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Hourly organic tracers-based source apportionment of PM$_{2.5}$ before and during the Covid-19 lockdown in suburban Shanghai, China: Insights into regional transport influences and response to urban emission reductions

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**HIGHLIGHTS**

- Covid lockdown lowered PM$_{2.5}$ and NO$_x$ while increased O$_3$ levels in suburban Shanghai.
- Nitrate was the most prominent contributor to PM$_{2.5}$ reduction during lockdown.
- Lockdown led to reduced PM$_{2.5}$ contributions by 8 primary and 5 secondary sources.
- Contribution percent drop from industrial and vehicular sources were as high as 87%.
- Lockdown effects on SOA formation likely differ in suburban and urban environments.

**GRAPHICAL ABSTRACT**

**ABSTRACT**

During the Covid-19 outbreak, strict lockdown measures led to notable reductions in transportation-related emissions and significantly altered atmospheric pollution characteristics in urban and suburban areas. In this work, we compare comprehensive online measurements of PM$_{2.5}$ major components and organic molecular markers in a suburban location in Shanghai, China before lockdown (Dec. 28, 2019 to Jan. 23, 2020) and during lockdown (Jan. 24 to Feb. 9, 2020). The NO$_x$ levels declined sharply by 59% from 44 to 18 ppb during the lockdown, while O$_3$ rose two times higher to 42 ppb. The PM$_{2.5}$ level dropped from 64 to 49 μg m$^{-3}$ (24%). The major components all showed reductions, with the reduction of nitrate most prominent at −58%, followed by organics at −19%, and sulfate at −17%. Positive matrix factorization analysis identifies fourteen source factors, including nine primary sources and five secondary sources. The secondary sources consist of sulfate-rich factor, nitrate-rich factor, and three secondary organic aerosol (SOA) factors, with SOA_I being anthropogenic SOA, SOA,II associated with later generation products of organic oxidation, and SOA,III being biogenic SOA. The combined secondary sources contributed to 69% and 63% (40 and 22 μg m$^{-3}$) of PM$_{2.5}$ before and during lockdown.
lockdown, respectively, among which the reductions in the nitrate-rich (~55%) factor was the most prominent. Among primary sources, large reductions (~80%) were observed in contributions from industrial, cooking, and vehicle emissions. Unlike some studies reporting that the restriction during the Covid-19 resulted in enhanced secondary sulfate and SOA formation, we observed decreases in both secondary inorganic and SOA formation despite the overall elevated oxidizing capacity in the suburban site. Our results indicate that the formation change in secondary inorganic and organic compounds in response to substantial reductions in urban primary precursors are different for urban and suburban environments.

1. Introduction

Fine particulate matter (i.e., PM$_{2.5}$, aerodynamic dimer less than 2.5 μm) is known to be associated with adverse human health effects and decreasing visibility (Nel, 2005; Pöschl, 2005; Seinfeld and Pandis, 2006; Friedlander and Lippmann, 1994). PM pollution in China has received significant attention, and numerous studies have been conducted to characterize the sources and formation pathways leading to excessive PM$_{2.5}$ concentrations. PM$_{2.5}$ can be either emitted primarily from natural and anthropogenic sources (Hu et al., 2010; Zhu et al., 2018) or produced secondarily via oxidation of gas precursors (Yu et al., 1999; Kroll and Seinfeld, 2008). Generally, vehicle emission, industrial processes, biomass burning, coal combustion, and cooking are major primary sources. Recent studies have highlighted the important role of enhanced secondary aerosol formation in driving severe haze pollution in China (Huang et al., 2014; Sun et al., 2014). Acquiring quantitative understanding of contributing sources is of fundamental importance to formulating control measures in reducing PM pollution. The secondary sources are diverse in their precursors and formation pathways. By the precursor chemical nature, they could be broadly classified as secondary inorganic aerosols (SIA) and secondary organic aerosols (SOA). Our knowledge of SOA sources has been rapidly expanding but remains limited due to the large number of organic precursors and each precursor having a multitude of formation processes.

Receptor models, e.g., chemical mass balance and positive matrix factorization (PMF), are widely used to apportion the sources of PM$_{2.5}$ using ambient chemical measurements at the receptor sites (Wong et al., 2019; Wang et al., 2019). Previous source apportionment studies are mainly based on offline filter measurements. The low-time resolution nature of such measurements (typically at a daily time resolution) limits our ability to study the diurnal pattern of the major sources and the rapid developing processes of haze formation in ambient environments. Recently, high-time resolution instruments, such as the Aerodyne Aerosol Mass Spectrometer (AMS) and Aerosol Chemical Speciation Monitor (ACSM), have been widely deployed to characterize aerosol composition and identify PM sources (Takegawa et al., 2009; Sun et al., 2013). However, the individual mass fragment ions by AMS and ACSM are less specific than molecular markers for source tracking and identification. In comparison, the thermal desorption aerosol gas chromatography–mass spectrometry (TAG) system, a recently commercialized instrument, provides hourly measurements of individual organic molecular markers (Williams et al., 2006; Zhao et al., 2012). The molecular data significantly improves our ability in differentiating organic aerosol sources and in tracking SOA sources specific to common organic precursors, e.g., isoprene, monoterpenes, and toluene (Lyu et al., 2020; He et al., 2020; Li et al., 2020; Wang et al., 2017a).

In the wake of the outbreak of COVID in late December 2019, China implemented strict controls (lockdown) nationwide starting on January 24, 2020 to drastically restrict the population’s outdoor activities and moving around for both within cities and between cities (Tian et al., 2020). Significant reductions in primary pollutants such as CO, NO$_x$, and anthropogenic volatile organic compounds (VOCs) were observed in varied environments (Huang et al., 2020, 2021; Jia et al., 2020; Chen et al., 2020; Sun et al., 2020; Chang et al., 2020). Many studies have investigated the impacts of lockdown controls on the reductions of PM$_{2.5}$ levels in megacity environments, where NO$_x$ and other gaseous pollutants (e.g., CO) play a major role in PM pollution (Huang et al., 2020; Xu et al., 2020; Chang et al., 2020). Expectedly, increased O$_3$ concentrations has been observed due to the weaker NO titration during the lockdown periods (Wang et al., 2022; Xu et al., 2020; Chang et al., 2020; Huang et al., 2020; Sun et al., 2020). The lockdown measures coupled with the more oxidative atmospheric conditions were shown to have noticeably increased the contributions of secondary sulfate and organic aerosols in urban locations, offsetting the large reductions in vehicle-related and other primary emissions (Le et al., 2020; Chang et al., 2020). In comparison to urban environments, the suburban regions are generally featured by relatively low levels of PM and NO$_x$ while O$_3$ levels are on the rise in recent years (Wang et al., 2017b). Such distinctive pollution characteristics in suburban regions may lead to different responses of PM pollution to control measures. Thus, field observation data in suburban environments are needed to help us understand the impact of reduction of urban pollution sources on the surrounding areas.

Here, we present a case study examining the impact of emission reduction from the COVID lockdown on PM$_{2.5}$ level and the underlying contributing sources at a suburban site of Shanghai, China. The measurement campaign covered before the lockdown (Dec. 28, 2019 to Jan. 23, 2020) and during the lockdown (Jan. 24 To Feb. 9, 2020). Detailed online measurements of PM$_{2.5}$ and its major components, along with the organic molecular markers were made. The objectives are to comparatively characterize PM$_{2.5}$ chemical compositions and to identify the potential sources and contribution variations from before to during control periods. The direct field observation evidence of the atmospheric responses under different ambient environments may provide new insights for developing more effective air pollution mitigation measures.

2. Method and materials

2.1. Sampling site

The campaign site was on the top of a 4-floor building (~15 m, above ground level) at the Dianshan Lake (DSL) supersite (31.09° N, 120.98° E) in western Shanghai, China. A map of the location is shown in Fig. S1. The sampling site is ~50 km away from downtown Shanghai, surrounded by farms and vegetable fields, with relatively low influences of local anthropogenic sources.

2.2. Sample collection and analysis

An array of online instruments were available at the site to monitor meteorological parameters (temperature, relative humidity (RH), and wind speed and direction), gaseous pollutants (i.e., NO, NO$_2$, SO$_2$, CO, and O$_3$), PM$_{2.5}$ mass, major chemical components, and organic molecular compounds. In this work, we focus measurements during two periods of time: before the lockdown phase (Dec. 28, 2019 to Jan. 23, 2020), and during the lockdown phase (Jan. 24 to Feb. 9, 2020).

The hourly concentrations of 96 organic molecular compounds in PM$_{2.5}$ were measured every 2 h by the TAG system (Aerodyne Research Inc, USA). The instrument was configured to collect PM$_{2.5}$ for an hour while the subsequent hour was spent on the gas-chromatographic (GC) separation and mass spectrometric detection of the sample. Thus, there are 12 sets of hourly TAG-measured organic compound concentrations.
3. Results and discussion

3.1. Measurement overview of the field campaign

The temporal variations of meteorological parameters, gaseous pollutants, along with the PM$_{2.5}$ and its major components, and the TAG-measured molecular markers are shown in Fig. 1. The average temperature was 8.0 ± 3.9 °C before the lockdown and decreased to 6.0 ± 3.3 °C during the lockdown, while RH and wind speed did not show a discernible change during the lockdown. The northwestern wind was dominant most of the time before the lockdown, while it occurred less frequently, and the wind direction shifted to the eastern/northeastern during the lockdown. The ventilation coefficient (wind speed multiplied by the PBL height), an index to examine the total atmospheric diffusion capacity (Tiwari et al., 2016; Dai et al., 2020) increased from 840 ± 299 m$^2$ s$^{-1}$ before the lockdown to 1117 ± 919 m$^2$ s$^{-1}$ during the lockdown, suggesting the slightly better pollution diffusion capability during the lockdown. The NO$_x$ concentration was reduced by 59% from 44 ± 35 ppb before the lockdown to 18 ± 4.2 ppb during the lockdown. Similarly, SO$_2$ and CO concentrations were reduced by 12% and 34%, respectively. These were mainly due to the significant reduction of the number of on-road vehicle, along with the lower productivities of power plants and the industrial sector. On the contrary, the O$_3$ concentration increased to 42 ± 11 ppb, almost two times that before the lockdown (20 ± 13 ppb), indicating enhanced atmospheric oxidation capacity during the lockdown (Fig. 2b).

The PM$_{2.5}$ concentrations decreased from 64 ± 43 μg m$^{-3}$ before the lockdown to 49 ± 31 μg m$^{-3}$ during the lockdown. The combined SIA species (i.e., SO$_2^+$, NO$_2^-$, and NH$_4^+$) accounted for 70% and 49% of PM$_{2.5}$ before and during the lockdown, respectively. The concentrations of nitrate, sulfate and ammonium were 24 ± 18 μg m$^{-3}$, 9.8 ± 5.7 μg m$^{-3}$, and 11 ± 7.5 μg m$^{-3}$ before the lockdown, and decreased by 58%, 18%, and 45% during the lockdown, respectively (Table 1). The concentrations of OC and EC were 5.6 ± 2.9 μg m$^{-3}$ and 2.6 ± 1.4 μg m$^{-3}$ before the lockdown and dropped to 4.5 ± 2.6 μg m$^{-3}$ and 1.6 ± 0.9 μg m$^{-3}$ during the lockdown, respectively. The organic matter (OM) was estimated by multiplying the OC mass with a converting factor of 1.6 (Feng et al., 2009). Unlike NO$_3^-$ and NH$_4^+$, OM did not show significant changes in either the absolute concentration or percentage contribution, with 9.0 ± 4.7 μg m$^{-3}$ (14%) and 7.3 ± 4.2 μg m$^{-3}$ (15%) before and during the lockdown, respectively. These results indicate different responses of SIA and OM to lockdown measures. Decreases were also observed for crustal material (2.49 [Si] + 1.63 [Ca] + 2.42 [Fe]) (He et al., 2020) and other elements (sum of V, Ni, Cr, Mn, Cu, Zn, As, Se, Ba, and Pb), with drops of 50% and 40%, respectively. In contrast, elemental K increased by 76% from 0.53 ± 0.39 μg m$^{-3}$ before the lockdown to 0.96 ± 0.99 μg m$^{-3}$ during the lockdown, reflecting influence of the uncontrollable use of fireworks during the Chinese New year holiday period.

The TAG-measured molecular markers were categorized into ten groups according to their common structures and sources, including SOA tracers, C$_{3,5}$ dicarboxylic acids (DCAs) and hydroxyl DCAs (hDCAs), C$_{6-9}$ DACs & hDCAs, aromatic acids, biomass burning tracers (BB tracers), fatty acids, saccharides, alkanes, hopane & sterane, and poly-cyclic aromatic hydrocarbons (PAHs). Detail descriptions of individual categories are provided in Text S1. Among the individual organic compounds, the groups of SOA tracers and DCAs & hDCAs are of secondary origins, while others are mainly emitted through primary processes. The average concentrations of a total of 53 polar and 43 non-polar molecular markers are shown in Table S2. Before the lockdown, the sum concentration of TAG-measured molecular markers was 501 ± 368 ng m$^{-3}$, fairly comparable to those observed in the same period in urban Shanghai (Zhu et al., 2020). The concentration decreased by 47%–264 ± 172 ng m$^{-3}$ during the lockdown.

The more abundant TAG-measured organic compounds are those associated with primary sources, with saccharides being the most abundant, accounting for 23% and 25% of the total TAG-measured...
compounds before and during the lockdown, respectively (Fig. 1d). Fatty acids, BB tracers, and aromatic acids also comprised a considerable fraction, with average contributions of 18%, 9.9%, and 9.1% before the lockdown, respectively. Their mass concentrations decreased by 78%, 20%, and 37% during the lockdown, while their percentage contributions changed in mixed directions, with fatty acids by -10%, BB tracers by +15%, and aromatic acids by +11%. This reflects the different degrees of concentration changes relative to the total TAG-measured compounds. As for the other primary categories of lesser percentage contributions (i.e., <6.0%), large reductions were observed in alkanes (81%), hopane & steranes (83%), and PAHs (73%). Among the three secondary organic categories, the concentrations of C3-5 DCAs & hDCAs were comparable to those of SOA tracers, and 4–5 times higher than C6-9 DCAs & hDCAs. During the lockdown, all these three secondary categories showed substantial decline in mass concentrations, by 43%, 40%, and 51%, respectively. The combined contributions of secondary organic compounds to OM were 1.9% before the lockdown and 1.3% during the lockdown period. These results suggest that the lockdown measures have significant impacts in lowering the primary anthropogenic emissions, which in turn have led to reduced formation of SOA during the lockdown period.

3.2. Source identification and quantification by PMF

3.2.1. Factor profile interpretation

Three to twenty factors were initially tested, and the 14-factors solution was determined to be optimal as it provides the most reasonable factor profiles and high stability (Fig. S3 and Table S2). Detailed description and discussion are provided in Text S3. The PMF-resolved factor profiles are shown in Fig. 2. The diurnal variations in individual factor contributions are compared before and during the lockdown and shown in Fig. 3. The first factor is interpreted as fireworks (F1), as signaled by the high loadings of K, Cu, and Ba, which are indicative tracers of fireworks emissions. The contributions of F1 were minor before the Spring Festival but increased sharply on the Lunar New Year’s Eve (Jan 24, 2020) and during the Lantern Festival (Fig. S5). During the lockdown, its diurnal variation showed much higher contributions at nighttime than daytime (Fig. 3). The industrial emission (F2) is identified by trace elements of Fe, Cr, Mn, Zn, and Ca. The elemental Fe, Cr, Mn, and Zn have been widely used to track industrial emissions in previous studies (Li et al., 2020). The Fe/Ca and Mn/Ca mass ratios in this factor (10 and 1.2) were much higher than those derived from natural soil and paved road dust samples (0.8–2.6, 0.01–0.08) (Ho et al., 2003), suggesting the anthropogenic features of this factor. This factor shows higher concentrations during

Fig. 1. Temporal variations of (a) meteorological parameters (i.e., temperature, RH, and ventilation coefficient); (b) O3 and NOx; (c) major chemical species in PM2.5; (d) TAG-measured molecular markers during the entire campaign.

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5.1. Meteorological parameters, and the average concentrations of gaseous pollutant
and chemical components of PM$_{2.5}$ before and during the lockdown, respectively.

| Meteorological parameters | Before lockdown | During lockdown | Percentage change (%) | significance level |
|---------------------------|----------------|----------------|-----------------------|-------------------|
| Temperature (°C)          | 7.9 ± 3.9      | 6.0 ± 3.3      | -24                   | ***               |
| Relative humidity (%)     | 82 ± 14        | 78 ± 18        | -4.9                  | -                 |
| Ventilation coefficient (m$^2$ s$^{-1}$) | 840 ± 929 | 1117 ± 919     | +33                   | *                 |

| Gaseous pollutants (ppb)  |                   |                |                      |                   |
|---------------------------|------------------|----------------|----------------------|-------------------|
| O$_3$                     | 20 ± 13          | 42 ± 11        | +106                 | ***               |
| NO$_2$                    | 44 ± 35          | 18 ± 4.2       | -59                  | -                 |
| SO$_2$                    | 2.9 ± 31         | 2.6 ± 0.8      | -12                  | **                |
| CO                        | 823 ± 333        | 544 ± 243      | -34                  | ***               |

Table 1: Meteorological parameters, and the average concentrations of gaseous pollutant and chemical components of PM$_{2.5}$ before and during the lockdown, respectively.

The biomass burning emission factor (F7) is identified by levoglucosan and mannose. A low loading of K is observed in this factor, reflecting that during our campaign period the main source of K was unrelated to biomass burning. Instead, K was primarily from firework (F1) and coal combustion (F3). This also demonstrates that the advantage of using bio-marker (e.g., levoglucosan) to trace biomass burning emissions. In the absence of organic markers, the biomass burning source would be unlikely resolved as a separate source, which in turn contributes to inaccuracy of source contribution estimates for other related sources. The eight factor (F8) contains high loading of crustal elements of Ca. The elemental Ca is derived from suspended dust (Zhang et al., 2003; Cheng et al., 2005), confirming the identification of dust source. The identification of vehicle emission (F9) is based on the high loading of hopanes and steranes. Before the lockdown, the morning rush hour peaked around 8:00 a.m. in the diurnal variation, supporting the impact of vehicle emissions on this factor (Fig. 3). The sulfate-rich factor (F10) is characterized by high concentrations of sulfate and ammonium, with a molar ratio of [NH$_4$]/[SO$_4^{2-}$] at 2.2. The nitrate-rich factor (F11) is identified by high loadings of nitrate and ammonium. The molar ratio of ([NH$_4$]/[NO$_3$]/[SO$_4^{2-}$]) was higher than 2 (2.5), suggesting the predominant formation of (NH$_4$)$_2$SO$_4$. The good correlation of ([NH$_4$]/[NO$_3$] - 2 × [SO$_4^{2-}$]) with [Cl] (r = 0.76, slope = 0.53) suggests that the excessive NH$_4$ is in the form of NH$_4$Cl, which is also confirmed by the high loadings of Cl$^-$ in F11.

The SOA$_1$ factor (F12) contains high loading of naphthalene-derived SOA tracer (phthalic acid, 79%), which is associated with anthropogenic VOC precursor origin. The SOA$_2$ factor (F13) is characterized by high abundance of LMW DCAs and HDCAs, which are common later generation oxidation products of hydrocarbons (Chim et al., 2017; Ervens et al., 2004). Such a SOA source factor is also related in our source apportionment study in urban Shanghai (Zhu et al., 2020). Finally, the SOA$_3$ factor (F14) is distinguished by α-pinene derived SOA tracer (79%) and β-caryophyllene derived SOA tracers (24%). This factor represents SOA with biogenic VOC precursor origin. All the three SOA factors show noticeably higher daytime concentrations (Fig. 3), likely reflecting more intensive photo-oxidation during the daytime.

3.2.2. Source characteristics by air mass origins

Clustering analysis of the 48-h backward air mass trajectories were conducted to investigate the pollution origins and transport pathways. The air masses that moved to the sampling site can be grouped into four clusters (i.e., CL#1–4) according to the similarity in spatial distribution (Fig. 4). The PM and gaseous pollutants, as well as the meteorological parameters under different clusters were compared in Fig. S6. Briefly, (1) CL#1 represents the local air mass. The highest NO$_x$ concentrations and temperature along with the lowest wind speed were observed in this cluster. (2) CL#2 represents air masses that originated inland and were affected by urban pollution in northwestern China. The concentrations of PM$_{2.5}$, PM$_{10}$, SO$_2$, and CO were at the highest levels compared with other clusters. (3) CL#3 represents the cleaner air masses originated from Siberia and Mongolia and moving rapidly to the sampling site via long-range transport. Relatively high wind speed, as well as low concentrations of PM mass and NO$_x$ were found in CL#3. (4) CL#4

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The mass of “others” is the difference between PM$_{2.5}$ and the above-listed major species.

See Table S1 for a full list of TAG-measured individual organic compounds.

Significance level (p by Wilcoxon test: * * * p < 0.001, * * p < 0.01, * p < 0.05, ** p < 0.1, −0.1 < p < 1.0 (not significant).
represents the mixed type, by which air mass originated from the eastern China sea and moved through the urban Shanghai before arriving at the sampling site. CL#4 was characterized by the highest levels of RH and the lowest SO\(_2\) during the entire observation period, in accordance with its origin. Overall, CL#1, #2, and #3 reflect the air masses passed over continental areas through short-, medium-, and long-distance transport, respectively, and CL#4 reflects the mixing of oceanic and eastern continental influence.

Before the lockdown, the occurrence percentages of CL#1 and #2 were 24% and 31%, higher than the other two relatively clean clusters. However, during the lockdown, the percentage of CL#1 decreased significantly to 7.6%, and the percentage of CL#4 increased to 42%. The percentage of CL #3 changed little, with 18% and 17% before and during the lockdown, respectively. Compared with the uncontrolled period, the occurrence frequencies of pollution air mass decreased significantly in the lockdown period. These data indicate that the reduction of PM\(_{2.5}\) mass and SIA during the lockdown might not be merely a result of stringent lockdown measures, the effects of meteorological factors likely contributed, as well.

The mass contributions of individual factors corresponding to each trajectory cluster are shown in Fig. 4. PM\(_{2.5}\) pollution was on average at the lowest level in CL#3 (37 \(\mu g \ m^{-3}\)) and CL#4 (38 \(\mu g \ m^{-3}\)), which are associated with the air masses covering the relatively clean regions. Much higher PM\(_{2.5}\) was observed under the impacts of medium-range transitional air mass (CL#2, 64 \(\mu g \ m^{-3}\)) than the local air mass (CL#1, 48 \(\mu g \ m^{-3}\)). Sulfate-rich factor, nitrate-rich factor, and coal combustion were invariably the major PM pollution sources under all clusters. The sulfate-rich factor (F10) showed the highest contribution under CL#2 (8.2 \(\mu g \ m^{-3}\)), and the lowest in CL#1 (6.1 \(\mu g \ m^{-3}\)) and CL#3 (7.0 \(\mu g \ m^{-3}\)). This variation pattern was similar to that of SO\(_2\), suggesting the strong sulfate formation processes during the regional air mass movement. The nitrate-rich factor (F11) showed much higher contributions under CL#2 (29 \(\mu g \ m^{-3}\)) and CL#1 (22 \(\mu g \ m^{-3}\)) than CL#4 (20 \(\mu g \ m^{-3}\)) and CL#3 (15 \(\mu g \ m^{-3}\)), indicating the combined contributions of local nitrate formation and influence from the polluted northwestern areas. The lowest mass contributions of the three SOA factors were observed when air mass moved from North China via long-range transport (CL#3), likely due to the combined result of less abundant VOCs precursors and stronger dilution process. The anthropogenic SOA factor (F12) and SOA-II (F13) showed the highest mass concentrations under CL#2, whereas higher concentration was observed under CL#1 for biogenic SOA factor (F14). These results highlight the varying prominence of the three different SOA factors during regional and local transport.

Fireworks (F1) showed higher contributions in the transported type of CL#2 and #3 than CL#1 and CL#4, consistent with its geographical characteristic. Mass contributions of industrial emission (F2), cooking emission (F5), PAH-rich (F6), and vehicle emission (F9) were at the highest level under CL#1, in agreement with the influence of local anthropogenic emissions. Coal combustion (F3) showed much higher mass contribution under CL#2 (8.7 \(\mu g \ m^{-3}\)) than CL#3, CL#1, and CL#4 (4.2, 3.8, and 2.9 \(\mu g \ m^{-3}\), respectively), confirming the strong input of coal combustion from northwestern regions. Residual oil combustion (F4) showed higher contributions under CL#1 and #4. This effect of ship/vessel emissions is consistent with the observation that both clusters are associated with air masses passing over the marine regions. Mass contributions from biomass burning (F7) showed higher contributions under CL#1 and CL#2 (2.4 and 2.0 \(\mu g \ m^{-3}\), respectively), suggesting the input from both local emissions and regional transport. Dust (F8) showed comparable mass contributions among all the four clusters (1.5–2.1 \(\mu g \ m^{-3}\)).

3.2.3. PM\(_{2.5}\) contributions before and during the lockdown

Fig. 5 compares the concentrations of the 14 source factors and their percentage contributions to PM\(_{2.5}\) before and during the lockdown, together with the concentration change. The numerical values are summarized in Table 2. All sources, except for the fireworks emission, showed reduction in mass concentration contributions during the lockdown (Fig. 5c). More details in individual sources are described below.

Secondary sources are dominant contributors to PM\(_{2.5}\) at our study location. The combined secondary sources were 2–3 times higher than those of primary sources, accounting for 40 ± 29 \(\mu g \ m^{-3}\) (69%) and 22 ± 15 \(\mu g \ m^{-3}\) (63%) of ambient PM\(_{2.5}\) in the two sub-periods, respectively. Among the secondary sources, the nitrate-rich factor was the most predominant contributor, with average contributions of 29 ± 25 \(\mu g \ m^{-3}\) (50%) and 13 ± 10 \(\mu g \ m^{-3}\) (38%) before and during the lockdown, respectively. The reduction rate of nitrate-rich factor was 55% (−16 \(\mu g \ m^{-3}\), Fig. 5b), in accordance with the reduction in the
corresponding precursors of NOx (~59%). The sulfate-rich factor was the second predominant contributor to PM$_{2.5}$, showing comparable mass concentrations with 7.4 ± 4.6 μg m$^{-3}$ before the lockdown and 7.3 ± 5.2 μg m$^{-3}$ during the lockdown. The combined three SOA factors were minor contributors, accounting for 3.5 ± 3.4 μg m$^{-3}$ before the lockdown and the mass concentration was reduced by 42% to 2.0 ± 1.6 μg m$^{-3}$ during the lockdown. The average concentrations of SOA_I, SOA_II, and SOA_III factors decreased by 44% (~1.3 μg m$^{-3}$), 41% (~0.05 μg m$^{-3}$), and 30% (~0.17 μg m$^{-3}$), respectively. The comparative difference among the three SOA factors likely reflects varying responses of lockdown measures to the reduction in varied origin of VOC precursors. SOA_I and SOA_II were more closely tied with oxidation products from anthropogenic VOC precursors, which was largely reduced during the lockdown, whereas SOA_III is of biogenic origin and less influenced during the lockdown, thus with the lower percentage drop.

With regards of the nine primary sources, the combined contribution was 18 ± 13 μg m$^{-3}$ before the lockdown. They decreased by ~34% to 12 ± 8 μg m$^{-3}$ during the lockdown. Coal combustion was the
predominant primary source of PM$_{2.5}$. Its mass concentrations dropped from 6.1 ± 5.5 μg m$^{-3}$ before the lockdown to 4.2 ± 3.8 μg m$^{-3}$ during the lockdown. Lockdown measures had larger impacts on the sources of industrial emission, cooking emission, PAHs-rich, and vehicle emission, showing obvious reductions of 87% (−4.5 μg m$^{-3}$), 83% (−0.36 μg m$^{-3}$), 87% (−0.41 μg m$^{-3}$), and 87% (−0.49 μg m$^{-3}$), respectively. The results were expected and strongly agreed with the corresponding reduction in electricity consumption, residential emission, power plant combustion, and the volume of on-road traffic in Shanghai (Zhu et al., 2020). The contribution of the dust factor descended from 2.2 ± 1.7 to 1.6 ± 1.0 μg m$^{-3}$. The reduction rate of dust (33%) was in accordance with the recorded reduction in suspended dust in Shanghai (Zhu et al., 2020). The average concentrations of BB did not show significant decrease, with 1.7 ± 1.7 μg m$^{-3}$ before the lockdown and 1.3 ± 1.5 μg m$^{-3}$ during the lockdown. This was confirmed by the uncontrolled open fire burning across the country (Fig. S7). The average concentration of fireworks during the lockdown (3.3 ± 5.0 μg m$^{-3}$) was five times higher than that before the lockdown (0.62 ± 0.62 μg m$^{-3}$). Considering the ban on fireworks in municipal Shanghai, the increased contribution during the lockdown was most likely from the transported fireworks emission from the less urbanized peripheral areas in the north during the Spring Festival. The residual oil combustion was a minor contributor to PM$_{2.5}$, accounting for 2.5% (1.5 ± 0.9 μg m$^{-3}$) before the lockdown and 2.9% (1.0 ± 0.8 μg m$^{-3}$) during the lockdown.

When examining the contributions by air clusters, the highest PM$_{2.5}$ concentrations were observed under CL#2 (81 μg m$^{-3}$), followed by CL#4 (59 μg m$^{-3}$), CL#1 (52 μg m$^{-3}$), and CL#3 (40 μg m$^{-3}$) before the lockdown, and dropped significantly (exceeding −18%) in all air clusters during the lockdown. Fig. 6a compared the mass concentrations of individual factors to PM$_{2.5}$ across different air clusters in two sub-periods. Under the CL#1, the concentrations of nitrate-rich factor were comparable before and during (23 and 22 μg m$^{-3}$, respectively), while the sulfate-rich factor decreased by 60% from (7.2 μg m$^{-3}$) to (2.9 μg m$^{-3}$) during the lockdown. A sharp contrast was observed for the other three clusters, with reduced mass concentrations of nitrate-rich factor, and increased contributions of sulfate-rich factor. These results indicate the strong secondary sulfate formation processes during atmospheric transport. SOA_I and SOA_II factors decreased by 67% and 44% in CL#1 during the lockdown period, while SOA_III factors showed comparable concentration in the two sub-periods (1.0–0.98 μg m$^{-3}$, respectively).

As for the primary sources, the coal combustion in mass concentration contribution decreased during the lockdown in all air clusters, with the largest reduction rate in CL#1 (~65%) and CL#2 (~43%). The mass concentrations of industrial emission, cooking emission, PAHs-rich factor, and vehicle emission decreased by >73% in all air clusters, in agreement with the reductions in anthropogenic emissions across the country during the lockdown. Opposite trend was observed for the fireworks factor, with remarkable enhancement in mass concentrations in all air mass clusters. The increase was less significant in local CL#1, which was expected as the local fireworks have been banned during the Chinese New Year holiday in Shanghai. The BB factor showed varied changes under different air clusters, with significant reductions in CL#1 (~29%) and #4 (~21%), while comparable concentrations in CL#2 and CL#3.

3.2.4. OC contributions before and during the lockdown

The PMF analysis enables examination of the OC source contributions from the 14 identified factors. The amount of OC apportioned to each source factor in the PMF analysis was considered as the
contribution of that source to ambient OC. The measured OC is well predicted by PMF-modeled OC, with the two showing excellent agreement ($r = 0.96$, Fig. S8). Fig. 7 compares the individual contributions of the 14 sources in both mass concentrations and percentage contributions between the two sub-periods. The OC was grouped into two categories for further analysis, i.e., primary OC (POC) and secondary OC (SOC). POC is taken to be the combined OC from 9 primary sources, and SOC is the sum of OC from the 5 secondary sources.

During the study period, POC was notably higher than SOC. The combined POC concentration was reduced slightly from $3.9 \pm 2.3 \mu g C m^{-3}$ before the lockdown to $3.1 \pm 1.6 \mu g C m^{-3}$ during the lockdown (Table S4). Coal combustion was the most predominant POC contributor, showing reduction in both absolute mass concentration (from $1.7 \pm 1.6$ to $1.0 \pm 1.0 \mu g C m^{-3}$) and in percentage contribution (from 32% to 30%) from before to during the lockdown. Dust and BB factors were the next significant POC contributors, each accounting for ~14% of OC. The contribution of dust was $0.80 \pm 0.69 \mu g C m^{-3}$ before the lockdown and decreased to $0.56 \pm 0.35 \mu g C m^{-3}$ during the lockdown. The BB contribution dropped by 21% from $0.72 \pm 0.69 \mu g C m^{-3}$ before the lockdown to $0.57 \pm 0.53 \mu g C m^{-3}$ during the lockdown. Reductions were also observed for the residual oil combustion factors, dropping from $0.60 \pm 0.42 \mu g C m^{-3}$ to $0.41 \pm 0.35 \mu g C m^{-3}$. Fireworks was a negligible contributor (1.4%) before the lockdown, while its contribution increased to a significant 10% during the lockdown (10%). Other sources including cooking, PAHs, and vehicle emission were very minor contributors, together contributing to 1.6% before the lockdown. Their concentrations decreased by >84% with 0.3% percentage contributions during the lockdown. The industrial emission had negligible contribution to OC.

The concentrations of SOC accounted for 25% of total OC with $1.3 \pm 1.0 \mu g C m^{-3}$ before the lockdown and dropped to $0.80 \pm 0.53 \mu g C m^{-3}$ and 20% during the lockdown. The factors of SOA_I, nitrate-rich, and SOA_III were the top three secondary contributors to OC. The contribution of SOA_I was $0.73 \pm 0.78 \mu g C m^{-3}$ (14%) before the lockdown and dropped to $0.40 \pm 0.38 \mu g C m^{-3}$ (10%) during the lockdown. The nitrate-rich factor had its contribution decreased by 56% from $0.23 \pm 0.19$ to $0.10 \pm 0.08 \mu g C m^{-3}$. SOA_III factor showed a relatively smaller decrease with percentage changes of 28% from $0.21 \pm 0.24$ to $0.15 \pm 0.11 \mu g C m^{-3}$. Sulfate-rich factor showed comparable concentrations with $0.14 \pm 0.09 \mu g C m^{-3}$ (2.6%) before the lockdown and $0.14 \pm 0.10 \mu g C m^{-3}$ (3.5%) during the lockdown. SOA_II factor was a minor OC contributor, contributing to 0.4% and 0.3%, respectively.
Table 2

| Source factors | Before lockdown | During lockdown | Percentage change (%) | Wilcoxon test significance level |
|----------------|----------------|----------------|-----------------------|-----------------------------------|
| Measured PM$_{2.5}$ | 64 ± 43 | 49 ± 31 | −24 | *** |
| PMF-modeled PM$_{2.5}$ | 59 ± 37 | 35 ± 21 | −41 | *** |
| F1_Fireworks | 0.62 ± 0.62 | 3.3 ± 5.0 | +438 | *** |
| F2_Industrial emission | 5.2 ± 5.3 | 0.67 ± 0.61 | −87 | *** |
| F3_Coal combustion | 6.1 ± 5.5 | 4.2 ± 3.8 | −31 | ** |
| F4_Residual oil combustion | 1.5 ± 0.9 | 1.0 ± 0.78 | −32 | *** |
| F5_Cooking emission | 0.44 ± 0.50 | 0.07 ± 0.09 | −83 | *** |
| F6_PAHs | 0.47 ± 0.44 | 0.06 ± 0.04 | −87 | *** |
| F7_Biomass burning | 1.7 ± 1.5 | 1.3 ± 1.1 | −22 | * |
| F8_Dust | 2.3 ± 1.7 | 1.5 ± 0.81 | −33 | *** |
| F9_Vehicle emission | 0.56 ± 0.64 | 0.07 ± 0.07 | −87 | *** |
| F10_Secondary sulfate | 7.4 ± 4.6 | 7.3 ± 5.2 | −1 | − |
| F12_SOAI (anthropogenic SOA) | 29 ± 25 | 13 ± 10 | −55 | *** |
| F13_SOAI II (late-generation SOA) | 3.0 ± 3.1 | 1.7 ± 1.5 | −44 | ** |
| F14_SOAI III (biogenic SOA) | 0.11 ± 0.13 | 0.07 ± 0.06 | −41 | *** |

**p < 0.001, *0.001 < p < 0.01, *0.01 < p < 0.05, *0.05 < p < 0.1, #: 0.1 < p < 1.0 (not significant).

Significance level (p) by Wilcoxon test.

The individual source contributions to OC were compared under different air clusters before and during the lockdown to investigate the influence of air masses of different origins (Fig. 6b). The total POC had higher levels when the air mass of CL#2 (5.7 μgC m$^{-3}$) and CL#1 prevailed (4.4 μgC m$^{-3}$) before the lockdown, and decreased by 26% and 36% during the lockdown, respectively. The POC concentrations were comparable in CL#3 (3.0 and 3.2 μgC m$^{-3}$) and CL#4 (2.6 and 2.4 μgC m$^{-3}$) in the two sub-periods. Coal combustion factor showed significant reductions in CL#1 (−64%), CL#2 (−42%), and CL#4 (−15%), while the concentration in CL#3 slightly increased from 1.2 μgC m$^{-3}$ to 1.3 μgC m$^{-3}$. Residual oil combustion factor increased by 51% in CL#1 but decreased by >30% in the other three clusters. The contributions of the BB factor in CL#1 and CL#2 were 2–3 times higher than CL#3 and CL#4 before the lockdown, and the levels were comparable in all air clusters during the lockdown. The contributions of cooking emission, PAHs, and vehicle emission decreased largely in all air clusters, with percentage reductions in the range of 77–93%.

Different from POC, the total SOC loading was the highest in CL#2 and the lowest in CL#3 in the two sub-periods. The SOC concentration was reduced in all air clusters during the lockdown. SOA_I factor showed higher concentration in CL#2 and CL#1 before the lockdown (1.6 and 0.51 μgC m$^{-3}$) and dropped by 74% and 71% with 0.41 and 0.15 μgC m$^{-3}$ during the lockdown, respectively. Under CL#1, the contributions of nitrate-rich factor showed small changes during the lockdown (~0.17 μgC m$^{-3}$), while 40–80% reductions were observed in other three clusters. In contrast, sulfate-rich factor dropped by 60% in CL#1 (0.13–0.06 μgC m$^{-3}$), whereas its concentrations remained unchanged in the other three clusters (0.13–0.17 μgC m$^{-3}$). The levels of SOA_III factor were higher in CL#1 and showed comparable contributions in the two sub-periods. As for the SOA_II factor, its OC contributions in all air clusters were relatively smaller and comparable (0.01–0.03 μgC m$^{-3}$).

A few recent studies in northern China (e.g., Beijing-Tianjin-Hebei regions) reported that the concentrations of PM$_{2.5}$ were relatively unchanged or even slightly increased from before to during the lockdown period despite the large reductions in primary anthropogenic emissions, and attributed this to the offsetting effect caused by the enhancement of secondary formation processes (Le et al., 2020; Wang et al., 2020b; Cui et al., 2020). Specifically, Le et al. (2020) found that the ground-based PM$_{2.5}$ concentrations in central Beijing increased by 55.1% (+30.6 μgC m$^{-3}$) during the lockdown, which was due to increase in sulfate (+26.3%) and SOA (+15.1%). Further model simulation results suggested that the unexpected PM$_{2.5}$ pollution increase was contributed from enhanced aerosol heterogeneous chemistry along with the stagnant atmospheric conditions and uninterrupted power plant emissions. Wang et al. (2020a) applied the CMAQ model to study the responses of PM$_{2.5}$ to the anthropogenic emission reductions and meteorological conditions in nine urban sites. Their results revealed that the effects of adverse meteorological conditions did not offset the reductions in anthropogenic emission. Comparing the observational PM$_{2.5}$ concentrations in “2 + 26 cities” in the Beijing-Tianjin-Hebei region (i.e., two megacities of Beijing and Tianjin plus 24 neighboring cities), Cui et al. (2020) found that the PM$_{2.5}$ levels were higher during the lockdown in rural Xianghe (+15 μg m$^{-3}$, 15%), urban Beijing (+11 μg m$^{-3}$, 19%), and urban Langfang (+10 μg m$^{-3}$, 13%). They attributed the increased PM$_{2.5}$ as a combined
result of the intensive secondary formation and unfavorable meteorological conditions. Our recent publication in urban Shanghai also observed enhancement of secondary sulfate and organic contributions to both PM$_{2.5}$ and OC during the lockdown (Zhu et al., 2020). In comparison, our current observation in suburban Shanghai found that the contributions of SOA sources to PM$_{2.5}$ and OC did not increase with the increasing oxidation capacity during the lockdown.

To understand such an urban-suburban difference, we first compared the contributions of individual SOC factors between the two sites. As shown in Fig. 8a, the total SOC in urban SAES during the lockdown increased by 23%, mainly due to the increase from sulfate-rich factor (from 1.2 to 2.5 μgC m$^{-3}$) and SOA II (from 0.40 to 0.65 μgC m$^{-3}$). In comparison, the total SOC in suburban DSL showed a significant decrease, with SOA I leading (from 0.73 to 0.40 μgC m$^{-3}$, -45%), followed by the nitrate-rich factor (from 0.23 to 0.10 μgC m$^{-3}$, -56%). The sulfate-rich factor exhibited less concentration difference (~0.14 μgC m$^{-3}$). The above distinct SOC variations imply different driving factors underlying secondary formation in the urban and suburban atmospheres.

It should be noted that the SOA formation in ambient air is complex and vary in different environments because formation pathways could involve multiple oxidants in addition to the numerous VOC precursors. Furthermore, meteorological conditions (e.g., temperature, RH, radiation, and pressure) also play important roles in affecting the reaction kinetics and medium (e.g., gas phase vs. aerosol liquid phase) as well as gas-particle partition of the secondary products (Boyd et al., 2017; Takekawa et al., 2003). During the lockdown, the levels of VOC precursors decreased significantly, by ~40% in urban Shanghai and ~50% in the suburban DSL site (Jia et al., 2020). It is possible that the formation of SOA in DSL site would be more limited by the less abundant precursors levels.

The site-to-site differences in meteorological conditions were small during the lockdown, with comparable levels in temperature (6.9 vs. 6.0 °C) and RH (72 vs. 78%) between the urban and the suburban sites (Fig. S9a). We thus examined other factors (e.g., air mass transport). During the wintertime, Shanghai was a receptor of the highly polluted air massed from northern China. The urban site is in downtown Shanghai, whereas the suburban site is situated 50 km west of the urban site and adjacent to Jiangsu and Zhejiang Provinces. During the lockdown, the prevailing wind from northwest direction brought in air masses that had passed through the regions with intensive coal burning (e.g., Tianjin, Anhui, Jiangsu province) for winter residential heating (Dai et al., 2020). The air masses reached the suburban DSL site before being further transported to the urban SAES site. This was supported by...
the slightly higher levels of SO2 in DSL site than SAES site (2.6 vs. 2.2 ppb). Moreover, better correlations were found between OC and As and Pb in DSL site ($r = 0.76$–$0.79$) than in SAES site ($r = 0.60$–$0.66$) during the lockdown, suggesting the stronger association of OC with coal burning in DSL site. In contrast, OC showed a stronger correlation with sulfate in SAES site ($r = 0.85$) than in DSL site ($r = 0.72$), reflecting the stronger contributions of sulfate formation process to the overall secondary formation in SAES site. Recent studies highlight the potentially important role of heterogeneous oxidation of SO2 during long-range transport leading to the sulfate accumulation (Liu et al., 2021; Willis et al., 2018). We further investigated the relationships between the SOC factors and ALWC. As shown in Fig. 8b and 8c, SOC in the sulfate-rich and SOA II factors showed positive correlations with ALWC at the SAES site, while no discernible correlations were observed at the DSL site. Previous field observations also revealed the important role of aqueous phase reactions in accelerating the sulfate formation in urban cities (Wang et al., 2021; Huang et al., 2019). These results implicated the potentially enhanced formation of the secondary sulfate and SOA through the aerosol heterogeneous reactions in urban SAES site.

In summary, our findings in suburban Shanghai are distinct from that reported in urban Shanghai and those in megacities in Eastern China, where both stronger oxidation capacity and higher secondary inorganic and organic aerosol formations were observed during the lockdown. The site-to-site differences likely reflect the location-specific complexity of interplaying of precursors, oxidants, and meteorological factors in the formation of SOA.

4. Conclusions

Hourly organic molecular markers in PM$_{2.5}$ were monitored by the TAG system before and during the Covid-19 lockdown in suburban Shanghai from Dec. 28, 2019 to Feb. 9, 2020, along with other major chemical components. This set of measurement data provides us an opportunity to study the response of aerosol composition and source contributions to lockdown emission controls. The mass concentrations of PM$_{2.5}$ dropped by 24% ($-15$ μg m$^{-3}$) from before to during control periods, with nitrate making the leading contribution ($-14$ μg m$^{-3}$). Other components also dropped except for the fireworks-related elements (e.g., $K$, $Ba$) as the lockdown coincided with the Chinese New Year holidays. With the incorporation of the TAG-measured organic source tracers, the PMF receptor modeling analysis identified 9 primary and 5 secondary source factors, among which nitrate-rich factor, sulfate-rich factor, and coal combustion were the major PM$_{2.5}$ contributors. The five secondary sources all decreased during the lockdown with the largest reductions occurring with the secondary nitrate-rich factor ($-16$ μg m$^{-3}$) and the anthropogenic SOA factor (SOA II) ($-13.3$ μg m$^{-3}$). The source analysis shows varying reductions from primary sources—industrial emission ($-4.5$ μg m$^{-3}$), coal combustion ($-1.9$ μg m$^{-3}$), dust ($-0.8$ μg m$^{-3}$), vehicular emissions ($-0.5$ μg m$^{-3}$), residual oil combustion ($-0.5$ μg m$^{-3}$), and cooking emission ($-0.4$ μg m$^{-3}$) consistent with the reduced human activities during the lockdown. The percentage dropping was most notable for industrial (–87%) and vehicular emissions (–87%), reflecting the steep impact of the COVID lockdown on these two sectors. One primary source—fireworks, however, had a significant increase (+2.7 μg m$^{-3}$) during the lockdown, which was an expected result of Chinese New Year-related celebration in the surrounding less urbanized areas.

Backward trajectories analysis suggested that the regional input from the northwestern regions was highly responsible to the PM$_{2.5}$ accumulation during the whole campaign. The strict controls across the country during the lockdown have significant effects on reducing both the regional air pollutants and local emissions. While previous studies that were conducted in megacities demonstrated the offset effects of the primary emissions by the enhanced secondary pollution and oxidation.
capacity during the Covid-19 lockdown, we observed reduced secondary inorganic aerosols as well as SOA. Cross-comparing in OC source apportionment between urban Shanghai and this study location in the suburban suggested that during the lockdown, the reductions in secondary formation in suburban DSL could be regulated by the reduced VOCs precursor and the meteorological parameters (e.g., ALWC and air mass transport). This case study provides direct evidence that the responses to substantial reductions in urban primary precursors are likely different in urban-influenced suburban locations from that in urban centers. Currently, our results indicate that controlling the primary emissions in the urban areas reduces both primary and secondary PM$_{2.5}$ components in suburban Shanghai. Further studies are essential to enhance our understanding of the complicated mechanisms driving the responses of primary and secondary aerosols to control measures in different environments.

CRediT authorship contribution statement

Shan Wang: TAG data collection, Data analysis, Investigation, Writing – original draft. Qiongqiong Wang: TAG data collection, Investigation. Shihui Zhu: TAG data collection. Min Zhou, Liping Qiao, Dandan Huang, Yingge Ma, and Yiquan Lu: Collection and processing of the PM$_{2.5}$ and its major chemical species data. Cheng Huang: Project and resources support. Qiongyan Fu and Yusen Duan: Collection and processing of the meteorological parameters and gaseous pollutants. Jian Zhen Yu: Conceptualization, Methodology, Supervision, Writing – review & editing the manuscript.

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.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

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