

Point by point Responses to RC2 (egusphere-2024-1385)

Measurement report: Sources and meteorology influencing highly-time resolved PM_{2.5} trace elements at 3 urban sites in extremely polluted Indo Gangetic Plain in India.

In this response we have shown:

The reviewer comments in Black text

The responses to comments are shown in Blue

Addition and changes to the original manuscript are shown in red colour.

Addition in the Supplementary material in red italic

The reviewer's insightful comments and suggestions are highly appreciated. We carefully considered all the comments, addressed each of the suggestions, and made the necessary changes. We believe that this process has significantly improved the manuscript's overall quality. Below is a collection of the detailed responses to each of the comments. (Black text shows reviewer comments, while the responses are shown in blue, and original manuscript and changes to the original manuscript are shown in red colour). Please note that the line numbers are with reference to the submitted manuscript and not the revised manuscript.

Review of “Measurement report: Sources and meteorology influencing highly-time resolved PM_{2.5} trace elements at 3 urban sites in extremely polluted Indo Gangetic Plain in India” by Shukla et al. This manuscript looks at the elemental composition of PM_{2.5} at three sites in the Indo-Gangetic Plain. One of the sampling sites is in a region that is less well represented in the literature. The authors used source apportionment to understand the different emission sources of elemental PM_{2.5} and explored the spatial and temporal variation to be used for mitigation strategies. The topic is interesting and of importance to the community, but revisions are necessary before publication.

We appreciate the reviewer's positive feedback. We thank the reviewer's insightful comments and suggestions. Following the reviewer's suggestion, we have made changes to the manuscript, the revisions made in response to the reviewer's suggestions have helped us in improving the overall quality of our work.

Major comments

#Comment 1

Section 3.3: It would help the reader to have a description of the different source profiles at the beginning of this section. This information is interspersed in the text or buried in the supplemental. In general, as this paper is focused on pollution sources, it is necessary to link these profiles to possible specific sources and/or atmospheric processes - Specific questions that I had in regard to this were:

-What is the difference (in composition and likely sources) between SFC1 and SFC2

-Are Cl-rich and S-rich also related to combustion, as it was discussed the main sources of Cl and S were combustion in the previous section, or are there sources?

-What are the sources of Cu-rich

Thank you for your detailed comments and suggestions. We have addressed the reviewer's concerns by adding a concise paragraph at the beginning of Section 3.3 to describe the different source profiles, including their compositions and likely origins. This provides readers with a clear context before delving into the subsequent sections. For detailed information on the sources, please refer to Supplementary Section S2, along with Figs. S10 to S13, which present the factor profiles, time series, and their correlations with major tracers.

Response to the specific questions:

Difference between SFC1 and SFC2: SFC1 and SFC2 represent different combustion-related processes. SFC1 is primarily associated with biomass burning, particularly crop residue burning, which is supported by the high levels of potassium and its correlation with PAHs. In contrast, SFC2 is characterized by high contributions of Zn, Cr, and Ni, and is more strongly influenced by industrial waste burning involving heavy metals.

Combustion Relevance of Cl-rich and S-rich Factors: Both Cl-rich and S-rich factors are linked to combustion activities. The Cl-rich factor is predominantly influenced by trash burning (including plastic/PVC) and brick kiln activities, while the S-rich factor is associated with coal combustion emissions, particularly from fly ash produced in thermal power plants.

Sources of the Cu-rich Factor: The Cu-rich factor is linked to industrial emissions involving metal recycling and electronic waste burning. The high levels of copper and lead suggest contributions from lead-acid battery recycling and the combustion of electronic waste such as cables and circuit boards.

We have added an additional paragraph, which summarise the sources in the main text section 3.3 from paragraph 2, while the detailed discussion is provided in Supplementary section S2:

“Based on the characteristics and correlation among source profiles (as discussed in detail in supplementary section S2 (Table S5)), similar resolved sources were identified, including Cl-rich (steel industry, trash burning), coal combustion, Cu-rich (metal industry/e-waste burning), Dust, SFC1 (Biomass burning), SFC2 (Industrial waste burning), and S-rich (powerplant and coal combustion) sources, across all three sites. The characteristics of the resolved factors will be discussed in subsequent sections, as well as in Supplementary section S2. Briefly, the Cl-rich factor was primarily dominated by Cl (Figs. S10-S12), with minor contributions from Br and K. This profile is associated with combustion-related processes, mainly from steel industry along with trash burning (specifically plastic/PVC) and brick kilns. According to several studies (Almeida et al., 2015; Ding et al., 2020; Gani et al., 2018), metallurgical activities and incomplete burning of plastic/PVC mostly influence Cl-rich emissions. The S-rich factor was defined by high S content, together with Se and V (Figs. S10–S12), which are suggestive of coal combustion emissions, especially from fly ash generated by thermal power plants (Li et al., 2022). The S-rich profile correlates well with CO₂ (measured using AMS at m/z 44) indicating it to be aged and regional transported, (measured using AMS at m/z 44) (Fig. S13). Cu, Pb, and Br each made significant contributions to the Cu-rich factor (Figs. S10–S12). Lead-acid battery recycling, electronic waste burning (Kolenčik et al., 2013), and industrial metal emissions emits significant amount of Cu and Pb (Julander et al., 2014).

Pb and Se dominated the coal combustion component, supported by As and Se (Figs. S10–S12). The coal combustion factor may be related to lead-smelting or coal burning (home as well as industries) (Manchanda et al., 2021; Rai et al., 2020). SFC1 factor mostly comprised K and S (Figs. S10–S12) with small contributions from Rb, Br, and Se. Strong connection between this profile and PAHs (measured using AMS) (Fig. S13) indicates biomass burning, especially crop residue burning, which is linked to agricultural residue burning (Lalchandani et al., 2022) is further supported by presence of K together with Rb, a marker of biomass burning (Shukla et al., 2021). High contributions of Zn, Cr, and Ni—which are linked to industrial waste burning in earlier studies at U-IGP (Rai et al., 2020; Sharma and Mandal, 2017) define the SFC2 factor. Unlike SFC1, which is connected to biomass burning, SFC2 is

more heavily influenced by industrial activities including the burning of waste materials. High concentrations of Si, Ca, and Fe found in the Dust-related source, however diurnal variation and the meteorology helped us define the dust source as road dust resuspension in U-IGP (both periods) and C-IGP (only cold period). While during warm period at C-IGP, it was related to dust storms. In summary, S-rich and Cl-rich sources connected to combustion activities including coal burning and garbage burning. Representing diverse combustion-related activities, SFC1 and SFC2 were mostly linked to biomass burning and industrial waste burning respectively. Supplementary section S2 addresses the thorough information of these sources backed by elemental composition and association with external tracers (Figs. S10–S13).”

#Comment 2

Section 3.2 focuses on the importance of Pb, Ni, As and (to a lesser extent) Cr in regard to health effects, but there is no discussion of the sources of these elements. This would be valuable to include, either based on distribution across the source profiles, or correlations with the time series of the different profiles.

Thank you for your valuable suggestion to include a discussion on the sources of Pb, Ni, As, and Cr in Section 3.2. We agree that providing information on the sources of these carcinogenic elements enhances the understanding of their health impacts. While our detailed source apportionment analysis is presented in Section 3.3, we recognize the importance of briefly discussing the sources in the preliminary health assessment. Therefore, we have revised Section 3.2 to include a discussion of the potential sources of these elements based on our findings. This addition offers context to the observed concentrations and their implications for public health, while maintaining the focus of the section. We hope this revision addresses your concern and enhances the clarity and comprehensiveness of the manuscript.

The revised section 3.2 discussing health effects is:

“Following USEPA guidelines, we concentrated our health impacts study on four main carcinogens: Pb, Ni, As, and Cr; only these elements have known inhalation reference concentrations (RfCs) for household air. Previous research including Rai et al. (2021) have investigated these metals in the U-IGP, thereby offering a chance to investigate their spatial variance between the U-IGP and C-IGP areas. The source apportionment results discussed in next section attributed the measured concentrations of Pb, Ni, As, and Cr to specific sources. Pb is mostly connected with coal combustion and industrial sources including

emissions from thermal power plants and lead smelting operations (Cui et al., 2019; Swanson et al., 2013). Ni and Cr, linked to industrial waste burning and metallurgical operations, helped to identify the SFC2 factor having high signals of Zn, Cr, and Ni (Rai et al., 2020; Sharma and Mandal, 2017). Coal combustion sources emit mostly As, further confirmed by the presence of Pb and Se in the coal combustion factor (Cui et al., 2019; Swanson et al., 2013). Identification of the sources for the elements with high health impacts from source apportionment is very important for restricting the emissions to reduce the health risks.

and

“At U-IGP2, the exceedance of As was observed to be 10–32% most likely resulting from coal combustion pollutants. Generally keeping below 1%, Ni and Cr exhibited a smaller number of data points surpassing the RfCs (Fig. 3). Still, their importance stems from industrial activity that influences the SFC2 factor. We found an interesting pattern of high exceedances of possibly hazardous amounts of carcinogenic substances during the warm seasons. Particularly, Pb levels surpassed hazardous criteria at the C-IGP1 site; Ni and As levels exceeded at both the U-IGP2 and C-IGP1 sites; and Cr levels exceeded at the U-IGP2 site only during the warm period.”

and

“This implies that public health is always at risk since emissions from sources include waste burning, coal combustion, and industrial operations throughout periods with different meteorological conditions.”

#Comment 3

The authors extensively discuss the observed differences between the warm and cold periods and frequently refer to the role of meteorology. However, they do not clearly propose what meteorological processes are driving the differences. For example, is it differences in temperature/RH, different wind speed and direction patterns, a difference in boundary layer height, etc. Furthermore – Can the authors exclude that it is not due to differences in seasonal trends in emissions rather than meteorology (for example the seasonal variation in crop-residue burning, house-hold combustion for heating). These differences would still be of importance, but they are not, strictly speaking, due to meteorology.

Thank you for your insightful comment regarding the role of meteorology in the observed differences between the warm and cold periods. We appreciate your suggestion to clarify the specific meteorological processes driving these differences and to distinguish between the

impacts of meteorology and seasonal emission trends. In response, we have revised Section 3.4 to explicitly describe the specific meteorological factors contributing to the observed variations. We have detailed how variations in temperature, relative humidity (RH), planetary boundary layer height (PBLH), and wind speed and direction influence pollutant dispersion, accumulation, and secondary formation processes. We have also clarified that while meteorological conditions play a significant role, seasonal differences in emission sources, such as crop residue burning and household combustion for heating, also contribute to the observed variations. Our revised section emphasizes that both meteorology and seasonal emission changes operate in conjunction to drive the seasonal differences observed. We have also subsequently modified the conclusions based on our revised discussion. In addition we have added a Fig. 5, which shows the diurnal variation of sources along with meteorological parameters, during cold and warm period. In the Figure 5, we can clearly observe the variation in the diurnal patterns as well as concentrations of the meteorological parameters (RH, Temperature, Solar radiation, PBLH) during warm and cold periods for all the three IGP sites.

The revised Figure 5 is:

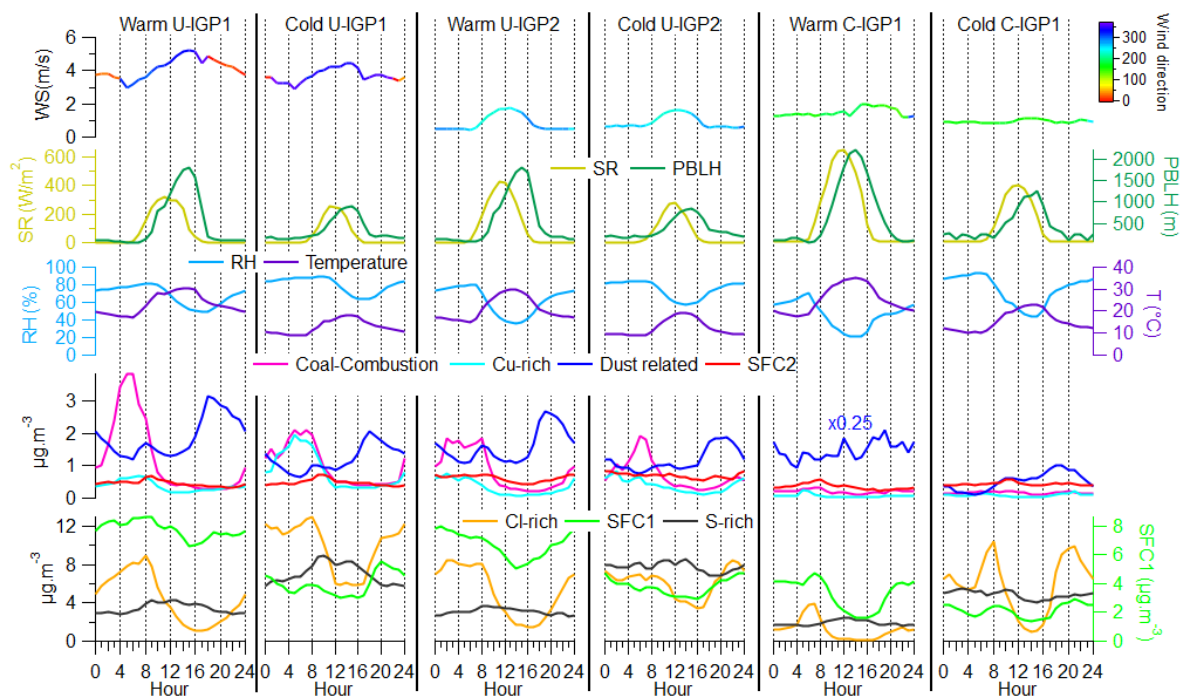


Figure 5. Diurnal variation of elemental sources and meteorological parameters (SR, PBLH, WS, WD, RH and T) at the three sites in the IGP region; U-IGP1, U-IGP2 and C-IGP1 during the warm and cold periods.

We have revised the section 3.4:

“The elevation of $PM_{2.5}$ during the warm period pollution episodes (EP1 and EP2) as compared to clean period C1 can be due to the change in meteorological parameters such as decrease in PBLH (465-657m) during pollution episodes as compared to warm clean period (C1 with PBLH 500-840m). Higher PBLH reduced pollution concentrations on the surface by means of vertical dispersion of pollutants. Dust-related sources, especially at C-IGP1 during EP2 were mainly from wind-induced dust resuspension, which in turn contributed to increase in $PM_{2.5}$ during warm period pollution episodes. Conversely, during clean periods S-rich and dust source (road-dust resuspension) were dominating. In EP1, during the last week of October, agricultural residue burning (SFC1) and other combustion source emissions are rather prevalent, as also reported by Manchanda et al. (2022), especially in significant dispersion conditions. As these observations during warm periods across the IGP indicates severe pollution events also depend on seasonal emission sources along with the meteorological conditions (Fig. 8).

As we investigated at the elemental source change from C1 to EP1, we found that Cl-rich sources at U-IGP1 and U-IGP2 contribution increased significantly. Furthermore, SFC1 increased significantly during EP1, which was explained by the increased burning of agricultural residue. During the warm period, although the temperature and PBLH is high and favorable for dispersion, Lalchandani et al. (2022) and Manchanda et al. (2022) found that this seasonal contribution from biomass combustion was major source of air pollution. Again, from C1 to EP1, secondary aerosol formation mechanisms had a considerable influence, as relative contribution of S-rich reduced but overall concentration increased. During the warm period, the sharp rise in dust-related contributions at C-IGP1 can be ascribed to the dust storms during EP2 resulting from the dry conditions (RH=46%) and windy conditions (WS=2 to 6 m/s). During EP2, the average concentration of the SFC1 source increased, indicating a shift in emission pattern even if its percentage contribution to EI- $PM_{2.5}$ decreased. These findings highlight how the burning of agricultural residue and other factors impact the pollution levels even during the warm period with less favorable meteorological conditions.

Lower temperatures (average 9–24°C), higher relative humidity (69–87%), lower planetary boundary layer height (as low as 167–386-m), and slower wind velocities during the cold phase (EP3 and EP4) as compared to C2, establish conditions favorable for aqueous phase formation of secondary inorganic aerosols and pollutant accumulation (Seinfeld & Pandis, 2006). $PM_{2.5}$ increased with the increase in Cl-rich during cold periods indicates the role of aqueous phase production of secondary aerosols at high relative humidity, as shown in Fig. 7. Further the Cl-rich increase may be due to the contribution from burning roadside trash (PVC/plastic-related waste) for heating purpose. Similarly, secondary aerosol formation from coal combustion emissions helped along with reduced dispersion, the increase of S-rich factor.

The daily fluctuations in the sulfur-rich has distinct trend, especially at C-IGP during cold period (Fig. 8). Usually, photochemical oxidation processes caused the afternoon increase in the S-rich during C2. While the late evening increase in sulfur-rich during the EP3 and EP4 pollution episodes suggests that higher relative humidity helped aqueous-phase formation (Seinfeld and Pandis, 2006) which was not observed during warm periods when temperatures were high and relative humidity was low (photochemical oxidation path were more favorable).

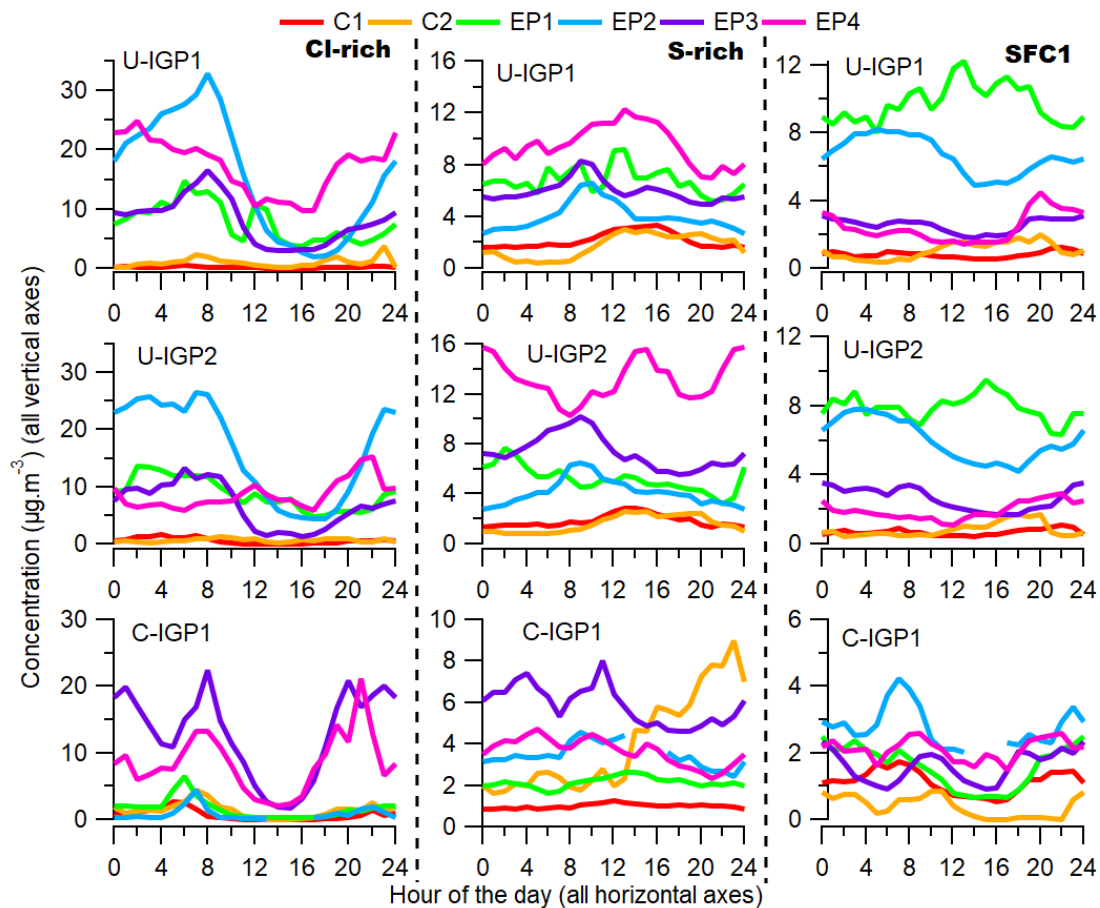


Figure 8. The diurnal variation of Cl-rich, S-rich and SFC1 sources having major role in the variation of EI-PM_{2.5} during the clean and polluted episodes C1, EP1, EP2, C2, EP3 and EP4 during both warm and cold period.

In most pollution episodes and IGP locations, the results show that wind speed, PBLH, temperature, and relative humidity have a significant impact on the amounts of EI-PM_{2.5} and elemental source contributions. Low temperatures, high relative humidity, lower wind speeds, and reduced PBL height all contributed to an increase in Cl-rich and S-rich factors during cold seasons, highlighting the importance of pollution accumulation and secondary production processes. The importance of dispersion mechanisms was highlighted during mild seasons when dust resuspension was more noticeable owing to drier conditions and stronger winds. However, it is impossible to overlook the significant impact that is caused by consistent shifts

in emission sources. The variations in pollution levels seen can be ascribed to events more often occurring in particular periods. Among these are industrial processes, combustion of trash and wood for domestic heating, and burning agricultural residue. Our results show that fluctuations in emissions during different periods along with meteorological conditions explain the cyclical oscillations seen in the IGP region. Variations in emissions across the year highlight the cyclical features of the IGP region's observed weather patterns.”

We have revised the section 4: Conclusion accordingly:

“During the cold period, the Cl-rich source had higher concentrations at night, indicating increased emissions from garbage burning (including PVC and plastics) and the involvement of low PBLH and high RH in pollutant accumulation and formation of secondary aerosols. Mostly, S-rich source concentrations was high during afternoon, possibly due to photochemical sulfate production despite the dilution conditions. Due to increased emissions from industrial processes and biomass burning along with favorable meteorology which enhanced the accumulation caused pollution episodes.

The U-IGP region showed higher concentrations of industrial-related sources, such as the Cu-rich factor associated with industrial metal processes and lead-acid battery recycling. The coal combustion was also higher in U-IGP linked to thermal power plants and industrial emissions highlighting spatial variability influenced by local emissions and meteorological factors in the two IGP regions. The multi-site Potential Source Contribution Function (PSCF) analysis demonstrated the role of regional transport and local emissions in contributing to variations in source origin regions, emphasizing the need for coordinated regional air quality management strategies. During pollution episodes, the relative contribution of Cl-rich increased as when PM_{2.5} concentrations showed sharp increase as compared to clean periods especially during cold, indicating the role of emissions from trash burning (plastic and PVC) and steel industries along with favorable meteorological conditions for formation and accumulation of pollutants.

During warm period, relative contribution of Cl-rich in the C-IGP has less contribution, but during cold period, both the U-IGP and C-IGP have significant Cl-rich source contribution indicating the role of chlorine responsible for hazy extreme air pollution in the whole IGP region. During warm periods high pollution events occurred across the IGP, with major contributions from SFC1 (crop residue burning) indicates that severe pollution events also depend on seasonal emission sources despite less favorable meteorological conditions. These findings underscore the significant impact of both meteorological conditions and seasonal emission sources such as industrial activities, waste burning, and agricultural residue burning on air quality.”

Minor Comments

#Comment 1

Line 356: What is meant by “conventional sources”?

Thank you for highlighting the ambiguity in our terminology. By "conventional sources," we intended to refer to the typical emission sources of sulfur (S) and chlorine (Cl) that are consistently present throughout the year. These include emissions from power plants (a primary source of sulfur) and steel industries (a significant source of chlorine), among others. We have removed the term and revised the manuscript to explicitly mention these sources for clarity.

The revised lines are:

“During the cold period, elevated levels of S and Cl can be attributed to increased anthropogenic combustion activities, including emissions from power plants and steel industries, respectively. However, specific winter meteorological conditions such as higher RH, lower temperatures, reduced planetary boundary layer height (PBLH), and decreased wind speed (Supplementary Fig. S3), enhance the formation and accumulation of sulfate and chloride. Higher RH promotes secondary aerosol formation through heterogeneous reactions (Seinfeld & Pandis, 2006), whereas lower temperatures increase particulate-phase partitioning of semi-volatile compounds (Pathak et al., 2011). A reduced PBLH limits vertical mixing, trapping pollutants near the surface (Zhang et al., 2014) and decreased wind speed limits pollutant dispersion, leading to accumulation (Wang et al., 2014).”

#Comment 2

Line 356: Due to the focus of meteorology in this paper, can you expand on what you mean by “favorable meteorological conditions” in this specific case?

Thank you for your insightful comment. By "favorable meteorological conditions," we refer to specific atmospheric factors during the cold period that enhance the formation and accumulation of sulfate and chloride in the atmosphere. Higher relative humidity promotes secondary aerosol formation through heterogeneous reactions on aerosol surfaces (Seinfeld & Pandis, 2006). Lower temperatures increase the partitioning of semi-volatile compounds into the particulate phase, leading to elevated concentrations of S and Cl in PM_{2.5} (Pathak et al., 2011). A reduced planetary boundary layer height limits vertical mixing, trapping pollutants near the surface and resulting in higher surface-level concentrations (Zhang et al., 2014).

Decreased wind speeds hinder the dispersion and dilution of pollutants, causing them to accumulate in the atmosphere (Wang et al., 2014). Collectively, these meteorological conditions create an environment that favors elevated levels of S and Cl during the cold period by enhancing both their formation and accumulation. We have revised the manuscript to clarify this explanation.

The revised lines are:

" During the cold period, elevated levels of S and Cl can be attributed to increased anthropogenic combustion activities, including emissions from power plants and steel industries, respectively. However, specific winter meteorological conditions such as higher RH, lower temperatures, reduced planetary boundary layer height (PBLH), and decreased wind speed (Supplementary Fig. S3), enhance the formation and accumulation of sulfate and chloride. Higher RH promotes secondary aerosol formation through heterogeneous reactions (Seinfeld & Pandis, 2006), whereas lower temperatures increase particulate-phase partitioning of semi-volatile compounds (Pathak et al., 2011). A reduced PBLH limits vertical mixing, trapping pollutants near the surface (Zhang et al., 2014) and decreased wind speed limits pollutant dispersion, leading to accumulation (Wang et al., 2014)."

#Comment 3

Line 434: Is there further evidence or previous research that supports that the Cl-rich aerosol is related to aqueous phase reactions, rather than increased emissions during this period?

Thank you for your insightful comment. Yes, there is evidence from previous research supporting the role of aqueous-phase reactions in the formation of Cl-rich aerosols during periods of high relative humidity (RH), particularly in the Delhi region of India. A study by Gunthe et al. (2021) published in Scientific Reports investigated the formation mechanisms of Cl-rich aerosols in Delhi. The researchers found that elevated RH during the cold season enhances aqueous-phase reactions, leading to the formation of secondary Cl-containing aerosols. These reactions involve the dissolution of gaseous chlorine-containing precursors and subsequent chemical transformations within the aqueous phase of aerosol particles. The study concluded that the increased levels of Cl-rich aerosols during the cold period are significantly influenced by aqueous-phase processes facilitated by high RH conditions, rather than solely by increased emissions. This finding aligns with our assertion that the elevated Cl-rich aerosols during the cold period are due to enhanced formation through aqueous-phase reactions under high RH conditions as also evident in Figure 5. Based on this evidence, we

have added this reference in line 434 to reflect the role of aqueous-phase reactions more accurately.

The revised lines is:

”The increased Cl-rich and S-rich during cold period can be due to the high RH conditions favour the rapid formation of these secondary inorganic sources as also reported by Lalchandani et al. (2022) through aqueous phase reactions (Gunthe et al., 2021) along with the low PBLH effect (Fig. 5).”

The Figure 5 is:

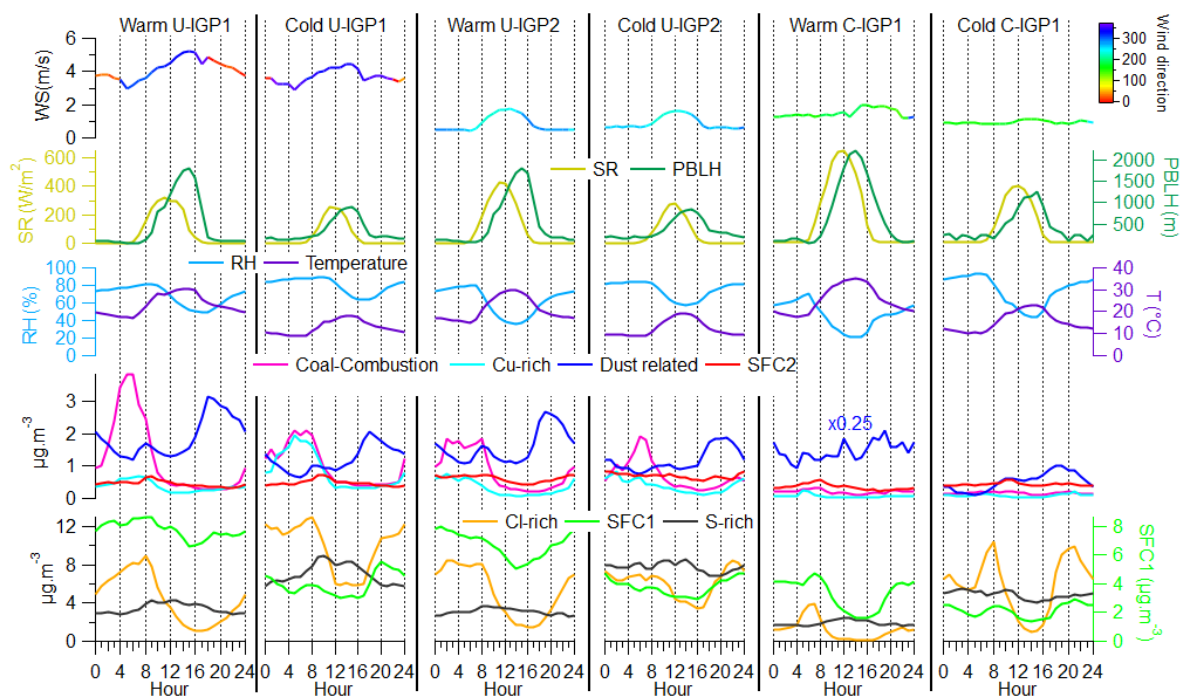


Figure 5. Diurnal variation of elemental sources and meteorological parameters (SR, PBLH, WS, WD, RH and T) at the three sites in the IGP region; U-IGP1, U-IGP2 and C-IGP1 during the warm and cold periods.

#Comment 4

Typographical:-There are numerous grammatical errors throughout the manuscript. Although the meaning is generally clear some sentences are hard to follow, and I recommend the authors thoroughly proofread the manuscript. Some specific recommendations are included below.

Thank you for your review of our manuscript and for indicating the grammatical errors. We have thoroughly checked the manuscript and corrected the grammatical errors. We believe these revisions have improved the clarity and readability of the manuscript.

#Comment 5

Sometimes PM_{2.5} is subscripted (i.e. line 103) while at other points (line 79) it is not. It should be consistent.

Thank you for bringing this inconsistency to our attention. We apologize for inconsistent formatting of "PM_{2.5}" in the manuscript. We have thoroughly reviewed the entire document and have standardized the formatting.

#Comment 6

Line 336: Parentheses do not match.

Thank you for the reviewer's comment. We have corrected the sentence.

The revised lines are:

"The measured elemental concentrations during both periods at two sites in the U-IGP region were nearly identical (at U-IGP1: 19.2 $\mu\text{g}\cdot\text{m}^{-3}$ in warm; 22.9 $\mu\text{g}\cdot\text{m}^{-3}$ in cold and U-IGP2: 17.4 $\mu\text{g}\cdot\text{m}^{-3}$; 19.2 $\mu\text{g}\cdot\text{m}^{-3}$)."

#Comment 7

Line 403: "These findings indicate..." I am having trouble following this sentence

Thank you for the reviewer's comment. We apologize for any confusion caused by the original sentence. We have revised the sentence.

The revised lines are:

" We found an interesting pattern of high exceedances of possibly hazardous amounts of carcinogenic substances during the warm seasons. Particularly, Pb levels surpassed hazardous criteria at the C-IGP1 site; Ni and As levels exceeded at both the U-IGP2 and C-IGP1 sites; and Cr levels exceeded at the U-IGP2 site only during the warm period."

#Comment 8

Line 423: What is RC?

Thank you for bringing this to our attention. "RC" stands for "Relative Contribution. However, to enhance clarity and avoid any confusion for readers, we have removed the abbreviation RC and used "Relative Contribution" in the whole manuscript.

The revised lines are:

"At the U-IGP1 site, significant differences were observed in the relative contribution of the elemental sources."

#Comment 9

I would recommend using the terminology "warm and cold periods" rather than just "warm and cold" for example at line 422, 434, etc

Thank you for your helpful suggestion. We agree that using "warm and cold periods" enhances clarity and consistency in the manuscript. We have revised the text accordingly in the whole manuscript.

The revised lines are:

Line 422: "3.3.1 Variation of elemental sources during warm and cold periods"

Line 434: "The increased Cl-rich and S-rich sources during the cold period can be attributed to high RH conditions that favor the fast formation of these sources through aqueous-phase reactions."

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