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Changes in source contributions to particle number concentrations after the COVID-19 outbreak: Insights from a dispersion normalized PMF

Qili Dai, Jing Ding, Congbo Song, Baoshuang Liu, Xiaohui Bi, Jianhui Wu, Yufen Zhang, Yinchang Feng, Philip K. Hopke

A dispersion normalized PMF was applied to PNCs dataset.

- DN-PMF enhanced the diel emission pattern and directionality of sources.
- Traffic emissions decreased by more than 40% after the COVID-19 outbreak.

GRAPHICAL ABSTRACT

Factor analysis models use the covariance of measured variables to identify and apportion sources. These models, particularly positive matrix factorization (PMF), have been extensively used for analyzing particle number concentrations (PNCs) datasets. However, the variation of observed PNCs and particle size distribution are driven by both the source emission rates and atmospheric dispersion as well as chemical and physical transformation processes. This variation in the observation data caused by meteorologically induced dilution reduces the ability to obtain accurate source apportionment results. To reduce the influence of dilution on quantitative source estimates, a methodology for improving the accuracy of source apportionment results by incorporating a measure of dispersion, the ventilation coefficient, into the PMF analysis (called dispersion normalized PMF, DN-PMF) was applied to a PNC dataset measured from a field campaign that includes the Spring Festival event and the start of the COVID-19 lockdown in Tianjin, China. The data also included gaseous pollutants and hourly PM2.5 compositional data. Eight factors were resolved and interpreted as municipal incinerator, traffic nucleation, secondary inorganic aerosol (SIA), traffic emissions, photonucleation, coal combustion, residential heating and festival emissions. The DN-PMF enhanced the diel patterns of photonucleation and the two traffic factors by enlarging the differences between daytime peak values and nighttime concentrations. The municipal incinerator plant, traffic emissions, and coal combustion have cleaner and more clearly defined directionalities after dispersion normalization. Thus, dispersion normalized PMF is capable of enhancing the source emission patterns.
1. Introduction

Positive associations between ambient particulate matter (PM) pollution and subsequent detrimental health outcomes have been well documented in many epidemiologic studies (Song et al., 2017; Yin et al., 2019). Given the severe fine PM (PM$_{2.5}$, with aerodynamic diameter less than 2.5 μm) pollution in China, PM$_{2.5}$ was ranked as the leading mortality risk factor in 2015 (Cohen et al., 2017). Compared with larger sized particles with higher mass concentrations, smaller particles have lower mass concentrations but higher particle number concentrations (PNCs) and larger surface areas that are more likely resulting in increased adverse health effects (Meng et al., 2013; Yin et al., 2019). A recent time-series analysis in Northern China (Shenyang) reported that short-term exposure to increased PNCs for particles with size less than 0.5 μm were significantly associated with total and cardiovascular mortality (Meng et al., 2013). They found that these associations between mortality and PNCs tended to be independent of particle mass concentrations and simultaneous exposures to gaseous pollutants. Previous studies also highlighted that the limited availability of size-resolved PMC measurements prevented obtaining adequate epidemiologic evidence regarding associations with PNCs (Ibald-Mulli et al., 2002; Meng et al., 2013; Ibald-Mulli et al., 2002), particularly for source specific PNCs.

At the end of 2019, a novel coronavirus (COVID-19) was reported in Wuhan, China and quickly spread since the outbreak was coincident with the Chinese Spring Festival human migration (Huang et al., 2020). To limit the spread of COVID-19, strict control measures were enforced across the country that reduced transportation, economic and social activities. Most people were self-isolated at home. Emissions reductions from anthropogenic sources were reported to have substantially improved air quality globally (Isaifan, 2020; Venter et al., 2020). For example, air quality in China has improved with the reduction of non-essential industrial and motor vehicle usage as reported by Liu et al. (2020), who has investigated the spatial and temporal characteristics of nighttime light radiance and air quality index values before and during the pandemic in mainland China. The COVID-19 outbreak provides an opportunity to evaluate the public health benefits of air quality improvement. A recent study found that COVID-19 forced-industrial and anthropogenic activities lockdown are likely have saved more lives by preventing ambient air pollution than by preventing infection (Wu et al., 2020). Their cross-sectional study conducted in the United States indicated that an increase of only 1 μg m$^{-3}$ in PM$_{2.5}$ was associated with an 8% increase in the COVID-19 death rate. Thus, an emphasis is needed on the importance of continuing to enforce existing air pollution regulations to protect human health both during and after the COVID-19 crisis. The environmental impact of the pandemic is of particular interest to governments and the public since it is crucial for developing post-pandemic pollution control strategies. However, there are no reports to date on changes in source contributions to health risk relevant PNCs during the outbreak of COVID-19.

To identify particle sources and calculate their contributions, source apportionment studies have been conducted to support control strategies that can further reduce the health burden of PM. Positive matrix factorization (PMF) has been widely applied to apportion the PNCs to their sources/formation processes (Harrison et al., 2011; Leoni et al., 2018; Ogulei et al., 2006a; Ogulei et al., 2006b; Vu et al., 2015). The observed variation of PNCs and particle size distribution are driven by source emission rates and dilution in air as well as transformation processes include nucleation, coagulation, condensation, evaporation, deposition etc. (Ketzel and Berkowicz, 2004, 2005; Kumar et al., 2011; Lorelei de Jesus et al., 2020). Dilution is recognized as a crucial process that induces other processes to alter the particle number and size distributions (Gidhagen et al., 2005; Jacobson and Seinfeld, 2004; Ketzel and Berkowicz, 2004). Kumar et al. (2011) reviewed the dynamics and dispersion modelling of nanoparticles from road traffic in the urban atmospheric environment and concluded that dilution is the most important process that needed to be considered with highest priority in dispersion models irrespective of any spatial scales. Freshly emitted particles and new particles formed via nucleation process are diluted in the air and undergone transformation processes. The conventional use of PMF to apportion PNCs is to extract size distribution profiles from the observed particle size distributional data on the basis of the internal covariance of particles from different detected sizes. Due to the variation in dispersion, some of the information content in the observation data will certainly be lost. To investigate the changes in source contributions to PNCs after the outbreak of COVID-19, a newly proposed dispersion normalized PMF (DN-PMF) that incorporated the ventilation coefficient (VC) into the PMF analysis aimed of reducing meteorological influence on the analysis was applied to a PNCs dataset measured from January 15, 2020 to February 13, 2020 that included the start of the COVID-19 lockdown in Tianjin, China. The database also included pollutant gases and hourly PM$_{2.5}$ speciated composition data.

2. Methodology

2.1. Site description and instrumentation

Data were collected at an air quality supersite (117°24′N 38°59′E) on the campus of Nankai University in the jinan district of Tianjin, China. The supersite is ~25 km southeast of the Tianjin city center and ~30 km west of Bohai Bay (Fig. S1). It is a suburban site surrounded by several universities, and distant from major highways and high traffic zones. A suite of instruments was operated in the supersite building with sampling inlets on the roof terrace.

Particle number concentrations with sizes ranging from 7.2 nm to 778 nm were measured using a universal scanning mobility particle sizer spectrometer (U-SMPS) (PALAS, Germany). Other air pollutants, including PM$_{1}$ and PM$_{2.5}$, sulfur dioxide (SO$_2$), nitrogen oxides (NO$_x$), carbon monoxide (CO) and ozone (O$_3$) were also recorded hourly. Organic carbon (OC) and element carbon (EC) were measured hourly using a semicontinuous thermal-optical carbon analyzer (Focused Photonics Inc., China) and water-soluble ions (SO$_4^{2-}$, NO$_3^-$, NH$_4^+$, Cl$^-$, K$^+$, Mg$^{2+}$, and Ca$^{2+}$) by an in situ ion chromatograph (Thermo Fisher Scientific Inc., USA). Inorganic elements were measured hourly using an X-ray fluorescence instrument (Focused Photonics Inc., China).

Meteorological parameters including relative humidity (RH), temperature (T), and wind direction/wind speed were measured using an automatic meteorological observation system (LUFFT Inc., Germany). Solar radiation was recorded with a sun photometer (Kipp & Zonen Inc., Netherlands). Mixing layer height (MLH) was measured using ceilometer (Vaisala Inc., Finland) at the Tianjin Eco-Environmental Monitoring Station. Daily checks were conducted for all instruments to ensure their normal functioning. The detailed instrumentation information is presented in Dai et al. (2020) and details regarding the quality assurance and control are also available in Tian et al. (2020).
2.2. Dispersion normalized positive matrix factorization

PMF effectively apportioned total PNC to the extracted factors (sources/processes) on the basis of the covariances among the observed variables. The variation resulted from dilution could be reduced by incorporating the ventilation coefficient into the PMF analysis as described by Dai et al. (2020). Briefly, dilution can be estimated as a ventilation coefficient, an index of the potential volume into which source material undergoes dilution after release into ambient air per unit time. The VC for the area in which measurements were made is defined as the product of mixed layer height and the average wind speed during measurement time period $i$ (Asghari et al., 2009):

$$\text{VC}_i = MLH_i \times \pi_i$$

(1)

where $MLH_i$ and $\pi_i$ are the mixed layer height and mean wind speed during time period $i$, respectively. VC values were calculated hourly to match the time resolution of the other variables. To reduce the influence of local dispersion, the measured data were normalized to the average VC over the measurement campaign to obtain dispersion normalized concentrations:

$$C_{VCj} = \frac{C_i}{\text{VCmean}}$$

(2)

where $C_{VCj}$ is dispersion normalized concentrations, $C_i$ is the concentration observed during period $i$, and $\text{VCmean}$ is the average value of VC over the entire measurement campaign. In this case, $\text{VCmean}$ is $582 \text{m}^2 \text{s}^{-1}$. The concentrations and uncertainties of PNC dataset were normalized on a sample by sample basis. The dispersion normalized concentrations and uncertainties were used as the input matrix to the PMF analysis.

The time series of PNC, PM1 mass concentration and VC values are shown in Fig. 1. The hourly average total PNC from 7.2 to 778 nm over the measurement campaign was $14,397 \pm 5695 \text{pt cm}^{-3}$, ranged from 5490 to 48,372 pt cm$^{-3}$. The average mass concentration of PM1 was $52.2 \pm 42.6 \mu g \text{m}^{-3}$, ranged from 1.0 to 224.1 $\mu g \text{m}^{-3}$. In general, PM1 mass concentrations increased under poor dispersion conditions (low VC values) across the whole campaign. While periods with significantly elevated total PNC, such as the afternoons of January 29th, January 30th and February 3rd, were characterized by increases of nucleation mode particles with increased VC values (increased wind speed and MLH). These afternoon nucleation processes appear to be driven by photochemistry on clean days with strong solar radiation with relatively low PM mass concentrations. Increase PM concentrations would increase the amount of particle surface area that serves as a condensation sink (McMurry and Friedlander, 1979). As shown in Fig. 2, most particles in the nucleation and Aitken modes were less positively correlated with VC than accumulation mode particles. Given that particles in different modes originated from different sources/processes (Kumar et al., 2011; Vu et al., 2015), this result highlights the fact that particles have undergone different effects of dilution within a given meteorological condition after release to or formation in the ambient air. Thus, dispersion normalized PMF was applied to PNC data to reduce the effect of dilution on source estimates.

3.2. Solution selection

Solutions using PMF with four to nine factors were explored for the original data and dispersion normalized data. The best solution with the optimal number of factors was evaluated with selection criteria of appropriately narrow distributions of scaled residuals of PNCs and the physical interpretability of factors in terms of (a) examination of size factor profiles and its association with external variables, (b) source directionality from CBPF plots, and (c) diel patterns.

Eight-factor models were finally selected as the best solutions for both the unnormalized and normalized data sets. For the DN-PMF, the source contributions were unnormalized by dividing the normalization ratio shown in Eq. (2). The eight resolved factors were interpreted as: municipal incinerator, traffic nucleation, secondary inorganic aerosol (SIA), traffic emissions, photonucleation, coal combustion, residential heating and festival emissions. The factor of municipal incinerator was method detection limit (MDL) values were replaced by half of the MDL value. The corresponding uncertainties of these values were set at five sixths of the MDL values (Polissar et al., 1998). Missing chemical species values were estimated by linearly interpolating the closet non-missing values and the corresponding uncertainties were increased by a factor of 3. All auxiliary variables were assigned as “weak” and the total PNC was set as the “total variable” with uncertainty tripled. The F-value for total PNC was used to normalize the size bin data and factor contributions. The F-value for PM2.5 was used to normalize the chemical species. The gaseous species were not normalized. The effects of measurement error and rotation ambiguity on the selected solutions were examined via the bootstrap (BS) and displacement (DISP) analyses function in PMF v5.0. The original data matrix and dispersion normalized concentrations and uncertainties were subjected to PMF analyses, hereafter called unnormalized PMF and DN-PMF.

Conditional bivariate probability function (CBPF) analysis was applied to the PMF-modelled factor contributions so as to examining the identity of factors and investigating the source directionality. CBPF plots were computed using the openair package in R and details of the CBPF analyses is available elsewhere (Carslaw and Ropkins, 2012; Uria-Tellaetxe and Carslaw, 2014).

3. Results and discussion

3.1. Overview of the measurement

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Solutions using PMF with four to nine factors were explored for the original data and dispersion normalized data. The best solution with the optimal number of factors was evaluated with selection criteria of appropriately narrow distributions of scaled residuals of PNCs and the physical interpretability of factors in terms of (a) examination of size factor profiles and its association with external variables, (b) source directionality from CBPF plots, and (c) diel patterns.

Eight-factor models were finally selected as the best solutions for both the unnormalized and normalized data sets. For the DN-PMF, the source contributions were unnormalized by dividing the normalization ratio shown in Eq. (2). The eight resolved factors were interpreted as: municipal incinerator, traffic nucleation, secondary inorganic aerosol (SIA), traffic emissions, photonucleation, coal combustion, residential heating and festival emissions. The factor of municipal incinerator was
mixed with SIA in seven-factor solution, while nine-factor solution includes an unknown factor split from the residential heating factor in eight-factor solution (Fig. S2). Constraints were applied to PM mass values to maximally pull them up for several factors that had PM values of zero, which is unlikely exist in reality, such as photonucleation. The selected eight-factor solutions were then subjected to BS and DISP tests.

PMF predicted total PNC strongly correlated with the observed values with squared correlation coefficients ($r^2$) of 0.994 for both data sets (Fig. S3). For both PMF and DN-PMF analyses, there were no DISP swaps. The BS solutions for PMF and DN-PMF had at least 75% and 95% agreement with the base case results ($n = 200$ runs), respectively. Factor size profiles and associated chemical species and other auxiliary variables profiles derived from unnormalized PMF and DN-PMF are shown in Figs. S4 and 3, respectively. The factors are interpreted and discussed individually in the following section.

3.3. Factor interpretation

3.3.1. Municipal incinerator

This factor presented three major size modes: a nucleation mode peaked at ~8–20 nm, a small sized mode peaked around 30–50 nm and an accumulation mode peaked at 100–200 nm (Fig. 3). On-line measurements of the size distribution of particles in flue and stack gas of a municipal waste incineration plant in Europe showed a bimodal size distribution with peak mode at 90–140 nm and a smaller mode at approximately 40 nm (Maguhn et al., 2003). It was also found that particles tend to growth by absorption and coagulation after leaving the boiler. The chemical species profile had high contributions of Cr and Cu with narrow DISP intervals (Fig. 3), which are tracers of municipal waste incineration. Lu et al. (2018) reported abundant Cr in bottom ash from plastic materials incineration. The CBPF plot of incinerator also shows a very clear NW wind direction pointing to the municipal incineration plant. Thus, this factor was assigned to municipal incinerator. On average, municipal incinerator had the lowest contribution to total PNC during the campaign (3.2%, Fig. S5).

3.3.2. Traffic nucleation and traffic emissions

Traffic has been recognized as one of the major sources of PNC. Two traffic factors were identified as traffic nucleation and traffic emissions with particle major size modes about 10–30 nm and 30–150 nm, respectively. As been previously reported in literature (Morawska et al., 2008; Vu et al., 2015), particles formed from the hot exhaust gases and consequently cool down and condense to produce large numbers of particles in the nucleation mode with sizes less than 30 nm. The size of the traffic nucleation factor mode is similar with that observed in London, UK (Harrison et al., 2011), Brisbane, Australia (Friend et al., 2012), Fahaheel, Kuwait (Al-Dabbous and Kumar, 2015) and other locations (Rivas et al., 2020; Vu et al., 2015). The particle size range of traffic emissions is also within the size range as previously reported in other studies, in which exhaust particles from vehicles are reported in the

![Fig. 1. Time series of PM$_1$ mass concentrations, total PNC, ventilation coefficient (VC) (a), and size spectra of PNC (b).](image)
size range of 30–500 nm (Casati et al., 2007; Rivas et al., 2020; Vu et al., 2015; Zhang et al., 2020). These particles are likely dominated by agglomerates of solid carbon materials. The diel pattern of the traffic nucleation factor has a sharp morning rush hour peak (6:00–8:00 am) and had enhanced contributions more broadly distributed beginning in the afternoon. While the traffic emissions factor showed a strong peak in the morning after the rush hour (8:00–10:00 am) and a minor peak in the evening rush hour. Both traffic factors had high concentrations of NO\textsubscript{2} and explained 5–10% of the EC variation, indicating its traffic exhaust nature including diesel emissions (Zhang et al., 2020). The presence of some O\textsubscript{3} and radiation in the traffic emissions factor suggests that particles exhaust from vehicle emitted in the morning were possibly subjected to photochemistry processes.

The CBPF plot of traffic nucleation indicates its directionality corresponded to the roads located ~1.5 km and 3.0 km southwest of the measurement site (Fig. S1). The traffic emissions factor has a stronger association with southeasterly winds and a predominant occurrence for wind speeds between 3 and 5 m s\textsuperscript{-1}. There are two major highways situated ~18 km east of the measurement site and a highway extending to the southeast. Both traffic factors are heavily affected by local traffic emissions.

### 3.3.3. Secondary inorganic aerosol (SIA)

The SIA (ammonium nitrate and sulfate) had three modes in the particle number size profiles with the major size mode between 180 and 600 nm, an Aitken mode and an ultrafine mode, which is similar with observations such as in Rochester, NY (Squizzato et al., 2019). The explained variations of particles in the accumulated mode increased with increasing size. The presence of high nitrate, sulfate and ammonium in the species profile support its assignment as secondary inorganic material. The wide particle size range of SIA factor suggests that it originated from both gas-to-particle conversion of local NO\textsubscript{X} and regional transportation of sulfate. Some EC present in the species factor may be caused by the condensation of secondary material on freshly emitted EC particles. The morning peak of SIA at 9:00–10:00 am likely resulted from downmixing of transported secondary particles from aloft after the breakup of overnight inversions. The broad afternoon minimum was likely due to the mixing layer height dynamics. The CBPF plot of SIA shows higher probability values with moderate speed winds (~2 m s\textsuperscript{-1}) come from downtown Tianjin (NW) more than other directions, highlighting the regional transport nature of SIA during heavy particulate pollution episodes. It was the largest contributor to PM mass concentration but accounted for only 6.2% of total PNC over the measurement campaign.

### 3.3.4. Photonucleation

In addition to new particle formation from traffic emissions with a dominant particle size ranged from ~10–30 nm as typically observed during rush hour as discussed above, a factor with particle size peaking in the nucleation mode size range (<20 nm), and characterized by high explained variations of O\textsubscript{3} and solar radiation with narrow DISP bands was identified as new particle formation (NPF) through photochemistry. It shows a strong sharp increase around midday (12:00) concurrent with the highest solar intensities that drive photochemical processes. Photonucleation occurred primarily during clean days with low PM mass concentrations (Rivas et al., 2020). The low PM concentrations result in low condensation sinks and allow for NPF to occur (McMurry and Friedlander, 1979). As suggested by the CBPF plot, this factor is associated primarily with relatively higher wind velocities for east-southeasterly winds and southerly winds. The CBPF plot of photonucleation overlaps with traffic nucleation in south-south-west direction, suggesting air masses from this direction likely facilitate NPF. Easterly winds often associated with relatively clean Bohai Bay air possibly favor the nucleation process because of its low condensation sink. Interestingly, the non-local feature of photonucleation tends to indicate that the measurement site is likely well separated from the emission sources. On average, this factor contributed 7.8% of total PNC.

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**Fig. 3.** Particle number size distribution profiles (left panel), chemical species profiles (middle panel) and other auxiliary variables profiles (right panel) derived from DN-PMF. The solid black dot lines and grey bars are the base values, the unfilled red circle lines are the mean fractional displacement (DISP) values, the asymmetric error bars represent the maximum and minimum DISP values. The pink triangles represent the % explained variation. Particle number size distribution data were normalized as fractions on the total PNC. The other variables are shown in their corresponding values. Among the chemical species data, Ba and As were scaled up by 100 to better visualize their values (dark grey bars).
3.3.5. Coal combustion
Coal combustion was reported as an important source of ambient particles with size ranged from nanoparticles to coarse particles, depending upon various factors such as coal type, combustion condition, dilution ratio and residence time, and pollution control device (Vu et al., 2015). The formation of particles emitted from coal combustion involved in complex chemistry as it will undergo vaporization-nucleation-condensation-growth processes (Carbone et al., 2010). Lipsky et al. (2002) reported a nucleation peak mode around 10 nm in the stack plume of coal combustion. Particles tend to shift to large sizes after release into ambient air experiencing dilution, cooling, and condensation/coagulation processes. An experimental measurement of pulverized coal combustion suggested that ultrafine particles exhibited a unimodal distribution under 1500 K with size peak shifting from 10.76 nm to 38.46 nm as time evolved (Gao et al., 2017). There is a paper mill with hot steam supplied by pulverized coal boilers situated ~6.5 km SE from the measurement site (Fig. S1). The CBPF plot presents a clear SE direction linking this factor to the paper mill. Thus, this factor with particle sizes peaking around 30 nm was attributed to coal combustion. The gaseous and species profiles corroborate the assignment of this factor to coal combustion since it had relatively high concentrations of NO2, EC, SO4 2− and NO3 −, and explained some of the SO2, CO, and As variations. The diel pattern presents a middle day peak as the particles can be transported by the afternoon sea breeze from Bohai Bay.

3.3.6. Residential heating
The factor has a size mode ranging from ~50 nm to 300 nm and peaked in the accumulation mode (~120 nm). It was identified as residential heating. Its size distribution is similar with particles emitted from residential wood pellet boilers (Chandrasekaran et al., 2011; Wang et al., 2018). It is associated with some SO2, NO2, and CO with tight DISP bands, and moderate loading of PM mass concentration. The chemical species profile also supports coal as it explained ~15% of Cl − and ~10% OC with small intervals in mass fractions. Cl is a common marker of coals in northern China (Yu et al., 2013; Li et al., 2019).

The diel pattern for this factor presents a clear morning peak (7:00–8:00 am) during stable atmosphere conditions with low wind speeds. PNC of residential heating during nighttime appears to be higher than that during daytime. The CBPF plot of residential heating factor has an association with all wind directions and low wind speeds (<2 m s−1).

3.3.7. Festival emissions
The festival emissions factor is attributed largely on the basis of strong chemical association with OC, EC, NO3 −, SO4 2−, K +, Mg2+, Cl −, and Ba. It explained relatively large proportions of the variation in OC, EC, K+, Mg2+, Ba and As. K and Ba are tracers of firework emissions (Kong et al., 2015; Tian et al., 2014) and Mg is indicative of firework emission as well (Kong et al., 2015). High concentrations of these melliferous species were observed during firework displays days in many locations (Joshi et al., 2019; Kong et al., 2015; Moreno et al., 2010; Tian et al., 2014). These species have relatively narrow DISP intervals in the species profiles supporting the inclusion of firework emissions in this factor. OC, EC and sulfate in the profile were also likely emitted from coal/wood burning in large bond fires to celebrate the festival as reported in Dai et al. (2020). Fireworks were generally banned in urban areas and Tianjin implemented a no-fireworks policy this year. However, people still likely utilize fireworks during these holiday periods. Based on the CBPF plot, the origins of these particles were located in residential areas where coal/biomass was burned for heating/cooking during the Spring Festival. People were required to stay at home after the implement of lockdown measures to prevent the spread of COVID-19 and thus, more people were in residential areas throughout the day requiring more heating and cooking than under normal circumstances. Fireworks emissions were correlated with residential coal/biomass burning. It should be noted that this factor contributed significantly to PM mass concentration, particularly PM1, in the period after the start of the Spring Festival (January 24, 2020).

3.4. Source directionality and diel emission patterns
Since the DN-PMF reduced the influence of dispersion, the CBPF plots derived from DN-PMF results tend to have better defined directionality than the regular PMF based plots. Fig. 5 presents the CBPF plots for the eight factors resolved from unnormalized PMF and DN-PMF. The directionality of municipal incinerator plant, pulverized coal combustion plant, and traffic emission are clearer in the dispersion normalized results.

Fig. 4 presented the diel patterns for the eight factors resolved from unnormalized PMF and DN-PMF. Generally, pollutants are more abundant during nighttime due to the low VC values given low wind speeds and mixed layer heights. Concentrations tend to decrease in the afternoon when the VC values are high. The DN-PMF was expected to scale down the nighttime concentrations and scale up the middle day

![Fig. 4. Diel profiles of residential heating (a), traffic nucleation (b), photoneucleation (c), municipal incinerator (d), secondary inorganic aerosol (e), traffic emissions (f), coal combustion (g), and festival emissions (h) estimated using PMF and DN-PMF.](image-url)
concentrations. The DN-PMF enhanced the diel pattern of photonucleation by elevating its midday peak when VC values are high and lowering down its nighttime values when VC values are low. The nighttime peak of pulverized coal combustion present in the unnormalized results was obviously lowered compared to the daytime values. It is our understanding that the paper mill only operates on a 12-h per day basis so their emissions would be anticipated to diminish at night. DN-PMF also enhanced the morning and evening rush hour peaks of traffic emissions. The morning and evening rush hour peaks of traffic emission increased significantly compared with the values at other times. Through visual inspection, there are no notable changes in the diel profiles of municipal incinerator, secondary inorganic aerosol, and festival emissions in the dispersion normalization results. The residential heating factor presented a strong morning peak likely the result of enhanced transport supported by increased wind speed. The DN-PMF decreased the wind effect by reducing its peak value and brought down its evening values.

3.5. Changes in source contribution after COVID-19 outbreak

This measurement campaign included the unusual event of the Chinese Spring Festival (SF) and lockdown measures enforced to prevent the spread of COVID-19. Unlike the common sources under the ‘business as usual’ scenario prior to January 24, there was a change in the number of sources due to the fireworks displayed during the SF. Source emission strengths also changed after the COVID-19 outbreak as emergency responses were implemented beginning on January 25 in Tianjin. To evaluate the effects of response measures on PNCs, the whole campaign was divided into three periods: before SF, during SF, and after SF to assess the changes in source contributions to total PNC, as shown in Fig. 6.
Residential heating was the largest source of PNC before the SF and the second largest source during and after the SF, accounting for an average of 23.6%, 21.8%, and 21.2% of the total PNC, respectively. Combining traffic nucleation and traffic emissions, traffic was the predominant source of PNC over the whole campaign with average fractions of 36.7%, 26.5%, and 33.2% of the total PNC before, during, and after the SF, respectively. Contribution of the municipal incinerator decreased continuously from 3.6% of total PNC before the SF to be the smallest contributor (2.7%) after the SF. Similarly, SIA also decreased in importance over time. After the SF, the contribution of SIA declined to 3.3% as the next to smallest source. Festival emissions accounted for 11.7% of the total PNC during the SF.

After the outbreak of COVID-19, the PNCs of traffic emissions and traffic nucleation dropped from 2907 and 3222 pt cm\(^{-3}\) to 1727 and 1814 pt cm\(^{-3}\), respectively, representing decreases of 44% and 41%. The reduced traffic volume after the implement of lockdown measures was responsible for these reductions in traffic contributions. The NO\(_x\) emitted from traffic were also reduced. The decline of 46% in SIA associated particles after the COVID-19 outbreak was likely due to the reduced local production of nitrate from NO\(_x\), along with decreased transported SIA from areas with closed industrial facilities. The number of photonucleation produced particles increased from 750 pt cm\(^{-3}\) (4.5%) before the SF to 1503 pt cm\(^{-3}\) after the SF (11.3%).

Coal combustion, together with residential heating was the predominant source of PNC after the SF (43.2%). Therefore, further reduction in coal burning pollution is needed to reduce PNC and improve public health.

### 4. Conclusions

To reduce the influence of meteorology on quantitative source estimates, dispersion normalized PMF incorporating data normalized with the ventilation coefficient into PMF analyses, was applied to a PNCs dataset measured from a field campaign that includes the Spring Festival event and the start of the COVID-19 outbreak in Tianjin, China. In addition to PNCs data, other variables include solar radiation, gaseous pollutants, particle mass concentrations and chemical compositional data were measured and included to facilitate the interpretation of factors. The PNCs dataset combined with these additional variables were normalized by VC values and then subjected to PMF analysis. Eight factors were resolved and labelled as municipal incinerator, traffic nucleation, secondary inorganic aerosol (SIA), traffic emissions, photonucleation, coal combustion, residential heating and festival emissions.

To examine the effects of reducing meteorological influence on estimated source patterns, the diel pattern and source directionality for each source derived from conventional PMF (unnormalized) and DN-PMF were compared. The DN-PMF enhanced the diel patterns of photonucleation and the two traffic factors by sharpening the differences between daytime peak values and nighttime concentrations. After dispersion normalization, the PMF results yielded better defined directionality of sources. The directionality of municipal incinerator plant, traffic emissions and coal combustion plant are cleaner using the dispersion normalized results. Thus, the DN-PMF was able to improve the apportionment of PNCs to its sources.

Given the significant reduction of traffic volume after the implement of lockdown measures, the PNCs of traffic emissions and traffic nucleation decreased by 44% and 41%, respectively, after the outbreak of COVID-19. Nevertheless, traffic was still to be the predominant source of PNC after the COVID-19 outbreak.

### CRediT authorship contribution statement

Qili Dai: Methodology, Writing - original draft preparation
Jing Ding: Assisting in sample collection, Data curation
Congbo Song: Result interpretation, Writing - review & editing
Baoshuang Liu: Writing - review & editing
Xiaohui Bi: Resources, Investigation
Jianhui Wu: Sample collection and analysis
Yufen Zhang: Writing - review & editing
Yinchang Feng: Resources, Supervision and leadership
Philip K Hopke: Result interpretation, 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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Appendix A. Supplementary data

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