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Assessment of Spatio-Temporal Variability of Total Columnar Ozone Over Jammu and Srinagar, India

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KEYWORDS

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ABSTRACT:

Total Columnar Ozone (TCO) is an important factor for air quality studies. This paper presents the trend analysis of TCO over the regions of Jammu and Srinagar. TCO datasets from the Atmospheric Infrared Sounder (AIRS) sensor spanning from 2003 to 2022 have been used to ascertain trends in TCO without eliminating the seasonal discrepancies, quasi-biennial oscillation (QBO), solar cycle impacts and ENSO factors. Long term trends obtained with the TCO time series indicate a decline in ozone concentration by -0.03 DU/year over Jammu, while no statistically significant trend is observed over Srinagar. Although there is a natural variation in ozone levels with changing seasons, trend analysis suggests a higher level of TCO concentrations in the winter and the pre-monsoon season as compared to the succeeding periods. The variability of TCO with respect to the meteorological parameters establishes an inverse correlation with the temperature. Thus, contributing to understanding the intricate interplay of factors influencing TCO variations, making a significant stride in understanding atmospheric dynamics.

1. Introduction

The existence of life on this planet cannot be assumed without the presence of a favourable atmosphere (Kasting 1993). A number of biogeochemical cycles occurring over Earth have transformed the atmosphere from its initial hostile phase to its present composition (Shakoor et al. 2020). However, over the last century, especially after the industrial revolution, progressions in human technology have caused serious perturbations in the natural composition of atmosphere (Manisalidis et al. 2020; Allenby 2013). For instance, one of the serious consequences is the depletion of the stratospheric ozone also termed as 'ozone hole' (Ghosh and Midya 1994).

Ozone is a natural gaseous layer which acts as a blanket against the harmful ultraviolet rays produced by the sun (Petrescu et al. 2018). Ozone is present in the atmosphere in trace quantities. The presence of ozone in the atmosphere has severe impacts on human health (Kumar et al. 2015; Zhang et al. 2004), agricultural productivity (Feng et al. 2016; Burney and Ramanathan 2014), infrastructure and materials (Kambezidis and Kalliampakos 2013). Furthermore, it is an air pollutant as it aids photochemical smog formation (Geddes and Murphy 2012; Crutzen 1974), acts as a greenhouse gas (McFarlane 2008; IPCC 2014), is a primary species for the formation of hydroxyl radicals (Seinfeld and Pandis 2016) and is also a regulator of the tropospheric oxidative potential (Thompson 1992).

TCO is the total ozone measured in Dobson Unit (DU), present over 1 cm^2 of base area in a vertical air column (Hegglin et al. 2015; Mahendranth and Bharathi 2012). Stratospheric ozone makes up 90% of the TCO (de Forster et al. 1997; Shukla et al. 2017). Most ozone is produced in the tropical regions (Grewe 2006), which is carried by the stratospheric winds towards the poles (Richter et al. 2017). Thus, the protection received from the harmful UV radiation is reduced near the equator (Narayanan et al. 2010). A natural variation in levels of ozone is observed, with peak concentrations in early spring and lowest concentrations in the season of fall (Lee and Park 2022).

Globally, the influence of anthropogenic activities has led to a decline in the TCO over the last few decades (Collins et al. 2000). This decrease is predominantly noticeable at the poles and mid-latitudes (WMO 2018).

Since the late 1970s, the functioning of stratospheric ozone layer as a protective layer has been periodically found to be disrupted by the emission of chlorine-containing synthetics by humans into the atmosphere (Parson 2003; VanLoon and Duffy 2017). These have been found to cause an ozone hole over the Southern Pole and the appearance of this hole has been realised in the spring season annually (Farman et al. 1985).

The first ozone hole was discovered in mid-1980 over Antarctica by Dr. Joe C. Farman and his team (Godrej 2001). The British Antarctic Survey had been monitoring ozone levels since 1957. Data from satellite and ground-based stations confirmed the gradual fall of ozone levels between mid-September and mid-October, with abrupt declines starting in the late 1970s (Stolarski et al. 1986, Farman et al. 1985). Recent research indicates ozone depletion as a universal issue, although its effects cannot be extrapolated to other locations (Atkinson et al. 1989; Bojkov et al. 1990; Stolarski et al. 1991, 1992).

However, signs of ozone recovery have been reported in the mid and high latitudes owing to the concerted efforts made under the Montreal Protocol and its associated mechanisms (Nair et al. 2015; Chipperfield et al. 2015; Solomon et al. 2016; de Laat et al. 2017; Kuttippurath and Nair 2017; Pazmiño et al. 2018; Kuttippurath et al. 2018; Weber et al. 2018). Nonetheless there is no uniformity in this revival across different latitudes (WMO 2018), specifically with persistent inconclusiveness in the existing trends at the lower latitudes.

Reporting the deviation and trend of the total ozone column is significant in discovering its behaviour (Reinsel et al. 2005; Solomon 1988). One method is by studying the abrupt alterations, which are observed when a swift change results in a temporally prolonged new condition of the climate system due to crossing of a tipping point (Alley et al. 2003). Another method for the study of TCO is achieved by utilising the trend analysis approach (Bojkov and Fioletov 1995; Reinsel et al. 1994) and has been made imperative owing to the escalating and intensifying apprehensions of the declining levels of stratospheric ozone and growing concentrations of tropospheric ozone. However, to understand the variations in the ozone levels in totality and thus, the dynamism of the process, a foray into measures such as means, seasonal alterations and extreme values is required.

The emission and variability of ozone are inadequately documented over South Asia fundamentally limiting its use for modelling of air and climate studies (Beig and Ali 2006). Therefore, the present work focuses on analysing a satellite derived TCO product spatially as well as temporally and to realise the associations to meteorological parameters over two urban locales in North-western India. Another objective is to analyse the trend of TCO variability over the study locations.

2. Study Area

For the purpose of this investigation two urban locations of North-western Himalayas located in the Union Territory of Jammu and Kashmir (**Table 1**, **Fig. 1**) have been specifically chosen for measuring the spatial and temporal variation in the vertical ozone column in Dobson Units (DU).

The city of Jammu is located on the banks of river Tawi. Geographically it is situated at 32.73°N 74.87°E with an average altitude of 350 m (Alam et al. 2021) above the mean sea level. Jammu city is situated on the jagged ridges of muted heights of the Shivalik hills which are present to its north, east and southeast. The Trikuta range borders it in the north-west (Gupta and Gupta 2017). Similar to other parts of north-western India, Jammu is characterised by extreme summer temperatures reaching 46°C, while the winter months record temperatures occasionally falling below 4°C (Maharana and Ray 2013). Climatologically, the area is part of the humid subtropics.

The second site is Srinagar and its co-ordinates are 34°5′24″N 74°47′24″E (Bona and Lone 2022). Located in the Kashmir Valley, Srinagar lies on the banks of the Jhelum River, and has several lakes including the famous Dal and Anchar lakes. The average elevation of the city is 1,585 m (Anees 2022) above the mean sea level. Despite the humid subtropical climate, it experiences cool winters, with average daytime temperature touching 2.5°C (Savio et al. 2022). However, at night the temperatures drop below freezing point owing to its setting amidst the Himalayas which surround it on all four sides (Negi 1998).

Table 1. The latitudes and longitudes of the two urban locations

Study Sites	Latitude		Longitude	
Jammu	33.1˚N	32.34˚N	74.67˚E	75.044˚E
Srinagar	34.270656˚N	33.896656˚N	74.41737˚E	75.177371˚E

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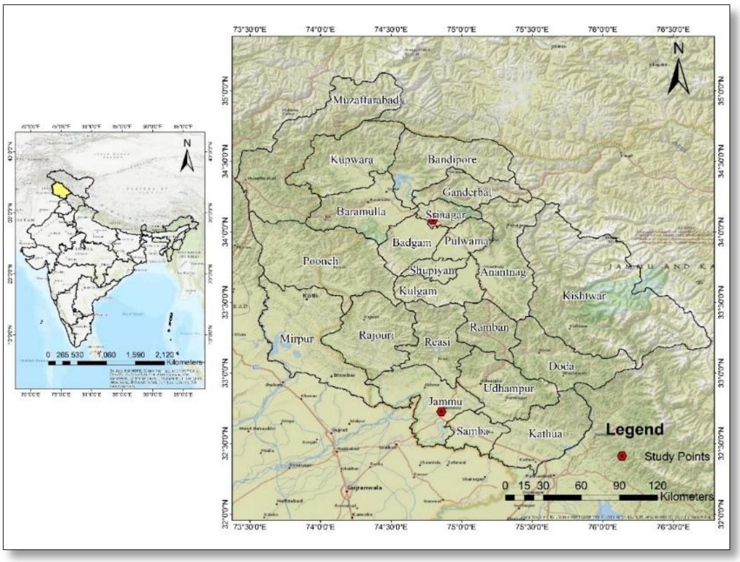

Fig. 1 Study area map depicting study sites Jammu and Srinagar

The urban centres of Jammu and Srinagar are the two largest cities in the Union Territory of Jammu and Kashmir, however the lack of ground-based observation sites for the measurement of TCO, makes the analysis of TCO difficult. In absence of previously conducted studies to assess the long-term variability of TCO over the region, the current study attempts to draw inference by utilising satellite datasets.

3. Data and Methods

3.1 Datasets

(a) AIRS3STD V7.0 products

AIRS, i.e., Atmospheric Infrared Sounder sensor, is among the six instruments which are aboard EOS Aqua (Earth Observing System Aqua), a sun synchronous, near polar orbit satellite. AIRS functions as a grating spectrometer with a spectral resolution (R) of 1200. The AIRS dataset used is AIRS3STD, which is short name for AIRS/Aqua L3 Daily Standard Physical Retrieval (AIRS-only) 1-degree x 1-degree V7.0. It has a spatial coverage over the entire globe ranging from -180.0, -90.0, 180.0, 90.0 and has been operational since 01-09-2002 to present. The AIRS L3 daily product has been employed to tackle the high occurrence of variations in the climatic system. The data is resolved at 1° x 1° spatially and 1 day temporally. The product is however, separated depending on the crusading of the satellite sub-point in its track, into ascending and descending orbit. The ascending node has an equatorial traversing time of 1330 hours local time whereas the descending node crosses the equator at 0130 hours local time. (GES DISC n.d.; Tian et al. 2020)

Time averaged diurnal data of AIRS, on a daily basis has been downloaded for a period of 20 years starting from January 2003 up till December 2022 from the Giovanni and the Earth data search tool portals of the Goddard Earth Services Centre, NASA, USA.

(b) OMTO3d V3 products

The Ozone Monitoring Instrument (OMI) is designed as UV/VIS imaging spectrometer onboard EOS-Aura satellite launched on 15, July 2004 (Torres et al. 2007). OMI has been operational since August 9, 2004. With a swath of approximately 2600 km OMI covers the globe daily and crosses the equator at 1345 hours. However, with the occurrence of the row anomaly since June 25th, 2007, OMI's temporal coverage to cover the globe has increased to two to three days (Torres et al. 2013). The spectral resolution of OMI is of the range of 0.42 nm-0.63 nm. The mapping of the ozone columns is performed for two profiles: 13 km x 24 km and 13 km x 48 km (Ahmad et al. 2003).

(c) CRU TS 4.05 products

Meteorological data utilised in the research were acquired from Climate Research Unit (CRU). Extraction of the data for each grid point of resolution 0.5° by 0.5° was carried out. The mean monthly temperature measured in degree celsius (°C) along with the precipitation in millimetres (mm) has been computed (Harris et al. 2020).

3.2 Statistical Analysis

The time averaged diurnal AIRS TCO dataset was first imputed for missing values and then averaged to get a daily average value of TCO for each of the stations. From these daily TCO observations, monthly and annual means of TCO were calculated. Other parameters such as the largest and smallest values, the annualised standard deviation (ASD), the annual coefficient of relative variation (ACRV), were calculated. The ACRV

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was computed (equation 1) to probe the relationship between total ozone variability and the climate. The percentage variability of ozone (A_i) was also calculated using equation 2 (Eresanya et al. 2017).

$$ACRV = \frac{ASD}{X_a} \times 100 \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (1)$$

$$A_i = \frac{R_i}{Q_i} \times 100 \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (2)$$

Where, X_a is the annual mean TCO in DU, R_i is the range of TCO over a year i and Q_i is the maximum TCO concentration for the year i.

3.3 Trend Analysis

(a) Mann-Kendall test

Mann Kendall test is a frequently employed and popular technique for trend analysis (Das et al. 2020; Das and Bhattacharya 2018; Rahman et al. 2016, 2017). It compares every value of the temporal dataset with all the remaining values, in a chronological manner. It then calculates the number of times that the residual temporal terms are greater than the dataset under consideration (Jaswal et al. 2014). The variance S of a series is evaluated using equation 3 (Mann 1945; Kendall 1975):

$$S = \sum_{x=1}^{n-1} \sum_{y=j+1}^{n} sign(p_y - p_x) \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (3)$$

Where n is the total number of observations, and p_y and p_x are the corresponding values at time y and x,

$$\text{Sign}(p_y - p_x) \begin{cases} = +1 \text{ if } p_y - p_x > 0 \\ = 0 \text{ if } p_y - p_x = 0 \\ = -1 \text{ if } p_y - p_x < 0 \end{cases} \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (4)$$

and

$$Var(S) = \frac{1}{18}\{n(n-1)(2n+5) - \sum_{j=1}^{z} t_j(t_j-1)(2t_j+5)\} \ldots\ldots\ldots\ldots\ldots (5)$$

Where, Var(S) measures the variance of the statistic S, z is the number of tied groups and t_j is the size of the j^{th} tied number. Tied groups are a set of data having similar values.

Kendall's tau (τ) is used for the identification of the statistic S and is given as:

$$\tau = \frac{S}{B} \ldots\ldots (6)$$

and the standardized Z statistic is used to determine the presence of a statistically significant trend and is given by:

$$Z \begin{cases} = \frac{S-1}{\sqrt{Var(S)}} & \text{if } S > 0 \\ = 0 & \text{if } S = 0 \\ = \frac{S-1}{\sqrt{Var(S)}} & \text{if } S < 0 \end{cases} \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (7)$$

A positive value of the Z statistic signifies a rising trend, whereas a negative value shows the presence of a decreasing trend.

(b) Autocorrelation function

The existence of autocorrelation in a data series poses a problem in trend detection. A positive autocorrelation strengthens the occurrence of a Type I error (false positive) in the analysis. It detects the presence of a significant trend when in actuality there is an absence of such a trend (Yue et al. 2002). Hence, it becomes imperative to test the datasets for presence of serial correlation. For the purpose of this study lag-1 autocorrelation has been evaluated and analysed at the 0.05 significance level (Das et al. 2021).

$$r_k = \frac{\sum_{k=1}^{N-k}(x_t - x_t)(x_{t+k} - x_{t+k})}{[\sum_{k=1}^{n-k}(x_t - x_t)^2(x_{t+k} - x_{t+k})^2]^{0.5}} \ldots\ldots\ldots\ldots\ldots\ldots (8)$$

Where r_k is the autocorrelation coefficient at lag k for the time series x_t. N is the total length of the time series.

The dataset is considered to be autocorrelated if r_k falls between the extreme margins of the confidence intervals (Anderson 1954), else it is considered to be serially independent.

(c) Modified Mann-Kendall test

The modified Mann-Kendall test is used when serial correlation occurs in the dataset (Hamed and Rao, 1998). The modified variance (S) is calculated by using equation 9 (Das et al. 2020).

$$Var(S) = \frac{1}{18} n(n-1)(2n+5).(\frac{n}{n_e^*}) \ldots\ldots\ldots\ldots\ldots (9)$$

$(\frac{n}{n_e^*})$ is the correction factor which has been adjusted to the autocorrelated data and

$$(\frac{n}{n_e^*}) = 1 + (\frac{2}{n^3 - 3n^2 + 2n}) \sum_{f=1}^{n-1}(n-f)(n-f-1)(n-f-2)\, \rho_e(f) \ldots\ldots\ldots\ldots (10)$$

Where ρ_e (f) measures the autocorrelation between the ranks of the observed values and is given as,

$$\rho(f) = 2\sin(\frac{\pi}{6}\rho_e(f)) \ldots\ldots\ldots\ldots\ldots (11)$$

(d) Sen's Slope estimator

Sen's method (Sen, 1968) is employed to evaluate the rising or falling slope of the time series TCO datasets.

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The magnitude of the slope is calculated by considering the median of slopes of all data pairs (Jaswal et al. 2014).

$$m = median\left[\frac{P_y - P_x}{y - x}\right], for\ all\ y < x \ldots\ldots\ldots\ldots (12)$$

Where, m is the slope between points P_y and P_x

4. Results and Discussion

4.1. Spatiotemporal variation of TCO over Jammu

The TCO trend over the years shows no statistically significant variation when compared to their annual averages. However, the year 2015 shows a maximum level of mean annual TCO observed over this region with a value of 295.2 ±18.51 DU while the lowest value for the same has been observed subsequently in the year of 2016 with an average TCO value of 283.3 ± 18.1 DU (**Table 2**). The greatest variation from the measure of central tendency i.e., mean, is observed in 2005 which shows a dispersal of 21.79 from the mean. Analysing the seasonal trends, it was observed that during winters (January, February) and the pre-monsoon (March, April and May) relatively higher values of TCO was observed as compared to the months of monsoon and post monsoon (**Fig. 2**).

Table 2. Statistical Analysis of ozone over Jammu

Year	Annual Average TCO (DU)	Maximum Mean Monthly TCO (DU)	Minimum Mean Monthly TCO (DU)	Annual Standard Deviation (ASD)	Average Temperature (°C)	Average Rainfall (mm)	Annual Coefficient of Relative Variation (ACRV)	% variability of TCO
2003	291.75	310.07	270.87	19.39	21.97	88.52	6.65	12.64
2004	286.16	296.42	275.08	15.03	22.68	66.78	5.25	7.20
2005	290.43	316.04	264.87	21.79	21.84	74.43	7.50	16.19
2006	288.21	302.85	280.61	15.81	22.61	118.35	5.49	7.34
2007	286.19	302.09	261.59	19.97	22.63	72.38	6.98	13.41
2008	286.81	305.38	267.52	16.85	22.31	87.58	5.88	12.40
2009	288.74	298.88	274.22	13.94	22.54	54.44	4.83	8.25
2010	290.40	316.43	268.37	20.12	22.69	90.01	6.93	15.19
2011	285.76	307.58	266.31	17.62	22.48	84.82	6.17	13.42
2012	291.73	304.36	277.64	16.45	22.08	68.50	5.64	8.78
2013	288.60	298.64	273.12	16.76	22.43	97.43	5.81	8.55
2014	291.16	317.02	269.80	18.27	22.18	85.39	6.27	14.90
2015	295.16	309.97	281.13	18.51	22.16	105.11	6.27	9.30
2016	283.28	302.00	253.18	18.12	23.16	74.43	6.40	16.16
2017	285.85	300.63	273.23	15.26	22.88	76.31	5.34	9.12
2018	291.42	309.20	270.46	17.90	22.94	89.47	6.14	12.53
2019	290.87	303.62	278.47	19.03	21.98	115.74	6.54	8.28
2020	291.07	313.40	265.40	21.42	21.93	117.57	7.36	15.32
2021	290.23	307.99	272.94	17.72	22.67	106.43	6.11	11.38
2022	288.28	312.39	268.81	17.72	23.09	129.69	6.15	13.95

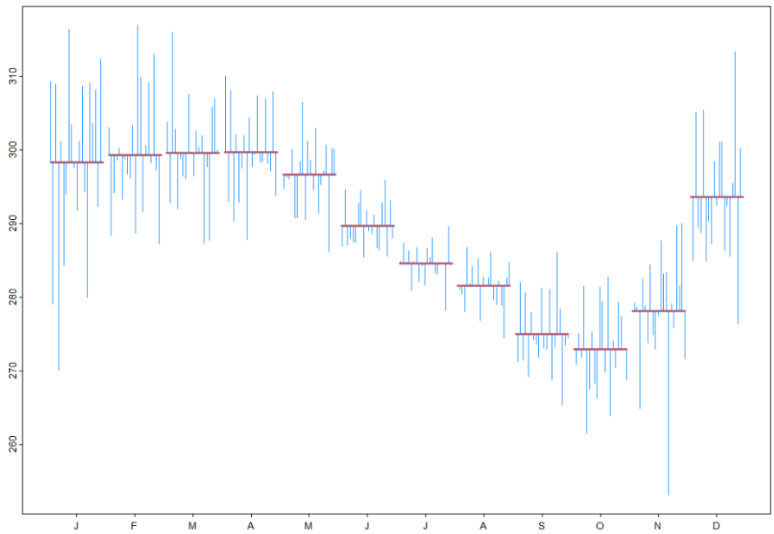

Fig. 2 Monthly distribution of TCO over Jammu for 2003-2022

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4.2. Spatiotemporal variation of TCO over Srinagar

The urban centre of Srinagar records the TCO in the range of 279.5 ±18.7 DU to 289.1± 17.9 DU. It has the highest mean annual value of TCO for 2015, and the lowest normative value in the following year of 2016. The upper limit of the spread of the TCO values is witnessed in the year of 2005 holding a value of 21.5 (**Table 3**). The February, March and April showcase highest values for the TCO as compared to the remaining months (**Fig. 3**). These trends are similar to those observed in the Jammu region.

Table 3. Statistical Analysis of ozone over Srinagar

Year	Annual Average TCO (DU)	Maximum Mean Monthly TCO (DU)	Minimum Mean Monthly TCO (DU)	Annual Standard Deviation (ASD)	Average Temperature (°C)	Average Rainfall (mm)	Annual Coefficient of Relative Variation (ACRV)	% variability of TCO
2003	286.79	306.91	267.32	20.41	9.63	80.43	7.12	12.90
2004	280.07	286.77	271.12	14.93	10.33	57.40	5.33	5.46
2005	285.17	313.03	266.22	21.50	9.47	62.48	7.54	14.95
2006	282.77	296.87	270.55	15.85	10.23	86.29	5.60	8.87
2007	280.73	295.71	259.17	19.11	10.31	53.70	6.81	12.35
2008	281.55	301.99	264.63	16.83	10.26	63.43	5.98	12.37
2009	284.93	298.51	269.06	16.44	10.31	54.91	5.77	9.87
2010	285.37	307.78	264.26	20.19	10.41	85.33	7.08	14.14
2011	280.06	297.87	263.46	17.84	10.15	60.13	6.37	11.55
2012	287.31	301.16	275.38	17.80	9.76	51.38	6.19	8.56
2013	283.50	290.34	268.95	15.66	10.39	71.38	5.53	7.37
2014	286.17	309.36	268.60	18.19	9.97	63.28	6.36	13.18
2015	289.10	304.99	275.80	17.93	9.98	88.13	6.20	9.57
2016	279.51	301.99	251.45	18.70	10.76	49.82	6.69	16.74
2017	280.63	296.40	267.04	14.75	10.47	57.82	5.26	9.91
2018	286.63	311.88	267.73	18.66	11.79	35.37	6.51	14.16
2019	285.96	297.56	272.95	18.07	10.74	55.38	6.32	8.27
2020	286.25	306.49	261.00	21.33	10.73	58.82	7.45	14.84
2021	285.61	303.51	269.43	18.58	11.38	42.50	6.51	11.23
2022	281.93	310.87	265.07	18.23	11.97	48.38	6.46	14.73

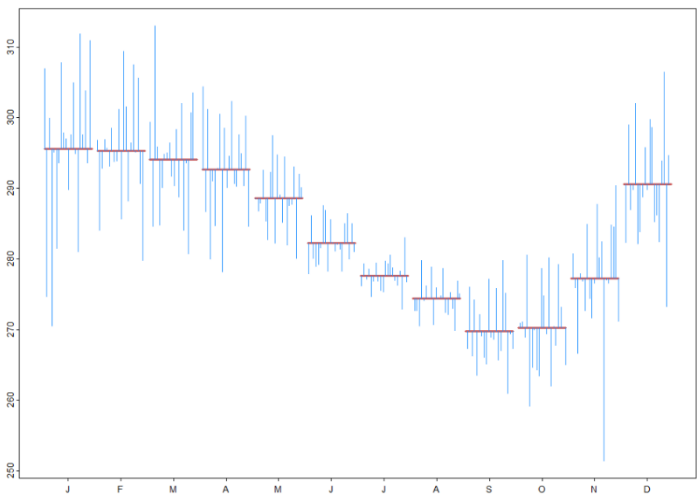

Fig. 3 Monthly distribution of TCO over Srinagar for 2003-2022

4.3. Comparison of TCO levels over Jammu and Srinagar

In general, the TCO levels over Jammu are greater than the TCO levels over Srinagar, however, this distribution is heterogeneous. The distribution of TCO over the entire area of Jammu and Kashmir for the period of 2003-2022 is shown in **Fig. 4**. It clearly shows the variation in concentrations of TCO for two different spatial resolutions, viz. AIRS (1°× 1°) and OMI (0.25°× 0.25°). Further analysis using the Empirical Cumulative

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Distribution Function (ECDF) and Box plots is performed (**Fig. 5**). For Jammu, 20 to 80% of TCO was distributed between 270-300 DU. The ECDF plot for Srinagar was shifted to the left to that of Jammu, representing lower values of observed TCO over Srinagar as compared to Jammu, with 20-80% values lying between the TCO data range of 270-290 DU. Moreover, the boxplots also reveal the higher TCO concentrations observed over Jammu city as compared to Srinagar city.

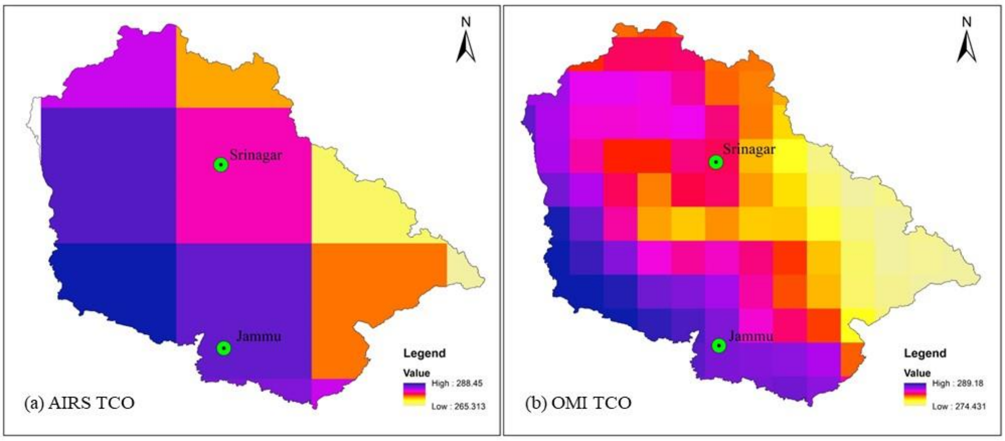

Fig. 4 Spatial distribution of time averaged (2003-2022) TCO (DU)

The seasonal trends for both the stations follow a similar pattern with higher TCO levels observed in the winter months (January and December) and in the pre-monsoon months (March, April and May). The latitudinal location, tropopause height and atmospheric crescendos (Tian et al. 2008) are the major drivers controlling the variation in TCO. Other factors controlling the seasonal variation are the weather conditions and topography (Rajab et al. 2013). Srinagar lies in a cold mountainous region, having low temperatures and subsequently low tropopause height (Tian et al. 2008), causing concentrations of ozone to increase in winters and spring. This observation is in concurrence with the findings of Chen et al. (2014). However, the seasonal disparity in TCO intensity is complex, as the major contribution of TCO comes from the lower stratosphere where the life time of photochemical species range from several months to years (Banks and Kockarts 1973). The higher TCO levels over Jammu can also be ascribed to the relative industrialisation and congestion in its urban zone (Fishman et al. 1996; Rajab et al. 2013) as compared to the urban zone of Srinagar.

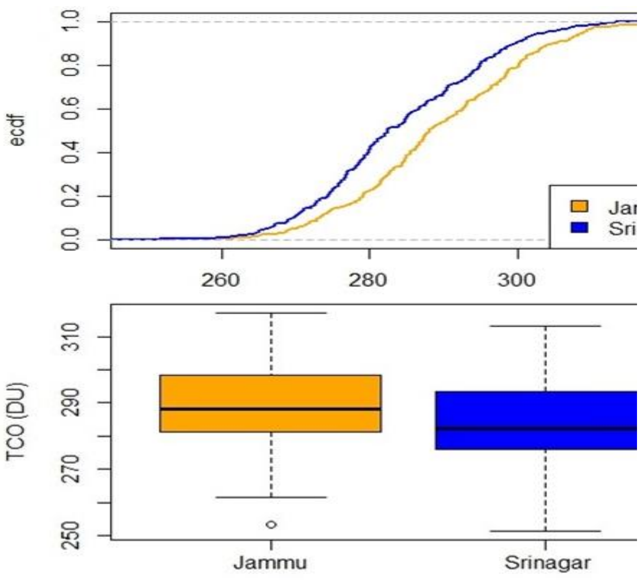

Fig. 5 ECDF and boxplot of TCO over the study sites

The formation of ozone is favoured in pre-monsoon as compared to winters, owing to enhanced incoming solar radiation which results in elevated temperatures (Ahmed and Yaseen 2005), however the TCO variability in the pre-monsoon is controlled by the atmospheric dynamics and not by the chemistry of ozone formation (Liu et al. 2010). Furthermore, a decline in ozone concentration and appearance of small holes occur during pre-monsoon due to seasonal alteration of the tropopause height and increased convective activity (Zou 1996). Additionally, the presence of increased amount of solar flux which reacts with the accretion of NOx and hydrocarbon built up during winter may result in localised production of ozone in the pre-monsoon months (Vingarzan 2004). The upper tropospheric and lower stratospheric zones are also under the influence of the South Asia high monsoon anticyclonic circulation (Randel and Park 2006). TCO levels over South Asia continue to fall during the monsoon, but a homogenous and direct relationship cannot be established between the two.

The increased residence time of ozone during winters also contributes to an elevated amount of ozone during winters (Li et al. 2021). This is further facilitated by the position of the sun and the angle at which its rays are incident during the winters, protecting the ozone from destruction (Rowland 1990). Another factor which can be encompassed is the inter-continental pollution and its transfer and transport from a source to a destination, with the help of winds which change their direction seasonally (Fu et al. 2012; Sharma et al. 2017).

The findings of this investigation are in consonance with the previous studies conducted in the nearby regions of the Hindukush Himalayas and Tien Shan Mountain regions, which corroborates the seasonal and regional variation in TCO (Rafiq et al. 2017; Songa et al. 2015; Zahid and Rasul 2010). The northern regions, latitudes greater than 30°N, record higher TCO (≥310 DU) during winters and spring as compared to lower TCO values (285–300 DU) in summers and autumn. The elevation in the TCO in the northern regions can be attributed to the mechanism of blockage of the movement of ozone by subtropical westerly jet towards its north (Kuttippurath et al. 2023). The Southern regions which are impacted by the Tropospheric and Stratospheric dynamics (Liu et al. 2010), shows lower TCO values during winter and higher concentrations of TCO in spring.

Furthermore, seasonal variations over different temporal and spatial scales have been reported by Zou et al. (2020), Zhang et al. (2014), Zou (1996) with consistent findings as the findings of the current study. An upsurge in TCO levels was observed in winters and spring and a fall in the concentrations of TCO in the monsoon and post-monsoon seasons by Midya et al. (2003). A higher amount of reduction in TCO was observed by Midya et al. (2011) and Midya and Saha (2011) during the monsoons in comparison to post-monsoons. However, contrary to our study which records lower level of TCO over high latitudes (Srinagar) and a higher level of TCO at lower latitudes (Jammu), certain studies report a reverse trend (Rafiq et al. 2017; Chen et al. 2014; Siddiqui et al. 2001) and this can be attributed to the geographical and topographical differences in the regions under study.

4.4. TCO vs. Meteorological Parameters

For the purpose of establishing a link between the variability of TCO with climate, bivariate correlation was performed between the meteorological parameters which included the average annual temperature and average annual precipitation with the ACRV for both the stations (**Table 4**). For the entire duration of the study, both Jammu and Srinagar showcased a negative correlation, varying from very high to moderate, between the average annual temperature and the ACRV, indicating that temperature and the ozone variability are inversely proportional (Barnett et al. 1975). These results corroborate with earlier researches that the variability of TCO is concomitant with the photochemical coupling that exists between ozone and temperature (Steinbrecht et al. 2003; 2006; Azeem et al. 2001; Chandra et al. 1996) and the seasonal atmospheric circulations are responsible for movement of ozone between locations causing variations in the concentration of ozone (Shangguan et al. 2019; Brunamonti et al. 2018; Varotsos et al. 2017a; 2017b; Varotsos and Kirk-Davidoff 2006; Varotsos 2004).

Table 4. Correlation between meteorological parameters and ACRV

Year	Jammu		Srinagar	
	Correlation of ACRV of ozone with average annual temperature	Correlation of ACRV of ozone with average annual rainfall	Correlation of ACRV of ozone with average annual temperature	Correlation of ACRV of ozone with average annual rainfall
2003	-0.78	-0.50	-0.69	0.20
2004	-0.76	-0.60	-0.69	-0.17
2005	-0.64	-0.41	-0.76	0.31
2006	-0.81	-0.30	-0.72	-0.32
2007	-0.77	-0.35	-0.84	-0.15
2008	-0.80	-0.55	-0.97	-0.43
2009	-0.85	-0.54	-0.78	0.26
2010	-0.74	-0.44	-0.69	-0.30
2011	-0.78	-0.53	-0.83	-0.07
2012	-0.73	-0.48	-0.68	0.21
2013	-0.80	-0.43	-0.78	-0.45
2014	-0.77	-0.45	-0.61	-0.02
2015	-0.82	-0.49	-0.91	-0.19
2016	-0.84	-0.59	-0.80	-0.64
2017	-0.73	-0.35	-0.69	-0.06

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2018	-0.76	-0.79	-0.93	-0.41
2019	-0.82	-0.20	-0.86	0.47
2020	-0.89	-0.29	-0.91	0.35
2021	-0.74	-0.62	-0.88	0.26
2022	-0.81	-0.34	-0.89	0.12

The correlation of ACRV with the average annual rainfall, resulted in a negative moderate to negligible values for the correlation coefficient for the study site of Jammu. In case of Srinagar the correlation coefficient values varied with both positive and negative values, however, the correlation was not found to be significant, thus, negating any relationship between the ACRV and the precipitation.

4.5. Trend Analysis of TCO

The time series under consideration extends from 2003-2022 with a duration of 20 years, for both the locations of Jammu and Srinagar. The trend for Jammu is statistically significant at 0.05 level and shows a waning in the TCO intensity, with a decay of -0.03 DU/year (**Table 5**), analogous to the observations by Tandon et al. (2010), affecting over 40% of the Indian landmass, but no statistically significant trend was observed over South India. A declining trend for TCO has been observed over twelve stations in India for the period of 1979-2010, with TCO levels ranging from 210 DU to 380 DU and the variation in the slope of the trend line varying from 0.004 to 0.008 (Bhattacharya et al. 2012). Similarly, Ningombam et al. (2018) and Sahoo et al. (2005) have also conveyed a significant declining trend in TCO levels over northern India and Ladakh region respectively. In case of Srinagar, the trend is insignificant.

Table 5. Trend analysis of TCO levels over Jammu and Srinagar

Test	Parameter	Jammu	Srinagar
Mann-Kendall Test	Z-statistics	**-2.32**	0.15
	P-value	**0.02**	0.88
	Variance	1545525.67	1545533.33
Modified Mann-Kendall Test	Lag-1 ACF	0.48	0.65
	N/N*	0.23	0.10
	Z-statistics	**-4.86**	0.48
	P-value	**0.000001**	0.63
	Variance	351465.12	156342.77
	Tau	-0.10	0.01
	Sen's slope	-0.03	0.001563

The multiplicative decomposition of the time series into its individual constituents viz. trend, seasonal and random components is also performed for both Jammu and Srinagar (**Fig. 6** and **Fig. 7**). The decomposition helps remove the impact of seasonality from the observed dataset to exhibit a trend free from the seasonal influences. No homogenous unidirectional trends exist for both Jammu and Srinagar.

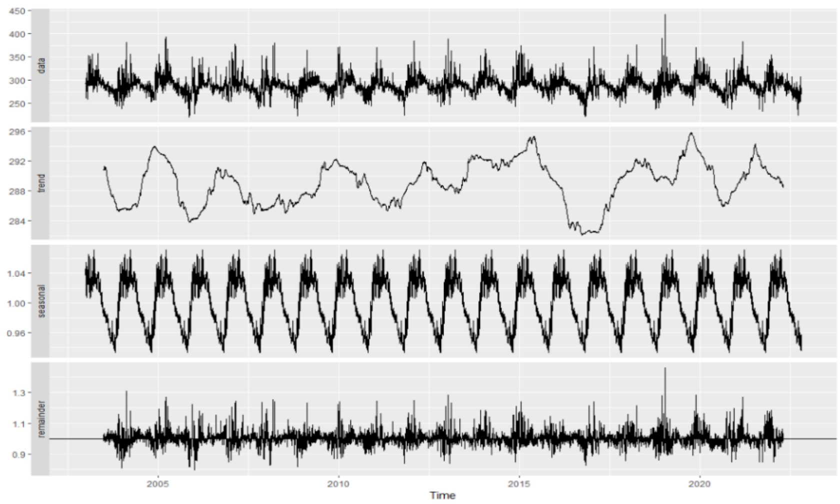

Fig. 6 Multiplicative decomposition of TCO trend over Jammu

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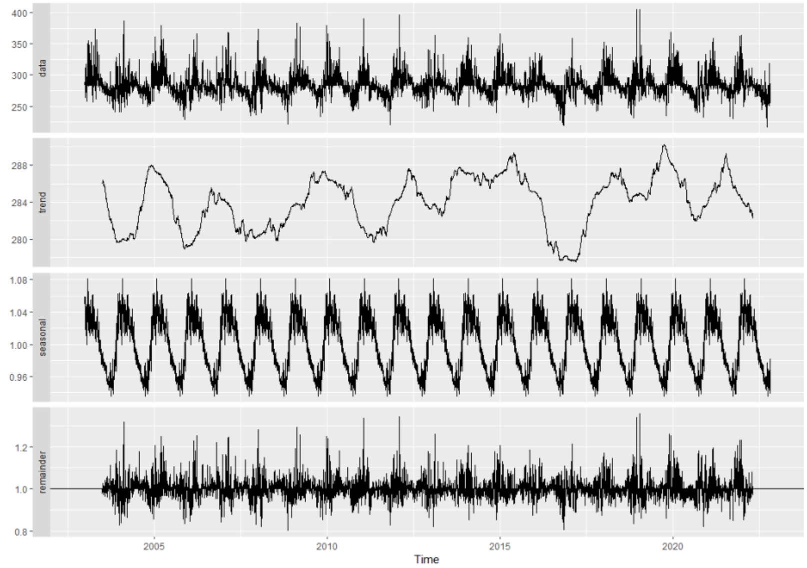

Fig. 7 Multiplicative decomposition of TCO trend over Srinagar

A decline in the trend of TCO, has substantial and extensive impacts not only on the environment, but also on human health, and climate (McKenzie et al. 2011; Martens, 1998). The Montreal Protocol has significantly contributed to the reduction of ozone-depleting substances (ODS) production and consumption (Victor, 2011). However, the annual variation in TCO conceals the attribution of the trends to the diminishing ODS concentrations (Bernhard et al. 2023) necessitating further investigation into their impact on humans and the environment.

4.6. Correlation analysis of AIRS TCO and OMI TCO

Validation of AIRS TCO and OMI TCO datasets for both the study sites of Jammu and Srinagar was done using correlation analysis. Both Jammu and Srinagar recorded strong correlation at very high levels of significance (α<0.01) between the AIRS and OMI TCO datasets. Scatterplot for the average monthly TCO datasets of AIRS and OMI shows high correlation (red points) and low correlation (green points) with the LOESS smoothed fit line (red line) (**Fig. 8).**

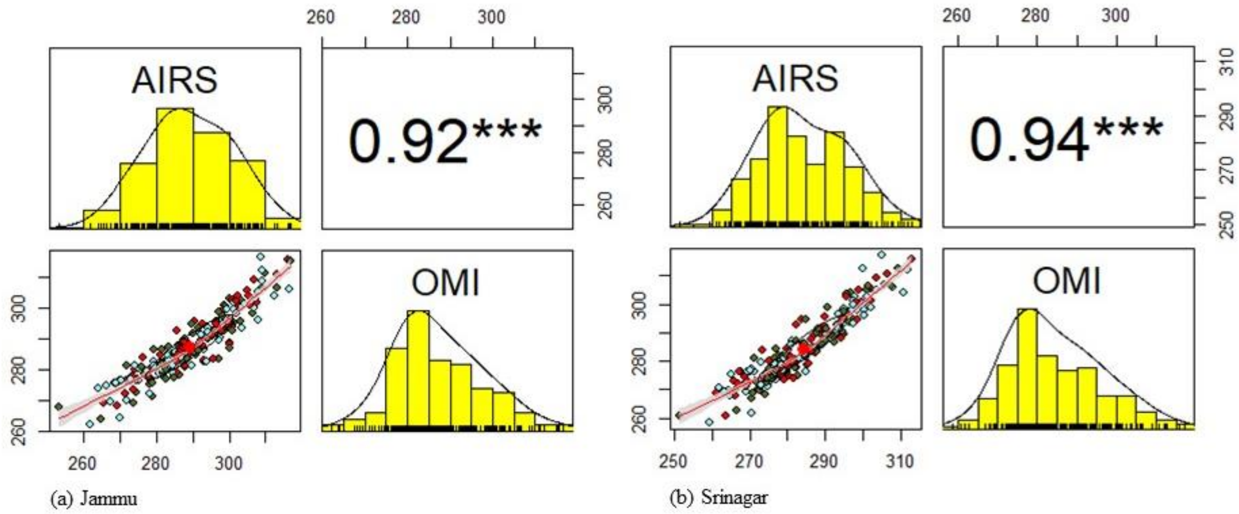

Fig. 8 Validation of TCO datasets of two satellites for Jammu and Srinagar

5. Concluding remarks

The long-term trend of TCO at two geographical locations experiencing varying environmental conditions was investigated. A logical variability in the concentrations of TCO is observed with maximum in December-February and March-May and minimum in

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the other periods. Profusion of variation is greater over Jammu as compared to Srinagar. Measurements show that heightened TCO over Jammu can be attributed to the demographic factors such as the population density and other human induced influences along with the influence of geographical, climatological and topographical factors. Long term trend of TCO computed using the non-parametric tests (Mann-Kendall and modified Mann-Kendall), without accommodating seasonal alterations, solar cycle, QBO, and ENSO, indicates that the TCO trends over Jammu for the period of 2003-2022 are declining significantly ($\alpha<0.05$) at -0.03 DU/year and no significant trend is observed over Srinagar. However, the real mechanism of the production and decomposition of ozone is a complex one and requires further investigation so as to develop a comprehensive understanding along with determining the effects of various other factors such as the QBO, ENSO, solar activity etc. Furthermore, the unavailability of IMD ground-based measurement datasets for Jammu and Kashmir, to further validate the findings of the present study, needs to be addressed. Satellite measurements are currently the only sources for providing continuous TCO observations and have been utilised in the course of this study without validation from ground-based installations.

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Statements and Declarations

- **Ethics approval and consent to participate:** *Ethical review and consent to participate was not required for the study.*
- **Ethical Responsibilities of Authors:** *All authors have read, understood, and have complied as applicable with the statement on "Ethical responsibilities of Authors" as found in the Instructions for Authors.*
- **Availability of data and material:** *The original contributions presented in this study are done using open-source data, included in the article, further inquiries can be directed to the corresponding author.*
- **Competing interests:** *The authors declare that they have no conflict of interest*
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- **Authors' contributions:** *All authors contributed to the study's conception and design. Nazuk Bhasin and Sudhanshu Kumar analysed the data and wrote the main manuscript text. Amit Kumar Tiwari helped with the statistical analysis and the interpretation of results. Anil Barla, Rakesh Kumar and Gopal Shankar Singh reviewed and improved the draft substantially. All authors reviewed and approved the final manuscript.*

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