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Evaluating the Impacts of Regional Transport and Monsoons on the Air Quality in Nanjing Based on VAR Model

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Abstract

Heavy air pollution occurred frequently in Nanjing in recent years. To estimate local and regional source contributions of air pollutants ($PM_{2.5}$, SO_2, NO_2, CO) in Nanjing during different seasons, the air monitoring data between January 2015 and April 2016 was collected from China National Environmental Monitoring Center and analyzed with time series approach of Granger causality test and vector autoregressive (VAR) model. It is found that the correlations of pollutions among cities during Southeast prevailing wind (SP) period were weaker than North prevailing wind (NP) period, partly due to low pollution level in SP period. The Granger causality tests indicate that there were more causal relationships of air pollutants between Nanjing and 9 surrounding cities in NP period partly due to heavier regional pollution, and the causalities were not always consistent with prevailing wind direction. Variance decomposition in VAR model suggests that local source played a more important role than regional source during SP period, as it donated 68.6%, 65.3%, 69.7% and 76.9% for $PM_{2.5}$, SO_2, NO_2 and CO, respectively. However, Nanjing was strongly affected by regional transport during NP period, as North direction contributed 64.4%, 58.1%, 60.2% and 56.8% of $PM_{2.5}$, SO_2, NO_2 and CO in Nanjing, respectively. Furthermore, the observed air pollutant concentrations in Nanjing were well consistent with the model-simulated results, except highly active pollutant of NO_2. These results indicated that the time series approach could be a simple tool for understanding the impacts of regional transport and monsoons on the air quality in a particular city.

Keywords: Regional transport; Air quality; VAR model; Granger causality test; Monsoons

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Introduction

The Yangtze River Delta (YRD) region, one of the most developed and fastest growing economic development regions in China, has been suffering poor air quality in the recent decades [1-4], especially for severe haze pollution [5-8]. Being the capital of Jiangsu Province, Nanjing covers an area of 6587 km^2 and has a resident population of 8.22 million by the end of 2014 (Nanjing Statistical Yearbook 2015). Based on surface observations obtained from Nanjing Meteorological Station and five suburban stations, the annual occurrence of hazy days in Nanjing increased from 1961 to 2005, with a negative correlation with visibility during hazy periods [9]. In the recent decades, Nanjing has been suffering from heavy air pollution, which has induced public health risk and even caused mortality in Nanjing [10-12].

Nanjing is located in the lower reaches of the YRD region, a subtropical monsoons climate region with the southeast monsoons in summer and the northwest monsoons in winter. As significant relationship was found between urban air quality and monsoons [13], air quality in Nanjing would be also influenced by the monsoons. Generally, Nanjing is downwind of most developed mid-YRD regions including the megacity Shanghai and Suzhou-Wuxi-Changzhou City cluster in summer, and downwind of the North China Plain (NCP) region in winter. The local/regional source analysis of air pollution is important if air quality in Nanjing is to be effectively improved. Many research papers have investigated the main factors of heavy air pollution in Nanjing. Chen et al. [14] reported that burning of crop straw coupled with temperature inversion was the main reason for the heaviest pollution episode of Nanjing in 2007. Zhu et al. [4] pointed out that the transport of air pollutants releasing from crop residue burning in the central and north parts of Jiangsu Province, combining with unfavorable weather condition was the main reason for a serious air pollution in Nanjing and surrounding regions between October 28 and 29, 2008. Based on back trajectory analysis, Kang et al. [6] figured out both local emission and regional transport were the main factors for a long-lasting haze episode in Nanjing and its surrounding areas from October 15 to 31, 2009. Based on Lagrangian dispersion simulations, a calculation of potential source contributions suggested that the YRD region contributed over 70% of CO; the North-YRD and the NCP region were the main contributors to $PM_{2.5}$ pollution at the SORPES station in Nanjing [15]. Additionally, based on "sawtooth cycle" of aerosol and its component, Tang et al. [7] estimated that the northeastern areas on average contributed 46% of total non-refractory-PM_1 mass in Nanjing during winter haze, among which aged low-volatility organics and sulfate presented the largest regional contributions, accounting for 74% and 65%, respectively. Therefore, the regional transport could be a serious pollution source in Nanjing.

In the previous research [16], we applied vector autoregressive (VAR) model and Granger causality test to investigate the impacts of climate (prevailing wind) on the inter-influence of $PM_{2.5}$ among four cities (Shanghai, Lin'an, Ningbo and Nanjing) in the YRD region; and attempted to evaluate the relative source contributions of $PM_{2.5}$ from each city. In that study, we concluded that, to better evaluate the impacts of transport of haze pollutants to a specific city, it is ideal to choose the receptor city at the center of a system that is surrounded by the other cities, covering the entire possible transport pathway. Consequently, we construct an improved VAR model for the air quality of Nanjing by selecting the system including Nanjing and its 9 surrounding cities (**Figure 1**). Additionally, to make the source contributions more reasonable, only the variance decomposition for Nanjing was analyzed, because the contributions of regional transport from all possible directions for Nanjing were considered.

Data and Methods

Sites description and air pollutant data

The impact of regional transport and local emission on air quality in Nanjing (NJ) is studied by time series of pollutant concentrations in NJ and its 9 surrounding cities of Bengbu (BB), Huaian (HA), Yancheng (YC), Nantong (NT), Suzhou (SZ), Huzhou (HZ), Xuancheng (XC), Tongling (TL) and Hefei (HF) (**Figure 1**), which cover every possible transport pathways to Nanjing and with similar distances to Nanjing. These cities are all located in the Yangtze Plain, facilitating the connection of air pollutants and formation of regional air pollution. However, topography conditions of some cities are little different. For instance, TL and XC are located in the piedmont plain which might favor convergence of air pollutants; NT and YC are located on the coast which might benefit diffusion of air pollutants.

The monitoring data of cities above were collected from China National Environmental Monitoring Center (CNEMC), during January 1st, 2015 and April 30th, 2016. The pollutant data were used for time series analysis with several statistical methods.

Identifying the prevailing wind

Since the meteorological conditions often play an important role in the transport and diffusion of air pollutants, the regional transport direction of air pollutants is expected to change across monsoons. In this paper, the ratio of wind direction in Nanjing was identified by backward trajectory statistics. 48-h backward air trajectory arriving at Nanjing city (32.063°N, 118.782°E) was calculated by Hysplit-4 model from the National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory (http://ready.arl.noaa.gov/HYSPLIT.php). And meteorological data came from NCEP/NCAR Reanalysis meteorological database. The model was run four times per day at UTC times of 0:00, 06:00,12:00 and 18:00 (local times 08:00, 14:00, 20:00 and 02:00 of the next day, respectively), between 2013/6/01 and 2016/05/30, with starting height of 500 m above ground level. Trajectory clustering and statistics were performed by the geographic information system (GIS) based software, TrajStat [17]. The clustering result was shown in **Figure S1.** And these clusters were classified as north, east, south and west clusters, representing wind direction in regional scale. In Nanjing, the probability of clusters in every direction and every month were calculated (**Table S1)** to identify southeast prevailing wind control (SP) months and north prevailing wind control (NP) months.

Data analysis and process

Combining with prevailing wind control months (**Table S1**) and period of the available monitoring data (2015/01/01-2016/04/30), the reference time was classified as SP period (2015/05/01-2015/08/31) and NP period (2015/11/01-2016/02/29), which are also recognized as summer monsoons and winter monsoons.

It should be noted that the meteorological conditions in SP and NP period are different. As shown in **Figure S2**, the temperature in SP period was relatively high, while the sea level pressure was relatively low. What's more, the precipitation in SP period was higher than NP period, which would help precipitation of air pollutants. It is well known that monsoons dominates air pollutant transport characteristics [18,19], influences the weather and air quality of cities in monsoons climate region [13]. Tu et al. [20] found that the variations of O_3 and its precursors in Nanjing were significantly affected by local meteorological conditions and distant transports associated with Asian monsoons. Thus, the monsoons condition (summer and winter monsoons) was considered to be the main factor to influence air quality in Nanjing.

To identify the impact of prevailing wind on the pollutants transport and inter-influence between Nanjing and surrounding cities, and describe pollutants source contributions in Nanjing, several sets of time series analysis were carried out during SP and NP period as follows:

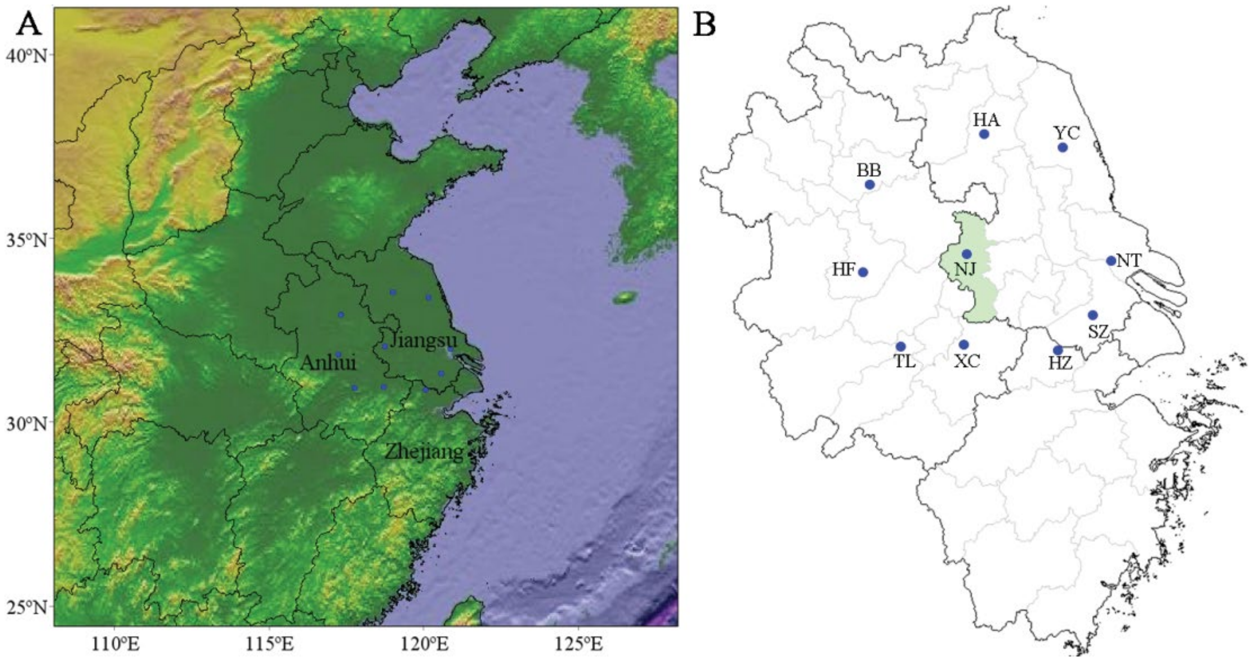

Figure 1 Topography (A) and location (B) map of the research area, including Nanjing (NJ) and surrounding city of Bengbu (BB), Huaian (HA), Yancheng (YC), Nantong (NT), Suzhou (SZ), Huzhou (HZ), Xuancheng (XC), Tongling (TL) and Hefei (HF).

(1) Correlation analysis of air pollutants among cities: The Pearson correlation analysis is the most widely-used type of technical methods in the multivariate data analysis to describe the dependence between variables. And it's used to characterize the relationship of daily pollutant concentrations among cities during SP and NP period, respectively.

(2) Inter-influence of air pollutants between Nanjing and other cities: The causality between Nanjing and its surrounding cities was tested by Granger causality test, which was first proposed in 1969 [21] and became a popular method for possible causal relationships between variables of time series [22]. The optimal lag of the Granger causality test is the same as the optimal order of the VAR model.

(3) VAR model design for the local/regional contributions of air pollutants in Nanjing: The VAR model is an econometric model used to analyze stationary time series [23]. In previous research, we presented a VAR model to describe $PM_{2.5}$ source contribution from surrounding cities, which assumed that the concentrations at the receptor city could be explained by the transport and mixing of pollutants from all cities in the model from previous days (Xiao et al.) i.e.,

$$C_{i,o} = \sum_{j} \left(a_{ij}.C_{j,-1}\right) + R_i \qquad (1)$$

Where the subscripts *i* and *j* represent any two cities within the system, while *i* could equal to *j*; $C_{i,0}$ and $C_{j,-1}$ are the $PM_{2.5}$ concentrations at present and the previous day respectively for any two cities in the model; while ; a_{ij} are the regression coefficients; and R_i is the regression residue for city *i*.

In this study, since only the present pollutant concentrations at Nanjing were discussed, the model could be simplified as

$$C_0 = \sum_{j} \left(a_j.C_{j-1}\right) + R \qquad (2)$$

Where the subscripts *j* represent any city within the system, including Nanjing itself; a_j are the regression coefficients, which describes the mixing process; and *R* is the residue, which represents the influence to air pollutant at Nanjing, that could not be explained by transport and mixing processes during the 1 day interval, including reactions and variations in emissions.

The VAR model is used to describe the local/regional contributions of air pollutants ($PM_{2.5}$, SO_2, NO_2 and CO) in Nanjing by variance decomposition. Variables of NJ, BB, HA, YC, NT, SZ, HZ, XC, TL and HF were used to establish VAR model A, with NJ itself for local contribution; BB, HA and YC for North direction contribution; NT, SZ and HZ for Southeast direction contribution; XC, TL and HF for Southwest contribution. Additionally, VAR model B was constructed, with BB, HZ and XC excluded for North, Southeast and Southwest direction, respectively. Both VAR model A and B were built during SP and NP period, respectively.

The optimal lag orders of Granger causality test and VAR models were presented in **Tables S2 and S3.** The lag order selected by the criterion was generally 1 (1 day), especially in VAR model B. Thus, 1 lag order was considered as the optimal lag orders. And AR root test showed that all the reciprocal absolute value of AR characteristic root was less than one (**Figure S4**), which means the established VAR models are stable.

(4) Model-simulated pollutant concentrations.

VAR model was also used to simulate pollutant concentrations in NJ, and the results were compared with observed pollutant concentrations. Additionally, VAR model in NP period was used to predict pollutant concentrations between 2015/01/01 and 2015/02/28. And linear regression model is used to evaluate the VAR model simulation.

Results and Discussion

Regional distributions of pollutants

Figure 2 plots the box chart of air pollutant ($PM_{2.5}$, SO_2, NO_2, and CO) concentrations in 10 cities of the YRD region during SP and NP period. Monthly variation of $PM_{2.5}$, SO_2, NO_2 and CO concentrations in 10 cities over the YRD region was shown in **Figure S3**.

In 2012, China issued the new Ambient Air Quality Standards (GB3095-2012), with an annual mean of 35 μ g/m³ and the 24-h mean of 75 μ g/m³ for ambient $PM_{2.5}$ concentration. It's obvious that average $PM_{2.5}$ concentrations in 10 cities were higher than the annual mean standard (35 μ g/m³) during SP period, and the top three polluted cities were BB (53.51 ± 20.28 μ g/m³), TL (50.77 ± 31.09 μ g/m³) and HF (47.55 ± 22.80 μ g/m³), which are located in the western area of NJ. As pollutant concentrations in

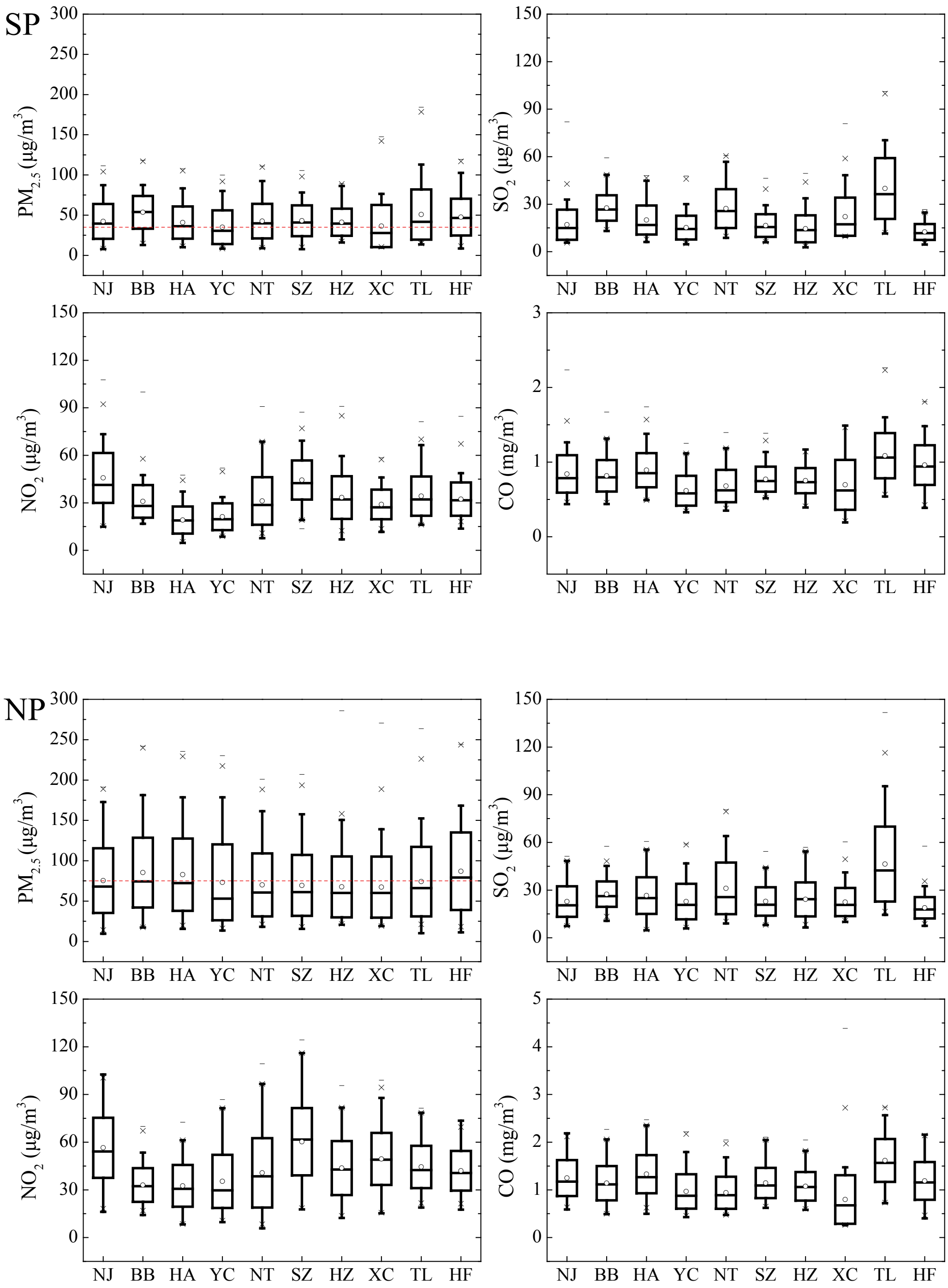

Figure 2 Box chart of pollutant concentrations in 10 cities. SP represents days between 2015/5/1 and 2015/8/31, NP represents days between 2015/11/1 and 2016/2/29.

SP period were relatively low in one year cycle, the annual mean $PM_{2.5}$ concentration would be higher than the national standard. Furthermore, the average $PM_{2.5}$ concentrations in 10 cities were adjacent to the 24-h mean standard (75 μ g/m^3) during NP period, and the top three polluted cities were HF (87.00 ± 48.03 μ g/m^3), BB (85.31 ± 43.30 μ g/m^3), HA (82.24 ± 44.81 μ g/m^3), which are located in the northwest area of NJ. It seemed that cities of HF and BB had more severe $PM_{2.5}$ pollution during SP and NP period, partly due to their locations of inland.

It's interesting that SO_2 level in all cities showed little difference between SP and NP period. The top three polluted cities in SP period were TL (38.43 ± 18.19 μ g/m^3), NT (27.07 ± 12.49 μ g/m^3) and BB (24.61 ± 9.00 μ g/m^3). Similarly, TL (46.37 ± 23.56 μ g/m^3), NT (31.06 ± 16.24 μ g/m^3) and BB (27.46 ± 8.01 μ g/m^3) were the top three polluted cities in NP period. Additionally, the top clean city was HF, with 10.90 ± 4.94 μ g/m^3 in SP period and 18.87 ± 6.70 μ g/m^3 in NP period. Since coal-fired plants were the major source for SO_2 emission in the YRD region, regional distribution of SO_2 might be mainly dependent on local emission, as the top three polluted cities and the top clean city were the same in SP and NP period. Lin et al. [2] displayed the distribution of power plants over YRD with annual energy intensities and pointed out high SO_2 level occur in the area with more power plants.

The regional distribution of NO_2 was different from SO_2, as the most polluted cities were SZ and NJ, with 60.29 ± 21.20 μ g/m^3, 56.47 ± 18.88 μ g/m^3 during NP period; 37.26 ± 10.60 μ g/m^3, 38.96 ± 10.91 μ g/m^3 during SP period. NO_2 level was low in HA and YC during SP period, while HA and BB had low NO_2 concentration during NP period. Emission inventory of primary air pollutants in the YRD region in 2010 showed that NOx emission in SZ and NJ was higher than NT, HA, YC and HZ [24]. Higher NO_2 concentration was found in SZ and NJ, probably due to more vehicles stocks in these two cities.

Similar to SO_2, the highest CO-polluted city was TL during SP and NP period, with a concentration of 0.91 ± 0.27 mg/m^3 and 1.62 ± 0.45 mg/m^3. And the lowest CO concentration was XC. Both TL and XC are located at the piedmont (**Figure 1A**). It seemed that high concentration of SO_2 and CO in TL came from power plants.

Generally, pollution level ($PM_{2.5}$, SO_2, NO_2 and CO) in 10 cities was relatively high during NP period, compared with SP period (**Figure 2**), which might be partly due to unfavorable meteorological dispersion conditions in winter [1].

Temporal relationship of pollutants among cities

Temporal relationship of pollutants among cities was performed by Pearson correlation analysis, and results were presented in **Figure 3** (detail results were shown in **Table S2**). Generally, correlations of pollutants among 10 cities during SP period were weaker than NP period, partly due to low pollution level in SP period (**Figure 2**). During SP period, the correlation of SO_2 among 10 cities was weaker, compared with other pollutants ($PM_{2.5}$, NO_2, CO). And no correlation of SO_2 was found between BB and other cities (Pearson's correlation coefficients were less than 0.2), even BB was relatively high SO_2 polluted city (**Figure 2**).

However, In NP period, nearly all Pearson correlation coefficients of $PM_{2.5}$ and NO_2 were higher than 0.4, indicating moderate regional pollution. It was noted that the highest SO_2 polluted city of TL still had poor correlation with other cities except NJ and NT, while the lowest CO level city of XC had poor correlation with other cities, partly due to the topographic condition.

Generally, in SP period, air mass come from the South YRD region and East China Sea wound be relatively clean and the pollution in each cities wound be more dependent on local emission, which might result in weaker correlation or even no correlation of pollutant concentrations between cities. In NP period, air mass mainly comes from the North YRD or NCP region would bring high-level pollutants, combining with other unfavorable meteorological conditions (such as less precipitation, **Figure S2**), which would induce high pollutant concentrations in Nanjing and surrounding cities and stronger correlations among cities.

Inter-influence of air pollutants between Nanjing and other cities

Granger causality test was introduced to investigate the causal relationship (inter-influence) of pollutants between NJ and other cities, during SP and NP period. **Figure 4** displayed the results of Granger causality test with arrows, during SP and NP period (detail results were shown in **Table S4**).

For $PM_{2.5}$, the results of Granger causality test between NJ and other cities were consistent with prevailing wind direction, except BB and HA. NJ was influenced by BB, HA and TL during SP period, not consistent with the prevailing wind. During NP period, NJ was influenced by HA and YC, and influence SZ, HZ, XC and TL, consistent with prevailing wind direction.

As the SO_2 pollution level was relatively low during SP and NP period, NJ was influenced by particular cities and not always consistent with the prevailing wind direction. Interestingly, the highest SO_2 polluted city, TL, would not influence NJ, which may be partly due to lag order of the Granger causality test.

For NO_2, NJ was influenced by particular surrounding cities during SP period. But in NP period, NJ would influence SZ, HZ and XC, consistent with prevailing wind direction.

For CO, only a few causal relationships were found between NJ and particular surrounding cities, which were not always consistent with prevailing detection.

Generally, more interrelationships were found in NP period than that in SP period, indicating the monsoons (prevailing wind) facilitate the transport of air pollution. It is noted that the Granger test here was based on daily pollutant concentrations and the lag order was one day. Actually, more Granger causality was found with different lag order.

The cumulative impact on air pollutants in Nanjing

The discussion above showed that air pollutants in NJ were influenced by surrounding cities, especially in NP period. In order to estimate the cumulative impact on air pollutants in Nanjing from itself and other cities, VAR model was constructed during SP and NP period (VAR model estimates were shown in **Table S5**) and impulse response function was carried out. The results of

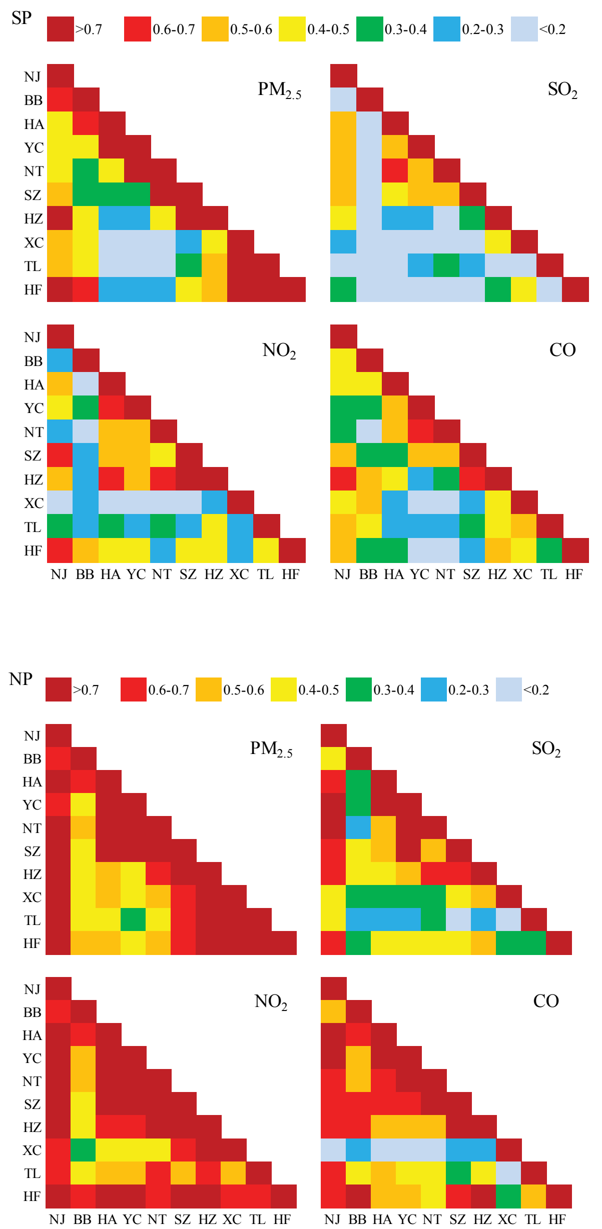

Figure 3 Pearson correlations of pollutant concentrations among 10 cities. SP represents days between 2015/5/1 and 2015/8/31, NP represents days between 2015/11/1 and 2016/2/29. Detail results were shown in Table S2.

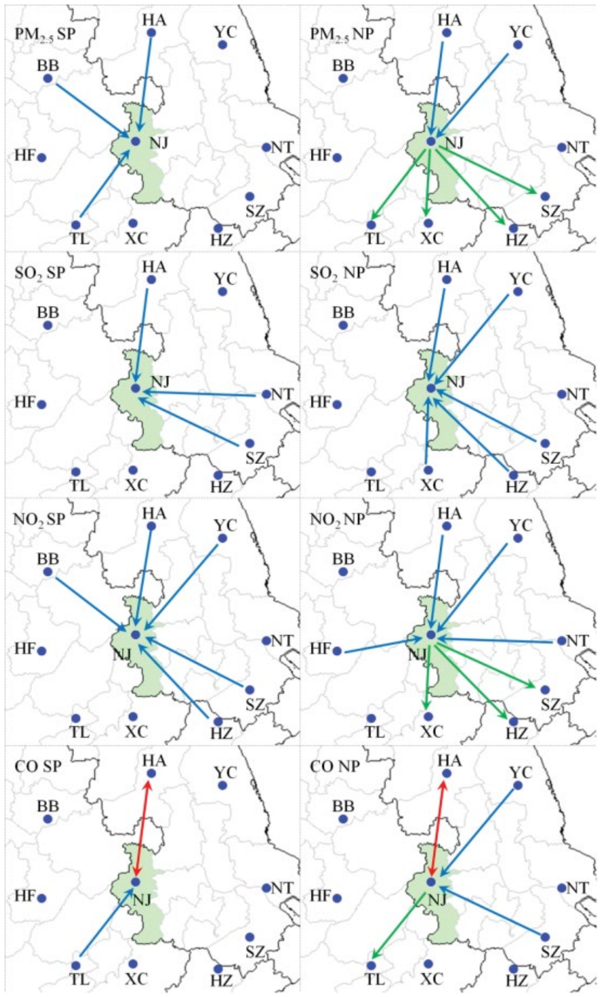

Figure 4 Results of Granger causality test of air pollutants between NJ and other cities. The arrow indicates the direction of causality. SP represents date between 2015/5/1 and 2015/8/31, NP represents days between 2015/11/1 and 2016/2/29. Detail results were shown in Table S4.

Impulse response showed that the strongest response at the first period is generally from NJ itself, and the impact would decrease after one or few periods, then attain nearly zero at the 21st period (**Figure S5**). Thus, 21-period response values were summed as the cumulative response of NJ (cumulative impact to NJ), and results were shown in **Figure 5**. Generally, the cumulative impact on NJ in NP period was higher than that in SP period, due to the larger standard deviation of variables in NP period. Results of VAR model B were similar to VAR model A.

For $PM_{2.5}$, the top three factors were TL, NJ and XC during SP period. And the top three factors were HA, NJ and SZ during NP period, with cumulative response value of 67.18, 67.03 and 57.33. The largest cumulative impact was from the southwest direction during SP period and the north direction during NP period, which seemed to be consistent with prevailing wind direction. However, for SO_2, the top three factors were HA, NJ and SZ during SP period, YC, NJ and SZ during NP period. The largest cumulative impulse was from the north direction area during SP and NP period, not consistent with prevailing wind direction. For NO_2, the three top factors were NJ, HA, SZ during SP period, and HF, YC and HA during NP period. That means the largest cumulative impulse was from NJ itself during SP period, due to heavy traffic pollution. For CO, the top three factors were NJ, XC and HA during SP period, and HA, NJ and YC during NP period. Similar to NO_2, NJ itself was the biggest factors during SP period.

Generally, during SP period the biggest factor for $PM_{2.5}$, SO_2, NO_2 and CO in NJ was TL, HA, NJ and NJ, respectively, not related to the prevailing wind direction. Furthermore, NJ itself was the second maximum factor for $PM_{2.5}$ and SO_2, which indicated that the

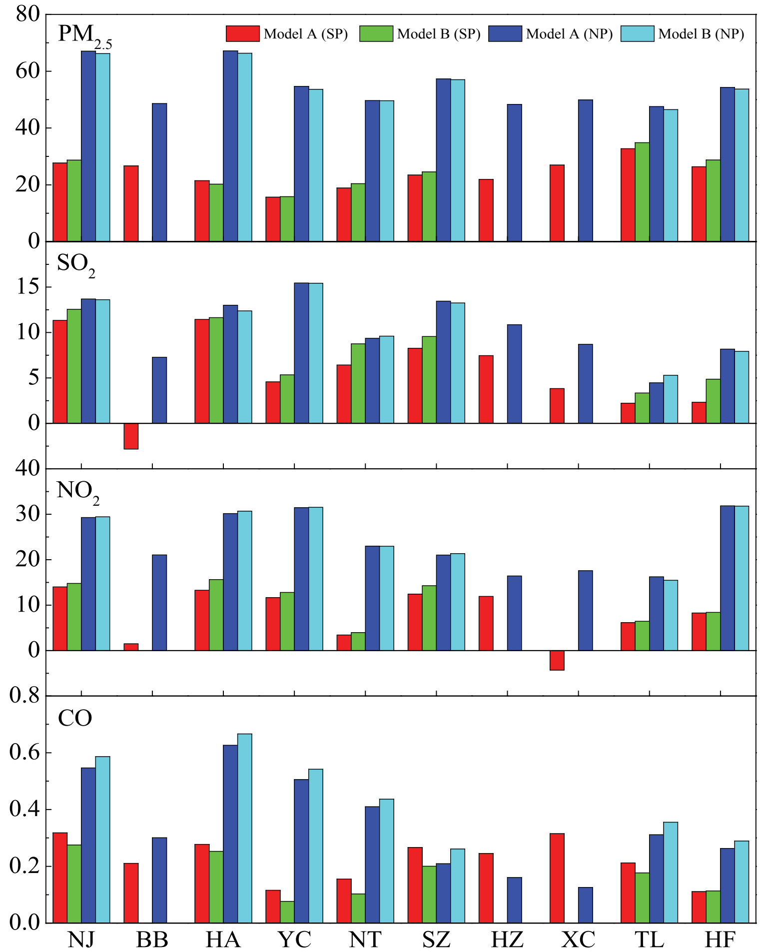

Figure 5 Accumulated response of NJ to generalized one standard deviation innovations. SP represents date between 2015/5/1 and 2015/8/31, NP represents days between 2015/11/1 and 2016/2/29. Detail results of impulse response were shown in Figure S4.

local emission might be the major factor for air quality change in Nanjing during SP period. During NP period, however, the largest factor for $PM_{2.5}$, SO_2, NO_2 and CO in NJ was HA, YC, HF and HA, respectively, consistent with prevailing wind direction, except HF. The results of impulse response function indicated that the north direction area might be the major factor for air quality change in Nanjing during NP period.

Local and regional contributions of air pollutants in Nanjing

The results of impulse response function have shown local emission and air pollutant change in north direction area might play a major role in air quality change in Nanjing during SP and NP period, respectively. In order to estimate the relative source contributions of air pollutants in Nanjing, variance decomposition in VAR model was carried out, and results were shown in **Figure 6** (detail results were shown in **Table S6** and **Figure S6**). Both VAR model A (10 variables) and B (7 variables, excluding BB, HZ and XC) demonstrated major source contributions of air pollutants came from the local emission in SP period, and the North direction in NP period. Source analysis of pollutants in Nanjing with VAR model A was shown below.

For $PM_{2.5}$, local source (NJ) contributed 68.6% in SP period, indicating local emission was the major source contribution for Nanjing. And the major foreign source was from the Southwest direction (19.7%), consistent with prevailing wind direction. However, during NP period, the local source contributed only 28.4%, and the North direction contribution was 64.4%. It's reasonable, as north prevailing wind transport $PM_{2.5}$ pollution from the NCP region in winter. Based on "sawtooth cycle" of aerosol, Tang et al. [7] indicated that the northeastern areas on average contributed 46% of total non-refractory-PM_1 in Nanjing during 2013 winter haze.

For SO_2, local source contributions were 65.3% and 28.5% in SP and NP period, respectively. It's interesting that the major foreign source contribution in SP period was from the North direction (26.5%), not the Southwest direction (5.4%). Possible causes were as follows. SO_2 concentration in TL was highest (**Figure 2**), but the correlation between TL and NJ was poor in SP period (**Figure 3**) and no Granger cause found between TL and NJ (**Figure 4**). Furthermore, the cumulative impact from TL was relatively low in SP and NP period, compared with other variables (**Figure 5**). Thus, from the perspective of time series analysis,

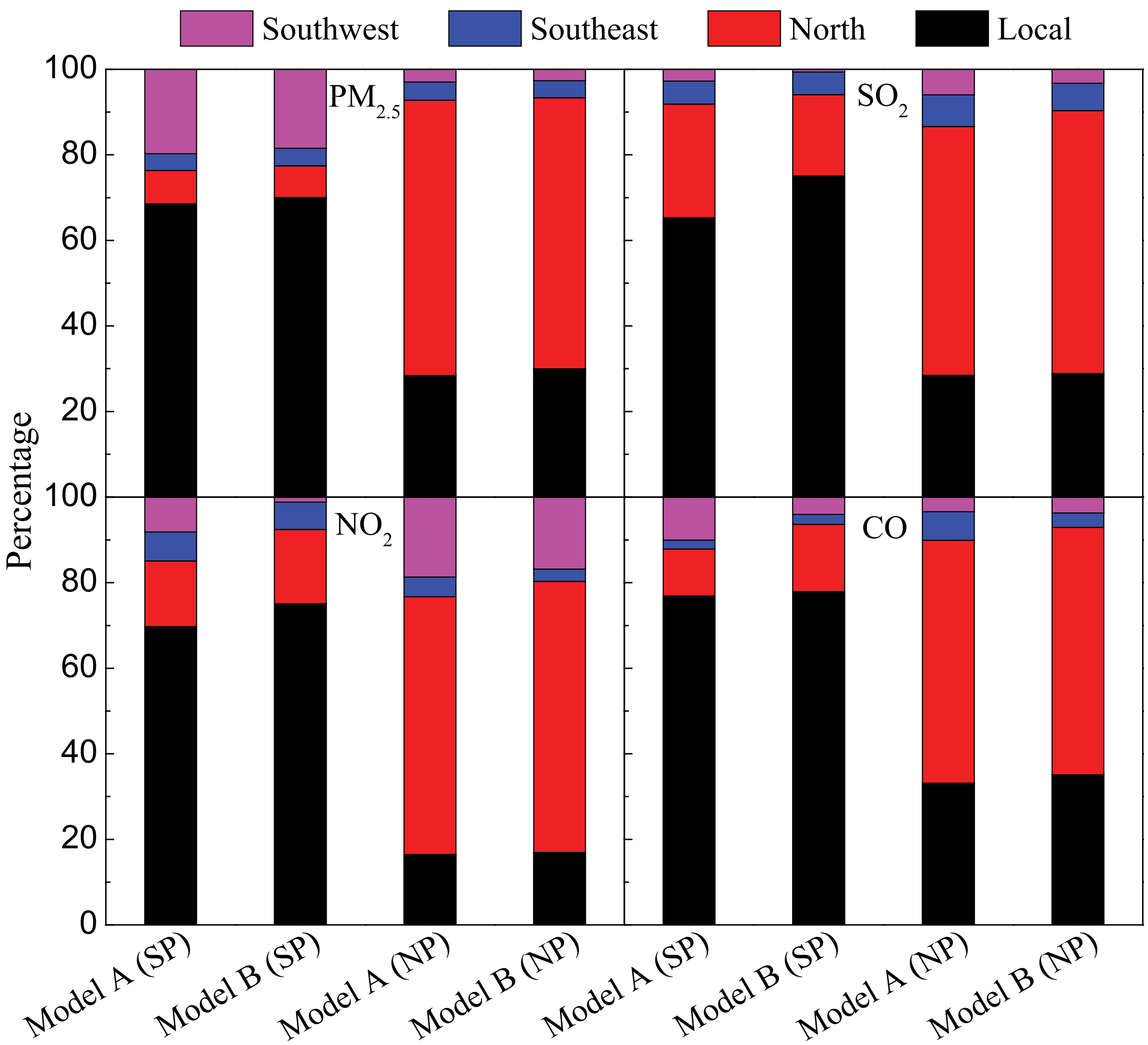

Figure 6 The local and regional contributions of air pollutants in Nanjing. SP represents date between 2015/5/1 and 2015/8/31, NP represents days between 2015/11/1 and 2016/2/29. Detail variance decomposition was shown in Figure S6 and Table S6.

the contribution from TL could be neglected and induced low contribution from the Southwest direction (**Table S6**). For NO_2, local source contribution was 69.7% and 16.5% in SP and NP period, respectively. Similar to SO_2 source contribution, the North direction was the major foreign source, contributing 15.4% and 60.2% in SP and NP period, respectively.

For CO, local source contributions were 76.9% and 33.1% in SP and NP period, respectively. The North direction and Southwest direction were the major foreign source in SP period, contributing 11.0% and 10.0%, respectively. In NP period, the North direction contributed 56.8%, became the major foreign source contributor.

Model simulation for air pollutants in Nanjing

VAR model could be a simple tool to point out regional source contributions in Nanjing from three major directions. In this part, both VAR Model A and Model B were used to simulate air pollutant concentrations in Nanjing. Because $PM_{2.5}$ is the primary and secondary particle pollutant, NO_2 is primary and secondary gas pollutant with high activation. Therefore, the variation of $PM_{2.5}$ and NO_2 would be more complex than SO_2 and CO (primary gas pollutants), and it was supposed that the model simulation would perform better for SO_2 and CO. **Figure 7** displayed the comparison between the observed and model-fitted pollutant concentrations in NJ. It can be found that most of the data dots are adjacently distributed along the y=x line. The adjusted R^2 of line regression models ranged from 0.371 to 0.552, and slopes ranged from 0.376 to 0.555. The best model-fitted pollutant was SO_2, a primary gas pollutant, while the worst one was CO, also a primary gas pollutant. Model B performance was similar to model A, as SO_2 was the best model-fitted pollutant (adjusted R^2=0.527), while CO was still the worst one (adjusted R^2=0.362).

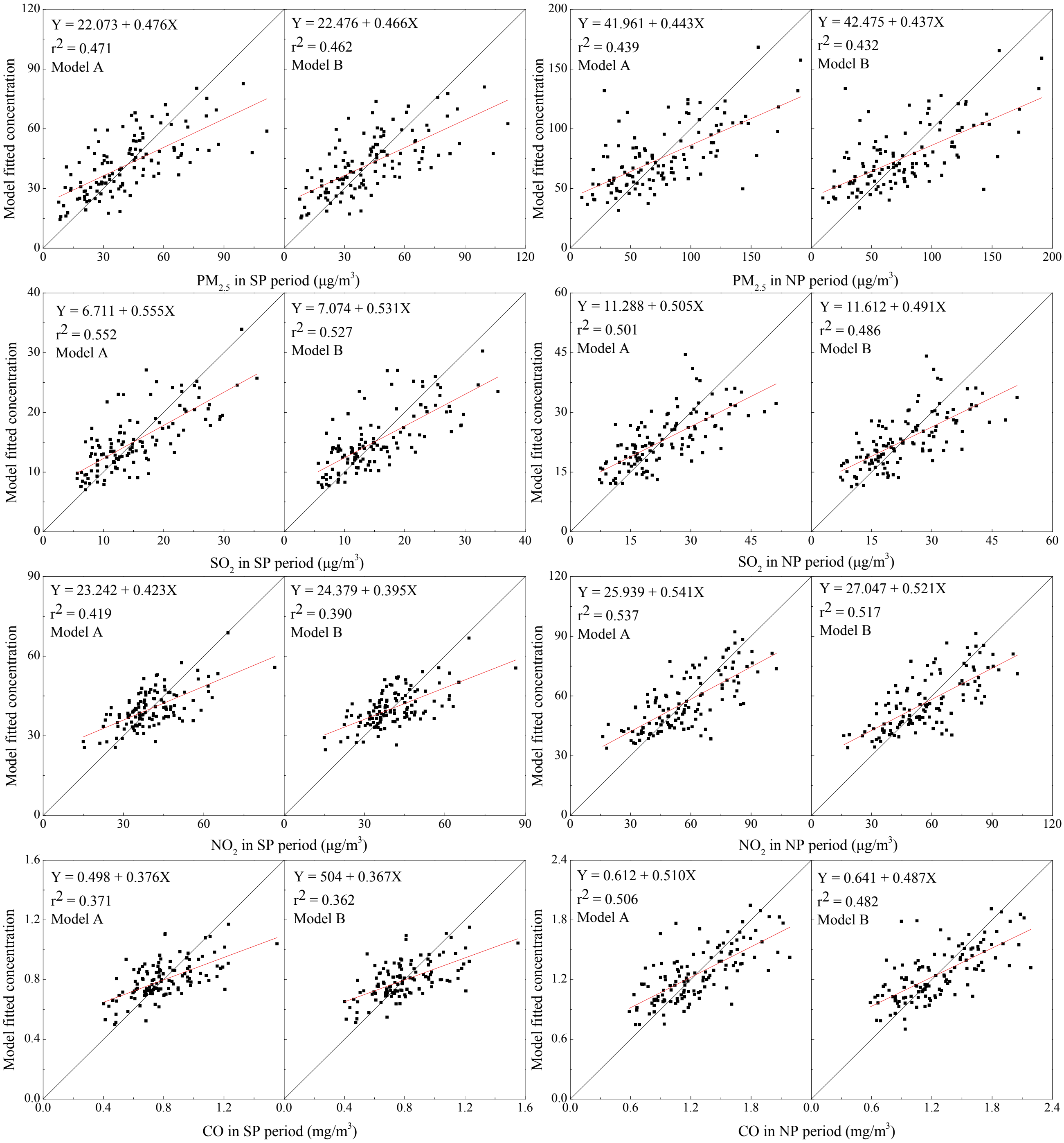

Figure 7 Comparison between the observed and model-fitted pollutant concentrations in Nanjing. SP period represents date between 2015/5/1 and 2015/8/31, NP period represents days between 2015/11/1 and 2016/2/29.

Additionally, the adjusted R^2 for every pollutant in model A was a little better than that in model B, as model A was constructed with 10 variables and might simulate pollutant concentration better.

Then, Model A and Model B during NP period were used to predict pollutant concentrations in NJ between 2015/1/1 and 2015/2/28, during which it also prevails north wind. And the comparison results were presented in **Figure 8**. In Model A, prediction of $PM_{2.5}$ was poor, as the adjusted R^2 was just 0.138. And adjusted R^2 was a little better for NO_2 (R^2=0.236), but the slope of line regressive model was only 0.110, with data dots deviating from the straight line. This indicated the prediction of Model A could not perform well for primary and secondary pollutants. Generally, VAR Model B was better in prediction **(Figure 8).** This result may be attributed to fewer variables in VAR Model B, and then the model performance was less affected by meteorological condition change during different periods **(Figure S2).**

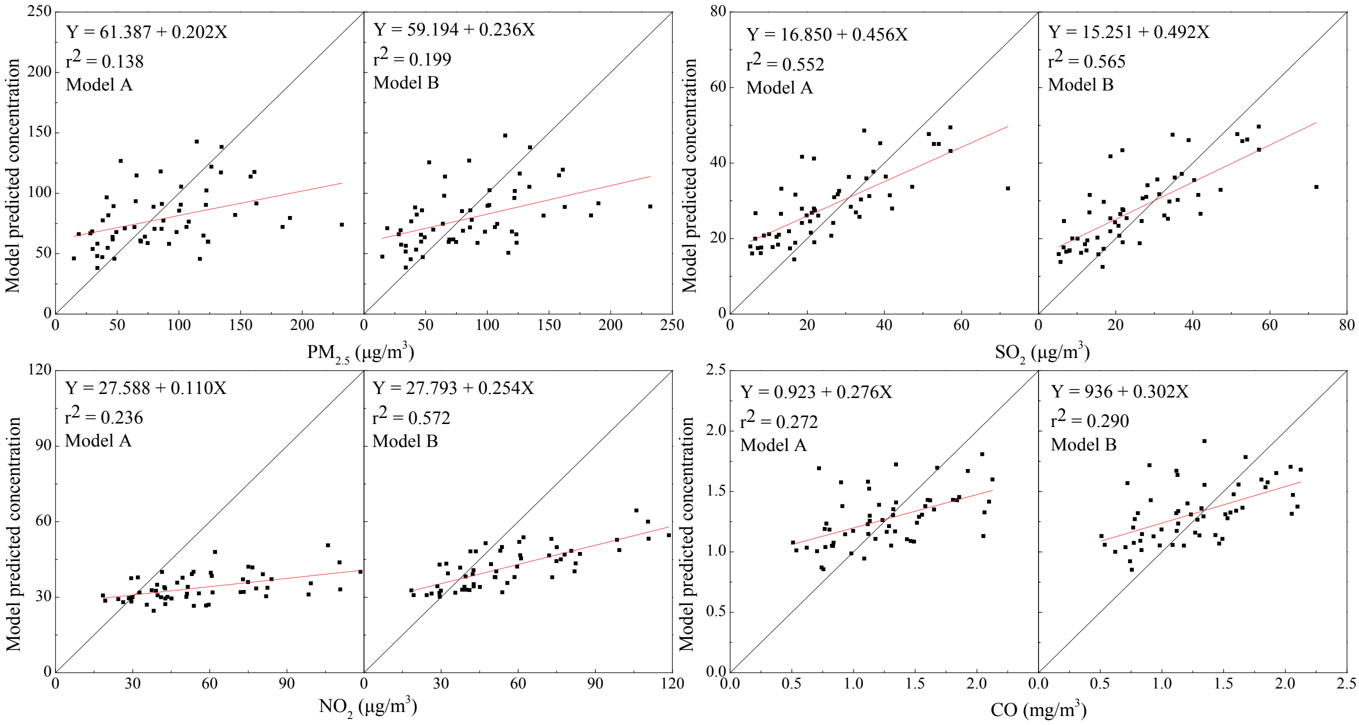

Figure 8 Comparison between the observed and model-predicted pollutant concentrations in Nanjing. VAR models were constructed during NP period and used to predict pollutant concentrations between 2015/1/1 and 2015/2/28.

Conclusions

In this study, we present the impacts of regional transport and meteorological condition (Prevailing wind) on the air pollution in Nanjing, based on the VAR model and Granger causality test with daily air monitoring data between January 2015 and April 2016. The results suggested that relatively low pollution level induces weaker correlations of pollutants among cities during SP period, as well as less inter-relationship between NJ and surrounding cities. And the inter-relationships were not always according to prevailing wind direction in both SP and NP period, partly due to distribution or emission of pollutants. The variance decomposition in VAR model indicated that local source contributed the largest fraction in SP period, with the percentage of 68.6%, 65.3%, 69.7% and 76.9% for $PM_{2.5}$, SO_2, NO_2 and CO, respectively. However, regional transport dominated the pollution source in NP period, as the North direction contributed more than 55% of all these pollutants in Nanjing. Furthermore, the VAR model could also be used to simulate air pollutant concentrations in Nanjing. Therefore, based on time series analysis, this paper highlights the important impact of regional transport and monsoons on air pollution in Nanjing and suggests that collaborative control measures among different cities are needed to improve air quality in the YRD region.

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