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Ensemble source apportionment of air pollutants and carbon dioxide based on online measurements

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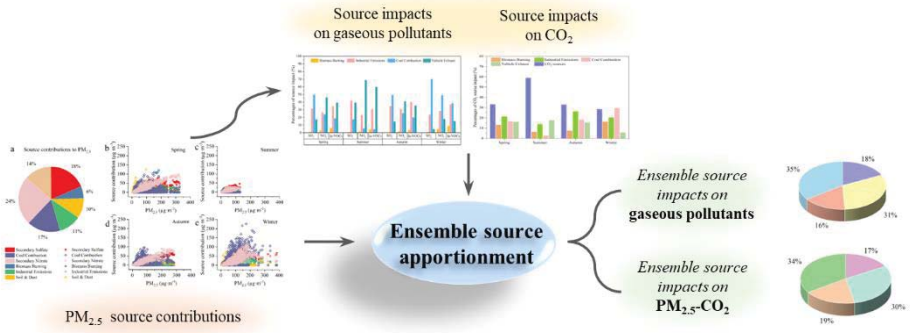
Abstract

Air pollution and climate change have attracted worldwide attention due to their significant threats to human health and the environment. To maximize the co-benefits of clean air policies on greenhouse gas emissions and vice versa, it is imperative to coordinate emission control measures for air pollutants and CO_2. To do this, we first need to better quantify the impacts of different sources to air pollutants and CO_2 at once. Based on a 2-year observation of fine particulate matter ($PM_{2.5}$), gaseous pollutants (including sulfur dioxide (SO_2), nitrogen oxides (NO_x), initial volatile organic compounds (In-VOCs)), as well as carbon dioxide (CO_2), we apportioned the sources of criteria air pollutants and CO_2 simultaneously. We then developed a new source apportionment method to quantitatively determine the relative impacts of different emission sources to (1) a gaseous pollutant ensemble (SO_2-NO_x-In-VOCs) and (2) $PM_{2.5}$ and CO_2 ensemble ($PM_{2.5}$-CO_2). The results demonstrate that vehicle exhaust (35%), industrial emissions (31%), biomass burning (18%), and coal combustion (16%) were the main control targets for the SO_2-NO_x-In-VOCs ensemble, and they were also the dominant contributors to $PM_{2.5}$-CO_2 ensemble with similar contribution rates. Not surprisingly, the source impacts are substantially different for the two studied ensembles to those of individual air pollutant alone. This study provides a new source apportionment method to deliver scientific evidence for developing a coordinated strategy to maximize the benefits of clean air and carbon policies to air quality and the climate.

Keywords

Ensemble source apportionment; Particulate matter; Gaseous pollutants; Carbon dioxide; Coordinated control

Graphical abstract



Highlights

- Concentrations and sources of $PM_{2.5}$, gaseous pollutants and CO_2 were analyzed.
- Developed weighted average method that can quantify ensemble source impacts.
- Reduce combustion source emissions benefit control of gaseous pollutants and $PM_{2.5}$-CO_2.

Manuscript words: 8498（including the whole text file, table and figure captions, and references)

1. Introduction

Air pollution and climate change substantially threaten public health and the environment (Chen et al., 2020; Schnell and Prather, 2017), which is commonly attributed to the increase in anthropogenic source emissions caused by rapid economic development (Zheng et al., 2015). As a major air pollutant, fine particulate matter ($PM_{2.5}$) has detrimental impacts on the cardiovascular, immune and nervous systems, increases morbidity and mortality, and causes massive disruption to economic activity (Aguilera et al., 2021; Shi, Y. et al., 2018; Wang, Y. et al., 2021). To reduce $PM_{2.5}$ pollution, China implemented an action plan, the Air Pollution Prevention and Control Action Plan in 2013 (http://www.gov.cn/zwgk/2013-09/12/content_2486773.htm) and launched the Three-Year Action Plan for Winning the Blue Sky War in 2018 (http://www.gov.cn/zhengce/content/2018-07/03/content_5303158.htm). Although $PM_{2.5}$ concentrations have significantly decreased by 30-40% (Li, K. et al., 2019; Xue et al., 2019; Zhai et al., 2019; Zhang et al., 2019), a large enhancement in secondary components was reported for Northern China due to the formation of sulfates, nitrates, ammonium and secondary organic aerosols (SOA) through nonlinear photochemistry of gaseous precursors (sulfur dioxide (SO_2), nitrogen oxides (NO_x) and volatile organic compounds (VOCs)) (Qu et al., 2021; Wang, B. et al., 2021). Secondary aerosols typically contribute more than half of the total $PM_{2.5}$ mass in China (Huang et al., 2014; Zheng et al., 2016), thus, variations of $PM_{2.5}$ can be largely driven by the changes in secondary aerosols.

In addition, climate change has been demonstrated to adversely affect human health,

including a rise in vector-borne diseases, increased heat-related morbidity and mortality, and potential negative impacts on water resources and crop production (Hayes and Poland, 2018; Piao et al., 2010; Watts et al., 2015), which may affect human habitability in the future (Kang and Eltahir, 2018; Pal and Eltahir, 2016). Since the industrial revolution, the increasing population, intensive agricultural activities and the use of energy (such as fossil fuels) have brought unprecedented climate warming (Scheutz et al., 2009). Carbon dioxide (CO_2), a typical greenhouse gas (Kramer et al., 1999), is released into the atmosphere and forces the climate out of balance by interfering with the earth's natural carbon cycle (Rastogi et al., 2002). Studies have shown that the global average CO_2 concentration in the 21st century has exceeded that of the industrial age (1750 AD, 278 ppm) by nearly 45% (Liu et al., 2021). Although CO_2 is not considered an air pollutant, it has become a research hotspot due to its effect on the climate. Simulation studies showed that low-carbon policies oriented toward carbon emission reduction have co-benefits that improve air quality (Shi et al., 2021). Therefore, CO_2 emission reduction is urgently needed.

It is worth noting that climate change, particulate matter and gaseous pollutants are intricately linked. Increased CO_2 in the atmosphere creates a greenhouse effect by absorbing solar radiation reflected from the earth's surface and also releases long-wave radiation, causing global warming. Climate change leads to changes in temperature, radiation, precipitation, wind speed and other meteorological factors that affect the generation, accumulation and diffusion of pollutants (Cai et al., 2017; Zou et al., 2017). Moreover, pollutants (particularly aerosols) affected by the photochemical generation

of gaseous precursors can further affect the climate system by altering the atmospheric radiation budget and influencing cloud formation (Zhao et al., 2017). Therefore, a set of collaborative approaches to carbon dioxide, particulate matter and gaseous pollutants reduction need to be developed.

Air pollution has the similar root and source as greenhouse gases. Human activities can not only emit CO_2 but also simultaneously emit air pollutants such as by $PM_{2.5}$, SO_2, NO_x, and VOCs (Shi et al., 2021). Although many recent publications have reported on the source contributions of the aforementioned pollutants (Li et al., 2020; Sun et al., 2020; Yang et al., 2021), only general conclusions can be drawn about the importance of individual pollutants. So far, there is still a lack of source apportionment technology to directly quantify the synergistic effects of common sources of multiple pollutants using detailed $PM_{2.5}$, SO_2, NO_x, VOCs and CO_2 concentration data. Controlling emissions sources of pollution is an effective way to fulfill coordinated emission reduction of greenhouse gas and air pollution. In this study, we make use of comprehensive observations from June 1, 2017 to May 6, 2019 at Tianjin, China, to quantitatively estimate the source impacts of particulate matter, gaseous precursors (SO_2, NO_x, VOCs) and CO_2, respectively. Based on analytical results of individual species, an ensemble source apportionment technique for coordinated source traceability of multiple air pollutants and greenhouse gas is developed. Ultimately, the purpose is to identify common sources across different seasons to achieve collaborative carbon emission reduction and clean air goals. Our research will provide implications for China and other countries to formulate coordinated air pollutant-greenhouse gas

control strategies in the future.

2. Material and methods

2.1. Site description

The campaign was carried out based on annual sampling (from June 1st 2017 to May 6th 2019) at the air quality supersite (117°24′N, 38°59′E) in Nankai University of Tianjin, a megacity in Northern China, which is located in a typical suburban residential area and surrounded by agricultural land. The location of the sampling site is shown in Fig. S1. About 1.5 km southwest and 2.5 km northwest of the site are major roadways with dense automobile traffic. To the northwest of the sampling site, there are industrial parks specializing in manufacturing and chemical industries. Therefore, the sampling point is mostly affected by local air pollution sources.

2.2. Measurement and analysis

In order to understand the common sources of air pollutants and greenhouse gas, multiple instruments were deployed in the campaign. $PM_{2.5}$ mass concentrations were collected using the online beta attenuation particle monitor (Focused Photonies Inc., BPM-200, China). Elements (Ca, K, Pb, Cr, Cd, Zn, Cu, Ni, Fe, Mn, Ti, etc), key components in $PM_{2.5}$, were measured by an online X-ray fluorescence (Focused Photonies Inc., AMMS-100, China). Water-soluble ions in $PM_{2.5}$ (Na^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, F^-, NO_3^-, SO_4^{2-}, and NH_4^+) were recorded using an online ion chromatograph

(Thermo Fisher Scientific Inc., URG 9000D, USA), with cation detection using 20 mM methanesulfonic acid, and anion analysis using 0.08 mM Na_2CO_3/0.01 mM $NaHCO_3$. Organic carbon (OC) and element carbon (EC) of $PM_{2.5}$ were carried out by a thermal-optical carbon analyzer (Focused Photonies Inc., OCEC-100, China). Concentrations of CO_2, carbon monoxide (CO), nitrogen oxides (NO_x = NO + NO_2), and gaseous pollutants, including SO_2 and hydrogen sulfide (H_2S) were detected by online instruments (API Inc. T360, T300, T201, T101, USA). Volatile organic compounds (VOCs) were measured using a set of GC955-611/811 series instruments (Syntech Spectras Inc., Holland). The time resolutions of the measurements collected for this study are summarized in Table S1.

2.3. Source apportionment

Factor analysis models, such as the positive matrix factorization (PMF), are statistical methods that use the chemical composition data at the receptor to obtain source profile and source contribution (equation (1)) (Paatero, 1997; Paatero and Tapper, 1994).

$$x_{ij}=\sum_{k=1}^{p} g_{ik}f_{kj}+e_{ij} \quad (1)$$

where, x_{ij} (μg·m^{-3}) is the concentration of the *j*th species in *i*th sample, g_{ik} (μg·m^{-3}) is the contribution of the *k*th source to the *i*th sample, f_{kj} (μg·m^{-3}/μg·m^{-3}) is the fraction of the *j*th species from the *k*th source, e_{ij} (μg·m^{-3}) are the residuals, and *p* is the number of factors. In the process of PMF calculation, the discrepancy between the extracted factor profile and the actual source profile may be attributed to a number of reasons (Reff et

al., 2007; Shi et al., 2016), including similar source markers (i.e., collinear sources, such as dust and soil sand) and emission characteristics of some pollution sources, as well as measurement error, sampling variability and modeling process (i.e., rotational ambiguity).

The PMF/Multilinear Engine 2-Species Ratios (PMF/ME2-SR) model based on linear fitting is a scientific computing platform that allows the implementation of external constraints (Amato and Hopke, 2012; Amato et al., 2009; Sofowote et al., 2018). The model incorporates species ratios into the model as constraint conditions of the extraction process, which makes the extracted factors more physically interpretable and acceptable (Liu et al., 2015), and solves the problem of PMF to a certain extent. Since some pollution sources emit both PM and gaseous pollutants, the latter can also reflect the emission characteristics of pollution sources. In view of this feature, the species concentration and the ratios of gaseous pollutants (SO_2/CO) based on the 2014 emission inventory in Tianjin (see Table S2) (Shi, G. et al., 2018) were incorporated into the model to help explore the influence of sources. In our implementation, a total of 5203 multi-species data in the same period were selected for source apportionment calculation. PMF/ME2-SR was applied with the aim of analyzing the common source impact of individual species while quantifying the ensemble-derived source impacts of multiple-species.

2.4. Ensemble-derived source impacts

We developed an ensemble source apportionment method for gaseous pollutants and

$PM_{2.5}$-CO_2, by using the weighted average method proposed by Lee et al (Lee et al., 2009). In this method, the common source is defined as the source that contributes to both species 1, 2, …, and *n*. The source that contributes to only one of the substances will not be identified as the common source. The ensemble source apportionment consists of three main steps:

Stage 1. Using the PMF/ME2-SR model to analyze the source impact of individual species ($PM_{2.5}$, SO_2, NO_x, total initial VOCs (In-VOCs), and CO_2). It should be noted that, the photochemical loss of VOCs can introduce additional uncertainties in the trend of source impact. We corrected the photochemical loss of different VOCs species and used the obtained In-VOCs to identify their real sources (more details of the method are described in our previous study (Wang et al., 2022)). The trend of gaseous data in source apportionment results reflects the influence of $PM_{2.5}$ sources, but not the real contributions to gas species, so it is defined as source impact in this study. We regard particulate matter and its components as a total variable, and SO_2, NO_x, In-VOCs, and CO_2 are used as their own total variables to participate in the calculation. More of the calculation is detailed in Supporting Section S1 and Section S2.

Stage 2. Based on the principle of the weighted average method, the weighting coefficients of $PM_{2.5}$, SO_2, NO_x, In-VOCs, and CO_2 are calculated using standard deviation and concentration range thresholds (equation (2)).

$$W_{kj}=\frac{C_j}{\sigma_{S_{jk}} \cdot RT_j} \tag{2}$$

where C_j is the concentration of species *j* during sampling; $\sigma_{S_{jk}}$ is the standard deviation of source impact of species *j* in source *k*; RT_j is range threshold of species *j*.

Here, $RT_{PM_{2.5}}$, RT_{SO_2} and RT_{NO_x} corresponds to GB 3095-2012 "Ambient Air Quality Standard" 24-hour average I grade standard limit (35 μg·m^{-3}, 50 μg·m^{-3}, and 100 μg·m^{-3}). $RT_{In\text{-}VOCs}$ uses the maximum allowable emission concentration of pollution sources, which is extracted from GB 16297-1996. Since there is no commonly accepted standard value for CO_2, we made the estimate based on the percentile of 35 μg·m^{-3} in all $PM_{2.5}$ data (the estimated value was 754836 μg·m^{-3}).

Stage 3. Combining individual species source impact and weighting coefficient, the coordinated source traceability formula (equation (3)) is proposed to quantify the ensemble-derived source impacts of multiple species (SO_2-NO_x-In-VOCs and $PM_{2.5}$-CO_2). More details about the calculation principle are described in Section S3.

$$\bar{S}_k = \frac{W_{kj} \cdot S_{jk}}{\sum_{l=1}^{L} W_{kj} \cdot S_{jk}} \times 100\% \quad (3)$$

where $\bar{S}_k$ is the ensemble impact of source k (μg·m^{-3}) during sampling; S_{jk} is the impact of species j in source k; W_{kj} is the calculated weight of the impact of source k and is calculated by standard deviation and range threshold in this study, as described in equation (2).

3. Results and discussion

3.1. Pollutant levels

The monthly variations of $PM_{2.5}$ compositions, main gaseous precursors (SO_2, NO_x, and VOCs), and CO_2 during the campaign are illustrated in Fig. 1. The average concentration of $PM_{2.5}$ was 61.7 μg·m^{-3}. Between November to March, $PM_{2.5}$

concentrations were 2.2~3.0 times of the National Air Quality I Grade Standard (35 μg·m^{-3}). The seasonal pattern was consistent with previous studies in other regions (Feng et al., 2021), suggesting fine particle pollution is still severe in winter. Fig. 1c provides additional statistics on the mass concentrations of major inorganic elements and ionic components. The results showed that the concentrations of inorganic elements were in the order of winter (2.9 μg·m^{-3}) > spring (2.0 μg·m^{-3}) > autumn (1.9 μg·m^{-3}) > summer (0.9 μg·m^{-3}). Their concentration peaked in February at 3.1 μg·m^{-3} and was the lowest in July at 0.6 μg·m^{-3}. Compared with other elements, K (0.7 μg·m^{-3}), Fe (0.6 μg·m^{-3}), Ca (0.3 μg·m^{-3}) and Zn (0.2 μg·m^{-3}) accounted for 34%, 28%, 16% and 11% of the total elements respectively. In Fig. 1d, the concentrations of water-soluble ions ranked as: winter (38.0 μg·m^{-3}) > autumn (31.3 μg·m^{-3}) > spring (31.1 μg·m^{-3}) > summer (16.5 μg·m^{-3}). They peaked in March (46.1 μg·m^{-3}), and decreased to the lowest value in June (12.9 μg·m^{-3}). Among them, the mass concentrations of secondary ions such as NO_3^-, NH_4^+ and SO_4^{2-} were 10.6 μg·m^{-3}, 7.9 μg·m^{-3} and 6.7 μg·m^{-3}, respectively, contributing to 17.2%, 12.3%, and 10.8% of $PM_{2.5}$ mass. This suggests a severe secondary pollution. Thus, it is necessary to further explore gaseous pollutants that are precursors in the formation of secondary aerosol.

As shown in Fig. 1, during the monitoring period, the average concentrations of SO_2, NO_x and VOCs were 9.7 μg·m^{-3}, 86.0 μg·m^{-3} and 55.7 μg·m^{-3}, respectively. NO_x, SO_2, and VOCs were significantly correlated with $PM_{2.5}$ (Fig. S2), and the correlation coefficients (r_1, r_2 and r_3) were 0.83, 0.81 and 0.80, respectively. These correlations revealed that the variations in $PM_{2.5}$ concentrations are strongly influenced by local

meteorological conditions (Dai et al., 2020). Understanding the impact of gaseous pollutant source emissions could contribute to the formulation of $PM_{2.5}$ pollution source control strategies. Furthermore, by analyzing observational data, we found that CO_2 average concentration was 433.7 ppm. The concentrations from November to January were higher than that in other months, which were 479.0 ppm, 499.5 ppm and 507.1 ppm, respectively. Coincidentally, the temporal pattern of CO_2 correlated with $PM_{2.5}$ which suggests that the main sources of CO_2 and $PM_{2.5}$ are similar in the study area. Thus, it is possible to apportion the sources of both CO_2 and $PM_{2.5}$ at the same time, which is conducive to the understanding of the impact of source changes on the coordinated emission reduction of CO_2 and pollutants.

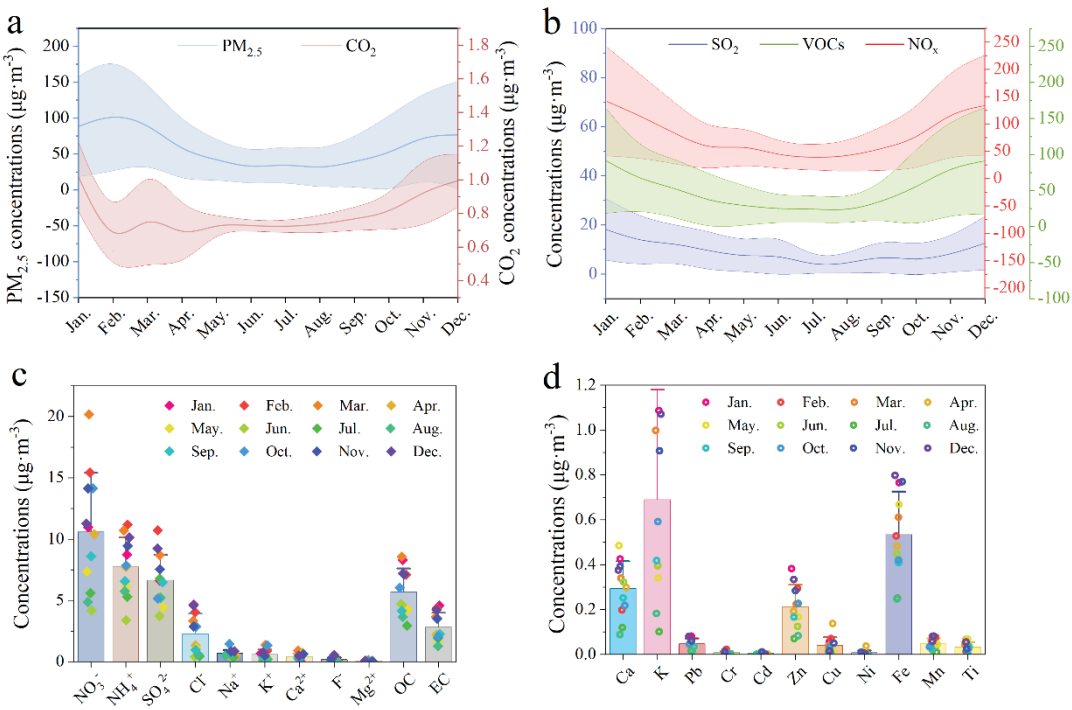


Fig. 1. Monthly concentrations of monitored species from June 2017 to May 2019. (a): Concentrations ($\mu g \cdot m^{-3}$) of $PM_{2.5}$ and CO_2. (b): Concentrations ($\mu g \cdot m^{-3}$) of sulfur dioxide (SO_2), nitrogen oxides (NO_x), and volatile organic compounds (VOCs). (c): Concentrations ($\mu g \cdot m^{-3}$) of elements in $PM_{2.5}$. And (d): concentrations ($\mu g \cdot m^{-3}$) of ions, organic carbon (OC), and element carbon (EC) in $PM_{2.5}$.

3.2. Source impacts on individual species

In order to further understand the influence of pollution sources on air pollutants and CO_2, online measurement datasets that include ions, elements, OC, EC concentrations in particle phases, and gaseous species (SO_2, NO_x, In-VOCs, CO, and CO_2) were inputted into the PMF/ME2-SR model. The calculation principles are available in Section S1 and Section S2. As shown in Fig. S3, the positive correlation (r=0.95) between simulated $PM_{2.5}$ and measured $PM_{2.5}$ concentrations confirmed the reliability of the source apportionment results. More discussion on the reliability of our results is provided in Supporting Information Section S4.

3.2.1. Source contributions to $PM_{2.5}$

A detailed analysis of $PM_{2.5}$ source apportionment was carried out to further quantify the impact of common sources. Sources of $PM_{2.5}$ were identified according to source markers and ratios of gaseous species in factors (Table S2). After running the model, 8 factors were identified and the factor profiles were showed in Fig. 2. Factor 1 was dominated by high proportions of OC, EC, NO_x, CO, and CO_2. Studies suggested that OC and EC are marker species for vehicles, and a large amount of nitrogen oxides will be emitted during driving too (Li, X. et al., 2019; Yuan et al., 2006). In addition, the SO_2/CO mass ratio was close to the value from vehicle emissions (Table S2). Hence, factor 1 was classified as vehicle exhaust (Shi, G. et al., 2018). Factor 2 was identified as secondary nitrate owing to its high abundance of NO_3^- and NH_4^+ (Shi et al., 2017). Factor 3 was considered to be coal combustion, which was strongly enriched with As,

OC, EC, Cl^-, and SO_2 (Shi et al., 2019), and the ratio of SO_2/CO was similar to the value (Table S2). Factor 4 was assigned to industrial emissions, which had a high content of heavy metal elements such as Fe, Zn, Mn, Pb, and Cu. In factor 5, Ca^{2+}, and Mg^{2+} exhibited high weighting and these species are generally used as the markers for soil & dust (Srivastava et al., 2021). Factor 6 was characterized by high levels of K^+, OC, EC, and accompanied by a high proportion of CO_2, common indicators of biomass burning (Li, X. et al., 2019; Liu et al., 2019). Factor 7 had high loadings of SO_4^{2-} and NH_4^+, and the high proportion of SO_2 in this source was apparent, which was consistent with the features of secondary sulfate (Feng et al., 2022; Gao et al., 2016). Factor 8 was only accompanied by high levels of CO_2 and it was therefore judged to be a CO_2-sources, which is normally related to the natural emissions other than the aforementioned anthropogenic emissions.

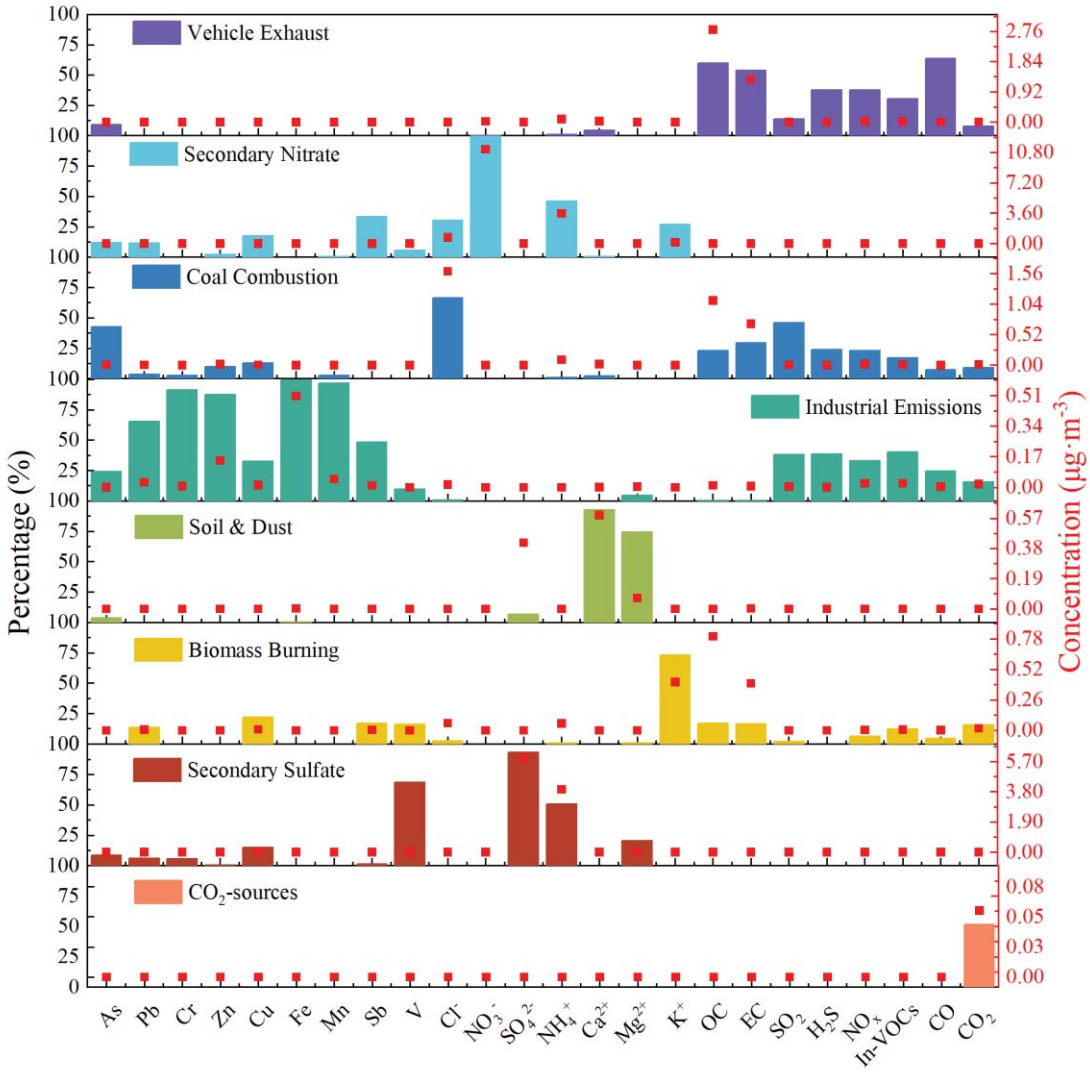


Fig. 2. Factor profiles of $PM_{2.5}$ derived by PMF/ME2-SR. Bars represent the normalized percentage (%) of species apportioned to each species in the corresponding source. Red dots represent the mass concentration ($\mu g \cdot m^{-3}$) of the component resolved from PMF/ME2-SR.

Based on the above information, the contributions of each pollution source to $PM_{2.5}$ in the entire sampling period were calculated. As shown in Fig. 3a, secondary nitrate and secondary sulfate were the dominant contributors among seven sources, contributing 24% and 18% to $PM_{2.5}$. The average contribution of coal combustion to $PM_{2.5}$ mass is 17%, which is comparable to a previous study (Liu et al., 2018). The $PM_{2.5}$ contribution from industrial emissions is 11%, which is in good agreement with

the results that the source contribution from industry to $PM_{2.5}$ in Tianjin was in the range of 10-20% (Liu et al., 2018). Vehicle exhaust, soil & dust, and biomass burning also contributed significantly to $PM_{2.5}$, accounting for 14%, 10%, and 6%, respectively. Fig. 3b-e summarized the averages of $PM_{2.5}$ in major source contributions during four seasons. It can be clearly seen from the figure that the contributions of various pollution sources to $PM_{2.5}$ in winter were significantly more serious than that in summer. Fig. S4 presents a corresponding bar graph for Fig. 3b-e, which provides a numerical relationship between the seasonal changes in $PM_{2.5}$ and source contributions. Soil & dust in spring (14%) had a greater impact on $PM_{2.5}$ source contribution than in other seasons, which is related to the resuspension of dust from the earth's crust by strong winds (wind speed in different seasons are shown in Fig. S5). In summer, secondary sulfate was the main source during the study period, accounting for 30% of the total source contributions. Strong photochemical reactions in summer increased the oxidation capacity of the atmosphere and accelerated the oxidation of SO_2 into sulfate (Cao et al., 2021). In autumn, secondary nitrate made a major contribution to $PM_{2.5}$ (25%), which may enhance the formation of nitrate by heterogeneous reaction with increased humidity and decreased photochemical activity (Petetin et al., 2015; Pun and Seigneur, 2001). During winter, coal combustion (27%) and biomass burning (9%) played major roles in $PM_{2.5}$, especially the latter, whose proportion increased significantly compared with other seasons, due mostly to the increase of residential heating. The relative contributions of different sources to $PM_{2.5}$ concentrations varied significantly with seasons, which helped to quickly identify pollution sources for

priority control and thus promoted the implementation of cleaner production policies.

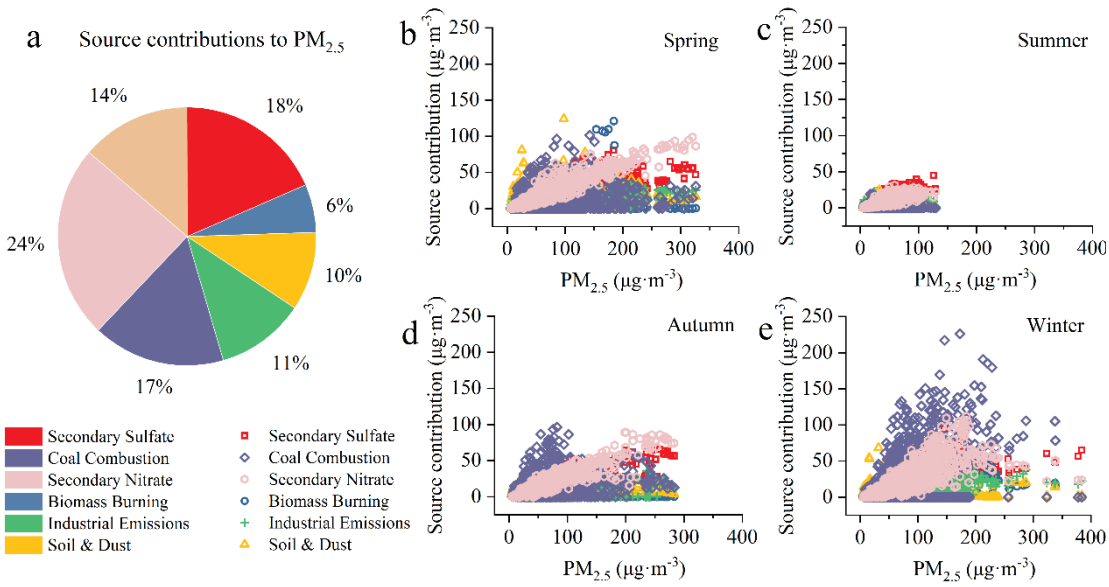


Fig. 3. Source contributions to $PM_{2.5}$. (a): Average source contribution to $PM_{2.5}$ during the sampling period. (b-e): Contribution concentrations of different pollution sources to $PM_{2.5}$ in spring, summer, autumn, and winter, respectively. The corresponding average percentages are presented in Fig. S4.

3.2.2. Source impacts on gaseous pollutants

Unlike primary pollutants, secondary aerosol species, such as sulfate, nitrate, and SOA, are produced by photochemical processes from atmospheric oxidation of precursor gases (e.g., SO_2, NO_x, and VOCs). Studies have shown that excessive emissions of gaseous pollutants may affect human health and aggravate atmospheric particulate matter pollution (Anenberg et al., 2017). Therefore, exploring the sources of gaseous precursors of secondary aerosol is also helpful to further analyze the sophisticated effects of gaseous pollutants on $PM_{2.5}$ generation. To this end, we further compared the impacts of common sources associated with $PM_{2.5}$ on SO_2, NO_x and In-VOCs.

The effects of different pollution sources on various gaseous precursors showed

relatively apparent seasonal differences as shown in Fig. 4, and the average percentages of their source impacts are provided in Fig. S6 and Fig. S7. For SO_2, the impact of coal combustion fluctuated between 50-70% in spring, autumn, and winter, revealing a leading role of coal combustion on SO_2, which agrees with earlier findings (Ma et al., 2017); industrial emissions maintain a high level of influence on SO_2, ranging from 24% to 42%; impacts of vehicle exhaust were the highest in summer (39%) and lowest in winter (5%); the influence of biomass burning remain relatively consistent at about 1%. In addition, nitrogen compounds in fuels tend to be oxidized to produce NO_x after thermal decomposition under high temperature and oxygen-rich conditions. Studies have found that more than 20% of global anthropogenic emissions of NO_x are produced by on-road vehicles (Anenberg et al., 2017). The results in Fig. 4e-h also provide evidence that vehicle exhaust have a significant impact on NO_x, with the average seasonal impacts ranging from 18% in winter (lowest) to 69% in summer (highest), suggesting that more aggressive measures should be taken to reduce the effect of vehicle emissions on atmospheric NO_x concentration in the study region. It is noted that NO_x production was also affected by biomass burning, although the average proportion is small (3%), which may be related to the to the combustion process of fuel nitrogen. Besides, In-VOCs were mainly affected by industrial emissions, vehicle exhaust, coal combustion and biomass burning, whose average impacts were 37%, 34%, 23% and 6%, respectively. Similar to the source contribution to $PM_{2.5}$, we found that biomass burning and coal combustion rose to 9% and 39% in winter mainly due to the increased heating, which could cause the sources to become the main control targets of In-VOCs

in the study area during winter. Overall, the dominant role of combustion-related sources on air quality improvement during the sampling period in the study area is evident.

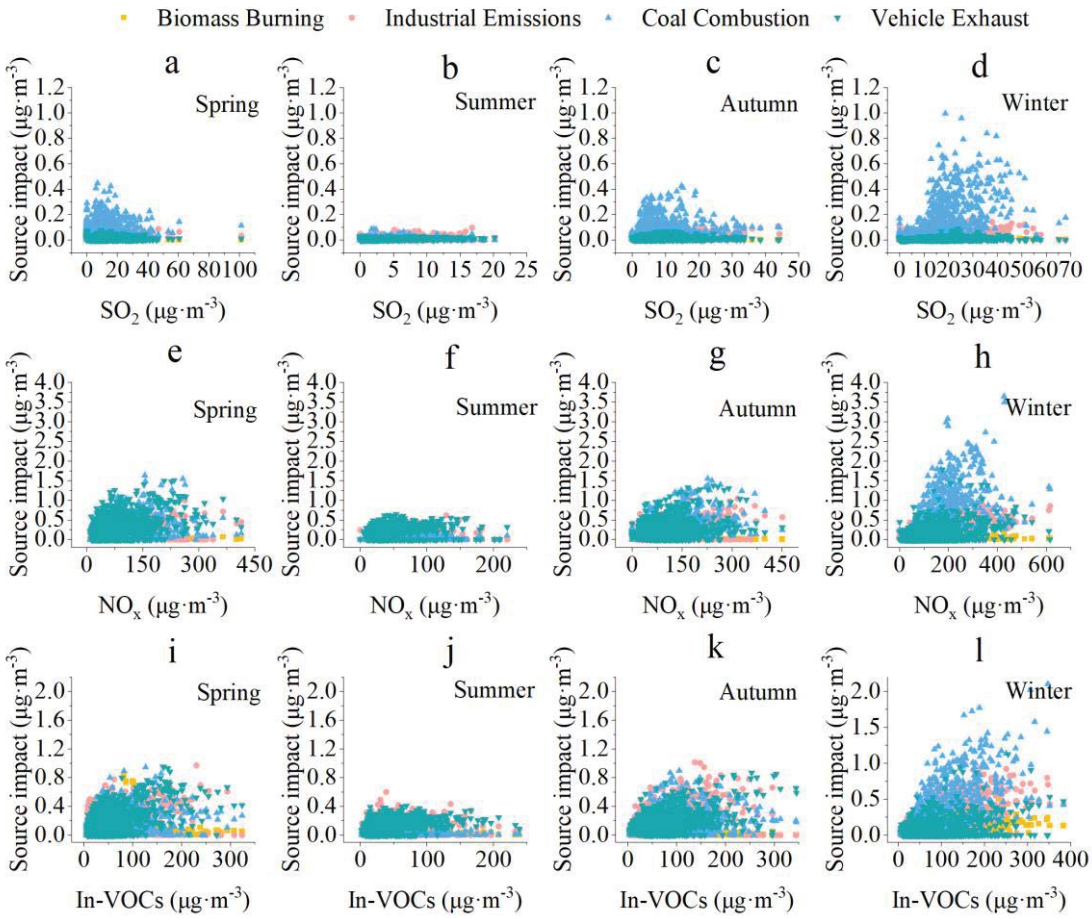


Fig. 4. Impacts of different pollution sources on SO_2, NO_x, and In-VOCs. The scatter plots (a-d) show the source impacts on CO_2 in spring, summer, autumn, and winter respectively during the sampling period. (e-h) correspond to the source impacts on NO_x in four seasons. (i-l) correspond to the source impacts on In-VOCs in four seasons.

3.2.3. Source impacts on CO_2

Affected by human activities, emission reduction at the source is an effective way to control CO_2 pollution. In this study, one natural source and four common anthropogenic sources related to $PM_{2.5}$ were identified (Fig. 5a). Among them, CO_2-sources were the source with the greatest impact on CO_2, reaching 36%, which tends to the CO_2

emissions reported by Wang, F. et al. (2021). This is followed by industrial emissions (21%), coal combustion (18%), vehicle exhaust (13%), and biomass burning (12%). Referring to previous studies (Zheng et al., 2018), industrial sector account for about 20% of total CO_2 emission in Tianjin and the coal combustion and transportation are important contributors to CO_2 emissions, which supports our research results. From Fig. 5b-e, we found a strong effect of combustion-related sources emissions on CO_2, similar to that on gaseous pollutants. As a common feature throughout the year, the leading role of CO_2-sources during the observation period in the study region was clear; the impact of industrial emissions on CO_2 was stable at 20-26% in spring, autumn and winter, except that it was low at 14% in summer; the impacts of coal combustion on CO_2 in autumn and winter were greater than that in spring and summer; however, the highest impact of vehicle exhaust appeared in summer (18%), and the lowest impact occurred in winter (6%); unlike other sources, the impact of biomass burning showed a considerable increase in winter (by 1.2 to 2.6 times, compared to other seasons). According to the percentage contributions of CO_2 sources in four seasons provided in Fig. S8, we found that in spring and autumn, but not in summer and winter, the orders of the top four source categories were CO_2-sources > industrial emissions > coal combustion > vehicle exhaust. In contrast, the proportion of biomass burning and coal combustion in winter increased to 16% and 29%.

Our study results emphasized and quantified that fossil fuel combustion through human activities is the main cause of the increase in atmospheric CO_2 concentration (Fig. 5a), and the impacts of these common sources on CO_2 were higher in autumn and

winter than in spring and summer (Fig. S9). This finding may be due to continued heating in early spring and that CO_2 is easier to accumulate under adverse meteorological conditions such as low mixing height in winter (Wang et al., 2016). Therefore, in order to reduce CO_2, strengthening the control of winter combustion-related source emissions may be the key to achieve carbon emission reduction target.

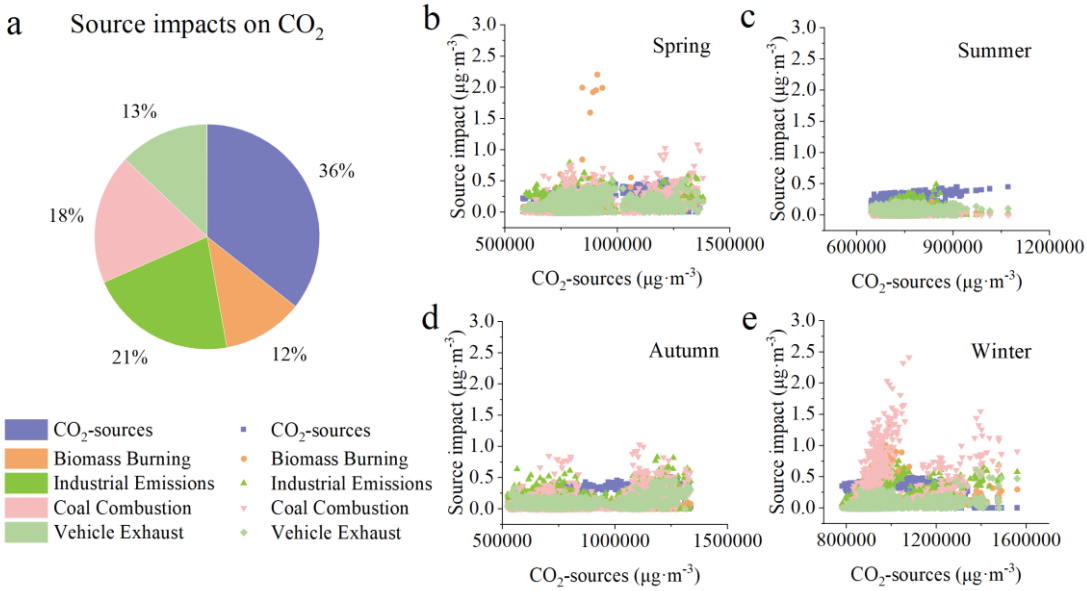


Fig. 5. Impacts of identified sources on CO_2. (a): The normalized impact (100%) of identified sources on CO_2. (b-e): Impacts of different pollution sources on CO_2 in spring, summer, autumn, and winter, respectively.

3.3. Ensemble source apportionment

Through the above analysis of the sources of air pollutants and CO_2, we can conclude that they may have the shared characteristics of common sources and common processes. In the context of China's commitment to carbon emission reduction, it is essential to make good use of the synergistic effects of air pollutants control and greenhouse gas reduction. In order to maximize the benefit of CO_2 control on air pollutants and vice versa, this study further calculated the ensemble source

apportionment results of (1) key gaseous precursors (SO_2, NO_x and In-VOCs) and (2) $PM_{2.5}$ and CO_2 based on the method presented in section 2.4. The methodology developed here can directly and quantitatively analyze the common sources of different pollution systems, which will facilitate the formulation of coordinated control policies to maximize the efficiencies of carbon and clean air control.

3.3.1. Ensemble source impacts on gaseous pollutants

First, in order to directly explore the impact of gaseous precursors on secondary aerosols, we applied the weighted averaging method as reported by Lee et al. (2009) to derive ensembles of source impact results of three gaseous pollutants (SO_2-NO_x-In-VOCs) that have important effects on the environment. Fig. 6a shows four common sources associated with particulate emissions, among which vehicle exhaust was the largest source, accounting for 35%. The second dominant source was industrial emissions at 31%. The other source contributions, in descending order, were biomass burning (18%) and coal combustion (16%). As illustrated in Fig. 6b, the impacts of biomass burning sources showed clear seasonal characteristics, peaking in autumn (25%). Vehicle exhaust was the main anthropogenic source causing serious air pollution of gaseous pollutants in summer (44%), while industrial emissions were important in winter (37%). In order to facilitate direct comparison, Table 1 summarized the proportions of diverse source impacts on various gaseous pollutants. Comparing the synergistic effect of SO_2-NO_x-In-VOCs with the impacts of individual species, the results show large discrepancies. The relative importance of coal combustion is lower

in the SO_2-NO_x-In-VOCs ensemble in comparison to the SO_2 alone (decreased from 55% to 16%). The role of vehicles became more prominent in the ensemble than SO_2 alone (increased to 35%). In contrast to NO_x alone, the ensemble effect of coal combustion decreased considerably by 46% due to the influence of lower NO_x weighting on the weighted result, while industrial emissions increased by a similar degree (about 12%). Compared with In-VOCs alone, the ensemble source impacts of coal combustion and biomass burning sources decreased from 23% to 16% and increased from 6% to 18%, respectively. The results on the ensemble source impact of SO_2-NO_x-In-VOCs provide the scientific basis to develop strategies with maximum impacts on secondary particulate pollution.

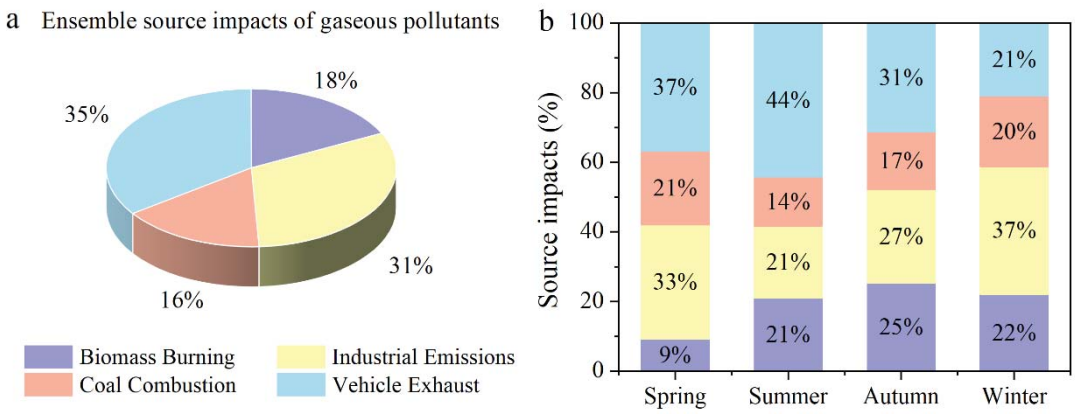


Fig. 6. Ensemble source impacts on gaseous pollutants (SO_2-NO_x-In-VOCs). (a): Average source impacts on gaseous pollutants during the sampling period. (b) Source impacts on gaseous pollutants in four seasons.

Table 1.

Source categories and impacts on $PM_{2.5}$, CO_2, SO_2, NO_x, In-VOCs, ensemble results of gaseous pollutants (SO_2-NO_x-In-VOCs) and $PM_{2.5}$-CO_2 as resolved in this study (%).

Source Impacts	CO_2 (%)	Secondary Sulfate (%)	Biomass Burning (%)	Soil & Dust (%)	Industrial Emissions (%)	Coal Combust ion (%)	Secondary Nitrate (%)	Vehicle Exhaust (%)
$PM_{2.5}$	-	18	6	10	11	17	24	14

CO_2	36	-	12	-	21	18	-	13
SO_2	-	-	1	-	30	55	-	14
NO_x	-	-	3	-	28	29	-	40
In-VOCs	-	-	6	-	37	23	-	34
SO_2-NO_x-In-VOCs	-	-	18	-	31	16	-	35
$PM_{2.5}$-CO_2	-	-	17	-	30	19	-	34

3.3.2. Ensemble source impacts on $PM_{2.5}$ and CO_2

In order to promote coordinated $PM_{2.5}$ and carbon emission reduction, we combined source contribution to $PM_{2.5}$ and source impact on CO_2 to calculate the ensemble source impact on $PM_{2.5}$-CO_2, which directly quantifies the contributions from common sources, and explores the co-benefits of atmospheric pollutants and greenhouse gases are of great significance to air quality.

There were four sources that affect $PM_{2.5}$ and CO_2 ensemble, with percentages ranging from 17% to 34% (Fig. 7a). Fig. 7b further illustrated the impacts of common sources on $PM_{2.5}$-CO_2 in different seasons. Vehicle exhaust was dominant in spring (33%), summer (44%), and autumn (34%). The impact of industrial emissions on $PM_{2.5}$-CO_2 fluctuates between 21-35% and that of coal combustion between 14-24%, respectively. Biomass burning showed a greater impact in autumn (23%) and winter (21%) due to straw burning and local heating, but its proportion is lower in spring (i.e., 16%). These above common source categories play important roles in a particular season. In contrast, considering that secondary nitrate, secondary sulfate, and soil & dust sources only contribute to $PM_{2.5}$ and do not contribute to CO_2; and that CO_2-

sources only have an effect on CO_2 concentration. Therefore, the above four sources were not identified as common sources with ensemble impacts on $PM_{2.5}$-CO_2. Overall, compared to $PM_{2.5}$ source contributions, the ensemble results (summarized in Table 1) showed a moderate contribution increase in biomass burning, industrial emissions, coal combustion, and vehicle exhaust. With regard to the sources impacts of CO_2, biomass burning, industrial emissions, coal combustion, and vehicle exhaust increased by 1.4, 1.4, 1.1, and 2.6 times.

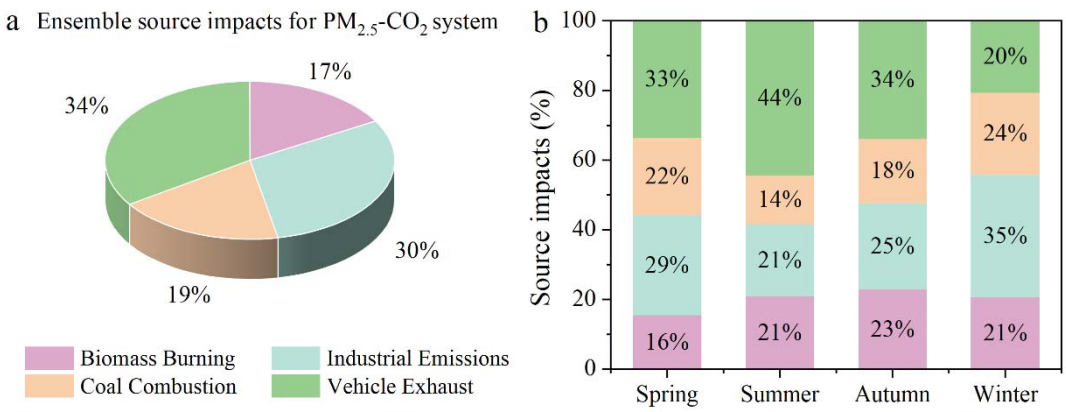


Fig. 7. Ensemble source impacts on $PM_{2.5}$-CO_2. (a): Average source impacts on $PM_{2.5}$-CO_2 during the sampling period. (b): Source impacts on $PM_{2.5}$-CO_2 in spring, summer, autumn, and winter, respectively.

4. Conclusions

Promoting the simultaneous reduction of CO_2 and air pollutants emissions are an inevitable choice for future climate and environmental governance. In this study, we investigated the concentration characteristics and source impacts of $PM_{2.5}$, SO_2, NO_x, In-VOCs and CO_2 using a combination of 2-year ground-based online measurements and model calculations. Furthermore, an ensemble source apportionment method was used to quantify the common source impacts of SO_2-NO_x-In-VOCs and $PM_{2.5}$-CO_2.

The results indicated that combustion-related sources (including coal combustion, industrial emissions, vehicle exhaust, and biomass burning) have significant impacts on multiple pollutants, and the focus of pollution prevention and control should be changed accordingly for different pollutants. For example, in order to reduce the secondary components of particulate matter, focusing on the source categories that play leading roles in the SO_2-NO_x-In-VOCs emissions, including vehicle exhaust (35%), industrial emissions (31%), biomass burning (18%), and coal combustion (16%), as estimated in this study. From the perspective of $PM_{2.5}$-CO_2 prevention, vehicle exhaust in spring, summer, and autumn was the key contributor (33%, 44%, 34%). In winter, the primary control targets should be industrial emissions and coal combustion (35% and 24%). The technique of ensemble source apportionment can prioritize the identification of sources that contribute the most to pollutant emissions when selecting greenhouse gas emission reduction measures, which can not only reduce pollutant emissions, but also bring carbon emission reduction benefits, so as to achieve higher air quality co-benefits.

This paper provides a feasible method for source tracing based on online observation dataset of $PM_{2.5}$ and CO_2. In prospective studies, the method can also be extended to multi-field collaborative trace studies, which may have significant potentials. We believe the findings in this study will contribute to a better understanding of the reduction of emission sources in environments, and provide quantitative scientific information for cleaner production.

CRediT authorship contribution statement

Zhenyu Wang: Data curation, Writing - original draft, Writing - review & editing. **Haofei Yu**: Writing - review & editing. **Weiqing Liang:** Investigation, Literature collection. **Feng Wang:** Data curation, Writing - review & editing. **Gen Wang:** Supervision. **Da Chen:** Supervision. **Weichao Wang:** Supervision. **Huan Zhao:** Investigation. **Yinchang Feng:** Conceptualization, Supervision. **Zongbo Shi:** Supervision, Writing - review & editing. **Guoliang Shi:** Conceptualization, Methodology, Writing - review & editing.

Declaration of competing interest

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

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CRediT authorship contribution statement

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