

Research Article

Statistical Analysis of Total Column Ozone over Uttarakhand: Environment of Himalaya

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ABSTRACT Total Column Ozone (TCO) is a critical factor affecting the earth's atmosphere, especially in the Himalayan region. A comprehensive study of TCO trend analysis and corresponding consequences in the Himalayan atmosphere needs to be analyzed. We statistically examine TCO variability by analyzing the daily TCO dataset of the last 15 years (2005–2019) over the crucial region of the Himalayan environment i.e. Uttarakhand, India. Obtained results indicate that TCO values are at peak during the spring season whereas it shows the least value during the winter season. The highest and lowest value of Coefficient of Relative Variance (CRV) is estimated as 3.14 and 1.09 during winter and monsoon season, respectively. Air mass trajectories have been estimated using Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT), which shows the existence of strong seasonal variability of Ozone corresponding to continental and maritime transportation towards Uttarakhand. Moreover, Least Square Method (LSM) and the Mann-Kendall test estimate a high correlation (86%) for the seasonal and annual trend of TCO with a negative rate. The obtained decreasing rate is very low which indicates recovery of TCO during the study period. Further results imply that the inter-annual oscillation pattern of TCO is similar to Quasi-Biennial Oscillation (QBO) significantly. In addition, a comparative study has been performed for the data measured by two TCO measuring instruments i.e. Ozone Monitoring Instrument (OMI) and Ozone Mapping Profiler Suite (OMPS). TCO values measured from both instruments are highly correlated (96%) with an average relative difference of around 3%. The outcomes of this study are expected to be beneficial for future study of TCO over other crucial regions of Himalayan territory.

KEY WORDS Total Column Ozone, Himalayan region, Temporal variation, Correlation coefficient, Mass trajectories

1. INTRODUCTION

Ozone is a robust component of the earth's atmosphere and can be found primarily in two layers of the earth's atmosphere. A major part i.e. 90% of its mass is found in the stratosphere lying within 16–35 kilometers above the surface of the earth whereas the remaining 10% ozone is found in the troposphere of earth. Stratospheric ozone is considered as a shielding layer (good ozone) as it hinders the high-energy radiations (2000 Å to 3000 Å) from penetrating the earth's main environment. On

the other hand, Ultra Violet (UV)-B radiations are biologically harmful solar radiation and the ozone layer prevents these hazardous radiations to reach on the surface of the earth (Madronich *et al.*, 1998; Dobson 1973, 1968). Most ozone is formed in the tropic region where it moves from lower to higher latitude by the Brewer-Dobson circulation (Weber *et al.*, 2011). TCO is the amount of total ozone (troposphere and stratospheric ozone) containing in a vertical column from the earth's surface to the top of the atmosphere at standard temperature and pressure (STP) (Fahey and Hegglin, 2014; Brasseur and Hitchman, 1988). It is measured using data obtained from ground-based stations and satellites in terms of Dobson unit (DU), which describes the thickness of a layer of pure ozone at STP. One DU is 2.69×10^{16} ozone molecules/cm² in a vertical column extended till the atmospheric limit or 0.01 mm thick layer of pure ozone to the surface of the earth at standard condition (STP). The value of TCO significantly varies from 200 to 300 DU all over the globe. Meanwhile, if its value reduces to 220 DU it is considered as a condition for occurring Ozone hole at any location.

A series of reactions of ozone formation and distortion was proposed by Sydney Chapman in 1930. It depicts that the photolysis of Oxygen molecules produces nascent radicals in presence of UV radiation which further reacts with oxygen and results in ozone molecule. Moreover, ozone depletion involves the reverse process of conversion of ozone molecules into atomic oxygen due to catalyst reaction (Groves and Tuck, 1980). Apart from this ozone abundance are the key parameters for estimating the formation and depletion of ozone at any location of the globe. The processes of formation and deterioration of ozone are found to be naturogenic as well as anthropogenic. However, both can occur simultaneously (Rubin, 2001). Anthropogenic chemicals such as chlorofluorocarbons (CFCs), halocarbons, and other volatile organic compounds can destroy stratospheric ozone significantly by reacting with ozone (Chipperfield *et al.*, 2020; Said Ismail and Hameed, 2013; Solomon, 1999; Cicerone *et al.*, 1974; Crutzen 1974). Apparently, few studies suggest long term variability of ozone with geophysical parameters, including the Quasi-Biennial Oscillation, solar activity, the stratosphere-troposphere exchange, and volcano eruption (Brenna *et al.*, 2019; Bisht *et al.*, 2014; Kalita *et al.*, 2011; Ningombam, 2011; Garcia and Solomon, 1987; Willett, 1962).

Despite a small portion of ozone contributes to the earth's atmospheric composition, TCO plays a vital role

in climate change and ecology (Ogunjobi *et al.*, 2007; Wayne, 2000, 1989). The depletion of the ozone layer results in severe serious health issues i.e. skin cancer, cataract, deficient immunity, respiratory disorders for species (UNEP 2016, 1998; Van der Leun *et al.*, 2003). It can also contaminate the marine ecosystem as well (cell-damaging life of phytoplankton and zooplankton). Besides, ozone depletion permits UV-B radiation which may retard the physiological and developmental processes of plants (Sivasakthivel and Siva Kumar Reddy, 2011; Schmalwieser *et al.*, 2003). Therefore, ozone layer conservation is mandatory to sustain an ideal ecological surrounding on earth (Venkanna *et al.*, 2015; WMO, 2011, 1989; Freijer *et al.*, 2002).

Numerous studies on atmospheric ozone have been carried out all across the world. The Antarctic ozone hole was first observed by the British Antarctic Survey in the 1980s (Farman *et al.*, 1985). Several studies emphasize seasonal depletion in the stratospheric ozone layer in Antarctica and have observed depletion during the spring season (Newman *et al.*, 1991; Rowland, 1989; Iwasaka and Kondoh, 1987). Over the last two decades, minute depletion in ozone was also observed over the Arctic during the late winter and early spring (Goutail *et al.*, 1999; Brune *et al.*, 1990). But these studies are limited to the polar region only (Newman *et al.*, 2006). Such studies have been extended for other locations of the globe and established statistical trends of TCO over the northern and southern mid-latitudes (Tohir *et al.*, 2018; Badawy *et al.*, 2017; Tan *et al.*, 2014; Nair *et al.*, 2013; Oluleye and Okogbue, 2013; Stein, 2007; Chakrabarty *et al.*, 1998; Rowland, 1991). These studies suggest that the depletion of ozone is not only confined in the Polar Regions but varies significantly in another latitudinal region as well.

The study of ozone layer depletion and variation has emerged as a major global scientific and environmental issue in the recent past. In this framework, various international protocols have been designed (UNEP, 1987). Montreal Protocol is an international treaty to protect the ozone layer by phasing out the production and limited use of ozone-depleting substances (ODS) (1989). This agreement was executed on 1 Jan 1989 and has been signed by most of the countries including India. This protocol has reduced the risk of further ozone depletion to a large extent (Pinedo-Vega *et al.*, 2017; Velders *et al.*, 2007; Egorova *et al.*, 2001; Anderson *et al.*, 2000). From the perspective of India, a few studies on ozone variation and depletion have been reported to date (Madhu, 2014;

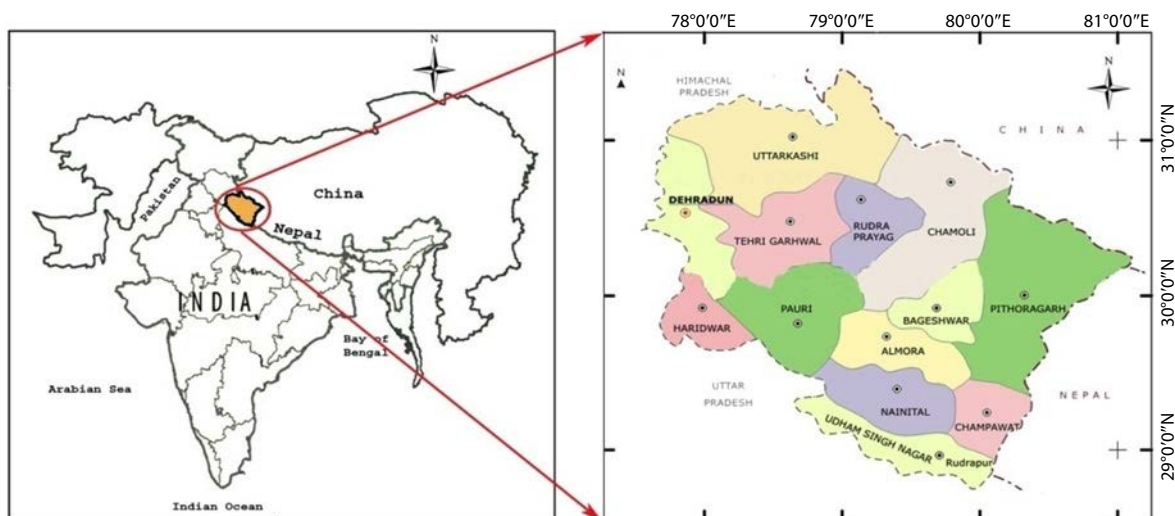


Fig. 1. Map of Uttarakhand with latitude and longitude of the study area.

Pulikesi *et al.*, 2006; Kundu and Jain, 1993; Mani and Sreedharan, 1973). These studies are limited to industrial areas with high population density. However, there is a paucity of such studies in Himalayan regions i.e. areas with lower population density and enhanced ecological versatility. The ozone climatology is supposed to be crucial in these regions due to climatic disparity. Therefore, a comprehensive study of ozone (TCO variation) over the Himalayan region is needed to monitor the atmospheric ozone concentration in a regular period.

In this paper, we have statistically analyzed the spatial and temporal trend of TCO variability using the TCO dataset of the last 15 years i.e. 2005–2019 over the crucial region of the Himalayan environment i.e. Uttarakhand, India. Air mass trajectories have been estimated using Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) to understand the source of air pollutants and corresponding continental and maritime transportation towards Uttarakhand with maximum ozone concentration under various climatic conditions has been investigated. We have compared the data measured by two TCO measuring instruments i.e. Ozone Monitoring Instrument (OMI) and Ozone Mapping Profiler Suite (OMPS) and corresponding results have been analyzed.

2. MATERIALS AND METHODS

2.1 Study Area

Uttarakhand, a state in northern India crossed by the

Himalayas extended from (28.71°N to 31.45°N and 77.56°E to 81.03°E) is the study area shown in Fig. 1. It is situated in southern Asia (North of Indo Gangetic Plain (IGP), East of China, and foothill of Himalayas). It has a total geographic area of 53,483 km² of which 86% is mountainous and 65% is covered by forest with an average population density of 189 people/km² (Census, 2011). It is also surrounded by major Indian industrialized cities i.e. Delhi, Lucknow, and Chandigarh. Along with more industrial development the agricultural activities, burning of crop residues, and forest fire are the primary source of pollutants over Uttarakhand. The mountainous topography and complex land-sea interactions are responsible for phenomenal changes in weather over Uttarakhand. The average temperature varies within a range of 21.8°C to 5°C with the highest in June while the lowest in December and for few stations the temperature falls below 0°C. Maximum rainfall occurs from July to September i.e. early NEM with an average precipitation of 160–200 cm. Uttarakhand holds a rich heritage of India and is known as Devbhumi i.e. “Land Of God”. Its snow-clad peaks, beautiful hill stations, and charming weather make it a favorite destination for tourism. Nanda Devi, Valley of flowers and National Park (1988, 2005) are world heritage sites declared by UNESCO are the most fascinating places of Uttarakhand. Uttarakhand is the most popular Himalayan state of India, situated in the natural environment of Himalaya and it plays a vital role in hosting many animals, plants, and rare herbs. Most importantly, Uttarakhand covers a major portion of the

Himalayan range and therefore represents a crucial study station (area) to analyze atmospheric phenomena i.e. ozone variation over the Himalayan region.

2.2 Data Sources

The daily data of TCO has been acquired from the open resource database of NASA and NOAA. The used data is recorded with two TCO measuring instruments i.e. Ozone Monitoring Instrument (OMI) and Ozone Mapping and Profiler Suite (OMPS). The TCO value is measured in Dobson Unit (DU). OMI aboard in AURA Spacecraft which continue the TOMS record of TCO and other atmospheric parameters related to ozone chemistry and climate have spatial coverage of Global (0.25° longitudes × 0.25° latitudes) whereas OMPS instrument fitted in Suomi NPP& NOAA-20 satellite with a spatial resolution of (1° longitudes × 1° latitudes) and it continues Solar Backscatter Ultraviolet (SBUV) series data.

The monthly and annual mean for all the stations are derived by taking the cumulative mean of daily data. The principle of measurement of TCO is based on the reflection of solar radiation on certain spectrum bands (Bhartia *et al.*, 2013; Heath *et al.*, 1975), some unresolved data corresponding to polar night and low light area cannot be measured and recorded as zero (Pinedo-Vega *et al.*, 2017). These unresolved data are few and removed into account for monthly and annual mean. To examine the seasonal variability of ozone we divide TCO data into five seasons' i.e. winter (Dec–Feb), spring (March–April), summer (May–June), monsoon (July–Sep), and autumn (Oct–Nov). Also, to investigate the variation in ozone profile i.e. the latitudinal variation of TCO, we studied four stations of India i.e. Leh Laddak (34.10°N-77.4°E), Dehradun (28.37°N-77.13°E), Bangalore (12.58°N-77.34°E), and Kanyakumari (8.4°N-77.32°E) from northward to southward for the year 2014. The coefficient of Relative Variance (CRV), correlation coefficient, probability distribution, least square method i.e. regression analysis method, and Mann-Kendall test (MK) are used for statistical analysis and trend analysis of TCO variation over Uttarakhand during a period of 15 years i.e. from 2005 to 2019.

2.3 Methods

a) Percentage relative difference is calculated as;

$$\% \text{ difference} = \frac{TCO_{(OMI)} - TCO_{(OMPS)}}{TCO_{(OMPS)}} \quad (1)$$

Here 'X' is the value of ozone concentration and N is the number of years.

b) A coefficient of relative variation (CRV) for each site has been determined as;

$$CRV = \frac{SD}{\mu} \times 100 \quad (2)$$

Here 'SD' is the standard deviation and 'μ' is the mean for N years.

c) Trend analysis and fluctuation of ozone concentration have been estimated by Linear Regression (Least Square method) and Mann-Kendall test for all the selective sites.

d) Statistical models are used for the probability distribution (Normal, Lognormal, and Gamma) of TCO. We check their deviation from normality using KST (Kolmogorov-Smirnov D-test) probability distribution over Uttarakhand.

2.4 Back Trajectory Calculation

The origin of air quality and transportation patterns of air mass arriving towards Uttarakhand has been analyzed using Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) backward trajectories. The HYSPLIT Model (Draxler and Rolph, 2010) was developed by Air Resource Laboratory (NOAA) and these trajectories have been simulated from the Web site (http://ready.arl.noaa.gov/HYSPLIT_traj.php). Based on the highest concentrations of ozone in the study period, multiple air mass back trajectories for 14th May 2014, 13th June 2014, and 22nd December 2014 are simulated at different elevations: 500 and 1,000 m a.g.l. using HYSPLIT models. The analysis has been performed with the GDAS meteorological dataset at a starting time of 00:00 UTC with a total run time of 120 hours.

3. RESULTS AND DISCUSSION

The monthly mean of TCO from OMI and OMPS datasets (January 2012 until December 2019) has been compared by estimating relative differences based on Equation (1) tabulated in Table 1. A range of relative difference 0.51% to 5.29% for the study period has been found which is within the acceptable interval. Moreover, the average relative difference of monthly TCO data via both datasets is merely 3.5%. Also, we found a high positive correlation between the values of TCO retrieved

Table 1. Comparison of TCO values retrieved from OMI and OMPS datasets.

Year/Month	2012	2013	2014	2015	2016	2017	2018	2019
% difference between OMI and OMPS data of TCO								
January	0.88	8.25	2.24	1.35	3.56	2.65	0.12	1.06
February	1.16	0.6	0.14	0.34	4.59	6.4	0.89	2.9
March	1.81	4.8	2.49	5.87	1.55	3.51	1.75	5.52
April	10.11	0.02	1.01	1.57	4.06	5.32	1.05	0.17
May	4.28	10.47	10.45	5.71	9.47	5.41	1.23	0.98
June	5.13	5.92	7.02	6.32	8.2	5.24	0.31	0.17
July	9.17	10.43	8	7.4	10.18	4.42	0.13	0.46
August	7.59	8.1	5.76	5.33	6.75	5.58	0.09	0.23
September	4.1	4.98	2.3	1.14	2.16	2.59	0.02	0.85
October	1.62	3.69	4.41	0.03	1.39	2.09	0.32	0.25
November	2.77	3.79	5.03	3.24	4.59	5.29	0	1.86
December	1.02	2.44	0.99	7.7	4.64	1.47	0.26	1.06
Average	4.14	5.29	4.15	3.83	5.1	4.16	0.51	1.29
Correlation coefficient between the value of TCO with OMI and OMPS data								
	0.86	0.69	0.68	0.73	0.85	0.86	0.99	0.96

from both the instruments having a different resolution. We have not observed an average monthly mean of TCO value less than 220 DU (Condition for occurring Ozone hole at any location on the globe). It implies that However, we noted TCO value less than 220 DU for one or two days in December 2008 and 2016 and January 2019 from the OMI data set while in December 2008 and January 2019 from the OMPS data set, which can be ignored for accounting it as the ozone hole. For the total number of years considered average ozone maximum and minimum value have observed 395 DU and 212 DU from OMI and 382 DU and 208 DU from OMPS, respectively.

3.1 Monthly Variation

The monthly mean of TCO is found in the range from (297 ± 13.78) DU to (260 ± 8.73) DU for OMI and (300 ± 16.08) DU to (258 ± 6.07) DU from OMPS datasets. The average maximum and minimum value of ozone concentration are measured in May and December, respectively from both the instruments over Uttarakhand. Further, a bell-shaped normal distributed curve for monthly ozone mean of 15 years (2005–2019) has been observed as shown in Fig. 2. We have observed similar results as the average TCO value from OMI increased, the ozone value measured from OMPS was also increased. Therefore, the quality of OMPS datasets is analogous to OMI data of ozone.

We have statistically analyzed the data and estimated

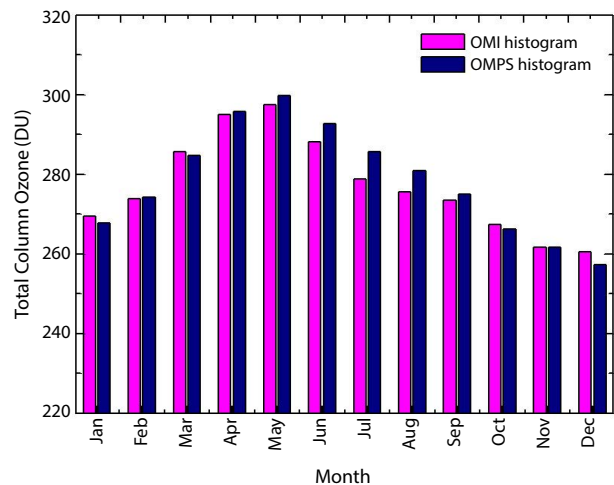


Fig. 2. Monthly variation of TCO over Uttarakhand via OMI and OMPS datasets.

final distributions comply with the mean monthly series of TCO by applying the Kolmogorov-Smirnov goodness of fit test of distribution. It is noticed that monthly TCO distribution exhibits lognormal probability distribution in Uttarakhand. The basic statistics (maximum, minimum, average, standard deviation, and distributions) for the Interannual monthly average TCO (2005–2019) from both data sets are mentioned in Table 2. On observing Table 2, we have found that the set of monthly series of TCO follows different distributions and changes from

Table 2. Probability distribution with statistical parameters of TCO via OMI and OMPS datasets (2005–2019) for Uttarakhand.

Month	OMPS					OMI				
	Max.	Min.	Standard deviation	Distribution	KST-value $\alpha = 0.05$	Max.	Min.	Standard deviation	Distribution	KST-value $\alpha = 0.05$
January	291	245	11.47	Normal	0.18	279	251	9.25	Normal	0.20
February	298	242	15.47	Lognormal	0.16	298	255	13.70	Lognormal	0.19
March	296	274	6.12	Lognormal	0.10	306	269	8.94	Lognormal	0.13
April	319	282	10.71	Gamma	0.18	313	282	10.33	Lognormal	0.15
May	322	274	16.08	Normal	0.16	319	278	13.78	Gamma	0.17
June	309	275	11.40	Lognormal	0.16	312	276	11.33	Lognormal	0.21
July	301	268	12.44	Lognormal	0.21	302	265	12.25	Lognormal	0.19
August	295	269	9.66	Lognormal	0.18	295	266	10.44	Lognormal	0.18
September	287	267	5.79	Gamma	0.14	288	266	6.74	Lognormal	0.18
October	278	257	6.08	Lognormal	0.16	278	258	6.34	Lognormal	0.25
November	277	250	7.57	Gamma	0.13	276	249	8.10	Gamma	0.18
December	268	248	6.07	Gamma	0.14	273	241	8.73	Normal	0.12

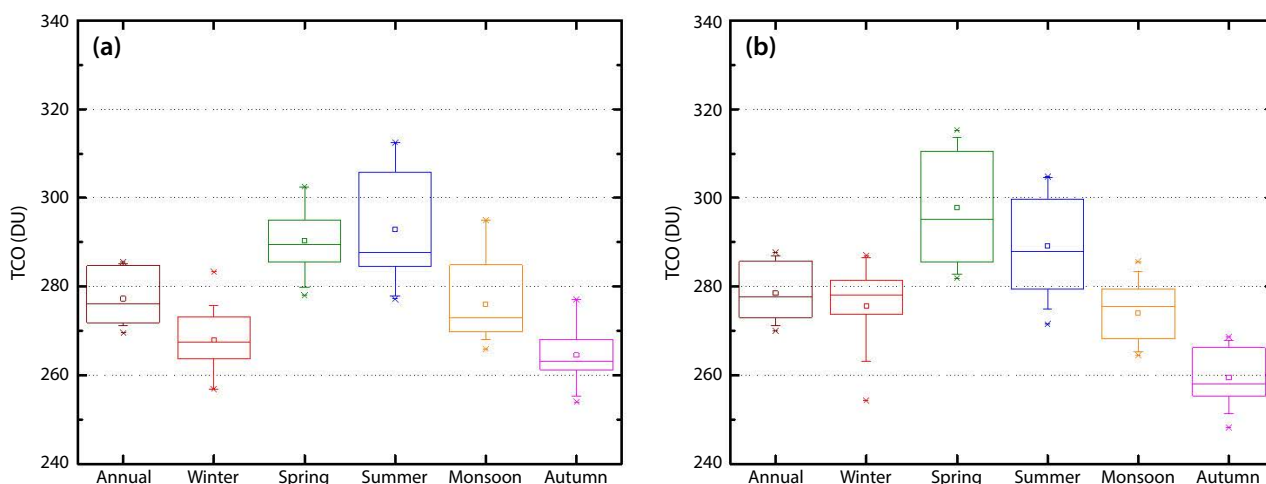


Fig. 3. Annual and the seasonal average of TCO (2005–2019) from (a) OMI and (b) OMPS datasets.

normal, lognormal to gamma distribution which indicates that the distribution is progressively approaching heavier tailed from January to December.

3.2 Inter Annual Variation and Seasonal Variation

Box plots of average annual and seasonal values of TCO (2005–2019) from both the datasets were illustrated in Fig. 3. It is observed that the summer and autumn seasons have the highest and lowest average TCO value from both the sensors. Seasonal average time series shows that the average TCO in the summer season was maximum (292.81 ± 11.82) DU and minimum in autumn (264.53 ± 6.87) DU from OMI dataset whereas OMPS

shows that the maximum average TCO in the spring season was (297.73 ± 12.16) DU and the minimum was (259.48 ± 6.58) autumn DU. In addition, the sensors recorded annual maximum and minimum TCO measurements in the same years 2015 and 2008, respectively. The behavior is not repeated in any of the seasons (spring, summer, monsoon, and autumn) except winters i.e. the maximum and minimum TCO values from two sensors do not overlap in the same year again.

Interannual variability of TCO retrieved from the OMI dataset (2005–2019) showed merely the same oscillating behavior for all the stations along the latitudinal and longitudinal belt of Uttarakhand illustrated in Fig. 4. From

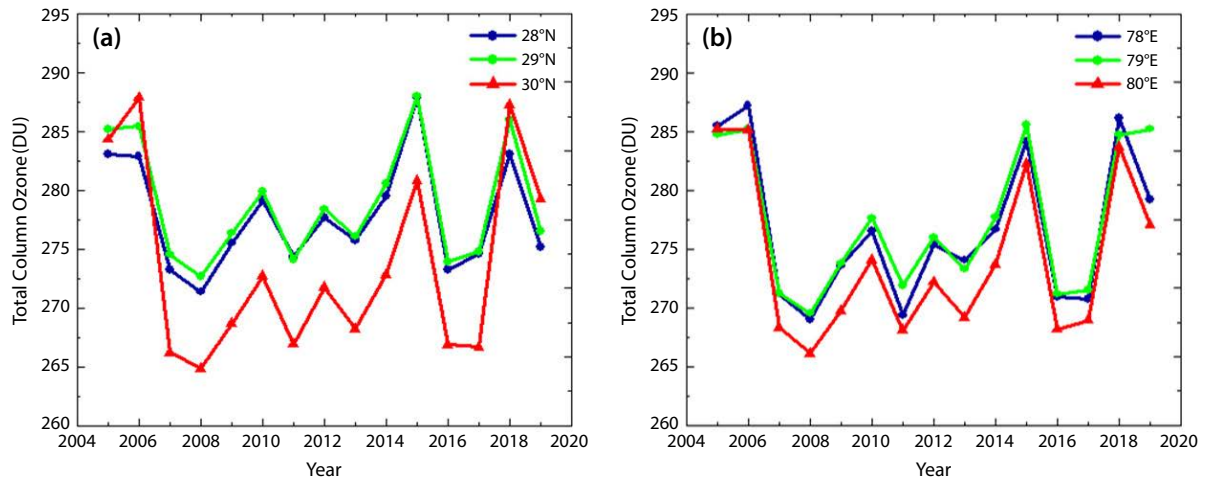


Fig. 4. The Interannual variation of TCO across (a) latitudinal and (b) longitudinal belt of Uttarakhand.

Table 3. Annual and seasonal trend of TCO by least square method and MK Test from 2005–2019.

Method	Season	Dehradun	Almora	Haridwar	Pithoragarh	U.S. Nagar	Chamoli
LSM	Annual	0.030	0.162	-0.045	-0.057	-0.001	0.033
	Winter	-0.589	-0.144	-0.495	-0.574	-0.579	-0.489
	Spring	0.090	0.226	-0.041	-0.114	-0.459	0.320
	Summer	0.078	0.231	0.690	0.013	0.140	0.058
	Monsoon	0.084	0.146	0.191	0.095	0.027	0.089
	Autumn	0.051	0.285	0.018	0.078	0.159	0.056
MK TEST	Annual	0.297	0.595	0.198	0.099	0.000	0.297
	Winter	-0.495	-0.248	-0.297	-0.495	-0.693	-0.495
	Spring	0.000	0.397	0.000	-0.297	-1.437	0.495
	Summer	0.594	0.595	0.792	0.594	0.248	0.594
	Monsoon	0.594	0.397	0.693	0.396	0.545	0.099
	Autumn	0.495	0.794	0.198	0.495	1.041	0.396

2005 to 2019 ozone variability repeats its ascending and descending pattern at a period of two years. This fluctuation of TCO follows the analogous pattern of the quasi-biennial oscillation (QBO) significantly. Further, we observed an abrupt change in TCO in 2007 and 2016. It suggests this fluctuation might be due to interruption in QBO for the respective years.

Table 3 depicts the trend of TCO annual and seasonal (retrieved from OMI from 2005 to 2019) for all the stations by the least square method and MK Test. We obtained a significant negative trend of TCO from LSM with rate 0.03 DU/year, 0.162 DU/year, -0.045 DU/year, -0.057 DU/year, -0.001 DU/year, and 0.033 DU/year for Dehradun, Almora, Haridwar, Pithoragarh, U.S. Nagar and Chamoli, respectively. However, the annual decrease-

ing rate is very low over Uttarakhand which indicates a significant recovery of TCO during the study period. Also, we observed a negative trend for winters and spring seasons for most of the stations while the increasing trend in summers, monsoon, and autumn seasons for all the stations. Similarly, results obtained from the Mann-Kendall test are illustrated in Table 3. The trend by LSM and MK test is highly correlated in all the seasons as well as annually except monsoon. The correlation coefficient between trends obtained from both the methods has been calculated 0.86 annually, 0.72 in winters, 0.97 in spring, 0.53 in summer, 0.24 in monsoon, and 0.75 in autumn seasons, respectively. Moreover, for most of the year, the value of TCO has been observed to fluctuate about its mean value excluding in 2008 and 2015.

Table 4. Coefficient of the relative variance of TCO from 2005–2019.

CRV	Annual	Winter	Spring	Summer	Monsoon	Autumn
Dehradun	1.6	3.11	2.35	2.29	1.18	2.35
Almora	1.67	2.93	2.32	2.36	1.24	2.08
Haridwar	1.58	2.95	2.24	2.09	1.19	2.03
Pithoragarh	1.66	3.13	2.35	2.38	1.21	2.01
U.S. Nagar	1.64	2.77	2.58	2.11	1.24	1.85
Chamoli	1.7	3.14	2.68	2.42	1.09	2.37

3.3 Spatial Variation (Coefficient of Relative Variation (CRV))

The variability of ozone concentration over Uttarakhand has been studied. Annual and seasonal CRV of TCO for six stations i.e. Dehradun, Almora, Haridwar, Pithoragarh, U S Nagar, and Chamoli is shown in Table 4. A significant variation in TCO has been observed among these study areas. Further, it is clear from the observations that the highest value of CRV is obtained in winter due to the maximum fall in temperature during winters, while the lowest value has been estimated at monsoon for all the selective stations. These Outcomes suggest that climatic parameters temperature and rainfall have a significant impact on the variability of TCO. Moreover, a gradual ascending trend has been observed for the annual and spring variation of CRV from east to west stations. Further observations reveal that CRV of ozone concentration increases gradually from north to south in summer, autumn and winters, whereas decreases in monsoon season from north to south. This concludes TCO varies as a function of latitude for most of the seasons. Fig. 5 provides a more clear insight into the spatial distribution of CRV.

3.4 Latitudinal Variation

To characterize the ozone profile i.e. latitudinal distribution of TCO, we have estimated the monthly mean of ozone concentration for four Indian subcontinent Lehigh Laddak (34.10°N-77.4°E), Dehradun (28.37°N-77.13°E), Bangalore (12.58°N-77.34°E), and Kanyakumari (8.4°N-77.32°E) for the year 2014. Fig. 6 indicates the logging of two months in maximum ozone value for Lehigh (high latitudinal area) and Kanyakumari (low latitudinal area). We have statistically analyzed the data and estimated skewness for distribution for northward region i.e. Lehigh Laddak and Dehradun the value of skewness is found positive (0.515 and 0.183), respectively. Similarly, the skewness is negative i.e. -0.539 and -0.600 for Bangalore and Kanyakumari, respectively. However, a quantitative variation in

TCO of approx 16% has been calculated from a latitudinal belt of India i.e. from Kanyakumari to Lehigh Laddak. The probable reason for this variation might be the processes of formation and transportation of ozone from the tropic to the pole region (Ganguly and Iyer, 2005). Also, CRV has been estimated at 6.58, 4.8, 4.48, and 4.17 for Lehigh Laddak, Dehradun, Bangalore, and Kanyakumari, respectively.

3.5 Back Trajectory Analysis

Back trajectories are simulated for correlating it with seasonal change of TCO and to understand the air mass transport and its source of origin (Stohl, 1998). Further observing the influence of continental and maritime air mass on seasonal variation of surface ozone, backward air trajectories are retrieved from the surface of Uttarakhand Fig. 7(a), 7(b), and 7(c) during seasons i.e. spring, summer, and winter. These back trajectories imply that the air mass moves from west of Uttarakhand's summer and winter seasons. During the spring season (14 May 2014), trajectories are continental and short-range. Air masses at low altitudes (500 m, 1000 m) appear to be originated from a western direction crossing Afghanistan and Pakistan following through the Indian state Punjab and Haryana. Fig. 7(a) and influenced by pollutants from nearby industrialized areas. In summer (13 June 2014), trajectories of air mass are marine type and occurred at heights (500 m, 1000 m) bring ozone and ozone precursors from southwestern directions i.e. Aarebic Sea and crossing some landmass area before reaching Uttarakhand. It is an indication of gasoline combustion originated from the Middle East and transport by the air mass trajectories at different altitudes Fig. 7(b). Trajectories followed by air mass in the last week of June (end of summer) indicated the onset of monsoon from the Arabian Sea. Similarly, during the winter season (22 December 2014), trajectories are long-range and marine type transports from, originating from the Caspian Sea (north-western direction

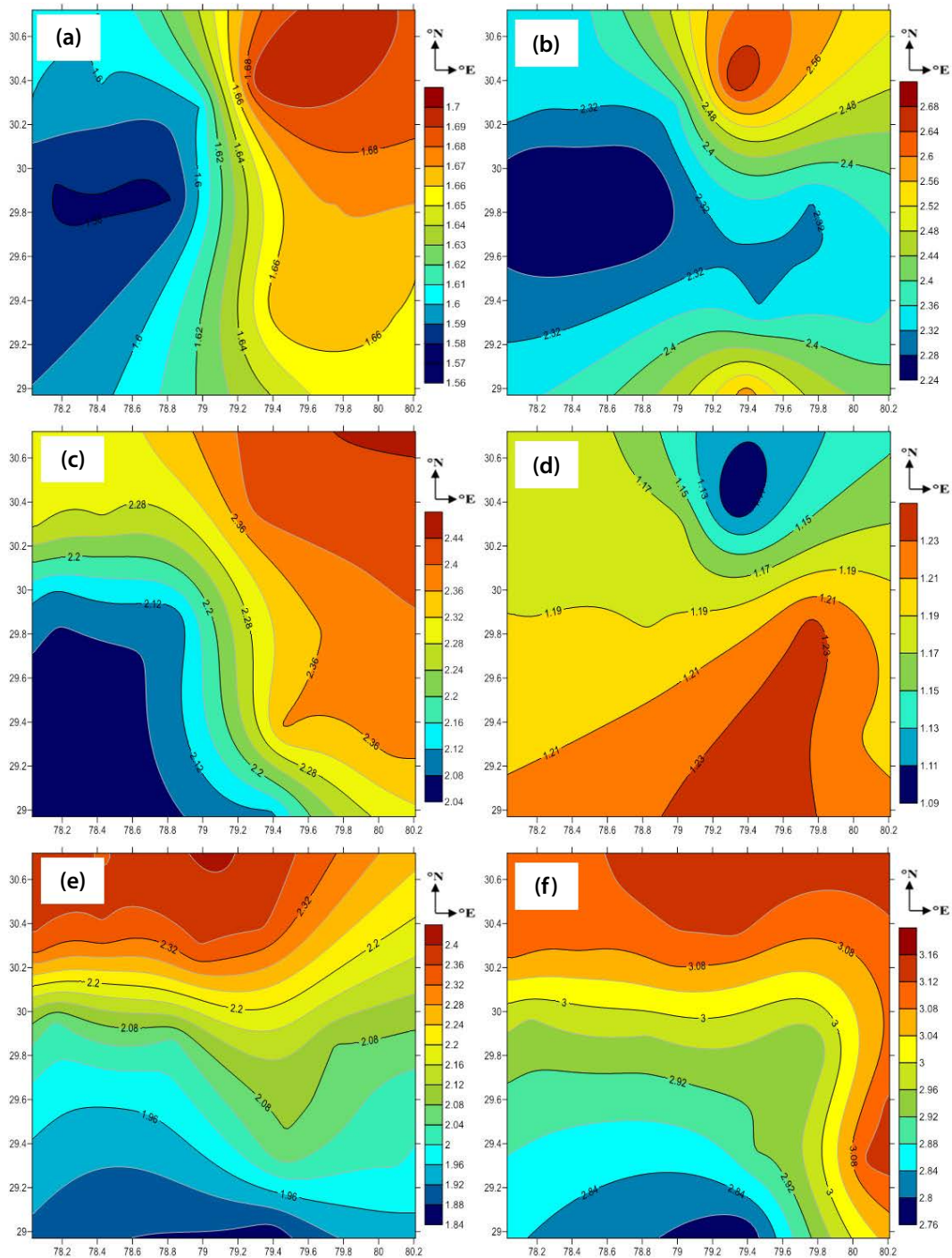


Fig. 5. CRV distribution of ozone over Uttarakhand (a) Annual, (b) Winter, (c) Summer, (d) Spring, (e) Monsoon, (f) Autumn.

of receptor site) and crossing Afghanistan Pakistan, and Indian states Punjab and also contributes to the observed low O_3 levels due to oceanic influence Fig. 7(c).

Since marine air mass is relatively clean compared to continental air mass. Therefore, with this oceanic influ-

ence, the air mass increases the composition of hydroxyl radicals (OH). As ozone is a highly reactive and unstable gas so it reacts with OH radicals and converted into atomic oxygen or molecular oxygen. This causes depletion of O_3 to a larger extent and might be one of the reasons for

the reduction of ozone concentration observed during the summer and winter seasons at this location. This complements our result illustrated in Fig. 2 i.e. maximum TCO value in spring then further decreases till winters. It concludes that maximum ozone concentrations in different seasons were a consequence of the transport effect of these trajectories.

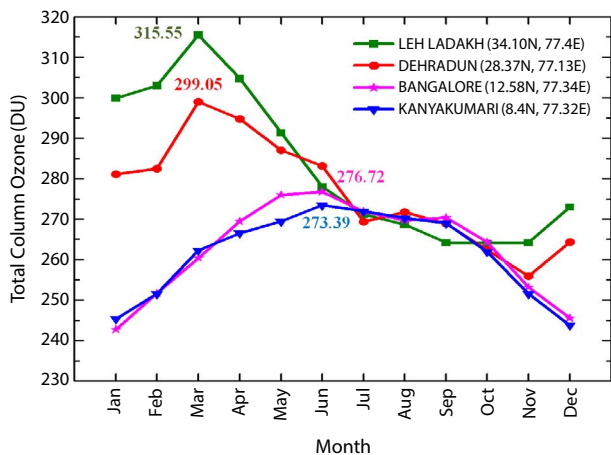


Fig. 6. Latitudinal variation of TCO from southward to northward regions in the year 2014.

4. CONCLUSION

The statistical analysis of temporal variability of TCO estimated a significant seasonal and annual cycle with a minimum in December and maximum in May. During the study period, we have obtained a monthly mean of TCO data retrieved from OMI was within (297 ± 13.78) DU to (260 ± 8.73) DU from OMI and (300 ± 16.08) DU to (258 ± 6.07) DU from OMPS, respectively. Kolmogorov-Smirnov goodness of fit test found that most of the months follow lognormal probability distribution at Uttarakhand for both the datasets of TCO. Interannual variability of TCO showed merely the similar oscillating behavior along the latitudinal and longitudinal belt of Uttarakhand which identical to the quasi-biennial oscillation (QBO) pattern. Further, the abrupt change noticed in TCO during the year (2007 and 2016) might be due to interruption in quasi-biennial oscillation for the respective years.

Analysis of Seasonal and annual trend of TCO using the least square method and Mann-Kendall test yield a negative trends for annual, winters, and spring season with the maximum in winter while the positive trend for the rest of the seasons. Although a negative annual trend

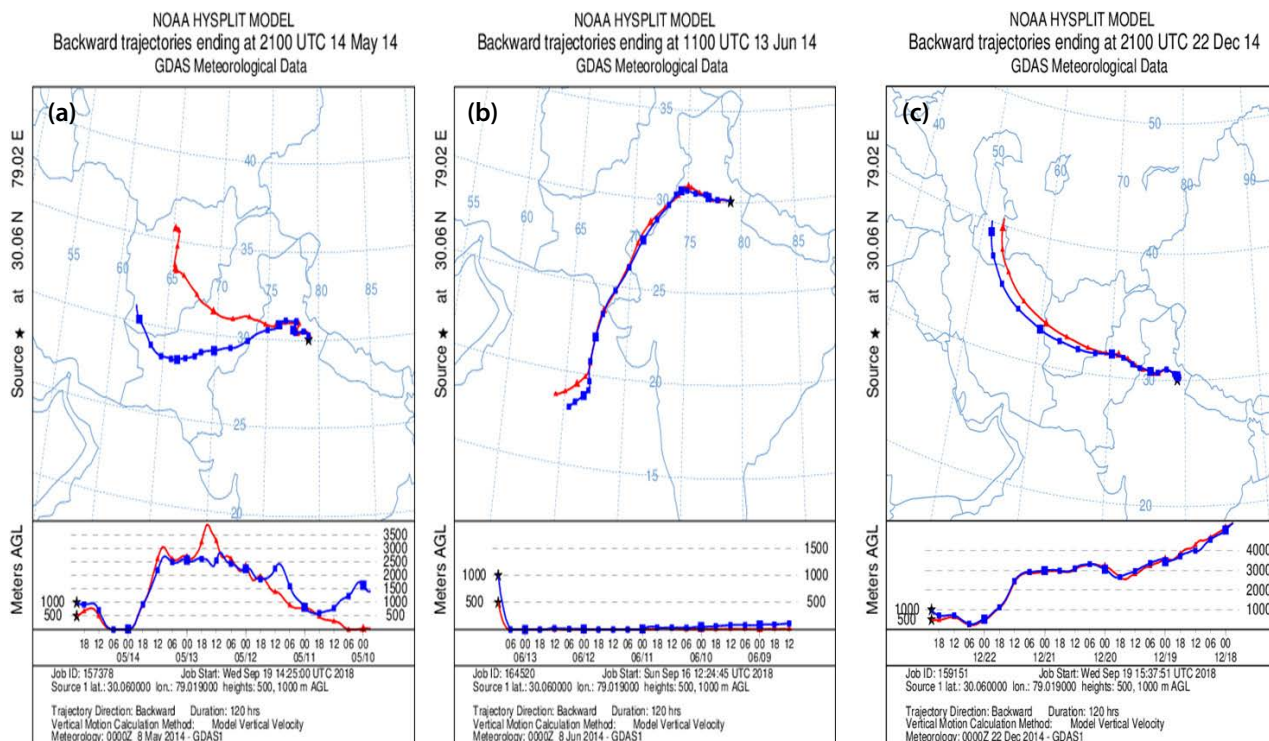


Fig. 7. Back trajectories arriving in Uttarakhand (a) summer season, (b) rainy season, (c) winter season.

was observed but corresponding rates were very low which suggests a recovered TCO level in Uttarakhand from 2005 to 2019. These obtained trends from both the methods were highly correlated (0.86%) with each other. It validates our obtained results from both methods.

Furthermore, the annual and seasonal COV pattern of TCO showed a significant variation as a function of latitude which was increased from low to high latitude stations. The higher value of COV of ozone concentration occurred in winter whereas the lowest values occur at monsoon for all selected sites. Besides, seasonal variation was investigated through backward trajectories in association with continental and maritime during the study period. It showed that maximum ozone concentrations in different seasons are a consequence of the transportation of air mass trajectories. These results are footprints to correlate the TCO variation and climatic factors such as temperature, relative humidity, rainfall, and zonal wind, etc. Apart from this, a comparative study for two data sets i.e. OMI and OMPS estimated average relative difference was merely 3%. Indeed, the daily data TCO from 2012–2019 was highly correlated for every year in the range of 68% to 99%. The outcomes of TCO distribution and trends were found similar to both the datasets. These results suggest that although both data sets used in this study i.e. OMI and OMPS have different resolutions but their measurements complement each other. Highly correlated outcomes from both these datasets enhance the validity of the results obtained in this study as well. This study provides a detailed statistical interpretation and estimation of temporal and spatial variations of TCO over the Himalayan region i.e. Uttarakhand, India. The estimated results confirm a crucial variation in TCO in Uttarakhand over the study period. It is expected that the obtained outcomes would be helpful for the development of further studies of ozone variation (TCO variation) over other geographical regions as well.

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