40% of the region has declining TOZ trends.

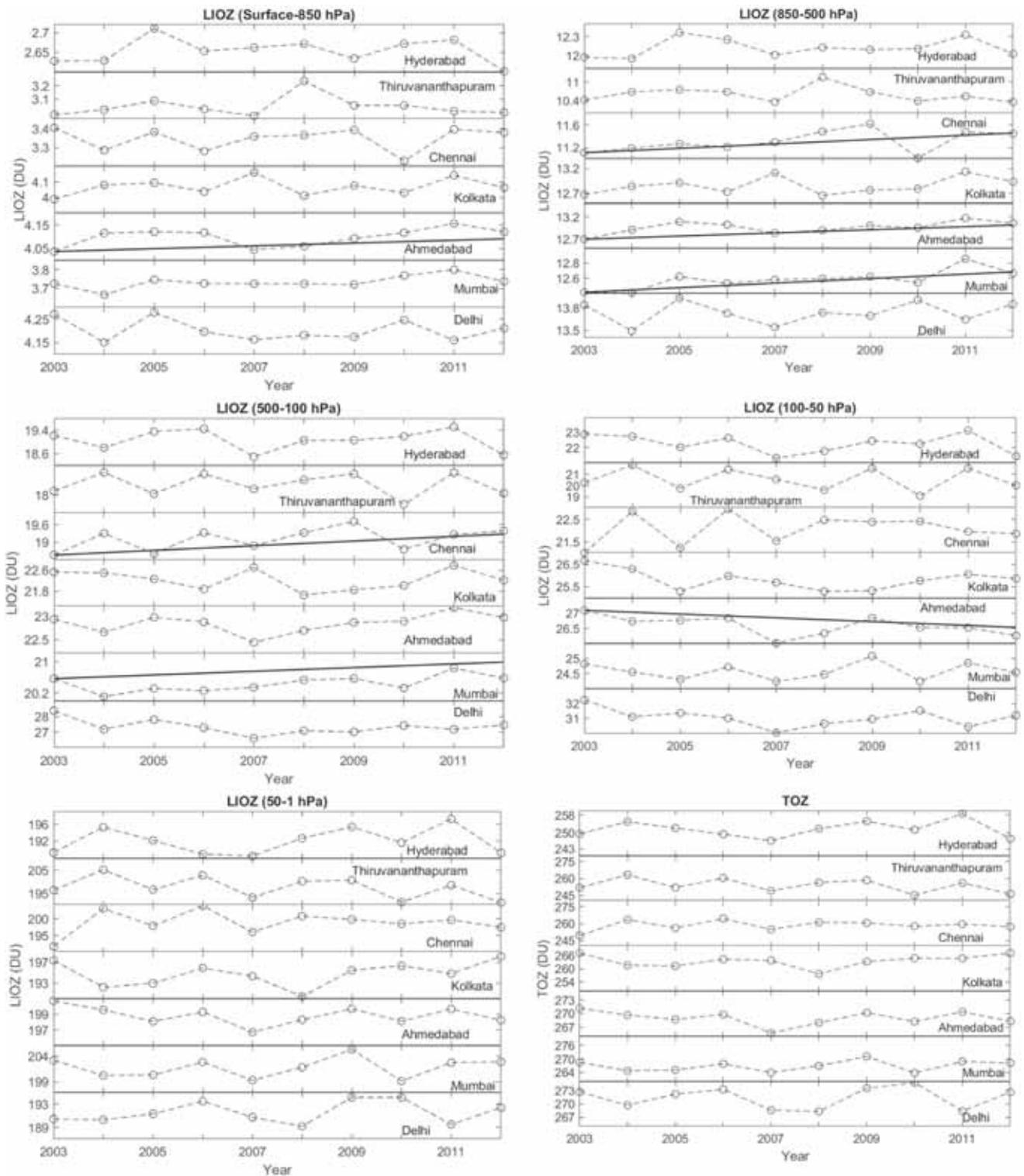


Figure. 6. Time series plot of annual means (circles) of layer integrated ozone in different atmospheric layers and total column ozone over various cities of India. Annual trend line (thick line) is also plotted for the cities where statistically significant trend was observed.

As discussed in previous subsections, the decrease in ozone in lower stratosphere is one of the possible reasons for decrease in temperature there,

and the increase in ozone in lower troposphere is also responsible for increasing temperature in lower troposphere by acting as greenhouse gas.

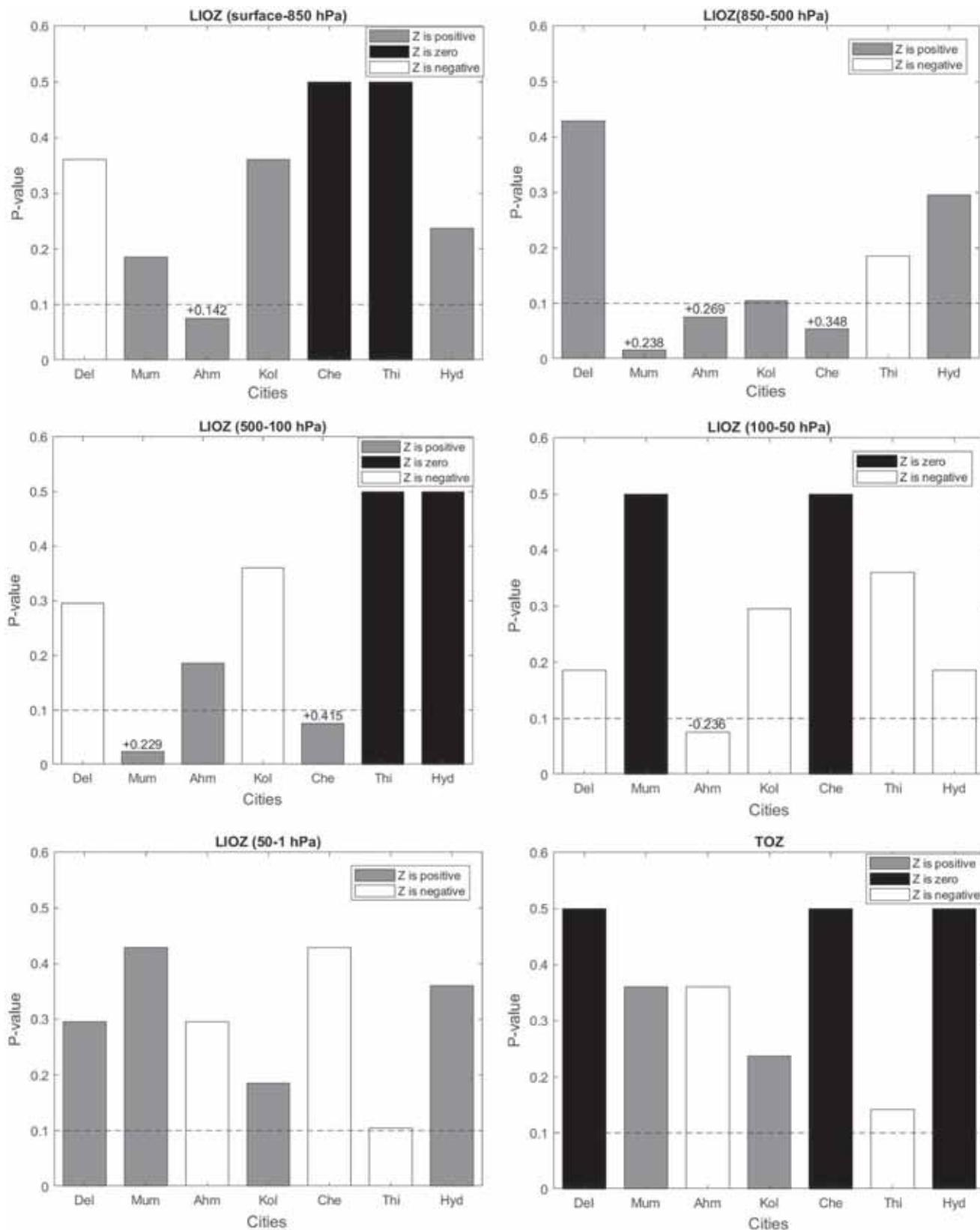


Figure 7. Mann-Kendall test results and Sen's slope estimate for LIOZ in various atmospheric layers and TOZ. P-value corresponding to Z statistics is plotted in Y-axis for various cities on X-axis. Grey bar corresponds to positive Z value, black bar corresponds to zero Z value and white bar corresponds to negative Z value. The values written on top of few bars are slopes (in %/year) of statistically significant trends.

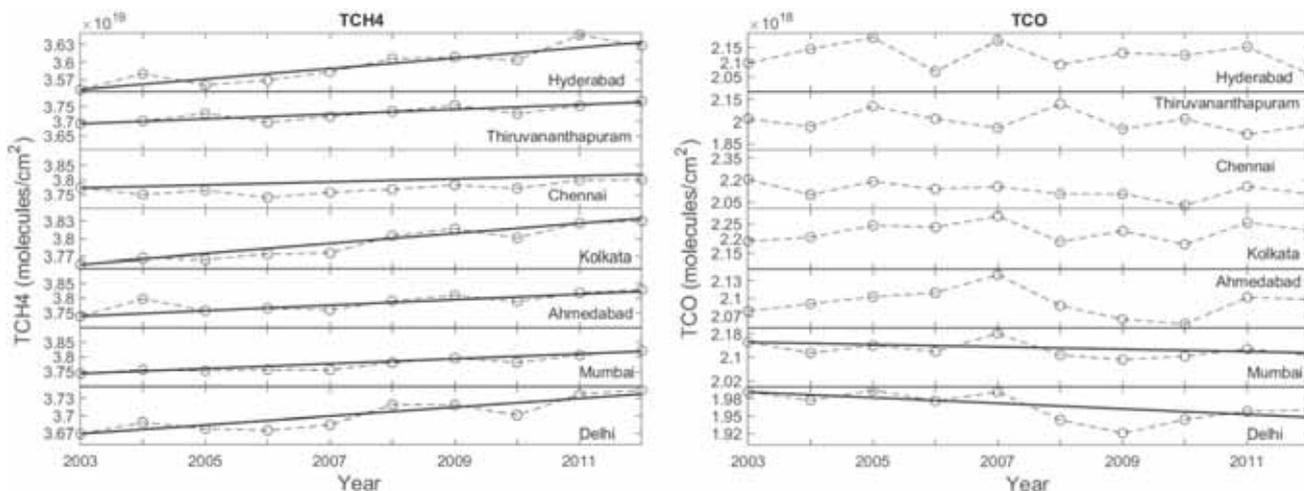


Figure 8. Time series plot of annual means (circles) of total column methane and total column carbon-monoxide over various cities of India. Annual trend line (thick line) is also plotted for the cities where statistically significant trend was observed.

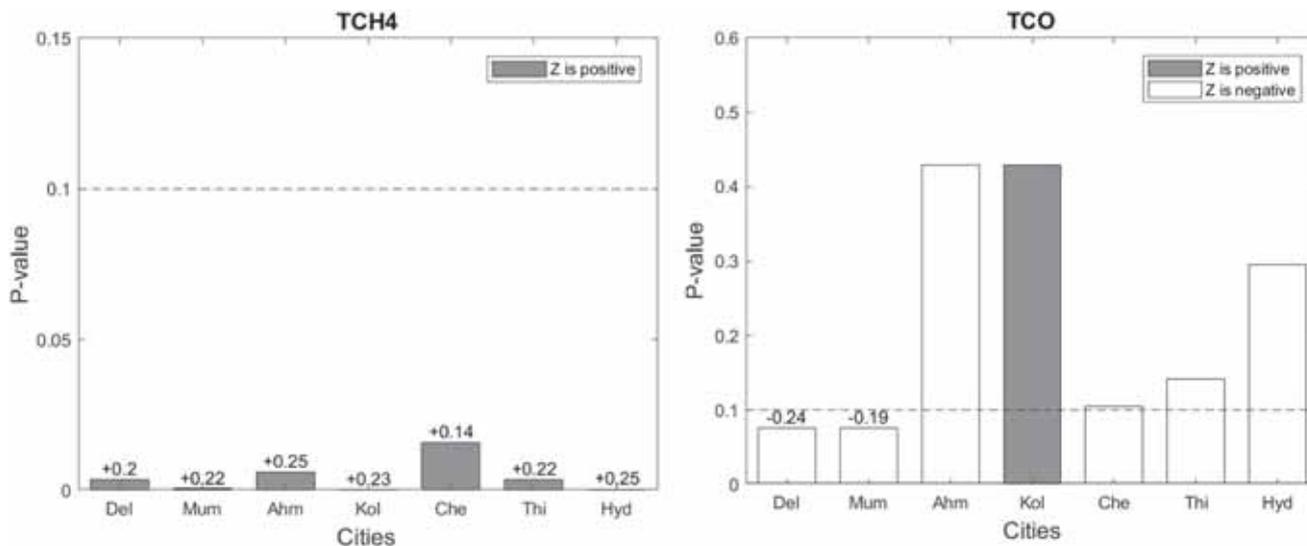


Figure 9. Mann–Kendall test results and Sen's slope estimate for TCH_4 and TCO. P -value corresponding to Z statistics is plotted in Y-axis for various cities on X-axis. Grey bar corresponds to positive Z value and white bar corresponds to negative Z value. The values written on top of few bars are slopes (in %/year) of statistically significant trends.

4.4 Total column methane and total column carbon-monoxide

Here, the results of Mann–Kendall test for TCH_4 and TCO are discussed. The time series plots and statistically significant annual trend lines for TCH_4 and TCO are shown in figure 8 over various cities. Figure 9 presents Mann–Kendall test statistics for TCH_4 and TCO over different cities of India and Sen's slope estimates for statistically significant trends. TCH_4 has shown the increasing trend over all cities of the country with very good significance level. The increased rate of TCH_4 is 0.20%/year, 0.22%/year, 0.25%/year, 0.23%/year, 0.14%/year,

0.22%/year and 0.25%/year over Delhi, Mumbai, Ahmedabad, Kolkata, Chennai, Thiruvananthapuram and Hyderabad, respectively. Many studies over different parts of the world have shown increasing trend of methane (Blake and Rowland 1988; Etheridge *et al.* 1998; Bousquet *et al.* 2011). This increasing trend of methane may be due to increase in methane emissions. Singh *et al.* (2018) have observed that the annual methane emission from landfills have increased from 404 Gg in 1999–2000 to 1084 Gg in 2015 over India. This increased methane act as a greenhouse gas in troposphere, and contribute for increase in tropospheric temperature. Also, the increased

tropospheric methane lower the thermal radiation passing through troposphere to reach stratosphere, thereby decreasing lower stratospheric temperature. Moreover, the increased methane in stratosphere is oxidised by ozone to form water vapour, thereby reducing stratospheric ozone and this reduced ozone further decrease the stratospheric temperature by reduction in absorption of UV radiations.

TCO trends have been observed to be mostly decreasing and the decreasing trends are significant over Delhi and Mumbai. The rate of decrease of TCO is 0.24%/year and 0.19%/year over Delhi and Mumbai. The decreasing trends of carbon-monoxide has also been demonstrated in previous studies (Novelli *et al.* 2003; Zellweger *et al.* 2009; Angelbratt *et al.* 2011; Li and Liu 2011; Warner *et al.* 2013; Worden *et al.* 2013; Yoon and Pozzer 2014). The decreasing trends of TCO over most of the Indian region have also been shown by Girach and Nair (2014) using MOPITT data. They have also reported that the decreased trend of TCO may be partly due to increase in water vapour and/or ozone in troposphere. The increasing trend of tropospheric water vapour and ozone has been shown in the present study.

5. Conclusions

In the present study, 10 yrs of Aqua-AIRS sounding products, like atmospheric temperature, water vapour and ozone profiles, and total column CH₄ and total column CO have been analysed for their trend analysis over few major Indian cities using non-parametric Mann–Kendall test. The following conclusions can be made for various atmospheric parameters in different atmospheric layers in a decade from 2003 to 2012.

1. The LAT has been found to be increasing in lower troposphere (surface–850 hPa) and decreasing in lower stratosphere (100–50 hPa). The decreasing trend in lower stratosphere (100–50 hPa) is more prominent than increasing temperature in lower troposphere (surface–850 hPa). The increasing temperature trend in surface–850 hPa layer is found to be statistically significant only over the Chennai which is due higher urbanization and, hence larger CO₂ emissions in Chennai. The warming over Chennai is not limited to lower tropospheric region as the LAT has significant increasing trend over Chennai in 850–500 hPa layer also. This is due

to greenhouse effect of increased ozone in this layer which has been shown as statistically significant increasing trend of LIOZ (850–500 hPa) over Chennai in this study. Over Thiruvananthapuram, the LAT (850–500 hPa) has decreasing trend which may be due to decreasing trend of ozone with *P*-value of 0.186 in 850–500 hPa layer. LAT in 500–100 hPa has significant decreasing trend only over Ahmedabad which is due to decrease of water vapour trend over Ahmedabad in 500–100 hPa. Also, Ahmedabad is the only city where water vapour has significant decreasing trend.

2. The water vapour has been observed to be increasing mainly in the lower and lower-mid part of the troposphere. It has also been noticed that the both LAT and LIWV have significant decreasing trend over Ahmedabad in 500–100 hPa atmospheric layer which confirms the positive feedback effect of temperature and water vapour. For TWV, the trends are mostly increasing; however, it is statistically significant only over Hyderabad. The water vapour increasing trend is most prominent over Hyderabad as it is statistically significant in surface–850 and 850–500 hPa layers, and hence for TWV. All the statistically significant water vapour trends are increasing, except the trend over Ahmedabad in 500–100 hPa layer. This is possibly the main reason for decreasing temperature trend over Ahmedabad in the layer.
3. The ozone has been found to be increasing in troposphere and decreasing in lower stratosphere. The increasing trend of ozone in troposphere is most prominent in lower-mid tropospheric region (850–500 hPa layer). The larger increase of ozone concentration in 850–500 hPa layer than surface–850 hPa layer has also been demonstrated by earlier studies (Oltmans *et al.* 2013). Tropospheric increase in ozone concentration is mainly due to increase in pollutants including methane (increase in methane is shown in the present study also), and lower stratospheric decrease in ozone is due to tropical upwelling and stratospheric meridional circulation (Fadnavis *et al.* 2010). However, another reason for decrease in stratospheric ozone may be increase in methane which is oxidised by ozone to form water vapour in stratosphere. The TOZ is not found to have any significant trend, may be due to contrasting increase and decrease of ozone in troposphere and stratosphere. As already said, this

increasing ozone concentration in troposphere has contribution in increase of tropospheric temperature and this decreasing ozone concentration in lower stratosphere is one of the major reasons for decrease in lower-stratospheric temperature.

4. It has been observed that TCH₄ has increasing trend over all cities of the country with very good significance level. Increasing methane emissions with time (Singh *et al.* 2018) may be the major reason for increasing trends of methane. For TCO, the trends have been found to be decreasing and the decreasing trends are significant over Delhi and Mumbai. Girach and Nair (2014) have reported that the increasing trend of water vapour and ozone in troposphere is partly responsible for decreasing TCO trends.

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