Since January 2020 Elsevier has created a COVID-19 resource centre with free information in English and Mandarin on the novel coronavirus COVID-19. The COVID-19 resource centre is hosted on Elsevier Connect, the company's public news and information website.

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1. Introduction

Fine particulate matters (PM$_{2.5}$) attract extensive attention worldwide due to their hazards and risks involving the deterioration of climate conditions, human health, and the environment (Jimenez et al., 2009). Cardiovascular disease was found to be associated with exposure to fine particles (PM$_{2.5}$) through the direct deposition of these particles in the lungs. The systemic inflammation and oxidative stress were found to be induced by exposure to PM$_{2.5}$ (Li et al., 2019; Pope III et al., 2004). For the purpose of social health and the sustainable environment development, the Taiwan EPA has promulgated the PM$_{2.5}$ air quality standard with annual and daily averages of 15 and 35 μg/m$^3$, respectively (Taiwan, 2012). Consequently, the annual variation ratio and the exceedance days of the PM$_{2.5}$ concentrations in central Taiwan from 2006 to 2017 was found to be –3.42% and –4.67, respectively (Cheng and Hsu, 2019). However, a previous study found the secondary PM$_{2.5}$ concentrations such as nitrate (NO$_3$) and ozone (O$_3$) concentrations increased during fall and winter in central Taiwan. It was because the formation of O$_3$ concentrations was dominated by the VOC-limited chemical reaction (Lin et al., 2022b). The O$_3$ production mechanism in the troposphere through the reaction between volatile organic compound (VOC), nitrogen oxide (NO$_x$ = NO+NO$_2$), and solar radiation (SR) is complex and nonlinear (Seinfeld and Pandis, 2006). Under the NO$_x$-limited O$_3$ production regime, the high VOC concentrations and the low NO$_x$ level condition enhance the production of O$_3$ (Kroll et al., 2020). Under the condition that the VOC concentrations are low and the NO$_x$ concentrations are high, the O$_3$ production mechanism shifts to the VOC-limited regime. More O$_3$ and secondary NO$_x$ could be produced due to a decrease in the depletion of atmospheric oxidant including OH and O$_3$ by the reduction of NO$_x$ concentrations (Bhatti et al., 2022). Owing to previous studies’ findings, there are many uncertainties in the production and dissipation of PM$_{2.5}$ and precursors in the atmosphere. The chemical reactions between NO$_x$, sulfur dioxide (SO$_2$), O$_3$, and VOCs in the atmosphere need further investigation.

Due to the epidemic outbreak of the COVID-19 at the end of 2019, partial or whole lockdown measures were inevitable to be implemented for many cities to control contagious viruses. Previous studies showed that the COVID-19 alert reduced anthropogenic activities, resulting in a significant reduction in primary or secondary air pollutants concentrations in many cities (Faridi et al., 2021; Jephcote et al., 2021; Zangari...
2. Materials and methods

2.1. Sampling site and datasets

The sampling sites are shown in Fig. S1. The datasets used in the present study were extracted from an air quality monitoring stations located at Taiwan Boulevard (24°10′53.97″ N, 120°35′47.02″ E) in Taichung city from January 2019 to August 2021, which can be downloaded on http://taqm.epb.taichung.gov.tw/.

Hourly air pollutant concentrations datasets were provided by the Environmental Protection Bureau of the Taichung City Government, who carried out the air quality monitoring campaign. The beta gauge monitor (BAM-1020, Met One Instruments, Inc.) was deployed to monitor the air quality monitoring stations located at Taiwan Boulevard (24°10′53.97″ N, 120°35′47.02″ E) in Taichung city from January 2019 to August 2021, which can be downloaded on http://taqm.epb.taichung.gov.tw/.

2.2. Kinetic analysis of the secondary NO$_3^-$ and SO$_4^{2-}$

To better understand the atmospheric chemistry of secondary PM$_{2.5}$, including NO$_2$ and SO$_2$ during the Level 3 COVID-19 alert, the diurnal variation of the kinetic production rate of HNO$_3$ (g) (μg/m$^3$/h), R[HNO$_3$], and the HSO$_4^-$ production rate (μg/m$^3$/h), R[HSO$_4^-$], were calculated and compared with the measured NO$_3^-$ and SO$_4^{2-}$ concentrations, respectively. The calculation method developed by Lin et al. (2022b) is shown at section S1 in the Supplementary data and is briefly summarized here. The total hourly production rate of [HNO$_3$] (μg/m$^3$) concentration is given by:

$$R[HNO_3] = R[HNO_3]_{\text{daytime}} + U(SR) \times R[HNO_3]_{\text{night}}$$

where U(SR) is the unit step function, which is given as follows:

$$U(SR) = \begin{cases} 1, & SR > 10 \\ 0, & 0 \leq SR \leq 10 \end{cases}$$

For the SO$_4^{2-}$ concentration predictions, the HSO$_4^-$ production rate was calculated by the following equation (Seinfeld and Pandis, 2006):

$$R[HSO_4^-] = k_{SO_2-\text{OH}}[OH][SO_2]$$

where $k_{SO_2-\text{OH}}$ is the bimolecular rate constant (cm$^3$ molecule$^{-2}$ s$^{-1}$) (Table S1).

2.3. Artificial neural network model

In this study, four models, model 1 to model 4, based on the ANN technique, were developed to predict NO$_3^-$, O$_3$, nitrate (NO$_3^-$), and sulfate (SO$_4^{2-}$) concentrations. Spearman’s ρ between the predicted targets, including NO$_3^-$, O$_3$, SO$_4^{2-}$, and NO$_3^-$ with input variables, was calculated to select the critical input parameters for model development, as shown in Table S2. The present model was developed based on the procedures as shown in Fig. 1.

The ANN technique has been applied to develop ambient PM$_{2.5}$ concentrations forecasting models in previous studies (Biancofiore et al., 2017; Kim et al., 2012; Lin et al., 2022b; Park et al., 2018; Soh et al., 2018; Xing et al., 2020; Zhao et al., 2019). The prediction accuracy of the ANN is dependent on the network structure comprising user-defined input, hidden, and output layers in the model (Bishop, 2007). The input datasets prepared by users are imported into the input layer. In the hidden layers, a sigmoid transfer function transforms and adjusts the input variables by a weight and transmits data to the hidden layers.

The traffic flows datasets were provided by the Transportation Bureau of TCG, which can be downloaded on the website of http://e-traffic.taiw.gov.tw/RoadGrid/Pages/VD/History2.html. With limited traffic flow monitoring stations in Taichung City, the traffic flows at the intersection between Chaoma Road and Huanzhong Road, marked in red (Fig. S1) were extracted. The effect of the traffic volume on the variation in NO$_2$, O$_3$, SO$_4^{2-}$, and NO$_3^-$ concentrations during the Level 3 COVID-19 alert was investigated.

Further investigation of various trends in the secondary PM$_{2.5}$ formation and secondary inorganic aerosols, including sulfate (SO$_4^{2-}$) and nitrate (NO$_3^-$), during the lockdown is needed (Kroll et al., 2020). However, most previous studies focused on the variation of PM$_{2.5}$, O$_3$, and NO$_2$ emissions and stronger photochemical reactions, respectively. The calculation method developed by Lin et al. (2022b) is shown at section S1 in the Supplementary data and is briefly summarized here. The total hourly production rate of [HNO$_3$] (μg/m$^3$) concentration is given by:

$$R[HNO_3] = R[HNO_3]_{\text{daytime}} + U(SR) \times R[HNO_3]_{\text{night}}$$

where U(SR) is the unit step function, which is given as follows:

$$U(SR) = \begin{cases} 1, & SR > 10 \\ 0, & 0 \leq SR \leq 10 \end{cases}$$

For the SO$_4^{2-}$ concentration predictions, the HSO$_4^-$ production rate was calculated by the following equation (Seinfeld and Pandis, 2006):

$$R[HSO_4^-] = k_{SO_2-\text{OH}}[OH][SO_2]$$

where $k_{SO_2-\text{OH}}$ is the bimolecular rate constant (cm$^3$ molecule$^{-2}$ s$^{-1}$) (Table S1).

The AIM technique has been applied to develop ambient PM$_{2.5}$ concentrations forecasting models in previous studies (Biancofiore et al., 2017; Kim et al., 2012; Lin et al., 2022b; Park et al., 2018; Soh et al., 2018; Xing et al., 2020; Zhao et al., 2019). The prediction accuracy of the ANN is dependent on the network structure comprising user-defined input, hidden, and output layers in the model (Bishop, 2007). The input datasets prepared by users are imported into the input layer. In the hidden layers, a sigmoid transfer function transforms and adjusts the input variables by a weight and transmits data to the hidden layers.
An algorithm that uses a feed-forward neural network to learn is updated its weights until it reaches an iteration convergence (Fig. 1(a)). In this study, the MATLAB ANN toolbox (2021a) was used to train, validate, and test the NN. The datasets were divided randomly into 70, 15, and 15% for testing, validation, and testing, respectively. The number of neurons from 10 to 100 in the hidden layer was examined 15, and 15% for testing, validation, and testing, respectively. The updated its weights until it reaches an iteration convergence (Fig. 1 (a)).

The contribution of each input variable to NO\textsubscript{3}, O\textsubscript{3}, NO\textsubscript{2}, and SO\textsubscript{2}\textsuperscript{2} was investigated through the sensitivity analysis to verify the effect of meteorological variables and precursors on targets including NO\textsubscript{3}, O\textsubscript{3}, SO\textsubscript{2}\textsuperscript{2}, and NO\textsubscript{2}. The variation of each input variable was calculated using the equation as follows:

\[ \Delta \delta_i = |y_i,2020 - y_i,2021| \]  
\[ \Delta \delta_T = |y_T,2020 - y_T,2021| \]

where \( \delta_i \) is the input variable, \( \delta_T \) is the target variable, \( y_i \) and \( y_T \) are the targets measured in 2020, and \( y_i \) and \( y_T \) are the targets measured in 2021, respectively.

All datasets were then normalized using the equation as follows:

\[ \xi_i = \frac{\Delta \delta_i - \Delta \delta_{i, \text{min}}}{\Delta \delta_{i, \text{max}} - \Delta \delta_{i, \text{min}}} \]

where \( \xi_i \) is the normalized value; \( \Delta \delta_{i, \text{min}} \) is the minimum value of the hourly variation, and \( \Delta \delta_{i, \text{max}} \) is the maximum value of the hourly variation. The ANN model is then trained and tested based on \( \xi_i \) according to the same procedures described in Section 2.3 to assess the contribution of various inputs \( \Delta \delta_{i,j} \) to the targets \( \Delta \delta_{T,j} \). The sensitivity of each input variable, \( S \), then can be calculated as follows:

\[ S = |U^*_j - U^*_i| \]

where \( U^*_j \) is the average of the model output when the tested input variable is set to be 1 and \( U^*_i \) is the average of the model output when the tested input variable is set to be 0. The output was then denormalized back to the original scale for each input variable. The method to develop the present model was derived referring to the concept proposed in previous studies (Lung et al., 2020; Zwack et al., 2011a; Zwack et al., 2011b), in which a multi-variable linear regression model was established for assessing the importance of multiple emission sources to PM\textsubscript{2.5} concentrations.
3. Results

3.1. Mean monthly variation in air pollutants

Fig. 2 shows the monthly mean PM$_{2.5}$, WIS, and precursors concentrations to investigate the effect of local meteorological conditions on the air quality of Taichung City from 1st January 2019 to 31st August 2021. The mean values and standard deviation as shown in Fig. 2 are listed in Table S3-S5. In 2019, the highest PM$_{2.5}$ concentration of 31.3 $\mu$g/m$^3$ and NO$_3^-$ concentrations of 9.48 $\mu$g/m$^3$ occurred in March (spring), decreased to the lowest concentration of 6.82 and 3.04 $\mu$g/m$^3$ for PM$_{2.5}$ and NO$_3^-$, respectively in summer, and then rose again in fall and winter, as shown in Fig. 2 (c). Similar results were also detected in 2020. These results implicated that the seasonal variations of air pollutant concentrations were significantly affected by the regional weather, cross-boundary transport of air pollutants, and mass transfer conditions (Cheng and Hsu, 2019; Lee et al., 2020). The prevailing wind changed to the northeastern monsoon during spring and winter in Taichung City, where was in the windward side of southwest monsoon. Under this meteorological condition, the convective and diffusive effects was enhanced to reduce air pollutant concentrations. In Fig. 2 (a) and (e), similar mean monthly variation trends for gaseous pollutants are observed. For the variation of O$_3$ concentrations, although the weather in summer grows warm with the aid of strong solar radiation to improve the photochemical reaction, the O$_3$ concentrations declined to the lowest level of 22.87 ppb in 2019 and 20.21 ppb in 2020 due to an effective convective flow conditions (Tsai et al., 2008) and a high rain intensity (Yen and Chen, 2000) (Fig. 2 (a), (e), (i)). During the fall season, the largest monthly mean O$_3$ concentration of 35.44 ppb, in 2019 and 2020 was observed. It was because a weak convection-dispersion condition and a presence of strong solar radiation were in favor of an increase in O$_3$ concentrations (Hsu and Cheng, 2020).

3.2. Diurnal variation in air pollutants

The diurnal variation of PM$_{2.5}$, WIS, precursors concentrations, meteorological parameters, and traffic volume from May 19 to July 27 over the period 2019–2021 is shown in Fig. 3. In the sight of the diurnal variation of each year, the NO$_x$ concentrations peaked at 6:00–8:00, decreased to the lowest level at noon, and then increased from 14:00–18:00 (Fig. 3 (a), (e), and (f)), which was associated with the traffic volume, as shown in Fig. 3 (d), (h), and (l). The diurnal variation of O$_3$ concentrations was observed to show an inverse trend as compared with that of NO$_x$ concentrations, wherein the O$_3$ concentrations increased to the highest level at noon due to the strong photochemical reactions. PM$_{2.5}$ concentrations exhibited a similar diurnal variation during the same period within three years, where the PM$_{2.5}$ concentration increased to the highest level around noon (Fig. 3 (b), (f), (j)). According to this result, the formation of secondary PM$_{2.5}$ could be influenced by traffic emissions, boundary layer height, and the photochemical reaction. NO$_x$ concentrations have similar variation trends as that of PM$_{2.5}$ concentrations, as shown in Fig. 3 (b), (f), and (j), in which a significant peak at noon is observed. It was because the homogeneous reaction between gaseous NH$_3$ and HNO$_3$ with the aid of sunlight enhanced the secondary particle formation (Lin et al., 2022a).

There is a divergent diurnal variation trend for SO$_2^-$ concentrations in 2021 as compared to that in 2019 and 2020 (Fig. 3 (b), (f), and (j)). In 2019 and 2020, the SO$_2^-$ concentrations peaked at noon due to the gas to particle partitioning through the photochemical reaction between HSO$_4^-$ and OH$^-$ radicals (Lin et al., 2022a). After 21:00, an insignificant diurnal pattern of SO$_2^-$ was observed because the regional accumulation of nonvolatile sulfate was observed to be critical to the diurnal trend of SO$_2^-$ in Taichung City (Lin et al., 2022b). A significant increase in the SO$_2^-$ concentration in 2021 (Fig. 3 (j)) by 41.6% (Fig. 3 (b)) and 60.7% (Fig. 3 (f)) at 13:00 was observed relative to 2019 and 2020, respectively. This result implicated that an increase in SO$_2^-$ concentration at noon during the pandemic of COVID-19 was enhanced by the photochemical reaction.

3.3. The kinetic model for NO$_3^-$ and SO$_2^-$ formation mechanism investigation

Fig. 4a shows the variations of P$_{[NO_3]}$ and the measured NO$_3^-$ concentrations in the diurnal pattern during the Level 3 COVID-19 alert. From 9:00 to 13:00, the variation trend of P$_{[NO_3]}$ tracks the NO$_3^-$ concentration very well, suggesting that the secondary PM$_{2.5}$ formation is dominated by the photochemical reaction. From 14:00 to 18:00, the boundary layer height increases due to high temperature to generate a dilution effect on air quality to compensate the photochemical formation of HNO$_3$, resulting in a decrease in the NO$_3^-$ concentration (Leung et al., 2020). From 20:00 to 4:00, an increase in both NO$_3^-$ concentration and P$_{[NO_3]}$ further indicates a heterogeneous reaction between N$_2$O$_5$ and H$_2$O controls the formation of NO$_3^-$ at night. In Fig. 4 (c), from 8:00 to 12:00, the SO$_2^-$ concentration was observed to slightly increase with increasing P$_{[HNO_3]}$, by the homogeneous reaction of gaseous H$_2$SO$_4$, Gaseous H$_2$SO$_4$ rapidly condenses into existing atmospheric particulate matters or forms secondary particles (Salcedo et al., 2006). After 18:00, an insignificant diurnal variation of SO$_2^-$ suggests that the local SO$_2^-$ are accumulated aged particles rather than secondary particles because a large deviation between the measured SO$_2^-$ concentration and P$_{[HNO_3]}$ is observed as shown in Fig. 4 (e).

It is noted that although the theoretical kinetic analysis for the diurnal variation of NO$_3^-$ and SO$_2^-$ can provide reliable scientific evidence to explain the possible formation mechanism for the local secondary PM$_{2.5}$, the R$^2$ between the calculated and measured NO$_3^-$ and SO$_2^-$ concentration is as low as 0.04 and 0.58, respectively. Therefore, a more accurate prediction model should be developed to track the diurnal variation of NO$_3^-$ and SO$_2^-$ concentrations.

3.4. The ANN prediction model development

Fig. 5 shows a linear correlation between observed values and predicted NO$_x$, O$_3$, NO$_3^-$, SO$_2^-$ calculated by Model 1 (Fig. 5(a)), Model 2 (Fig. 5(b), Model 3 (Fig. 5(c)), and Model 4 (Fig. 5(d)), respectively. The input variables in each model are shown in Table S6. After screening out some redundant variables, the input variables including PM$_{10}$, PM$_{2.5}$, SO$_2$, CO, O$_3$, THC, CH$_4$, NMHC, SO$_2^-$, NO$_3^-$, NH$_4^+$, Cl$^-$, RH, AT, and traffic volume were selected to develop model 1 (Table S6). A total of 886, 111, and 111 data points were randomly divided for training, validation, and testing, respectively. As shown in Fig. 5 (a), Model 1 can predict 80.1% of NO$_x$, with RMSE and MAE of 2.51 and 2.41 ppbv, respectively. For model 2, the input variables were PM$_{10}$, PM$_{2.5}$, SO$_2$, CO, NO$_x$, THC, CH$_4$, NMHC, SO$_2^-$, NO$_3^-$, NH$_4^+$, Cl$^-$, RH, AT, and traffic volume (Table S6). Model 2 can predict 77.0% of O$_3$, with RMSE and MAE of 3.60 and 4.65 ppbv, respectively (Fig. 5(b)). The input variables for model 3 and model 4 were PM$_{10}$, PM$_{2.5}$, SO$_2$, CO, NO$_x$, O$_3$, THC, CH$_4$, NMHC, RH, AT, and traffic volume (Table S6). Model 3 and model 4 can predict 72.6 and 67.2% of NO$_3^-$ and SO$_2^-$ concentrations, respectively. The RMSE and MAE for model 3 is 0.61 and 0.95 $\mu$g/m$^3$, respectively. For Model 4, the RMSE and MAE are 0.05 and 0.07 g/m$^3$, respectively. Fig. 6 shows a comparison of the diurnal variation between the predicted values and the measured concentrations for NO$_x$ (Fig. 6(a)), NO$_3^-$ (Fig. 6(b)), NO$_2$ (Fig. 6(c)), and SO$_2^-$ (Fig. 6(d)) during the Level 3 COVID-19 alert. Clearly, the diurnal trends of predicted concentrations agree reasonably with measured values with a deviation of 0.26–13.1% for NO$_x$, 0.01–21.1% for O$_3$, 0.10–14.5% for NO$_3^-$, and 0.11–19.4% for SO$_2^-$ in general, the largest deviation occurred at the turning point. For example, for NO$_3^-$, the largest deviation between the measured and the
Fig. 2. The mean monthly variation in PM$_{2.5}$, WIS, precursors concentrations, and meteorological parameters. (a) The mean monthly variation in NO$_x$, O$_3$, and SO$_2$ concentrations, (b) The mean monthly variation in CO, NMHC, and THC concentrations, (c) The mean monthly variation in PM$_{2.5}$ and WIS concentrations, and (d) The mean monthly variation in RH and T.
Fig. 3. Diurnal variation in PM$_{2.5}$, WIS, precursors concentrations, meteorological parameters, and traffic volume from May 19 to July 27, 2019–2021. (a), (e), (i) Diurnal variation in NO$_2$, O$_3$, and SO$_2$ concentrations; (b), (f), (j) Diurnal variation in PM$_{2.5}$ and WIS concentrations; (c), (g), (k) Diurnal variation in RH and T; (d), (h), (l) Diurnal variation of traffic volume.
Fig. 4. (a) Relationship between the diurnal variation of $P_{\text{HNO}_3}$ ($\mu g/m^3/h$), NOR, and the measured NO$_3^-$ concentration ($\mu g/m^3$) and (b) Relationship between the diurnal variation of $P_{\text{HSO}_3}$ ($\mu g/m^3/h$), SOR, and the measured SO$_4^{2-}$ concentration ($\mu g/m^3$) during the Level 3 COVID-19 alert.
Fig. 5. Correlation between observed values and predicted values by (a) Model 1, (b) Model 2, (c) Model 3, and (d) Model 4 based on the current time variables.
predicted value of 14.5% was at 11:00 (Fig. 6(c)), in agreement with the occurrence of the peak value of $P_{\text{[HNO}_3\text{]}}$ as shown in Fig. 4(a). A similar result was also observed for $\text{SO}_4^{2-}$ with the highest deviation of 19.4%, as shown in Fig. 6(d). This result suggests that the complex atmospheric chemical reaction for NO$_3^-$ and SO$_4^{2-}$ production, and meteorological factors such as RH, T, and boundary layer height contribute to the diurnal variation of the secondary PM$_{2.5}$. Therefore, to enhance the accuracy and applicability of the ANN model, long-term datasets for the WIS, OH radical concentrations, precursors for the secondary inorganic particle formation, including NH$_3$, HNO$_3$, and H$_2$SO$_4$, and the boundary layer height are recommended to be taken as the input variables to develop the secondary PM$_{2.5}$ prediction models.

4. Discussion

4.1. Impact of the Level 3 alert on the variations in air pollutant concentrations

In previous studies (Vu et al., 2019; Zheng et al., 2020; Liu et al., 2020a, 2020b; Shi et al., 2021), the de-weather model excluded the meteorological factors based on boosted regression tree (RF) approach was utilized to investigate the effect of primary emissions on the variations in air pollutants. It was because the differences of meteorological conditions between their study periods were very high. The reduced air pollutant concentrations were dominated by the emission control measures implemented in Wuhan (Zheng et al., 2020). During the COVID lockdown, the predicted de-weather values in Hangzhou showed that the reduction of NO$_x$ concentrations resulted majorly due to a decrease in traffic volume (Liu et al., 2020a, 2020b). The reduction of NO$_x$ resulted in an increase in O$_3$ concentrations (Liu et al., 2020a, 2020b). The reduction of NO$_x$ resulted in an increase in O$_3$ concentrations (Liu et al., 2020a, 2020b), which weakened the NO-titration effect on O$_3$ and contributed to the significant increase in O$_3$ (Li et al., 2019). An increase in O$_3$ concentrations further enhanced the formation of secondary aerosols (Liu et al., 2020a, 2020b).

To investigate the effect of the meteorological factors on the variations in air pollution concentrations, the weather conditions were analyzed. Figs. S2, S3, and S4 show the time series of hourly average meteorological parameters, PM$_{2.5}$, WIS, and gaseous pollutants concentrations in the same period in 2019, 2020, and 2021, respectively. The monthly average T and RH were in the range of 28.7–29.5 °C and 78.1%–87.5% for the three years, respectively. The average WS was 2.50 m/s in 2019, 1.3 m/s in 2020, and 1.2 m/s in 2021. The prevailing
WD was dominated by the southwest monsoon within three years. These statistical results revealed that the meteorological parameters were under the same condition for three different years during the observation period. That is, the uncertainties in the effect of the meteorological parameters on the variation of air pollutant concentrations during the Level 3 COVID-19 alert were comparable to the same period in 2019 and 2020 and could be excluded. The method to isolate the influences of the meteorological factors on the air pollutant concentrations in the present study was according to the concept used in several previous studies (Baldasano et al., 2020; Berman and Ebisu, 2020; Dantas et al., 2020; Sharma et al., 2020; Huang et al., 2021). For example, Dantas et al. (2020) studied the effect of the city lockdown on the air pollution concentrations in a city of Brazil, by comparing the PM$_{10}$, CO, NO$_2$, and O$_3$ concentrations with values obtained in the same period of 2019. They found that the meteorological interferences in the air quality were dominated by the air pollutants transported from an industrial park (Dantas et al., 2020).

During the Level 3 COVID-19 alert in June 2021, the average NO$_2$ concentration decreased by 20.0% and 25.9% as compared to that in June 2019 and June 2020, respectively, it was not surprising that, due to the work from home policy, distance learning, and a reduction in the traffic flow. This decreasing level was similar to 25.5% in the Continental United States (Berman and Ebisu, 2020) and 20% in Bakersfield and San Francisco (Naeger and Murphy, 2020). Although a decrease in PM$_{2.5}$ concentrations from 9.73 μg/m$^3$ in 2019 to 6.60 μg/m$^3$ in 2021 was observed, there was no significant difference in the PM$_{2.5}$ concentration between June–July in 2020 (6.05 μg/m$^3$) and June–July in 2021 (6.60 μg/m$^3$). This result reveals that traffic emissions, as well as local stationary pollution sources, controlled the variation of PM$_{2.5}$ concentration in Taichung City. In 2021, the average O$_3$ concentration slightly increased by 5.76% and 13.2% as compared to that in 2019 and 2020, respectively. Besides, the NO$_3$ concentration increased by 46.4% and 98.2%, compared to the equivalent period in 2019 and 2020 (Fig. 2 (e), (g), and (k)), respectively. This was because the O$_3$ production is under the VOC-limited reaction, in which a reduction of the NO$_3$ concentration enhanced the production of precursors such as OH radicals to produce more O$_3$ and secondary PM$_{2.5}$ (Bhatti et al., 2022).

A comparison of the traffic volume in the diurnal pattern during the Level 3 COVID-19 alert in 2021 (Fig. 3 (l) with that in the same period in 2019 (Fig. 3 (l)) and 2020 (Fig. 3 (h)) revealed that the traffic volume reduced by 19.8% and 34.9% during rush hour at 8:00 relative to 2019 and 2020, respectively. This traffic volume reduction at 8:00 in 2021 caused a decrease in NO$_x$ by 27.9% and 20.2% relative to 2019 and 2020, respectively. There was an insignificant change in the O$_3$ concentration from the perspective of diurnal variation within three years. However, a significant increase in the NO$_3$ concentration in 2021 (Fig. 3 (b)) by 38.2% (Fig. 3 (f)) and 63.1% (Fig. 3 (j)) at 12:00 was also observed relative to 2019 and 2020, respectively. This finding further revealed that the production of secondary inorganic particles was under the NO$_3$-saturated regime during the Level 3 COVID-19 alert, wherein a reduction of NO$_x$ emission could decrease the exhaustion of OH and O$_3$ by the chemical reaction with NO$_x$, resulting in an increase in NO$_3$ production (Lin et al., 2022a).

### 4.2. Homogeneous formation of NO$_3$ and SO$_4^{2-}$

As shown in Section 3.3, the present model could be used to simulate the homogeneous and photochemical formation of the diurnal variations in NO$_3$ and SO$_4^{2-}$ concentrations. To further investigate the heterogeneous formation of the NO$_3$ and SO$_4^{2-}$, the nitrogen oxidation ratio (NOR) and sulfur oxidation ratio (SOR) were calculated as follows:

$$\text{NOR} = \frac{[\text{NO}_3^-]_{\text{mol}}}{[\text{NO}_2^+]_{\text{mol}} + [\text{NO}_3^-]_{\text{mol}}} \quad (11)$$

$$\text{SOR} = \frac{[\text{SO}_4^{2-}]_{\text{mol}}}{[\text{SO}_2^+]_{\text{mol}} + [\text{SO}_4^{2-}]_{\text{mol}}} \quad (12)$$

where $[\text{NO}_3^-]_{\text{mol}}$, $[\text{NO}_2^+]_{\text{mol}}$, $[\text{SO}_2^+]_{\text{mol}}$, and $[\text{SO}_4^{2-}]_{\text{mol}}$ are concentrations in mmol/m$^3$. The production of the secondary inorganic aerosols by heterogeneous mechanism occurs accompanied by an increase in NOR/SOR, humidity, and aerosol mass concentration (Zhang et al., 2022). As shown in Fig. 4 (b), the NOR was in the range of 0.04–0.09, which was a very low level compared to 0.19–0.43 calculated in China during the lockdown period (Liu et al., 2020a, 2020b). An insignificant increasing trend was observed in NO$_3$ concentrations with increasing RH and NOR (Fig. 4(a)), suggesting that the diurnal variation of the secondary NO$_3$ was dominated by the photochemical reactions during the Level 3 COVID-19 alert. This result is in good agreement with the diurnal variation trend in the NO$_3$ concentrations predicted by the present kinetic model.

The SOR was calculated to be 0.31–0.43, very close to the level of about 0.26–0.49 calculated in China (Liu et al., 2020a, 2020b). An increase in SO$_4^{2-}$ concentration with increasing SOR and RH occurred at 20:00–22:00 (Fig. 4(a)), indicating that the heterogeneous reaction dominated the nighttime formation of SO$_4^{2-}$.

### 4.3. Effect of the meteorological conditions and the primary emissions on the air pollution indicators

In the present study, (as discussed in Section 4.1), the uncertainties in the effect of the meteorological factors on air pollutant concentrations were excluded. To further verify the contributions of input variables to the variations of air pollutants in Taichung City, an easy-to-use ANN model was developed. Fig. 7 shows the importance of each input variable to the variations of primary traffic emissions. This result agrees with the previous findings (Liu et al., 2020a, 2020b; Zheng et al., 2020). As shown in Fig. 7 (b)-(d), VOCs dominate the variations in O$_3$, NO$_x$, and SO$_4^{2-}$ concentrations between 2020 and 2021 using sensitivity analysis based on the ANN model. This analysis is applicable for the verification of the variation of air pollutants controlled by meteorological parameters or primary emissions. As shown in Fig. 7(a), the two important contribution factors are Δ$\delta$, NMHC (6.45 μg/m$^3$) and Δ$\delta$, CO (6.24 μg/m$^3$), which are the primary tracers for traffic emissions (DeCarlo et al., 2010). This result suggests that the variations of NO$_3$ concentrations during the Level 3 COVID-19 alert were dominated by the variations of primary traffic emissions. This result agrees with the previous findings (Liu et al., 2020a, 2020b; Zheng et al., 2020). As shown in Fig. 7 (b)-(d), VOCs dominate the variations in O$_3$, NO$_x$, and SO$_4^{2-}$ concentrations, in which the contributing factors are 22.9 ppb to Δ$\delta$, O$_3$, 4.25 μg/m$^3$ to Δ$\delta$, NO$_3$, and 2.16 μg/m$^3$ to Δ$\delta$, SO$_4^{2-}$. This result sheds light on the variations of secondary inorganic aerosols controlled by the local air pollutants through the photochemical reaction between precursors and oxidants in the atmosphere. The present findings implicate that the VOCs and O$_3$ play important roles in the secondary aerosol formation in Taichung City, which needs effective air pollution control strategies for the abatement of emissions to improve air quality. The present ANN model, coupled with the kinetic NO$_3$ and SO$_4^{2-}$ formation model, is applicable for decision makers to investigate the formation mechanism of the secondary aerosols and verify the contribution of each input variable to the predicted targets.

There are some limitations to the present ANN model. Unlike the Random Forest (RF) de-weather model developed in Grange et al. (2018), the present model is incapable of providing time-series meteorological-normalized values for air pollutants variation trend analysis. The RF de-weather model was trained and developed by input variables, including air pollutants measured data and meteorological factors such as T, RH, WD, WS, pressure, and boundary layer height (BLH). After the model was developed, the meteorological factors were removed by users to predict the time-series air pollutant concentrations without the weather factors effect (Grange et al., 2018; Grange et al., 2018; Zhang et al., 2022).
Fig. 7. Contribution of input variables to (a) NO<sub>x</sub>, (b) O<sub>3</sub>, (c) NO<sub>y</sub>, and (d) SO<sub>4</sub><sup>2-</sup> concentrations.
et al., 2020; Zheng et al., 2020). However, the RF-based de-weather model should be operated based on the R-language program, which would be a challenging task for decision makers and government, who are dedicated to building a sustainable environment and city for citizens. The present model is developed using the ANN tool kit in Matlab software, which is easy to operate with a convenient operation interface and clear operation guidelines. The present model could be easily applied by decision makers to realize the contribution of each factor to the variations of air pollutant concentrations. Future works are warranted to develop a user-friendly de-weather air pollutant concentrations prediction model based on the ANN technique for decision makers to assess the effectiveness of air pollution control policies.

5. Conclusions

This study investigated the mean monthly and diurnal variations in the concentrations of air pollution indicators during the level 3 COVID-19 alert in Taichung City. The ANN technique was further applied to develop models for time series NO$_2$, O$_3$, NO$_x$, and SO$_2^-$ concentrations prediction. We found a significant decline in the NO$_2$ concentrations by 20.0% and 25.9% during the Level 3 COVID-19 alert from June to July 2021 as compared to that during the same period in 2019 and 2020, respectively, due to the work from home policy, distance learning, and a reduction in the traffic flow. From June to July 2021, the average O$_3$ and NO$_2$ concentrations increased by 13.2% and 98.2%, respectively, as compared to those during the same period in 2020. The result depicts that the VOC-limited O$_3$ production regime with a reduction of NO$_x$ concentration improved OH radical formations to produce more O$_3$ and secondary aerosols (Lin et al., 2022a).

Based on the statistical results of the diurnal variation in air pollutants, NO$_x$ decreased by 27.9% and 20.2%, respectively, during rush hour at 8:00 relative to 2019 and 2020, due to a reduction in the traffic volume by 19.8% and 34.9% relative to 2019 and 2020, respectively. A significant increase in the NO$_3$ concentration in 2021 by 38.2% and 63.1% at 12:00 was also observed relative to 2019 and 2020, respectively. Thus, during the Level 3 COVID-19 alert, the production of secondary particles was under the VOC-limited regime to enhance NO$_x$ production (Leung et al., 2020; Lin et al., 2022a).

The present models can predict 67.2–80.1% of NO$_x$, O$_3$, NO$_y$, and SO$_2^-$ concentrations. To further improve the accuracy of the models for NO$_x$, O$_3$, NO$_y$, and SO$_2^-$ concentrations prediction, more long-term real-time datasets are recommended, including WIS, OH radicals, NH$_3$, HNO$_3$, and H$_2$SO$_4$ concentrations, solar radiation, and boundary layer height should be taken as the input variables for prediction model development. To conclude this study provides an insight into the complex secondary pollutant formation mechanism to verify the COVID-19 induced changes in anthropogenic emissions. The current easy-to-use and well-developed ANN model can be applied by decision makers to develop the most efficient strategy to improve the local air quality in the future.

Declaration of Competing Interest

None.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.ecoinf.2022.101674.
1. Lung, S.C.C., Wang, W.C.V., Wen, T.Y.J., Liu, C.H., Hu, S.C., 2020. A versatile low-cost sensing device for assessing PM\textsubscript{2.5} spatiotemporal variation and quantifying source contribution. Sci. Total Environ. 716, 137145. https://doi.org/10.1016/j.scitotenv.2020.137145.

2. Marques, M., Domingo, J.L., 2022. Positive association between outdoor air pollution and the incidence and severity of COVID-19. A review of the recent scientific evidences. Environ. Res. 203, 111930. https://doi.org/10.1016/j.envres.2021.111930.

3. Menut, L., Bessagnet, B., Siour, G., Mailler, S., Pennel, R., Cholakian, A., 2020. Impact of lockdown measures to combat COVID-19 on air quality over western Europe. Sci. Total Environ. 741, 140426. https://doi.org/10.1016/j.scitotenv.2020.140426.

4. Naqvi, H.R., Datta, M., Mutreja, G., Siddiqui, M.A., Naqvi, D.F., Naqvi, A.R., 2021. Improved air quality and associated mortalities in India under COVID-19 lockdown. Environ. Pollut. 268, 116591. https://doi.org/10.1016/j.envpol.2020.116591.

5. Park, S., Kim, M., Kim, M., Namgung, H.G., Kim, K.T., Cho, K.H., Kwon, S.B., 2018. Predicting PM\textsubscript{10} concentration in Seoul metropolitan subway stations using artificial neural network (ANN). J. Hazard. Mater. 341, 75–82. https://doi.org/10.1016/j.jhazmat.2017.07.050.

6. Pope III, C.A., Burnett, R.T., Thurston, G.D., Thun, M.J., Calle, E.E., Krewski, D., Gapstur, S.M., Meier, R.M., Brandt, J., 2004. Cardiovascular Mortality and Long-Term Exposure to Particulate Air Pollution: Epidemiological Evidence of General Pathophysiological Pathways of Disease, 109, pp. 71–77. https://doi.org/10.1161/01.HT.

7. Represa, N.S., Della Coca, L.S., Abril, G., García Ferreyra, M.F., Scavuzzo, C.M., 2021. Atmospheric pollutants assessment during the covid-19 lockdown using remote sensing and ground-based measurements in Buenos Aires, Argentina. Aerosol Air Qual. Res. 21, 200486. https://doi.org/10.4209/aaqr.2020.07.0486.

8. Sácedo, D., Onasch, T.B., Dzepina, K., Canagaratna, M.R., Zhang, Q., Huffman, J.A., DeCarlo, P.F., Jayne, J.T., Worsnop, D.R., Kolb, C.E., Robinson, N.L., Zuberi, B., Marr, L.C., Volkamer, R., Molina, L.T., Molina, M.J., Cardenas, B., Bernabé e, R.M., Marquez, C., Gaffney, J.S., Marley, N.A., Laskin, A., Shukhanandand, V., Xie, Y., Brune, W., Leibowitch, E., Shirley, T., Jimenez, J.L., 2006. Characterization of ambient aerosols in Mexico City during the MCM-2003 campaign with aerosol mass spectrometry: results from the CENICA supersite. Atmos. Chem. Phys. 6, 925–946. https://doi.org/10.5194/acp-6-925-2006.

9. Seinfeld, J.H., Pandis, S.N., 2000. Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, first ed. John Wiley & Sons, New York, USA.

10. Sharifi, A., Khavarian-Garmsir, A.R., 2020. The COVID-19 pandemic: impacts on cities and major lessons for urban planning, design, and management. Sci. Total Environ. 749, 142391. https://doi.org/10.1016/j.scitotenv.2020.142391.

11. Sharma, S., Zhang, M., Amhika, Gao, J., Zhang, H., Kota, S.H., 2020. Effect of restricted emissions during COVID-19 on air quality in India. Sci. Total Environ. 728, 138878. https://doi.org/10.1016/j.scitotenv.2020.138878.

12. Shi, Z., Song, C., Liu, B., Lu, G., Xu, J., et al., 2021. Abrupt but smaller than expected changes in surface air quality attributable to COVID-19 lockdowns. Sci. Adv. 7, eabd6696. https://doi.org/10.1126/sciadv.abe6696.

13. Sicard, P., Marco, A.D., Agathokleous, E., Feng, Z., Xu, X., et al., 2020. Amplified ozone pollution in cities during the COVID-19 lockdown. Sci. Total Environ. 735, 135942. https://doi.org/10.1016/j.scitotenv.2020.135942.

14. Soh, P.W., Chang, J.W., Huang, J.W., 2018. Adaptive deep learning-based air quality prediction model using the most relevant spatial-temporal relations. IEEE Access 6, 38186–38199. https://doi.org/10.1109/ACCESS.2018.2849820.

15. Taiwan, E.P.A., 2012. Air Quality Index (AQI). https://reurl.cc/b5a1qM.

16. Tian, X., An, C., Chen, Z., Tian, Z., 2021. Assessing the impact of COVID-19 pandemic on urban transportation and air quality in Canada. Sci. Total Environ. 765, 144270. https://doi.org/10.1016/j.scitotenv.2020.144270.

17. Tsai, D.H., Wang, J.J., Wang, C.H., Chan, C.C., 2008. A study of ground-level ozone pollution, ozone precursors and subtropical meteorological conditions in Central Taiwan. J. Environ. Monit. 10, 109–118. https://doi.org/10.1039/b714479b.

18. Vu, T.V., Shi, Z., Cheng, J., Zhang, Q., He, K., Wang, S., Harrison, R.M., 2019. Assessing the impact of clean air action on air quality trends in Beijing using a machine learning technique. Atmos. Chem. Phys. 19, 11303–11314. https://doi.org/10.5194/acp-19-11303-2019.

19. Xing, J., Zheng, S., Ding, D., Kelly, J.T., Wang, S., Li, S., Qin, T., Ma, M., Dong, Z., Kang, C., Zhu, Y., Zheng, H., Ren, L., Liu, T.Y., Hao, J., 2020. Deep learning for prediction of the air quality response to emission changes. Environ. Sci. Technol. 54, 8589–8600. https://doi.org/10.1021/acs.est.0c02923.

20. Yen, M.C., Chen, T.C., 2000. Seasonal variation of the rainfall over Taiwan. Int. J. Climatol. 20, 803–809. https://doi.org/10.1002/1097-0088(20000615)20:7<803::AID-JOC525>3.0.CO;2-4.

21. Zangari, S., Hill, D.T., Charette, A.T., Miroswsky, J.E., 2020. Air quality changes in New York City during the COVID-19 pandemic. Sci. Total Environ. 742, 140496. https://doi.org/10.1016/j.scitotenv.2020.140496.

22. Zhang, Y., Yu, T.V., Sun, J., He, J., Shen, X., Lin, W., Zhang, X., Zhong, J., Gao, W., Wang, Y., Fu, T.M., Ma, Y., Li, W., Shi, Z., 2020. Significant changes in chemistry of fine particles in wintertime Beijing from 2007 to 2017: impact of clean air actions. Environ. Sci. Technol. 54, 1344–1352. https://doi.org/10.1021/acs.est.9b04678.

23. Zhang, Y., Tang, A., Wang, C., Ma, X., Li, Y., et al., 2022. PM\textsubscript{2.5} and water-soluble inorganic ion concentrations decreased faster in urban than rural areas in China. J. Geophys. Res. 122, 83–91. https://doi.org/10.1029/2021JD036716.

24. Zhao, J., Deng, F., Cai, Y., Chen, J., 2019. Long short-term memory - fully connected (LSTM-FC) neural network for PM\textsubscript{2.5} concentration prediction. Chemosphere 220, 486–492. https://doi.org/10.1016/j.chemosphere.2018.12.128.

25. Zheng, H., Kong, S., Chen, N., Yan, Y., Liu, D., et al., 2020. Significant changes in the chemical compositions and sources of PM\textsubscript{2.5} in Wuhan since the city lockdown as COVID-19. Sci. Total Environ. 739, 140000. https://doi.org/10.1016/j.scitotenv.2020.140000.

26. Zwack, L.M., Faciorek, C.J., Spengler, J.D., et al., 2011a. Characterizing local traffic contributions to particulate air pollution in street canyons using mobile monitoring techniques. Atmos. Environ. 45 (15), 2507–2514. https://doi.org/10.1016/j.atmosenv.2011.02.035.

27. Zwack, L.M., Faciorek, C.J., Spengler, J.D., et al., 2011b. Modeling spatial patterns of traffic-related air pollutants in complex urban terrain. Environ. Health Perspect. 119 (6), 852–859. https://doi.org/10.1289/ehp.1002519.