Implications of the Improvement in Atmospheric Fine Particles: A Case Study of COVID-19 Pandemic in Northern Taiwan

Chuan-Hsiu Huang1, Yi-Ru Ko1, Tzu-Chi Lin1, Yu-Hsiang Cheng2,3, Yu-Cheng Chen4,5, Yu-Chieh Ting1*

1 Graduate Institute of Environmental Engineering, National Taiwan University, Taipei, Taiwan
2 Department of Safety, Health and Environmental Engineering, Ming Chi University of Technology, New Taipei, Taiwan
3 Center for Environmental Sustainability and Human Health, Ming Chi University of Technology, New Taipei, Taiwan
4 National Institute of Environmental Health Sciences, National Health Research Institutes, Miaoli, Taiwan
5 Department of Occupational Safety and Health, China Medical University, Taichung, Taiwan

ABSTRACT

The outbreak of COVID-19 pandemic in northern Taiwan led to the implementation of Level 3 alert measures during 2021 and thereby impacted the air quality significantly, which provided an unprecedented opportunity to better understand the control strategies on air pollutants in the future. This study investigated the variations in sources, chemical characteristics and human health risks of PM2.5 comprehensively. The PM2.5 mass concentrations decreased from pre-alert to Level 3 alert by 49.4%, and the inorganic ions, i.e., NH4+, NO3– and SO42–, dropped even more by 71%, 90% and 52%, respectively. Nonetheless, organic matter (OM) and elemental carbon (EC) simply decreased by 36% and 13%, which caused the chemical composition of PM2.5 to change so that the carbonaceous matter in PM2.5 dominated instead of the inorganic ions. Correlation-based hierarchical clustering analysis further showed that PM2.5 was clustered with carbonaceous matter during the Level 3 alert, while that clustered with inorganic ions during both pre-alert and post-alert periods. Moreover, 6 sources of PM2.5 were identified by positive matrix factorization (PMF), in which secondary nitrate (i.e., aging traffic aerosols) exhibited the most significant decrease and yet primary traffic-related emissions, dominated by carbonaceous matter, changed insignificantly. This implied that secondary traffic-related aerosols could be easily controlled when traffic volume declined, while primary traffic source needs more efforts in the future, especially for the reduction of carbonaceous matter. Therefore, cleaner energy for vehicles is still needed. Assessments of both carcinogenic risk and non-carcinogenic risk induced by the trace elements in PM2.5 showed insignificant decrease, which can be attributed to the factories that did not shut down during Level 3 alert. This study serves as a metric to underpin the mitigation strategies of air pollution in the future and highlights the importance of carbonaceous matter for the reduction in PM2.5.

Keywords: COVID-19 Level 3 alert, PM2.5, Chemical composition, Source apportionment, Mitigation strategies

1 INTRODUCTION

Air pollution is a high-profile problem that has been widely studied in the past few decades. Air pollutants can be classified as particulate matter (PM) and gaseous pollutants [e.g., nitrogen oxides (NOx), carbon monoxide (CO), sulfur dioxide (SO2), ozone (O3)], of which both have been...
reported to pose great impact to human health, air quality and climate (Feng et al., 2016; Huang et al., 2018; Lefohn et al., 2017; Tseng et al., 2019). In particular, PM$_{2.5}$ has become a central issue in many countries because of its complex chemical composition and significant impacts (ChooChuay et al., 2020; Hsu et al., 2017; Lee et al., 2019; Xiang et al., 2020). One of the major components of PM$_{2.5}$ is water-soluble inorganic ions (WSIs), which usually exhibit higher mass concentrations on hazy days (Cheng et al., 2021; Xu et al., 2019). Ammonium nitrate (NH$_4$NO$_3$) and ammonium sulfate ((NH$_4$)$_2$SO$_4$) play important roles in the quality of atmospheric visibility (Cheng and Tsai, 2000; Khanna et al., 2018). Moreover, NH$_4$NO$_3$ has been reported to be more sensitive to visibility degradation, requiring more effective control strategies (Ting et al., 2022). Another important composition of PM$_{2.5}$ is carbonaceous aerosols, which can be generally classified as organic carbon (OC) and elemental carbon (EC). In recent years, there has been a growing body of literature that stated the importance of carbonaceous aerosols because of their adverse effect on human health and the light absorbability, affecting the radiative budget in the atmosphere (Huang et al., 2021; Jo et al., 2016; Yan et al., 2021). In terms of the PM$_{2.5}$-bound trace elements, although they usually account for less than 10% of the total PM$_{2.5}$ mass concentration, most of these elements are toxic and associated with several health issues (Liu et al., 2018). Additionally, the trace elements can induce the formation of strong oxidants such as reactive oxygen species, adversely affecting human health to a certain extent (Cigánková et al., 2021; Liu et al., 2019; Xie et al., 2020).

Because of the impact of complicated chemical components in PM$_{2.5}$ on human health, air quality and climate, it is urgent to execute effective control measures of air pollution, especially in Taiwan. Since the incidence of adenocarcinoma lung cancer in Taiwan is growing and more than half of the lung cancer patients never smoked that could be associated with the exposure to PM$_{2.5}$ (Tseng et al., 2019). Although previous studies investigated the chemical compositions, sources and formation mechanisms of PM$_{2.5}$ using various approaches, there have been still some biases in the control strategies owing to seasonal variation and different sampling approaches (Liao et al., 2021; Liu et al., 2018). Moreover, it is difficult to well understand how the air quality can be achieved after implementing various emission control measures.

The emergence of COVID-19 (SARS-CoV-2–coronavirus disease 2019) caused severe social and economic shocks due to the lockdown measures in many countries worldwide. However, the unprecedented pandemic could provide a rare opportunity to study the impact of human activities to air pollution. Recently, there has been a proliferation of studies that investigated the variations of air pollution due to the pandemic (Bera et al., 2022; Cui et al., 2020; Grange et al., 2021; Liu et al., 2021; Sharma et al., 2020). Most of the studies indicated that most of the criteria air pollutants dropped significantly during the lockdown periods, with the enhanced level of atmospheric oxidation capacity (AOC) and ozone (O$_3$) (Hong et al., 2021; Liu et al., 2021; Zhu et al., 2021a). However, the situation in Taiwan might be different with those lockdown countries since the regulation policies were not as strict as lockdown. Taiwan National Health Command Center announced the Level 3 alert policy in Taipei from May 15th to July 26th, 2021, to avoid the spread of COVID-19, which was the first and only Level 3 alert period in Taipei. The chemical compositions, sources and health risk of PM$_{2.5}$ were investigated, seizing this rare opportunity to understand how air quality will change...
while human activities significantly drop. This study will provide an insight into the implementation of more effective mitigation strategies of air pollution in the future.

2 METHODS

2.1 Sampling Site and Period

This study was conducted in the Taipei urban area, the epicenter of the COVID-19 outbreak in Taiwan. The sampling site was located on the rooftop of a building (~15 m high above surface) in National Taiwan University (25°1’2”N, 121°32’38”E) and the surrounding environment is shown in Fig. 1. In order to make intercomparison of atmospheric pollutants, the sampling time was divided into three periods, which were pre-alert (March 4th to May 12th, 2021), Level 3 alert (May 15th to July 26th, 2021) and post-alert (July 29th to October 5th, 2021), in which there were 21 filter samples for each period. A high-volume sampler (Tisch Environmental, Inc.) was used to collect 24 h PM$_{2.5}$ samples on 8 × 10 inches quartz fiber filters, with a flow rate of 1.13 m$^3$ min$^{-1}$. The filters were prebaked at 900°C for 3 hours to eliminate any impurity. The total PM$_{2.5}$ samples were 63 and were wrapped in aluminum foil bags at −20°C after sampling. Meteorological conditions and gaseous pollutants were obtained from the adjacent monitoring station (Guting station, about 1.5 km from the sampling site) of Taiwan Environmental Protection Administration (TEPA). Wind rose plots for pre-alert, Level 3 alert and post-alert are shown in Fig. S2 and illustrate that the wind directions most frequently come from the east side, especially from the northeast.

2.2 Chemical Analysis

A punch of quartz filters (0.5 cm$^2$) was used to analyze the mass concentrations of OC and EC by thermal/optical carbon analyzer (DRI 2015, Magee Scientific) using the IMPROVE_A reflectance protocol (Chow et al., 2007). OM was estimated as 1.8 × OC in this study (Malm et al., 2017).

Fig. 1. Map of the sampling site and the surrounding environment. The blue line on the right marked the border of Taipei city.
A portion of filter (1/15) was cut and extracted with 10 mL ultrapure water (Milli-Q, 18.2 MΩ) in sonic bath for 45 mins. The extract was then filtered with a 0.22 µm pore syringe filter for the analysis of WSIIs using ion chromatography (Dionex ICS-1100, Thermo Scientific), including NH₄⁺, NO₃⁻, SO₄²⁻, Na⁺, Cl⁻, K⁺, Mg²⁺, and Ca²⁺.

Another portion of filter (1/30) was extracted with microwave digestion system (CEM Mars, CEM corporation) for the trace element analysis. Digestion procedure started from adding 10 mL HNO₃ and 0.3 mL HF for the first 60 minutes at 200°C, and then extra 2.8 mL of 5% H₃BO₃ was added for another 50 minutes at 140°C. After digestion, the extracts were filtered with 0.45 µm PTFE filters and then diluted to 25 mL with ultrapure water. The total 18 trace elements, including Al, K, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Sr, Cd, Sb, Ba, and Pb were analyzed by inductively coupled plasma-mass spectrometry (ICP-MS, Agilent Technologies 7700x). Details of the experimental information and QA/QC can be found in a previous study (Hsu et al., 2021). Blanks were conducted for every analysis in this study.

The nitrogen oxidation ratio (NOR) and sulfur oxidation ratio (SOR) have been widely used to evaluate the formation rates of nitrate and sulfate. The following formulae Eq. (1) and Eq. (2) were used to calculate NOR and SOR (Ohta and Okita, 1990). The concentrations of each following species were converted as molar concentrations.

\[
\text{NOR} = \frac{[\text{NO}_3^-]}{[\text{NO}_2^-]+[\text{NO}_3^-]} \quad (1)
\]

\[
\text{SOR} = \frac{[\text{SO}_4^{2-}]}{[\text{SO}_2^-]+[\text{SO}_4^{2-}]} \quad (2)
\]

The neutralization of aerosol acidity was evaluated by the neutralization ratio (NR), which are expressed as Eq. (3) and Eq. (4). More details of this method can be found in the previous study (Cheng and Tsai, 2000).

\[
\text{nss-SO}_4^{2-} = \text{SO}_4^{2-} - 0.231 \times \text{Na}^+ \quad \text{(the units are µg m}^{-3}\text{)} \quad (3)
\]

\[
\text{NR} = \frac{\text{NH}_3}{\text{nss-SO}_4^{2-} + \text{NO}_3^-} \quad \text{(the units are neq m}^{-3}\text{)} \quad (4)
\]

The EC-tracer method has been widely used to estimate primary OC (POC) and secondary OC (SOC) by Eq. (5) and Eq. (6) (Zhang et al., 2019):

\[
\text{POC} = \text{EC} \times (\text{OC/EC})_{\text{min}} \quad (5)
\]

\[
\text{SOC} = \text{OC}_{\text{tot}} - \text{POC} \quad (6)
\]

where OC_{tot} represents the total mass concentration of OC, and (OC/EC)_{min} is the lowest value of OC/EC in the sampling period.

The atmospheric oxidation capacity (AOC) is an important factor to understand the oxidation level of the atmosphere and can further serve as an indicator for the formation of secondary pollutants (Qin et al., 2022; Wang et al., 2021). In this study, AOC was evaluated by Eq. (7):

\[
\text{AOC} = \frac{\text{O}_3 + \text{NO}_2}{\text{CO}} \quad (7)
\]

where O₃ = O₃ + NO₂. O₃ was normalized by CO (carbon monoxide) since it remains stable in the atmosphere, and the normalization of CO can eliminate the boundary layer effect (Feng et al., 2022; Yao et al., 2020).
2.3 Correlation-based Hierarchical Clustering

Cluster analysis is a useful method when it comes to air pollution data analysis, such as k-means clustering and hierarchical clustering (Carslaw and Beevers, 2013; Qiao et al., 2018). In this study, hierarchical clustering was performed to distinguish the differences and characteristics of air pollutants between pre-alert, Level 3 alert and post-alert periods. The distance of the species categorized was provided by Spearman’s correlation coefficient and with the “complete” approach for hierarchical clustering. Details of the different kinds of approaches for clustering the air pollution data can be found elsewhere (Govender and Sivakumar, 2020).

In order to understand the composition and formation of PM$_{2.5}$ during different periods, PM$_{2.5}$ and its major components (NH$_4^+$, Na$^+$, K$^+$, Mg$^{2+}$, Ca$^{2+}$, Cl$^-$, NO$_3^-$, SO$_4^{2-}$, POC, SOC, EC) and the gaseous pollutants were used to carry out a correlation matrix. Spearman's correlation coefficients were presented as the matrix's scale due to the non-normal distribution of most species (with the Shapiro–Wilk normality test). It is a non-parametric measurement that shows the relationship and similarity of each species, with the value ranging from –1 to +1. The correlation-based hierarchical clustering method can serve as a graphical insight into the variations of PM$_{2.5}$ and its highly correlated composition.

2.4 Source Apportionment and Data Analysis

In this study, the positive matrix factorization (PMF 5.0) model developed by USEPA was used to identify the sources of PM$_{2.5}$ during the sampling period. PMF is based on the least squares algorithm to provide robust results of factor analysis (Paatero, 1997; Paatero and Tapper, 1994). 24 dominant components (NH$_4^+$, Na$^+$, Cl$^-$, NO$_3^-$, SO$_4^{2-}$, OC, EC, Al, K, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Cd, Sb, Ba, and Pb) in PM$_{2.5}$ were set as input files for PMF modelling. In this study, the PMF model was run with three to eight factors to get reasonable and statistically robust sources in the Taipei urban area. The optimal number of factors is six factors with the Q/Q$_{ref}$ closest to 1.

Bivariate polar plot (BPP) is a graphical method to understand the characteristics of different type of sources (Carslaw and Beevers, 2013). BPP has been widely used in many previous studies for the purpose of source identification (Chang et al., 2018; Leoni et al., 2018). BPP treats the data with wind speed and wind direction to find out the potential sources of pollution in a polar coordinate. This study uses BPP as an auxiliary way to interpret the factors of PMF and therefore the time series results from PMF were input as the pollution data. We distributed the daily PMF data to 24 hours in order to combine with the hourly wind speed and direction.

The statistical analysis of this study was performed by R language (R Core Team, version 4.0.5), including Spearman’s correlation coefficient, Mann-Whitney U test and Shapiro–Wilk normality test. Several R packages were used for the data visualization such as “openair” (Carslaw and Ropkins, 2012), “corrplot”, “ggplot2”, “ggpubr”, “magrittr” and “ggstatsplot” (Patil, 2021).

2.5 Health Risk Assessment of PM$_{2.5}$-bound Elements

In this study, health risks caused by PM$_{2.5}$-bound elements were characterized as carcinogenic risks (lung cancer) and non-carcinogenic risks (such as some chronic effects). Carcinogenic risks through inhalation were evaluated by excess cancer risk (ECR), as shown in Eq. (8) (Duan et al., 2021). The carcinogenic elements include Co, As, Pb, Ni, Cr (VI) and Cd according to the classification of IARC (IARC, 2022). The concentration of Cr (VI) was calculated as 1/7 of the concentration of Cr detected in the atmosphere (Park et al., 2008). Non-carcinogenic risks through inhalation were evaluated as hazard quotient (HQ), as expressed in Eq. (9) (Duan et al., 2021).

$$ECR = EC \times IUR$$  \hspace{1cm} (8)

$$HQ = \frac{EC}{RfC \times 1000}$$  \hspace{1cm} (9)

The corresponding inhalation unit risk (IUR) and reference concentration (RfC) values of each element were obtained from U.S. EPA and Xie et al. (2020). EC can be calculated through Eq. (10).
where $C$ is the concentration of elements ($\mu g m^{-3}$), $ET$ is the exposure time (8 hour per day), $EF$ is the exposure frequency, $ED$ is the exposure duration (24 years) and $AT_n$ is the average lifetime (70 years for ECR and 24 years for HQ).

3 RESULTS AND DISCUSSION

3.1 Overview of Chemical Composition in PM$_{2.5}$

Table S1 presents all species that were analyzed in three sampling periods, most of the species decreased apparently from pre-alert to Level 3 alert and increased from Level 3 alert to post-alert. Fig. S3 shows the time series of the air pollutants for the whole sampling period, and the variations of PM$_{2.5}$, NO$_x$ and CO were very similar in that all showed lower concentrations during the Level 3 alert period. This implied that the lower level of traffic congestion could have an impact on both particulate matter and gaseous pollutants. The concentrations of OC and EC were also lower during Level 3 alert but did not change as much as PM$_{2.5}$, which will be further discussed in the following paragraph.

Fig. 2 illustrates variations in PM$_{2.5}$ and its major compositions among three sampling periods. Nonparametric Mann-Whitney U test was used to verify the significance of the changes. As can be seen in the figure, the concentrations of PM$_{2.5}$, NO$_x$, CO, and EC were significantly lower during Level 3 alert compared to pre-alert and post-alert periods. OC and SO$_2$ concentrations also showed a similar trend, although not as pronounced as PM$_{2.5}$. The decrease in these pollutants during Level 3 alert can be attributed to the reduction in traffic congestion, which in turn reduced the emissions of these pollutants. The concentrations of NH$_4^+$ and SO$_4^2-$ showed a different pattern, with a slight increase during Level 3 alert, which may be due to increased emissions from other sources or changes in meteorological conditions.

![Fig. 2. Variations of average mass concentrations in PM$_{2.5}$ and chemical compositions during Pre-alert, Level 3 alert and Post-alert periods. Error bars represent the standard deviation. The numbers above the brackets show the $p$-value between Pre-alert and Level 3 alert; Level 3 alert and Post-alert.](image-url)
be seen, PM$_{2.5}$, NH$_4^+$, NO$_3^-$ and SO$_4^{2-}$ exhibited significant decreases from pre-alert to Level 3 alert period, which were reductions of 49.4%, 70.9%, 90.3% and 51.7% ($p < 0.01$), respectively. The concentration of NO$_3^-$ showed the greatest reduction, which could be attributed to the significant drop of NOx emissions and this was similar to the urban areas of Spain (Clemente et al., 2022) and China (Tian et al., 2021). NO$_3^-$ has commonly been shown to have lower concentrations in summer, while the variations between spring and summer in previous studies were –38% (Hsu et al., 2017) and –42% (Chang et al., 2010) in Taiwan, which were much smaller than this study (~90%). While seasonal variation and meteorological conditions may affect the concentrations of air pollutants, the impact of COVID-19 Level 3 alert was still the main contributor. In addition, Fig. 3(a) shows that NOR also decreased dramatically and this was different from those in the related COVID-19 lockdown studies (Liu et al., 2021; Ren et al., 2021). It demonstrated that both the declines of precursors and production rates contributed to the significant change in NO$_3^-$. It is interesting that the concentration of NO$_3^-$ was even lower at the post-alert period, which was different from the patterns of PM$_{2.5}$, NH$_4^+$ and SO$_4^{2-}$. The rise of SOR (Fig. 3(b)) from Level 3 alert to post-alert period might be a main reason since the formation rate and concentration of SO$_4^{2-}$ increased hindering the formation of NO$_3^-$. (Qiao et al., 2019). Moreover, the Neutralization Ratio was 0.73, 0.61 and 0.74 for pre-alert, Level 3 alert and post-alert periods and this could further prove that there was not enough NH$_4^+$ to neutralize NO$_3^-$ in the post-alert period. It was indicated that (NH$_4$)$_2$SO$_4$ and NH$_4$HSO$_4$ took priority over NH$_4$NO$_3$ since the formation tendency of SO$_4^{2-}$ with NH$_4^+$ was higher than NO$_3^-$. (Zheng et al., 2015). As for the carbonaceous aerosols, OC and EC, they also decreased from pre-alert to Level 3 alert by 35.9% and 13.2%, respectively, but it

![Fig. 3](https://example.com/fig3.png)  

**Fig. 3.** Boxplots of (a) NOR and (b) SOR of the three periods. Box elements present the median, lower (25th) and upper (75th) quartiles, curves outside the box show the distribution of the raw data and brown dots display the mean values.
Fig. 4. Chemical compositions in PM$_{2.5}$ of the three periods. Inorganic ions and trace elements include all the species mentioned in methodology.

showed no statistical significance ($p > 0.01$). Therefore, the weakening of secondary inorganic aerosols had a greater impact on the decline of PM$_{2.5}$ than that of carbonaceous aerosols. In terms of the changes in OC, POC and SOC decreased similarly by 38.9% and 32.4%, respectively. As shown in Fig. S4, AOC changed insignificantly ($p > 0.01$) from pre-alert to Level 3 alert, which indicated that the level of AOC was approximately equal in three periods and thus the formation of SOC was not enhanced during the Level 3 alert. This was different from those in other COVID-19 studies that the enhanced level of AOC resulted in increased mass concentrations of SOA during the lockdown period (Feng et al., 2022; Meng et al., 2021; Zhu et al., 2021b). Furthermore, the decrease in precursors, such as NO$_x$, did not cause a notably non-linear increase as response to O$_3$, which could be attributed to the dominated formation mechanism of O$_3$ being VOC-limited, instead of NO$_x$-limited (Li et al., 2020). As for the comparison between Level 3 alert and post-alert periods, the differences were insignificant ($p > 0.01$) for all species. The patterns of NH$_4^+$ and SO$_4^{2-}$ were quite similar, proving that the formation of (NH$_4$)$_2$SO$_4$ and NH$_4$HSO$_4$ took precedence over NH$_4$NO$_3$.

The changes of chemical compositions in PM$_{2.5}$ indicated that the significant decrease in PM$_{2.5}$ during Level 3 alert period was mainly due to the reduction in inorganic ions, especially NO$_3^-$. This was similar to those in previous COVID-19 studies suggesting that the short-term declines in human activities could be highly effective for the betterment of air quality (Chen et al., 2020; Sharma et al., 2020).

The variations in chemical compositions of PM$_{2.5}$ for the three periods were shown in Fig. 4. During the pre-alert and post-alert periods, inorganic ions were the dominant species, accounting for 41.9% and 35.0% in PM$_{2.5}$, respectively. However, it decreased to 29.3% with the increase in the fractions of OM and EC during the Level 3 alert period, and OM showed the highest percentage in Level 3 alert period compared to the other periods. Although the mass concentrations of carbonaceous matter declined from pre-alert to Level 3 alert, the fractions of OM and EC in PM$_{2.5}$ increased to become the dominant species in Level 3 alert period. This result provided insights into the chemical characteristic of PM$_{2.5}$ and revealed that carbonaceous matter could be a more challenging issue in the future.

### 3.2 Clustering Analysis

Fig. S5 shows the correlation-based hierarchical clustering results during pre-alert and post-alert periods. PM$_{2.5}$ was clustered with NO$_3^-$, NH$_4^+$, SO$_4^{2-}$ and K$^+$ during the pre-alert period (Fig. S5(a)), and PM$_{2.5}$ was clustered with NH$_4^+$, SO$_4^{2-}$ and K$^+$ during the post-alert period (Fig. S5(b)). It was observed that inorganic ions were grouped in the clusters containing PM$_{2.5}$, indicating that the formation of secondary inorganic aerosols was prevailing during pre-alert and post-alert periods. This was similar with that in the previous studies suggesting the formation mechanism of PM$_{2.5}$...
in urban area was mainly dominated by secondary inorganic aerosol (Guo et al., 2014; Zheng et al., 2015), especially in the haze episodes (Zhou et al., 2022). In contrast, PM$_{2.5}$ was clustered with EC, SOC and POC, without any inorganic ions during the Level 3 alert period, as shown in Fig. 5. It can be elucidated that the amount of PM$_{2.5}$ during the Level 3 alert period were more subject to the carbonaceous matter rather than secondary inorganic aerosols. This phenomenon was consistent with the changes in chemical fraction, as described in Section 3.1, which implied the difficulty in the reduction of carbonaceous aerosols. Therefore, it is essential to pay more considerations on mitigation strategies of carbonaceous aerosols. Moreover, it also provided further evidence that the changes brought by Level 3 alert were not only in the mass concentration of particles and gaseous pollutants but also the chemical compositions.

### 3.3 Source Identification and Contribution of PM$_{2.5}$

**3.3.1 PMF analysis**

The source profiles over the sampling period and the contribution of six sources to PM$_{2.5}$ identified by PMF are shown in Fig. 6. The sources were verified based on the tracers for specific sources and the BPP (Fig. S6). The detailed discussion of each source is described as follows.

Factor 1 was the greatest contributor (36.7%), which was dominated by OC, EC, Cu, Zn and Sb. OC and EC were regarded as indicators of vehicle exhaust from tail pipe (Wang et al., 2018), while Cu and Sb were identified as the tracers of brake wear (Gugamsetty et al., 2012). As for Zn, it is a tracer for tire wear (Wik and Dave, 2009). BPP plot of this factor indicated that the highest concentration existed at the near-center point, which could be attributed to traffic-related emissions since the sampling site was located at the metropolitan area nearby the main traffic artery. This factor also correlated well with NO$_x$ and CO (R = 0.73 and 0.72), suggesting that Factor 1 should be a traffic-related source.
Factor 2 had the highest proportion of Na\textsuperscript{+} and Cl\textsuperscript{−}, which could be identified as sea salt from the marine source (Liu et al., 2020; Sharma et al., 2016). BPP showed that this source mainly came from the east of the sampling site when the wind speed was relatively high. The wind rose plots in Fig. S2 have already shown that the frequencies of eastern wind were the highest and thus this factor might originate from the ocean.

Factor 3 was related to the mixing of soil dust and industrial sources, which were characterized by high percentage of Al, Co, Cr, Ti, Mn and Fe. The crustal elements, e.g., Al, Ti and Fe, were usually attributed to soil and road dust (Hsu et al., 2017), and yet Co, Cr and Mn were more likely to be the tracers of industrial source (Gugamsetty et al., 2012; Hsu et al., 2017; Sharma et al., 2016). BPP exhibited that the high concentrations came from the northwest of the sampling site and there were also some industrial parks at the similar direction of New Taipei City. Therefore, this factor was identified as a mixing source of soil dust and industry.

Factor 4 was associated with secondary sulfate, contributed by high percentages of NH\textsubscript{4}\textsuperscript{+}, SO\textsubscript{4}\textsuperscript{2−}, V and Se. This factor was the second largest contributor of PM\textsubscript{2.5} (27.9%). NH\textsubscript{4}\textsuperscript{+} and SO\textsubscript{4}\textsubscript{2−} are typical indicators of secondary inorganic aerosols, which could be integrated as ammonium sulfate. V has been referred to as tracers of heavy oil combustion, and Se was often treated as markers of coal combustion from power plants (Jeong et al., 2017; Park et al., 2019). Recent studies in Taipei city indicated that heavy oil combustion was related to shipping or port activities (Liao and Wu, 2020; Liao et al., 2021). BPP revealed that the highest contribution of this factor came from the northeast of the sampling site, which could be attributed to the aging aerosol due to regional or long-range transport, or oil combustion from power plant and Keelung port. It has also been reported that secondary pollutants from these sources may be able to transport about 30 to 50 kilometers (Hsu et al., 2017).

Factor 5 can be interpreted as burning activities with high contents of Ni and K. Ni could be representative of industrial boiler combustion and K was usually reported to originate from different kinds of biomass burning (Dall'Osto et al., 2013; Phillips-Smith et al., 2017). BPP illustrated the high concentrations of this factor coming from the north and southeast, and there’s a crematorium at the southeast of the sampling site.

Factor 6 was referred to as secondary nitrate based on the high contributions of NO\textsubscript{3−}, Cl\textsuperscript{−} and NH\textsubscript{4}\textsuperscript{+} (Gu et al., 2011). The high correlation coefficient (R = 0.88) between Factor 6 and NOR can further support this interpretation. BPP also showed high mass concentrations at the near-center

Fig. 6. Source profiles of six factors resolved by PMF model.
point as discussed in Factor 1, hence the secondary nitrate could be referred to as aging traffic aerosol. This factor had the third largest contribution to the mass concentration of PM$_{2.5}$, which was 18.1%. Factor 1 and Factor 6 accounted for 54.8% of the total PM$_{2.5}$ mass concentration, implying that sources related to traffic had the greatest impact on the air quality of Taipei metropolitan area.

### 3.3.2 Variations in source contributions of three periods

Time series of six factors resolved by PMF are shown as Fig. S7. Factor 6 exhibited the most significant decrease (95.8%) from pre-alert to Level 3 alert period, attributed to the reduction of secondary nitrate derived from aging traffic aerosols. Moreover, it can be seen that the patterns of Factor 6 in the three periods were nearly identical to NOR (Fig. 3(a)). Although Factor 1 was a traffic-related emission source, it did not change as much as Factor 6. Average contributions and distributions of each source are shown in Fig. 7. The mean value of Factor 1 for pre-alert was 3.75 µg m$^{-3}$, while those of Level 3 alert and post-alert were 3.04 µg m$^{-3}$ and 3.10 µg m$^{-3}$, respectively. As for Factor 6, there was a clear slump from pre-alert to Level 3 alert (4.77 µg m$^{-3}$ to 0.20 µg m$^{-3}$) and even lower (0.09 µg m$^{-3}$) during the post-alert period. This has been discussed in the previous section and implied that secondary pollutants from traffic sources decreased more noticeably than primary pollutants. The reduction of traffic-related sources (Factor 1 + Factor 6) was 5.28 µg m$^{-3}$ from pre-alert to Level 3 alert, contributing to 44.7% of the decrease of PM$_{2.5}$. It was observed that the contribution of Factor 2 existed a slight change in the Level 3 alert period. This might be due to the weakening of wind from east and northeast. Factor 3 exhibited a small increase in Level 3 alert period, which was the only source that increased. It might be partially due to one of

![Fig. 7. Comparison of 6 factors from PMF results in the three periods. Each y-axis represents the concentration of PM$_{2.5}$ (µg m$^{-3}$).](https://example.com/fig7.png)
the important differences between the regulations of Level 3 alert and lockdown since most of the factories did not shut down during Level 3 alert period. The changes of Factor 4 during Level 3 alert were notable, with contributions of 2.91, 1.56 and 3.07 µg m\(^{-3}\) to PM\(_{2.5}\) for pre-alert, Level 3 alert and post-alert periods, respectively. This could result from the reduction of regional & long-range transport (Griffith et al., 2020) and the declined shipping activities. Factor 5 was burning activities and its contribution was the lowest during Level 3 alert period, which could be expected since these kinds of human activities indeed diminished. It was noticeable that, even in Level 3 alert period, PM\(_{2.5}\) was still higher than the threshold recommended by World Health Organization (5 µg m\(^{-3}\)). Hence, traffic-related emission (Factor 1), especially for the carbonaceous matter, should be a vital role to deal with. This also implied that cleaner energy for vehicles is needed in the future to reduce traffic-related emission (Wong et al., 2022).

### 3.4 Health Risk Assessment of Trace Elements

The carcinogenic risk and non-carcinogenic risk through inhalation exposure of trace elements are shown in Fig. 8. The values of ECR were 7.89 × 10\(^{-7}\), 5.55 × 10\(^{-7}\) and 5.66 × 10\(^{-7}\) in pre-alert, Level 3 alert and post-alert periods, HQ was 1.03 × 10\(^{-1}\), 8.4 × 10\(^{-2}\) and 1.44 × 10\(^{-1}\) respectively. Although the health risk was all below the acceptable level in three periods (ECR < 10\(^{-6}\), HQ < 1), both ECR and HQ exhibited a pattern of the lowest values during the Level 3 alert period. Cr and As were the dominant species for ECR, while Ni and Mn significantly contributed to HQ. According to PMF results, the health risk in Taipei was mainly caused by Factor 3 and Factor 5, which were identified as soil dust, industrial and burning activities. In addition, the dominant species contributing to health risk were the same in all periods and were also similar to a recent study in New Taipei city (Hsu et al., 2019), indicating the importance of industry-related trace elements to human health. Although the sum of Factor 3 and Factor 5 contributed to less than 10% of the PM\(_{2.5}\) mass concentrations, they should be taken care of to level down the human’s health risk. This indicated that the priority in air pollution control are different between PM\(_{2.5}\) mass concentrations and

![Fig. 8. Bar plots and changes for health risk assessment results. (a) ECR, (b) HQ.](image)
health risk. More efforts should be made on industrial sources in the future to reduce these trace elements, especially some carcinogenic heavy metals.

As for the variations in the health impact induced by trace elements, ECR and HQ showed slighter and insignificant decrease (−29.6% and −18.9%, both $p > 0.01$) compared to PM$_{2.5}$ from pre-alert to Level 3 alert period. It indicated that health risks caused by the trace elements were somewhat more difficult to control than the total PM$_{2.5}$ (Wang et al., 2022), which might be associated with soil dust & industrial sources. This further verified that some of the minor sources of PM$_{2.5}$ were non-negligible regarding to human health risk. The results could serve as implication for policy maker that the concentrations of trace elements should be considered more carefully as there was no significant decrease even during the unprecedented Level 3 alert period.

4 CONCLUSIONS

In this study, the variations of chemical compositions, sources and health risk of PM$_{2.5}$ in Taipei were investigated across the pre-alert, Level 3 alert and post-alert periods. Significant differences were found in the sources, mass concentrations and chemical compositions of PM$_{2.5}$ among these three periods. According to the PMF results, secondary nitrate (Factor 6) and secondary sulfate (Factor 4) decreased most significantly and contributed to the remarkable reduction in PM$_{2.5}$ (−49.4%). Among the 6 factors, secondary nitrate was the greatest contributor to the decrease in PM$_{2.5}$, which could be due to the decline in traffic congestion level. However, traffic-related emission (Factor 1) changed insignificantly ($p > 0.01$), indicating that secondary pollution of traffic emission could be more easily controlled than primary traffic emission. In the future, air pollution control strategies should consider traffic-related carbonaceous emissions and the energy of vehicles more comprehensively to better improve the air quality.

Compared to pre-alert and post-alert, the fractions of chemical compositions in PM$_{2.5}$ were different during Level 3 alert, when the fraction of inorganic ions was the lowest with higher percentages of OM and EC. In addition, the clustering analysis showed that the variation in PM$_{2.5}$ was associated with carbonaceous matter during Level 3 alert period while inorganic ions clustered with PM$_{2.5}$ during both pre-alert and post-alert periods. Such outcomes can be concluded that more attention should be paid to reducing carbonaceous matter in future work since secondary inorganic aerosols could decrease more significantly than carbonaceous aerosols when human activities declined. In addition, ECR and HQ of the trace elements were the lowest during the Level 3 alert period, yet the degrees of changes were smaller than those of PM$_{2.5}$ and inorganic ions. As a result, the characteristics and concentrations of atmospheric trace elements need to be better understood and reduced since the number of patients with lung cancer is still growing in Taiwan. This study elucidated the variations in chemical characteristics and sources contributions of PM$_{2.5}$ during an unprecedented condition compared to the normal periods, which helps in prioritizing mitigation strategies of air pollutants in the future.

ACKNOWLEDGEMENTS

This research was funded by the National Science and Technology Council, Taiwan, under grant No. NSTC 111-2222-E-002-012.

SUPPLEMENTARY MATERIAL

Supplementary material for this article can be found in the online version at https://doi.org/10.4209/aaqr.220329

REFERENCES

Bera, B., Bhattacharjee, S., Sengupta, N., Saha, S. (2022). Variation and dispersal of PM$_{10}$ and PM$_{2.5}$ during COVID-19 lockdown over Kolkata metropolitan city, India investigated through HYSPLIT


