Chemosphere 316 (2023) 137878

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

First insight into seasonal variability of urban air quality of northern Pakistan: An emerging issue associated with health risks in Karakoram-Hindukush-Himalaya region

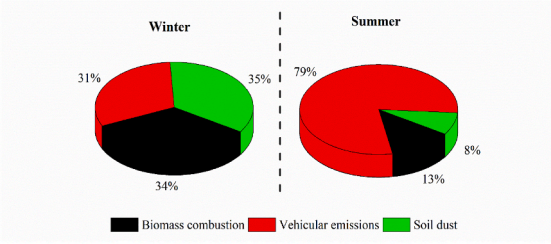
Nasir Hussain [a,b,1], Masroor Ahmad [a,1], Hassaan Sipra [c], Shuakat Ali [a], Jabir Hussain Syed [d,e,*], Khadim Hussain [b], Syed Waqar Hassan [b]

[a] Department of Environmental Sciences, Karakorum International University, Gilgit, Pakistan
[b] Gilgit-Baltistan Environmental Protection Agency (GB-EPA), Gilgit, Pakistan
[c] Centre for Climate Research and Development, COMSATS University Islamabad, Park Road Tarlai Kalan, 45550, Islamabad, Pakistan
[d] Department of Meteorology, COMSATS University Islamabad, Park Road Tarlai Kalan, 45550, Islamabad, Pakistan
[e] Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, 11 Yuk Choi Rd, Hung Hom, Hong Kong

HIGHLIGHTS

- First baseline urban air quality results from Gilgit city in northern Pakistan.
- Our findings depict $PM_{2.5}$ health crossed national and WHO daily permissible limits.
- Air Pollutant levels showed a downward winter-summer trends.
- Source apportionment showed the role of biomass burning and vehicular emissions.
- Weak regulatory framework seeks urgent attention to improve air quality of HKH region.

GRAPHICAL ABSTRACT



Source Apportionment of Atmospheric Emissions in Karakoram-Hindukush-Himalaya Region, Northern Pakistan

ARTICLE INFO

Handling Editor: Volker Matthias

Keywords:
Air quality
$PM_{2.5}$
Health risks
Biomass burning
Vehicular emissions
HKH region

ABSTRACT

There is a dire need of air quality monitoring in the high-mountain areas of Karakoram-Hindu Kush-Himalaya (HKH) region, particularly related to the recent activities undergoing the China-Pakistan Economic Corridor (CPEC). This study presents the first baseline monitoring and evaluation findings from Gilgit city, Gilgit-Baltistan. Hourly data collection for air quality parameters ($PM_{2.5}$, NO, NO_2, SO_2, O_3 and CO) were measured using air-pointer (recordum, Austria) from 1 Jan 2018 to 31 Mar 2018 (winter) and 1 Jun 2018 to 31 Aug 2018 (summer). Our findings depict $PM_{2.5}$ health limits were crossed in the winter season, while NO, NO_2 and SO_2 remained below their health limits. O_3 and CO showed a rising trend in summer months, crossing the 8-h health limits during the season. Seasonal correlation in meteorology found an inverse relationship between most parameters and temperatures; reverse was true for O_3 and CO. In parallel, thermal optical carbon analysis filter-based sampling characterized air quality into mass concentrations of $PM_{2.5}$, organic carbon (OC), elemental carbon (EC) and various heavy metals. Filter-based $PM_{2.5}$ correlated well with analyzer-based $PM_{2.5}$ for all months that were studied, except February and March 2018. $PM_{2.5}$, OC and EC were higher in summer as compared to winter, whereas higher heavy metal contributions were measured predominantly during summer.

* Corresponding author. Department of Meteorology, COMSATS University Islamabad, Park Road Tarlai Kalan, 45550, Islamabad, Pakistan.
E-mail address: jabir.syed@comsats.edu.pk (J.H. Syed).
[1] Equally contributed.

https://doi.org/10.1016/j.chemosphere.2023.137878
Received 17 October 2022; Received in revised form 30 December 2022; Accepted 13 January 2023
Available online 13 January 2023

Health impacts were found to be above health limits for Ni in children only. Furthermore, principal component analysis-multiple linear regression (PCA-MLR) technique was applied to determine source apportionment, confirming the role of biomass burning in winters, and vehicular emissions in summers, highlighting the need for flexible monitoring of technologies/approaches, and communications among the various public, private agencies, and all relevant stakeholders.

1. Introduction

Growing evidence in the recent literature points to rapid deterioration of air quality in Pakistan (Ghauri et al., 2007; Colbeck et al., 2010; Jabeen and Khokhar, 2019; Hashmi et al., 2020; Tabinda et al., 2020; Anjum et al., 2021). This is of concern for policy makers, the private sector and civil society, with reference to urban planning, public health, energy generation, transportation fuel standards, industrial emissions, and regional (transboundary) cooperation (Raza et al., 2021). Air pollution implications in Pakistan are growing – many studies focus on health, weak regulatory framework, and low investments in alternatives, but also on the urban environments of Pakistan, or rural indoor air pollution (Parek et al., 2001; Waheed et al., 2009; Ali and Athar, 2010; Majid et al., 2012; Rasheed et al., 2014; Shi et al., 2020). There exists a significant gap in the literature with respect to the role of ambient air quality monitoring and enforcement, particularly in the northern, high mountain areas of the country.

The documented impacts of air pollution in the Karakoram, Hindu Kush, and Himalayan (HKH) ranges, which extend into Pakistan, relate to rising radiative forcing and accelerated snowmelt in the region, at the atmospheric scale (Lau et al., 2010). Pakistan is responding to this challenge through adaptation measures under the Green Climate Fund (GCF) sponsored by the Glacier Lake Outburst Flooding Project (Green Climate Fund, 2016). However, within the funding proposal, no mention of air quality and its role in increasing glacial melt is found. Primarily, air pollution studies conducted at various sites across the HKH in India and Nepal, reveal concentrations of air pollutants higher than the established national and international air quality standards (Gajananda et al., 2005; Panwar et al., 2013; Kumar et al., 2020). Pakistan's Ministry of Environment (now Ministry of Climate Change) made available a set of National Environmental Quality Standards (NEQS) for ambient air, which place limits on hourly basis, 8-h, daily, and annual averages, depending on the type of pollutant (Pak-EPA, 2009). The World Health Organization (WHO) has its own limits, which are more stringent than Pakistan's, based on health impacts (WHO, 2018). Other studies point to the role of meteorological parameters, mainly temperature and wind speed, in shaping air pollution in the South Asian region, observed as increasing wintertime smog, increasing summertime pollution levels, and changes in diurnal cycles (Adak et al., 2014; Bhat et al., 2017; Nasir et al., 2019). However, such studies focus on black carbon, satellite analysis and aerosol transport models; few in-situ results are available across the region.

Local air pollution presents challenges in the north and share characteristics with urban and rural environments of Pakistan, with mitigation potential that produce co-benefits for health and local environment. Increasing tourism, trade and investment in the region are causing urbanization, and raising emissions from vehicles, waste generation/burning and wood fuel use in Northern Pakistan. Implementation of sound policy interventions requires an assessment of the current local air quality scenario. This is important as the area will be drastically altered by Pakistan's largest ever infrastructure investment plan, the $50 billion China-Pakistan Economic Corridor (CPEC), part of the Belt and Road Initiative. The corridor connects three highway alignments that span across all of Pakistan and fall into a singular transition highway between Pakistan and China, via the GB region. An estimated 7000 trucks are expected to use this road on a weekly basis (Li et al., 2021; Awais et al., 2019). Substandard fuel consumption, agriculture practices, inefficient management of waste have been recognized as potential sources of atmospheric aerosols in Pakistan and over northern India, particularly with reference to seasonality (Rengarajan et al., 2007; Ram et al., 2012; Venkataraman et al., 2018). It is important to conduct source apportionment studies to provide context to policy and decision makers with reference to actions they may take to mitigate air pollution and its health and environmental impacts. Limited source apportionment studies are undertaken in Pakistan, with none being conducted in the northern areas on air quality. The composition and characterization of atmospheric particles contribution to the formation of winter – time haze layer, is still unspecified. A baseline assessment of the ambient air quality in Gilgit City will build a context for interventions that align with Pakistan's CPEC obligations and national commitments to sustainable development goals (SDGs) and the Paris Agreement through Nationally Determined Contributions. In-situ analysis of ambient air quality of Gilgit City remains missing, with only a geospatial study on air quality thus far (Hussain et al., 2014).

Climate implications of atmospheric pollutants including trace gases and aerosol ($PM_{2.5}$) emissions in Northern areas of Pakistan are considered of regional importance, but the dynamics of the system are not well understood with lacking research studies and monitoring of atmospheric pollutants (Cong et al., 2010; Kumar and Attri, 2016; Tripathee et al., 2017; Kaushal et al., 2018). There continue to be questions about the magnitude, its contribution, and the source provenance of the atmospheric emissions from urban centers in Northern Pakistan. Although understanding the process involved in atmospheric pollution is a main research area in atmospheric science and has been investigated widely in the past decades throughout the world, there are still gaps in knowledge of actual production and scientific estimation of particulate matter in the environment of Pakistan. An in-depth understanding of the emissions and their formation process under different environmental conditions is urgently needed. Therefore, this study has been designed to mainly focus on field measurements of $PM_{2.5}$, its characterization and trace gases, their seasonal behavior and to understand the role of sources and meteorology in the formation and chemistry of air pollutants in the study region. The results from the field campaign were inter-compared to improve our understanding of the regional impact of urban pollution under diverse pollution and weather conditions in different parts of this region. To our knowledge, this is the first study of its kind among the fast-growing urban center in the Northern Pakistan.

2. Materials and methods

2.1. Description of the study area

Gilgit City (35°55′7.28″N, 74°18′44.82″E), the capital of the autonomous Gilgit-Baltistan territory (GB region), with a population of 216,760 (Gilgit Baltistan Scouts, 2021), is the first major city of Pakistan on the CPEC route, along the Chinese border (Figure S1). The city is located in a broad valley near the confluence of the Gilgit River and Hunza River. Gilgit is a major tourist destination in Pakistan, and serves as a hub for trekking and mountaineering expeditions in the Karakoram mountain range. The city is projected to expand in size, given its proximity to the planned Moqpondass Special Economic Zone being established (35 km away) in Gilgit-Baltistan, where high emission industries (marble/granite, iron ore, steel, mineral and leather) are to be installed (CPEC, 2021). Being a mountainous region, the weather conditions in Gilgit are cold during the winter season (−2.3 °C), consisting of eight to nine months per year. Average rainfall is 120–240 mm yearly, low due to

its high altitude. Despite lacking rainfall, the region has plenty of water for irrigation via rivers and springs, abundant with melting snow water from high altitude glaciers. Summer is hot and short, with minimal intense sunny days where temperatures reach 40 °C. The city has more than 11,000 registered vehicles, making vehicular traffic emissions an important concern for urban planning. Burning of biomass, wood and vehicular emissions are considered the source of a constant winter haze layer in the atmosphere throughout the winter season (GB-EPA, 2012; GB-EPA, 2013; GB-EPA, 2019).

2.2. Air sampling

2.2.1. Real-time measurements

Air quality measurements for ambient air quality parameters ($PM_{2.5}$, NO_x, CO, O_3 and SO_x) were taken on an hourly basis at the Gilgit City Environment Protection Agency's (EPA) office rooftop (Figure S1) from 1 Jan 2018 to 31 Mar 2018 (winter) and 1 Jun 2018 to 31 Aug 2018 (summer), using analyzer (Airpointer; recordum, Austria). The building's surroundings comprise of houses, institutions, and commercial places within a 5-km radius. Missing data on an hourly time scale for air pollutants resulted from electrical outages and other technical issues at the sampling site. Out of a total of 4368 h in the study months, 3923 hourly measurements were taken, with a completion rate for each month: January (88%), February (100%), March (94%), June (69%), July (90%) and August (99%). The hourly measurements were converted to daily averages and were considered missing if more than a third of the day was missing.

2.2.2. Filter-based $PM_{2.5}$ measurements

Particulate matter ($PM_{2.5}$) samples were collected using High Volume-Active Air Sampler (Guangzhou Mingye Huanbao Technology Company) equipped with a $PM_{2.5}$ inlet. All samples were collected on the same roof top as the analyzer method (~4 m height). Air sampling was performed with ~24 h intervals at a flow of ~18 m^3/h. Quartz fiber filters (20 × 25 cm^2, 2600 QAT, PALL, USA; prebaked (450 °C, 6 h, muffle furnace), was used to collect the particulate phase samples. A total of 32 filters were collected in the winter and 22 in the summer of 2018. Field blanks (6 filters; one for each sampling month) were also assembled in the active sampler (Turned-OFF mode) at the sampling site, and then disassembled after approximately 5 min. Samples were refrigerated at −20 °C until analysis. Samples were conditioned at 22 ± 1 °C in relative humidity of 35 ± 2% for 24 h before and after each sampling, and then weighed using an electronic gravimetric balance with a detection limit of 1 μg (Sartorius, Göttingen, Germany). The corresponding $PM_{2.5}$ mass concentration of each filter is presented in Equation (1);

$$\begin{aligned} PM_{2.5}\ \text{mass concentration} = {} & \text{Weight after sampling (W2)} \\ & - \text{Weight before sampling (W1)} / \text{Sampling Volume} \end{aligned} \quad (1)$$

More details on OC/EC analysis, heavy metals analysis, methodology of calculating health risk assessment and QC/QA summary for heavy metal analysis have been given in the Supplementary Information (S1–S4).

2.3. Meteorological parameters

Hourly meteorological parameters (temperature, relative humidity, wind direction and wind speed) were provided by the Pakistan Meteorological Department (PMD) (Gilgit office) for the sampling period. The data had a 100% completion rate.

2.4. Statistical analysis

Basic statistical analysis techniques illustrate the extent of air pollution from a daily to monthly time scale, alongside a comparison of diurnal winter and summer concentrations. Hourly data was extracted from the Air-pointer via the recordum dashboard in. csv format; the data was cleaned and processed using Microsoft Excel before correlation analysis was conducted using R x64 3.5.0 software. Origin 9.0 software was used for data analysis and graphing. Filter-based data was analyzed in Microsoft Excel. Source apportionment analysis were done through Principal Component Analysis combined with Multiple Linear Regression (PCA-MLR) and details are provided in the Supplementary Information (S5).

3. Results and discussion

3.1. Comments on seasonal trends

3.1.1. $PM_{2.5}$

In general, the winter $PM_{2.5}$ concentrations were higher than the summer season. For analyzer method (Fig. 2), maximum daily average $PM_{2.5}$ limit set by Pakistan (35.0 μg m^{-3}) was exceeded, 91% of days recorded for January, followed by 30% of days in February, all of which occurred in the first third of the month. No days exceeded the $PM_{2.5}$ limit in the remaining study months, including summer. Consequently, Fig. 1 also displays the sharp downward trend of daily average $PM_{2.5}$ concentrations from January (32.9 μg m^{-3} – 74.7 μg m^{-3}) to March (7.50 μg m^{-3} – 23.8 μg m^{-3}); it is in contrast with the rising $PM_{2.5}$ concentrations from June (5.00 μg m^{-3} – 17.4 μg m^{-3}) to August (6.80 μg m^{-3} – 21.3 μg m^{-3}), albeit slower. Against the WHO maximum daily average $PM_{2.5}$ limit (25.0 μg m^{-3}), the number of days exceeding the limit rose to 100% in January and 67% in February, with no change in the remaining study months. Variability (as a measure of standard deviation) was higher in winter (16.7 μg m^{-3}) as compared to summer (3.20 μg m^{-3}). Variability reduced for each consecutive month from January (15.4 μg m^{-3}) to March (4.10 μg m^{-3}), while variability declined from June (3.70 μg m^{-3}) to July (2.70 μg m^{-3}) and rose in August (3.10 μg m^{-3}). No relationship was found between daily $PM_{2.5}$ concentrations and weekdays versus weekend days.

Similarly, daily averaged $PM_{2.5}$ concentrations for the filter-based method ranged from 6.69 μg m^{-3} to 98.1 μg m^{-3}, as shown in Fig. 2. $PM_{2.5}$ concentrations were higher in winter (51.8 μg m^{-3}) as compared to summer (12.5 μg m^{-3}), for the filter-based analysis as well. However, for filter-based analysis, $PM_{2.5}$ concentrations were highest in March (63.0 μg m^{-3}), then January (48.2 μg m^{-3}) and February (45.2 μg m^{-3}). Further, significant differences arose in the $PM_{2.5}$ concentrations between the filter-based and analyzer methods in February and March (CL> 95%). The summer months showed less variability, with highest concentrations in June (13.9 μg m^{-3}), then August (12.1 μg m^{-3}) and July (11.6 μg m^{-3}). Filter paper $PM_{2.5}$ concentrations exceed the health limit set by Pakistan (WHO) in 81% (94%) of monitored days in the winter. All March days monitored were above either limit. No days monitored during summer were above either limit.

3.1.2. Trace gases

Daily averaged concentrations of analyzer-based NO, NO_2 and SO_2, seen in Fig. 1, did not exceed the national limits (40.0 μg m^{-3}, 80.0 μg m^{-3} and 120 μg m^{-3}, respectively); all showed a decreasing trend from January to March. NO and NO_2 showed an increasing trend from June to August, while SO_2 continued its downward trend, with two spikes noted in June. Although overall variability for NO was similar for winter and summer at 5.70 μg m^{-3}, highest variability was observed in June (6.70 μg m^{-3}), followed by July (6.10 μg m^{-3}), then February (5.00 μg m^{-3}), January (4.70 μg m^{-3}), August (4.70 μg m^{-3}) and March (4.00 μg m^{-3}). Similar ordering of variability was observed for NO_2. For SO_2, variability in winter (4.30 μg m^{-3}) was higher than in summer (2.30 μg m^{-3}).

In addition, winter and summer 1-h O_3 concentrations recorded were below the limits set by Pakistan (130 μg m^{-3}), with a larger concentration range in the summer (128 μg m^{-3}), as compared to winter (80.4

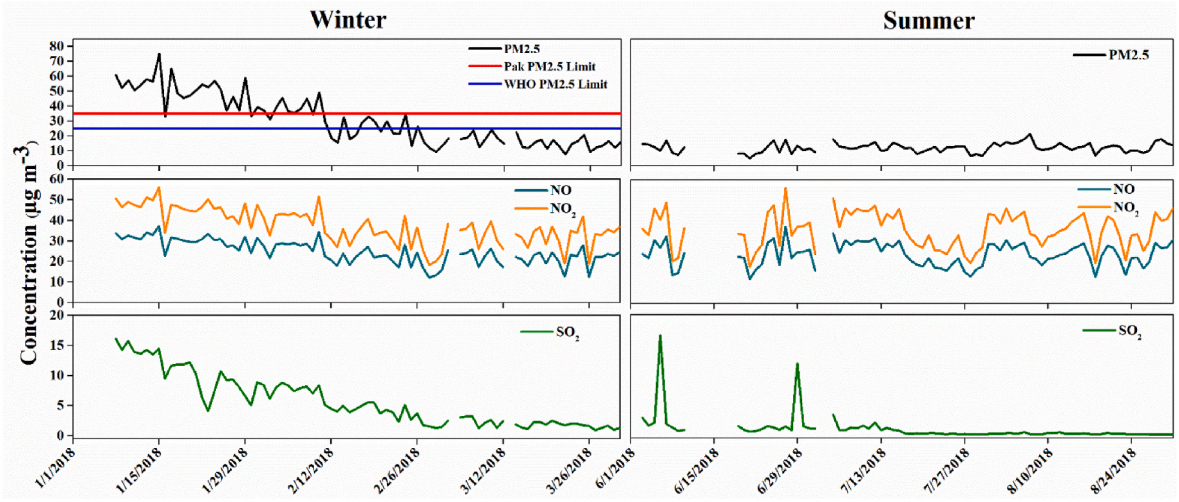


Fig. 1. Daily mean $PM_{2.5}$, NO, NO_2 and SO_2 concentrations from 1 January to 31 March 2018 (top) and 1 June to 31 August 2018 (bottom).

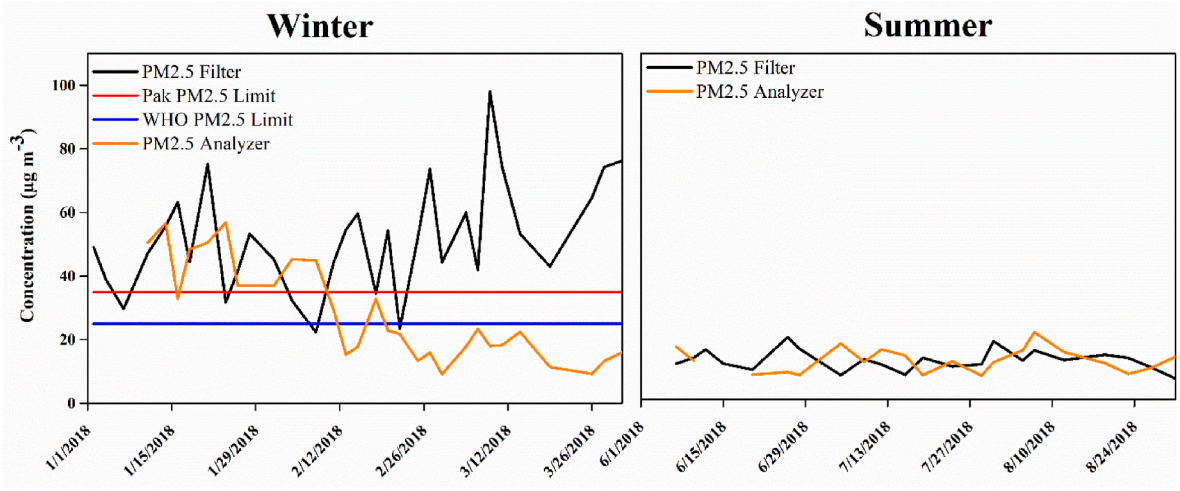


Fig. 2. Time series of $PM_{2.5}$ filter and analyzer results for common dates.

μg m^{-3}). Summer 1-h O_3 concentrations were higher in 21% of hours, against the maximum 1-h O_3 concentrations in winter. Although Pakistan provides no standards for 8-h mean O_3 concentrations, given the strong link between lower ozone concentrations and mortality (Huang et al., 2019), an assessment against WHO's recommended limit of 100 μg m^{-3} is made (Fig. 3). Only 14 h in June and 18 h in July were above the WHO limit. A rising trend was noted from January to March for 8-h mean O_3 concentrations, but in summer, no trend is observed.

Ground level O_3 is a secondary pollutant resulting from chemical reactions between NO_x and volatile organic compounds (VOCs), as well as CO (Kovač-Andrić et al., 2013; Melkonyan and Kuttler, 2012; Anjum et al., 2021). Winter and summer both presented a rising trend for 1-h CO concentrations, with only two instances in January, and one instance in February, March, and June each, where 1-h CO concentrations exceeded the limit set by Pakistan (10 mg m^{-3}). Hourly CO concentrations exceeded the hourly limit in 1.5% and 4.3% of the hours recorded for July and August, respectively. Hourly CO concentrations were lower in winter as compared to summer, ranging from 0.00 mg m^{-3} to 14.7 mg m^{-3} in January and from 1.65 mg m^{-3} to 13.0 mg m^{-3} in March. In June, the hourly CO concentrations ranged from 4.30 mg m^{-3} to 10.4 mg m^{-3} and from 5.70 mg m^{-3} to 12.4 mg m^{-3} in August. 8-hour mean CO concentrations (Fig. 3) exceeded the limit set by Pakistan (5.00 mg m^{-3}) in a more pronounced manner, while retaining the trends presented for 1-h CO concentrations. The 8-h mean CO limit was exceeded in January (by 3.3% of the hours recorded), February (1.2%), March (2.0%), June (78.3%), July (97.3%) and August (100%).

3.1.3. Carbon species

OC concentrations ranged from 1.70 μg m^{-3} to 23.4 μg m^{-3} and were higher in winter as compared to summer (Figure S2). OC concentrations displayed similar characteristics to $PM_{2.5}$ concentrations in the winter months. Daily averaged OC concentrations were highest in March (15.0 μg m^{-3}), then January (14.1 μg m^{-3}) and February (12.0 μg m^{-3}). In summer, OC concentrations were highest in June (8.19 μg m^{-3}), then August (5.07 μg m^{-3}) and July (3.67 μg m^{-3}). EC concentrations were lower than OC concentrations throughout the study period. EC concentrations ranged from 0.01 μg m^{-3} to 2.01 μg m^{-3} and were higher in winter as compared to summer, with daily averaged concentrations highest in March (1.23 μg m^{-3}), then February (1.22 μg m^{-3}) and January (1.06 μg m^{-3}). In summer, EC concentrations were highest in August (0.89 μg m^{-3}), then June (0.56 μg m^{-3}) and July (0.20 μg m^{-3}).

The OC/EC ratio presented a range from 1.77 μg m^{-3} to 163 μg m^{-3}. OC/EC ratio was higher in summer as compared to winter, with highest ratio calculated for June (51.8 μg m^{-3}), then July (37.0 μg m^{-3}). The high standard deviation for June and July was indicative of the few instances in both months where the EC concentrations dropped to nearly zero. The lowest OC/EC ratio for all study months was in August (7.39 μg m^{-3}). In winter, January (14.3 μg m^{-3}) presented the highest OC/EC

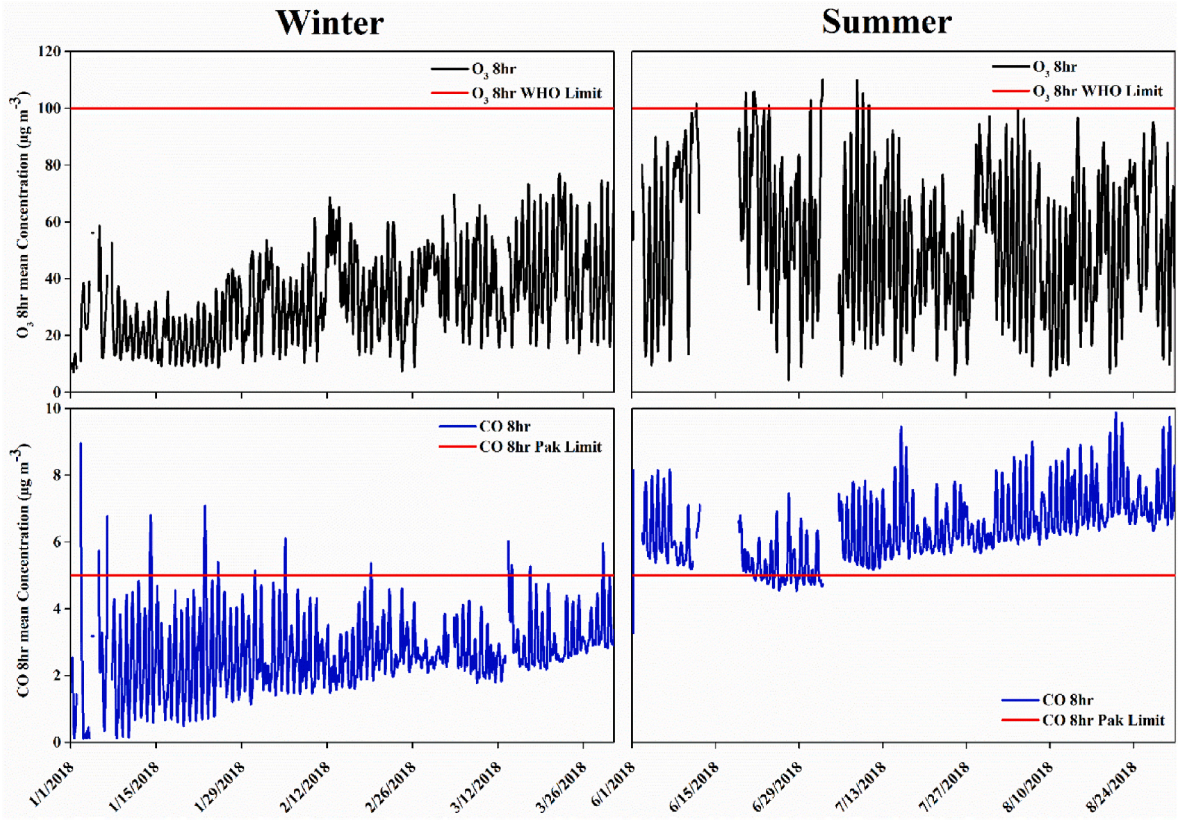


Fig. 3. 8-h mean O_3 and CO concentrations for the study months.

ratio, followed by March (13.0 µg m^{-3}) and February (9.87 µg m^{-3}). The high averages for June and July stemmed from six consecutive monitoring days with an average of 91.3 µg m^{-3}).

The OC/EC ratio determines the strength of the source emissions, with values larger than 5 indicating dominance of biomass burning, and values of less than or equal to 2 indicating fossil fuel combustion (Rastogi et al., 2016). The high values of the OC/EC ratio in summer were indicative of increasing fossil fuel combustion, which was validated by the increasing emissions of CO, the result of increasing vehicular emissions in that season.

3.1.4. Heavy metals

Filter-based analysis revealed mass concentrations for 8 heavy metals, with high variability. The daily time series (Figure S3) presented extremely low mass concentrations for Cd, Cr, Cu, and Mn, only in January. Percentage-wise appearance for these low mass concentrations were as follows: Cd (0.1%), Cr (1.5%), Cu (2.4%) and Mn (8.4%). The remaining heavy metals (Ni, Pb, Zn and Fe) displayed higher mass concentration and variability, with Fe dominating across all study months. Ni, Pb and Zn had highest contributions in February (8.0%, 16.9%, and 22.8%, respectively). Ni contributions in January were second highest (3.8%), while Pb, Zn and Fe had second highest contributions in March (13.0%, 17.8% and 68.3%, respectively). Ni contributed well below Pb, Zn and Fe for June (0.4%), July (0.2%) and August (0.4%). For Pb, Zn, July was the summer month with the largest contributions (11.8% and 10.1%, respectively). Pb and Fe contributions in June (4.5% and 88.9%, respectively) were higher than in August (2.7% and 87.9%, respectively), while the reverse was true for Zn (June – 6.2% and August – 9.0%). As shown in Fig. 4, mass concentrations averaged by season were higher in summer as compared to winter, with a difference in increasing order for Ni (0.03 µg m^{-3}), Zn (0.97 µg m^{-3}), and Fe (35.6 µg m^{-3}). Mass concentrations were higher in winter as compared to summer for Pb, with a difference of 0.69 µg m^{-3}.

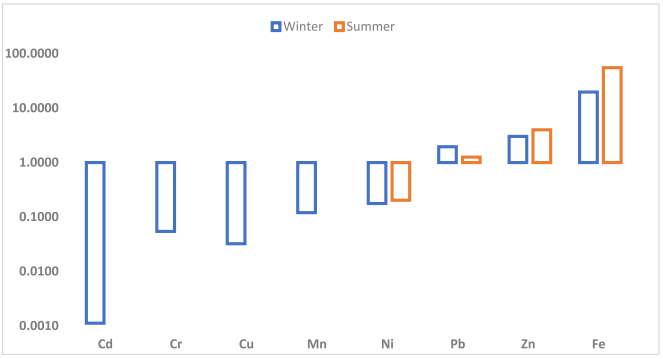


Fig. 4. Comparative analysis of heavy metals concentrations by season.

3.1.5. Diurnal variations

Hourly concentrations averaged over winter and summer for all air pollutants are presented in Figure S4, as box-and-whisker plots. Winter concentrations were higher (lower) for $PM_{2.5}$, NO_x, and SO_2 (CO and O_3) concentrations. In winter, $PM_{2.5}$, NO_x, CO and SO_2 share similar diurnal characteristics, with a rise, peak and fall in concentrations occurring from 14:00 to 17:00, indicating increasing activity in the afternoon, due to the colder temperatures in the winter mornings and evenings (Rasheed et al., 2014; Shrestra et al., 2016; Zhao et al., 2016). A second rise in concentrations occurs from 20:00 to its 01:00 peak across $PM_{2.5}$, NO_x, CO and SO_2, indicating a stronger role for evening activities in increasing pollutant concentrations. Concentrations for these pollutants declined from 02:00 to 07:00 and remained low from 08:00 to 13:00. In winter, diurnal O_3 concentrations presented inversely to the other pollutants, rising, peaking, and falling from 07:00 to 14:00, and repeating with higher variability from 15:00 to 03:00. In summer, $PM_{2.5}$, NO_x, CO and SO_2 peaked at 22:00, with higher variability observed from 21:00 to

01:00. These pollutant concentrations showed lower variability in the remaining hours, with highest median values observed at 08:00. Whereas in winter, the highest observed median values were found at 0:00, and in summer the highest observed median values were found at 22:00, indicating earlier cession in air pollutant generating activities. Diurnal O_3 concentrations in summer presented larger interquartile ranges across all hours, indicating increasing role of sunlight and higher temperatures in O_3 formation (Wang et al., 2017).

A comparison of the quantile bias ($Bias_{quantile}$ = $Winter_{quantile}$ − $Summer_{quantile}$) at each hour revealed that all SO_2 quantile biases were positive. $PM_{2.5}$ produced negative quantile bias in all quantiles from 07:00 to 10:00, with other negative quantile biases noted at 11:00, 12:00 and from 20:00 to 22:00. The bias is negative for NO_x across more hours and quantiles as compared to $PM_{2.5}$. From 07:00 to 12:00, all quantiles were negatively biased, and the interquartile range was negatively biased from 18:00 to 22:00. In the case of CO, 99% of quantiles reported negative bias, given the higher summer CO concentrations. For O_3, 71% of all quantile-hour combinations reported negative bias. The 75th and 100th quantile reported negative bias across all hours, while the interquartile range reported negative bias 01:00 to 06:00 and from 09:00 to 17:00.

3.2. $PM_{2.5}$ and US air quality index (USAQI)

As an additional analysis, $PM_{2.5}$ is considered for health purposes at the hourly level. While the USAQI standard is applicable to the daily mean, it was applied to the hourly $PM_{2.5}$ concentrations (Figure S5) to elucidate intensity and exposure potential by day. It revealed high intraday and week-on-week variability. In line with daily analysis, winter presented a higher proportion of values above 55.5 μg m^{-3} (Unhealthy, Very Unhealthy and Hazardous) whereas in summer, a higher proportion of values categorized as between 0 μg m^{-3} and 55.5 μg m^{-3} (Good, Moderate and Unhealthy for Sensitive Groups). January showed Unhealthy $PM_{2.5}$ concentrations between the hours of 13:00 to 17:00 and 20:00 to 04:00, with few Very Unhealthy $PM_{2.5}$ concentrations recorded at 15:00. Varying degrees of less impactful $PM_{2.5}$ concentrations were found in the remaining hours. By March, fewer hours recorded Unhealthy $PM_{2.5}$, with Good $PM_{2.5}$ consistently recorded from 17:00 to 00:00, indicating a reduction in evening pollutant related activity over the winter months. Biomass burning remains the primary source of cooking and heating in much of Northern Pakistan and may account for the declining trend in winter $PM_{2.5}$ concentrations. In summer, June recorded the highest proportion of Good $PM_{2.5}$ (70%) of any study month, followed by Moderate (28%) and Unhealthy for Sensitive Groups (2%), both of which clustered between 01:00 and 07:00. Moderate $PM_{2.5}$ concentrations as a proportion of the total increased in July (31%) and August (40%), largely between 20:00 and 01:00.

3.3. Relationship with meteorological parameters

The relationship of daily mean air pollutant concentrations and meteorological parameters is explored through frequency distribution and correlation analysis (Fig. 5). $PM_{2.5}$ and SO_2 concentrations are right-skewed with highest frequency at 10–20 μg m^{-3} and 0–2.5 μg m^{-3}. NOx (75–80 μg m^{-3}) and O_3 (40–50 μg m^{-3}) are less right-skewed. CO presented right-skewed bimodal distribution, while Temp and RH presented left-skewed bimodal distributions, indicating seasonal change. WS presented left-skewed unimodal distribution; therefore, wind speed is rarely beyond 0.5 knots. $PM_{2.5}$ is significantly correlated with all air pollutants, with highest positive correlations with NO_x and SO_2, 0.82 for

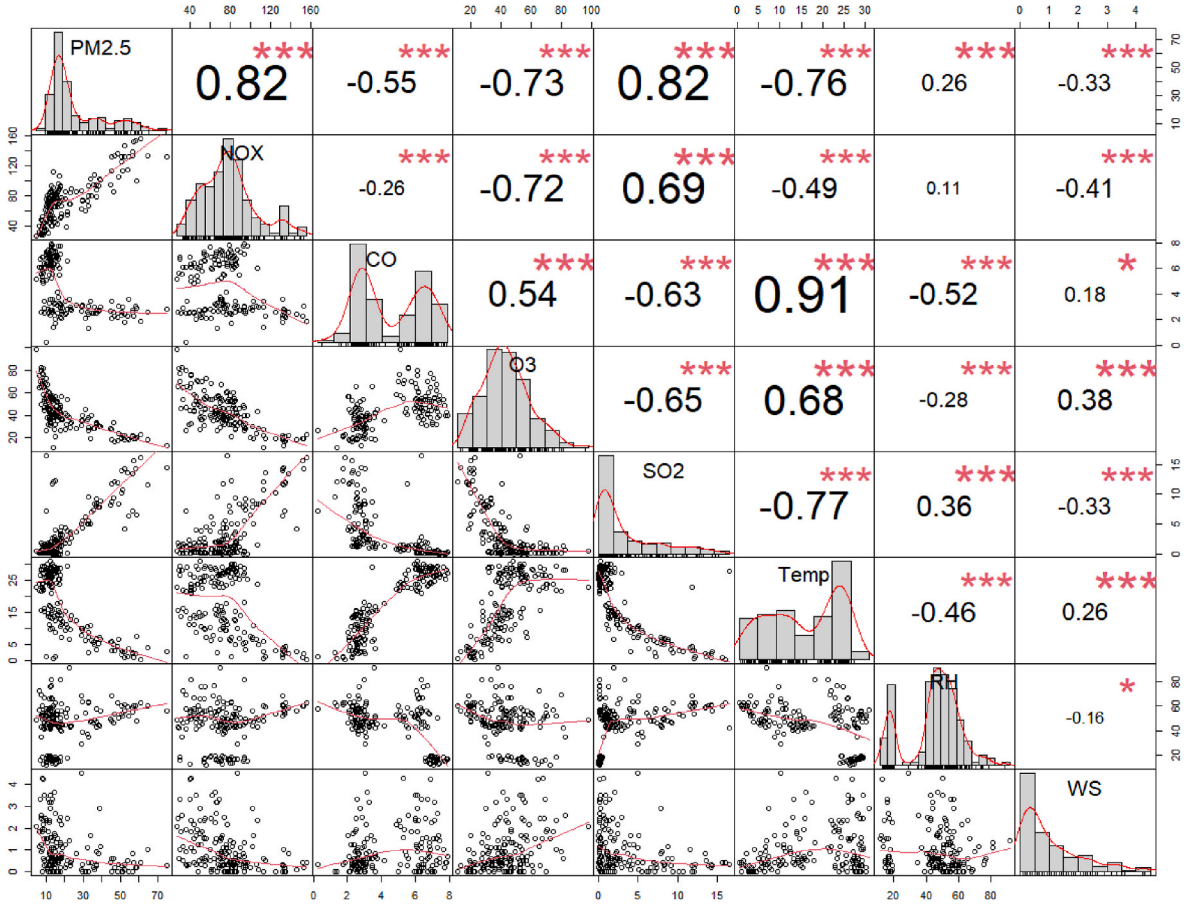


Fig. 5. Correlation analysis of daily mean air pollutants concentrations and meteorological parameters for all studied months.

both. NOx and SO_2 are also positively correlated (0.69). $PM_{2.5}$ is negatively correlated with O_3 (−0.73) and CO (−0.55). NO_x is negatively correlated with O_3 (−0.72) and CO (−0.26). CO and O_3 are positively correlated (0.54), thus SO_2 is negatively correlated with CO (−0.63) and O_3 (−0.65). Strongest correlations are seen among pollutants and Temp – highest for CO (0.91), followed by SO_2 (−0.77), $PM_{2.5}$ (−0.76), O_3 (0.68), then NOx (−0.49). RH showed weaker, inverse correlation, as compared to Temp, for all pollutants: highest for CO (−0.52), followed by SO_2 (0.36), O_3 (−0.28), $PM_{2.5}$ (0.26), with no significant relationship for NOx. WS was also weaker than Temp for all pollutants, with strongest correlation for NOx (−0.41), followed by O_3 (0.38), $PM_{2.5}$ and SO_2 both (−0.33), with no significant relationship for CO.

Similarly, correlation was checked among $PM_{2.5}$, OC, EC, and meteorology parameters, which showed significant negative correlation with temperature, except wind speed, where the correlation was positive and weakest (0.30). $PM_{2.5}$ had exhibited the strongest correlation (−0.64) with temperature. $PM_{2.5}$, OC, and EC, displayed the strong positive correlations with each other as shown in Table S1.

3.4. Health risk assessment for heavy metals

To assess threats to the health of residents triggered by heavy metals in $PM_{2.5}$, non-carcinogenic risks to children and adults through direct inhalation, ingestion and dermal exposure are summarized in Table 1; Table S2). Among the three exposure paths i.e., inhalation, ingestion and dermal; inhalation exposure presented the highest non-carcinogenic effects, followed by ingestion, and then lowest was dermal exposure pathway for both children and adults. The non-carcinogenic risk posed by Ni through inhalation for children is higher than 1 mg/kg/day limit. The rest of the heavy metals via inhalation, ingestion and dermal exposure are below the threshold limits. Only the HI (the sum of HQs) for children through inhalation exceeded threshold limits and vice versa.

3.5. Source apportionment using PCA-MLR

For PCA-MLR analysis, As, Cd, Cr and Cu were not considered because of none-to-low concentrations. Three primary elements were extracted using PCA with varimax rotation accounting for 71% of total variance for the winter, and 58% of the total variance for the summer samples (Table S3). In winter, Factor 1 explained 71% variance dominated with $PM_{2.5}$, Ni, Pb, and Fe, suggesting the role of vehicular combustion (Nagendra and Khare, 2003). Factor 2 in winter showed variance of 61% with maximum loadings of $PM_{2.5}$, EC, and OC, indicating biomass combustion (Zheng et al., 2005). Factor 3 with variance of 38% explained maximum load of Zn and Mg, indicated mixed sources (Jiang et al., 2014; Li et al., 2017). In summer, Factor 1 contributed 58% variance with being heavily weighted by EC, OC, and Ni suggesting a biomass combustion source (Kong et al., 2010; He et al., 2014, 2018). Factor 2 represented 45% variance with the emission load of $PM_{2.5}$, EC and Pb suggesting a vehicular emissions source (Seneviratne et al., 2011). Similarly, Factor 3 explained 31% variance, with the dominance of Zn heavy metals, indicating soil dust (Begum et al., 2011; Kothai et al., 2011). In winter, emissions sources are evenly distributed (Figure S6), while vehicular emissions dominate in summertime.

Table 1
Summary of the non-carcinogenic risk of each element for children and adult through inhalation, ingestion, and dermal exposure.

Non-Carcinogenic (HQ)		
Heavy metals	Children	Adults
Inhalation exposure		
Cr	0.0657	0.0374
Mn	0.282	0.1592
Ni	**1.054**	0.5385
Ingestion exposure		
Cr	0.058×10^{-3}	0.006×10^{-3}
Mn	0.264×10^{-5}	0.02×10^{-5}
Ni	0.25×10^{-4}	0.02×10^{-4}
Cu	0.031×10^{-4}	0.002×10^{-4}
Zn	7.21×10^{-5}	0.77×10^{-5}
Pb	3.06×10^{-3}	0.32×10^{-3}
Dermal exposure		
Cr	0.001×10^{-4}	0.002×10^{-4}
Mn	0.013×10^{-6}	0.015×10^{-6}
Ni	0.008×10^{-4}	0.014×10^{-4}
Cu	0.002×10^{-6}	0.004×10^{-6}
Zn	0.60×10^{-6}	0.926×10^{-6}
Pb	0.293×10^{-6}	0.456×10^{-6}

4. Conclusions

Rising air pollution in the GB region is of concern for Pakistan. This first assessment of air quality finds inter-seasonal variability in $PM_{2.5}$, NO_x, CO, O_3, and SO_2, through the analyzer method. We find that $PM_{2.5}$ levels were higher than the national and WHO daily permissible limits in January and February only, while the remaining NO, NO_2, and SO_2 remained below these health limits. All showed a downward winter-summer trend. CO and O_3 showed a rising trend from winter to summer, with CO being beyond the 8-hourly limit set by Pakistan in all study months, and O_3 crossing its 8-hourly Pakistan limit occasionally in June and July. The results indicated a strong, positive correlation between $PM_{2.5}$, NO_x and SO_2 and a strong, negative correlation with CO and O_3. These results are in line with other studies. Strong correlations with temperature were found across all five criteria pollutants tested, with weaker correlations for wind speed. Diurnal variations in wintertime $PM_{2.5}$, NO_x, and CO and SO_2 found at a peak during the late afternoon and over the midnight period, highlighting the need of protective measures to be employed at these times. Comparatively, O_3 showed opposite peaking trends, with higher variability during wintertime. In Summertime $PM_{2.5}$, NO_x, and CO were found at a peak during the periods of 20:00 to 23:00.

Filter-based analysis revealed similar trends for $PM_{2.5}$, as compared to the analyzer method, except in February and March, where the filter-based $PM_{2.5}$ levels showed a rising trend and exceeded the national and WHO daily permissible limits. Characterization of carbon species (OC and EC) revealed higher variability in winter, as compared to summer, and the OC/EC ratio was indicative of biomass burning and vehicular combustion. Heavy metal concentrations were dominated by Fe, followed by Zn and Pb, with highest concentrations found in the summer, which is when tourism activity rises in the city and region. Heavy metals health impacts found Ni to have significant inhalation exposure in children, which suggests the need for increased funding focused on child healthcare programs. Finally, PCA-MLR technique confirmed OC/EC results, with approximately equal contributions of biomass and vehicular combustion and soil dust in winter, and significantly higher contributions of vehicular combustion in summer.

These results present the first in-depth assessment of seasonal variation in air quality in the city of Gilgit and serve as a baseline against the backdrop of CPEC investments, rising population and tourism influx. The research work has relevance for the Gilgit Environmental Protection Agency (EPA), from the perspective on enhancing rules and regulations, ensuring robust monitoring mechanisms, as well as the need for raising public awareness on this critical public health and environmental protection issue. Within the weak regulatory framework of GB, attention must be paid towards providing alternatives to biomass burning, and vehicular combustion. Building these requirements within the various facets of the burgeoning GB economy will reduce the long-term public health burden, while meeting Pakistan's SDGs commitment in one of its least developed areas. More targeted studies that incorporate the local universities' infrastructure and regional cooperation with HKH neighbors will yield results, particularly with respect to raising the national ambient air quality standards, and incorporation of multiple heavy metals concentrations into discussions of regulatory nature.

Credit author statement

JHS has conceptualized the main idea and acquired funding; NH, MA, SA, KH & SWH have collected and analyzed the samples. NH, MA & HS have prepared the first draft of the manuscript; JHS & SA have reviewed, edited, and finalized the draft for submission.

Funding

JH Syed is thankful to Hong Kong Polytechnic University for Post-Doctoral fellowship funding (#K64160).

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgements

The authors are very grateful to Prof. Guo Hai (Hong Kong Polytechnic University) and Dr. Furrukh Amin (CCRD, Comsats University Islamabad) for their support to complete this research and in preparation of the manuscript, respectively. We are also thankful to Pakistan Meteorological Department (Gilgit Office) and Environmental Protection Agency-Gilgit Baltistan (GB-EPA) for providing the meteorology data and support in installation of air quality monitors and collection of data for the studied period, respectively.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2023.137878.

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