Point by point Responses to RC1 (egusphere-2024-1385)

Measurement report: Sources and meteorology influencing highly-time resolved PM2.5 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.

This manuscript investigates atmospheric particulate matter (PM) and associated trace elements in the Upper Indo-Gangetic Plain (U-IGP) and Central Indo-Gangetic Plain (I-IGP), aiming to understand their concentrations, seasonal variations, and sources.

While the study covers an important topic, there are several critical issues that need to be addressed. The rationale for selecting the U-IGP region over other areas is not well explained, leading to logical inconsistencies in the study's focus. Furthermore, the manuscript conflates the definitions of heavy metals and trace elements and does not clearly delineate their significance or relationship. The analysis of seasonal variations, especially concerning crustal elements and dust, lacks clarity, and the use of the term "dust" is ambiguous—it's unclear whether it refers to natural or anthropogenic sources. The discussion on meteorological influences, health impacts, and the PMF method is either insufficient or overly generalized. The presentation quality also needs improvement, with figures that do not effectively convey the key findings, and a confusing structure that hampers the overall readability.

In its current form, the manuscript does not meet the standards required for publication. And in particular the authors need to think about why it should be published as a MEASUREMENT REPORT type of article, or where the scientific highlights need to be highlighted for this type of article. Major revisions are necessary to address the scientific and presentation-related issues.

We appreciate the reviewer's feedback. We thank the reviewer's insightful comments and suggestions. Following the reviewer’s suggestion, we have made significant 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.

Here are my specific recommendations:

#Comment 1

Lines 39-40: Why is it UPPER IGP and not examining IGPs of other orientations? There is a logical problem with the reason for the point selection here.

Thank you for your comment. To clarify, the study aims to fill the gap in research coverage by expanding the scope to both the Upper and Central Indo-Gangetic Plain (U-IGP and C-IGP). Our intention was to emphasize that while previous research has predominantly focused on the U-IGP (mostly offline and a few high-resolution studies with no spatial coverage), we extended our investigation to the C-IGP (only a few offline and no high-time resolution measurements) in order to capture a comprehensive picture of the aerosol dynamics across the region. Following the Rai et al. (2021) study, which compared the one site in the U-IGP (same as our U-IGP1 site) with the major cities globally (London, Krakow and Beijing) and found that it was three times higher, when we compared our C-IGP concentrations with that study we found that although the C-IGP concentrations were lower than those in the U-IGP, they were significantly higher than the mean concentration measured in Beijing (2.6 times), Krakow (3.1 times), and London (15 times). This difference clearly tells the importance of high-time resolution studies in the other part of the IGP regions. We have revised the abstract to make this clearer and hope this addresses Reviewer’s concern.

We have modified the Lines 38-42 in the revised MS:

“However, studies (mostly offline) have been predominantly concentrated in the Upper IGP (U-IGP), with limited spatial coverage across the Central IGP (C-IGP). This work aims to bridge the gap by examining both U-IGP and C-IGP regions, offering an in-depth analysis of elemental concentrations, sources, regional comparisons, seasonal fluctuations, meteorological impacts, and health hazards through the use of Xact at three urban locations.”

#Comment 2

Lines 42-44: Has there been a change in major contributors? If not, then the word while should not be used after the comma.

Thank you for pointing this out. We agree that the use of 'while' may be misleading. We have revised the sentence to clarify the seasonal differences in elemental contributions and removed any ambiguity.

We have modified the Lines 42-44 in the revised MS:

“S, Cl, and K were prominent in elemental $PM_{2.5}$ during cold seasons; warm seasons showed notable changes in Al, Si, Sr, and Ba concentrations, so highlighting the influence of meteorological conditions and different emission sources on pollution levels.”

#Comment 3

Lines 64-73: The review is confusing and here confuses the definitions of heavy metals and trace elements and does not systematically address the relationship between the two and their respective importance.

Thank you for highlighting this point. We agree that the previous text could have led to confusion regarding the definitions of heavy metals and trace elements. In response, we have used only one term “elements” in the whole manuscript.

We have modified the Lines 64-73 in the revised MS:

“Though elements make a small portion of $PM_{2.5}$, they are helpful in source apportionment as they are unique marker of the air pollution sources (Rai et al., 2020). Some elements such as lead, cadmium, and mercury, are especially worrisome because of their toxicity and possibility for bioaccumulation in vital organs once absorbed into the bloodstream (Giordano et al., 2013; Kastury et al., 2017; Rehman et al., 2018). The toxicity and exposure risks of elements are influenced by weather conditions and emission sources (Holden et al., 2016; Tchounwou et al., 2012).”

#Comment 4

Lines 74-75: Just because PM is elevated doesn't mean it's not important. The order of magnitude that PM can reach needs to be given to help the reader determine how important it is.

Thank you for your helpful comment. We have revised the sentence to include the order of magnitude of PM concentrations, particularly during pollution episodes, to highlight the significance of elevated PM levels in the Indo-Gangetic Plain. This provides the necessary perspective to better point to the importance of the issue.

We have modified the Lines 74-75 in the revised MS:

"An alarming rise in atmospheric PM concentrations, which can exceed 500 µg/m³ during pollution episodes in India's Indo-Gangetic Plain (IGP), such as the extreme episode from October 31 to November 8, 2016, when hourly $PM_{2.5}$ levels exceeded 500 µg/m³ on several days (Agarwal et al., 2024). Urbanization, industry, and biomass burning (Pant et al., 2015) all contribute to this pollution situation including extreme haze events. Pollution levels are influenced by major meteorological variations between the warm and cold periods (Bhat et al., 2017; Dumka et al., 2021; Kumar et al., 2020)."

#Comment 5

Line 77: I don't think it's climate change (on a long-term scale), more that it's a change in meteorological conditions.

We thank the reviewer for highlighting this point. We have modified the sentence.

We have modified the Lines 77 in the revised MS:

"Pollution levels are influenced by major meteorological variations between the warm and cold periods (Bhat et al., 2017; Kumar et al., 2020; Dumka et al., 2021)."

#Comment 6

Lines 80-82: I still don't understand why the U-IGP was chosen - was it because the atmospheric issues were not prominent, or was it for some other reason?

Thank you for your insightful comment. This study was focused on elemental variation between both U-IGP and C-IGP. By comparing these two regions, this study aims to provide a more comprehensive understanding of the spatiotemporal variability in elemental sources and their meteorological drivers. The Upper Indo-Gangetic Plain (U-IGP) was chosen for this study because it represents a region with a substantial body of research, particularly on particulate-bound organic aerosols. However, studies focusing on elemental sources and high-time resolution measurements are still limited, especially in the context of seasonal variations and meteorological influences. The Central Indo-Gangetic Plain (C-IGP), on the other hand, has been largely understudied, despite its distinct pollution profile influenced by industrial activities, vehicular emissions, regional transport of dust, and crop-residue burning.

Additionally, most previous studies in the U-IGP have relied on offline filter analysis, whereas this study employs high-time resolution measurements, providing new insights into short-term variability in elemental $PM_{2.5}$.

The Indo-Gangetic Plain (IGP) spans over 255 million hectares of densely populated and agriculturally rich land across northern India, Pakistan, and Bangladesh. The U-IGP, covering Punjab, Haryana, and Delhi, is characterized by significant urban emissions, including traffic, industries, brick kilns, and biomass-based energy. In contrast, the C-IGP (Uttar Pradesh and Bihar) faces a combination of regional and long-range dust transport, power plant emissions, crop-residue burning, and other sources. The inclusion of both regions in this study allows for a detailed examination of how local meteorology and varying emission sources influence $PM_{2.5}$ composition."

We have modified and expanded the Lines 80-82 in the revised MS:

" Although most PM based research has focused on the Upper IGP (U-IGP), notably on particulate-bound organic aerosols, there is still a major knowledge gap on elemental contributions, particularly on high-time resolution observations. Conversely, the Central IGP (C-IGP) has gotten even less attention; just few offline research on either organic or elemental PM. The present work intends to close the literature gap between U-IGP and C-IGP by means of elemental sources and their seasonal fluctuations as well as the impact of meteorological conditions on pollution levels. High population density, industrial emissions, traffic exhaust, and biomass burning define the U-IGP; additional influences such long-range mineral dust transportation, crop-residue burning, and power plant emissions, which vary greatly with meteorological conditions, define the C-IGP."

#Comment 7

Line 94: What's the meaning of SA?

We thank the reviewer for highlighting this abbreviation, we have expanded it. SA abbreviation in the manuscript is used for Source apportionment. We have thoroughly checked the manuscript and put abbreviation and its expanded form at the point of its first use for reference.

Line 94:

“Few source apportionment (SA) investigations on elemental data with high-time resolution recently had been conducted in U-IGP (Delhi) exclusively (Manchanda et al., 2022; Rai et al., 2020; Shukla et al., 2021).”

#Comment 8

Lines 95-98: Are these statements intrinsically related? What do the differences between warm and cold season crustal elements and Cl indicate? Is dust here dust or anthropogenic source dust and does the 50%+ contribution of dust conflict with the results of this paper? Also, are some of the important sources covered in this paper all reported in previous work with source profiles? That is, are the sources in this paper reliably and consistently referenced?

Thank you for your comment. The observed differences between warm and cold season crustal elements and Cl are likely due to role of both meteorology and distinct emissions during both periods. Higher temperatures and lower RH in the warm period contributed to increased resuspension of crustal elements, while Cl levels were higher in the cold period due to increased combustion emissions and a lower boundary layer trapping pollutant.

The Shukla et al. (2021) study was conducted during June dust storms, reported high dust contributions (52.5%). In our study, warm and cold periods are based on relative temperatures, and dust also contributed significantly when dust storms were observed in the C-IGP during warm periods. However, it's important to clarify that dust is not included in the secondary source category. The dust factor is reported to be dominated by Si, Ca, Fe, Al and K, these elements have been used as markers for crustal dust/soil (Sharma et al., 2016; Sun et al., 2019). Instead, we grouped Cl-rich and SO_4 sources, which contributed 27% during the study period. Similar trends were found for secondary organic aerosols (SOA), highlighting the need to reduce secondary aerosol precursors in Delhi. We have also included some more studies which have reported these sources. The presence of S and Se in the S-rich factor is consistent with the findings of previous studies (Hao et al., 2020; Weber et al., 2019). These previous studies were referenced to provide context for U-IGP research, not for direct comparison. Our study uses bootstrapping for uncertainty analysis and provides detailed discussions on source profiles, diurnal variations, and correlations in the supplementary materials (section S2).

We have modified and expanded the Lines 95-98 in the revised MS:

"Rai et al. (2020) found that during the warm period crustal elements more contributed to elemental $PM_{2.5}$, may be due to higher temperatures and reduced RH facilitating their resuspension. The dust factor is reported to be dominated by Si, Ca, Fe, Al and K, these elements have been used as markers for crustal dust/soil (Sharma et al., 2016; Sun et al., 2019). In contrast, Cl-rich factor dominated by Cl, linked to combustion and industrial activities was more prominent during the cold period (Ding et al., 2020). Shukla et al. (2021), reported that during summer (June), dust source was prominent as several dust storms occurred associated with high wind speed and temperature along with low RH. In our work also, dust made a major contribution during warm C-IGP seasons. Furthermore, chloride and sulfate (classified as secondary oxidized aerosols) contributed 27%, which emphasizes the significance of lowering secondary aerosol precursors in U-IGP."

The supplementary section S2 is:

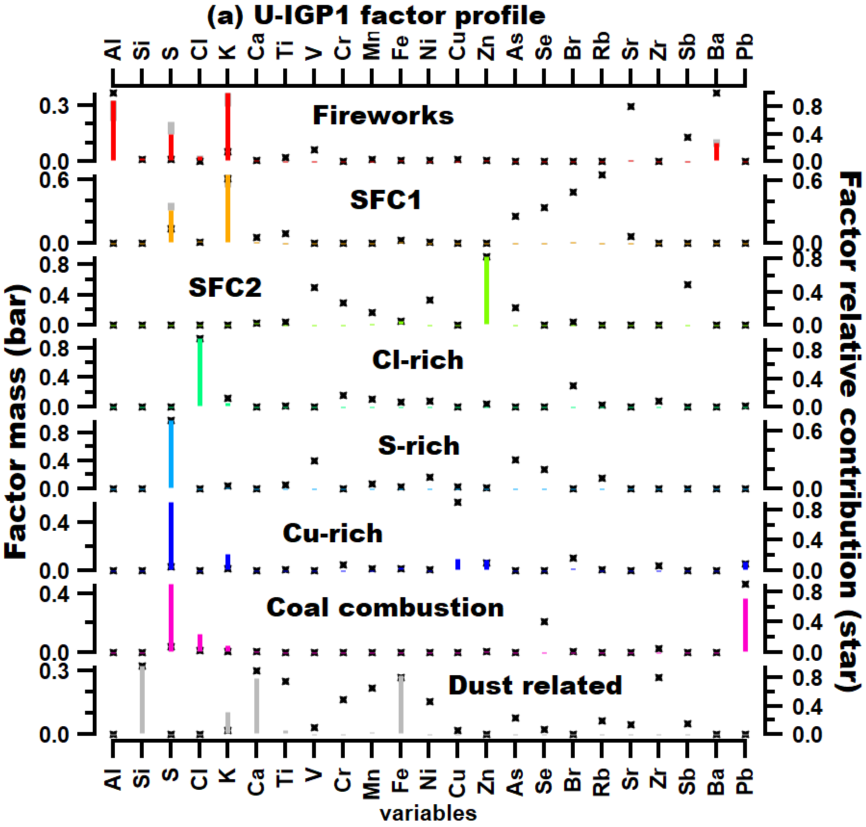
"The 7 factors Cl-rich, S-rich, Cu-rich, Coal combustion, SFC1, SFC2 and dust related sources are resolved at all the three sites, while an additional fireworks factor was resolved at the U-IGP1 and U-IGP2 sites. The seven factors resolved at all three sites are Cl-rich, S-rich, Cu-rich, Coal combustion, SFC1, SFC2, and dust-related sources. Additionally, a fireworks factor was identified at the U-IGP1 and U-IGP2 sites. Our study focuses on the variation of these seven common factors, which will be further discussed in the section describing factor profiles. The role of meteorology in the variation of elemental sources between the three sites in the IGP region is also investigated. It is important to note that a separate study on the fireworks factor during Diwali at two sites in the U-IGP was already conducted by Manchanda et al. (2022).

The Cl-rich factor profile mass (U-IGP1, U-IGP2, and C-IGP1) is primarily dominated by high levels of Cl (93%, 94%, and 87%). It also exhibits relative contributions from Br (30%, 38%, and 17%) and K (12%, 13%, and 16%), which are markers for combustion related to trash burning (specifically plastic/PVC) and brick kilns. A source apportionment study by Wang et al. (2023) investigated the composition of the Cl-rich factor and identified contributions from combustion processes, such as trash burning and brick kilns. Their findings support that the Cl-rich factor is predominantly influenced by emissions from these combustion-related sources. The good correlation (R=0.6-0.7) of Cl-rich with PAHs indicates incomplete combustion of plastic/PVC products mixed with other waste materials, especially from waste incineration.Jin et al. (2020) found that waste combustion and metallurgical smelting are the primary anthropogenic sources of chlorinated and brominated PAH emissions. Additionally, e-waste dismantling has been suggested as an emerging source of chlorinated PAHs.

The S-rich factor mass primarily consists of high signals from S (98%, 98%, and 88%). It also shows relative contributions from Se (20%, 17%, and 32%) and V (29%, 44%, and 12%). These elements are associated with fly ash from coal combustion in thermal power plants. (Yi et al., 2022) conducted a source apportionment analysis on the S-rich factor and reported a strong association with fly ash emissions from coal combustion. The presence of selenium and vanadium in the S-rich factor, as identified in their study, provides additional evidence linking this factor to coal combustion sources. The S-rich factor demonstrates a good correlation with CO2 (R=0.65) at all three sites, indicating its aged nature. The diurnal peak during the afternoon suggests that it undergoes transport. The Cu-rich source exhibits a very *high relative contribution of Cu (90%, 91%, and 91%) along with Pb (9%, 5%, and 5%) and Br (16%, 8%, and 2%). Cu and Pb are related to industrial metal sources, lead-acid battery recycling, and burning of electrical/electronic waste containing Cu (such as cables and circuit boards). Chen et al. (2022) conducted a source apportionment analysis and found that the high levels of lead and copper in the Cu-rich factor can be attributed to industrial emissions, particularly from metal-related industries and lead-acid battery recycling. This finding aligns with the hypothesis that the Cu-rich factor is influenced by industrial metal sources.*

The coal combustion factor is characterized by high signals of S (46%, 48%, and 39%) and Pb (36%, 44%, and 3.4%). It also shows significant relative contributions from Pb (90%, 86%, and 10%), As (1%, 26%, and 94%), and Se (40%, 24%, and 29%). These elements indicate emissions from industrial and domestic coal combustion, as well as from lead smelting. As and Se are commonly used as markers for coal combustion, and lead in coal is mainly present in the form of sulfides (e.g., galena and pyrite) (Cui et al., 2019). Zhang et al. (2014), the coal combustion factor was found to be dominated by sulfur and lead, confirming the presence of emissions from coal combustion sources. The identification of arsenic and selenium in the coal combustion factor further supports that these elements serve as markers for industrial/domestic coal combustion activities and lead smelting emissions. The SFC1 factor profile mass (U-IGP1, U-IGP2, and C-IGP1) is mainly dominated by high signals of K (67%, 55%, and 64%), followed by S (32%, 41%, and 26%). It also exhibits high relative contributions from K (62%, 64%, and 49%), Rb (66%, 65%, and 55%), Br (49%, 43%, and 49%), and Se (34%, 46%, and 39%). The good correlation with PAHs (R=0.5-0.7) and the presence of these elements indicate that the SFC1 factor is associated with biomass burning, especially crop residue burning. In a recent source apportionment analysis conducted by Liu et al. (2023), the SFC1 factor was found to be strongly associated with biomass burning, particularly crop residue burning. The high levels of potassium and sulfur, as well as the correlation with PAHs, observed in their study provide additional evidence linking the SFC1 factor to biomass burning sources.

The SFC2 factor is characterized by a high relative contribution of Zn (83%, 85%, and 70%), Cr (27%, 26%, and 16%), and Ni (30%, 18%, and 15%). The presence of Zn, Cr, and Ni indicates the role of industrial waste burning (Rai et al., 2020). Investigating the SFC2 factor, a source apportionment study by Sharma et al. (2016) revealed significant contributions from industrial waste burning sources. The elevated levels of zinc, chromium, and nickel identified in their analysis support the hypothesis that the SFC2 factor is influenced by emissions from such industrial waste burning activities. The dust factor profile consists of mass from Si (32%, 32%, and 57%), Ca (26%, 24%, and 19%), and Fe (28%, 24%, and 19%). It also demonstrates high relative contributions from Si (97%, 96%, and 100%), Ca (90%, 98%, and 88%), Fe (80%, *78%, and 80%), Ti (74%, 83%, and 84%), Ni (47%, 57%, and 67%), Cr (50%, 64%, and 61%), and Mn (65%, 66%, and 71%). All these elements are related to dust sources, and they exhibit a very high correlation with each other, as shown in Fig. S1."*



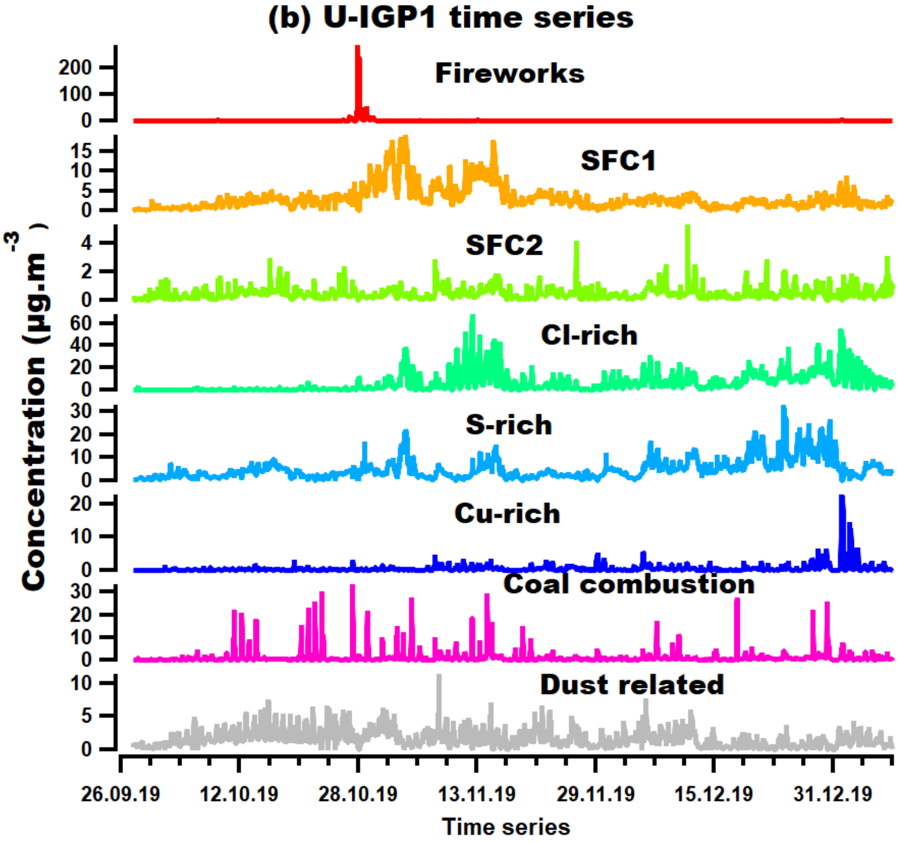
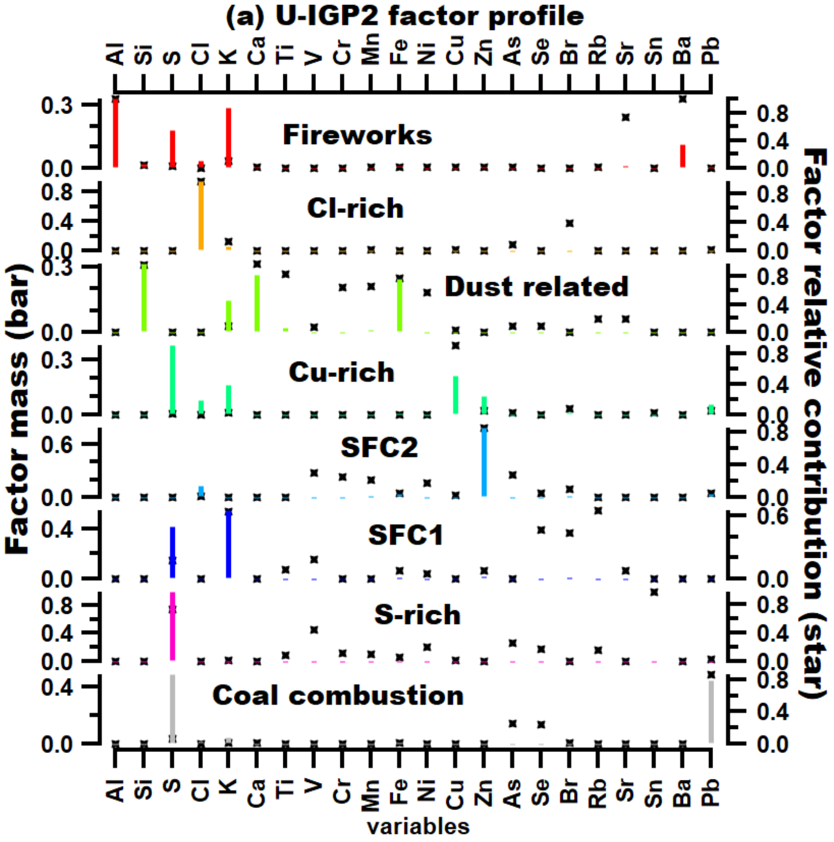


Figure S10. (a) Source profile of the 8-factor solution and (b) time series of the sources from the elemental source apportionment at U-IGP1.



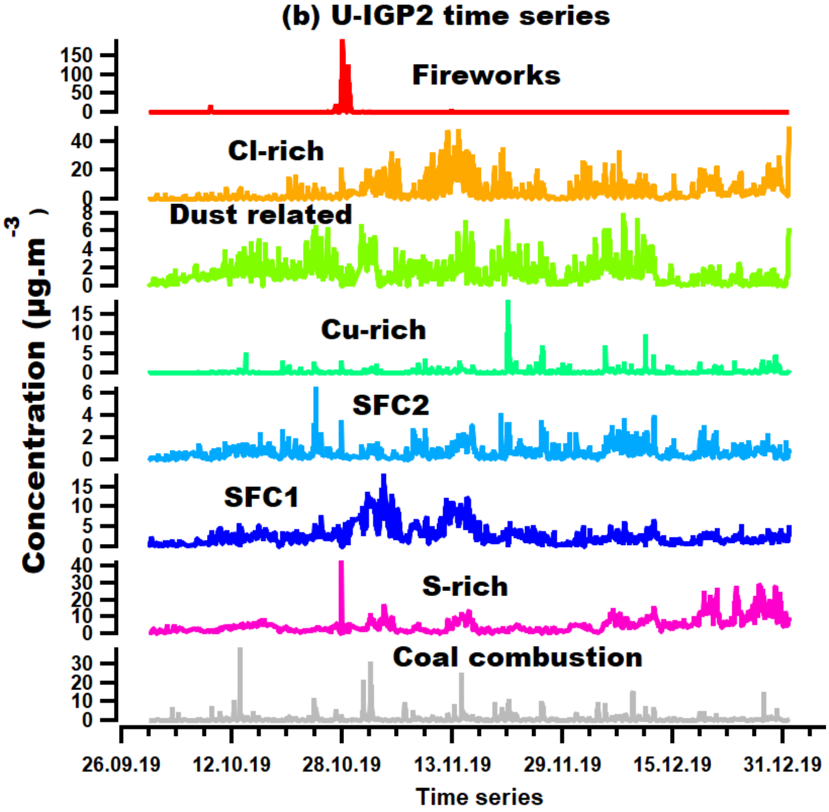
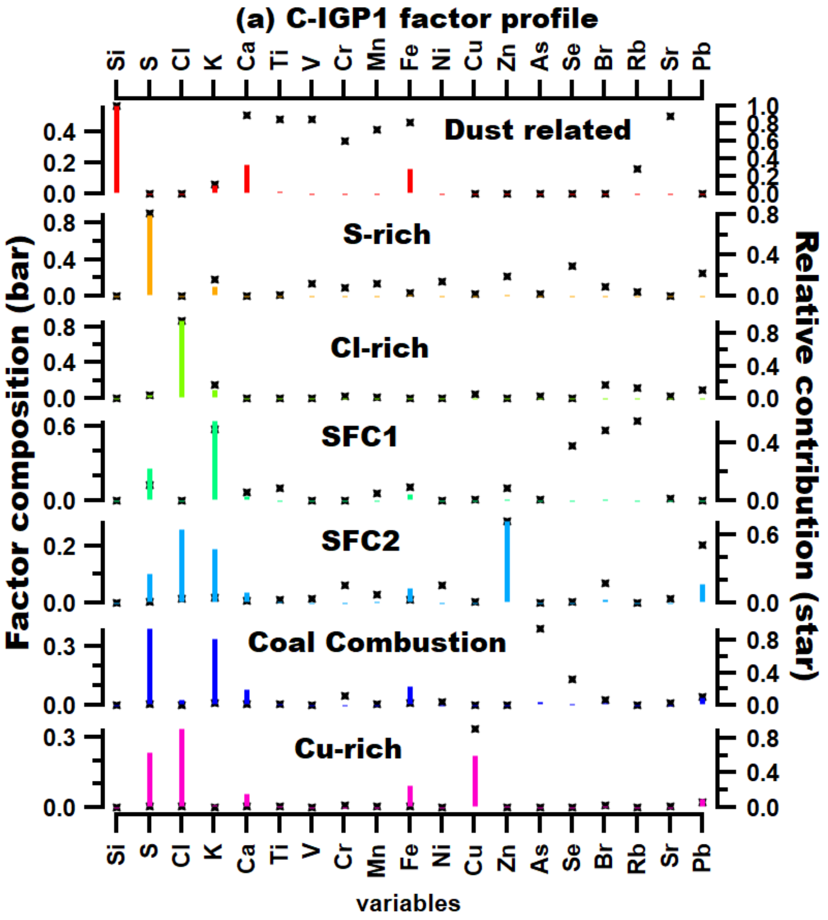


Figure S11. (a) Source profile of the 8-factor solution and (b) time series of the sources from the elemental source apportionment at U-IGP2.



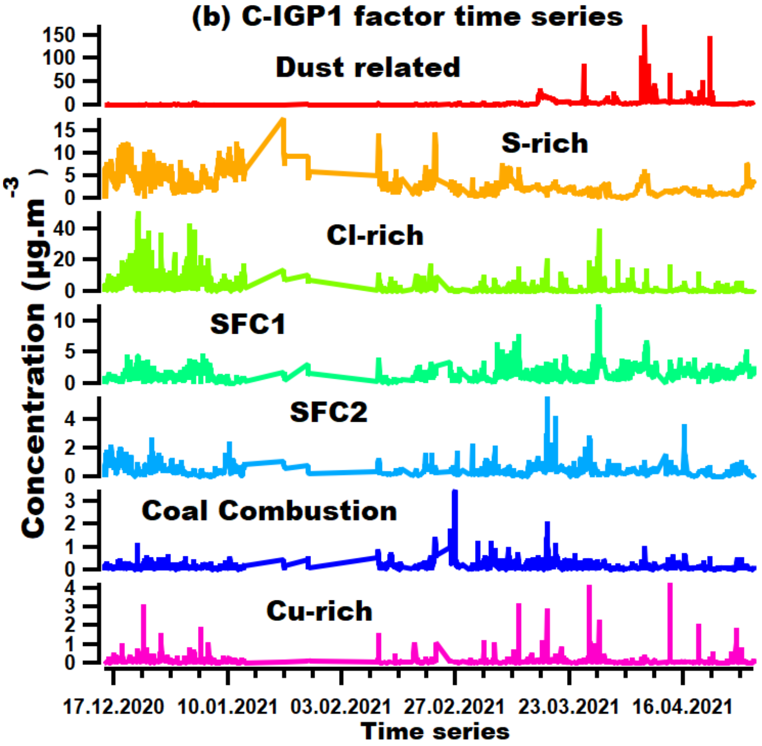
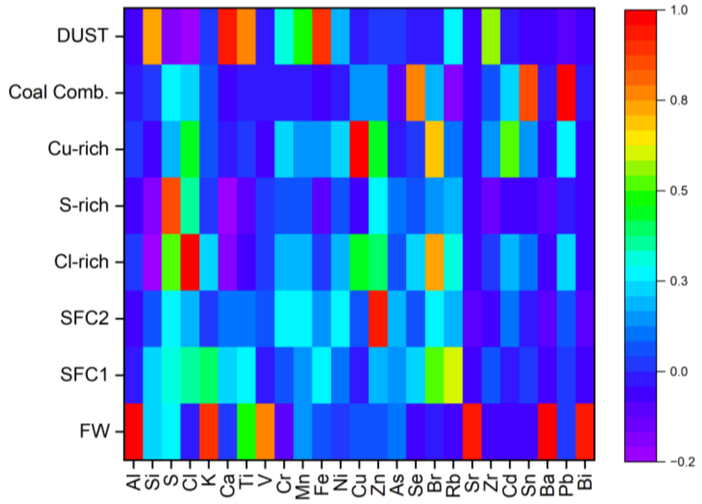
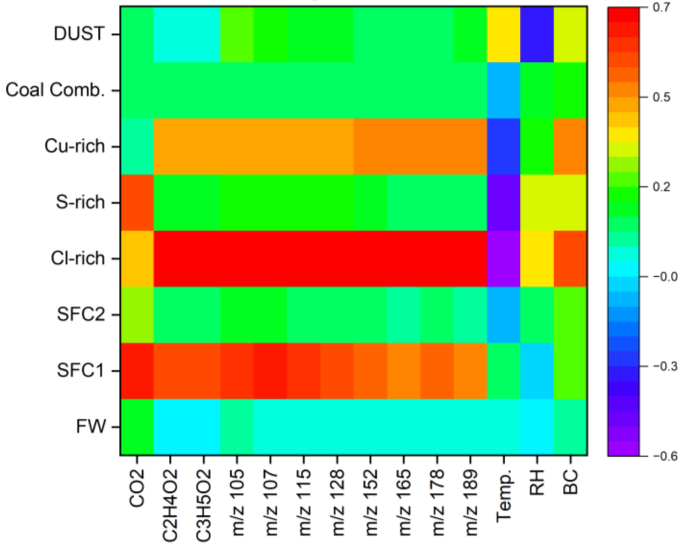


Figure S12. (a) Source profile of the 7-factor solution and (b) time series of the sources from the elemental source apportionment at C-IGP1.

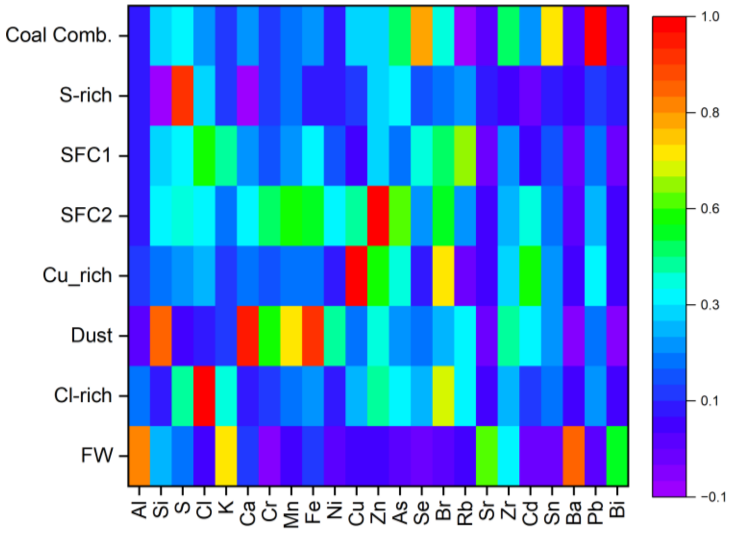
(a) Heatmap for correlation of sources at U-IGP1 with elements



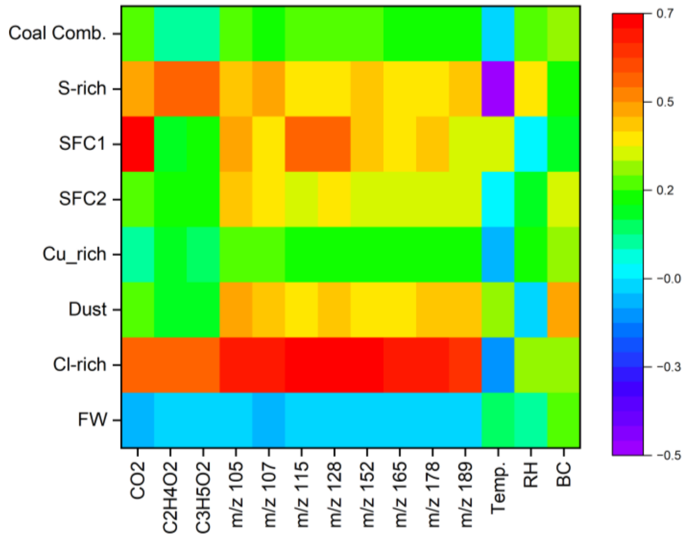
(b) Heatmap for correlation of sources at U-IGP1 with AMS markers, Meteorological markers and BC



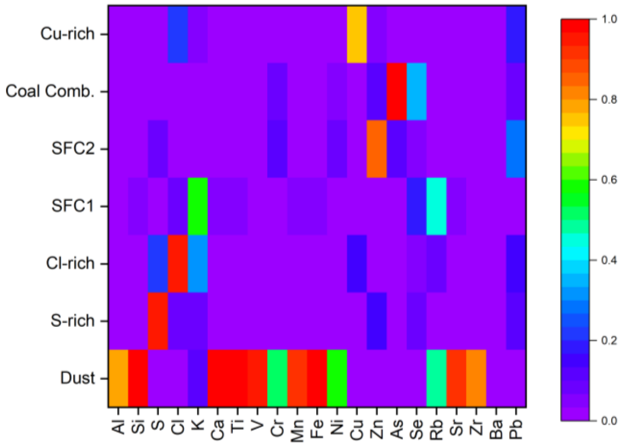
(c) Heatmap for correlation of sources at C-IGP1 with elements



(d) Heatmap for correlation of sources at U-IGP1 with AMS markers, Meteorological markers and BC



(e) Heatmap for correlation of sources at C-IGP1 with elements



(f) Heatmap for correlation of sources at C-IGP1 with AMS markers and BC

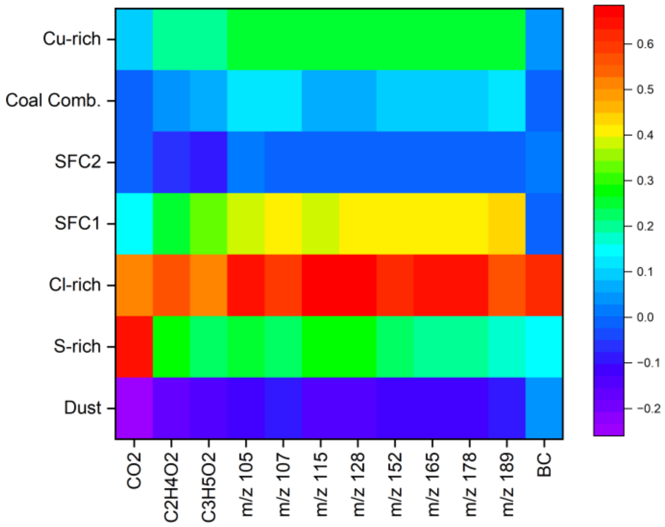


Figure S13. Heatmap showing correlation of resolved elemental sources with external tracers (a-f)

#Comment 9

Lines 99-100: The reference here is strange. Dust itself is not a secondary source of oxidation.

Thank you for your observation. We agree that dust is not a secondary source of oxidation. To clarify, in our study, dust is treated as a separate primary source, and it is not grouped under secondary sources. The secondary sources we refer to consist of Cl-rich and S-rich, which are associated with secondary oxidation processes. This differentiation ensures that the contribution of dust and secondary oxidized aerosols is accurately represented, and their distinct impacts are clearly communicated.

We have modified lines 99-100:

"Furthermore, chloride and sulfate (classified as secondary oxidized aerosols) contributed 27%, which emphasizes the significance of lowering secondary aerosol precursors in U-IGP."

#Comment 10

Lines 125-126: Please label in Figure S1, which regions are represented by U, C, and L respectively. If the authors consider the U-IGP to be important, then it is perfectly acceptable to put a diagram of the study area in the text.

Thank you for your valuable suggestion. We have updated Figure S1 to clearly label the Upper (U-IGP), Central (C-IGP), and Lower (L-IGP) Indo-Gangetic Plain regions. To enhance clarity and provide a better geographical context, we have included a detailed map of the study area in the main text, highlighting all three IGP regions and our sampling sites. We have moved the previously zoomed-in inset of the U-IGP region to the Supplementary Materials to avoid confusion, as our primary focus is on the C-IGP and U-IGP regions. These changes should make it easier for readers to understand the regional distinctions and the importance of both the C-IGP and U-IGP in our study.

The Revised Figure 1 is:

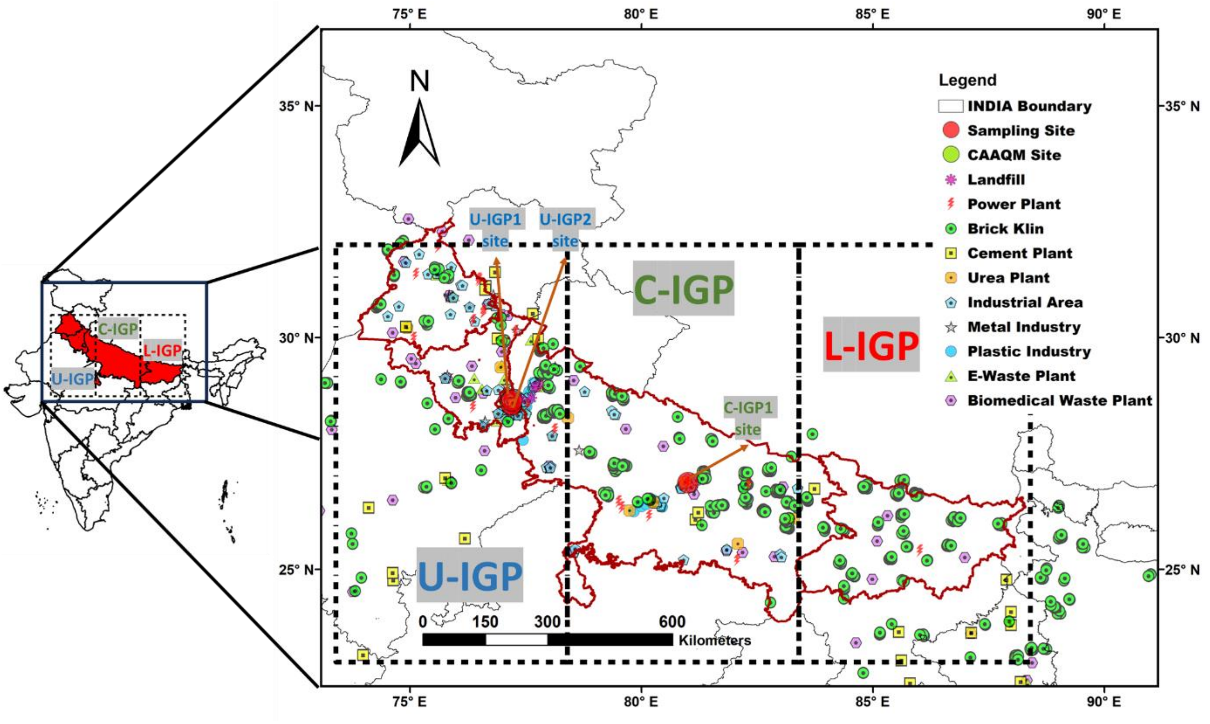


Figure 1. Detailed map with the sampling locations and the nearest government monitoring CAAQMS stations (shown in red circle) in the IGP region. The possible trace element sources have also been shown in the map. The inset picture zooms the sampling sites and industrial emission sources in the U-IGP region.

#Comment 11

Lines 140-150:Because this paper is a MEASUREMENT REPORT, it is still very different from a typical research paper. So please elaborate on the representativeness of the sites selected for this study to confirm the scientific increment and value of this report.

Thank you for your insightful comment. The selected sites are representative of the diverse emission sources in both U-IGP and C-IGP regions, which adds scientific value to this study. In U-IGP, the sites in Delhi cover both central and southern regions, capturing emissions from residential, educational, industrial, and major traffic sources, as well as seasonal agricultural burning. The C-IGP site in Lucknow represents a mix of urban, commercial, and industrial activities, including steel, cement, and chemical plants. These varied sources allow the study to provide a comprehensive overview of regional pollution dynamics, making the findings applicable to a broader context.

We have revised the lines 140-150:

"The Indian Institute of Technology Delhi, Hauz Khas (U-IGP1), and the Indian Institute of Tropical Meteorology, New Delhi branch, Pusa Forest (U-IGP2), were the two designated sample locations in U-IGP. U-IGP1 is located in South Delhi, among residential neighbourhoods, educational establishments, and a major highway (Outer Ring Road). This site displays urban emissions, specifically those originating from industrial sources, vehicular traffic, and residential cookery. U-IGP2 in downtown Delhi is influenced by the residential, woodland, and transportation sectors. Delhi's urban emissions are documented on both sites, which also include seasonal agricultural burning from neighbouring regions. The sampling location (C-IGP1) in Lucknow is located in a diverse urban environment, adjacent to residential zones, business sectors, and major roads, within the C-IGP. This is indicative of the typical urban-industrial pollution sources, as adjacent industries, such as steel, cement, and chemical factories, contribute to a combination of emissions. These sampling sites offer a comprehensive overview of pollution throughout the IGP."

#Comment 12

Line 158: Please check that the number of valid digits is the same.

We thank the reviewer for pointing this out. We have fixed the number of valid digit upto 1 decimal point for all the numbers.

Revised Table 1:

	U-IGP1		U-IGP2		C-IGP1	
(Mean±S.D)	Warm	Cold	Warm	Cold	Warm	Cold
Temp (°C)	23.3±5.2	12.9±4.9	21.2±6.1	12.5±5.3	25.6±6.7	15.4±5.2
R.H (%)	67.9±13.0	79.7±13.3	61.6±17.4	74.8±12.5	46.0±19.7	74.2±20.8
PBLH (m)	506.2±663.2	347.0±365.4	546.6±78	376.1±338.8	697.4±900.1	394.0±416.8
Solar radiation (W/m^2)	86.8±125.2	60.7±97.3	115.0±167.0	68.0±102.0	186.0±246.5	106.0±156.2
Sampling period	1st Oct- 26th Nov	27th Nov-08th Jan	1st Oct- 26th Nov	27th Nov-08th Jan	25th Feb-30th Apr	15th Dec-25th Feb

#Comment 13

Section 2.2: There is also confusion here about the formulation of the instrument. For example, lines 168-178 are also all about instruments, so why are they not integrated into subsection 2.2?

Thank you for your comment. We have revised the content between lines 164-177 to better distinguish the sampling details from the instrumentation. Specifically, we retained the relevant information about the sampling sites and their external environments, such as the height of the sampling locations, in the "Sampling Details" section. The remaining information, which pertains to the setup and operational details of the instruments (e.g., temperature control, humidity regulation, and instrument enclosures), has been moved to and integrated within Section 2.2, which discusses instrumentation. This revision ensures a clearer distinction between the site-specific sampling conditions and the technical aspects of instrument operation.

In section 2.1 sampling details, we have the lines:

"The U-IGP1 sampling site was located on the third floor of the Centre for Atmospheric Science (CAS) building at IIT Delhi, collected aerosols using a $PM_{2.5}$ cut-off cyclone mounted on the rooftop. The U-IGP2 location housed similar instrumentation on the second floor within a temperature-controlled laboratory and used an expanded $PM_{2.5}$ sampling tube. Instruments were situated on the rooftop laboratory at the C-IGP1 site, located within the office building (~12 m above ground level) of the Uttar Pradesh Pollution Control Board (UPPCB). These sampling sites provide a summary of emissions driven by local traffic, residential, industrial, and institutional activities."

While in section 2.2 Instruments, first paragraph we have added the lines:

"At each of the three sites in the IGP, the Xact ambient metal monitor was employed to sample aerosols with a $PM_{2.5}$ cutoff cyclone. The sample system included a heater to maintain a relative humidity (RH) of 45% with a temperature-regulated laboratories at U-IGP1 and U-IGP2, while at C-IGP1 enclosure maintaining a temperature range of 20°C to 25°C was used at rooftop. Elaborate discussion of the sampling site and instrumentation can be found in our earlier literatures (Jain et al., 2022; Lalchandani et al., 2022; Manchanda et al., 2022; Shukla et al., 2023)."

#Comment 14

Line 218:Why don't you just write here that you use the XX-based PMF method, which seems to be a more mainstream term? I know there has been a lot of recent research based on a priori information, so please refer to a more specialized way of presenting it.

Thank you for the reviewer's comment. We agree that specifying the exact method used for source apportionment (SA) will add clarity. In the revised manuscript, we have mentioned the "element-based ME-2" model, which is widely recognized and aligns with recent research utilizing a priori information for source apportionment.

We have revised the lines 218:

"The element-based ME-2 model was applied in this study to perform source apportionment."

#Comment 15

Lines 232-234: So please describe what a priori information was included and what uncertainties, or key issues, were resolved.

Thank you for the reviewer's comment. We have provided a detailed explanation of the a priori information and its role in the factor selection process in Supplementary Section 3.1. In particular, we used constrained factor profiles for certain sources, such as fireworks and SFC1, by applying the a-value approach, which allowed us to refine factor identification and resolve mixing issues. For instance, the fireworks factor was constrained with an a-value of 0.2, while SFC1 was constrained at 0.1. These constraints were chosen based on explained variance, residuals diurnal patterns, and correlation with external tracers. This approach is used extensively in the source apportionment studies (Rai et al.,2021; Tobler et al.,2021; Kumar et al.,2022). Bootstrapping results further confirmed the stability of these solutions. A detailed discussion on selection of final solution is provided in the Supplementary Section 3.1.

We have added this information after line 234:

"Details on factor selection and constraints are discussed in Supplementary section S1."

The Supplementary section S1 is:

"The unconstrained Positive Matrix Factorization (PMF) was applied to the sub-hourly elemental dataset at all three sites, ranging from three to eleven factors. The initial step in the solution selection process involved determining the minimum number of factors for further investigation, focusing on the factor that exhibited the maximum change in the Q/Qexp value. Specifically, at U-IGP1, a 37.9% change was observed at factor nb 7, while at U-IGP2 and C-IGP1, the changes were 28.3% and 22.2%, respectively, also at factor nb 7, as depicted in graph S3. Consequently, the minimum number of factors for subsequent investigation was determined to be seven.

At the U-IGP1 site, the 7-factor solution successfully distinguished Pb-rich, Cu-rich, Cl-rich, and S-rich sources. However, SFC2 exhibited a mixture with Al (which displayed a strong correlation with K, Sr, and Ba, as shown in Fig.S1, elements associated with fireworks), and SFC1 was mixed with Si (which showed a good correlation with Ca and other dust-related *elements), as well as one Ca-rich dust factor. Upon transitioning to the 8-factor solution, an additional dust factor was resolved, but the S-rich factor remained mixed, similar to the 7-factor solution. To assess the stability of factor profiles, the 8-factor solution was executed with ten different seeds. It was discovered that the primary cause of mixing was the transition of Al and Si from one factor to another (Al was part of SFC1 in the 7-factor solution but appeared in the S-rich factor in the 8-factor solution). Therefore, the clean fireworks factor with Al was derived from the 10-factor solution and incorporated into the 8-factor solution. The fireworks factor profile was constrained with the a-value ranging from 0 to 1, in increments of 0.1. Ultimately, an a-value of 0.2 was determined to be the optimal solution. However, SFC1 still exhibited mixing, so its profile was constrained as well. Consequently, a solution with a=0.2 for fireworks and a=0.1 for SFC1 was selected as the environmentally reasonable solution based on the Understanding Explained Variance (UEV), physical interpretation of factors, time series, correlation with external tracers, and diurnal variation. The correlation of elements among themselves also helped in selection of factors as shown in Fig. S1. To evaluate the reproducibility of the 8-factor solution with a=0.2 for fireworks and a=0.1 for SFC1, ten seed runs were conducted. The solution was deemed optimal, with a variation in Q/Qexp of less than 2.3% at U-IGP1. Bootstrapping was performed on the final solution, and a detailed discussion of the results can be found in section S2.*

At the U-IGP2 site, in the 7-factor solution, the sulfur-rich component was mixed with aluminum, and SFC1 was also combined due to its contributions from crustal elements. Moving on to the 8-factor solution, the silicon from the dust factor was mixed with SFC1, as well as chromium, manganese, nickel, and iron, which were previously associated with dust. However, these elements were separated. Similar to the U-IGP1 site, the fireworks during Diwali caused issues. When the unconstrained 8-factor solution was run with 10 different initializations, elements such as aluminum, silicon, and other dust-related components could not be cleanly resolved and were getting mixed with other factor profiles. Consequently, to obtain a cleaner fireworks factor, it was constrained in the 8-factor solution. The fireworks profile was constrained using an 'a-value' ranging from 0 to 1, with a step size of 0.1. After considering factors such as the unexplained variance (UEV), physical interpretation, time series, correlation with external tracers, and diurnal fluctuations, the solution with an 'a-value' of 0.4 for fireworks was selected as the most environmentally appropriate. To assess the reproducibility of the 8-factor solution with an 'a-value' of 0.4 for fireworks, ten runs were performed with different random seeds. The solution was considered the best one when the variance in Q/Qexp was less than 0.01% at U-IGP2 (Fig. S8 (a)). Finally, bootstrapping was conducted on the final solution, and the detailed results are discussed in section S2.

At the C-IGP1 site, in the 7-factor solution, the following factors were resolved without any mixing: chlorine-rich, sulfur-rich, SFC1, SFC2, copper-rich, coal combustion, and dust. When moving to the 8-factor solution, the dust factor split into two components, one dominated by potassium, calcium, and iron, and the other dominated by aluminum and silicon. However, when observing the time series of the two dust factors, similar peaks were observed (Fig. S3 (b)), and there was a strong correlation between silicon and calcium as shown in Fig. 1(c), which did not support the separation of the two dust factors. To further confirm this, an attempt was made to constrain the high potassium signal in one of the dust factors, but no sufficient evidence was found to justify separate dust factors. After conducting a thorough investigation considering physical interpretability, diurnal patterns, time series, and residuals, it was determined that the seven-factor solution was the most environmentally viable option. To evaluate the reproducibility of the seven-factor solution, ten preliminary runs with random initializations were performed. The solution was considered optimal when the variation in Q/Qexp at C-IGP1 was less than 10.3% (Fig. S8 (a)). Bootstrapping was then applied to the final solution, and the detailed results are discussed in section S2.

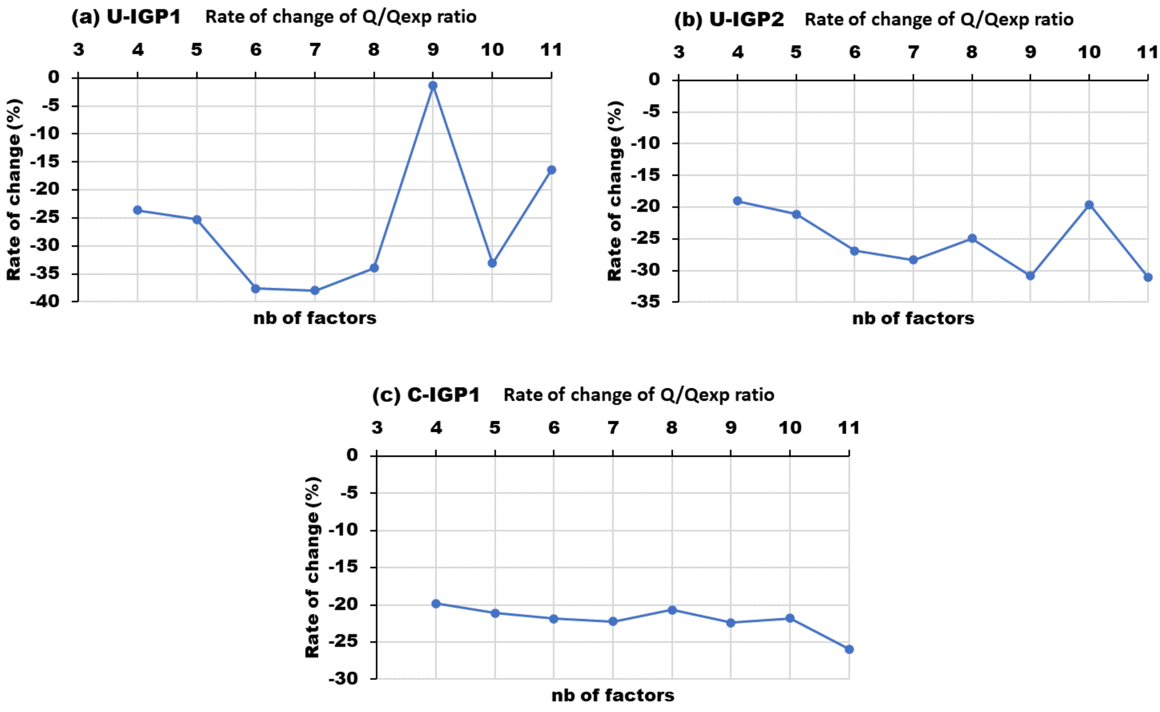


Figure S6. Relative percentage change in Q/Qexp ratio for the unconstrained PMF run from 3-factor to 11-factor solution at site (a) U-IGP1 (b) U-IGP2 and (c) C-IGP1.

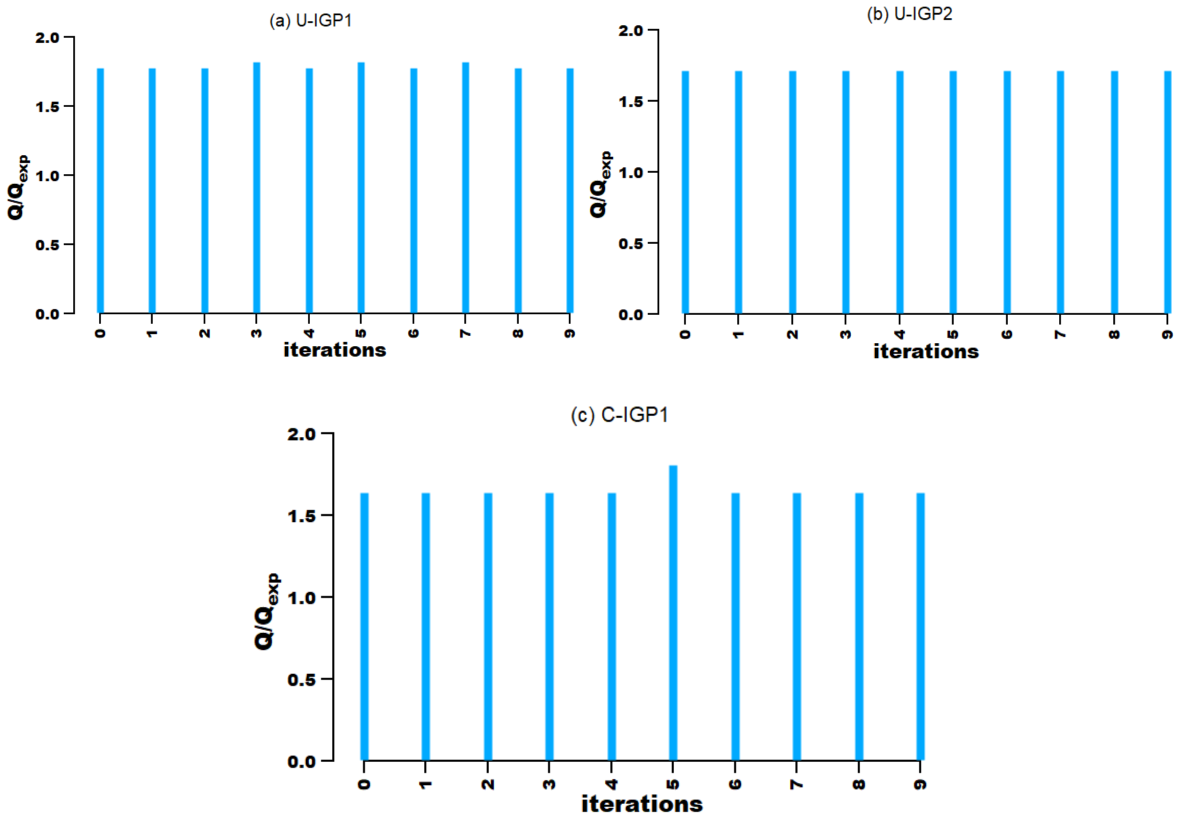
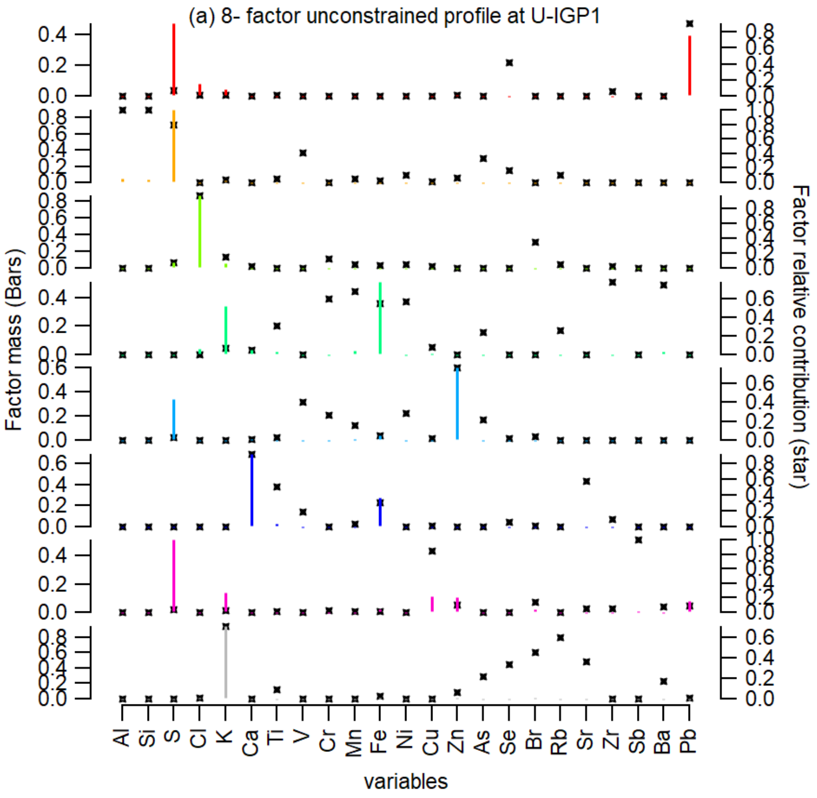
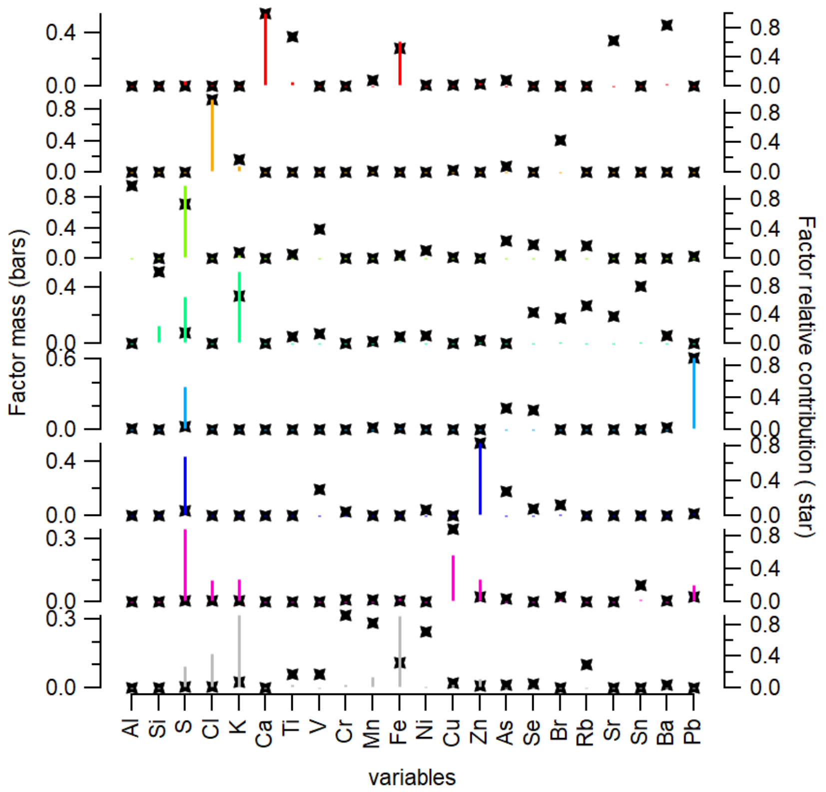
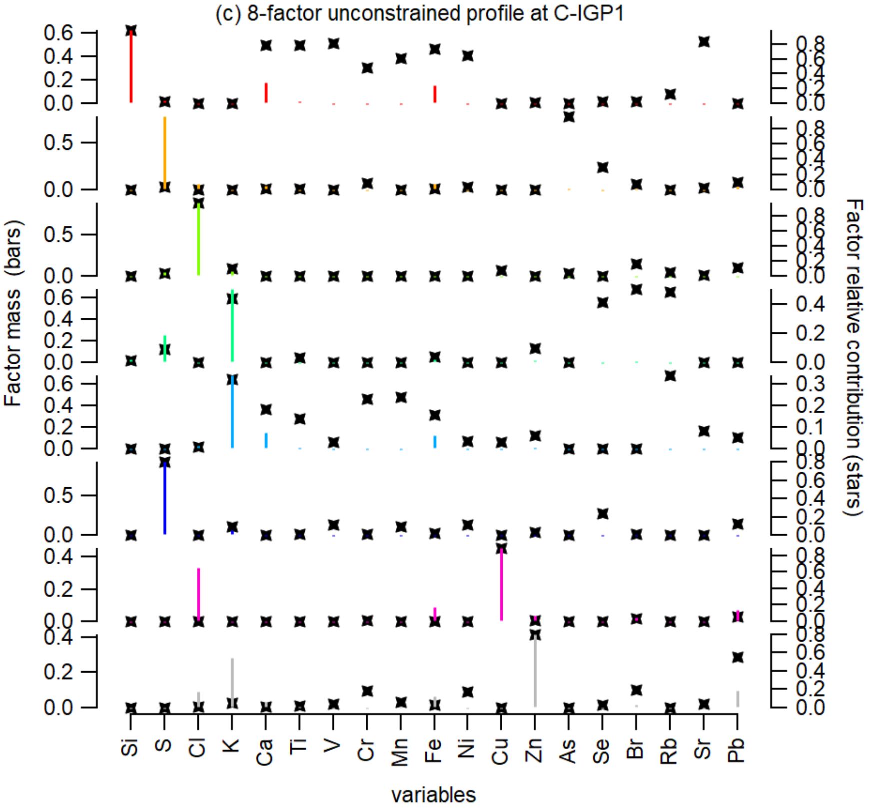


Figure S7. For elemental unconstrained PMF factors, the variation in Q/Qexp for 10 seeds (a) U-IGP1, (b) U-IGP2 and (c) C-IGP1.





(b) 8-factor unconstrained profile at U-IGP2



(c) 8-factor unconstrained profile at C-IGP1

Figure S8. The source profile of the 8-factor unconstrained solution used in the analysis while selecting the final number of factors at (a) U-IGP1, (b) U-IGP2 and (c) C-IGP1.

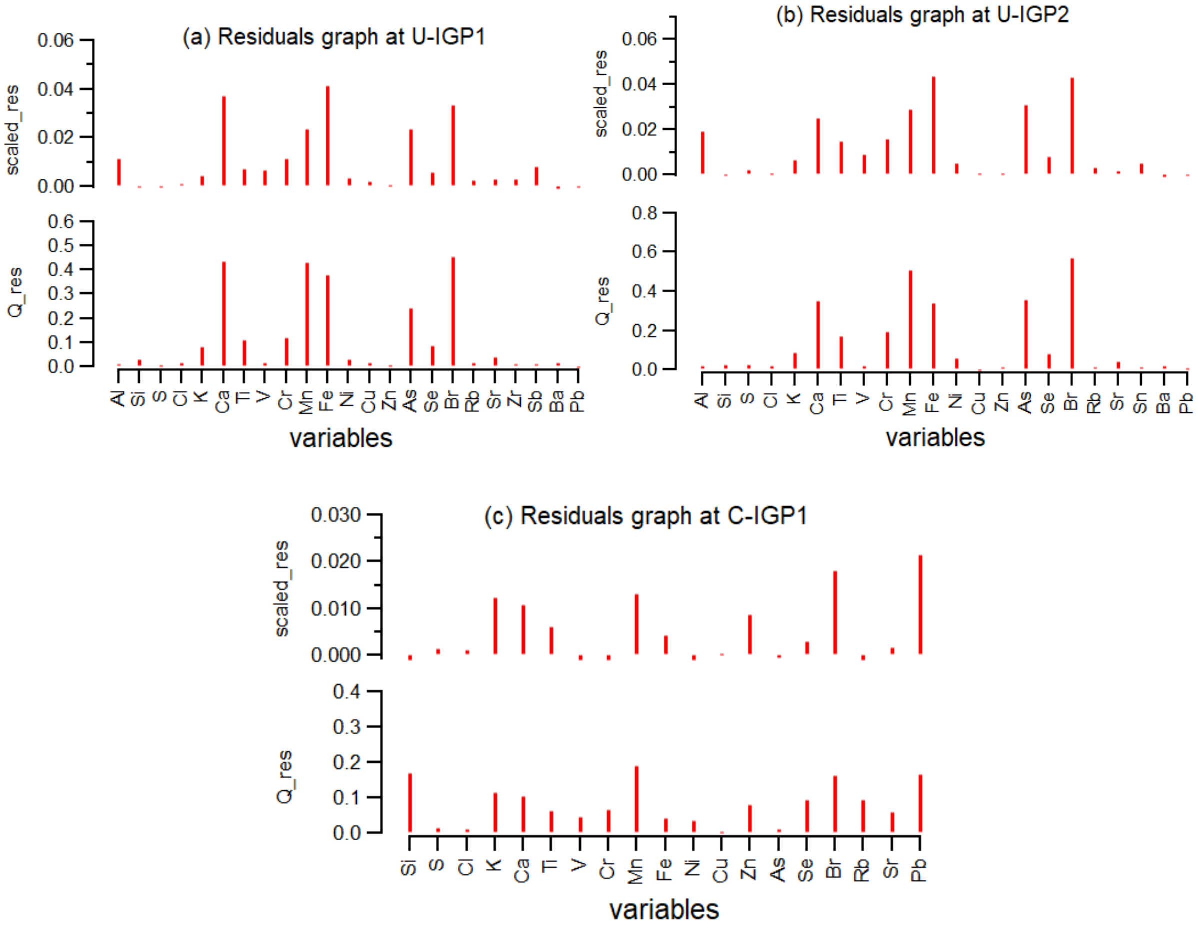


Figure S9. The variation of Q_reisdual and scaled_residual for each elemental species at (a) U-IGP1, (b) U-IGP2 and (c) C-IGP1 sites.

#Comment 16

Lines 329-334: Is it reasonable to need to consider COLD and WARM? Is it necessary to give the change in temperature (not Table 1) to testify to the reasonableness of the division, except for Figure 2c because there is a break in time, and both a and b are continuous? Or consider bringing up the junction date discussion.

We thank the reviewer for the comment. Apart from Table 1, we have included a revised Figure 5, which shows the diurnal variation of sources along with meteorological parameters, during cold and warm period. In the Figure 5, It was clearly observed 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. While, this study primarily focused on the C-IGP region, where high-resolution measurements were previously lacking, as noted in the introduction. During the source apportionment analysis of the C-IGP data, we found that the resolved factor profiles were quite similar to those obtained in U-IGP (Supplementary Figs. S10a, S11a, S12a), despite variations in measurement periods and emission sources. This prompted us to expand our investigation to the U-IGP region. Just to note that, the results from the U-IGP data was used for external correlation only with the combined PMF results from Shukla et al. (2023), while 10 days of Diwali data were analyzed by Manchanda et al. to study the festival's effects. Subsequently, we utilized the full U-IGP dataset, which revealed distinct differences between the warm and cold periods, influenced by changes in emission sources and meteorological conditions between these two IGP regions. These findings align with previous research, such as Rai et al. (2021), which focused on U-IGP but used a limited dataset. Our three-month dataset from both U-IGP and C-IGP, however, allowed us to draw more comprehensive conclusions about elemental sources and meteorological impacts across both regions.

We have revised the line 329-334:

“The elemental data was subdivided into warm and cold periods to analyze the variations in meteorological factors, emission sources, and probable formation mechanisms, particularly sulfur and chlorine (Supplementary Fig. S3). This subdivision was based on the distinct temporal changes observed in the resolved factors across both U-IGP and C-IGP sites. Despite differences in IGP regions, emission sources and meteorological conditions, similar factor profiles were identified at both sites (Supplementary Figs. S10a, S11a, S12a). This provided a basis for a detailed examination of how meteorology and source contributions are varying between the two periods, providing deeper insights into regional source characteristics. The summarized results in Table S2 further demonstrates the importance of this temporal division for interpreting elemental concentrations.”

The Figure showing the variation of elemental sources with meteorology is:

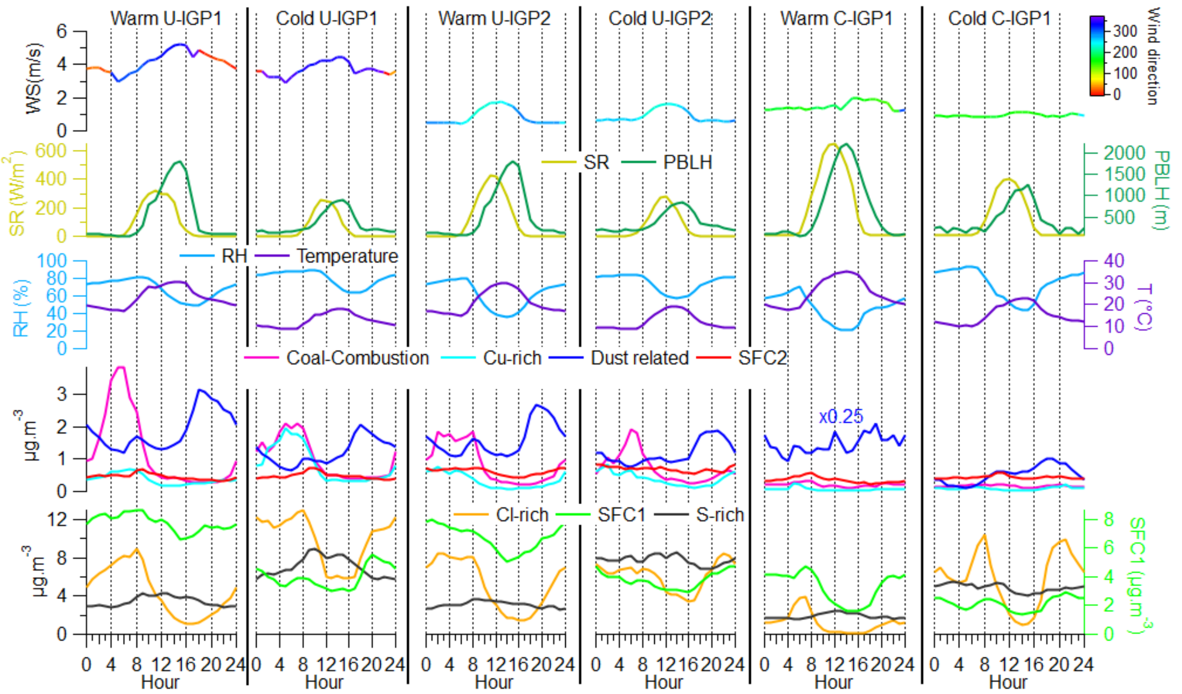


Figure 5. Diurnal variation of elemental sources and meteorological parameters (SR, PBLH, WS, WD, RH and T) at the three sites in the IGP region; U-IGP1, U-IGP2 and C-IGP1 during the warm and cold periods.

#Comment 17

Line 351: Why is Si, a typical crustal element, relevant to combustion?

We appreciate the reviewer's comment and acknowledge the confusion regarding the inclusion of Si in relation to combustion sources. The sentence originally intended to provide a statistical update on the contribution percentage after including silica. However, the primary focus was on S, Cl, and K as combustion-related elements. To avoid any further confusion, we have revised the sentence and removed the reference to Si in relation to combustion.

We have revised the line 351:

"In the warm period, these elements contributed over 73% in the U-IGP region and 52% in the C-IGP region."

#Comment 18

Line 356: Is there strict evidence of favorable meteorological conditions? Supporting evidence on meteorological conditions in the text also needs attention.

We thank the reviewer for pointing this out. Yes, there is clear evidence supporting the role of meteorological conditions during the cold period, which are illustrated in Figure 5 and

Supplementary Figure S3. We have revised the diurnal figure 5, which clearly shows the effect of meteorology in the variation of the diurnal concentrations of the elemental sources. These favorable conditions include higher relative humidity (RH), lower temperature, reduced planetary boundary layer (PBL) height, and decreased wind speed, all of which contribute to both the formation and accumulation of pollutants such as sulfur (S) and chlorine (Cl). Higher RH facilitates secondary aerosol formation, particularly through heterogeneous reactions on aerosol surfaces (Seinfeld & Pandis, 2006). Lower temperatures reduce the volatility of semi-volatile compounds, leading to increased partitioning into the particulate phase (Pathak et al., 2011). The reduced PBLH during winter limits vertical mixing, trapping pollutants closer to the surface and increasing their concentrations (Zhang et al., 2014). Furthermore, lower wind speeds hinder the dispersion of pollutants, leading to their accumulation in the atmosphere (Wang et al., 2014). All these factors combine to create an environment that favors the formation and persistence of elevated concentrations of S and Cl during the cold period.

We have revised the line 356:

"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)."

The Figure showing the variation of elemental sources with meteorology is:

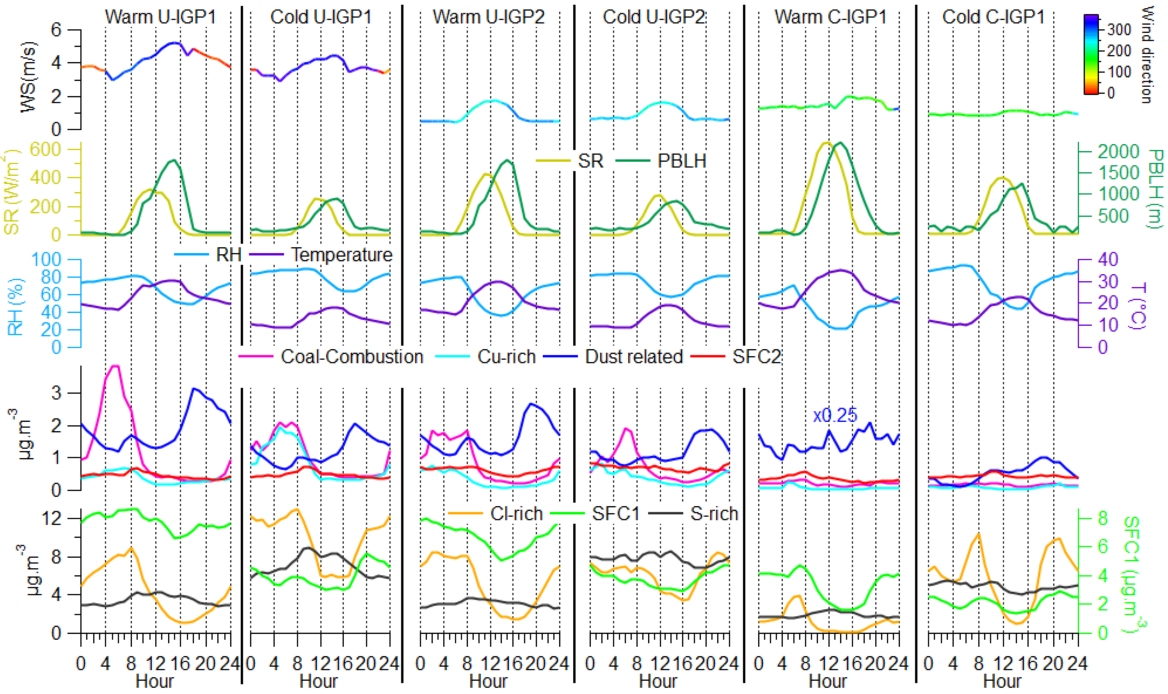


Figure 5. Diurnal variation of elemental sources and meteorological parameters (SR, PBLH, WS, WD, RH and T) at the three sites in the IGP region; U-IGP1, U-IGP2 and C-IGP1 during the warm and cold periods.

#Comment 19

Lines 371-373: What does EF have to do with the source?

We thank the reviewer for the insightful comment. While we recognize that source apportionment using PMF is a robust and widely accepted technique for identifying pollution sources, the inclusion of the enrichment factor (EF) serves as an additional step to assess the elemental data preliminarily. EF helps provide context by offering a quantitative measure of whether certain elements are enriched relative to crustal abundances. This distinction is crucial for elements such as Al, Si, and Fe, which are strongly tied to crustal sources, compared to those like S, Cl, and Pb, which are significantly enriched by anthropogenic activities. While the PMF analysis remains the primary method of source attribution in this study, the EF helps validate and support these findings by identifying general trends in elemental enrichment, particularly in cases where source resolution through PMF may be less distinct. By integrating EF with PMF results, we can cross-check the consistency of source categorization, particularly for elements influenced by both natural and anthropogenic processes.

We have revised the lines 367-377 incorporating this and next comment:

“Using Ti as a reference element (Fomba et al., 2013; Majewski & Rogula-Kozłowska, 2016), the enrichment factor (EF) was computed for each measured element to assess the effect of anthropogenic emissions on atmospheric elemental levels. The EF helps to evaluate the role of human activities by acting as a quantitative indicator of how much some elements are raised above their native crustal levels. Although source apportionment is covered in the next section, the EF offers a initial estimate of crustal versus anthropogenic impacts and also used to support the source apportionment results. Al, Si, Fe, and Zr displayed EF values between 0.4 and 2, indicating a high correlation with crustal sources across the IGP areas and time periods (Fig. S14). By contrast, K, Cr, Mn, and Ni displayed EF values ranging from 2 to 20, implying a mix of crustal and anthropogenic inputs. Particularly from industrial and combustion activities. S, Cl, Cu, Zn, As, Se, Br, Cd, Sn, Sb, and Pb showed EF values surpassing 20, suggesting major anthropogenic influences. Although EF emphasizes anthropogenic causes, climatic aspects also affect the transit and deposition of these elements; so, this interaction will be discussed in the section on source apportionment result. Supplementary section S4 offers further thorough treatment on EF of elements."

#Comment 20

Lines 377-378: I think if EF is relevant to the source, or how to reveal enhancement, it is necessary to put it in the text. Besides, considering meteorological effects, how can the authors define it only as an effect of anthropogenic activities?

We appreciate the reviewer's comment regarding the need for clarification on the relevance of the enrichment factor (EF) in relation to source identification and the effects of meteorology. The EF is a valuable tool for distinguishing between crustal and anthropogenic contributions to elemental concentrations. The use of EF allows us to quantitatively assess how much certain elements are enriched relative to their crustal abundances, providing insight into the potential enhancement due to human activities. So, since EF is a relative measure, it should not be affected by meteorology. This approach complements the source apportionment performed through PMF by establishing a baseline understanding of elemental enrichment, which can then be further explored in the context of prevailing meteorological conditions.

We have revised the lines 367-377 incorporating this and previous comment:

"Using Ti as a reference element (Majewski & Rogula-Kozłowska, 2016; Fomba et al., 2013), the enrichment factor (EF) was computed for each measured element to assess the effect of anthropogenic emissions on atmospheric elemental levels. The EF helps to evaluate the role of human activities by acting as a quantitative indicator of how much some elements are raised above their native crustal levels. Although source apportionment is covered in the next section, the EF offers a initial estimate of crustal versus anthropogenic impacts and also used to support the source apportionment results. Al, Si, Fe, and Zr displayed EF values between 0.4 and 2, indicating a high correlation with crustal sources across the IGP areas and time periods

(Fig. S14). By contrast, K, Cr, Mn, and Ni displayed EF values ranging from 2 to 20, implying a mix of crustal and anthropogenic inputs. Particularly from industrial and combustion activities. S, Cl, Cu, Zn, As, Se, Br, Cd, Sn, Sb, and Pb showed EF values surpassing 20, suggesting major anthropogenic influences. Although EF emphasizes anthropogenic causes, climatic aspects also affect the transit and deposition of these elements; so, this interaction will be discussed in the section on source apportionment result. Supplementary section S4 offers further thorough treatment on EF of elements."

#Comment 21

Lines 378-388: How do authors consider health effects? Especially other elements not mentioned here. I.e., how do the authors synthesize elements with high concentrations but weak health effects, and elements with low concentrations but strong effects? Or looking back is the representation in 3.2 appropriate?

We appreciate the reviewer's question regarding the selection of elements for health effects assessment. Our decision to focus on lead (Pb), nickel (Ni), arsenic (As), and chromium (Cr) was guided by the availability of inhalation reference concentrations (RfCs) from the US EPA, which specifically highlight these elements as significant due to their carcinogenic potential (USEPA, 2020). These RfCs provide clear regulatory benchmarks for evaluating potential health risks. Additionally, Rai et al. (2021) conducted a similar analysis in the U-IGP, focusing on these four metals, which gave us an opportunity to compare our findings across the U-IGP and C-IGP sites. By doing so, we aimed to observe how these carcinogenic metals vary not only within the U-IGP but also in the understudied C-IGP region. Thus, this approach allowed us to contextualize our results within existing health guidelines and contribute to a better understanding of regional variations in elemental concentrations.

We have added few lines before line 377:

" 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."

#Comment 22

Lines 411-412: Cl-rich is not a source, just a phenomenon. Here's hoping the authors can parse to the source of the emissions.

We appreciate the reviewer's comment regarding the "Cl-rich" factor. To clarify, the "Cl-rich" label refers to a combination of possible sources, predominantly from the steel industry and trash burning (particularly PVC), along with contributions from other sources such as brick kilns. We chose the term "Cl-rich" to describe this factor based on the dominant presence of chlorine in its profile, as detailed in Supplementary Section S2. This section outlines the potential sources contributing to this factor, supported by the findings of Rai et al. (2021), Ding et al. (2020) and Jin et al. (2020), which link chlorine-enriched emissions to combustion processes and metallurgical activities. The correlation of Cl-rich with PAHs further supports its association with incomplete combustion, particularly from waste incineration and plastic/PVC materials. We have revised the lines and mentioned in the bracket their possible sources. We have also included a brief summary of the source profiles in main text while the detailed discussion is provided in supplementary section S2.

The revised Lines 411-412 and the additional brief summary of the factors are:

"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 CO_2 (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 (Kolenčík 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-smeleting 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 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 & 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 23

What is the difference between SFC1 and SFC2? What sources are characterized respectively?

We appreciate the reviewer’s comment on the distinction between SFC1 and SFC2. While the detailed discussion on these factors is provided in Supplementary Section S2, we have now added a summary in the main text to further clarify their sources. SFC1 is associated with biomass burning, primarily crop residue burning, as indicated by high potassium levels and correlation with PAHs. SFC2, on the other hand, is linked to industrial waste burning, characterized by high contributions of zinc, chromium, and nickel. Both factors represent different combustion activities, highlighting the diverse emission sources in the region.

We have added an additional paragraph, which briefly summarise the sources in the main text section 3.3, 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 (Kolenčík 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-smeleting 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 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 & 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 24

Figures 4 and 5: I don't see much point in doing diurnal differences. Because the source contributions don't look like much of a percentage change, except for a change in concentration. The authors need to reconsider integrating the two graphs.

We thank the reviewer for the suggestion and have integrated the two graphs. The diurnal differences were studied because variations were observed between day and night average concentrations of sources, such as Cl-rich, which is semi-volatile and has a higher average concentration at night than during the day (Fig. 5). In addition, the S-rich source revealed a higher concentration during the day than at night. These observations also assisted us in analyzing the difference between diurnal variations, where we can see distinct changes between different sources, even if the contribution was consistent throughout the day and night, such as a distinct diurnal pattern in Cl-rich variation between warm and cold periods. During the warm period, Cl-rich concentrations began to climb around 8:00 PM, but during the cold period, they began to rise around 4:00 PM, most likely due to climatic changes. Figures 4 and 5 depict the fluctuation in absolute concentrations; the top part of the graphs represents the primary sources, while the bottom part represents the minor sources. However, in response to the reviewers' comments, we integrated Figures 4 and 5 to represent both the warm and cold periods.

The Figure 4 (Fig.4 and Fig. 5 integrated) is:

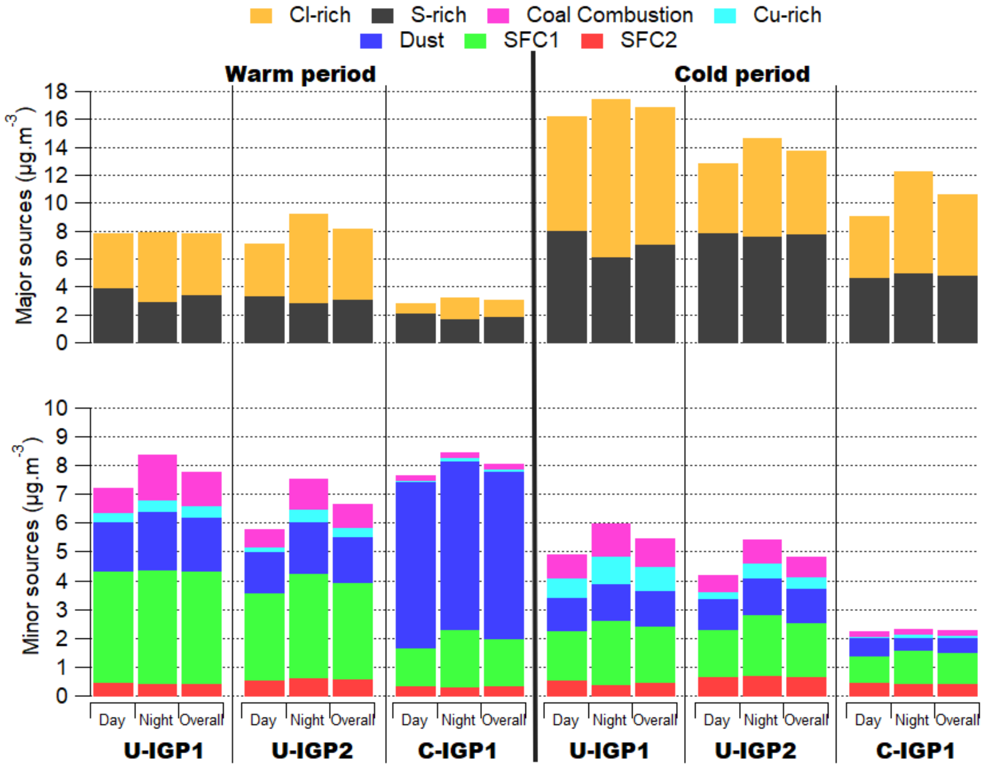


Figure 4. Variation in composition (µg.m-3) of elemental sources at the three sites in the IGP region; U-IGP1, U-IGP2 and C-IGP1 between day, night and overall period during Warm and Cold period.

#Comment 25

Lines 462-465: It is not very meaningful to discuss concentration variations because the boundary layer itself has daily variations, as in Table 1. What needs to be considered is the contribution, or percentage, to the total PM.

We thank the reviewer for pointing out regarding discussion of relative contribution rather than concentration. We have added the discussion as well as a figure in the supplementary section to highlight the variation in the relative contribution of the elements especially between day and night for both warm and cold period.

We have added the relative contribution discussion after the line and also the other discussions related to relative contributions are discussed at suitable places in the manuscript:

“From day to night, significant changes in relative source contributions are observed during both warm and cold periods across the different IGP regions as shown in Supplementary Fig. S16. The S-rich source showed higher cold-daytime contribution (~40%) as compared to the warm-daytime contribution (~24%). The Cl-rich source showed significant increase at nighttime (50%) as compared to the daytime (39%) at the U-IGP1 and C-IGP1 site. The relative contribution of other sources was almost consistent as shown in Fig. S16.”

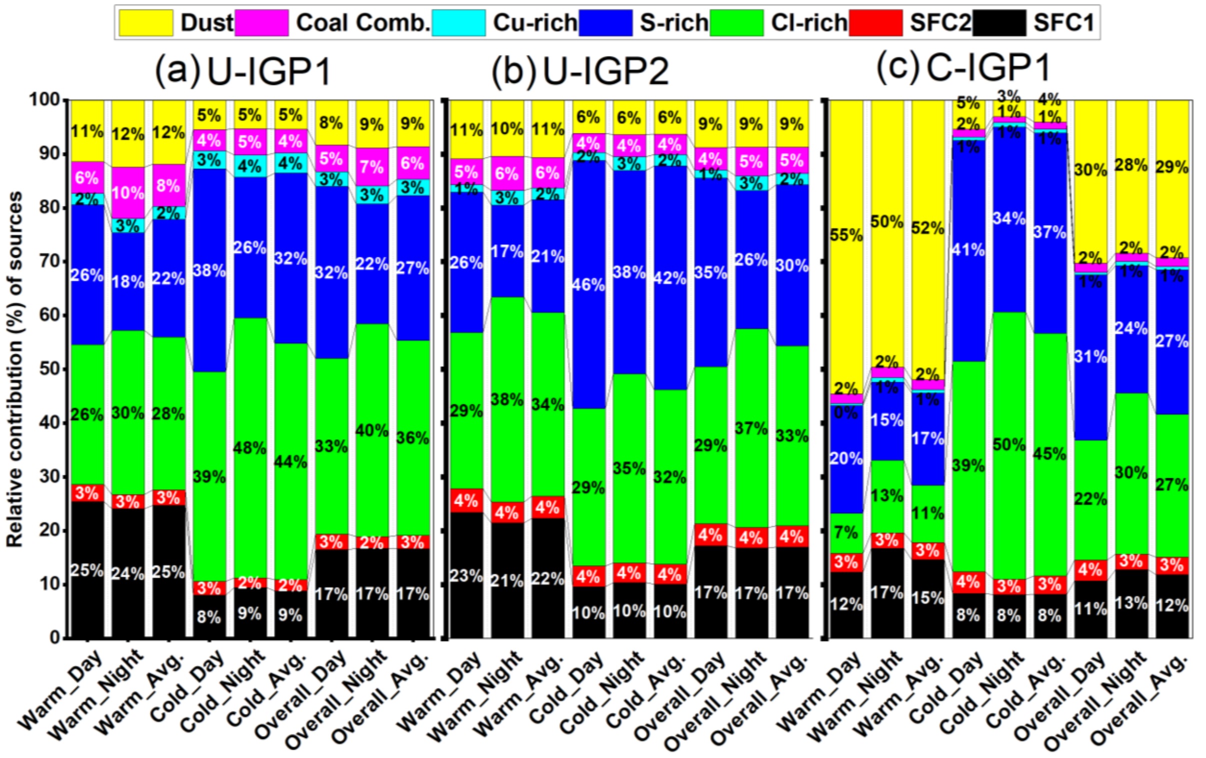


Figure S16. Variation in RC (%) of elemental sources at the three sites in the IGP region; U-IGP1, U-IGP2 and C-IGP1 between day, night and overall period during Cold and Warm period.

#Comment 26

Figure 6: The vertical coordinate of this figure is strange, if it is µg.m-3, the contribution of individual sources to the mass concentration does not correspond to Figures 4-5. In addition, uncertainty ranges for daily changes need to be given.

We apologize for the confusion regarding Figure 6. The diurnal variation in this figure was normalized to its peak concentration to better illustrate the differences between the three sites across the two IGP regions. However, we have changed the diurnal Figure 5 (Fig. 6 number changed due to the integration of Fig. 4 and Fig. 5) from normalised concentration to show the variation of elemental sources with meteorology during warm and cold period in the IGP regions. Additionally, we have included uncertainty ranges for daily changes in the supplementary Figure S15.

The Revised Figures are:

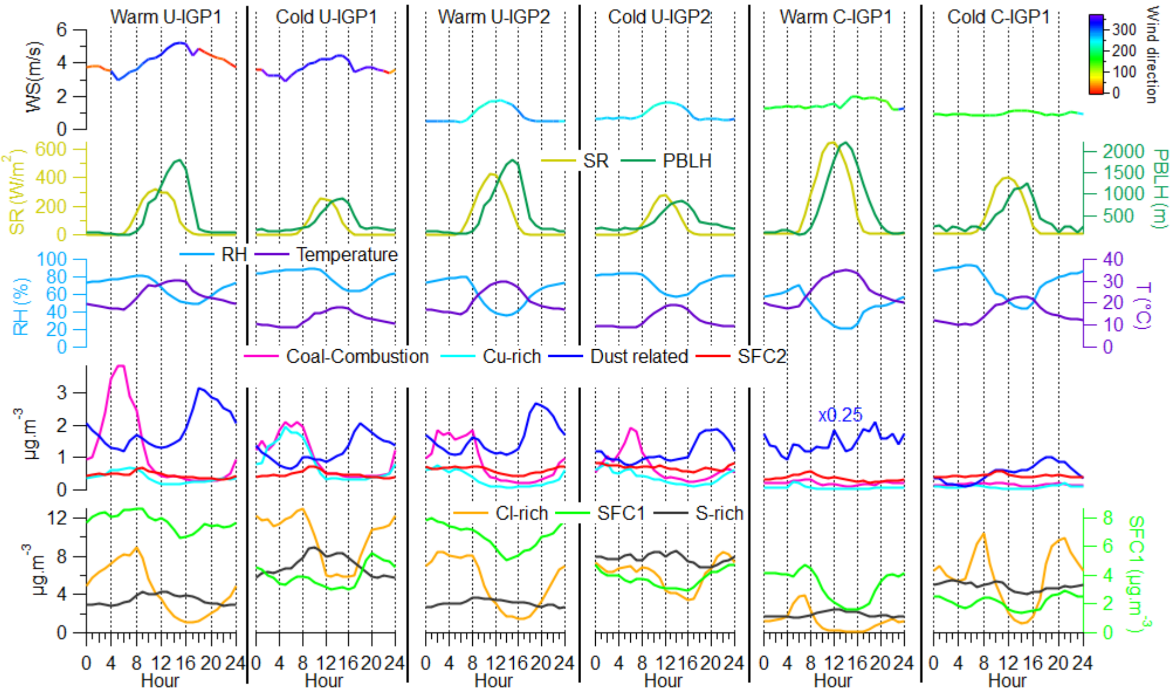


Figure 5. Diurnal variation of elemental sources and meteorological parameters (SR, PBLH, WS, WD, RH and T) at the three sites in the IGP region; U-IGP1, U-IGP2 and C-IGP1 during the warm and cold periods.

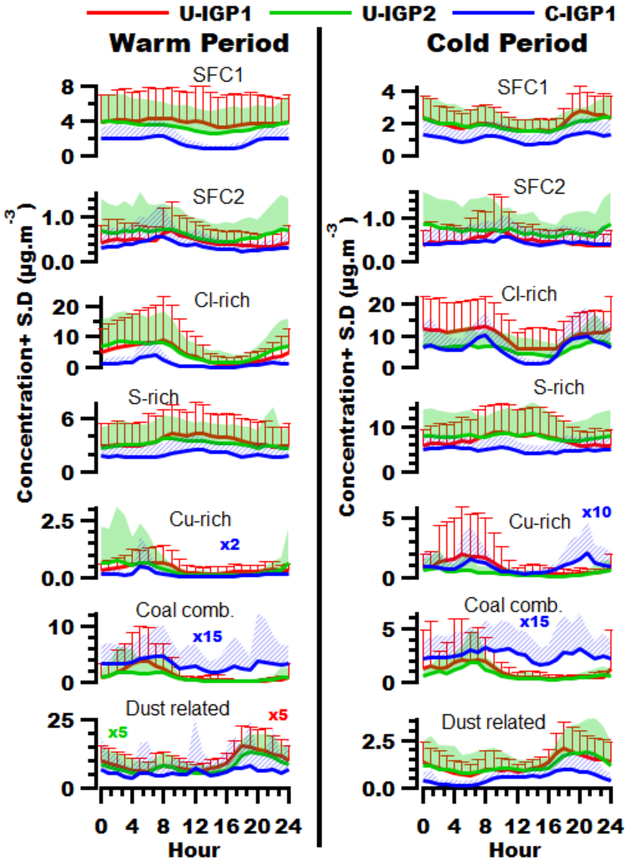


Figure S15. Diurnal variation (Avg Conc+S.D (Bar for U-IGP1,shade for U-IGP2 and slanted line for C-IGP1)) of elemental sources at the three sites in the IGP region; U-IGP1, U-IGP2 and C-IGP1 during the warm and cold periods.

#Comment 27

Section 3.4: I think there is a need to highlight the core innovative points or findings about METEOROLOGY. The authors didn't find out what the real meteorological drivers of PM changes in the cold/warm seasons are, and too much reporting on the rise and fall of PM will only increase the time spent reading the literature, making the real innovation of this section obscured.

Thank you for your insightful comments regarding Section 3.4. We appreciate your feedback and have made changes to better highlight the core findings related to meteorological influences on pollution episodes.

We have revised Section 3.4 to reduce repetitive descriptions of the rise and fall of PM$_{2.5}$ concentrations and focused more on the meteorological drivers and their specific impact on elemental sources. Specifically, we have streamlined the discussion to avoid redundant mentions of concentration changes and instead highlighted the mechanisms behind these variations. Additionally, we have incorporated new scientific insights and emphasized the novel findings derived from our observations. Notably, we clarified how cold-period conditions, including low temperatures, high RH, and reduced PBL height, led to the increased contribution of Cl-rich and S-rich sources. This highlights the significance of accumulation and secondary aerosol formation under specific meteorological conditions. In contrast, dust resuspension was more pronounced during warm periods, driven by drier conditions and increased wind speeds. We have also highlighted the enhanced contribution of S-rich sources during nighttime in the cold periods, attributing this to aqueous-phase oxidation facilitated by elevated RH, a process that is less active during the warm periods when photochemical oxidation dominates. This understanding of how meteorology influences source contributions provide a critical perspective on air quality dynamics in the IGP region. We hope the revised section 3.4 now presents a clearer and more insightful analysis; we have also revised the conclusions based on the revised section 3.4.

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). PM2.5 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).

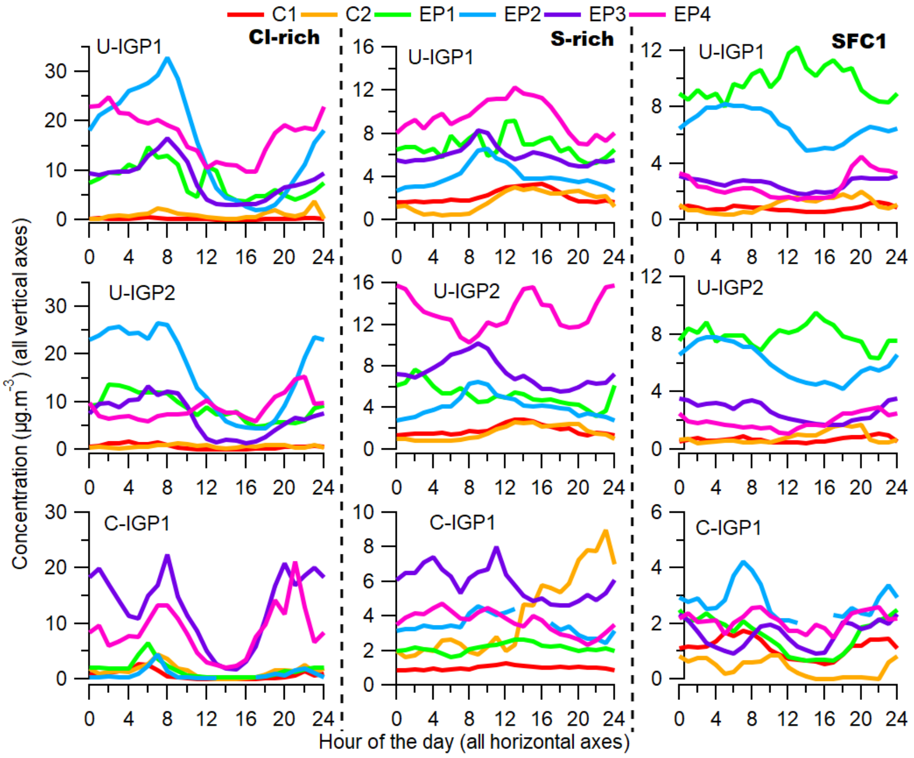


Figure 8. The diurnal variation of Cl-rich, S-rich and SFC1 sources having major role in the variation of El-$PM_{2.5}$ during the clean and polluted episodes C1, EP1, EP2, C2, EP3 and EP4 during both warm and cold period.

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 El-$PM_{2.5}$ 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 section 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 PM2.5 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."

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