Chemosphere 314 (2023) 137638

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Assessing the ambient air quality patterns associated to the COVID-19 outbreak in the Yangtze River Delta: A random forest approach

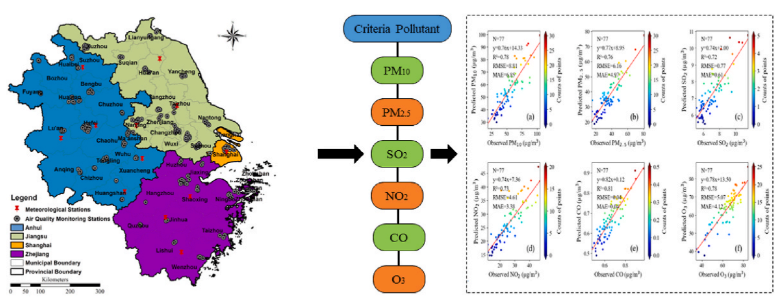
Ahmad Hasnain [a,b,c], Yehua Sheng [a,b,c,*], Muhammad Zaffar Hashmi [d,**], Uzair Aslam Bhatti [e], Zulkifl Ahmed [f], Yong Zha [a,b,c]

[a] *Key Laboratory of Virtual Geographic Environment, Ministry of Education, Nanjing Normal University, Nanjing 210023, China*
[b] *School of Geography, Nanjing Normal University, Nanjing 210023, China*
[c] *Jiangsu Center for Collaborative Innovation in Geographical Information, Resource Development and Application, Nanjing 210023, China*
[d] *Department of Chemistry, COMSATS University Islamabad, Pakistan*
[e] *School of Information and Communication Engineering, Hainan University, Haikou, China*
[f] *Department of Civil Technology, Mir Chakar Khan Rind University of Technology, DG Khan 32200, Pakistan*

HIGHLIGHTS

- Our random forest (RF) model showed high performance in the prediction of air pollutants in the Yangtze River Delta.
- Significant reductions were recorded in air pollutants during the COVID-19 phase.
- The Yangtze River Delta experienced rising trends in air pollutant concentrations in 2021–22.
- Meteorological parameters indicated mixed behavior during the study period.

GRAPHICAL ABSTRACT



ARTICLE INFO

Handling Editor: Volker Matthias

Keywords:
Air quality
Random forest model
COVID-19
Air pollution
Yangtze river delta

ABSTRACT

The novel coronavirus (COVID-19), first identified at the end of December 2019, has significant impacts on all aspects of human society. In this study, we aimed to assess the ambient air quality patterns associated to the COVID-19 outbreak in the Yangtze River Delta (YRD) region using a random forest (RF) model. To estimate the accuracy of the model, the cross-validation (CV), determination coefficient R^2, root mean squared error (RMSE) and mean absolute error (MAE) were used. The results demonstrate that the RF model achieved the best performance in the prediction of PM_{10} ($R^2 = 0.78$, RMSE = 8.81 μg/m^3), $PM_{2.5}$ ($R^2 = 0.76$, RMSE = 6.16 μg/m^3), SO_2 ($R^2 = 0.76$, RMSE = 0.70 μg/m^3), NO_2 ($R^2 = 0.75$, RMSE = 4.25 μg/m^3), CO ($R^2 = 0.81$, RMSE = 0.4 μg/m^3) and O_3 ($R^2 = 0.79$, RMSE = 6.24 μg/m^3) concentrations in the YRD region. Compared with the prior two years (2018–19), significant reductions were recorded in air pollutants, such as SO_2 (−36.37%), followed by PM_{10} (−33.95%), $PM_{2.5}$ (−32.86%), NO_2 (−32.65%) and CO (−20.48%), while an increase in O_3 was observed (6.70%) during the COVID-19 period (first phase). Moreover, the YRD experienced rising trends in the concentrations of PM_{10}, $PM_{2.5}$, NO_2 and CO, while SO_2 and O_3 levels decreased in 2021–22 (second phase). These findings provide credible outcomes and encourage the efforts to mitigate air pollution problems in the future.

* Corresponding author. Key Laboratory of Virtual Geographic Environment, Ministry of Education, Nanjing Normal University, Nanjing 210023, China.
** Corresponding author. Department of Chemistry, COMSATS University Islamabad, Pakistan.
E-mail addresses: shengyehua@njnu.edu.cn (Y. Sheng), zaffar.hashmi@comsats.edu.pk (M.Z. Hashmi).

https://doi.org/10.1016/j.chemosphere.2022.137638
Received 15 October 2022; Received in revised form 8 December 2022; Accepted 21 December 2022
Available online 21 December 2022
0045-6535/

1. Introduction

The novel coronavirus disease 2019 (COVID-19) (later named as SARS-CoV-2), which was first reported at the end of December 2019 in Wuhan, China, has significant impacts on all aspects of human society (Lu et al., 2020). Due to its high transmission rate between humans and linked to the mortality, the World Health Organization (WHO) called it the coronavirus disease 2019 (COVID-19) (WHO, 2020a). To combat its large-scale spread and contagious effects, many countries around the world, implemented strict prevention and control policies such as community limitations, self-quarantine, social distancing, restrictions on traffic and lockdowns for public health (He et al., 2020; Hasnain et al., 2021). Being one of the largest populated countries in the World, the Chinese authorities executed a series of strict prevention and control measures on January 23, 2020 in the capital city of China's Hubei Province, Wuhan and were subsequently followed by other cities and provinces (Le et al., 2020; Zhao et al., 2020a).

The strict and stringent actions during the COVID-19 period positively impacted the environment due to reduced anthropogenic activities. Previously, many studies have reported a sudden decrease in air quality levels during the COVID-19 control period in different areas and regions across the globe, such as in China (He et al., 2020; Le et al., 2020; Zhao et al., 2020a; Hasnain et al., 2021; Hua et al., 2021; Wang et al., 2021a), Turkey (Ghasempour et al., 2021; Orak and Ozdemir, 2021), India (Singh et al., 2020; Mor et al., 2021; Pal et al., 2021a, 2021b), United Arab Emirates (Teixidó et al., 2021), Spain (Tobías et al., 2020; Briz-Redón et al., 2021), Ireland (Spohn et al., 2022), USA (Bauwens et al., 2020; Berman and Ebisu, 2020), etc. Hua et al. (2021) studied the impact of the COVID-19 lockdown on NO_2 and $PM_{2.5}$ using a Generalized Additive Models (GAM) in Beijing, China. Singh et al. (2020) estimated the temporal and diurnal changes of the air pollutants and found a considerable decline in the concentrations of PM_{10}, $PM_{2.5}$, SO_2, NO_2 and CO using real-time data in India. Briz-Redón et al. (2021) investigated the short-term impact of the COVID-19 lockdown on air pollution in Spain. The study reported that the lockdown had significant impact on reducing the levels of different air pollutants. Another study documented by Sulaymon et al. (2021) revealed that the concentration levels of PM_{10}, $PM_{2.5}$, SO_2, NO_2 and CO decreased, while O_3 levels increased during the lockdown period in Wuhan, China.

Liu et al. (2022) investigated the $PM_{2.5}$ level using a machine learning approach in Hubei Province, China. The study reported that changes in anthropogenic emissions have reduced the concentrations of $PM_{2.5}$ in February and March 2020 by 33.3% compared with the last year, 2019. Huang et al. (2021) studied the variation of air pollutant concentrations and its formation mechanism during the period of COVID-19 in Wuhan. The authors of this found a significant increase in O_3 levels (43.9%), while the concentration of $PM_{2.5}$ decreased (31.7%).

In developing countries air pollution is one of key issues, which poses a major threat to human health (He et al., 2017). The World Air Quality Report indicated that many Asian countries such as India, Pakistan, Bangladesh and China experienced high levels of air pollution in recent years (AirVisual, 2019). China is one of the largest developing countries in the world, with a large population, transportation and industries. In the last 3 decades, many areas and regions of the country have experienced high air pollution levels (Zhao et al., 2020b). In the last few years, due to strict restrictions on transportation, heating activities and industrial emissions, a slight decline in air pollution was observed in China, but significant measures are still required to protect the environment at a significant level (Wu et al., 2020). After the first confirmed case of the COVID-19 pandemic and to retard its rapid spread, the Chinese authorities imposed stringent prevention and control measures. No doubt, the country effectively controlled to this infectious disease due to these appropriate measures, but as of July 18, 2022, China reported 237 new confirmed cases of the COVID-19, which shows that the country is still struggling with the COVID-19 outbreak (NHC).

The present study analyzed the six air pollutant parameters (PM_{10}, $PM_{2.5}$, SO_2, NO_2, CO and O_3) along with meteorological variables (air temperature, precipitation, relative humidity and wind speed) using daily average data in the YRD region. As discussed in the above lines, previously numerous studies have been documented, associated with the COVID-19 outbreak. Most of these studies have reported that the concentrations of different air pollutants were decreased during the lockdown period in different areas and regions across the globe, while some studies have discussed only a few air pollutant parameters to evaluate the effects of the COVID-19 lockdown or air pollution. However, compared with the prior studies, the current work presents a deeper analysis with a new paradigm, which reports the COVID-19 period (2020) and compared the results with the previous 2 years (2018–19) and the following two years (2021–22) to discover new findings using a machine learning model. In this study, we aimed to (1) assess the ambient air quality patterns in the YRD region using a random forest model; (2) explore the concentrations of different air pollutants during the same dates in the previous two years (2018–19); (3) to examine the status of air quality during the same period in the following two years (2021–22); (4) to evaluate the relationship between ambient air pollutants and meteorological variables during the study period. These findings provide a strong reference for the scientific community, policymakers and local authorities to mitigate air pollution problems in the future.

2. Materials and methods

2.1. Study area

The Yangtze River Delta, with its ancient history, is located in the north-central subtropical zone and the economic hub of China (Fig. 1). According to the "Development Plan of the Yangtze River Delta City Cluster" there are 26 cities, including 8 cities of Anhui, 9 cities of Jiangsu, 8 cities of Zhejiang, and Shanghai, which belongs to the Yangtze River Delta city cluster, while the core area has 16 cities. The region accounts for 11.7% of the national population and 2.2% of the national land. According to statistics, the Yangtze River Delta contributing about 21% gross domestic product (GDP) of the country. The Yangtze River Delta is one of the most important and leading economic development areas in China. In recent years, the region has been experienced with severe air pollution due to rapid development in industrialization and urbanization.

2.2. Air quality data and periods of study

Data on the daily average concentration for the six air pollutant parameters, including PM_{10} (particulate matter with the diameters of $\leq$10 μm), $PM_{2.5}$ (particulate matter with the diameters of $\leq$2.5 μm), SO_2 (sulphur dioxide), NO_2 (nitrogen dioxide), CO (carbon monoxide) and O_3 (ozone), were retrieved from the China Environmental Monitoring Station through 196 monitoring stations (CNEMC, 2019). These monitoring stations are distributed over 13 cities of Jiangsu Province, 16 cities of Anhui Province, 11 cities of Zhejiang Province and Shanghai. The data on the selected air pollutants were collected from 2018 to 2022. The COVID-19 period (23 January 2020–8 April 2020) was researched over the course of five years with the purpose of observing the change and evaluating the patterns of ambient air quality in the Yangtze River Delta region. To acquire more accurate results from the study, the missing values were replaced with the average values and the daily average of each pollutant was used.

2.3. Meteorological data

Meteorological data were downloaded from the NASA meteorological data service (https://power.larc.nasa.gov), including air temperature (T), precipitation (PP), relative humidity (RH) and wind speed (WS) through 12 meteorological monitoring stations (Fig. 1).

2.4. Random forest model and validation

The random forest is a new machine learning model, which contains multiple classification and regression tree (CART) integrations (Brokamp et al., 2018). CART has three distinct features. First, the model creates numerous trees, a bootstrap sample generates these trees in the original dataset. All raw data are used to develop only one tree in CART. Second, the model performed the segmentation of tree nodes each time based on an optimum variant, while to segment the tree nodes CART picks the optimum variant among all forecasters. Finally, the trees in the model are fully grown. This helps the model not easy to overfit (Liu et al., 2018). There are three training parameters which can be defined in the random forest model: n_estimators, the trees number in the forest-based on a bootstrap observations sample; max_features, the features number to be considered for the superlative split ("auto" is the default setting: then max_features = n_features) and min_samples_lea, the number of minimum samples which required to be at a leaf node (one is the default value). The two key parameters (n_estimators and max_features) in predicting the concentrations of air pollutants were optimized and estimated based on the out-of-bag (OOB) calibration error rate. The entire model-fitting dataset was randomly split into a training set and test set, where 90% of the data comprised the initial training set and the test set comprised 10% of the data. Cross-validation was used to check to overfit of the model.

2.5. Statistical analysis

Determination coefficient R^2, root mean squared error (RMSE) and mean absolute error (MAE) were used to evaluate the model's performance. The larger the R^2, the smaller the RMSE and MAE demonstrating that the prediction accuracy of the model is higher. These matrices used the following formulas:

$$R^2 = \sum_{i=1}^{n} (P_i - \overline{M})^2 \Big/ \sum_{i=1}^{n} (M_i - \overline{M})^2 \tag{1}$$

$$RMSE = \sqrt{\frac{1}{n}\sum_{i=1}^{n} |M_i - P_i|^2} \tag{2}$$

$$MAE = \frac{1}{n}\sum_{i=1}^{n} |M_i - P_i| \tag{3}$$

where M and P are the measured and predicted values and n denotes the number of samples in the validation set.

3. Results and discussion

3.1. Model performance

Our random forest (RF) model showed the best performance in the prediction of air pollutant concentrations in the Yangtze River Delta region. The results indicate that RF predicted PM_{10} and $PM_{2.5}$ with R^2 values of 0.78 and 0.76, RMSE values of 8.81 and 6.16 μg/m^3 and MAE values of 6.85 and 4.97 μg/m^3, respectively, during the period of COVID-19 (Fig. 2). Among other pollutants, the predicted R^2 values by RF for SO_2, NO_2, CO and O_3 were 0.72, 0.73, 0.81 and 0.78, RMSE values were 0.77, 4.61, 0.04 and 5.07 μg/m^3 and MAE values were 0.61, 3.38, 0.04 and 4.12 μg/m^3, respectively, during the corresponding period (Fig. 2). The performance of the model was also estimated at the provincial and municipal levels for all the air pollutants (Figs. 4 and 5). From Figs. 4 and 5, it is evident that the actual and predicted values significantly fitted during the period of COVID-19 (first phase-2020),

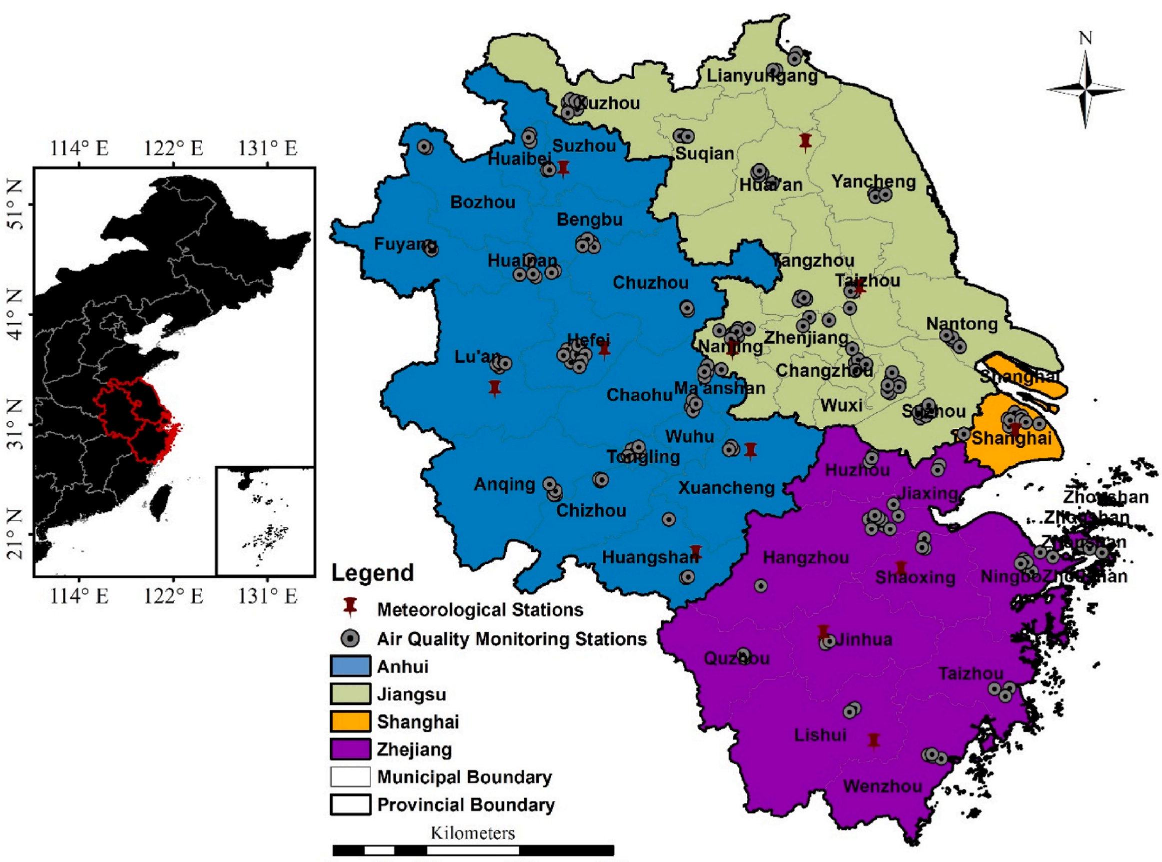


Fig. 1. Study area and distribution of meteorological and air quality monitoring stations.

and the same period in 2022 (second phase-2022) in Jiangsu, Anhui, Zhejiang and Shanghai. Compared with many studies, our model showed high performance covering a large area. For instance, Lu et al. (2021) compared three models in the prediction of $PM_{2.5}$ concentration and found by RF, the predicted value of R^2 was 0.69, followed by support vector regression ($R^2 = 0.57$) and artificial neural network ($R^2 = 0.53$). Huang et al. (2017) predicted the concentration of O_3 using LUR models in Nanjing, China and the value of R^2 was 0.65. The concentrations of PM were predicted based on generalized additive models and a simplified FLEXPART model combined with the Bayesian-RAT method in different areas of China (Guo et al., 2020; Zeng et al., 2020). However, our model provides superior performance than those of other comparable studies.

Moreover, RF predicts PM_{10} and $PM_{2.5}$ concentrations with $R^2 = 0.76$ and 0.74, RMSE = 13.65 and 7.73 μg/m^3 and MAE = 11.33 and 6.18 μg/m^3, respectively during the second phase (Figs. 3 and 5). These findings demonstrate that RF provides better performance during the first phase compared with the second phase. For example, during the first phase, the cross-validation R^2 values of RF for the PM_{10} and $PM_{2.5}$ prediction are 0.78 and 0.76, which are greater than that of values during the second phase. Similarly, by RF the values of RMSE and MAE for PM_{10} and $PM_{2.5}$ are better during the first phase compared to the second phase in the YRD region. Among the selected pollutants, RF predicts SO_2 concentration with $R^2 = 0.76$, RMSE = 0.70 μg/m^3 and MAE = 0.55 μg/m^3, NO_2 concentration with $R^2 = 0.75$, RMSE = 4.25 μg/m^3 and MAE = 3.58 μg/m^3, CO concentration with $R^2 = 0.80$, RMSE = 0.05 μg/m^3 and MAE = 0.04 μg/m^3 and O_3 concentration with $R^2 = 0.79$, RMSE = 6.24 μg/m^3 and MAE = 5.17 μg/m^3 during the second phase (Figs. 3 and 5). The model showed high performance in the prediction of SO_2 concentration during the second phase, while the R^2 and RMSE values of RF were better for NO_2 prediction during the second phase but the MAE value was poorer during the first phase than that of the second phase. RF exhibited similar results in the prediction of CO during the both phases with a small difference, while for O_3, the value of R^2 was higher during the second phase, but the RMSE and MAE values were relatively poorer during this phase compared to the first phase. In summary, RF provides superior performance in the prediction of air pollutant concentrations during the two phases in the YRD region.

3.2. *Spatial concentration pattern of air pollutants in Jiangsu Province*

The COVID-19 outbreak in China and worldwide, has provided a great opportunity to study its impacts on air pollution by comparing the concentrations of air pollutants. In this study, the concentration levels of different air pollutants during the COVID-19 outbreak (2020) were compared with the previous two years (2018–19) and the following two years (2021–22) in the Yangtze River Delta (YRD) region. For a deeper analysis, we further discussed the concentration pattern of air pollutants at the provincial and municipal levels. The comparisons of the years 2020 vs. 2018–19 indicate that the air pollutant concentrations decreased significantly in Jiangsu Province, such as PM_{10}, $PM_{2.5}$, SO_2, NO_2 and CO decreased by an average of −35.81%, −32.29%, −40.95%, −34.00% and −20.00% respectively, while O_3 was the only pollutant, which exhibited an increasing trend (7.67%) (Table S1, Fig. 6 and 7, S1-4). The reduction in different air pollutants during the first phase was due to the reduction in all industrial and construction activities, decreased human mobility and the stoppage of traffic (Zhao et al., 2020a; Wang et al., 2021b). The highest drop was observed in the

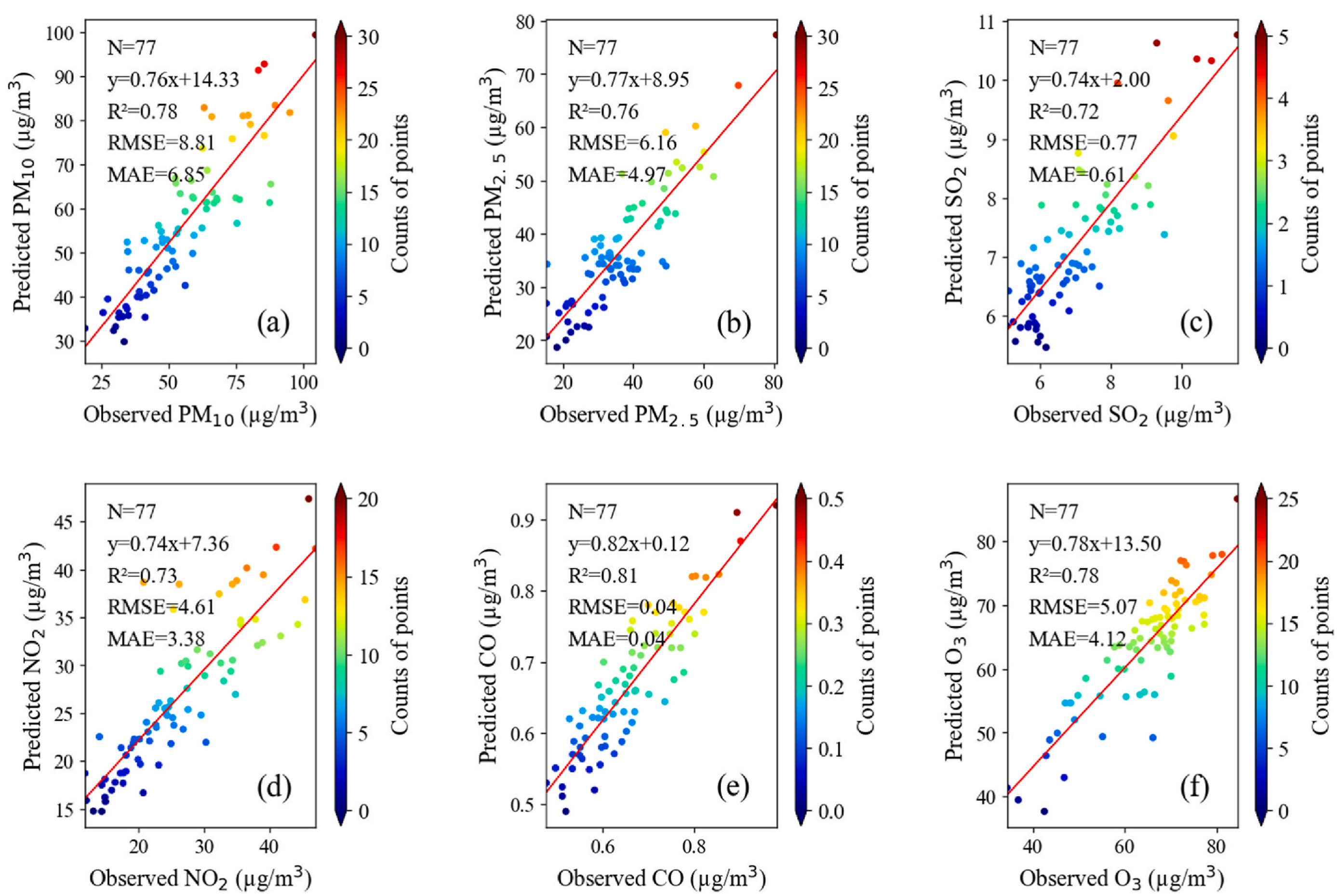


Fig. 2. Validation between actual and predicted air pollutants by random forest model in COVID-19 (first phase-2020) period in the Yangtze River Delta region (a: PM_{10}, b: $PM_{2.5}$, c: SO_2, d: NO_2, e: CO and f: O_3).

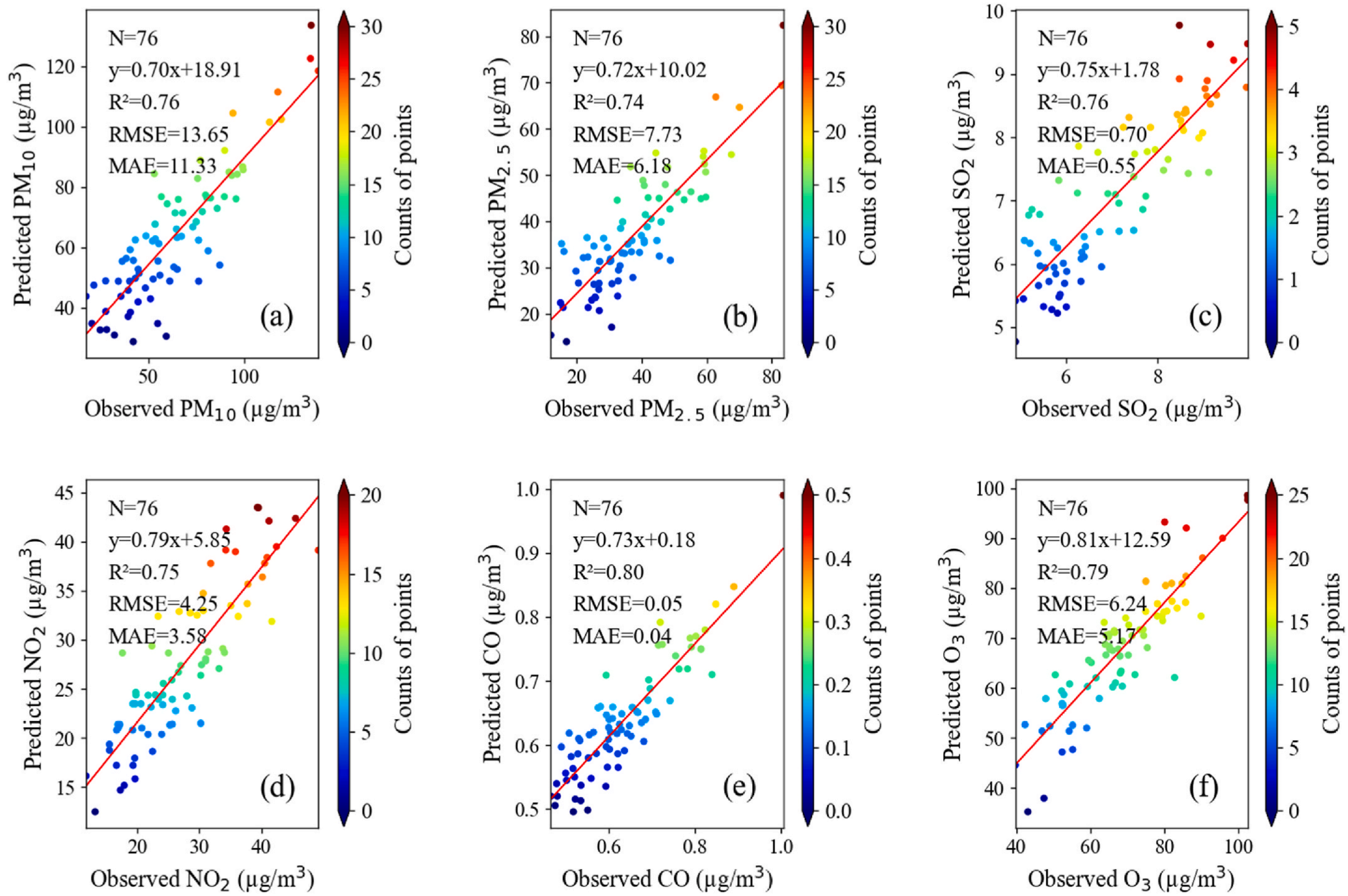


Fig. 3. Validation between actual and predicted air pollutants by random forest model in 2022 (second phase-2022) in the Yangtze River Delta region (a: PM_{10}, b: $PM_{2.5}$, c: SO_2, d: NO_2, e: CO and f: O_3).

concentration of SO_2, while particulate matter (PM_{10} and $PM_{2.5}$) and NO_2 showed almost similar tendencies with slight differences. The fall in the concentration of CO was lower compared to other pollutant parameters during this window of time in Jiangsu Province. In contrast to other pollutants, we found an increase in O_3 levels. Past studies also reported similar findings for Jiangsu Province. For instance, Wu et al. (2022) found a substantial reduction in $PM_{2.5}$, PM_{10}, CO, SO_2 and NO_2, while O_3 levels increased during the first phase compared to the prior years in Jiangsu. Another study reported by Hasnain et al. (2021) indicated that during the COVID-19 period, an abrupt fall in air pollutant concentrations was observed in Nanjing, the capital of Jiangsu Province (see Fig. 7).

Moreover, all the air pollutants presented rising trends during the second phase in Jiangsu. The concentrations of PM_{10} and $PM_{2.5}$ were increased by 22.36% and 5.94% respectively (Table S1,Figs. 6 and 7, S1-4). Among other pollutants, a slight increase in SO_2 was found, to be 0.07%, while NO_2, CO and O_3 levels increased by an average of 9.02%, 2.21% and 0.13% respectively (Table S1). The results suggest that the increase in different air pollutants during the second phase was due to the enhancement in anthropogenic sources such as biofuel burning, power plants, traffic and industrial emissions in Jiangsu (Yao et al., 2021; Hasnain et al., 2021; Wu et al., 2022). The rise in the concentration of PM_{10} was significantly greater than other pollutant concentrations. NO_2, a primary pollutant released from vehicle emissions, presented the second highest increasing value, while a slight rise in SO_2 and O_3 was observed during the second phase. In summary, the comparison of the years indicate that the air quality significantly improved during the first phase due to the stringent restrictions and shutdown policies, while an increase in air quality can be seen during the second phase in Jiangsu Province, which is worthy of attention.

3.3. Spatial concentration pattern of air pollutants in Anhui Province

Similar findings were observed for Anhui Province with small differences (Table S1, Figs. 6 and 7, S1-4). The levels of different air pollutants remarkably decrease during the first phase except for O_3. The results demonstrate that the reductions in PM_{10} and $PM_{2.5}$ were −31.25% and −31.37% respectively, during the first phase (Table S1). It should be noted that the decreasing spell of PM_{10} and $PM_{2.5}$ was almost similar during the corresponding period. The drops in SO_2, NO_2 and CO were −32.96%, −31.27% and −18.56% respectively, while a slight rise in O_3 levels was observed (1.74%) during the first phase in Anhui (Table S1, Figs. 6 and 7, S1-4). These findings agreed well with the prior studies documented by Hasnain et al. (2021) and Bhatti et al. (2022). Compared with the air quality status of Jiangsu Province, the reductions in the concentrations of different air pollutants in Anhui were relatively lower during the first phase (Table S1). The highest fall was recorded for SO_2, followed by $PM_{2.5}$, NO_2, PM_{10} and CO while O_3 exhibited a slight increase during the first phase in Anhui.

Moreover, the second phase shows comparatively a different pattern of air quality in Anhui Province compared with Jiangsu. An obvious escalation in PM_{10} concentration was observed, to be 21.77%, while the rise in $PM_{2.5}$ was 0.78% during the second phase in Anhui (Table S1, Figs. 6 and 7, S1-4). A large difference in the rise of PM_{10} and $PM_{2.5}$ was found. The increment in the movement of vehicles, construction and industrial activities can be responsible for this increase in PM_{10} concentration (Singh et al., 2020; Bilal et al., 2021). Among the selected pollutants, SO_2, CO and O_3 showed differing trends, which were

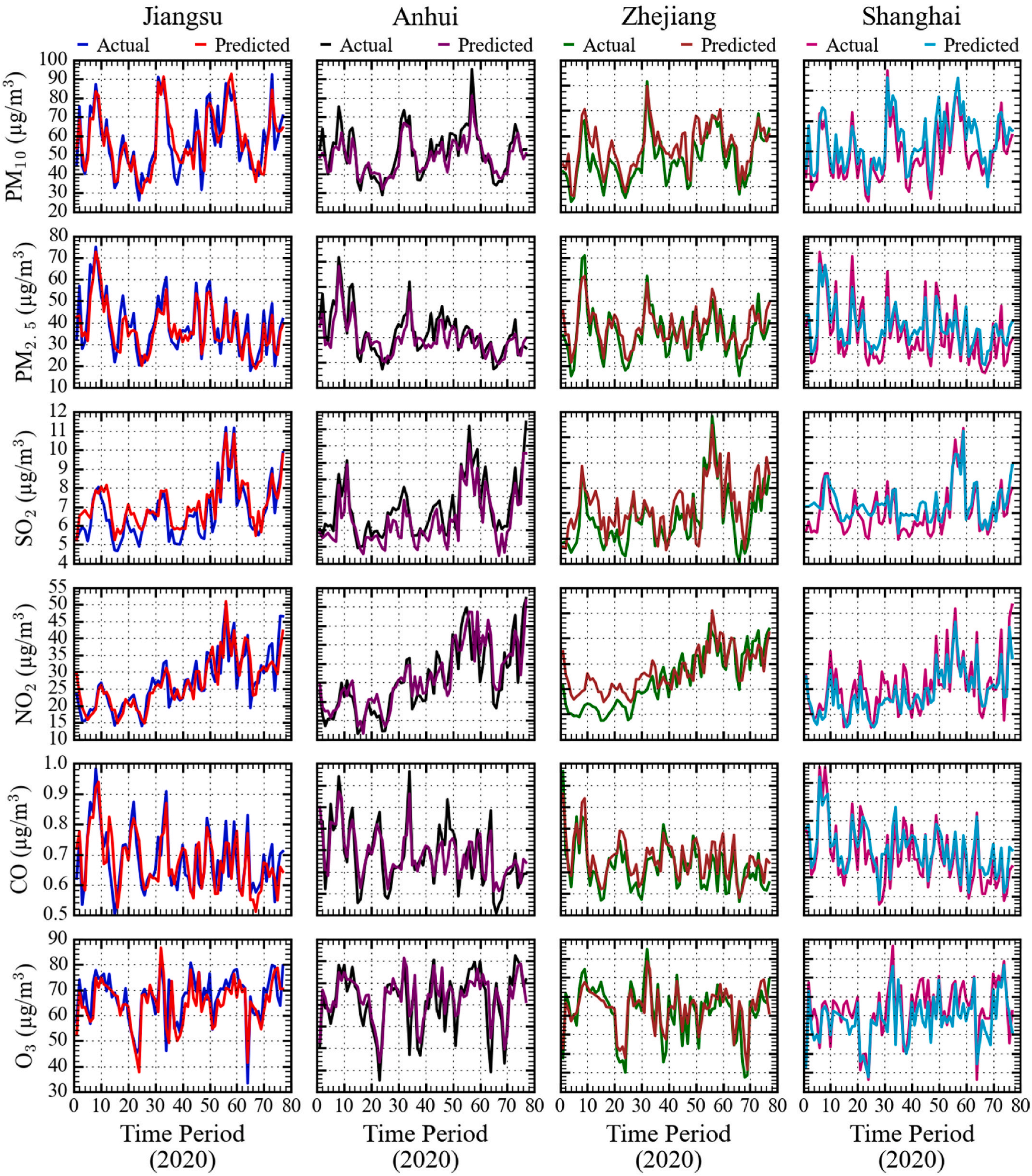


Fig. 4. Actual and predicted concentrations of different air pollutants during the COVID-19 period (first phase-2020) in Jiangsu, Anhui, Zhejiang and Shanghai.

decreased by −6.57%, −2.94 and −1.05% respectively, during the second phase in Anhui (Table S1). Compared with other pollutants, SO_2 exhibited a significant drop in Anhui, having the highest decreasing value in reduction scenario. The major sources of SO_2 are fuel burning, power plants, transport and industry (Huang et al., 2017). Hence, it can be inferred that the drop in SO_2 could be attributed to reduced and controlled thermal, transportation and industrial sectors. The levels of NO_2 increased by 7.78% in Anhui (Table S1). A slight difference can be seen in the increase of NO_2 in Anhui and Jiangsu. Overall, a sudden reduction in air quality was found during the first phase, while the air pollutants showed mixed behavior during the second phase in Anhui Province.

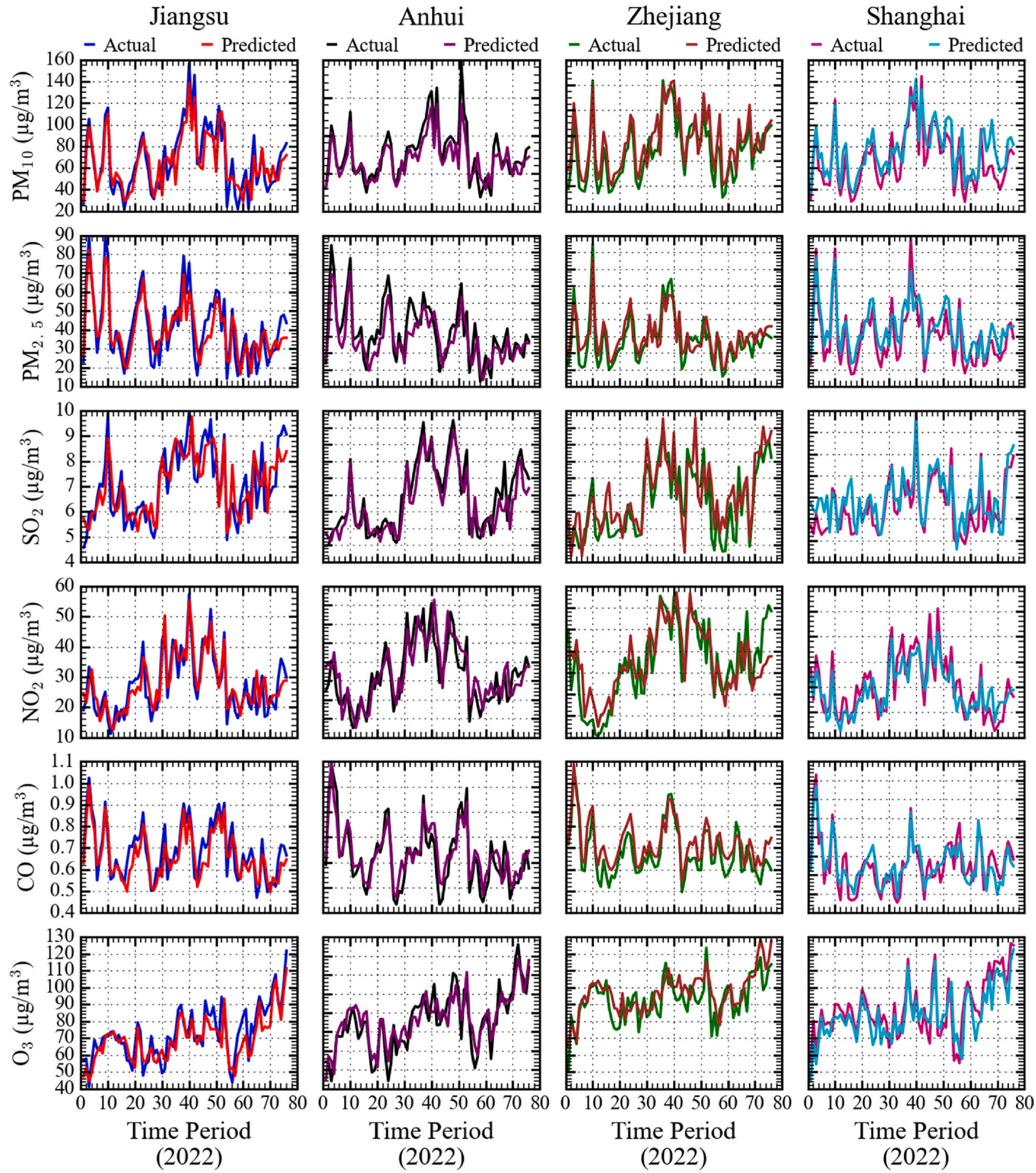


Fig. 5. Actual and predicted concentrations of different air pollutants in 2022 (second phase-2022) in Jiangsu, Anhui, Zhejiang and Shanghai.

3.4. Spatial concentration pattern of air pollutants in Zhejiang Province

A substantial decline in the levels of different air pollutants was experienced but O_3 showed similar performance during the first phase in Zhejiang Province (Table S1). Air pollutants exhibited a drastic drop such as $PM_{2.5}$ (−37.59%), followed by PM_{10} (−34.24%), NO_2 (−32.51%), SO_2 (−31.56%) and CO (−22.01%), while an increase in O_3 levels was experienced, to be 13.05% during the first phase in Zhejiang (Table S1, Figs. 6 and 7, S1-4). O_3, a secondary pollutant, is formed in the presence of sunlight and its precursors such as volatile organic compounds (VOCs) and nitrogen oxides (NOx), and its concentration varies according to physical/chemical removal, photochemistry and

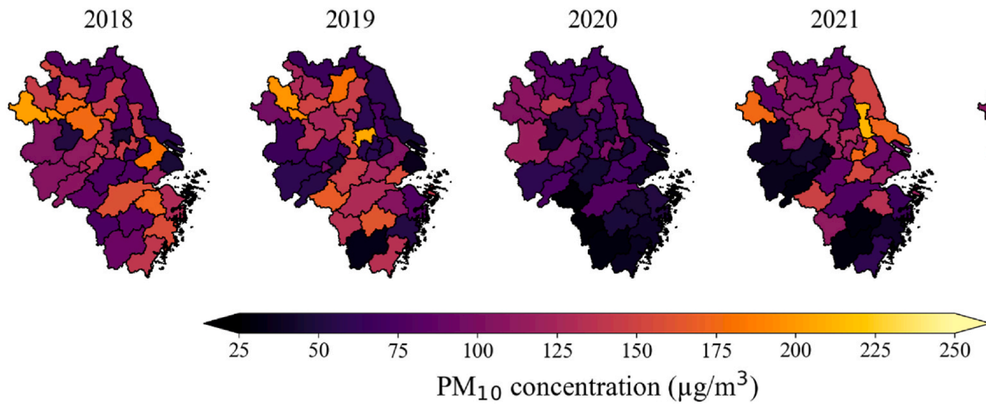


Fig. 6. Spatial distribution of PM_{10} concentrations in the Yangtze River Delta.

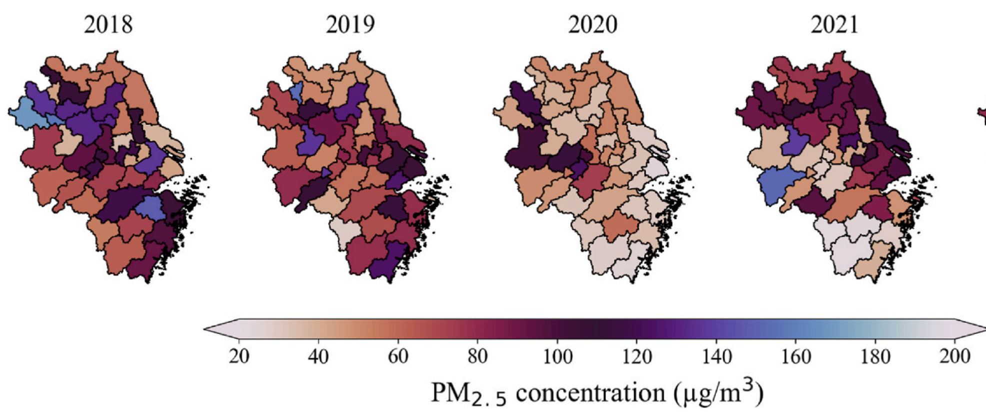


Fig. 7. Spatial distribution of $PM_{2.5}$ concentrations in the Yangtze River Delta.

transport over regional, and global scales (Reddy et al., 2012; Lal et al., 2000). The comparison analysis indicates that the concentration of O_3 increased significantly during the first phase in Zhejiang Province than Jiangsu and Anhui. The reduction ratio for $PM_{2.5}$ concentration was greater than other pollutants, while PM_{10}, SO_2 and NO_2 indicated relatively similar tendencies with some fluctuations in Zhejiang. Past studies also reported that except for O_3, the concentrations of different air pollutants were decreased during the COVID-19 control period compared with the prior years in different areas and regions across China (Chen et al., 2020; Zhao et al., 2020a; Hasnain et al., 2021; Bhatti et al., 2022).

The results demonstrate that compared with the first phase, a new pattern of air quality was observed in Zhejiang (Table S1). The concentration of PM_{10} increased remarkably by up to 19.74%, while $PM_{2.5}$ levels increased by 9.44% during the second phase (Table S1). Among other pollutants, the increasing values for SO_2 and NO_2 were 6.29% and 22.83%, respectively, while the levels of CO and O_3 reduced by an average of −2.42% and −5.71%, respectively, during the second phase in Zhejiang. It should be noted that the rising spell of NO_2 was greater than other pollutants in Zhejiang. Compared with Jiangsu and Anhui, an increase in PM_{10} concentrations was comparatively lower during the second phase. An almost similar trend was observed for CO with a slight difference in Anhui and Zhejiang. CO is a colorless gas emitted from biofuel burning, waste burning, forest fires and vehicular emissions (Singh et al., 2020; Hasnain et al., 2022). O_3 presented an opposite pattern, it decreased during the second phase, while an increase in O_3 was found during the first phase in Zhejiang. In general, except for O_3, the province observed a significant drop in air pollutant concentrations during the first phase, while except for CO and O_3, a remarkable rise in air pollutants was experienced during the second phase particularly PM_{10} and NO_2.

3.5. *Spatial concentration pattern of air pollutants in Shanghai*

Shanghai, the economic hub of the country, has been faced with severe and extreme air pollution in recent years, due to rapid economic development (Wang et al., 2021b). An almost similar pattern of air quality was noted for Shanghai during the first phase. Large reductions were found in different air pollutants, such as SO_2 (−38.53%), followed by PM_{10} (−37.61%), NO_2 (−31.96%), $PM_{2.5}$ (−31.54%) and CO (−17.57%), while O_3 increased (6.04%) during the first phase in Shanghai (Table S1, Figs. 6 and 7, S1-4). Li et al. (2020) reported that the limited vehicular, industrial and construction activities, among others, leading to a drastic drop and notable improvement in air quality during the COVID-19 period in the YRD region. Chu et al. (2021) found that the decrease in traffic, power plants, steel and iron production resulted in a marked decline in the concentrations of $PM_{2.5}$, SO_2, NO_2 and CO during the COVID-19 periods in China. Our results indicate that compared with other pollutants, SO_2 and PM_{10} reduced sharply, while $PM_{2.5}$ and NO_2 presented relatively similar behavior in reduction scenario. O_3 showed a constantly increasing trend, while it also exhibited similar tendencies in Jiangsu, Anhui and Zhejiang.

Compared with the previous three regions, a prominent increase in PM_{10} was found, to be 28.35% in Shanghai. (Table S1). PM_{10} contains polycyclic aromatic hydrocarbons and transition metals (zinc, copper, manganese) and can penetrate the lower air ways in the shape of thoracic particles with an aerodynamic diameter of less than 10 μm (Pal et al., 2021b; Hasnain et al., 2022). PM_{10} causes numerous lung diseases such as cardiovascular and asthma (Mandal et al., 2022). Our results reveal that the increment ratio for PM_{10} was significantly greater than other pollutants during the second phase in all the regions. $PM_{2.5}$, a primary pollutant, showed a slight increase, by approximately 0.63%, while NO_2 and CO increased by 12.51% and 6.56% respectively, during the second phase (Table S1). Among other pollutants, the reductions in the concentrations of SO_2 and O_3 were −7.69% and −4.53%, respectively (Table S1). The comparison analysis indicate that Anhui and Shanghai were found to experience a decline in SO_2 levels during the second phase. Overall, except for SO_2 and O_3, the concentration levels of air pollutants increased during the second phase in Shanghai.

3.6. Changes in pollutant concentrations in the Yangtze River Delta region

Here, we attempt to further examine and combine the entire YRD region and find out the changes in pollutant concentrations during the two phases. From an air pollution perspective, the YRD region has attracted wide interest from researchers and scholars due to its significant importance in the country and rapid economic development. The statistical analyses for the six air pollutants during the two phases are summarized in Table S2. The results indicate that the concentrations of different air pollutants reduced sharply, while O_3 revealed similar performance during the first phase in the YRD region (Table S2). The reductions in the concentrations of PM_{10}, $PM_{2.5}$, NO_2, CO and SO_2 were −33.95%, −32.86%, −32.65%, −20.48% and −36.37% respectively, while the concentration of O_3 increased by approximately 6.70% (Table S2, Figs. 6 and 7, S1-4). A study addressed by Wang et al. (2021b) revealed a significant drop in air quality during the COVID-19 period in the YRD region due to strict control measures and reduced vehicular, construction and industrial activities. In our findings, the highest drop was found for SO_2, while PM_{10}, $PM_{2.5}$ and NO_2 presented similar trends with slight differences. CO decline was relatively smaller than others pollutants, while O_3 exhibited a rising trend. These findings are very close to Yao et al. (2021) study, which indicated a significant reduction and improvement in air quality during the COVID-19 pandemic in the YRD region.

Moreover, a sharp increase in PM_{10} concentration was found, to be 21.91%, while the second highest increasing value was observed for NO_2 (11.60%) during the second phase (Table S2). Among other pollutants, the levels of $PM_{2.5}$ increased by an average of 4.03%, while surprisingly, there was no change in CO concentration. In contrast to other pollutants, SO_2 and O_3 reduced by −1.79% and −1.96% respectively, during the second phase. The decreasing spell of O_3 was higher than SO_2. A significant difference can be seen in the rise of PM_{10} and $PM_{2.5}$ during the second phase, while a considerable reduction in particulate matter was experienced during the first phase due to reduced transportation, construction and industrial activities. SO_2 was the only pollutant, which showed a constantly decreasing trend during the both phases. In summary, the present study concludes that the YRD region experienced a prominent decline and improvement in air pollutant levels, except for O_3 during the first phase, while the behavior of the air pollutants was different during the second phase.

3.7. Changes in meteorological parameters

Fig. S5 shows the daily average temperature, relative humidity, wind speed and total rainfall during the study period in the YRD region. Overall, air temperature indicated a constantly increasing trend with some fluctuations during all the years. The increasing ratio for temperature was higher in 2018 and 2019 than the other years. Large fluctuations were observed for RH, however, it exhibited a declining trend in 2020 (COVID-19 period) with some differences. A similar pattern for WS was also experienced, however, the fluctuations of WS were lower than RH. The WS in the YRD region for the years 2019, 2020 and 2022 was relatively higher than that found in the other years. From Fig. S5 it is evident that the rain was high in 2020 and 2022, while it showed decreasing tendencies in 2019 and 2021. Moreover, a slight increase in rainfall can be seen in 2018. Overall, the meteorological parameters revealed mixed behavior in the YRD region.

3.8. The relationship between ambient air pollutants and meteorological variables

Meteorological variables significantly affect the air pollutants and play an important role in their formation, dispersion and transportation (Chen et al., 2019a,b; Barzeghar et al., 2022). To evaluate the relationship between ambient air pollutants and meteorological variables, Pearson's correlation analysis was performed (Fig. S6). A strong correlation was observed between PM_{10} and $PM_{2.5}$ ($r = 0.75$), NO_2 and SO_2 ($r = 0.75$), while SO_2 and RH had a significant negative relationship ($r = -0.71$) in 2022. PM_{10}, $PM_{2.5}$, SO_2 and NO_2 were positively correlated, while there was a significant negative correlation between SO_2 and RH ($r = -0.65$), O_3 and RH ($r = -0.59$) in 2021. The concentrations of CO and $PM_{2.5}$ were highly correlated ($r = 0.80$), NO_2 and T ($r = 0.77$), while RH and SO_2 had a strong negative correlation ($r = -0.68$) in 2020. High RH favors the creation of temperature inversion, which makes the atmospheric stratification constant and hampers the vertical dispersal of pollutants (Yang et al., 2019). PM_{10} and $PM_{2.5}$ had a strong significant positive relationship ($r = 0.91$), while the concentration of SO_2 was also highly correlated with NO_2 concentration ($r = 0.81$) in 2019. PM_{10}, $PM_{2.5}$, SO_2 and NO_2 had a strong significant positive relationship, while a strong negative correlation between WS and NO_2 ($r = -0.64$), WS and SO_2 ($r = -0.64$) was observed in 2018.

Hu et al. (2021) reported that wind speed was the major meteorological factor affecting pollutant distribution. NO_2 and CO indicated a positive relationship with T during the COVID-19 period, while there was a significant negative correlation between NO_2 and RH (Fig. S6). CO showed a weak and negative correlation with RH. The concentrations of NO_2 and CO were negatively correlated with WS and PP. These findings are very similar to Zhou et al. (2020) study, who found a negative correlation between air pollutants and wind speed in Beijing and Nanjing. O_3 had a significant positive correlation with T due to the important role of temperature in the formation of O_3 (Chen et al., 2019a). Moreover, the concentration of O_3 was negatively correlated with RH, WS and PP in 2020. The results indicate that most of the pollutants negatively correlated with the meteorological parameters in the YRD region.

4. Conclusions

The COVID-19 pandemic in different areas and regions of the world has provided an inordinate opportunity to work in this direction. In the current study, we used a random forest model to assess the ambient air quality patterns associated with the COVID-19 outbreak in the YRD region. The innovative aspect of the current study is that we predicted the ambient air quality in the two phases using a random forest approach and compared the results with the previous two years and the following two years, which makes it different compared to the earlier studies related to the COVID-19. According to the obtained results, the model showed the best performance in the prediction of air pollutants. The actual and predicted values at the provincial and municipal levels also indicated the effectiveness of the RF model. Moreover, the YRD region experienced a significant drop in the concentrations of air pollutants, except for O_3, during the COVID-19 period in 2020, compared with the concentrations of air pollutants during the same period in the previous two years 2018–19. The concentrations of PM_{10}, $PM_{2.5}$, NO_2 and CO were increased, while SO_2 and O_3 levels slightly reduced in 2021–22 compared with the same dates of COVID-19. The present study concludes that the air pollutants showed different behavior during the two phases in the YRD region. The results of this research will be helpful for the scientific community, policymakers, planners and local authorities to mitigate air pollution problems in the future. This study can be further extended to other regions and countries by findings new correlations factors between ambient air pollutants and meteorological parameters.

Credit author statement

Ahmad Hasnain: Conceptualization, Methodology, Data curation, Formal analysis, Writing - original draft, Writing - review & editing, Validation, Visualization; Yehua Sheng: Supervision, Conceptualization, Resources, Investigation, Project administration, Funding acquisition; Muhammad Zaffar Hashmi: Supervision, Conceptualization, Investigation, Writing - review & editing; Uzair Aslam Bhatti: Validation, Investigation, Data curation, Writing - review & editing; Zulkifl Ahmed: Validation, Data curation, Formal analysis, Writing - review & editing; Yong Zha: Supervision, Conceptualization, Resources, Investigation.

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

The authors do not have permission to share data.

Acknowledgements

This research was supported by the Key Fund of National Natural Science Foundation of China (grant No. 41631175).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2022.137638.

References

AirVisual, 2019. Airvisual–air quality monitor and information you can trust. Available at: https://www.airvisual.com/. (Accessed 26 August 2019). Accessed.

Barzeghar, V., Hassanvand, M.S., Faridi, S., Abbasi, S., Gholampour, A., 2022. Long-term trends in ambient air pollutants and the effect of meteorological parameters in Tabriz, Iran. Urban Clim. 42, 101119 https://doi.org/10.1016/j.uclim.2022.101119.

Bauwens, M., Compernolle, S., Stavrakou, T., Müller, J.F., et al., 2020. Impact of coronavirus outbreak on NO2 pollution assessed using TROPOMI and OMI observations. Geophys. Res. Lett. 47 (11) https://doi.org/10.1029/2020GL087978, 0–3.

Berman, J.D., Ebisu, K., 2020. Changes in US air pollution during the COVID-19 pandemic. Sci. Total Environ. 739, 139864.

Bhatti, U.A., Wu, G., Bazai, S.U., Nawaz, S.A., et al., 2022. A pre- to post-COVID-19 change of air quality patterns in Anhui province using path analysis and regression. Pol. J. Environ. Stud. 31 (5), 1–14. https://doi.org/10.15244/pjoes/148065.

Bilal, M., Mhawish, A., Nichol, J.E., Qiu, Z., et al., 2021. Air pollution scenario over Pakistan: characterization and ranking of extremely polluted cities using long-term concentrations of aerosols and trace gases. Remote Sens. Environ. 264, 112617 https://doi.org/10.1016/j.rse.2021.112617.

Briz-Redón, A., Belenguer-Sapiña, C., Serrano-Aroca, A., 2021. Changes in air pollution during COVID-19 lockdown in Spain: a multi-city study. J. Environ. Sci. 101, 16–26. https://doi.org/10.1016/j.jes.2020.07.029.

Brokamp, C., Jandarov, R., Hossain, M., Ryan, P., 2018. Predicting daily urban fine particulate matter concentrations using a random forest model. Environ. Sci. Technol. 52, 4173–4179.

Chen, Y., Zheng, M., Lv, J., Shi, T., et al., 2019b. Interactions between ambient air pollutants and temperature on emergency department visits: analysis of varying-coefficient model in Guangzhou. China. Sci. Total Environ. 668, 825–834. https://doi.org/10.1016/j.scitotenv.2019.03.049.

Chen, X., Situ, S., Zhang, Q., Wang, X., Sha, C., Zhouc, L., Wu, L., Wu, L., Ye, L., Li, C., 2019a. The synergetic control of NO2 and O3 concentrations in a manufacturing city of southern China. Atmos. Environ. 201, 402–416.

Chen, K., Wang, M., Huang, C., Kinney, P.L., Anastas, P.T., 2020. Air pollution reduction and mortality benefit during the COVID-19 outbreak in China. Lancet Planet. Health 4, e210–e212.

Chu, B., Zhang, S., Liu, J., Ma, Q., He, H., 2021. Significant concurrent decrease in PM2.5 and NO2 concentrations in China during COVID-19 epidemic. J. Environ. Sci. 99, 346–353.

CNEMC, 2019. China National Environmental Monitoring Centre. http://www.cnemc.cn/. (Accessed 8 August 2019). Accessed.

Ghasempour, F., Sekertekin, A., Kutoglu, S.H., 2021. Google Earth Engine based spatio-temporal analysis of air pollutants before and during the first wave COVID-19 outbreak over Turkey via remote sensing. J. Clean. Prod. 319, 128599 https://doi.org/10.1016/j.jclepro.2021.128599.

Guo, L., Chen, B., Zhang, H., Zhang, Y., 2020. A new approach combining a simplified FLEXPART model and a Bayesian-RAT method for forecasting PM10 and PM2.5. Environ. Sci. Pollut. Res. 27, 2165–2183. https://doi.org/10.1007/s11356-019-06605-w.

Hasnain, A., Sheng, Y., Hashmi, M.Z., Bhatti, U.A., et al., 2022. Time series analysis and forecasting of air pollutants based on prophet forecasting model in Jiangsu province, China. Front. Environ. Sci. 10, 945628 https://doi.org/10.3389/fenvs.2022.945628.

Hasnain, A., Hashmi, M.Z., Bhatti, U.A., Nadeem, B., Wei, G., Zha, Y., et al., 2021. Assessment of air pollution before, during and after the COVID-19 pandemic lockdown in nanjing, China. Atmosphere 12, 743. https://doi.org/10.3390/atmos12060743.

He, G., Pan, Y., Tanaka, T., 2020. COVID-19, city lockdowns, and air pollution: evidence from China. In: MedRxiv. https://doi.org/10.1101/2020.03.29.20046649.

He, W., Wang, Y., Zuo, J., Luo, Y., 2017. Sectoral linkage analysis of three main air pollutants in China's industry: comparing 2010 with 2002. J. Environ. Manag. 202, 232–241.

Hu, M., Chen, Z., Cui, H., Wang, T., Zhang, C., Yun, K., 2021. Air pollution and critical air pollutant assessment during and after COVID-19 lockdowns: evidence from pandemic hotspots in China, the Republic of Korea, Japan, and India. Atmos. Pollut. Res. 12, 316–329. https://doi.org/10.1016/j.apr.2020.11.013.

Hua, J., Zhang, Y., Foy, B.D., Shang, J., et al., 2021. Quantitative estimation of meteorological impacts and the COVID-19 lockdown reductions on NO2 and PM2.5 over the Beijing area using Generalized Additive Models (GAM). J. Environ. Manag. 291, 112676 https://doi.org/10.1016/j.jenvman.2021.112676.

Huang, C., Wang, T., Niu, T., Li, M., Liu, H., Ma, C., 2021. Study on the variation of air pollutant concentration and its formation mechanism during the COVID-19 period in Wuhan. Atmos. Environ. 251, 118276 https://doi.org/10.1016/j.atmosenv.2021.118276.

Huang, L., Zhang, C., Bi, J., 2017. Development of land use regression models for PM2.5, SO2, NO2 and O3 in Nanjing, China. Environ. Res. 158, 542e552.

Lal, S., Naja, M., Subbaraya, B.H., 2000. Seasonal variations in surface ozone and its precursors over an urban site in India. Atmos. Environ. 34, 2713e2724. https://doi.org/10.1016/S1352-2310(99)00510-5.

Le, T., Wang, Y., Liu, L., Yang, J., Yung, Y., Li, G., Seinfeld, J., 2020. Unexpected air pollution with marked emission reductions during the COVID-19 outbreak in China. Science. https://doi.org/10.1126/science.abb7431.

Li, L., Li, Q., Huang, L., Wang, Q., et al., 2020. Air quality changes during the COVID-19 lockdown over the Yangtze River Delta Region: an insight into the impact of human activity pattern changes on air pollution variation. Sci. Total Environ. 732, 139282 https://doi.org/10.1016/j.scitotenv.2020.139282.

Liu, H., Yue, F., Xie, Z., 2022. Quantify the role of anthropogenic emission and meteorology on air pollution using machine learning approach: a case study of PM2.5 during the COVID-19 outbreak in Hubei Province, China. Environ. Pollut. 300, 118932 https://doi.org/10.1016/j.envpol.2022.118932.

Liu, Y., Cao, G., Zhao, N., Mulligan, K., Ye, X., 2018. Improve ground-level PM2.5 concentration mapping using a random forests-based geostatistical approach. Environ. Pollut. 235, 272–282.

Lu, D., Mao, W., Zheng, L., Xiao, W., Zhang, L., Wei, J., 2021. Ambient PM2.5 estimates and variations during COVID-19 pandemic in the Yangtze River Delta using machine learning and big data. Rem. Sens. 13, 1423. https://doi.org/10.3390/rs13081423.

Lu, R., Zhao, X., Li, J., Niu, P., Yang, B., Wu, H., et al., 2020. Genomic characterisation and epidemiology of 2019 novel coronavirus: implications for virus origins and receptor binding. Lancet 395, 565–574. https://doi.org/10.1016/S0140-6736 (20) 30251-8.

Mandal, J., Chanda, A., Samanta, S., 2022. Air pollution in three megacities of India during the Diwali festival amidst COVID-19 pandemic. Sustain. Cities Soc. 76, 103504.

Mor, S., Kumar, S., Singh, T., Dogra, S., Pandey, V., Ravindra, K., 2021. Impact of COVID-19 lockdown on air quality in Chandigarh, India: understanding the emission sources during controlled anthropogenic activities. Chemosphere 263, 127978. https://doi.org/10.1016/j.chemosphere. 2020.127978 0045-6535/© 2020.

National Health Commission Of the People's Republic of China (NHC): http://www.nhc.gov.cn/xcs/yqtb/list_gzbd.shtml.

Orak, N.H., Ozdemir, O., 2021. The impacts of COVID-19 lockdown on PM10 and SO2 concentrations and association with human mobility across Turkey. Environ. Res. 197, 111018 https://doi.org/10.1016/j.envres.2021.111018.

Pal, S.C., Chowdhuri, I., Saha, I., Chakrabortty, R., et al., 2021a. Improvement in ambient-air-quality reduced temperature during the COVID-19 lockdown period in India. Environ. Dev. Sustain. 23, 9581–9608. https://doi.org/10.1007/s10668-020-01034-z.

Pal, S., Das, P., Mandal, I., Sarda, R., et al., 2021b. Effects of lockdown due to COVID-19 outbreak on air quality and anthropogenic heat in an industrial belt of India. J. Clean. Prod. 297, 126674 https://doi.org/10.1016/j.jclepro.2021.126674.

Reddy, B.S.K., Kumar, K.R., Balakrishnaiah, G., Gopal, K.R., et al., 2012. Analysis of diurnal and seasonal behavior of surface ozone and its precursors (NOx) at a semi-arid rural site in southern India. Aerosol Air Qual. Res. 12, 1081e1094.

Singh, V., Singh, S., Biswal, A., Kesarkar, A.P., Mor, S., Ravindra, K., 2020. Diurnal and temporal changes in air pollution during COVID-19 strict lockdown over different regions of India. Environ. Pollut. 266, 115368 https://doi.org/10.1016/j.envpol.2020.115368.

Spohn, T.K., Martin, D., Geever, M., O'Dowd, C., 2022. Efect of COVID-19 lockdown on regional pollution in Ireland. Air Qual. Atmos. Heal. 15, 221–234. https://doi.org/10.1007/s11869-021-01098-4.

Sulaymon, I.D., Zhang, Y., Hopke, P.K., Zhang, Y., Hua, J., Mei, X., 2021. COVID-19 pandemic in Wuhan: ambient air quality and the relationships between criteria air pollutants and meteorological variables before, during, and after lockdown. Atmos. Res. 250, 105362.

Teixidó, O., Tobías, A., Massagué, J., Mohamed, R., et al., 2021. The influence of COVID-19 preventive measures on the air quality in Abu Dhabi (United Arab Emirates). Air Qual. Atmos. Heal. https://doi.org/10.1007/s11869-021-01000-2.

Tobías, A., Carnerero, C., Reche, C., Massagúe, J., Via, M., et al., 2020. Changes in air quality during the Lockdown in Barcelona (Spain) one month into the SARS-CoV-2 epidemic. Sci. Total Environ. 726, 138540 https://doi.org/10.1016/j.scitotenv.2020.138540.

Wang, H., Tan, Y., Zhang, L., Shen, L., Zhao, T., et al., 2021a. Characteristics of air quality in different climatic zones of China during the COVID-19 lockdown. Atmos. Pollut. Res. 12, 101247 https://doi.org/10.1016/j.apr.2021.101247.

Wang, H., Miao, Q., Shen, L., Yang, Q., et al., 2021b. Characterization of the aerosol chemical composition during the COVID-19 lockdown period in Suzhou in the Yangtze River Delta, China. J. Environ. Sci. 102, 110–122. https://doi.org/10.1016/j.jes.2020.09.019.

WHO, 2020a. Director-general's opening remarks at the media briefing on covid-19–11 march 2020 Available: https://www.who.int/director-general/speeches/detail/who-director-general-s-opening-remarks-at-the-media-briefing-on-covid-19-11-march-2020.

Wu, X., Guo, J., Wei, G., Zou, Y., 2020. Economic losses and willingness to pay for haze: the data analysis based on 1123 residential families in Jiangsu province, China. Environ. Sci. Pollut. Res. 27, 17864–17877. https://doi.org/10.1007/s11356-020-08301-6.

Wu, G., Sahabuddin, M.D., Bhatti, U.A., Nawaz, S.A., et al., 2022. COVID-19 and air pollution: air quality impact in 13 cities of the Jiangsu province of China. Pol. J. Environ. Stud. 31 (5), 1–10. https://doi.org/10.15244/pjoes/149714.

Yang, J., Ji, Z., Kang, S., Zhang, Q., Chen, X., Lee, S.Y., 2019. Spatiotemporal variations of air pollutants in western China and their relationship to meteorological factors and emission sources. Environ. Pollut. 254, 112952.

Yao, L., Li, W., Du, Y., 2021. Impact of COVID-19 on air quality in the Yangtze River Delta, China. Environ. Monit. Assess. 193, 523. https://doi.org/10.1007/s10661-021-09342-1.

Zeng, Y., Jaffe, D.A., Qiao, X., Miao, Y., Tang, Y., 2020. Prediction of potentially high PM2.5 concentrations in chengdu, China. Aerosol Air Qual. Res. 20, 956–965. https://doi.org/10.4209/aaqr.2019.11.0586.

Zhao, N., Wang, G., Li, G., Lang, J., Zhang, H., 2020a. Air pollution episodes during the COVID-19 outbreak in the Beijinge-Tianjine-Hebei region of China: an insight into the transport pathways and source distribution. Environ. Pollut. 267, 115617 https://doi.org/10.1016/j.envpol.2020.115617.

Zhao, Z., Qin, J., He, Z., Li, H., Yang, Y., Zhang, R., 2020b. Combining forward with recurrent neural networks for hourly air quality prediction in northwest of China. Environ. Sci. Pollut. Res. 27, 28931–28948. https://doi.org/10.1007/s11356-020-08948-1.

Zhou, H., Yu, Y., Gu, X., Wu, Y., et al., 2020. Characteristics of air pollution and their relationship with meteorological parameters: northern versus southern cities of China. Atmosphere 11, 253. https://doi.org/10.3390/atmos11030253.