Regional Air Quality: Forest Fires Impacts of SO2 Emissions on Air Pollutants in the Himalayan Region of Uttarakhand, India

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Abstract

Sulfur dioxide (SO$_2$) is a toxic with adverse health effects on respiratory tract, eyes, mucous membranes and skin. In the present study, the continuous ground-based SO$_2$ monitoring has planned over Srinagar Garhwal Valley of Uttarakhand. In the monsoon (M-2018), Post-monsoon (PoM-2018), Winter (W-2019) & Pre-monsoon (PrM-2019) & M-2019 have high SO$_2$ concentrations (3.66 ± 2.05 µg/m$^3$, 5.54 ± 2.23 µg/m$^3$, 6.42 ± 1.79 µg/m$^3$, 7.56 ± 3.53 µg/m$^3$, 6.45 ± 3.49 µg/m$^3$) at 1900 LT, 2000 LT, 1800 LT, 1900 LT & 1900 LT attributed mainly due to biomass burning and long-range transportation of pollutants. A drastic change in the SO$_2$ concentration has been observed from 4.81 to 17.39 µg/m$^3$ during May 2019 with a strong correlation of 0.61 with fire-counts during extensive forest fire. Whereas Jul 2018 (1.07 ± 0.82 µg/m$^3$) showed the lowest SO$_2$ concentration due to wet scavenging process. Temperature, humidity and wind speed have significant correlation with SO$_2$ in different season. Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model trajectories and cluster analysis indicate the transportation of air mass from the Gulf region, Sahara Desert, Pakistan, Afghanistan to Srinagar with a significant contribution of 40.43–72.29% air mass. We have also observed weekend effects (reduction in the pollutant concentration) in Jul 2018, Sep 2018, Feb 2019, Apr 2019 and May 2019.

1. Introduction

According to the World Health Organization (WHO), approximately 3.8 to 4.2 million people die worldwide every year due to exposure to polluted air (WHO, 2018). Sulfur dioxide (SO$_2$) is one of the pollutants in ambient air, which mainly originate from the industrial power plant (Garg et al. 2001), transportation, combustion of solid fuel for domestic use (Datta et al. 2010), as well as biomass burning of agricultural wastes (Sahai et al. 2007; Sharma et al. 2010a; Jethva et al. 2018). Sulphur (S) has an enormous tendency to form many compounds in gaseous and particulate phases and is collectively known as oxides of Sulphur (SO$_X$) (Skinder 2014). Thus, SO$_X$ includes Sulphur monoxide (SO), Sulphur dioxide SO$_2$, Sulphur trioxide (SO$_3$), and higher Sulphur oxides (SO$_3$ + x). The highest concentration of SO$_2$ in the atmosphere can cause acute to chronic diseases, including respiratory problems, heart diseases, asthmatic problems, lung cancer, etc. (Kampa and Castanas 2008). The SO$_2$ played a significant role in atmospheric chemistry and was attributed in the form of toxic fog, acid rain, new particle formation, socio-economic impacts, etc. (Kumar et al. 2014; Rao et al. 2017; Shen et al. 2012; Singh et al. 2007; Gautam et al. 2018). The variation of SO$_2$ strongly depends upon local metrological parameters such as temperature humidity, wind speed, wind direction and precipitation (Rogalski et al. 2014). The variation of SO$_2$ concentration combined depends upon local and long-range transport of pollutants from industrial areas mainly located in the Indian Gangetic Plan (IGP) region (Tiwari et al. 2016). The Himalayan region is covered by a dense forest of Chir, Pine and Oaks at different elevation range (Vadrevu et al. 2012). Generally, residents burn the litters and pastures of pine in Mar to May of each year which may result in a potential forest fire that affects the water, ecology, air quality and economic system of the Uttarakhand (Bahuguna and Upadhyay 2002; Kumar et al. 2011; Vadrevu et al. 2012). In recent years, there has been
an increase in forest fore activities in the Himalayas from Mar to May which has led to deterioration in
the air quality in Uttarakhand and surrounding areas. In the past, several relevant studies have been
carried out in the foothill of Himalaya (Dehradun, Haridwar and Rishikesh) to estimate the SO$_2$
concentration using a dust sampler but failed to provide better high resolution of SO$_2$ (Chauhan et al.
2010; Deep et al. 2019).

(Naja et al. 2014) conducted the first continuous observation of SO$_2$ concentration from 2019 to 2011
over Nainital (at the elevation of 1958 m AMSL) and examined the impact of transportation of pollutants
at the site. But the Srinagar Garhwal Himalayan region was unexplored and also had an observational
gap of 6 years in the western Himalayan region; So we planned to conduct a continuous observation of
SO$_2$ to fill the gap and explore in the detail such as atmospheric new particle formation events, that have
increased wherein SO$_2$, through its oxidation in the atmosphere (Meng et al. 2015). However,
measurements of SO$_2$ and factors controlling its seasonal variability in Uttarakhand regions are scarce.
This is the first year-round online measurements SO$_2$ in Srinagar Garhwal Valley (SGV), wherein diurnal
and seasonal variation of SO$_2$ and their possible sources along with meteorological parameters have
been presented. Besides, we have also studied the impact of forest fire emissions on SO$_2$ over SGV in the
Himalayas.

2. Material And Methodology

2.1 Sampling area: Observations were carried out at Department of Physics, Hemvati Nandan Bahuguna
Garhwal University Srinagar Garhwal, Uttarakhand, India (30º13'36 N, 78º48'14 E, 640 m AMSL) (Fig. 1).
The Srinagar Garhwal Valley (SVG) is situated over the hard rocks and sand deposited by the Alaknanda
River is called Proterozoic phyllites. The SGV is surrounded by different kinds of vegetation mainly
covered by Chir Pines (Pinus roxburghii), Shecham (Dalbergia sissoo) and Oaks (Querus semicarpifolia)
etc. at a diverse range of elevation and inclination (Kumar and Khanduri 2012). The SGV is located in the
sub-tropical region and experiences a minimum temperature of 2ºC in winter (W). The maximum
temperature 43ºC in the summer season with annual precipitation of 1550 mm (Forest Survey of India,
2009). It is also impacted by the long-range transport of pollutants and western disturbance (relatively
high humidity in winter) every year (Hunt et al. 2018; Sandeep et al. 2020). SGV experiences few cloud
bursts, flash floods, i.e. Kedarnath Flood 2013 (Rana et al. 2013). The observation site is free from the
source of direct emission of pollutants i.e. the residential area and city center are located nearly 1
kilometer (km), 3 km distant apart. Srinagar is a residence of 53,689 people as per the census 2011.
Srinagar is connected by a national highway (NH-58) whose north end is spared to Mana pass (Indo-
China border).

The south end is limited to Ghaziabad via Rishikesh and Haridwar. Every year Srinagar also experiences
high traffic from Apr to Oct during a holy ancient Hindu and Sikh pilgrimage of shrines such as Badrinath,
Kedarnath, Gangotri, Yamunotri and Hemkund Sahib located in Uttarakhand.
2.2. Observations and methodological techniques: The SO$_2$ concentration was measured by using the classical fluorescence spectroscopy principle. When SO$_2$ allowed absorbing ultraviolet (UV) radiation at 200 to 240 nm wavelength, photon emission occurs at 300 to 420 nm in the form of fluorescence. The amount of fluorescence is directly proportional to the sampled SO$_2$ concentration. SO$_2$ analyzer (Echotech serinus 50) sucks sample air by 0.750 SLPM (Standard liter per minute) through the outlet. Further, passed from dust filter and hydrocarbon kicker to remove dust & hydrocarbons from sample air. Now sample air is exposed to UV radiation, which originated from the Zinc discharge tube (800 to 1100 V) in the chamber. The optical bandpass filter produces the same UV radiation at 214 nm and attributes to make photons in all directions. Another optical bandpass filter is used to allow wavelengths about 310–350 nm to photomultiplier for the measurement of the intensity of emitted radiation. The reference detector is also used for the correction of fluctuations in lamp intensity (Toth et al. 2020). The instrument is calibrated as per user manual guidelines and described by (Yousefian et al. 2020). The collected data was extracted by Airodis (software) in the 5 min resolution and further converted into a one-hour resolution for the observation period from Jul 15, 2018, to Jul 15, 2019. The operation range of SO$_2$ analyzer is from 0.3 ppb – 20 ppm and further output of data was converted into µg/m$^3$ (Tsai et al. 2000) by using the conversion factor (1 ppb = 2.62 µg/m$^3$).

The meteorological parameters were measured and recorded by Automatic Weather Station (Davis Vantage Pro2), installed at the roof of department building to avoid nearby buildings shadow and ensure zero obstacles in measurements (Ike 2014; Jenkins 2014; Sharma et al. 2018; Dutta et al. 2019). The observed data were extracted by using the weather link 2.0 (software) in a one-hour resolution. The nominal accuracy of temperature, humidity, rain, and wind direction sensors are 0.5 ºC, 2%, 0.2 mm and 3º (degree), respectively.

To identify the fire events over Uttarakhand, we have extracted the data of fire spots from Fire Information for Resource Management System (FIRMS) platform and Moderate Resolution Imaging Spectroradiometer (MODIS) data set over India (Morisette et al. 2005; Li et al. 2012; Eskandari et al. 2020).

The seasonal variation of SO$_2$ has a significant role in identifying the possible sources SO$_2$. The monsoon-M-2018 (Jul -Sep), post-monsoon–PoM-2018 (Oct-Dec), winter-W-2019 (Jan-Feb) and pre-monsoon- PrM-2019 (Mar-May) seasons were also considered for the current study (Attri et al. 2015). However, for M -2018 data was available from Jul 15, 2018 to Sep 31, 2018 and M -2019 was considered from Jun 1, 2019 to Jul 15, 2019. We have considered data sets from Jul 15, 2018, to Jul 15, 2019 for analysis and interpretation in the present manuscript.

3. Results And Discussion

3.1 General features of SO$_2$ concentration and meteorology: Brief details of background meteorological conditions and daily variation of SO$_2$ concentration in the atmosphere of SGV have represented in Fig. 2.
The SO$_2$ concentration exhibits a strong variability within the range from 1.10 μg/m$^3$ (On Jul 15, 2018) to the highest SO$_2$ concentration as 17.39 μg/m$^3$ (On May 29, 2019) and also showing an increasing trend (Upslope = 0.011 ± 7.1E-4) between this period. However, after May 29, 2019, we observed a declining trend of SO$_2$ with the downslope of -0.17 ± 0.02 until Jul 15, 2019 over SGV. The average daily SO$_2$ concentration was 5.09 ± 1.43 μg/m$^3$, which was lower than the nearest previous reported locations, i.e. Dehradun (12.84 ± 1.18 μg/m$^3$) & Haridwar (25 ± 2.51 μg/m$^3$) during the year 2009 and 2011 (Chauhan et al. 2010; Deep et al. 2019).

The temperature (Fig. 2b) shows a declining trend from Jul 15, 2018 (30.32 °C) to Dec 28, 2018 (8.78 °C) and increasing onwards up-to Jul 15, 2019 (28.52 °C). The highest daily temperature was reported on Jul 1, 2019 (34.87 °C) at SGV. Temperature also shows different correlation nature with SO$_2$ in different seasons. SO$_2$ shows significant (P-value = < 0.05) and negative correlation with the temperature in the M-2018 (r = -0.48 & p-value= 7.93E$^{-4}$) and PoM-2018 (r = -0.23 & p-value= 0.01). Whereas, W-2019 (r = 0.43 & p-value= < 0.05) and PrM-2019 (r = 0.32 & p-value= < 0.05) have positive and significant correlation with SO$_2$ at SGV (Table 1). In W-2018 low temperature supports the SO$_2$ due to less oxidation and shallow boundary layer height (Gaur et al. 2014).

The relative humidity (Fig. 2c) shows several peaks throughout the observation with a vertical bell shape type pattern. In Jul-Aug-Sep, 2018, the high humidity levels were recorded on Jul 28, 2018 (93.62%), Aug 4, 2018 (84.70%), Sep 23, 2018 (93.58%) and Sep 24, 2018 (93.04%). Whereas, on Jan 22, 2019 (87.88%), 24 Jan (77.56%), 7 Feb (81.91%), 14 Feb (88.69%), 21 Feb (88.98%) and 4 Jul 2019 (84.11%) respectively. Humidity has positive and significant correlation with SO$_2$ in the M-2018 (r = 0.26 & p-value= 0.01) & PoM-2018 (r = 0.24 & p-value= 0.10) season and negatively significant in the PrM-2019 (r = -0.33 & p-value= <0.05) during study (Table 1). (Chaudhuri and Dutta 2014) also observed the variation of pollutants (SO$_2$, NO$_2$, PM$_{10}$ & CO and O$_3$) over the Kolkata during year 2011 to 2012 and reported r= -0.03 and r = -0.09 with Humidity and temperature. However (Sharma et al. 2010) observed the a seasonal variation of SO$_2$, NO, NO$_2$, & NH$_3$ and found good correlation with temperature & humidity in the W-2008 (r= -0.92 & 0.92), PrM-2008 (r= -0.83 & 0.86) and PoM-2008 (r= -0.63 & 0.52) over Delhi during 2008.

The wind direction and wind speed have represented in the Fig. 2d & 2e. In Jul-Aug 2018 and Jul, 2019 the winds were coming from South East direction with a lower wind speed below 5 m/s. Whereas, In the Sep-Oct-Nov-Dec of 2018, Jan-Feb - early Mar-May of 2019, the air mass was originating from the southwest direction above 5 m/s. Wind speed has significant impact on SO$_2$ concentration during different season. Wind speed has a negative correlation (r= -0.29 & p-value = < 0.5) in the M-2018 and positive correlation (r= 0.28 & p-value = < 0.5) with SO$_2$ during observation. However wind direction shows weak correlation with SO$_2$ during M-2018 (r= 0.03 & p-value = 0.79), PoM-2018 (r= 0.05 & p-value = 0.75), W-2019 (r= 0.05 & p-value = 0.75), PrM-2019 (r= 0.13 & p-value = 0.21), and M-2019 (r= 0.07 & p-value = 0.62) respectively Table 1). (Gupta et al. 2004) also reported a negative correlation (r= -0.88) between SO$_2$ and wind speed during 1997 to 2000 over Mumbai. Whereas (Turalioğlu et al. 2005) also reported
negative correlation between SO$_2$ and wind speed ($r = -0.49$) along with temperature ($r = -0.75$) and humidity ($r = 0.028$) over Erzurum, Turkey. Furthermore, the simultaneous seasonal variations of wind speed, wind direction and SO$_2$ concentration have also explained in section 3.6.

3.2 Diurnal and monthly variations of SO$_2$ over SGV: In the M-2018 season (Fig 3a), the maximum SO$_2$ concentration was $3.66 \pm 2.05 \, \mu g/m^3$ at the 1900 LT and the minimum was reported of $3.01 \pm 1.31 \, \mu g/m^3$ in the morning time at 0500 LT. The SO$_2$ variation is almost constant, i.e. changed by only $0.64 \, \mu g/m^3$ with an average value of $3.39 \pm 1.64 \, \mu g/m^3$. The SO$_2$ starts fluctuating from 0000 LT and drops at 0700 LT, after that SO$_2$ was increasing up to 1200 LT may be due to local anthropogenic and photochemical activities as described in Fig. 3a (Husar and Patterson 1980). Furthermore, a dip was observed at 1400 LT due to change in traffic volume and SO$_2$ is increasing up to $6.45 \pm 3.49 \, \mu g/m^3$ at 1900 LT due to regional and long-range transport of pollutants (Gautam et al. 2018; Sandeep et al. 2020) over SGV (also explained in section 3.8). In the PoM season, The highest SO$_2$ concentration was $5.75 \pm 1.83 \, \mu g/m^3$ reported at 1300 LT which may be attributed to the effects of heavy traffic contribution on the national highway (NH) -58 (Fig. 3b). We also recorded another second peak of SO$_2$ ($5.54 \pm 2.23 \, \mu g/m^3$) at 2000 LT during the same observation period that is showing the effects of local and regional biomass burning in Punjab, Haryana and transport of pollutants (also explained in section 3.7 & 3.8). In the case of W-2019, the highest SO$_2$ concentration $6.42 \pm 1.79 \, \mu g/m^3$ has observed at 1800 LT. Similarly, we have recorded high SO$_2$ concentration at 1900 LT as well as second highest at 1300 LT due to the effect of long-range transport pollutant and photochemical production of SO$_2$ during PrM-2019 season (Fig. 3b). The identical deeps in SO$_2$ concentration were observed at 0700-0800 LT and 0500 LT during M-2018, PoM-2018, W-2019, PrM-2019 and M-2019 probably may be due to low anthropogenic activities in early morning hours at the observation site (Fig. 3a & b). In the M-2018, the SO$_2$ concentration ($3.39 \pm 1.64 \, \mu g/m^3$) was low as compare to PoM-2018 ($5.25 \pm 1.74 \, \mu g/m^3$), W-2019 ($5.71 \pm 1.56 \, \mu g/m^3$), PrM-2019 ($6.20 \pm 3.09 \, \mu g/m^3$) and M-2019 ($5.63 \pm 3.09 \, \mu g/m^3$) season. (Gaur et al. 2014) observed high seasonal concentration during W-2009 due to shallow boundary layer height and weak oxidation of sulfate and observed a similar valley shape pattern during afternoon hours in the PrM-2009 season as well as M-2009 has almost constant variation.

The monthly variation of SO$_2$ is represented by using box plots (Fig. 3c), to understand the standardized statistics and distribution. The low SO$_2$ concentrations were found in Jul 2018 ($1.07 \pm 0.82 \, \mu g/m^3$), and Aug 2018 ($3.67 \pm 0.80 \, \mu g/m^3$) may be due to the process of extensive wet deposition in M-2018 (Pandey et al. 2005; Rai et al. 2010; Gaur et al. 2014). However, in Jul 2019 the SO$_2$ concentration was high ($4.19 \pm 0.68 \, \mu g/m^3$) as compared to Jul 2018 because of fewer precipitation events and biomass burning activities (section 3.8). Whereas, the high SO$_2$ were reported in May 2019 ($7.58 \pm 3.30 \, \mu g/m^3$), followed by Jun 2019 ($6.51 \pm 2.08 \, \mu g/m^3$), Feb 2019 ($6.11 \pm 0.65 \, \mu g/m^3$) and Nov 2018 ($5.79 \pm 0.71 \, \mu g/m^3$) during the study. The mean SO$_2$ concentration during May and Jun 2019 was higher than median levels, i.e. SO$_2$ concentration has clustered at a higher level due to forest fire. The SO$_2$ levels mainly in Oct-Nov-
Dec 2018 were due to extensive biomass burning activities in the Punjab Haryana and rest of the IGP region (Mittal et al. 2009; Gadi et al. 2011). Whereas, in the case of Jan and Feb 2019, a higher level of SO$_2$ was observed due to shallow boundary layer dynamic over the monitoring site (Gaur et al. 2014; Deep et al. 2019). However, predominately high SO$_2$ concentrations have observed in May 2019 and Jun 2019 may be attributed due to extensive fire forest activities over the Uttarakhand (Jha et al. 2016; Yarragunta et al. 2020). The Central Pollution Control Board and WHO stands have also given in Table 2 and compared with other locations (Table 3).

3.3 Weekdays and Weekend analysis: Many researchers examined the variation of atmospheric pollutants on weekdays and weekends to get an idea of different pollutant sources over another part of the world’s due to change in vehicular emission (Gour et al. 2013; Filonchyk et al. 2016; Radin Mohamed et al. 2016; Özden Üzmez 2018). We have considered days from Monday to Saturday, weekend days have considered only for Sunday, and other holidays for our study for weekdays. One of the primary sources of SO$_2$ in the SGV is due to vehicular transport activities for holy pilgrims (Badrinath, Kedarnath & Hemkund sahib) and hill stations (Auli, Chopta, etc.) every year (Semwal and Upreti 2019). The weekend effect has been defined as the decline of pollutant concentration in the atmosphere due to low anthropogenic activities (Cleveland et al. 1974). The difference between weekdays and weekends clearly shows the different levels of transportation and other anthropogenic activities over SGV. In Aug 2018, Sep 2018, Feb 2019, Apr 2019 and May 2019 the weekend effect can be easily observed with the percentage change between weekdays and weekends as 42.77 %, 38.99%, 30.09%, 26.82%, 16.30%, respectively (Fig. 4). These reductions of SO$_2$ may be due to heavy rainfall, snowfall, summer vacations as well as change in traffic volume. However, some significant difference was also reported in Jul 2018 (-5.49%), Oct 2018 (-3.18%), Nov 2018 (-9.70%), Dec 2018 (-10.16%), Jan 2019 (-1.39%) Mar 2019 (-5.97%) and Jul 2019 (-27.01%) due to the significant vehicle moments for pilgrims/ hill stations (Chopta Auli, Badrinath & Kedarnath) along with firecracker activities in Diwali. In May 2019, the high mean than median indicated the presence of high SO$_2$ concentration during fire forest activity (Fig. 4). The notable weekend effect of SO$_2$ and other pollutants (O$_3$, NO$_2$, etc.) have already observed by many researchers due to change in traffic volume, industrial and transportation activities in the different part of the world (Riga-Karandinos et al. 2006; Gour et al. 2013; Özden Üzmez 2018).

3.4 Impact of Forest fire on SO$_2$: The MODIS and FIRMS data have been used to understand the daily variation of fire activities/fire count (Over Uttarakhand) and ground-based SO$_2$ (Fig. 5a). Furthermore, the significant fire days have been identified and considered as the high fire activity period (HFAP) based on the methodology suggested by Yarragunta et al. (2020). If the three-day rolling mean/ running mean exceeds the overall median values, then it has considered as HFAP. In the first HFAP (May 6 to May 14, 2019), the SO$_2$ and fire count shows a remarkable variation in the range of 4.81 to 7.95 μg/m$^3$ along with daily fire counts in the range of 8 to 98. The second HFAP (May 21 to Jun 4, 2019) shows a sharp increment in the fire count (4 to 150) and SO$_2$ (5.26 to 17.39 μg/m$^3$) with the highest values of 17.39 μg/m$^3$ on May 29, 2019, which is very close to daily WHO standards (20 μg/m$^3$ Daily average). However,
a slightly lower SO₂ concentration has observed in the third HFAP (Jun 10, 2019, to Jun 18, 2019). After, Jun 20, 2019, the SO₂ and fire count deceased may be due to wet deposition activities. The SO₂ concentration has reported 3.61 times higher than the lowest SO₂ concentration in May 2019 as well it is only 2.63 μg/m³ less than WHO standards.

### 3.5 Satellite versus ground-based SO₂

The satellite-based SO₂ data set were extracted from the National Aeronautics and Space Administration (NASA)’s Giovanni platform by using Modern-Era Retrospective analysis for Research and Applications (MEERA) -2 model between the 77 -79 E longitude and 29 -31 N latitude above SGV (Ma et al. 2020). The satellite-based SO₂ surface mass concentration (SMC) has compared with ground-based monitored SO₂ concentration (C) over SGV (Fig. 6).

A moderate (r = 0.36 to 0.67) and positive correlation (r = 0.46) was also observed between the SO₂ SMC and SO₂ C during observation (Taylor 1990; Ma et al. 2020). The variation trend matches with the Jul, Aug, Sep, Oct and Nov 2018 along with May, Jun and Jul 2019. The average SO₂ SMC & SO₂ C were 3.04 ± 0.76 μg/m³ & 5.25 ± 1.84 μg/m³ reported over the observation site. The SO₂ SMC was 1.68 times lower than SO₂ C during the study. The satellite-based SO₂ observations are very rare over Uttarakhand. (Naja et al. 2014) performed the ground-based and satellite comparison as well as model simulation of SO₂ in the nearest station over Nainital, Uttarakhand.

### 3.6 Wind dependency of SO₂

The seasonal variation of SO₂, wind speed and wind direction can be visualized by using the bivariate Polar plots (Fig. 7) in the R (open-air Package) open platform (Carslaw and Ropkins 2012; Grange et al. 2016). The SO₂ dominated in the southeast (SW) to northwest (NW) direction in the range of 5 to 6 μg/m³ with corresponding wind speed ranging from 4 m/s to 8 m/s in PoM-2018 season (Fig. 7b). The minimum SO₂ concentration has dispersed from the northeast (NE) direction below 3.5 μg/m³. However, in the case of the W-2019 season, the SO₂ was dominantly present in the South (S) to northwest (NW) direction in the range of 9.07 to 10.25 μg/m³ and low values have sprayed in the range of 3.20 to 4.37 μg/m³ at observation site (Fig. 7c).

However, in the PrM-2019 season, the SO₂ concentration was dominated in the north-east (NE) to the south-east (SE) in the range of 6.5 to 9.5 μg/m³ along with the 2 to 14 m/s. Whereas, low values of SO₂ have been reported in the south (S) to north (N) direction (Fig. 7d). Now M -2019 follows almost similar trends, but the concentration was high. (Fig. 7e). The wind speed less than 5 m/s indicates the local transport of pollutants over Nainital (Dumka et al. 2015) Therefore, SGV has been affected by the local and transportation of pollutants from different directions in different seasons. The high wind speed also supports the high SO₂ concentration over Srinagar valley possibly due to long-range transport of air mass to SGV (The origin of possible source and air mass have also been explained in section 3.8).

### 3.7 Air back mass trajectory and Cluster analysis

The seasonal air back mass trajectory (AMBT) has been extracted from Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model & Global
Data Assimilation System (GDAS) data set and further plotted to investigate the pathway of air mass (Stein et al. 2015). Seven days AMBTs have been calculated for local time 1200 LT at 500 m, 1000 m & 1500 m (AMSL) and plotted by using MeteoInfo software with TrajSet (version 1.5) plugin (Rico-Ramirez et al. 2007; Zachary et al. 2018). Fig. 8a suggested mass air arrival from the Bay of Bengal (BOB) and the Arabian ocean via IGP to Srinagar. However, at 1500 m, the air mass also contributed from the Sahara region in the M-2018.

In the case of the PoM-2018 and W-2018 seasons, the predominant air mass is coming from the Gulf region, Western Pakistan, Afghanistan, Eastern Rajasthan, Haryana and Punjab to the monitoring site (Fig. 8b & Fig 8c). But in the case of PrM-2019 season the transportation of air mass is from Iran, Iraq, western Pakistan, Rajasthan, Punjab via Haryana to Srinagar Garhwal in the as well from western Uttar Pradesh (Fig. 8d). A similar trend has been observed for the M-2019 season (Fig. 8e). This result influences the previous high SO$_2$ observation during PoM-2018, W-2019 seasons (Fig. 3 & 5). The SO$_2$ concentration at the monitoring has also been influenced by long-range transportation as well as regional emission of pollutants. Air back mass trajectory was well documented and examined (Gautam et al. 2018; Sandeep et al. 2020) also investigated the long-range and significant contribution of the pollutants from IGP to SGV.

Now to quantify the contribution of air mass we also performed cluster analysis based on the GDAS data set and TrajSet platform over the observation site (Rozwadowska et al. 2010; Sharma et al. 2016; Gautam et al. 2021). The southeasterly air mass from BOB has a remarkable contribution of 59.83% along with a contribution of 10.68 % from southeasterly air mass originating from Pakistan and the Arabian ocean to the receptor site during M-2018. However, local air mass from Punjab and Rajasthan also contributes up to 29.49% of the transportation (Fig. 9a). In the PoM-2018, the air mass has a dominant contribution of 40.43% from Central Asian and Northern African countries along with 59.56% contribution from Afghanistan side (Fig. 9b). Now, W-2019 & PrM-2019 have remarkable contributions in the range of 61.54% to 72.29% from the Central Asian, Northern African countries along with Afghanistan and Pakistan. But it should be also noticed that Punjab and Rajasthan have 20.60 % to 37.85% contribution during transportation of air mass (Fig. 9c-d). The local contribution of 31.16 % and 26.81% from BOB and Central Asia have observed during M-2019 (Fig. 9e). The air mass from the Arabian Ocean region has only a 14.79% contribution. (Gautam et al. 2021) has reported the significant contribution of air mass from the Central Asian, and neighbors countries such as Afghanistan and Pakistan through southwesterly wind over Himalayan cloud Observatory, Badshahithaul (Located 80 km from observation site).

3.8 Fire Spot over Uttarakhand and India: The MODIS and FIRMS data sets have plotted in the origin software (Serial Number GL3S4-6089-7,609,063) to represent the biomass burning activities (BBAs) over Uttarakhand (Demonstrated by green box) and India in Fig. 10 (Kharol et al. 2012; Shaik et al. 2019; Yarragunta et al. 2020). In M-2018, the BBAs are very rare over Uttarakhand and the rest part of India due to rainfall events during M-2018 (Fig. 10a). The Punjab, Haryana, Himachal Pradesh (HP) and Jammu & Kashmir (J& K) have significant BBAs and therefore, air mass helps to transport the pollutant to SGV.
during PoM-2018 (Previously explained in Fig. 8b & 9b of section 3.7). Dehradun, Haridwar and Udham Singh Nagar of Uttarakhand (Fig. 1) have also contributed to the local BBAs as depicted in Fig. 10b (Shaik et al. 2019). The impact of BBAs also reflects in the relatively high concentration SO$_2$ concentration as compare to M-2018 (Fig. 3b). In W-2019, The BBAs dominantly occur in Uttarakhand due to solid wood & coal burning for warming and cooking purpose. BBAs also contribute to local SO$_2$ emissions along with the long-range transport of pollutants (Fig. 9c & 10c). Whereas, In the PrM-2019 season (Fig. 10d), the BBAs have a predominant impact on SO$_2$ may be due to agricultural waste burning and intense forest fire in Uttarakhand, J& K, and HP (Yarragunta et al. 2020). According to Fig. 10e, BBAs have a dominant influence as compare to the M-2018, which results in the relatively high SO$_2$ concentration previously describe in Fig. 3a during M-2019. The BBAs in the PrM and PoM in the IGP also significantly contributes to the higher level of pollutant at high altitudes of the Himalayas (Kumar et al. 2011; Bhardwaj et al. 2018).

4. Conclusions

The first online measurement of SO$_2$ concentration in the ambient air of SGV was studied during 2018–2019 and provide important information regarding the seasonal variation, transport and forest fire activities of SO$_2$. This study provides a brief introduction of SO$_2$ dynamics in the valley configuration and helpful to draft future policies by the governments and non-government organizations. Based on our observations from Jul 15, 2018 to Jul 15, 2019 we conclude the following significant points.

1. During M-2018 & -2019, the maximum SO$_2$ concentrations were reported 3.66 ± 2.05 and 6.45 ± 3.49 µg/m$^3$ at the 1900 LT respectively due to transport of pollutants. Second higher SO$_2$ concentration 6.42 ± 3.36 µg/m$^3$ was reported at 1200 LT due to photochemical activities and vehicle moments during M-2019.

2. We have observed the high SO$_2$ concentration 5.54 ± 2.23 µg/m$^3$, 6.42 ± 1.798 µg/m$^3$ and 7.56 ± 3.53 µg/m$^3$ at 2000, 1800, 1900 LT may be attributed mainly due to local solid wood burning for cooking and long-range transportation of pollutants during PoM-2018 & W-2019 and PrM-2019 season. Whereas, the lowest SO$_2$ concentrations were observed at 0700, 0600, 0600, 0800 and 0500 LT during M-2018, PoM-2018, W-2019, PrM-2019 and M-2019 probably low anthropogenic activities in early morning hours.

3. The monthly box plots of SO$_2$ revels that the lower values in the Jul 2018 (1.07 ± 0.82 µg/m$^3$), Aug 2018 (3.67 ± 0.80 µg/m$^3$) and Jul 2019 (4.19 ± 0.68 µg/m$^3$) have observed due to the process of extensive wet deposition. Whereas, higher values were observed in the May 2019 (7.58 ± 3.34 µg/m$^3$) and Jun 2019 (6.51 ± 2.08 µg/m$^3$) due to extensive fire forest activities over Uttarakhand. However, The SO$_2$ concentrations were slightly higher in Jan, and Feb 2019 may be due to the shallow boundary layer dynamic over the monitoring site.

4. Temperature has different correlation nature with SO$_2$ during M-2018 (r = -0.48), PoM-2018 (r = -0.23), W-2019 (r = 0.43) and PrM-2019 (r = 0.32) respectively. In addition, Humidity also has
significant correlation in M-2018 \( (r = 0.26) \), PoM \( (r = 0.24) \) and PrM-2019 \( (r = -0.33) \) during study. Whereas wind speed has a positive correlation \( (r = 0.28) \) in PrM-2019 and negative correlation \( (r = -0.29) \) in the M-2018 season. However, wind direction shows weak correlation with SO2 throughout observation.

5. The weekend effect was dominating in Aug 2018, Sep 2018, Feb 2019, Apr 2019 and May 2019 with the percentage change as 42.77 %, 38.99%, 30.09%, 26.82%, 16.30%, respectively due to change in traffic volume, vacations & rainfall activities. But some other months show the different behavior in Jul 2018 (-5.49%), Oct 2018 (-3.18%), Nov 2018 (-9.70%), Dec 2018 (-10.16%), Jan 2019 (-1.39%) Mar 2019 (-5.97%) and Jul 2019 (-27.01%) due to the significant vehicle moments for pilgrims/hill stations and firecracker activities (Diwali).

6. The second HFAP demonstrates a similar trend of variation between fire count (4 to 150) and SO2 (5.26 to 17.39 µg/m³) and SO2 approaches to its highest concentration of 17.39 µg/m³ (May 29, 2019). The Fire count and SO2 show a strong correlation of \( r = 0.61 \) during observation. A moderate correlation between the satellite-derived SO2 CMD and ground-based SO2 C \( (r = 0.60) \) was observed along with a much similar trend.

7. The AMBT analysis suggests domination of air mass from Gulf region, Sahara Desert, western Pakistan, Afghanistan, and Eastern Rajasthan via Punjab and Haryana and from IGP, BOB & Arabic Ocean during different seasons to influence the pollutant concentration over the monitoring site. Westerly winds from Central Asia, northern African countries with Afghanistan & Pakistan dominate during PoM-2018, W-2019 & PrM-2019 and have a contribution of 40.43%, 61.54% and 72.29% respectively. Whereas, In M-2018 & M-2019 show contribution of 59.83% & 26.81% from BOB region.

8. The daily SO2 concentration (1.07 to 17.39 µg/m³) was under the World Health Organization (WHO) and National Ambient Air Quality Standards (NAAQS). The daily average SO2 concentration was 5.25 ± 1.84 µg/m³ reported, which was approximately 2 to 5 times lower than nearest reported locations, i.e. Dehradun & Haridwar during the year 2011 and 2009.

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Tables

Due to technical limitations, tables are only available as a download in the Supplemental Files section.

Figures
The topography map of the monitoring site around the Department of Physics Hemvati Nandan Bahuguna Garhwal University (A Central University), Srinagar Uttarakhand (India). The observational site is located at the 30º13’36 N (Longitude), 78º48’14 E (latitude) and the elevation of 640 m AMSL. Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.
Figure 2

The daily time series of (a) Sulphur dioxide (SO2) concentration, (b) temperature, (c) humidity, (d) wind direction, (e) wind speed recorded over Srinagar Garhwal Valley.
Figure 3

Diurnal variation of SO2 in the (a) monsoon - 2018 (July 15, 2018 to September 31, 2018) & monsoon - 2019 (June 1, 2019 to July 15, 2019), (b) post-monsoon (October 1, 2018 to December 31, 2019), winter (January 1, 2019 to February 28, 2019) and pre-monsoon season (March 1, 2019 to May 31, 2019), and (c) monthly variation of SO2 during July 15, 2018 to July 15, 2019 at Srinagar Garhwal valley.
Figure 4

Weekdays (Monday to Saturday) and weekend (Sunday and other holidays) variation of SO2 over Srinagar Garhwal valley from July 15, 2018 to July 15, 2019.
Figure 5

Time series of (a) SO2, fire count (Over Uttarakhand) during forest fire events over Srinagar Garhwal Valley.
Figure 6

Monthly variation of ground-based SO2, satellite-derived SO2 surface mass concentration SO2 over Srinagar Garhwal Valley.
Figure 7

Wind dependency of SO2 concentration during (a) monsoon -2018, (b) post-monsoon season, (c) winter, (d) pre-monsoon and (e) monsoon -2019 over Srinagar Garhwal Valley.
Figure 8

Air mass back trajectories during monsoon season - 2018 (a), post-monsoon (b), winter season (c), pre-monsoon season (d) and monsoon - 2019 (e) over Srinagar Garhwal Valley. Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or
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Figure 9
Cluster analysis of air mass during monsoon season - 2018 (a), post-monsoon (b), winter season (c), pre-monsoon season (d) and monsoon - 2019 (e) over Srinagar Garhwal Valley. Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion
whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

Figure 10

MODIS derived Fire spot / activities during (a) Monsoon 2018 (Jul 15, 2018 Aug & Sept 2018), (b) Post-monsoon (Oct, Nov & Dec 2018), (c) Winter (Jan & Feb 2019), (d) Pre-monsoon (Mar, Apr, & May 2019) and (e) Monsoon 2019 (Jun & Jul 15, 2019) over India. A green box represents the fire spot over Uttarakhand. Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

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