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1 Ensemble source apportionment of air pollutants and 2 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 CO₂. To do this, we first need to better quantify the impacts of different sources to air pollutants and CO₂ at once. Based on a 2-year observation of fine particulate matter (PM_{2.5}), gaseous pollutants (including sulfur dioxide (SO₂), nitrogen oxides (NO_x), initial volatile organic compounds (In-VOCs)), as well as carbon dioxide (CO₂), we apportioned the sources of criteria air pollutants and CO₂ 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 (SO₂-NO_x-In-VOCs) and (2) PM_{2.5} and CO₂ ensemble (PM_{2.5}-CO₂). The results demonstrate that vehicle exhaust (35%), industrial emissions (31%), biomass burning (18%), and coal combustion (16%) were the main control targets for the SO₂-NO_x-In-VOCs ensemble, and they were also the dominant contributors to PM_{2.5}-CO₂ 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.

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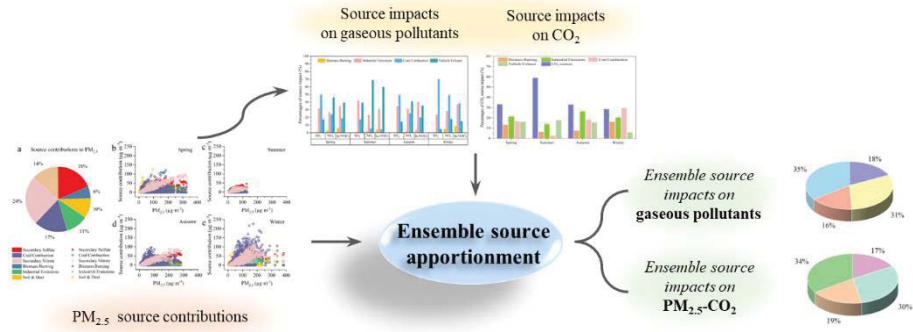
50 **Keywords**

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

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54 **Graphical abstract**

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57 **Highlights**

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- 59 ● Concentrations and sources of PM_{2.5}, gaseous pollutants and CO₂ were analyzed.
- 60 ● Developed weighted average method that can quantify ensemble source impacts.
- 61 ● Reduce combustion source emissions benefit control of gaseous pollutants and PM_{2.5}-CO₂.

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3 64 references)
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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/zwggk/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 (SO₂), 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,

1 88 including a rise in vector-borne diseases, increased heat-related morbidity and mortality,
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3 89 and potential negative impacts on water resources and crop production (Hayes and
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6 90 Poland, 2018; Piao et al., 2010; Watts et al., 2015), which may affect human habitability
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9 91 in the future (Kang and Eltahir, 2018; Pal and Eltahir, 2016). Since the industrial
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12 92 revolution, the increasing population, intensive agricultural activities and the use of
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15 93 energy (such as fossil fuels) have brought unprecedented climate warming (Scheutz et
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17 94 al., 2009). Carbon dioxide (CO₂), a typical greenhouse gas (Kramer et al., 1999), is
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20 95 released into the atmosphere and forces the climate out of balance by interfering with
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23 96 the earth's natural carbon cycle (Rastogi et al., 2002). Studies have shown that the
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26 97 global average CO₂ concentration in the 21st century has exceeded that of the industrial
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29 98 age (1750 AD, 278 ppm) by nearly 45% (Liu et al., 2021). Although CO₂ is not
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32 99 considered an air pollutant, it has become a research hotspot due to its effect on the
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35 100 climate. Simulation studies showed that low-carbon policies oriented toward carbon
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37 101 emission reduction have co-benefits that improve air quality (Shi et al., 2021).
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39 102 Therefore, CO₂ emission reduction is urgently needed.
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42 103 It is worth noting that climate change, particulate matter and gaseous pollutants are
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45 104 intricately linked. Increased CO₂ in the atmosphere creates a greenhouse effect by
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48 105 absorbing solar radiation reflected from the earth's surface and also releases long-wave
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51 106 radiation, causing global warming. Climate change leads to changes in temperature,
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54 107 radiation, precipitation, wind speed and other meteorological factors that affect the
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56 108 generation, accumulation and diffusion of pollutants (Cai et al., 2017; Zou et al., 2017).
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59 109 Moreover, pollutants (particularly aerosols) affected by the photochemical generation
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1 110 of gaseous precursors can further affect the climate system by altering the atmospheric
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3 111 radiation budget and influencing cloud formation (Zhao et al., 2017). Therefore, a set
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6 112 of collaborative approaches to carbon dioxide, particulate matter and gaseous pollutants
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9 113 reduction need to be developed.

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11 114 Air pollution has the similar root and source as greenhouse gases. Human activities
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13 115 can not only emit CO₂ but also simultaneously emit air pollutants such as by PM_{2.5},
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16 116 SO₂, NO_x, and VOCs (Shi et al., 2021). Although many recent publications have
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19 117 reported on the source contributions of the aforementioned pollutants (Li et al., 2020;
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22 118 Sun et al., 2020; Yang et al., 2021), only general conclusions can be drawn about the
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25 119 importance of individual pollutants. So far, there is still a lack of source apportionment
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28 120 technology to directly quantify the synergistic effects of common sources of multiple
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31 121 pollutants using detailed PM_{2.5}, SO₂, NO_x, VOCs and CO₂ concentration data.
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34 122 Controlling emissions sources of pollution is an effective way to fulfill coordinated
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37 123 emission reduction of greenhouse gas and air pollution. In this study, we make use of
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40 124 comprehensive observations from June 1, 2017 to May 6, 2019 at Tianjin, China, to
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43 125 quantitatively estimate the source impacts of particulate matter, gaseous precursors
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46 126 (SO₂, NO_x, VOCs) and CO₂, respectively. Based on analytical results of individual
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49 127 species, an ensemble source apportionment technique for coordinated source
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52 128 traceability of multiple air pollutants and greenhouse gas is developed. Ultimately, the
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55 129 purpose is to identify common sources across different seasons to achieve collaborative
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58 130 carbon emission reduction and clean air goals. Our research will provide implications
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61 131 for China and other countries to formulate coordinated air pollutant-greenhouse gas
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1 132 control strategies in the future.

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4 5 6 7 134 **2. Material and methods**

8 9 10 135 *2.1. Site description*

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13 136 The campaign was carried out based on annual sampling (from June 1st 2017 to May
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16 137 6th 2019) at the air quality supersite (117°24'N, 38°59'E) in Nankai University of
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19 138 Tianjin, a megacity in Northern China, which is located in a typical suburban residential
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22 139 area and surrounded by agricultural land. The location of the sampling site is shown in
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25 140 [Fig. S1](#). About 1.5 km southwest and 2.5 km northwest of the site are major roadways
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28 141 with dense automobile traffic. To the northwest of the sampling site, there are industrial
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31 142 parks specializing in manufacturing and chemical industries. Therefore, the sampling
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34 143 point is mostly affected by local air pollution sources.

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37 38 39 145 *2.2. Measurement and analysis*

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43 146 In order to understand the common sources of air pollutants and greenhouse gas,
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46 147 multiple instruments were deployed in the campaign. PM_{2.5} mass concentrations were
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49 148 collected using the online beta attenuation particle monitor (Focused Photonics Inc.,
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52 149 BPM-200, China). Elements (Ca, K, Pb, Cr, Cd, Zn, Cu, Ni, Fe, Mn, Ti, etc), key
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55 150 components in PM_{2.5}, were measured by an online X-ray fluorescence (Focused
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57
58 151 Photonics Inc., AMMS-100, China). Water-soluble ions in PM_{2.5} (Na⁺, K⁺, Mg²⁺, Ca²⁺,
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61 152 Cl⁻, F⁻, NO₃⁻, SO₄²⁻, and NH₄⁺) were recorded using an online ion chromatograph

1 153 (Thermo Fisher Scientific Inc., URG 9000D, USA), with cation detection using 20 mM
2
3 154 methanesulfonic acid, and anion analysis using 0.08 mM Na₂CO₃/0.01 mM NaHCO₃.
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6 155 Organic carbon (OC) and element carbon (EC) of PM_{2.5} were carried out by a thermal-
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9 156 optical carbon analyzer (Focused Photonics Inc., OCEC-100, China). Concentrations
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11 157 of CO₂, carbon monoxide (CO), nitrogen oxides (NO_x = NO + NO₂), and gaseous
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13 158 pollutants, including SO₂ and hydrogen sulfide (H₂S) were detected by online
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15 159 instruments (API Inc. T360, T300, T201, T101, USA). Volatile organic compounds
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17 160 (VOCs) were measured using a set of GC955-611/811 series instruments (Syntech
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19 161 Spectras Inc., Holland). The time resolutions of the measurements collected for this
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21 162 study are summarized in [Table S1](#).
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32 164 *2.3. Source apportionment*

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35 165 Factor analysis models, such as the positive matrix factorization (PMF), are
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37 166 statistical methods that use the chemical composition data at the receptor to obtain
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39 167 source profile and source contribution (equation (1)) ([Paatero, 1997](#); [Paatero and Tapper,](#)
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41 168 [1994](#)).
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$$46 169 x_{ij} = \sum_{k=1}^p g_{ik} f_{kj} + e_{ij} \quad (1)$$

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48 170 where, x_{ij} ($\mu\text{g}\cdot\text{m}^{-3}$) is the concentration of the j th species in i th sample, g_{ik} ($\mu\text{g}\cdot\text{m}^{-3}$) is
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50 171 the contribution of the k th source to the i th sample, f_{kj} ($\mu\text{g}\cdot\text{m}^{-3}/\mu\text{g}\cdot\text{m}^{-3}$) is the fraction of
51
52 172 the j th species from the k th source, e_{ij} ($\mu\text{g}\cdot\text{m}^{-3}$) are the residuals, and p is the number of
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54 173 factors. In the process of PMF calculation, the discrepancy between the extracted factor
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56 174 profile and the actual source profile may be attributed to a number of reasons ([Reff et](#)

1 175 [al., 2007](#); [Shi et al., 2016](#)), including similar source markers (i.e., collinear sources,
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3 176 such as dust and soil sand) and emission characteristics of some pollution sources, as
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6 177 well as measurement error, sampling variability and modeling process (i.e., rotational
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9 178 ambiguity).

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11 179 The PMF/Multilinear Engine 2-Species Ratios (PMF/ME2-SR) model based on
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13 180 linear fitting is a scientific computing platform that allows the implementation of
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17 181 external constraints ([Amato and Hopke, 2012](#); [Amato et al., 2009](#); [Sofowote et al.,](#)
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19
20 182 [2018](#)). The model incorporates species ratios into the model as constraint conditions of
21
22 183 the extraction process, which makes the extracted factors more physically interpretable
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24
25 184 and acceptable ([Liu et al., 2015](#)), and solves the problem of PMF to a certain extent.
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28 185 Since some pollution sources emit both PM and gaseous pollutants, the latter can also
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31 186 reflect the emission characteristics of pollution sources. In view of this feature, the
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34 187 species concentration and the ratios of gaseous pollutants (SO₂/CO) based on the 2014
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37 188 emission inventory in Tianjin (see [Table S2](#)) ([Shi, G. et al., 2018](#)) were incorporated
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40 189 into the model to help explore the influence of sources. In our implementation, a total
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43 190 of 5203 multi-species data in the same period were selected for source apportionment
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46 191 calculation. PMF/ME2-SR was applied with the aim of analyzing the common source
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49 192 impact of individual species while quantifying the ensemble-derived source impacts of
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52 193 multiple-species.

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56 195 *2.4. Ensemble-derived source impacts*

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60 196 We developed an ensemble source apportionment method for gaseous pollutants and

1 197 PM_{2.5}-CO₂, by using the weighted average method proposed by Lee et al (Lee et al.,
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3 198 2009). In this method, the common source is defined as the source that contributes to
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6 199 both species 1, 2, ..., and *n*. The source that contributes to only one of the substances
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9 200 will not be identified as the common source. The ensemble source apportionment
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12 201 consists of three main steps:

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14 202 **Stage 1.** Using the PMF/ME2-SR model to analyze the source impact of individual
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17 203 species (PM_{2.5}, SO₂, NO_x, total initial VOCs (In-VOCs), and CO₂). It should be noted
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20 204 that, the photochemical loss of VOCs can introduce additional uncertainties in the trend
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23 205 of source impact. We corrected the photochemical loss of different VOCs species and
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26 206 used the obtained In-VOCs to identify their real sources (more details of the method
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29 207 are described in our previous study (Wang et al., 2022)). The trend of gaseous data in
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31
32 208 source apportionment results reflects the influence of PM_{2.5} sources, but not the real
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35 209 contributions to gas species, so it is defined as source impact in this study. We regard
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38 210 particulate matter and its components as a total variable, and SO₂, NO_x, In-VOCs, and
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41 211 CO₂ are used as their own total variables to participate in the calculation. More of the
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44 212 calculation is detailed in Supporting [Section S1](#) and [Section S2](#).

45 213 **Stage 2.** Based on the principle of the weighted average method, the weighting
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48 214 coefficients of PM_{2.5}, SO₂, NO_x, In-VOCs, and CO₂ are calculated using standard
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51 215 deviation and concentration range thresholds (equation (2)).

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$$W_{kj} = \frac{C_j}{\sigma_{S_{jk}} \cdot RT_j} \quad (2)$$

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55 217 where C_j is the concentration of species j during sampling; $\sigma_{S_{jk}}$ is the standard
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58 218 deviation of source impact of species j in source k ; RT_j is range threshold of species j .

1 219 Here, $RT_{PM_{2.5}}$, RT_{SO_2} and RT_{NO_x} corresponds to GB 3095-2012 “Ambient Air
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3 220 Quality Standard” 24-hour average I grade standard limit ($35 \mu\text{g}\cdot\text{m}^{-3}$, $50 \mu\text{g}\cdot\text{m}^{-3}$, and
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6 221 $100 \mu\text{g}\cdot\text{m}^{-3}$). $RT_{In-VOCs}$ uses the maximum allowable emission concentration of
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9 222 pollution sources, which is extracted from GB 16297-1996. Since there is no commonly
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11 223 accepted standard value for CO_2 , we made the estimate based on the percentile of 35
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14 224 $\mu\text{g}\cdot\text{m}^{-3}$ in all $\text{PM}_{2.5}$ data (the estimated value was $754836 \mu\text{g}\cdot\text{m}^{-3}$).

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17 225 **Stage 3.** Combining individual species source impact and weighting coefficient, the
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19 226 coordinated source traceability formula (equation (3)) is proposed to quantify the
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21 227 ensemble-derived source impacts of multiple species (SO_2 - NO_x -In-VOCs and $\text{PM}_{2.5}$ -
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23 228 CO_2). More details about the calculation principle are described in [Section S3](#).

$$24 \bar{S}_k = \frac{W_{kj} \cdot S_{jk}}{\sum_{l=1}^L W_{kl} \cdot S_{lk}} \times 100\% \quad (3)$$

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26
27 229 where \bar{S}_k is the ensemble impact of source k ($\mu\text{g}\cdot\text{m}^{-3}$) during sampling; S_{jk} is the
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29 230 impact of species j in source k ; W_{kj} is the calculated weight of the impact of source k
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31 231 and is calculated by standard deviation and range threshold in this study, as described
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33 232 in equation (2).
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45 235 **3. Results and discussion**

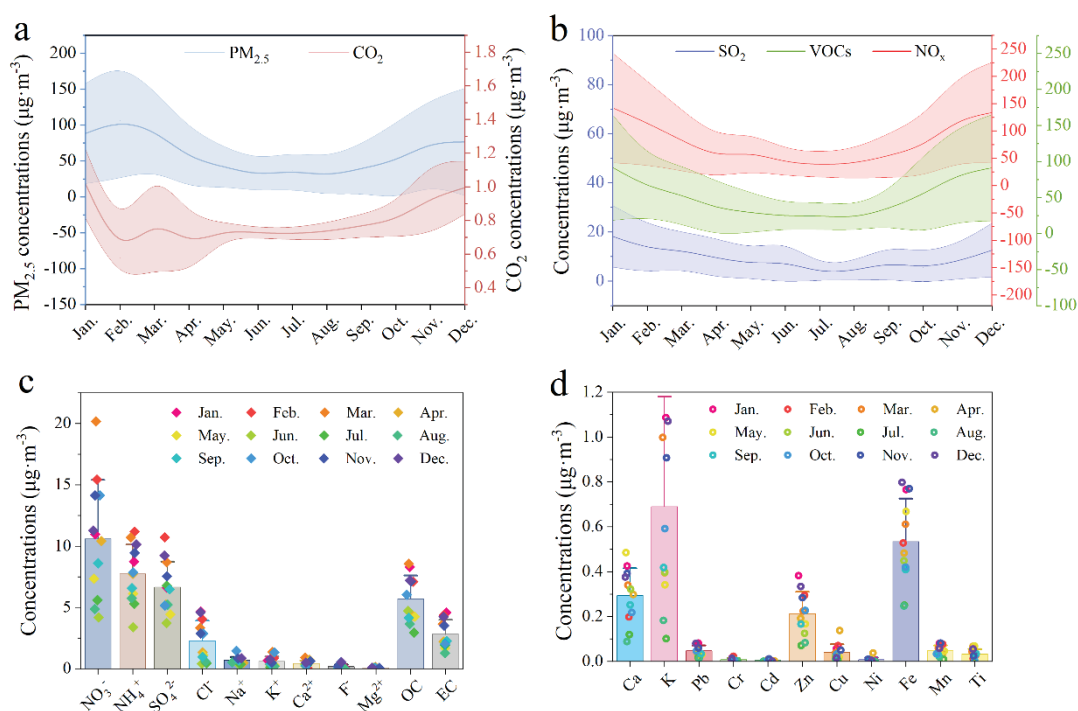
46 236 *3.1. Pollutant levels*

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49 237 The monthly variations of $\text{PM}_{2.5}$ compositions, main gaseous precursors (SO_2 , NO_x ,
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51 238 and VOCs), and CO_2 during the campaign are illustrated in [Fig. 1](#). The average
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55 239 concentration of $\text{PM}_{2.5}$ was $61.7 \mu\text{g}\cdot\text{m}^{-3}$. Between November to March, $\text{PM}_{2.5}$
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1 240 concentrations were 2.2~3.0 times of the National Air Quality I Grade Standard (35
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4 241 $\mu\text{g}\cdot\text{m}^{-3}$). The seasonal pattern was consistent with previous studies in other regions
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6 242 (Feng et al., 2021), suggesting fine particle pollution is still severe in winter. Fig. 1c
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9 243 provides additional statistics on the mass concentrations of major inorganic elements
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11 244 and ionic components. The results showed that the concentrations of inorganic elements
12
13 245 were in the order of winter ($2.9 \mu\text{g}\cdot\text{m}^{-3}$) > spring ($2.0 \mu\text{g}\cdot\text{m}^{-3}$) > autumn ($1.9 \mu\text{g}\cdot\text{m}^{-3}$) >
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15 246 summer ($0.9 \mu\text{g}\cdot\text{m}^{-3}$). Their concentration peaked in February at $3.1 \mu\text{g}\cdot\text{m}^{-3}$ and was the
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17 247 lowest in July at $0.6 \mu\text{g}\cdot\text{m}^{-3}$. Compared with other elements, K ($0.7 \mu\text{g}\cdot\text{m}^{-3}$), Fe (0.6
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19 248 $\mu\text{g}\cdot\text{m}^{-3}$), Ca ($0.3 \mu\text{g}\cdot\text{m}^{-3}$) and Zn ($0.2 \mu\text{g}\cdot\text{m}^{-3}$) accounted for 34%, 28%, 16% and 11%
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21 249 of the total elements respectively. In Fig. 1d, the concentrations of water-soluble ions
22
23 250 ranked as: winter ($38.0 \mu\text{g}\cdot\text{m}^{-3}$) > autumn ($31.3 \mu\text{g}\cdot\text{m}^{-3}$) > spring ($31.1 \mu\text{g}\cdot\text{m}^{-3}$) >
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25 251 summer ($16.5 \mu\text{g}\cdot\text{m}^{-3}$). They peaked in March ($46.1 \mu\text{g}\cdot\text{m}^{-3}$), and decreased to the
26
27 252 lowest value in June ($12.9 \mu\text{g}\cdot\text{m}^{-3}$). Among them, the mass concentrations of secondary
28
29 253 ions such as NO_3^- , NH_4^+ and SO_4^{2-} were $10.6 \mu\text{g}\cdot\text{m}^{-3}$, $7.9 \mu\text{g}\cdot\text{m}^{-3}$ and $6.7 \mu\text{g}\cdot\text{m}^{-3}$,
30
31 254 respectively, contributing to 17.2%, 12.3%, and 10.8% of $\text{PM}_{2.5}$ mass. This suggests a
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33 255 severe secondary pollution. Thus, it is necessary to further explore gaseous pollutants
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35 256 that are precursors in the formation of secondary aerosol.

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37 257 As shown in Fig. 1, during the monitoring period, the average concentrations of SO_2 ,
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39 258 NO_x and VOCs were $9.7 \mu\text{g}\cdot\text{m}^{-3}$, $86.0 \mu\text{g}\cdot\text{m}^{-3}$ and $55.7 \mu\text{g}\cdot\text{m}^{-3}$, respectively. NO_x , SO_2 ,
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41 259 and VOCs were significantly correlated with $\text{PM}_{2.5}$ (Fig. S2), and the correlation
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43 260 coefficients (r_1 , r_2 and r_3) were 0.83, 0.81 and 0.80, respectively. These correlations
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45 261 revealed that the variations in $\text{PM}_{2.5}$ concentrations are strongly influenced by local
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1 meteorological conditions (Dai et al., 2020). Understanding the impact of gaseous
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 3 pollutant source emissions could contribute to the formulation of PM_{2.5} pollution source
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 5 control strategies. Furthermore, by analyzing observational data, we found that CO₂
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 7 average concentration was 433.7 ppm. The concentrations from November to January
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 9 were higher than that in other months, which were 479.0 ppm, 499.5 ppm and 507.1
 10
 11 ppm, respectively. Coincidentally, the temporal pattern of CO₂ correlated with PM_{2.5}
 12
 13 which suggests that the main sources of CO₂ and PM_{2.5} are similar in the study area.
 14
 15 Thus, it is possible to apportion the sources of both CO₂ and PM_{2.5} at the same time,
 16
 17 which is conducive to the understanding of the impact of source changes on the
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 19 coordinated emission reduction of CO₂ and pollutants.
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272
 273 **Fig. 1.** Monthly concentrations of monitored species from June 2017 to May 2019. (a):
 274 Concentrations ($\mu\text{g}\cdot\text{m}^{-3}$) of PM_{2.5} and CO₂. (b): Concentrations ($\mu\text{g}\cdot\text{m}^{-3}$) of sulfur dioxide (SO₂),
 275 nitrogen oxides (NO_x), and volatile organic compounds (VOCs). (c): Concentrations ($\mu\text{g}\cdot\text{m}^{-3}$) of
 276 elements in PM_{2.5}. And (d): concentrations ($\mu\text{g}\cdot\text{m}^{-3}$) of ions, organic carbon (OC), and element
 277 carbon (EC) in PM_{2.5}.

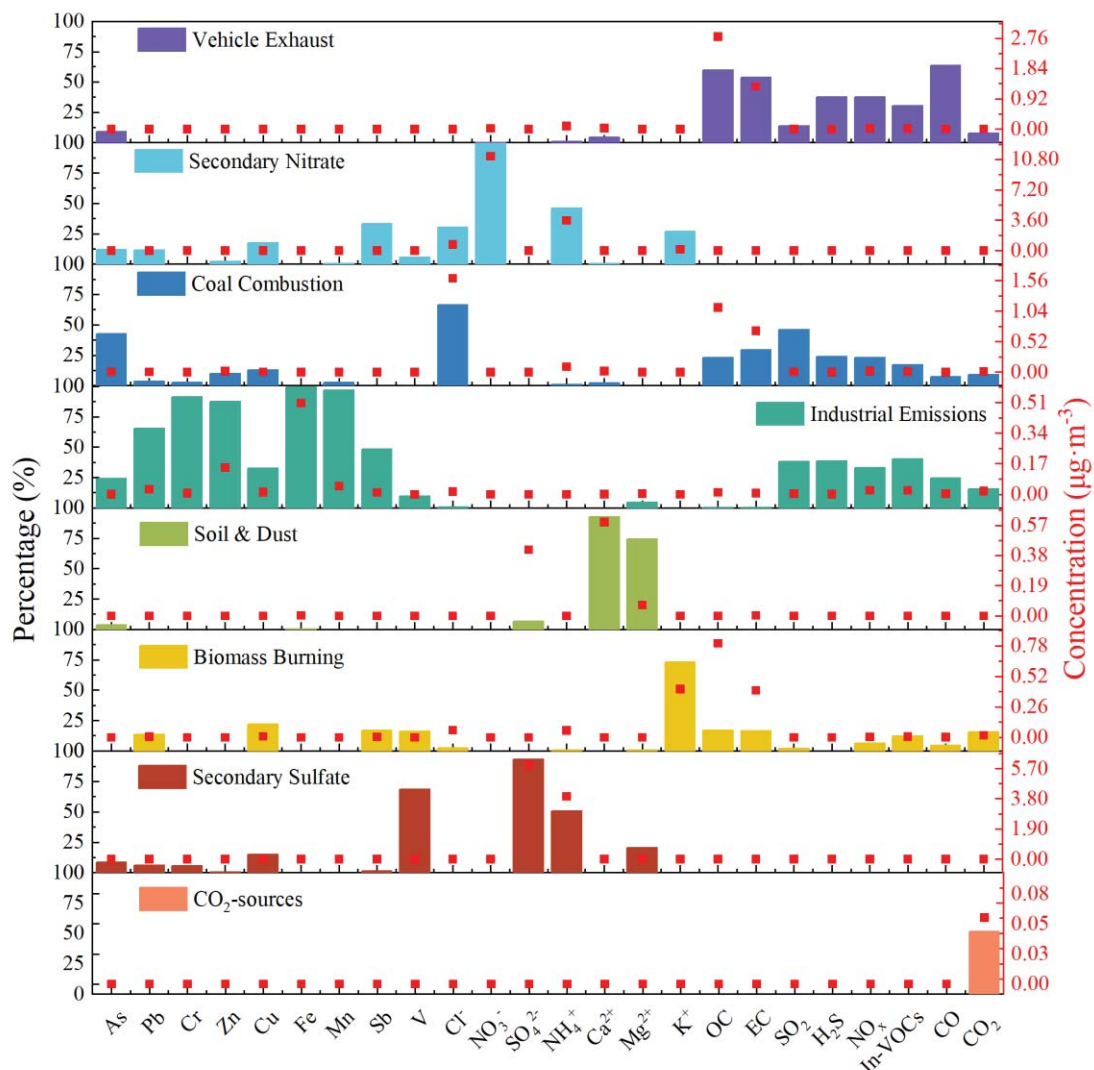
279 3.2. Source impacts on individual species

280 In order to further understand the influence of pollution sources on air pollutants and
281 CO₂, online measurement datasets that include ions, elements, OC, EC concentrations
282 in particle phases, and gaseous species (SO₂, NO_x, In-VOCs, CO, and CO₂) were
283 inputted into the PMF/ME2-SR model. The calculation principles are available in
284 [Section S1](#) and [Section S2](#). As shown in [Fig. S3](#), the positive correlation ($r=0.95$)
285 between simulated PM_{2.5} and measured PM_{2.5} concentrations confirmed the reliability
286 of the source apportionment results. More discussion on the reliability of our results is
287 provided in Supporting Information [Section S4](#).

289 3.2.1. Source contributions to PM_{2.5}

290 A detailed analysis of PM_{2.5} source apportionment was carried out to further quantify
291 the impact of common sources. Sources of PM_{2.5} were identified according to source
292 markers and ratios of gaseous species in factors ([Table S2](#)). After running the model, 8
293 factors were identified and the factor profiles were showed in [Fig. 2](#). Factor 1 was
294 dominated by high proportions of OC, EC, NO_x, CO, and CO₂. Studies suggested that
295 OC and EC are marker species for vehicles, and a large amount of nitrogen oxides will
296 be emitted during driving too (Li, X. et al., 2019; Yuan et al., 2006). In addition, the
297 SO₂/CO mass ratio was close to the value from vehicle emissions ([Table S2](#)). Hence,
298 factor 1 was classified as vehicle exhaust (Shi, G. et al., 2018). Factor 2 was identified
299 as secondary nitrate owing to its high abundance of NO₃⁻ and NH₄⁺ ([Shi et al., 2017](#)).
300 Factor 3 was considered to be coal combustion, which was strongly enriched with As,

1 301 OC, EC, Cl⁻, and SO₂ (Shi et al., 2019), and the ratio of SO₂/CO was similar to the value
2
3 302 (Table S2). Factor 4 was assigned to industrial emissions, which had a high content of
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5
6 303 heavy metal elements such as Fe, Zn, Mn, Pb, and Cu. In factor 5, Ca²⁺, and Mg²⁺
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8
9 304 exhibited high weighting and these species are generally used as the markers for soil &
10
11 305 dust (Srivastava et al., 2021). Factor 6 was characterized by high levels of K⁺, OC, EC,
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14 306 and accompanied by a high proportion of CO₂, common indicators of biomass burning
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17 307 (Li, X. et al., 2019; Liu et al., 2019). Factor 7 had high loadings of SO₄²⁻ and NH₄⁺, and
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20 308 the high proportion of SO₂ in this source was apparent, which was consistent with the
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23 309 features of secondary sulfate (Feng et al., 2022; Gao et al., 2016). Factor 8 was only
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26 310 accompanied by high levels of CO₂ and it was therefore judged to be a CO₂-sources,
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29 311 which is normally related to the natural emissions other than the aforementioned
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31 312 anthropogenic emissions.
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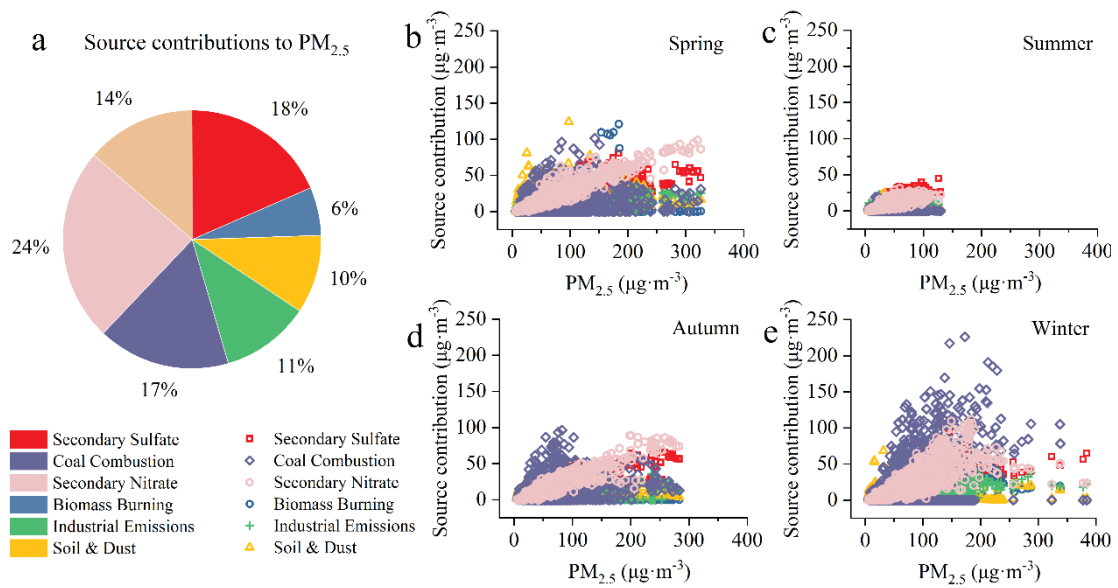


313
 314 **Fig. 2.** Factor profiles of PM_{2.5} derived by PMF/ME2-SR. Bars represent the normalized percentage
 315 (%) of species apportioned to each species in the corresponding source. Red dots represent the mass
 316 concentration ($\mu\text{g}\cdot\text{m}^{-3}$) of the component resolved from PMF/ME2-SR.

317
 318 Based on the above information, the contributions of each pollution source to PM_{2.5}
 319 in the entire sampling period were calculated. As shown in Fig. 3a, secondary nitrate
 320 and secondary sulfate were the dominant contributors among seven sources,
 321 contributing 24% and 18% to PM_{2.5}. The average contribution of coal combustion to
 322 PM_{2.5} mass is 17%, which is comparable to a previous study (Liu et al., 2018). The
 323 PM_{2.5} contribution from industrial emissions is 11%, which is in good agreement with

1 324 the results that the source contribution from industry to PM_{2.5} in Tianjin was in the
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3 325 range of 10-20% (Liu et al., 2018). Vehicle exhaust, soil & dust, and biomass burning
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6 326 also contributed significantly to PM_{2.5}, accounting for 14%, 10%, and 6%, respectively.
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8
9 327 Fig. 3b-e summarized the averages of PM_{2.5} in major source contributions during four
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11 328 seasons. It can be clearly seen from the figure that the contributions of various pollution
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13 329 sources to PM_{2.5} in winter were significantly more serious than that in summer. Fig. S4
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15 330 presents a corresponding bar graph for Fig. 3b-e, which provides a numerical
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17 331 relationship between the seasonal changes in PM_{2.5} and source contributions. Soil &
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19 332 dust in spring (14%) had a greater impact on PM_{2.5} source contribution than in other
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21 333 seasons, which is related to the resuspension of dust from the earth's crust by strong
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23 334 winds (wind speed in different seasons are shown in Fig. S5). In summer, secondary
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25 335 sulfate was the main source during the study period, accounting for 30% of the total
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27 336 source contributions. Strong photochemical reactions in summer increased the
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29 337 oxidation capacity of the atmosphere and accelerated the oxidation of SO₂ into sulfate
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31 338 (Cao et al., 2021). In autumn, secondary nitrate made a major contribution to PM_{2.5}
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33 339 (25%), which may enhance the formation of nitrate by heterogeneous reaction with
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35 340 increased humidity and decreased photochemical activity (Petetin et al., 2015; Pun and
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37 341 Seigneur, 2001). During winter, coal combustion (27%) and biomass burning (9%)
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39 342 played major roles in PM_{2.5}, especially the latter, whose proportion increased
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41 343 significantly compared with other seasons, due mostly to the increase of residential
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43 344 heating. The relative contributions of different sources to PM_{2.5} concentrations varied
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45 345 significantly with seasons, which helped to quickly identify pollution sources for
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346 priority control and thus promoted the implementation of cleaner production policies.



347
348 **Fig. 3.** Source contributions to PM_{2.5}. (a): Average source contribution to PM_{2.5} during the sampling
349 period. (b-e): Contribution concentrations of different pollution sources to PM_{2.5} in spring, summer,
350 autumn, and winter, respectively. The corresponding average percentages are presented in Fig. S4.

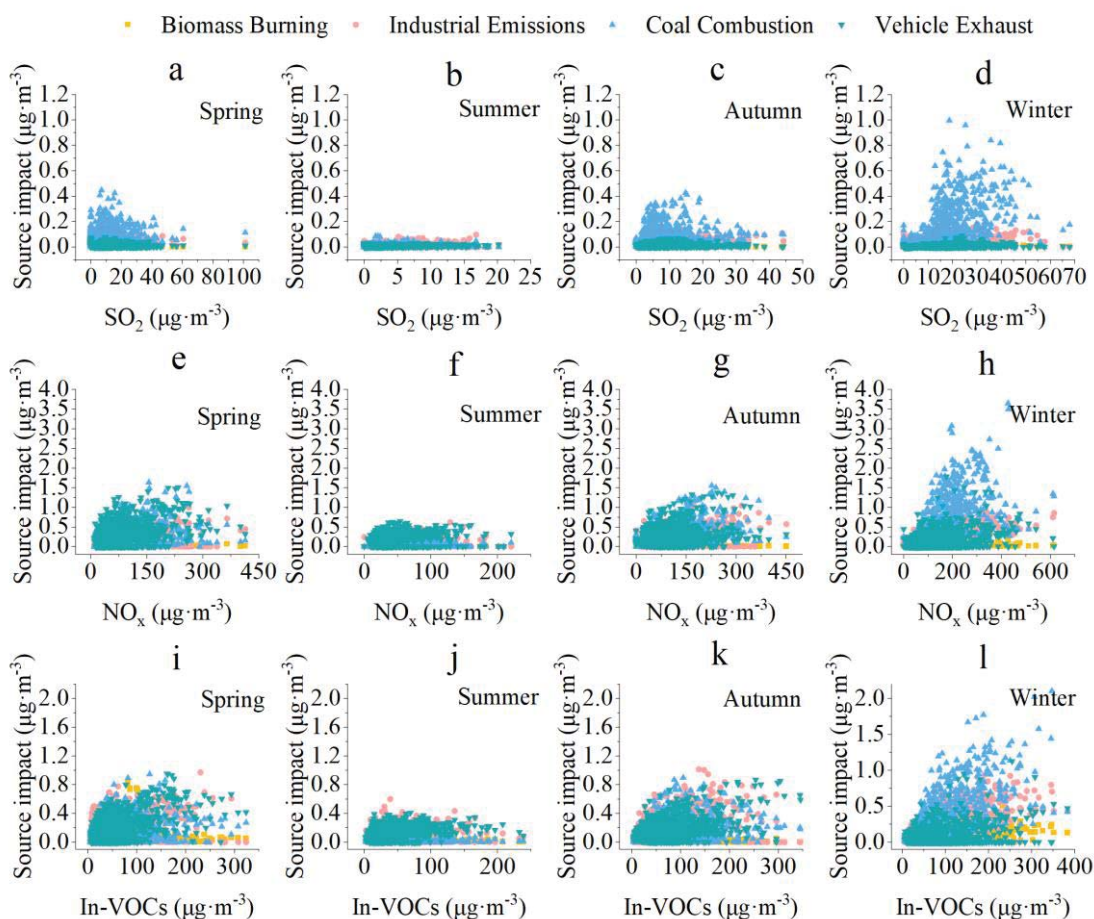
352 3.2.2. Source impacts on gaseous pollutants

353 Unlike primary pollutants, secondary aerosol species, such as sulfate, nitrate, and
354 SOA, are produced by photochemical processes from atmospheric oxidation of
355 precursor gases (e.g., SO₂, NO_x, and VOCs). Studies have shown that excessive
356 emissions of gaseous pollutants may affect human health and aggravate atmospheric
357 particulate matter pollution (Anenberg et al., 2017). Therefore, exploring the sources of
358 gaseous precursors of secondary aerosol is also helpful to further analyze the
359 sophisticated effects of gaseous pollutants on PM_{2.5} generation. To this end, we further
360 compared the impacts of common sources associated with PM_{2.5} on SO₂, NO_x and In-
361 VOCs.

362 The effects of different pollution sources on various gaseous precursors showed

1 363 relatively apparent seasonal differences as shown in Fig. 4, and the average percentages
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3 364 of their source impacts are provided in Fig. S6 and Fig. S7. For SO₂, the impact of coal
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6 365 combustion fluctuated between 50-70% in spring, autumn, and winter, revealing a
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9 366 leading role of coal combustion on SO₂, which agrees with earlier findings (Ma et al.,
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11 367 2017); industrial emissions maintain a high level of influence on SO₂, ranging from 24%
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14 368 to 42%; impacts of vehicle exhaust were the highest in summer (39%) and lowest in
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17 369 winter (5%); the influence of biomass burning remain relatively consistent at about 1%.
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19
20 370 In addition, nitrogen compounds in fuels tend to be oxidized to produce NO_x after
21
22 371 thermal decomposition under high temperature and oxygen-rich conditions. Studies
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24
25 372 have found that more than 20% of global anthropogenic emissions of NO_x are produced
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28 373 by on-road vehicles (Anenberg et al., 2017). The results in Fig. 4e-h also provide
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31 374 evidence that vehicle exhaust have a significant impact on NO_x, with the average
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34 375 seasonal impacts ranging from 18% in winter (lowest) to 69% in summer (highest),
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37 376 suggesting that more aggressive measures should be taken to reduce the effect of
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40 377 vehicle emissions on atmospheric NO_x concentration in the study region. It is noted that
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42 378 NO_x production was also affected by biomass burning, although the average proportion
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45 379 is small (3%), which may be related to the to the combustion process of fuel nitrogen.
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48 380 Besides, In-VOCs were mainly affected by industrial emissions, vehicle exhaust, coal
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51 381 combustion and biomass burning, whose average impacts were 37%, 34%, 23% and
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53 382 6%, respectively. Similar to the source contribution to PM_{2.5}, we found that biomass
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56 383 burning and coal combustion rose to 9% and 39% in winter mainly due to the increased
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59 384 heating, which could cause the sources to become the main control targets of In-VOCs
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385 in the study area during winter. Overall, the dominant role of combustion-related
 386 sources on air quality improvement during the sampling period in the study area is
 387 evident.



388
 389 **Fig. 4.** Impacts of different pollution sources on SO₂, NO_x, and In-VOCs. The scatter plots (a-d)
 390 show the source impacts on CO₂ in spring, summer, autumn, and winter respectively during the
 391 sampling period. (e-h) correspond to the source impacts on NO_x in four seasons. (i-l) correspond to
 392 the source impacts on In-VOCs in four seasons.

394 3.2.3. Source impacts on CO₂

395 Affected by human activities, emission reduction at the source is an effective way to
 396 control CO₂ pollution. In this study, one natural source and four common anthropogenic
 397 sources related to PM_{2.5} were identified (Fig. 5a). Among them, CO₂-sources were the
 398 source with the greatest impact on CO₂, reaching 36%, which tends to the CO₂

1 399 emissions reported by Wang, F. et al. (2021). This is followed by industrial emissions
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3 400 (21%), coal combustion (18%), vehicle exhaust (13%), and biomass burning (12%).
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6 401 Referring to previous studies (Zheng et al., 2018), industrial sector account for about
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9 402 20% of total CO₂ emission in Tianjin and the coal combustion and transportation are
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12 403 important contributors to CO₂ emissions, which supports our research results. From Fig.
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14 404 5b-e, we found a strong effect of combustion-related sources emissions on CO₂, similar
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17 405 to that on gaseous pollutants. As a common feature throughout the year, the leading role
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20 406 of CO₂-sources during the observation period in the study region was clear; the impact
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23 407 of industrial emissions on CO₂ was stable at 20-26% in spring, autumn and winter,
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26 408 except that it was low at 14% in summer; the impacts of coal combustion on CO₂ in
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29 409 autumn and winter were greater than that in spring and summer; however, the highest
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32 410 impact of vehicle exhaust appeared in summer (18%), and the lowest impact occurred
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35 411 in winter (6%); unlike other sources, the impact of biomass burning showed a
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38 412 considerable increase in winter (by 1.2 to 2.6 times, compared to other seasons).
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41 413 According to the percentage contributions of CO₂ sources in four seasons provided in
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44 414 Fig. S8, we found that in spring and autumn, but not in summer and winter, the orders
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47 415 of the top four source categories were CO₂-sources > industrial emissions > coal
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50 416 combustion > vehicle exhaust. In contrast, the proportion of biomass burning and coal
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53 417 combustion in winter increased to 16% and 29%.

54 418 Our study results emphasized and quantified that fossil fuel combustion through
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56 419 human activities is the main cause of the increase in atmospheric CO₂ concentration
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59 420 (Fig. 5a), and the impacts of these common sources on CO₂ were higher in autumn and
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421 winter than in spring and summer (Fig. S9). This finding may be due to continued
 422 heating in early spring and that CO₂ is easier to accumulate under adverse
 423 meteorological conditions such as low mixing height in winter (Wang et al., 2016).
 424 Therefore, in order to reduce CO₂, strengthening the control of winter combustion-
 425 related source emissions may be the key to achieve carbon emission reduction target.

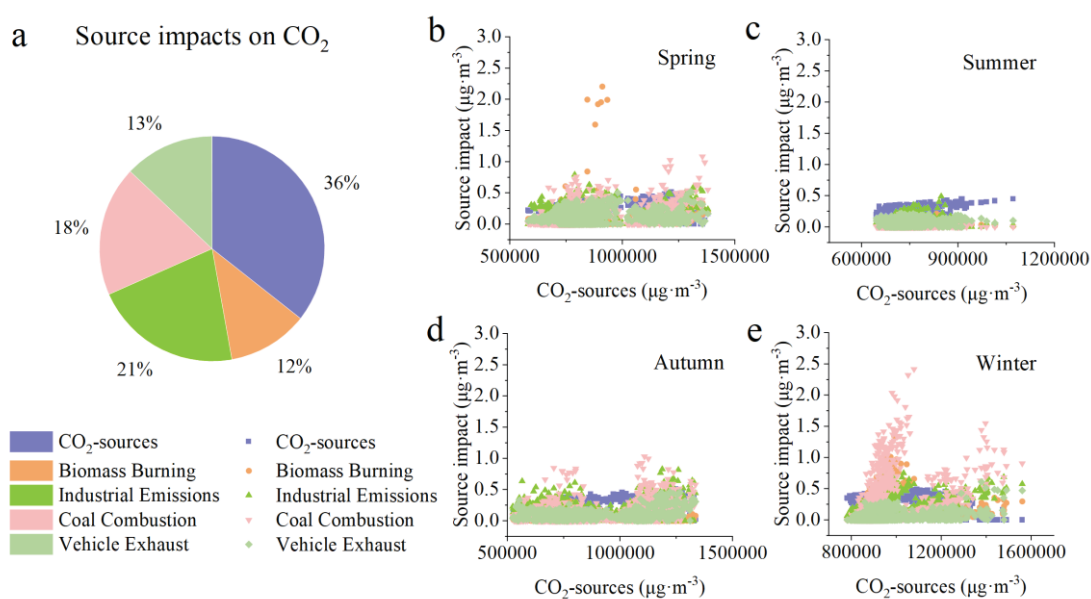


Fig. 5. Impacts of identified sources on CO₂. (a): The normalized impact (100%) of identified sources on CO₂. (b-e): Impacts of different pollution sources on CO₂ in spring, summer, autumn, and winter, respectively.

3.3. Ensemble source apportionment

432 Through the above analysis of the sources of air pollutants and CO₂, we can conclude
 433 that they may have the shared characteristics of common sources and common
 434 processes. In the context of China's commitment to carbon emission reduction, it is
 435 essential to make good use of the synergistic effects of air pollutants control and
 436 greenhouse gas reduction. In order to maximize the benefit of CO₂ control on air
 437 pollutants and vice versa, this study further calculated the ensemble source

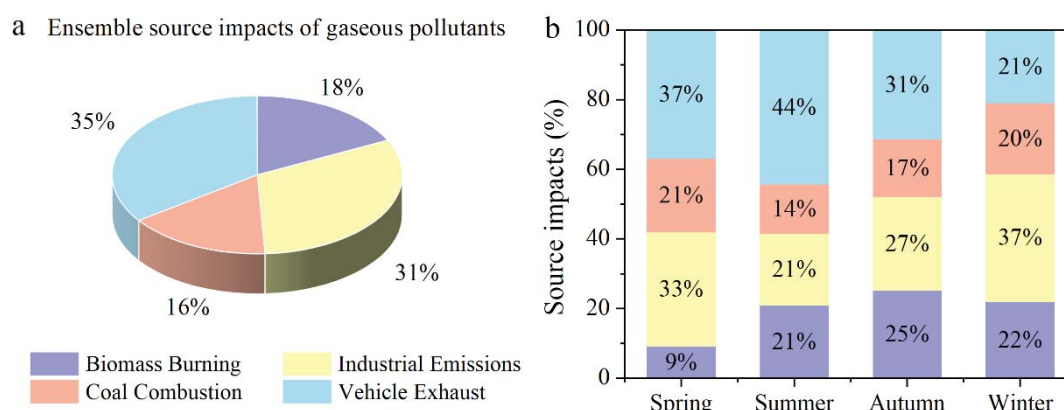
1 438 apportionment results of (1) key gaseous precursors (SO₂, NO_x and In-VOCs) and (2)
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3 439 PM_{2.5} and CO₂ based on the method presented in section 2.4. The methodology
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6 440 developed here can directly and quantitatively analyze the common sources of different
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9 441 pollution systems, which will facilitate the formulation of coordinated control policies
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12 442 to maximize the efficiencies of carbon and clean air control.
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17 444 *3.3.1. Ensemble source impacts on gaseous pollutants*

19 445 First, in order to directly explore the impact of gaseous precursors on secondary
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22 446 aerosols, we applied the weighted averaging method as reported by Lee et al. (2009) to
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25 447 derive ensembles of source impact results of three gaseous pollutants (SO₂-NO_x-In-
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28 448 VOCs) that have important effects on the environment. Fig. 6a shows four common
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31 449 sources associated with particulate emissions, among which vehicle exhaust was the
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34 450 largest source, accounting for 35%. The second dominant source was industrial
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37 451 emissions at 31%. The other source contributions, in descending order, were biomass
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39 452 burning (18%) and coal combustion (16%). As illustrated in Fig. 6b, the impacts of
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42 453 biomass burning sources showed clear seasonal characteristics, peaking in autumn
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45 454 (25%). Vehicle exhaust was the main anthropogenic source causing serious air pollution
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48 455 of gaseous pollutants in summer (44%), while industrial emissions were important in
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51 456 winter (37%). In order to facilitate direct comparison, Table 1 summarized the
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54 457 proportions of diverse source impacts on various gaseous pollutants. Comparing the
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56 458 synergistic effect of SO₂-NO_x-In-VOCs with the impacts of individual species, the
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59 459 results show large discrepancies. The relative importance of coal combustion is lower
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460 in the SO₂-NO_x-In-VOCs ensemble in comparison to the SO₂ alone (decreased from
 461 55% to 16%). The role of vehicles became more prominent in the ensemble than SO₂
 462 alone (increased to 35%). In contrast to NO_x alone, the ensemble effect of coal
 463 combustion decreased considerably by 46% due to the influence of lower NO_x
 464 weighting on the weighted result, while industrial emissions increased by a similar
 465 degree (about 12%). Compared with In-VOCs alone, the ensemble source impacts of
 466 coal combustion and biomass burning sources decreased from 23% to 16% and
 467 increased from 6% to 18%, respectively. The results on the ensemble source impact of
 468 SO₂-NO_x-In-VOCs provide the scientific basis to develop strategies with maximum
 469 impacts on secondary particulate pollution.



470
 471 **Fig. 6.** Ensemble source impacts on gaseous pollutants (SO₂-NO_x-In-VOCs). (a): Average source
 472 impacts on gaseous pollutants during the sampling period. (b) Source impacts on gaseous pollutants
 473 in four seasons.

474
 475 **Table 1.**

476 Source categories and impacts on PM_{2.5}, CO₂, SO₂, NO_x, In-VOCs, ensemble results of gaseous
 477 pollutants (SO₂-NO_x-In-VOCs) and PM_{2.5}-CO₂ as resolved in this study (%).

Source Impacts	CO ₂ (%)	Secondary Sulfate (%)	Biomass Burning (%)	Soil & Dust (%)	Industrial Emissions (%)	Coal Combustion (%)	Secondary Nitrate (%)	Vehicle Exhaust (%)
PM _{2.5}	-	18	6	10	11	17	24	14

1	CO ₂	36	-	12	-	21	18	-	13
2									
3	SO ₂	-	-	1	-	30	55	-	14
4									
5	NO _x	-	-	3	-	28	29	-	40
6									
7	In-VOCs	-	-	6	-	37	23	-	34
8									
9	SO ₂ -NO _x -In-VOCs	-	-	18	-	31	16	-	35
10									
11	PM _{2.5} -CO ₂	-	-	17	-	30	19	-	34
12									

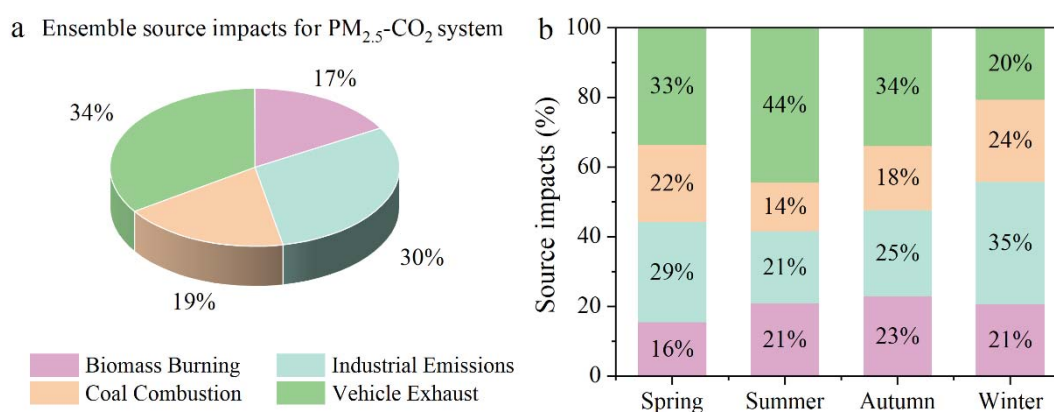
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17 479 *3.3.2. Ensemble source impacts on PM_{2.5} and CO₂*

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19 480 In order to promote coordinated PM_{2.5} and carbon emission reduction, we combined
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22 481 source contribution to PM_{2.5} and source impact on CO₂ to calculate the ensemble source
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25 482 impact on PM_{2.5}-CO₂, which directly quantifies the contributions from common sources,
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28 483 and explores the co-benefits of atmospheric pollutants and greenhouse gases are of
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31 484 great significance to air quality.

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33 485 There were four sources that affect PM_{2.5} and CO₂ ensemble, with percentages
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36 486 ranging from 17% to 34% (Fig. 7a). Fig. 7b further illustrated the impacts of common
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39 487 sources on PM_{2.5}-CO₂ in different seasons. Vehicle exhaust was dominant in spring
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42 488 (33%), summer (44%), and autumn (34%). The impact of industrial emissions on PM_{2.5}-
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45 489 CO₂ fluctuates between 21-35% and that of coal combustion between 14-24%,
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48 490 respectively. Biomass burning showed a greater impact in autumn (23%) and winter
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51 491 (21%) due to straw burning and local heating, but its proportion is lower in spring (i.e.,
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53 492 16%). These above common source categories play important roles in a particular
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56 493 season. In contrast, considering that secondary nitrate, secondary sulfate, and soil &
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59 494 dust sources only contribute to PM_{2.5} and do not contribute to CO₂; and that CO₂-

495 sources only have an effect on CO₂ concentration. Therefore, the above four sources
 496 were not identified as common sources with ensemble impacts on PM_{2.5}-CO₂. Overall,
 497 compared to PM_{2.5} source contributions, the ensemble results (summarized in Table 1)
 498 showed a moderate contribution increase in biomass burning, industrial emissions, coal
 499 combustion, and vehicle exhaust. With regard to the sources impacts of CO₂, biomass
 500 burning, industrial emissions, coal combustion, and vehicle exhaust increased by 1.4,
 501 1.4, 1.1, and 2.6 times.



502
 503 **Fig. 7.** Ensemble source impacts on PM_{2.5}-CO₂. (a): Average source impacts on PM_{2.5}-CO₂ during
 504 the sampling period. (b): Source impacts on PM_{2.5}-CO₂ in spring, summer, autumn, and winter,
 505 respectively.

507 4. Conclusions

508 Promoting the simultaneous reduction of CO₂ and air pollutants emissions are an
 509 inevitable choice for future climate and environmental governance. In this study, we
 510 investigated the concentration characteristics and source impacts of PM_{2.5}, SO₂, NO_x,
 511 In-VOCs and CO₂ using a combination of 2-year ground-based online measurements
 512 and model calculations. Furthermore, an ensemble source apportionment method was
 513 used to quantify the common source impacts of SO₂-NO_x-In-VOCs and PM_{2.5}-CO₂.

1 514 The results indicated that combustion-related sources (including coal combustion,
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3 515 industrial emissions, vehicle exhaust, and biomass burning) have significant impacts on
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6 516 multiple pollutants, and the focus of pollution prevention and control should be changed
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9 517 accordingly for different pollutants. For example, in order to reduce the secondary
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12 518 components of particulate matter, focusing on the source categories that play leading
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15 519 roles in the SO₂-NO_x-In-VOCs emissions, including vehicle exhaust (35%), industrial
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17 520 emissions (31%), biomass burning (18%), and coal combustion (16%), as estimated in
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20 521 this study. From the perspective of PM_{2.5}-CO₂ prevention, vehicle exhaust in spring,
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23 522 summer, and autumn was the key contributor (33%, 44%, 34%). In winter, the primary
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26 523 control targets should be industrial emissions and coal combustion (35% and 24%). The
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29 524 technique of ensemble source apportionment can prioritize the identification of sources
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32 525 that contribute the most to pollutant emissions when selecting greenhouse gas emission
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35 526 reduction measures, which can not only reduce pollutant emissions, but also bring
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37 527 carbon emission reduction benefits, so as to achieve higher air quality co-benefits.

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39 528 This paper provides a feasible method for source tracing based on online observation
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42 529 dataset of PM_{2.5} and CO₂. In prospective studies, the method can also be extended to
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45 530 multi-field collaborative trace studies, which may have significant potentials. We
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48 531 believe the findings in this study will contribute to a better understanding of the
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51 532 reduction of emission sources in environments, and provide quantitative scientific
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53 533 information for cleaner production.

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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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