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Response to the First Referee

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Reviewer #1:

This paper by Yan et al. investigated the characteristics of winter haze episodes in Jingzhou of Central China under typical potential synoptic controls (PSCs) during November 2013-February 2014. Furthermore, they examined the contributions of local and transport of pollutants from surrounding regions to PM2.5 under different PSCs by applying the GEOS-Chem model with a high resolution. This work also studied the effectiveness of different emission control strategies in Jingzhou, Central China, and other surrounding regions under different PSCs, and highlights the importance of collaborative actions for PM2.5 mitigation under server haze pollution. In general, the study is well organized and worthy of publication. However, I have some specific comments that I feel deserve attention.

We thank the reviewer for comments, which have been incorporated to improve the manuscript.

Major comments

1. The writing should be improved before publication.

We thank the referee for this comment. We have made necessary corrections to grammar throughout the text (see details in the revision manuscript). We have polished the manuscript for all the authors.

2. The configuration of the model is vague. How many nested domains were applied in each simulation? What is the geographic coverage of each domain and the corresponding resolution? What are the emission inventories for each domain? A figure showing each nested domain is also highly recommended.

We thank the referee for this comment. We have added a figure in the revised file of supporting information to explain the geographic coverage of each domain and the corresponding resolution for GEOS_Chem global model (2° × 2.5°, providing boundary condition to nested model) and nested model (70°E-140°E, 15°S-55°N; 0.25° × 0.3125°). The emission inventories for each domain are shown in the revised Table S1 and Table S2. We also revised the description in the text of Sect. 2.3.

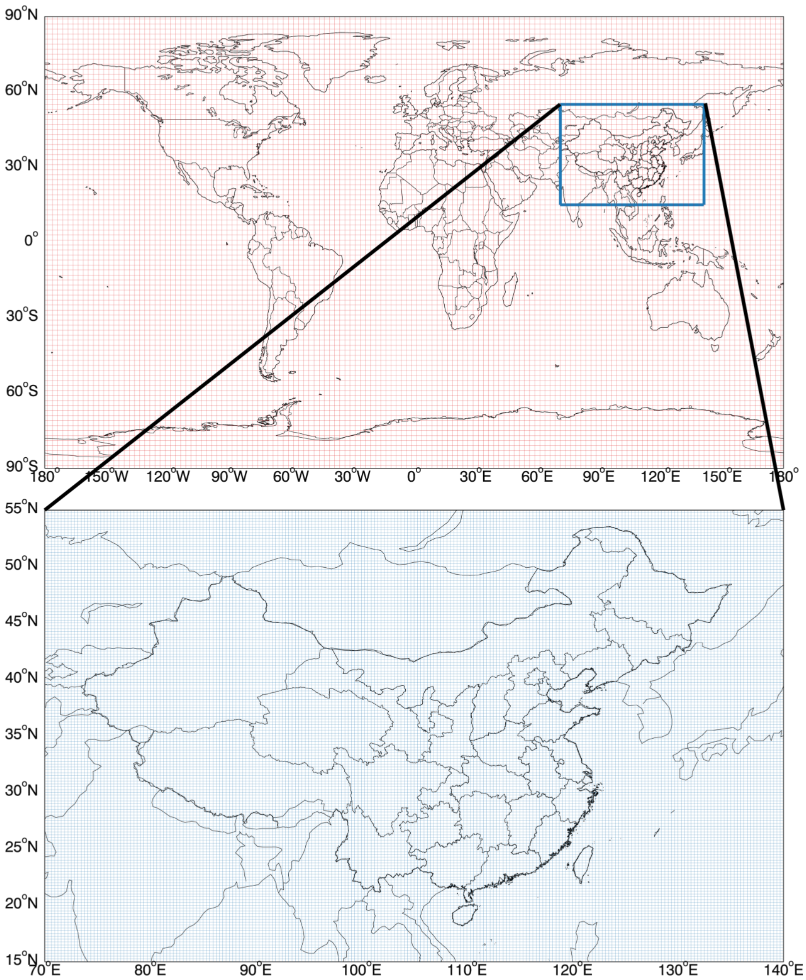

Figure S3 The geographic coverage of each domain and the corresponding resolution for GEOS_Chem global model (2° × 2.5°) and nested model (70°E-140°E, 15°S-55°N; 0.25° × 0.3125°).

Table S1 Anthropogenic and natural source emission inventories adopted in the GEOS-Chem global modelling of this study

Region	Abbreviation	Description	Resolution	Year	Species	Reference
Anthropogenic emission inventory						
Global	EDGAR	EDGAR v4.2 anthropogenic + biofuel	0.1°× 0.1°, monthly	2013-2014	NOx, SO_2, SO_4^{2-}, CO, NH_3	http://edgar.jrc.ec.europa.eu/overview.php?v=42
Global	BOND	BOND biofuel + anthropogenic BC + OC emissions	1°×1°, monthly	2000	BC and OC	Bond et al. (2007)
Global	RETRO	RETRO anthropogenic + biofuel	0.5°×0.5°, monthly	2000	NMVOCs[1] except C_2H_6 and C_3H_8	ftp://ftp.retro.enes.org/pub/emissions/aggregated/anthro/0.5x0.5/2000/
Global	SHIP	ICOADS ship emissions	1°×1°, monthly	2002	NO_x, SO_2, CO	Wang et al. (2008)
Global	AEIC	Aircraft emissions	1°×1°, monthly	2005	NO_x, SO_2, CO, NMVOCs[1], BC, OC	
China	MEIC	MEIC inventory for China	0.25°×0.25°, monthly	2013-2014	NO_x, SO_2, CO, NMVOCs[1], NH_3	http://www.meicmodel.org/.
USA	NEI2011	US EPA NEI-2011 emission inventory	0.1°× 0.1°, monthly	2013-2014	NO_x, SO_2, CO, NMVOCs[1], NH_3, BC, OC	https://www.epa.gov/air-emissions-inventories
Europe	EMEP	EMEP	1°×1°, annual	2013-2014	NO_x, SO_2, CO	Auvray and Bey (2005)
Biomass burning emission inventory						
Global	GFED4	GFED4 biomass burning inventory	0.25°× 0.25°, monthly	2013-2014	NO_x, SO_2, CO, NMVOCs, NH_3, BC, OC	http://www.globalfiredata.org, Giglio et al. (2013)
Biogenic emission inventory						

Global	MEGAN	MEGAN v2.1 biogenic emissions	—	2013-2014	ISOP, monoterpenes, sesquiterpenes, MOH, ACET, ETOH, CH_2O, ALD_2, HCOOH, C_2H_4, TOLU, PRPE	Guenther et al. (2012)
Other natural emission inventory						
Global	SoilNOx	Emission of NO_x from soils and fertiliser use	—	2013-2014	NO	Hudman et al. (2012)
Global	LightNOx	NO_x from lightning	—	2013-2014	NO	Murray et al. (2012)

1. RETRO includes PRPE, ALK_4, ALD_2, CH_2O and MEK; in the CTM, MEK emissions are further allocated to MEK (25 %) and ACET (75 %). AEIC and MEIC include PRPE, C_2H_6, C_3H_8, ALK_4, ALD_2, CH_2O, MEK and ACET. NEI2011 includes PRPE, C_3H_8, ALK_4, CH_2O, MEK and ACET. EMEP includes PRPE, ALK_4, ALD_2 and MEK. Emissions of C_2H_6 outside Asia are from Xiao et al. (2008).

Table S2 Anthropogenic and natural source emission inventories adopted in the GEOS-Chem nested modelling of this study

Region	Abbreviation	Description	Resolution	Year	Species	Reference
Anthropogenic emission inventory						
Non-China	EDGAR	EDGAR v4.2 anthropogenic + biofuel	0.1°× 0.1°, monthly	2013-2014	NOx, SO_2, SO_4^{2-}, CO, NH_3	http://edgar.jrc.ec.europa.eu/overview.php?v=42
Nested domain	BOND	BOND biofuel + anthropogenic BC + OC emissions	1°×1°, monthly	2000	BC and OC	Bond et al. (2007)
Non-China	RETRO	RETRO anthropogenic + biofuel	0.5°×0.5°, monthly	2000	NMVOCs[1] except C_2H_6 and C_3H_8	ftp://ftp.retro.enes.org/pub/emissions/aggregated/anthro/0.5x0.5/2000/
Nested domain	SHIP	ICOADS ship emissions	1°×1°, monthly	2002	NO_x, SO_2, CO	Wang et al. (2008)

Nested domain	AEIC	Aircraft emissions	1°×1°, monthly	2005	NO_x, SO_2, CO, NMVOCs[1], BC, OC	
China	MEIC	MEIC inventory for China	0.25°×0.25°, monthly	2013-2014	NO_x, SO_2, CO, NMVOCs[1], NH_3	http://www.meicmodel.org/.
Central China	SEEA	SEEA	0.1°× 0.1°, monthly	2017	NO_x, SO_2, CO, NH_3, VOCs	
Biomass burning emission inventory						
Nested domain	GFED4	GFED4 biomass burning inventory	0.25°× 0.25°, monthly	2013-2014	NO_x, SO_2, CO, NMVOCs, NH_3, BC, OC	http://www.globalfiredata.org, Giglio et al. (2013)
Biogenic emission inventory						
Nested domain	MEGAN	MEGAN v2.1 biogenic emissions	—	2013-2014	ISOP, monoterpenes, sesquiterpenes, MOH, ACET, ETOH, CH_2O, ALD_2, HCOOH, C_2H_4, TOLU, PRPE	Guenther et al. (2012)
Other natural emission inventory						
Nested domain	SoilNOx	Emission of NO_x from soils and fertiliser use	—	2013-2014	NO	Hudman et al. (2012)
Nested domain	LightNOx	NO_x from lightning	—	2013-2014	NO	Murray et al. (2012)

1. RETRO includes PRPE, ALK_4, ALD_2, CH_2O and MEK; in the CTM, MEK emissions are further allocated to MEK (25 %) and ACET (75 %). AEIC and MEIC include PRPE, C_2H_6, C_3H_8, ALK_4, ALD_2, CH_2O, MEK and ACET. NEI2011 includes PRPE, C_3H_8, ALK_4, CH_2O, MEK and ACET. EMEP includes PRPE, ALK_4, ALD_2 and MEK. Emissions of C_2H_6 outside Asia are from Xiao et al. (2008)

3. The circulation classification is the basis of all the analysis. Why did you choose the Lamb-Jenkension method? What are the advantages of this method compared to the ones used in other studies such as Chang and Zhan, 2017, Dai et al., 2021, etc.?

We thank the referee for this comment. We have reviewed the advantages of Lamb-Jenkension method with respect to the ones used in other studies in the revised Sect. 2.2: "Compared to the objective classification method PCA used in some studies (Chang and Zhan, 2017, Dai et al., 2021), this Lamb-Jenkension method is a combination of subjective and objective methods. After the objective judgment of the circulation, we also make subjective considerations to overcome the weaknesses of their respective, leading to better synoptic significance. Many works of circulation classification have used the Lamb-Jenkension method and reported that the analysis can well respond to the classification results (Philipp et al., 2016;Santurtun et al., 2015;Pope et al., 2015;Russo et al., 2014;Pope et al., 2014;Trigo and DaCamara, 2000)."

Philipp, A., Beck, C., Huth, R., and Jacobeit, J.: Development and comparison of circulation type classifications using the COST 733 dataset and software, International Journal of Climatology, 36, 2673-2691, 10.1002/joc.3920, 2016.

Pope, R. J., Savage, N. H., Chipperfield, M. P., Arnold, S. R., and Osborn, T. J.: The influence of synoptic weather regimes on UK air quality: analysis of satellite column NO_2, Atmospheric Science Letters, 15, 211-217, 10.1002/asl2.492, 2014.

Pope, R. J., Savage, N. H., Chipperfield, M. P., Ordonez, C., and Neal, L. S.: The influence of synoptic weather regimes on UK air quality: regional model studies of tropospheric column NO_2, Atmospheric Chemistry and Physics, 15, 11201-11215, 10.5194/acp-15-11201-2015, 2015.

Russo, A., Trigo, R. M., Martins, H., and Mendes, M. T.: NO_2, PM_{10} and O_3 urban concentrations and its association with circulation weather types in Portugal, Atmospheric Environment, 89, 768-785, 10.1016/j.atmosenv.2014.02.010, 2014.

Santurtun, A., Carlos Gonzalez-Hidalgo, J., Sanchez-Lorenzo, A., and Teresa Zarrabeitia, M.: Surface ozone concentration trends and its relationship with weather types in Spain (2001-2010), Atmospheric Environment, 101, 10-22, 10.1016/j.atmosenv.2014.11.005, 2015.

Trigo, R. M., and DaCamara, C. C.: Circulation weather types and their influence on the precipitation regime in Portugal, International Journal of Climatology, 20, 1559-1581, 10.1002/1097-0088(20001115)20:13<1559::aid-joc555>3.0.co;2-5, 2000.

4. The validation of model performances is very weak. The bias of the modeled PM2.5 in Jingzhou can be as high as more than 100 μg/m3, what are the possible reasons? The authors simply claimed the uncertainties in emissions, meteorology, and chemistry might cause this discrepancy without any details. What are the amount of the PM2.5

precursors emitted in this study and how are the values compared to the published literature? How about the meteorological parameters used by the model vs. observations? The authors claimed an improvement in sulfate by the increase in primarily emitted sulfate in the model, how is that compared with observations? They also analyzed the changes in the chemical composition of PM2.5 under different typical PSCs without examination of the model performances in the base case.

Thanks for this query and suggestion, which are valuable for us to improve this work.

In order to better evaluate the GEOS-Chem model performances, the spatial distribution of $PM_{2.5}$ concentrations averaged over the four typical heavy pollution processes simulated by the control (CON) simulation are compared with the observations (a total of 633 sites) from Ministry of Ecology and Environment of China (http://www.mee.gov.cn/) (revised Fig. 6). Similar to the underestimation in $PM_{2.5}$ at Jingzhou, the underestimation is on a national scale when compared with the MEE observations, with a bias of -29.3 μg/m^3, -18.7 μg/m^3, -39.0 μg/m^3 and -21.4 μg/m^3 on average for SW-type, NW-type, A-type and C-type synoptic pattern, respectively (Fig. 6).

In order to explain the causes of the model discrepancy, we have added Table S3 to show the observed (modeled) meteorological conditions averaged over these four pollution episodes controlled by SW-type, NW-type, A-type and C-type synoptic pattern, respectively. There is an overestimate in temperature and wind speed and an underestimate in humidity, which can partly contribute to the underestimation of modeled $PM_{2.5}$ concentrations. In addition, anthropogenic emissions for $PM_{2.5}$ precursors used here are for the year 2017 over Central China from our newly developed SEEA inventory (Table S4). From 2013 to 2017, anthropogenic NO_x, SO_2, and primary $PM_{2.5}$ emissions in Central China have declined substantially (Table S4), due to the implementation of stringent emission control measures for the 12th-13th Five-Year Plans (Zheng et al., 2018). The anthropogenic emissions biases may affect our simulations and $PM_{2.5}$ attribution results to some extent.

We have no observations of the chemical compositions of $PM_{2.5}$. In order to examine the model performances in the $PM_{2.5}$ chemical compositions, we have added Table 4 to review the reported concentrations of $PM_{2.5}$ and the three inorganic salts (sulfate, nitrate and ammonium) in other cities. The contributions of sulfate, nitrate and ammonium are 9.1%-31.9%, 5.7%-32.1% and 5.9%-13.3%, respectively. In the CON simulation, the fractions of each inorganic salt to $PM_{2.5}$ for these four typical heavy pollution processes are shown in revised Fig. S10, which are comparable to the previous results (Table 4).

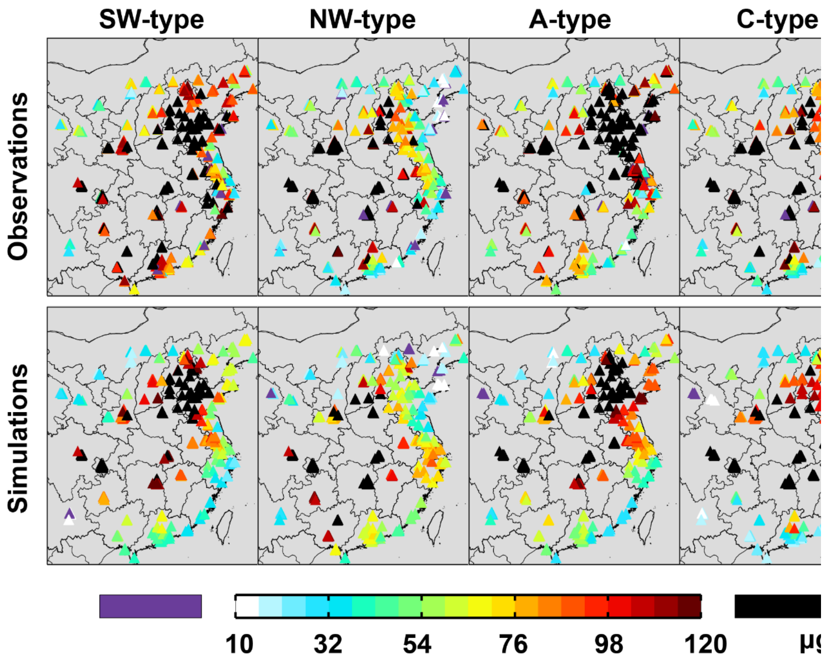

Figure 6 Spatial distribution of observed (top row) and modeled (bottom row, by CON case) $PM_{2.5}$ concentrations (μg/m^3) averaged over four severe pollution episodes controlled by SW-type (first column), NW-type (second column), A-type (third column) and C-type (forth column) synoptic pattern, respectively.

Table S3. The observed (modeled) meteorological conditions at Jingzhou averaged over these four pollution episodes controlled by SW-type, NW-type, A-type and C-type synoptic pattern, respectively.

PSC	Temperature (°C)	Humidity (%)	Pressure (kpa)	Wind speed (m/s)
SW	11.79 (12.96)	75.33 (69.25)	1018.33 (1024.06)	2.13 (3.09)
NW	3.61 (6.34)	71.16 (62.78)	1027.53 (1031.53)	1.44 (2.45)
A	5.81 (7.52)	64.96 (60.38)	1026.63 (1028.66)	1.45 (2.27)
C	9.60 (13.08)	78.10 (71.40)	1011.48 (1014.24)	1.88 (3.11)

Table S4. The emission amount of $PM_{2.5}$ precursors over Central China calculated from

SEEA (for the year 2017) and MEIC (for the years of 2013, 2014 and 2017) inventory (unit: 10^4 ton).

Category	SO_2	NO_X	NH_3	$PM_{2.5}$	CO	BC	OC	VOCs
SEEA (2017)	48.4	94.0	54.6	26.4	553.8	6.2	12.9	117.2
MEIC (2017)	52.0	70.4	57.5	35.2	629.2	6.8	11.7	116.4
MEIC (2013)	173.3	98.4	62.4	54.5	836.5	9.2	16.7	116.6
MEIC (2014)	97.0	80.0	61.1	46.8	744.2	8.3	15.3	116.4

Table 4 The reported concentrations of $PM_{2.5}$ and the three inorganic salts (sulfate, nitrate and ammonium, μg/m^3) in other cities.

References	Site	Time	$PM_{2.5}$	Sulfate	Nitrate	Ammonium
Cao et al., 2012	Beijing	01/03	115.6±46.6	20.0±4.2 (17.3%)	13.1±4.5 (11.3%)	9.4±4.1 (8.1%)
Cao et al., 2012	Qingdao	01/03	134.8±43.0	21.1±7.7 (15.7%)	19.3±9.2 (14.3%)	15.3±5.2 (11.4%)
Cao et al., 2012	Tianjin	01/03	203.1±76.2	32.5±15.1 (16.0%)	25.2±10.3 (12.4%)	22.2±9.8 (10.9%)
Cao et al., 2012	Xi'an	01/03	356.3±118.4	53.8±25.6 (15.1%)	29.0±10.0 (8.1%)	29.8±11.5 (8.4%)
Cao et al., 2012	Chongqing	01/03	316.6±101.2	60.9±19.6 (19.2%)	18.1±6.4 (5.7%)	28.8±8.9 (9.1%)
Cao et al., 2012	Hangzhou	01/03	177.3±59.5	33.4±16.7 (18.8%)	25.7±14.8 (14.5%)	19.1±10.7 (10.8%)

Cao et al., 2012	Shanghai	01/03	139.4±50.6	21.6±12.3 (15.5%)	17.5±8.7 (12.6%)	14.5±5.9 (10.4%)
Cao et al., 2012	Wuhan	01/03	172.3±67.0	31.4±15.6 (18.2%)	22.2±10.7 (12.9%)	18.4±10.2 (10.7%)
Zhang et al., 2011	Xi'an	03/06-03/07	194.1	35.6 (18.3%)	16.4 (8.4%)	11.4 (5.9%)
Huang et al., 2012	Xi'an	01/06-02/06	235.8±125.1	44.8±31.3 (19.0%)	20.5±14.2 (8.7%)	14.5±10.8 (6.1%)
Wang et al., 2020	Jinan	10/17	104±54	14.4±9.2 (13.8%)	33.4±23.2 (32.1%)	13.0±8.3 (12.5%)
Wang et al., 2020	Shijiazhuang	10/17	152±109	19.3±19.6 (12.7%)	42.8±41.1 (28.2%)	18.2±17.1 (12.0%)
Wang et al., 2020	Wuhan	12/17	117±33	13.6±3.2 (11.6%)	26.6±11.1 (22.7%)	13.1±3.8 (11.2%)
Wang et al., 2016a	Zhengzhou	01/11-02/11	297±160	48±36 (16.2%)	31±19 (10.4%)	21±16 (7.1%)
Wang et al., 2016a	Zhengzhou	01/12-02/12	234±125	23±10 (9.8%)	22±9 (9.4%)	16±5 (6.8%)
Wang et al., 2016a	Zhengzhou	01/13-02/13	337±168	56±39 (16.6%)	39±20 (11.6%)	31±18 (9.2%)
Luo et al., 2018	Zibo	12/06-02/07	224.9±85.4	40.1±19.2	18.1±9.0 (8.1%)	21.7±10.2 (9.7%)

				(17.9%)		
Wang et al., 2016b	Shanghai	12/11, 12/12, 12/13	73.9±57.5	12.2±9.2 (16.5%)	14.6±12.2 (19.8%)	8.2±6.7 (11.1%)
Xu et al., 2019	Beijing	02/17-03/17	180.5	20.1 (11.1%)	45.6 (25.3%)	22.5 (12.5%)
Xu et al., 2019	Beijing	05/17-09/17	186.7	20.2 (10.8%)	32.4 (17.4%)	17.1 (9.2%)
Xu et al., 2019	Beijing	10/17-11/17	167.5	17.9 (10.7%)	44.5 (26.6%)	20.9 (12.5%)
Zheng et al., 2016	Beijing	03/10-05/10	65.2±65.1	11.1±10.1 (17.0%)	11.1±11.0 (17.0%)	6.8±6.7 (10.4%)
Zheng et al., 2016	Beijing	07/09-08/09	88.9±39.1	23.0±13.9 (25.9%)	16.2±11.8 (18.2%)	11.8±6.8 (13.3%)
Zheng et al., 2016	Beijing	12/09-02/10	84.0±66.6	8.1±8.3 (9.1%)	8.0±9.6 (9.0%)	5.9±7.1 (6.6%)
Zheng et al., 2016	Guangzhou	11/10	73.3±16.5	16.6±4.0 (22.6%)	5.7±3.8 (7.8%)	6.2±2.0 (8.5%)
Zheng et al., 2016	Shenzhen	12/09	64.6±24.7	20.6±3.5 (31.9%)	4.9±3.5 (7.6%)	4.6±1.0 (7.1%)
Zheng et al., 2016	Wuxi	04/10-05/10	82.1±27.0	12.8±3.8 (15.6%)	9.9±6.3 (12.1%)	7.0±2.0 (8.5%)
Zheng et al., 2016	Jinhua	10/11-11/11	81.9±26.2	18.3±6.7 (22.3%)	12.6±7.0 (15.4%)	10.4±4.1 (12.7%)
Liu et al., 2018	Chongqing	2012-2013	73.5±30.5	19.7±9.6 (26.8%)	6.5±6.2 (8.8%)	6.1±2.7 (8.3%)

Liu et al., 2018	Shanghai	2012-2013	68.4±20.3	13.6±6.4 (19.9%)	11.9±5.0 (17.4%)	5.8±2.1 (8.5%)
Liu et al., 2018	Beijing	2012-2013	71.7±36.0	11.9±8.2 (16.6%)	9.3±7.5 (13.0%)	5.3±2.7 (7.4%)

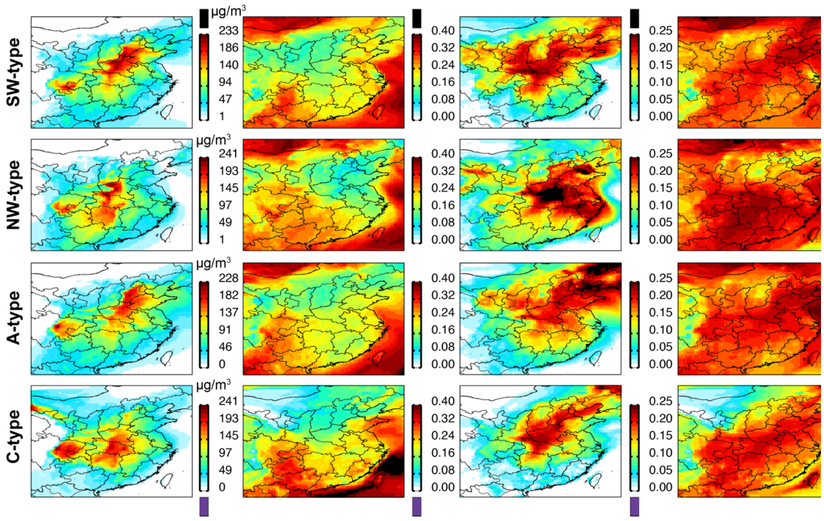

Figure S10 Spatial distribution of $PM_{2.5}$ concentrations and the fraction of each inorganic salt (sulfate: second column; nitrate: third column; ammonium: forth column) to $PM_{2.5}$ for these four typical heavy pollution processes simulated by GEOS-Chem control simulation.

Reference:

Cao, J.-J., Shen, Z.-X., Chow, J. C., Watson, J. G., Lee, S.-C., Tie, X.-X., Ho, K.-F., Wang, G.-H., and Han, Y.-M.: Winter and Summer PM2.5 Chemical Compositions in Fourteen Chinese Cities, Journal of the Air & Waste Management Association, 62, 1214-1226, 10.1080/10962247.2012.701193, 2012.

Huang, W., Cao, J., Tao, Y., Dai, L., Lu, S.-E., Hou, B., Wang, Z., and Zhu, T.: Seasonal Variation of Chemical Species Associated With Short-Term Mortality Effects of PM2.5 in Xi'an, a Central City in China, American Journal of Epidemiology, 175, 556-566, 10.1093/aje/kwr342, 2012.

Liu, Z., Gao, W., Yu, Y., Hu, B., Xin, J., Sun, Y., Wang, L., Wang, G., Bi, X., Zhang, G., Xu, H., Cong, Z., He, J., Xu, J., and Wang, Y.: Characteristics of PM2.5 mass concentrations and chemical species in urban and background areas of China: emerging results from the CARE-China network, Atmospheric Chemistry and Physics, 18, 8849-8871, 10.5194/acp-18-8849-2018, 2018.

Luo, Y., Zhou, X., Zhang, J., Xiao, Y., Wang, Z., Zhou, Y., and Wang, W.: PM2.5 pollution in a petrochemical industry city of northern China: Seasonal variation and source apportionment, Atmospheric Research, 212, 285-295, 10.1016/j.atmosres.2018.05.029, 2018.

Wang, H. L., Qiao, L. P., Lou, S. R., Zhou, M., Ding, A. J., Huang, H. Y., Chen, J. M., Wang, Q., Tao, S., Chen, C. H., Li, L., and Huang, C.: Chemical composition of PM2.5 and meteorological impact among three years in urban Shanghai, China, Journal of Cleaner Production, 112, 1302-1311, 10.1016/j.jclepro.2015.04.099, 2016a.

Wang, J., Li, X., Zhang, W., Jiang, N., Zhang, R., and Tang, X.: Secondary PM2.5 in Zhengzhou, China: Chemical Species Based on Three Years of Observations, Aerosol and Air Quality Research, 16, 91-104, 10.4209/aaqr.2015.01.0007, 2016b.

Wang, Q., Fang, J., Shi, W., and Dong, X.: Distribution characteristics and policy-related improvements of PM2.5 and its components in six Chinese cities, Environmental Pollution, 266, 10.1016/j.envpol.2020.115299, 2020.

Xu, Q., Wang, S., Jiang, J., Bhattarai, N., Li, X., Chang, X., Qiu, X., Zheng, M., Hua, Y., and Hao, J.: Nitrate dominates the chemical composition of PM2.5 during haze event in Beijing, China, Science of the Total Environment, 689, 1293-1303, 10.1016/j.scitotenv.2019.06.294, 2019.

Zhang, T., Cao, J. J., Tie, X. X., Shen, Z. X., Liu, S. X., Ding, H., Han, Y. M., Wang, G. H., Ho, K. F., Qiang, J., and Li, W. T.: Water-soluble ions in atmospheric aerosols measured in Xi'an, China: Seasonal variations and sources, Atmospheric Research, 102, 110-119, 10.1016/j.atmosres.2011.06.014, 2011.

Zheng, J., Hu, M., Peng, J., Wu, Z., Kumar, P., Li, M., Wang, Y., and Guo, S.: Spatial distributions and chemical properties of PM2.5 based on 21 field campaigns at 17 sites in China, Chemosphere, 159, 480-487, 10.1016/j.chemosphere.2016.06.032, 2016.

Minor comments:

Line 101-103: There must be many studies targeted the mitigation of PM2.5 at a regional scale (Ding et al., 2019; Zhang et al., 2019, Xing et al., 2018, 2019; Fu et al., 2017; etc.). Please rephrase this sentence.

We have rephrased this sentence: “Although there are many studies targeted $PM_{2.5}$ mitigations at a regional scale (Ding et al., 2019; Zhang et al., 2019, Xing et al., 2018, 2019; Fu et al., 2017; etc.), their results can not be directly applied to reduce winter $PM_{2.5}$ pollution under various synoptic controls."

Line 148-150: It is very confusing. The circulation classification is based on the meteorological data from November 2013 to February 2014, which is also the simulation episode. Why did you use the hourly PM2.5 data from 2013-2018?

We used the hourly $PM_{2.5}$ from November 2013 to December 2018 to screen the pollution days (daily mean $PM_{2.5}$ larger than 150 μg/m^3) and applied the daily mean sea level pressure between 2013 and 2018 from the NCEP/NCAR FNL Operational Global Analysis data to conduct the circulation classification. The meteorological observations at Jingzhou from November 2013 to February 2014 are used to analyze the meteorological characteristics during the period four severe particle pollution events occurred in succession over Central China. We have revised the Sect. 2.1:

"Hourly mass concentrations of $PM_{2.5}$ at Jingzhou (112.18°E, 30.33°N, 33.7 m) from November 2013 to December 2018 are obtained from Hubei Environmental Monitoring Central Station (http://sthjt.hubei.gov.cn/). We screen the pollution days with daily mean $PM_{2.5}$ concentrations larger than 150 μg/m^3 for circulation classification.

We use the daily mean sea level pressure (SLP) between 2013 and 2018 from the National Centers for Environmental Prediction/National Center for Atmospheric Research (NCEP/NCAR) Final (FNL) Operational Global Analysis data (horizontal resolution: 1° × 1°; temporal resolution: 6 hours; https://rda.ucar.edu/datasets/ds083.3/) to conduct the classification of Lamb-Jenkension circulation types.

The meteorological data of surface observations at Jingzhou, including ambient temperature, relative humidity, wind speed, wind direction and atmospheric pressure, are obtained from Hubei Meteorological Information and Technology Support Center (http://hb.cma.gov.cn/qxfw/index.html). The data from November 2013 to February 2014 are used to analyze the meteorological characteristics during the period four severe particle pollution events occurred in succession over Central China (Fig. S1)."

Line 195: Did you do nested runs or just one domain covering China? Please make this clear.

We have specified the model setups in the revised sentences: "The nested model, covering China (70°E-140°E, 15°S-55°N), is run with a horizontal resolution of 0.25° latitude × 0.3125° longitude and 72 vertical layers. The boundary condition of nested model is provided by the GEOS-Chem global model with a horizontal resolution of 2° latitude × 2.5° longitude (Fig. S3). Both global and nested simulations, driven by the GEOS-FP assimilated meteorological data, include detailed tropospheric Ozone-NO_x-VOCs-HO_x-aerosol chemistry."

Line 205: The SEEA inventory was developed for the year 2017. Did you use it directly without projection to the simulation episode? If you adjusted this inventory, what are the factors applied for the PM2.5 precursors and how did you obtain those data?

Yes, we have used the SEEA inventory of the year 2017 directly without projection to the simulation episode. The uncertainty discussion has been listed in Sect. 3.3: "Anthropogenic emissions for $PM_{2.5}$ precursors used here are for the year 2017 over Central China from SEEA inventory (Table S4). From 2013 to 2017, anthropogenic NO_x, SO_2, and primary $PM_{2.5}$ emissions in Central China have declined substantially (Table S4), due to implementation of stringent emission control measures for the 12^{th}-13^{th} Five-Year Plans (Zheng et al., 2018). The anthropogenic emissions biases may affect our simulations and $PM_{2.5}$ attribution results to some extent."

Line 215-217: Have you compared the modeled sulfate with observations, at least in Jingzhou? How about the model performances of the other components of PM2.5?

We have no observations of the chemical compositions of $PM_{2.5}$. In order to examine the model performances in the $PM_{2.5}$ chemical compositions, we have added Table 4 to review the reported concentrations of $PM_{2.5}$ and the three inorganic salts (sulfate, nitrate and ammonium) in other cities. The contributions of sulfate, nitrate and ammonium are 9.1%-31.9%, 5.7%-32.1% and 5.9%-13.3%, respectively. In the CON simulation, the fractions of each inorganic salt to $PM_{2.5}$ for these four typical heavy pollution processes are shown in revised Fig. S10, which are comparable to the previous observed results (Table 4). Please see details in the response of major comment#4.

Line 305: Again, I am confused about the emissions used in the CON case. You listed too many options for the anthropogenic source in Table S2. What inventories were EXACTLY selected for the CON case? Did you do a global/regional nested run? Please explain the choices of emissions in a separate column in the table.

We do a nested simulation, covering China (70°E-140°E, 15°S-55°N) with a horizontal resolution of 0.25° latitude × 0.3125° longitude. The boundary condition of nested model is provided by the GEOS-Chem global model with a horizontal resolution of 2° latitude × 2.5° longitude (Fig. S3). The emission inventories for each domain are shown in the revised Table S1 and Table S2. Please see details in the response of major comment#2.

Line 310: Please compare the meteorological field used in the model with observations to confirm that statement. Also, there are no perfect mechanisms, inventories, or parameterization of the model with no doubt. I suggest using "uncertainties".

We thank the referee for this comment. In order to explain the causes of the model discrepancy, we have added Table S3 to show the observed (modeled) meteorological conditions averaged over these four pollution episodes controlled by SW-type, NW-type, A-type and C-type synoptic pattern, respectively. There is an overestimate in temperature and wind speed and an underestimate in humidity, which can partly contribute to the underestimation of modeled $PM_{2.5}$ concentrations. In addition, anthropogenic emissions for $PM_{2.5}$ precursors used here are for the year 2017 over Central China from SEEA inventory (Table S4). From 2013 to 2017, anthropogenic NO_x, SO_2, and primary $PM_{2.5}$ emissions in Central China have declined substantially (Table S4), due to implementation of stringent emission control measures for the 12th-13th Five-Year Plans (Zheng et al., 2018). The anthropogenic emissions biases may affect our simulations and $PM_{2.5}$ attribution results to some extent. Additionally, the underestimation is on a national scale when compared with the MEE observations, with a bias of -29.3 μg/m^3, -18.7 μg/m^3, -39.0 μg/m^3 and -21.4 μg/m^3 on average for SW-type, NW-type, A-type and C-type synoptic pattern, respectively (Fig. 6). The national negative biases may be also attributed to insufficient resolution of the model (Yan et al., 2014) and imperfect chemical mechanisms (Yan et al., 2019). Please see details in the response of major comment#4.

Line 323-324: A comparison of the modeled fractions of the inorganic salts to observations, or reported values from other literature if no measurements are available.

We have no observations of the chemical compositions of $PM_{2.5}$. In order to examine the model performances in the $PM_{2.5}$ chemical compositions, we have added Table 4 to review the reported concentrations of $PM_{2.5}$ and the three inorganic salts (sulfate, nitrate and ammonium) in other cities. The contributions of sulfate, nitrate and ammonium are 9.1%-31.9%, 5.7%-32.1% and 5.9%-13.3%, respectively. In the CON simulation, the fractions of each inorganic salt to $PM_{2.5}$ for these four typical heavy pollution processes are shown in revised Fig. S10, which are comparable to the previous observed results (Table 4). Please see details in the response of major comment#4.

Line 324: "As shown in Table 3, ..."

Modified.

Line 358: How was this calculated? Please explain it.

We have added the explanation in the revised sentence: "In addition, the contributions from transboundary transport from non-Jingzhou Central China is simulated to be 12.0% by comparing the results of XJ0 and XCC0."

Line 415-417: How about the contributions of transported pollutants to the chemical composition of PM2.5 under the four PSCs?

We have discussed in the revised Sect. 3.4. During the pollution episodes of transmission-pollution characteristics (SW/NW-type), the contribution of transported pollutants to the chemical composition of $PM_{2.5}$ is significant. For the SW-type synoptic controlled pollution event, the transport of air pollutants from the south leads to the smallest proportion of the three inorganic salts (45.7%) in Jingzhou among the four pollution episodes (50.3%-55.5% for other three episodes), because the emissions of SO_2, NO_2 and NH_3 in the south (especially in Guangxi and Guizhou province) are smaller than those in Central China (Li et al., 2017a). However, during the NW-type synoptic controlled pollution episode, due to the transport contribution of pollutants from northern China (with much higher anthropogenic emissions of SO_2, NO_2 and NH_3) (Li et al., 2017a), the total proportion of the three inorganic salts is the highest (55.5%). For the other two types (A/C-type) synoptic controlled pollutions, local emission sources dominate the contributions and the contributions of transported pollutants to the chemical composition of $PM_{2.5}$ are small.

Line 424: The base year of emission reduction is 2015 for the 13th Five-year plan, which is quite different from your inventory. How effective is the designed reduction ratio of the anthropogenic emissions in this study?

Although the base year of emission reduction is 2015 for the 13^{th} Five-year plan, it does not affect to use the simulation results of emission scenarios (with the reduction ratio of 20% applied to the simulated year 2013/2014) to explore the emission reduction effect of specific haze pollution events. We have added this illustration in the revised Sect. 3.5.

Line 425 and 428-429: Please explain these abbreviations in the text as well.

We have revised these sentences as: "The differences in model results between CON (control simulation) and JSN/JSNN/JALL (emissions of (SO_2+NO_x)/(SO_2+NO_x+NH_3)/all pollution sources at Jingzhou are reduced by 20%) represent the environmental benefits caused by different local emission reduction scenarios. The potential $PM_{2.5}$ mitigations by joint prevention and control in different regions are calculated by sensitivity experiments of CCALL (emissions of all pollution sources over Central China are reduced by 20%), CNALL (over Central China and NCP region), CPALL (over Central China and PRD region) and TALL (over Central China, NCP, YRD, PRD and SCB regions)."

Line 437: I think an evaluation of the model performance in ammonium and/or ammonia is desired to confirm that.

We thank the referee for this comment. We have no observations of the chemical compositions of $PM_{2.5}$. Thus we have removed this statement in the revised text.

Figure 6, 8, 9, 10: I suggest to show the fraction of each inorganic salt to PM2.5 rather than their total mass.

We have shown the fraction of each inorganic salt to $PM_{2.5}$ in the revised Fig. S10.

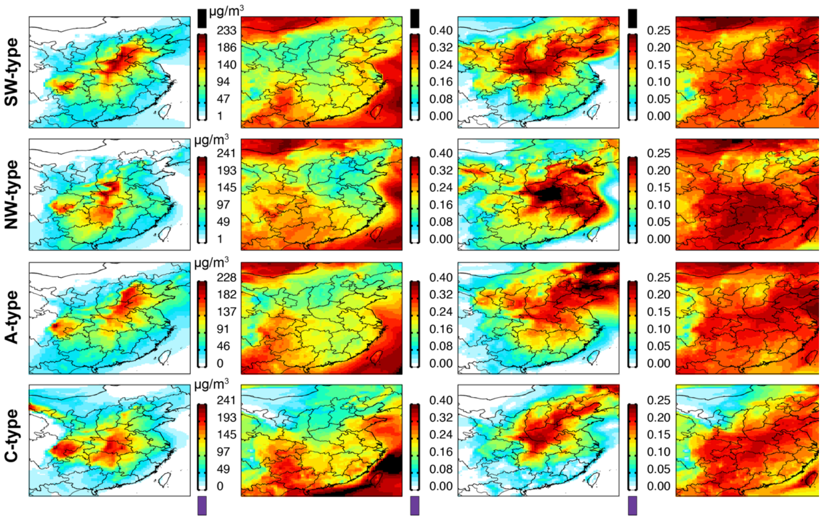

Figure S10 Spatial distribution of $PM_{2.5}$ concentrations and the fraction of each inorganic salt (sulfate: second column; nitrate: third column; ammonium: forth column) to $PM_{2.5}$ for these four typical heavy pollution processes simulated by GEOS-Chem control simulation.

Figure 11. It should be "TALL" in NW and C.

Modified.

References:

Wenyuan Chang and Jianqiong Zhan, The association of weather patterns with haze episodes: Recognition by $PM_{2.5}$ oriented circulation classification applied in Xiamen, Southeastern China, Atmospheric Research, 197, 425-436, 2017.

Huibin Dai, Jia Zhu, Hong Liao, Jiandong Li, Muxue Liang, Yang Yang, Xu Yue, Co-occurrence of ozone and $PM_{2.5}$ pollution in the Yangtze River Delta over 2013–2019: Spatiotemporal distribution and meteorological conditions, Atmospheric Research, 249, 105363, 2021.

Qiang Zhang, Yixuan Zheng, Dan Tong, et al., Drivers of improved $PM_{2.5}$ air quality in China from 2013 to 2017, Proceedings of the National Academy of Sciences, 116 (49) 24463-24469, 2019.

Aijun Ding, Xin Huang, Wei Nie, et al., Significant reduction of $PM_{2.5}$ in eastern China due to regional-scale emission control: evidence from SORPES in 2011–2018, Atmospheric Chemistry and Physics, 19, 11791– 11801, 2019.

Xing, J., Ding, D., Wang, S., Dong, Z., Kelly, J. T., Jang, C., ... & Hao, J. Development and application of observable response indicators for design of an effective ozone and fine-particle pollution control strategy in China. Atmospheric Chemistry & Physics, 19(21), 2019.

Xing, J., Ding, D., Wang, S., Zhao, B., Jang, C., Wu, W., ... & Hao, J. Quantification of the enhanced effectiveness of NOx control from simultaneous reductions of VOC and NH_3 for reducing air pollution in the Beijing–Tianjin–Hebei region, China. Atmospheric Chemistry and Physics, 18(11), 7799-7814, 2018.

Fu, X., Wang, S.,Xing, J., Zhang, X., Wang, T., & Hao, J. Increasing ammonia concentrations reduce the effectiveness of particle pollution control achieved via SO_2 and NOx emissions reduction in east China. Environmental Science & Technology Letters, 4(6), 221-227, 2017.

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Response to the Second Referee

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Reviewer #2:

This article analyses the potential synoptic controls over central China during winter haze pollution episodes by using Lamb-Jenkenson method and the NCEP/NCAR FNL operational global analysis data, and further evaluates the effectiveness of emission control to reduce PM2.5 under main synoptic conditions by GEOS-Chem model simulations. They found a substantial contribution of transportation in two synoptic patterns (SW-type and NW-type) and a dominated contribution of local emission sources in other two synoptic conditions (A-type and C-type). These results provide an opportunity to effectively mitigate haze pollution by local emission control actions in coordination with regional collaborative actions according to different synoptic patterns. The topic is of practical significance and the results are reliable. I would suggest for publication after addressing my comments below.

We thank the reviewer for comments, which have been incorporated to improve the manuscript.

1. The present comparison and verification of control simulation results in GEOS-Chem is not enough. It can be further verified by using PM2.5 observation data in a larger region of China or component observations of PM2.5 at some specific sites.

We thank the referee for his/her reading of our manuscript. The comments and suggestions are valuable for us to improve our work.

In order to better evaluate the GEOS-Chem model performances, the spatial distribution of $PM_{2.5}$ concentrations averaged over the four typical heavy pollution processes simulated by the control (CON) simulation are compared with the observations (a total of 633 sites) from Ministry of Ecology and Environment of China (http://www.mee.gov.cn/) (revised Fig. 6). Similar to the underestimation in $PM_{2.5}$ at Jingzhou, the underestimation is on a national scale when compared with the MEE observations, with a bias of -29.3 μg/m^3, -18.7 μg/m^3, -39.0 μg/m^3 and -21.4 μg/m^3 on average for SW-type, NW-type, A-type and C-type synoptic pattern, respectively (Fig. 6).

We have no observations of the chemical compositions of $PM_{2.5}$. In order to examine the model performances in the $PM_{2.5}$ chemical compositions, we have added Table 4 to review the reported concentrations of $PM_{2.5}$ and the three inorganic salts (sulfate, nitrate and ammonium) in other cities. The contributions of sulfate, nitrate and ammonium are 9.1%-31.9%, 5.7%-32.1% and 5.9%-13.3%, respectively. In the CON simulation, the fractions of each inorganic salt to $PM_{2.5}$ for these four typical heavy pollution processes are shown in revised Fig. S10, which are comparable to the previous observed results (Table 4).

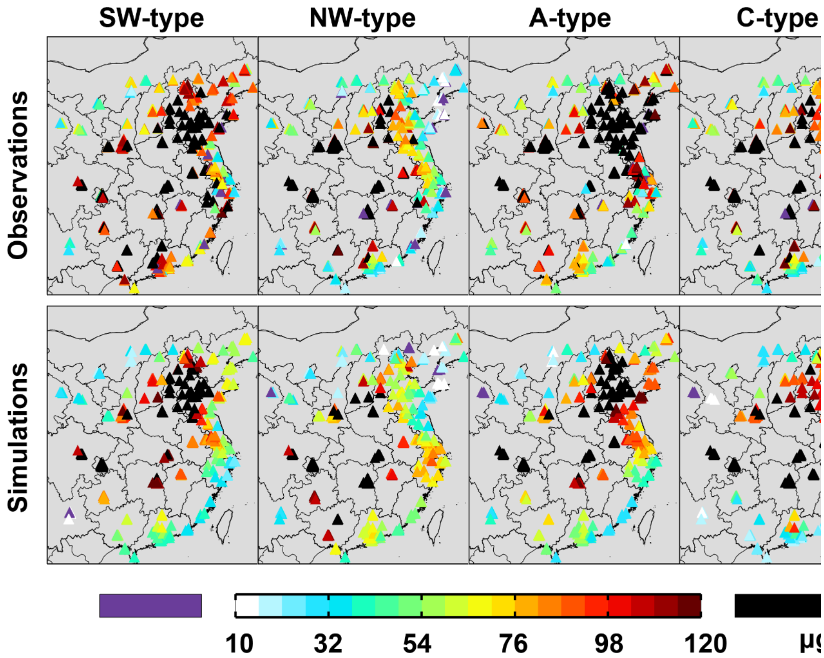

Figure 6 Spatial distribution of observed (top row) and modeled (bottom row, by CON case) $PM_{2.5}$ concentrations (μg/m^3) averaged over four severe pollution episodes controlled by SW-type (first column), NW-type (second column), A-type (third column) and C-type (forth column) synoptic pattern, respectively.

Table 4 The reported concentrations of $PM_{2.5}$ and the three inorganic salts (sulfate, nitrate and ammonium, μg/m^3) in other cities.

References	Site	Time	$PM_{2.5}$	Sulfate	Nitrate	Ammonium
Cao et al., 2012	Beijing	01/03	115.6±46.6	20.0±4.2 (17.3%)	13.1±4.5 (11.3%)	9.4±4.1 (8.1%)

Cao et al., 2012	Qingdao	01/03	134.8±43.0	21.1±7.7 (15.7%)	19.3±9.2 (14.3%)	15.3±5.2 (11.4%)
Cao et al., 2012	Tianjin	01/03	203.1±76.2	32.5±15.1 (16.0%)	25.2±10.3 (12.4%)	22.2±9.8 (10.9%)
Cao et al., 2012	Xi'an	01/03	356.3±118.4	53.8±25.6 (15.1%)	29.0±10.0 (8.1%)	29.8±11.5 (8.4%)
Cao et al., 2012	Chongqing	01/03	316.6±101.2	60.9±19.6 (19.2%)	18.1±6.4 (5.7%)	28.8±8.9 (9.1%)
Cao et al., 2012	Hangzhou	01/03	177.3±59.5	33.4±16.7 (18.8%)	25.7±14.8 (14.5%)	19.1±10.7 (10.8%)
Cao et al., 2012	Shanghai	01/03	139.4±50.6	21.6±12.3 (15.5%)	17.5±8.7 (12.6%)	14.5±5.9 (10.4%)
Cao et al., 2012	Wuhan	01/03	172.3±67.0	31.4±15.6 (18.2%)	22.2±10.7 (12.9%)	18.4±10.2 (10.7%)
Zhang et al., 2011	Xi'an	03/06-03/07	194.1	35.6 (18.3%)	16.4 (8.4%)	11.4 (5.9%)
Huang et al., 2012	Xi'an	01/06-02/06	235.8±125.1	44.8±31.3 (19.0%)	20.5±14.2 (8.7%)	14.5±10.8 (6.1%)
Wang et al., 2020	Jinan	10/17	104±54	14.4±9.2 (13.8%)	33.4±23.2 (32.1%)	13.0±8.3 (12.5%)

Wang et al., 2020	Shijiazhuang	10/17	152±109	19.3±19.6 (12.7%)	42.8±41.1 (28.2%)	18.2±17.1 (12.0%)
Wang et al., 2020	Wuhan	12/17	117±33	13.6±3.2 (11.6%)	26.6±11.1 (22.7%)	13.1±3.8 (11.2%)
Wang et al., 2016a	Zhengzhou	01/11-02/11	297±160	48±36 (16.2%)	31±19 (10.4%)	21±16 (7.1%)
Wang et al., 2016a	Zhengzhou	01/12-02/12	234±125	23±10 (9.8%)	22±9 (9.4%)	16±5 (6.8%)
Wang et al., 2016a	Zhengzhou	01/13-02/13	337±168	56±39 (16.6%)	39±20 (11.6%)	31±18 (9.2%)
Luo et al., 2018	Zibo	12/06-02/07	224.9±85.4	40.1±19.2 (17.9%)	18.1±9.0 (8.1%)	21.7±10.2 (9.7%)
Wang et al., 2016b	Shanghai	12/11, 12/12, 12/13	73.9±57.5	12.2±9.2 (16.5%)	14.6±12.2 (19.8%)	8.2±6.7 (11.1%)
Xu et al., 2019	Beijing	02/17-03/17	180.5	20.1 (11.1%)	45.6 (25.3%)	22.5 (12.5%)
Xu et al., 2019	Beijing	05/17-09/17	186.7	20.2 (10.8%)	32.4 (17.4%)	17.1 (9.2%)
Xu et al., 2019	Beijing	10/17-11/17	167.5	17.9 (10.7%)	44.5 (26.6%)	20.9 (12.5%)
Zheng et al., 2016	Beijing	03/10-05/10	65.2±65.1	11.1±10.1 (17.0%)	11.1±11.0 (17.0%)	6.8±6.7 (10.4%)

Zheng et al., 2016	Beijing	07/09-08/09	88.9±39.1	23.0±13.9 (25.9%)	16.2±11.8 (18.2%)	11.8±6.8 (13.3%)
Zheng et al., 2016	Beijing	12/09-02/10	84.0±66.6	8.1±8.3 (9.1%)	8.0±9.6 (9.0%)	5.9±7.1 (6.6%)
Zheng et al., 2016	Guangzhou	11/10	73.3±16.5	16.6±4.0 (22.6%)	5.7±3.8 (7.8%)	6.2±2.0 (8.5%)
Zheng et al., 2016	Shenzhen	12/09	64.6±24.7	20.6±3.5 (31.9%)	4.9±3.5 (7.6%)	4.6±1.0 (7.1%)
Zheng et al., 2016	Wuxi	04/10-05/10	82.1±27.0	12.8±3.8 (15.6%)	9.9±6.3 (12.1%)	7.0±2.0 (8.5%)
Zheng et al., 2016	Jinhua	10/11-11/11	81.9±26.2	18.3±6.7 (22.3%)	12.6±7.0 (15.4%)	10.4±4.1 (12.7%)
Liu et al., 2018	Chongqing	2012-2013	73.5±30.5	19.7±9.6 (26.8%)	6.5±6.2 (8.8%)	6.1±2.7 (8.3%)
Liu et al., 2018	Shanghai	2012-2013	68.4±20.3	13.6±6.4 (19.9%)	11.9±5.0 (17.4%)	5.8±2.1 (8.5%)
Liu et al., 2018	Beijing	2012-2013	71.7±36.0	11.9±8.2 (16.6%)	9.3±7.5 (13.0%)	5.3±2.7 (7.4%)

Reference:

Cao, J.-J., Shen, Z.-X., Chow, J. C., Watson, J. G., Lee, S.-C., Tie, X.-X., Ho, K.-F., Wang, G.-H., and Han, Y.-M.: Winter and Summer $PM_{2.5}$ Chemical Compositions in Fourteen Chinese Cities, Journal of the Air & Waste Management Association, 62, 1214-1226, 10.1080/10962247.2012.701193, 2012.

Huang, W., Cao, J., Tao, Y., Dai, L., Lu, S.-E., Hou, B., Wang, Z., and Zhu, T.: Seasonal Variation of Chemical Species Associated With Short-Term Mortality Effects of $PM_{2.5}$ in Xi'an, a Central City in China, American Journal of Epidemiology, 175, 556-566, 10.1093/aje/kwr342, 2012.

Liu, Z., Gao, W., Yu, Y., Hu, B., Xin, J., Sun, Y., Wang, L., Wang, G., Bi, X., Zhang, G., Xu, H., Cong, Z., He, J., Xu, J., and Wang, Y.: Characteristics of $PM_{2.5}$ mass concentrations and chemical species in urban and background areas of China: emerging results from the CARE-China network, Atmospheric Chemistry and Physics, 18, 8849-8871, 10.5194/acp-18-8849-2018, 2018.

Luo, Y., Zhou, X., Zhang, J., Xiao, Y., Wang, Z., Zhou, Y., and Wang, W.: $PM_{2.5}$ pollution in a petrochemical industry city of northern China: Seasonal variation and source apportionment, Atmospheric Research, 212, 285-295, 10.1016/j.atmosres.2018.05.029, 2018.

Wang, H. L., Qiao, L. P., Lou, S. R., Zhou, M., Ding, A. J., Huang, H. Y., Chen, J. M., Wang, Q., Tao, S., Chen, C. H., Li, L., and Huang, C.: Chemical composition of $PM_{2.5}$ and meteorological impact among three years in urban Shanghai, China, Journal of Cleaner Production, 112, 1302-1311, 10.1016/j.jclepro.2015.04.099, 2016a.

Wang, J., Li, X., Zhang, W., Jiang, N., Zhang, R., and Tang, X.: Secondary $PM_{2.5}$ in Zhengzhou, China: Chemical Species Based on Three Years of Observations, Aerosol and Air Quality Research, 16, 91-104, 10.4209/aaqr.2015.01.0007, 2016b.

Wang, Q., Fang, J., Shi, W., and Dong, X.: Distribution characteristics and policy-related improvements of $PM_{2.5}$ and its components in six Chinese cities, Environmental Pollution, 266, 10.1016/j.envpol.2020.115299, 2020.

Xu, Q., Wang, S., Jiang, J., Bhattarai, N., Li, X., Chang, X., Qiu, X., Zheng, M., Hua, Y., and Hao, J.: Nitrate dominates the chemical composition of $PM_{2.5}$ during haze event in Beijing, China, Science of the Total Environment, 689, 1293-1303, 10.1016/j.scitotenv.2019.06.294, 2019.

Zhang, T., Cao, J. J., Tie, X. X., Shen, Z. X., Liu, S. X., Ding, H., Han, Y. M., Wang, G. H., Ho, K. F., Qiang, J., and Li, W. T.: Water-soluble ions in atmospheric aerosols measured in Xi'an, China: Seasonal variations and sources, Atmospheric Research, 102, 110-119, 10.1016/j.atmosres.2011.06.014, 2011.

Zheng, J., Hu, M., Peng, J., Wu, Z., Kumar, P., Li, M., Wang, Y., and Guo, S.: Spatial distributions and chemical properties of $PM_{2.5}$ based on 21 field campaigns at 17 sites in China, Chemosphere, 159, 480-487, 10.1016/j.chemosphere.2016.06.032, 2016.

2. The novelty of this study need to be further clarified. New understanding or improvement of conclusion and application or in methods should be provided to reflect the general interests of the work rather than the local interests.

In Sect.1, we have further clarified and provided support that significant and new scientific merits are presented in this work. The $PM_{2.5}$ pollution in China has been continuously alleviating since 2013 as the implication of the Air Pollution Prevention and Control Action Plan. However, severe particle pollution still occurs frequently in autumn and winter, which is the major reason restricting the $PM_{2.5}$ to come up to national standard. Currently, how to effectively reduce emissions in autumn and winter is the key to mitigate haze pollution in China. Previous studies have highlighted that different levels of $PM_{2.5}$ pollutions are closely related to the dominant synoptic patterns in different regions, and they attribute the large spatial variability of pollution to the regional transport contributions, not only the different local sources of $PM_{2.5}$. Thus, heavy pollution prevention and control needs to consider the weather situation, otherwise local emission reduction measures would not work well. However, under different synoptic conditions, how to effectively reduce local and regional emissions to control haze pollution is rarely reported. In order to investigate the effectiveness of emission control to reduce $PM_{2.5}$ pollution under various potential synoptic controls, we take the severe particle pollution of winter haze episodes over Central China with transmission-pollution characteristics as an example. This study combines the atmospheric (circulation classification) and environmental (chemical transport modeling) research methods and could provide reference for emission control of severe winter haze pollution under different weather types, and provide basis for regional air quality policy-making.

3. Lines 105-109: several studies have investigated the potential effective emission reduction on ammonia, which should be reviewed here properly.

We have added the review of studies on potential efficiency of ammonia emission reduction in alleviating particulate pollution: "Moreover, current emission reduction policies in China mainly aimed at sulfur dioxide (SO_2) and nitrogen dioxide (NO_2), ignoring the effective emission reduction on ammonia (NH_3), although some modeling works have discussed the effectiveness of ammonia emission reduction for $PM_{2.5}$ mitigations (Liu et al., 2019; Ye et al., 2019; Xu et al., 2019; Bai et al., 2019)."

Bai, Z., Winiwarter, W., Klimont, Z., Velthof, G., Misselbrook, T., Zhao, Z., Jin, X., Oenema, O., Hu, C., and Ma, L.: Further Improvement of Air Quality in China Needs Clear Ammonia Mitigation Target, Environmental Science & Technology, 53, 10542-10544, 10.1021/acs.est.9b04725, 2019.

Liu, M., Huang, X., Song, Y., Tang, J., Cao, J., Zhang, X., Zhang, Q., Wang, S., Xu, T., Kang, L., Cai, X., Zhang, H., Yang, F., Wang, H., Yu, J. Z., Lau, A. K. H., He, L., Huang, X., Duan, L., Ding, A., Xue, L., Gao, J., Liu, B., and Zhu, T.: Ammonia emission control in China would mitigate haze pollution and nitrogen deposition, but worsen acid rain, Proceedings of the National Academy of Sciences of the United States of America, 116, 7760-7765, 10.1073/pnas.1814880116, 2019.

Xu, Z., Liu, M., Zhang, M., Song, Y., Wang, S., Zhang, L., Xu, T., Wang, T., Yan, C., Zhou, T., Sun, Y., Pan, Y., Hu, M., Zheng, M., and Zhu, T.: High efficiency of livestock ammonia emission controls in alleviating particulate nitrate during a severe winter haze episode in northern China, Atmospheric Chemistry and Physics, 19, 5605-5613, 10.5194/acp-19-5605-2019, 2019.

Ye, Z., Guo, X., Cheng, L., Cheng, S., Chen, D., Wang, W., and Liu, B.: Reducing $PM_{2.5}$ and secondary inorganic aerosols by agricultural ammonia emission mitigation within the Beijing-Tianjin-Hebei region, China, Atmospheric Environment, 219,

10.1016/j.atmosenv.2019.116989, 2019.

4. In Section 3.2, the mechanisms of heavy particle pollution caused by these four potential synoptic controls should be briefly discussed when describe characteristics of each synoptic pattern.

We have briefly discussed the mechanisms of heavy particle pollution caused by the four PSCs in the revised Section 3.2, when describe characteristics of each synoptic pattern:

"SW-type circulation is the predominant PSC of severe $PM_{2.5}$ pollution episodes. The circulation at 500 hPa is relatively flat and the whole East Asia region is affected by the westerly flow (Fig. S6a). Westerly belt fluctuates greatly at 700 hPa and there are two ridges and a southwest trough in the middle latitudes of Asia (Fig. S7a). Jingzhou is located in the front of a trough, prevailing the weak southwest airflow. At 850 hPa, the cold high pressure center is formed in Xinjiang of China. Warm low pressure appears in the low latitude area and weak high pressure appears in the East China Sea (Fig. 3a). In combination with the surface field, a high-low-high saddle like field forms from west to east (Fig. 4a). Such synoptic type is also the dominant weather system of eastern China (Shu et al., 2017; Yang et al., 2018). Jingzhou is located in the back of Bohai-northeast high pressure and the front of southwest warm low pressure. Thus it is affected by the southerly airflow, which could be conducive to the transport of air pollutants formed over southern China to Central China. Associated with small local surface wind speed (< 3 m/s) at Jingzhou, the dispersion of local and transported pollutants is inhibited.

NW-type circulation mainly occurs in the early winter (December and January). This synoptic pattern is also reported as one of the main types to affect the aerosol distributions over eastern China (Zheng et al., 2015). Circulation at 500 hPa is controlled by one trough and one ridge, with the weak ridge located in the northwest of China and the shallow trough located in the northeast of China (Fig. S6b). The whole East Asia is affected by the westerly current. The trough and ridge at 700 hPa are deepened. Jingzhou is located at the bottom of the shallow trough, prevailing the west-northwest airflow, affected by the flow around the plateau (Fig. S7b). At 850 hPa, the cold high pressure center is formed in Xinjiang, and Jingzhou is affected by the northerly airflow, due to being in the front of the high pressure (Fig. 3b). For the sea level pressure, the cold high pressure is located in the west of Mongolia and Xinjiang of China (Fig. 4b). Jingzhou is located at the region with weak fluctuation in the front of the high pressure, and the surface wind speed is smaller than 2 m/s. The haze episodes induced by NW-type synoptic pattern is similar to the transmission-accumulation pollution caused by SW-type, but the transmission path is from Northern China to Central China.

A-type circulation also mainly occurs in the early winter. The high-altitude circulation field is controlled by one trough and one ridge (Fig. S6c and S7c). East Asia is affected by west-northwest air flow, and the SLP is controlled by a huge high pressure, with the center located in the southwest of Baikal Lake (Fig. 4c). A surface high pressure favors the accumulation of air pollutants, especially over the regions of high pressure center (Leung et al., 2018). Jingzhou is in the sparse pressure field in front of the high pressure (Fig. 3c and 4c), with an average surface wind speed of ~1.3 m/s. The uniform west-northwest air flow at high altitude would lead to the low water vapor content and less cloud amount, which is conducive to radiation cooling at night. In addition, due to the weak high pressure ridge in the north, it is not conducive to the eastward and southward movement of cold air, leading to the stable weather situation and thus severe haze pollution at Jingzhou. This type is also responsible for most of the severe particulate pollution days in the BTH and YRD regions (Li et al., 2019).

C-type circulation mainly occurs in late winter and early spring, when the relative humidity is large with an average value of 74%. East Asia is controlled by the straight westerly flow, and the southwest shallow trough is obvious at 500 hPa (Fig. S6d). Additionally, the West Pacific subtropical high extends to the west, Central China is affected by the southwest flow. Southwest trough is deepened at 700 hPa, and Jingzhou is located in front of the trough and controlled by the southwest airflow (Fig. S7d). High pressure at the south of Xinjiang and the north of Plateau is strengthened at 850 hPa, and the southwest low pressure center is formed (Fig. 3d). Jingzhou is located in the low pressure system on the SLP field (Fig. 4d), with small surface wind speed (0-3 m/s). Together with the large relative humidity, which can promote the hygroscopic growth of particulate matter (Twohy et al., 2009; Zheng et al., 2015), the haze pollution is persistent and serious at Jingzhou. The impact of low-pressure systems on winter heavy air pollution have also been reported in the northwest Sichuan Basin (Ning et al., 2018)."

5. Lines 294-296: Why the four pollution episodes are selected?

We have explained in the revised sentences: "In order to reduce the simulation cost, the continuous four severe haze episodes occurred during November, 2013-February, 2014 are selected. These four haze episodes are controlled by the synoptic pattern of SW-type (18-25 November, 2013), NW-type (19-26 December, 2013), A-type (14-21 January, 2014) and C-type (26 January - 2 February, 2014), respectively."

6. Lines 304-308: The model control simulation is compared to PM2.5 observations at just one site (Jingzhou). Current comparison is insufficient to demonstrate the modeling performance.

In order to better evaluate the GEOS-Chem model performances, the spatial distribution of $PM_{2.5}$ concentrations averaged over the four typical heavy pollution processes simulated by the control (CON) simulation are compared with the observations (a total of 633 sites) from Ministry of Ecology and Environment of China (http://www.mee.gov.cn/) (revised Fig. 6). Please see details in the response of comment#1.

7. Line 308-311: Model biases are generally attributed to resolution, emission errors, meteorology and chemical mechanism without statistical results of further sensitivity simulations. Be careful to discuss the model deviation.

In order to explain the causes of the model discrepancy, we have added Table S3 to show the observed (modeled) meteorological conditions averaged over these four pollution episodes controlled by SW-type, NW-type, A-type and C-type synoptic pattern, respectively. There is an overestimate in temperature and wind speed and an underestimate in humidity, which can partly contribute to the underestimation of modeled $PM_{2.5}$ concentrations. In addition, anthropogenic emissions for $PM_{2.5}$ precursors used here are for the year 2017 over Central China from SEEA inventory (revised Table S4). From 2013 to 2017, anthropogenic NO_x, SO_2, and primary $PM_{2.5}$ emissions in Central China have declined substantially (revised Table S4), due to the implementation of stringent emission control measures for the 12th-13th Five-Year Plans (Zheng et al., 2018). The anthropogenic emissions biases may affect our simulations and $PM_{2.5}$ attribution results to some extent. Additionally, the underestimation is on a national scale when compared with the MEE observations, with a bias of -29.3 μg/m^3, -18.7 μg/m^3, -39.0 μg/m^3 and -21.4 μg/m^3 on average for SW-type, NW-type, A-type and C-type synoptic pattern, respectively (revised Fig. 6, see figure in the response of comment#1). The national negative biases may be also attributed to insufficient resolution of the model (Yan et al., 2014) and imperfect chemical mechanisms (Yan et al., 2019).

Table S3. The observed (modeled) meteorological conditions at Jingzhou averaged over these four pollution episodes controlled by SW-type, NW-type, A-type and C-type synoptic pattern, respectively.

PSC	Temperature (°C)	Humidity (%)	Pressure (kpa)	Wind speed (m/s)
SW	11.79 (12.96)	75.33 (69.25)	1018.33 (1024.06)	2.13 (3.09)
NW	3.61 (6.34)	71.16 (62.78)	1027.53 (1031.53)	1.44 (2.45)
A	5.81 (7.52)	64.96 (60.38)	1026.63 (1028.66)	1.45 (2.27)
C	9.60 (13.08)	78.10 (71.40)	1011.48 (1014.24)	1.88 (3.11)

Table S4. The emission amount of $PM_{2.5}$ precursors over Central China calculated from SEEA (for the year 2017) and MEIC (for the years of 2013, 2014 and 2017) inventory (unit: 10^4 ton).

Category	SO_2	NO_X	NH_3	$PM_{2.5}$	CO	BC	OC	VOCs
SEEA (2017)	48.4	94.0	54.6	26.4	553.8	6.2	12.9	117.2
MEIC (2017)	52.0	70.4	57.5	35.2	629.2	6.8	11.7	116.4
MEIC (2013)	173.3	98.4	62.4	54.5	836.5	9.2	16.7	116.6
MEIC (2014)	97.0	80.0	61.1	46.8	744.2	8.3	15.3	116.4

8. Line 337: PSC -> PSCs

Modified.

9. Line 359: The transportation of air pollutants from the south makes the proportion of the three inorganic salts (45.7%) in Jingzhou area the smallest. Consider revising it like: The transport of air pollutants from the south leads to the smallest proportion of the three inorganic salts (45.7%) in Jingzhou.

Modified.

10. Line 482: remove potential synoptic controls or (PSC)

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11. Line 494: contribute 82%/85% of PM2.5. Consider revising it like: dominate the contribution (82%/85%) to PM2.5.

Modified.

Effectiveness of emission control to reduce $PM_{2.5}$ pollution of Central China during winter haze episodes under various potential synoptic controls

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Abstract

Currently mitigating the severe particle pollution in autumn and winter is the key to further improve the air quality of China. The source contributions and transboundary transport of fine particles ($PM_{2.5}$) in pollution episodes are closely related to large-scale or synoptic-scale atmospheric circulation. Under different synoptic conditions, how to effectively reduce emissions to control haze pollution is rarely reported. In this study, we classify the synoptic conditions over Central China from 2013 to 2018 by using

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Lamb-Jenkension method and the NCEP/NCAR FNL operational global analysis data. The effectiveness of emission control to reduce $PM_{2.5}$ pollution during winter haze episodes under potential synoptic controls is simulated by GEOS-Chem model. Among the ten identified synoptic patterns, four types account for 87% of the total pollution days. Two typical synoptic modes of them are characterized by small surface wind speed and stable weather conditions/high relative humidity (A/C-type) over Central China due to a high-pressure system/a southwest trough low-pressure system, blocking pollutants dispersion. Sensitivity simulations show that these two heavy pollution processes are mainly contributed by local emission sources with ~82% for A-type and ~85% for C-type, respectively. The other two patterns lead to pollution of transport characteristics affected by northerly/southerly winds (NW/SW-type), carrying air pollution from northern/southern China to Central China. The contribution of pollution transmission from North/South China is 36.9%/7.6% of $PM_{2.5}$ and local emission sources contribute 41%/69%. We also estimate the effectiveness of emission reduction in these four typical severe pollution synoptic processes. By only reducing SO_2 and NO_x emission and not controlling NH_3, the enhanced nitrate counteracts the effect of sulfate reduction on $PM_{2.5}$ mitigations, with less than 4% decrease in $PM_{2.5}$. In addition, to effectively mitigate haze pollution of NW/SW-type synoptic controlled episodes, local emission control actions should be in coordination with regional collaborative actions.

1 Introduction

The regional pollution of fine particles ($PM_{2.5}$) has attracted worldwide attention in the public and in the scientific community (Cheng et al., 2016; Li et al., 2017c; Lin et al., 2018; Bi et al., 2019) due to its detrimental effect on visibility (Wang et al., 2020) and public health (Agarwal et al., 2017; Zhang et al., 2017). The $PM_{2.5}$ pollution in China has been continuously alleviating since 2013 as the implication of the Air Pollution Prevention and Control Action Plan (Zheng et al., 2018; Zhang et al., 2019),

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especially in the Beijing-Tianjin-Hebei region (BTH) (Li et al., 2017b; Cheng et al., 2019), the Yangtze River Delta (YRD) region and the Pearl River Delta (PRD) region. However, severe particle pollution still occurs frequently in autumn and winter, which is the major reason restricting the $PM_{2.5}$ to come up to national standard. For example, 12 extremely severe and persistent $PM_{2.5}$ pollution episodes occurred in Beijing in January 2013, February 2014, December 2015, December 2016 and January 2017 (Zhong et al., 2018; Sun et al., 2016; Wang et al., 2018). Currently, how to effectively reduce emissions in autumn and winter is the key to mitigate haze pollution in China.

The contribution of emission sources has been widely recognized as the decisive factor of $PM_{2.5}$ pollution over urban agglomerations, including industrial exhaust, urban transportation, residential emission, power plants, agricultural activities, and bio-combustion (Huang et al., 2014; Tian et al., 2016; Wu et al., 2018; An et al., 2019). While the outbreak, persistence and dissipation of particle pollution generally depends on the meteorological conditions and regional synoptic patterns, controlled by the large-scale or synoptic-scale atmospheric circulation (Chuang et al., 2008; Zhang et al., 2012; Russo et al., 2014; Zheng et al., 2015; Shu et al., 2017; Li et al., 2019).

Many studies have tried to reveal the relationship between synoptic patterns and severe particle pollution, and estimate the meteorological contributions to these pollution episodes. The YRD is mainly affected by pollutants transmitted from the northern and the southern China when the East Asian major trough is located at its front (Liao et al., 2017; Shu et al., 2017; Li et al., 2019). Liao et al. (2020) has confirmed that the relative position of the PRD to high-pressure systems imposes significant impacts on the diffusion conditions and the $PM_{2.5}$ distributions in the PRD region. For North China Plain (NCP), high frequency of stagnant weather accompanied by small pressure gradient and near-surface wind speed, and shallow mixing layer are major reasons of aerosol pollution over this region in winter (He et al., 2018). The aerosol pollution formation process in Sichuan Basin is often controlled by the large scale high-pressure circulation at sea level (Sun et al., 2020). In the Guanzhong basin, pollution event is

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generally governed by both the large-scale synoptic situation and the small-scale local circulation. The downhill wind not only forms a convergence zone in the basin, but also makes pollutants flow back from the mountain region to the basin (Bei et al., 2017). Leung et al. (2018) also find strong correlations of daily $PM_{2.5}$ variability with several synoptic patterns, including monsoon flows and cold front channels in northern China related to the Siberian High, onshore flows in eastern China, and frontal rainstorms in southern China. These previous studies have highlighted that different levels of $PM_{2.5}$ pollutions are closely related to the dominant synoptic patterns in different regions, and they attribute the large spatial variability of pollution to the regional transport contributions, not only the different local sources of $PM_{2.5}$. Thus, heavy pollution prevention and control needs to consider the weather situation, otherwise local emission reduction measures would not work well. However, under different synoptic conditions, how to effectively reduce local and regional emissions to control haze pollution is rarely reported.

Various key regions have issued the emergency preplan against the winter haze episodes, while these schemes can only be targeted at a certain city (The People's Government of Beijing Municipality, 2018; The People's Government of Shanghai Municipality, 2018) or a certain urban agglomeration (The People's Government of Guangdong Province, 2014). Although there are many studies targeted $PM_{2.5}$ mitigations at a regional scale (Ding et al., 2019; Zhang et al., 2019, Xing et al., 2018, 2019; Fu et al., 2017; etc.), their results can not be directly applied to reduce winter $PM_{2.5}$ pollution under various synoptic controls. Moreover, current emission reduction policies in China mainly aimed at sulfur dioxide (SO_2) and nitrogen dioxide (NO_2), ignoring the effective emission reduction on ammonia (NH_3), although some modeling works have discussed the effectiveness of ammonia emission reduction for $PM_{2.5}$ mitigations (Liu et al., 2019; Ye et al., 2019; Xu et al., 2019; Bai et al., 2019). Compared to remarkable reduction in SO_2, NO_2, and primary PM emissions, NH_3 emissions has remained stable during 2014–2018 in China (Zheng et al., 2018). In addition, given the

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important roles of other $PM_{2.5}$ precursors (eg., NMVOCs) in aerosol formation (Geng et al., 2019), cutting non-SO_2-NO_2-PM emissions should be proposed as a next-step mitigation strategy. Therefore, for $PM_{2.5}$ mitigations in a specific region during winter haze episodes forced by various synoptic conditions, whether the air pollution emergency management and control schemes are effective and how to improve them have become an urgent scientific question to be answered.

In order to investigate the effectiveness of emission control to reduce $PM_{2.5}$ pollution during winter haze episodes under various potential synoptic controls (PSCs), we take the severe particle pollution of winter haze episodes over Jingzhou, the hinterland of Yangtze River middle basin in Central China, as an example. Central China is geographically surrounded by major haze pollution regions, the SCB to the west, the PRD to the south, the YRD to the east, and the NCP to the north (Fig. 1). As a regional pollutant transport hub with sub basin topography, Central China is a region of transmission-pollution characteristics affected by two reported transport pathways from the vast flatland in central eastern China (Yu et al., 2020) and from the NCP region (Zheng et al., 2019a). In combination with high anthropogenic emissions (Wu et al., 2018) and secondary aerosol formation (Huang et al., 2020), Central China often suffers severe pollution episodes in winter caused by $PM_{2.5}$ (Gong et al., 2015; Xu et al., 2017). In this study, we conduct the circulation classification to differentiate the synoptic modes during the severe particle pollution episodes in winter over Central China from 2013 to 2018 by using Lamb-Jenkension method. Then we simulate the $PM_{2.5}$ chemical components, and the contributions of local sources as well as transboundary transport of $PM_{2.5}$ under different synoptic conditions. Finally, the effectiveness of emission reduction in main potential synoptic patterns are evaluated by GEOS-Chem model simulations. This study combines the atmospheric (circulation classification) and environmental (chemical transport modeling) research methods and could provide reference for emission control of severe winter haze pollution under different weather types, and provide basis for regional air quality policy-making.

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2 Data and Methods

2.1 Data

Hourly mass concentrations of $PM_{2.5}$ at Jingzhou (112.18°E, 30.33°N, 33.7 m) from November 2013 to December 2018 are obtained from Hubei Environmental Monitoring Central Station (http://sthjt.hubei.gov.cn/). We screen the pollution days with daily mean $PM_{2.5}$ concentrations larger than 150 μg/m^3 for circulation classification.

Figure 1

We use the daily mean sea level pressure (SLP) between 2013 and 2018 from the National Centers for Environmental Prediction/National Center for Atmospheric Research (NCEP/NCAR) Final (FNL) Operational Global Analysis data (horizontal resolution: 1° × 1°; temporal resolution: 6 hours; https://rda.ucar.edu/datasets/ds083.3/) to conduct the classification of Lamb-Jenkension circulation types.

The meteorological data of surface observations at Jingzhou, including ambient temperature, relative humidity, wind speed, wind direction and atmospheric pressure, are obtained from Hubei Meteorological Information and Technology Support Center (http://hb.cma.gov.cn/qxfw/index.html). The data from November 2013 to February 2014 are used to analyze the meteorological characteristics during the period four severe particle pollution events occurred in succession over Central China (Fig. S1).

In order to better evaluate the GEOS-Chem model performances, we also use the $PM_{2.5}$ observations (a total of 633 sites; from November 2013 to February 2014) from Ministry of Ecology and Environment of China (MEE, http://www.mee.gov.cn/) to conduct the model-observation comparison.

2.2 Lamb-Jenkension Circulation Classification

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Moved up [1]: We use the daily mean sea level pressures (SLP) from the National Centers for Environmental Prediction/National Center for Atmospheric Research (NCEP/NCAR) Final (FNL) Operational Global Analysis data (horizontal resolution: 1° × 1°; temporal resolution: 6 hours; https://rda.ucar.edu/datasets/ds083.3/) to conduct the classification of Lamb-Jenkension circulation types.

The atmospheric circulation classification adopts the Lamb-Jenkension method proposed by Lamb et al. (1950) and developed by Jenkension et al. (1977). Compared to the objective classification method PCA used in some studies (Chang and Zhan, 2017, Dai et al., 2021), this Lamb-Jenkension method is a combination of subjective and objective methods. After the objective judgment of the circulation, we also make subjective considerations to overcome the weaknesses of their respective, leading to better synoptic significance. Many works of circulation classification have used the Lamb-Jenkension method and reported that the analysis can well respond to the classification results (Philipp et al., 2016;Santurtun et al., 2015;Pope et al., 2015;Russo et al., 2014;Pope et al., 2014;Trigo and DaCamara, 2000).

To calculate the circulation types of Jingzhou, we mark total 16 points (97.5°E-127.5°E, 20°N-40°N) by every 10 longitudes and 5 latitudes and the center point located at 112.5° E and 30° N (Fig. S2). Using the sea level pressure of 16 points, we calculate six circulation indexes by scheme of central difference:

$$u = 0.5[P(12) + P(13) - P(4) - P(5)]$$

(1)

$$v = \frac{1}{\cos\alpha} \times \frac{1}{4}[P(4) + 2P(9) + P(13) - P(4) - 2P(8) - P(12)]$$

(2)

$$V = \sqrt{u^2 + v^2}$$

(3)

$$\xi_u = \frac{\sin\alpha}{2\sin\alpha_1}[P(15) + P(16) - P(8) - P(9)] - \frac{\sin\alpha}{2\sin\alpha_2}[P(8) + P(9) - P(1) - P(2)]$$

(4)

$$\xi_v = \frac{1}{8\cos^2\alpha}[P(6) + 2P(10) + P(14) - P(5) - 2P(9) - P(13) + P(3) + 2P(7) + P(11) - P(4) - 2P(8) - P(12)]$$

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$$\xi = \xi_u + \xi_v \tag{6}$$

Where $P(n)(n = 1, 2, 3 \cdots 16)$ is the sea level pressure at the n[th] point; α, α_1 and α_2 are the latitude values of points C, A_1 and A_2, respectively; V is the geostrophic wind, u and v are the latitudinal and meridional components of the geostrophic wind; ξ is the geostrophic vorticity; ζ_u is the u meridional gradient, and ζ_v is the v latitudinal gradient.

Taking the latitude of the center point as the reference frame, the unit of six circulation indexes is $hPa/(10^o lon)$, the direction of geostrophic wind can be determined by u and v, and cyclones and anticyclones can be determined by ξ. According to the geostrophic wind speed, wind direction and vorticity value, the circulation is divided into 10 types. The classification standard and corresponding types are shown in Table 1.

Table 1

2.3 GEOS-Chem simulations

The GEOS-Chem chemistry transport model is used (http://acmg.seas.harvard.edu/geos/) to simulate the spatiotemporal distribution of $PM_{2.5}$. The nested model, covering China (70°E-140°E, 15°S-55°N), is run with a horizontal resolution of 0.25° latitude × 0.3125° longitude and 72 vertical layers. The boundary condition of nested model is provided by the GEOS-Chem global model with a horizontal resolution of 2° latitude × 2.5° longitude (Fig. S3). Both global and nested simulations, driven by the GEOS-FP assimilated meteorological data, include detailed tropospheric Ozone-NO_x-VOCs-HO_x-aerosol chemistry. More details are shown in Yan et al. (2019). In the models, anthropogenic and natural sources are fully considered

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in GEOS-Chem. Table S1 and Table S2 show a list of emission inventories in the global model and nested simulation, respectively. In China, the monthly grid data of 0.25° × 0.25° from MEIC inventory (http://meicmodel.org) for CO, NO_x, SO_2 and non-methane volatile organic compounds (NMVOCs) in 2013-2014 is used. Over Central China, anthropogenic sources of these species are from our group SEEA (Source Emission and Environment Research) inventory with the grid data of 0.1° × 0.1° (not shown). The SEEA emission inventory was developed based on the year of 2017 for the Wuhan city cluster and it has been successfully adopted for the air quality simulating and forecasting of 7th CISM Military World Games in 2019. Other emission descriptions are shown in Supplementary Sect. S1.

In order to better simulate the spatiotemporal distribution of $PM_{2.5}$ over Central China, especially in winter heavy pollution periods, the standard v11-01 of GEOS-Chem is optimized according to the actual situation in China (see details in Supplementary Sect. S2), including optimizing $PM_{2.5}$ sources and increasing the proportion of sulfate primary emission (Yan et al., 2020). The $PM_{2.5}$ primary anthropogenic emissions enhance the $PM_{2.5}$ concentrations over Central China by 5-20 μg/m^3 in winter (Fig. S4). Compared with the results before the model optimization (Fig. S5), the sulfate concentration simulated by the optimized model increased from 10-20 μg/m^3 to 30-50 μg/m^3. Further comparisons of $PM_{2.5}$ with observations and inorganic salts (sulfate, nitrate and ammonium) with reported values from previous studies are shown in Sect.3.3.

3. Results and Discussion

3.1 Classification of PSCs

As shown in Fig. 2, among the circulation patterns of pollution-day at Jingzhou from 2013 to 2018, the frequency of SW-type circulation is the highest, accounting for 29% of the total pollution days. The frequencies of NW-type, A-type and C-type are also high, accounting for 27%, 19% and 12%, respectively. While the other six

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circulation patterns are less occurred, with the frequencies less than 5%. Thus, the above four typical circulation types are considered as the main potential synoptic controls of the severe particle pollution episodes over Central China.

Figure 2

3.2 Characteristics of the four main PSCs

SW-type circulation is the predominant PSC of severe $PM_{2.5}$ pollution episodes. The circulation at 500 hPa is relatively flat and the whole East Asia region is affected by the westerly flow (Fig. S6a). Westerly belt fluctuates greatly at 700 hPa and there are two ridges and a southwest trough in the middle latitudes of Asia (Fig. S7a). Jingzhou is located in the front of a trough, prevailing the weak southwest airflow. At 850 hPa, the cold high pressure center is formed in Xinjiang of China. Warm low pressure appears in the low latitude area and weak high pressure appears in the East China Sea (Fig. 3a). In combination with the surface field, a high-low-high saddle like field forms from west to east (Fig. 4a). Such synoptic type is also the dominant weather system of eastern China (Shu et al., 2017; Yang et al., 2018). Jingzhou is located in the back of Bohai-northeast high pressure and the front of southwest warm low pressure. Thus, it is affected by the southerly airflow, which could be conducive to the transport of air pollutants formed over southern China to Central China. Associated with small local surface wind speed (< 3 m/s) at Jingzhou, the dispersion of local and transported pollutants is inhibited.

Figure 3

NW-type circulation mainly occurs in the early winter (December and January). This synoptic pattern is also reported as one of the main types to affect the aerosol distributions over eastern China (Zheng et al., 2015). Circulation at 500 hPa is

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controlled by one trough and one ridge, with the weak ridge located in the northwest of China and the shallow trough located in the northeast of China (Fig. S6b). The whole East Asia is affected by the westerly current. The trough and ridge at 700 hPa are deepened. Jingzhou is located at the bottom of the shallow trough, prevailing the west-northwest airflow, affected by the flow around the plateau (Fig. S7b). At 850 hPa, the cold high pressure center is formed in Xinjiang, and Jingzhou is affected by the northerly airflow, due to being in the front of the high pressure (Fig. 3b). For the sea level pressure, the cold high pressure is located in the west of Mongolia and Xinjiang of China (Fig. 4b). Jingzhou is located at the region with weak fluctuation in the front of the high pressure, and the surface wind speed is smaller than 2 m/s. The haze episodes induced by NW-type synoptic pattern is similar to the transmission-accumulation pollution caused by SW-type, but the transmission path is from Northern China to Central China.

Figure 4

A-type circulation also mainly occurs in the early winter. The high-altitude circulation field is controlled by one trough and one ridge (Fig. S6c and S7c). East Asia is affected by west-northwest air flow, and the SLP is controlled by a huge high pressure, with the center located in the southwest of Baikal Lake (Fig. 4c). A surface high pressure favors accumulation of air pollutants, especially over the regions of high pressure center (Leung et al., 2018). Jingzhou is in the sparse pressure field in front of the high pressure (Fig. 3c and 4c), with an average surface wind speed of ~1.3 m/s. The uniform west-northwest air flow at high altitude would lead to the low water vapor content and less cloud amount, which is conducive to radiation cooling at night. In addition, due to the weak high pressure ridge in the north, it is not conducive to the eastward and southward movement of cold air, leading to the stable weather situation

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and thus severe haze pollution at Jingzhou. This type is also responsible for most of the severe particulate pollution days in the BTH and YRD regions (Li et al., 2019).

C-type circulation mainly occurs in late winter and early spring, when the relative humidity is large with an average value of 74%. East Asia is controlled by the straight westerly flow, and the southwest shallow trough is obvious at 500 hPa (Fig. S6d). Additionally, the West Pacific subtropical high extends to the west, Central China is affected by the southwest flow. Southwest trough is deepened at 700 hPa, and Jingzhou is located in front of the trough and controlled by the southwest airflow (Fig. S7d). High pressure at the south of Xinjiang and the north of Plateau is strengthened at 850 hPa, and the southwest low pressure center is formed (Fig. 3d). Jingzhou is located in the low pressure system on the SLP field (Fig. 4d), with small surface wind speed (0-3 m/s). Together with the large relative humidity, which can promote the hygroscopic growth of particulate matter (Twohy et al., 2009; Zheng et al., 2015), the haze pollution is persistent and serious at Jingzhou. The impact of low-pressure systems on winter heavy air pollution have also been reported in the northwest Sichuan Basin (Ning et al., 2018).

3.3 $PM_{2.5}$ and chemical components under the four main PSCs in control simulations

The spatiotemporal distribution of $PM_{2.5}$ and its components under the four typical synoptic controls over Central China were simulated by optimized GEOS-Chem model. In order to reduce the simulation cost, the continuous four severe haze episodes occurred during November, 2013-February, 2014 are selected. These four haze episodes are controlled by the synoptic pattern of SW-type (18-25 November, 2013), NW-type (19-26 December, 2013), A-type (14-21 January, 2014) and C-type (26 January - 2 February, 2014), respectively. The air quality at Jingzhou during the four pollution episodes is between grade 5 ($PM_{2.5}$ > 150 μg/m^3) and grade 6 heavy pollution ($PM_{2.5}$ > 250 μg/m^3, as Fig. 5a and S1a shown). The simulation time is started at

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November 1st, 2013, with the first two weeks used as spin up to eliminate the impact of initial conditions.

Figure 5

Figure 6

The daily/hourly mean $PM_{2.5}$ concentrations at Jingzhou in the four typical heavy pollution processes simulated by the control (CON) simulation (Table 2) are compared with the observations (Fig. 5a/Fig. S1a). The model underestimates the observed $PM_{2.5}$ concentrations (by 43.3 μg/m^3 on average), especially in the high $PM_{2.5}$ periods (by 116.8 μg/m^3 at the maximum occurring in November 21-23, 2013). The possible causes for underestimation are meteorological field deviations (an overestimate in temperature and wind speed and an underestimate in humidity; Table S3) and emission errors. Anthropogenic emissions for $PM_{2.5}$ precursors used here are for the year 2017 over Central China from SEEA inventory (Table S4). From 2013 to 2017, anthropogenic NO_x, SO_2, and primary $PM_{2.5}$ emissions in Central China have declined substantially (Table S4), due to implementation of stringent emission control measures for the 12th-13th Five-Year Plans (Zheng et al., 2018). The anthropogenic emissions biases may affect our simulations and $PM_{2.5}$ attribution results to some extent. Aditionally, the underestimation is on a national scale when compared with the MEE observations, with a bias of -29.3 μg/m^3, -18.7 μg/m^3, -39.0 μg/m^3 and -21.4 μg/m^3 on average for SW-type, NW-type, A-type and C-type synoptic controlled episodes, respectively (Fig. 6). The national negative biases may be also attributed to insufficient resolution of the model (Yan et al., 2014) and imperfect chemical mechanisms (Yan et al., 2019). Nevertheless, the model can reproduce the evolution of each severe particle pollution episode well, including the accumulation of pollutants, the continuing process and the gradual dissipation of pollution (Fig. 5a/Fig. S1a).

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Table 2

In order to examine the model performances in the $PM_{2.5}$ chemical compositions, we have reviewed the reported concentrations of $PM_{2.5}$ and the three inorganic salts (sulfate, nitrate and ammonium) in other cities (Table 4). The contributions of sulfate, nitrate and ammonium are 9.1%-31.9%, 5.7%-32.1% and 5.9%-13.3%, respectively. Figure S8/S9 shows the modeled spatial distribution of $PM_{2.5}$, sulfate, nitrate and ammonium concentrations averaged in the four typical heavy pollution processes over Jingzhou/China. The fractions of each inorganic salt to $PM_{2.5}$ for these four heavy pollution episodes are also shown in Fig. S10. Over Central China, the main components of $PM_{2.5}$ are the three inorganic salts in these pollution episodes, with the averaged contributions of sulfate, nitrate and ammonium being ~20%, ~18% and ~13%, respectively (Table 3). Our modelling results are comparable to the previous observed results (Table 4). Huang et al. (2014) have also reported that the three secondary inorganic particles rank the highest fraction among the $PM_{2.5}$ species in Central-Eastern China. As shown in Table 3, in addition to inorganic salts, other chemical components include dust (~15%), black carbon (~7%), primary organic aerosol (~14%) and second organic aerosol (~13%). In these four pollution events, the differences in mass percentages of each chemical component ranged from 0.1% (dust) to 6.2% (sulfate) (Table 3). See details in Sect. 3.4 for further analysis of the causes for the differences.

Table 3

Table 4

3.4 Local emissions versus transmission contributions to $PM_{2.5}$ under the four main PSCs

In order to investigate the effectiveness of emission control to reduce $PM_{2.5}$ pollution of Central China in the four typical severe particle pollution episodes, firstly

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3.4 Local emissions versus tran

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we estimate the local sources versus transmission contributions of $PM_{2.5}$ by GEOS-Chem sensitivity simulations (Table 2). Results of XJ0 (Emissions outside Jingzhou are zero) indicates the contribution of local emission sources to the $PM_{2.5}$ pollution over Jingzhou. The difference between CON and XCC0 (Emissions outside Central China are zero) shows the transmission contribution of $PM_{2.5}$ outside Central China to Jingzhou. The difference between CON and NCP0/YRD0/PRD0/SCB0 (Emissions over NCP/YRD/PRD/SCB are zero) represents the contribution of pollution transport from NCP/YRD/PRD/SCB regions to Jingzhou.

Figure 7

For the SW-type synoptic situation, differences between the simulation results of NCP0/YRD0/SCB0 and CON show that pollution controlled by SW-type circulation over Central China is almost not affected by the emission sources from North China/East China/Sichuan Basin. The concentrations of $PM_{2.5}$ and three inorganic salts simulated by NCP0/YRD0/SCB0 are similar to those simulated by CON, with a difference less than 3.0% (Fig. 8). However, affected by the southerly airflow at 850 hPa (Fig. 7), air pollutants formed over southern China could be transmitted to Central China, with the transport contribution of 7.6%. In addition, the contributions from transboundary transport from non-Jingzhou Central China is simulated to be 12.0% by comparing the results of XJ0 and XCC0. The transport of air pollutants from the south leads to the smallest proportion of the three inorganic salts (45.7%) in Jingzhou among the four pollution episodes (50.3%-55.5% for other three episodes), because the emissions of SO_2, NO_2 and NH_3 in the south (especially in Guangxi and Guizhou province) are smaller than those in Central China (Li et al., 2017a). Associated with the small surface wind speed of 2.1 m/s on average (Fig. 5) and the weak ascending in the vertical direction (Fig. 7) at Jingzhou, it is not conducive to the dispersion of local pollutants (Zheng et al., 2015). The high $PM_{2.5}$ concentrations are mainly accumulated

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by local emissions. The simulations of XJ0 and CON show that local emission sources over Jingzhou contribute ~70% to $PM_{2.5}$.

Figure 8

Figure 9

For the NW-type synoptic mode, affected by the northerly airflow (Fig. 9), it is conducive to the southward movement of air pollutants in northern China (He et al., 2018; Leung et al., 2018). Influenced by the local and surrounding terrain over Central China (Fig. 1), two transmission channels are formed from north to south and from northeast to southwest (Fig. 9). In addition, due to the local small wind speed (1.4 m/s on average) near the ground (Fig. 5), the weak convection and the warm ridge along the East Asia coast (Fig. 9), the local and transported pollutants accumulate in Central China. The average concentration of $PM_{2.5}$ in Jingzhou is 179.4 μg/m^3. Due to the transport contribution of pollutants from northern China (with much higher anthropogenic emissions of SO_2, NO_2 and NH_3) (Li et al., 2017a), the total proportion of the three inorganic salts is the highest (55.5%). The $PM_{2.5}$ concentration simulated in NCP0 is 63.1% of that by CON simulation (Fig. 8), indicating that the transmission contribution from North China in this heavy pollution episode is as high as 36.9%. The contribution of local emission sources is much smaller than that of SW-type synoptic pattern, only 41.2% (comparison between XJ0 and CON).

Figure 10

Under the A-type circulation, Jingzhou is controlled by a high pressure system (Fig. 10) which can lead to stable weather conditions caused by radiation inversion (Guo et al., 2015) and subsidence inversion (Kurita et al., 1985), being favorable to continuous accumulation of local pollutants (Guo et al., 2015). The distribution of $PM_{2.5}$

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in China is similar to that of SW-type weather condition, with an averaged $PM_{2.5}$ concentration of 128.6 μg/m^3 over Central China. Unlike SW-type, the $PM_{2.5}$ at Jingzhou in this synoptic pattern is less affected by transboundary transport, with the total transport contribution of the surrounding four major pollution regions being less than 9%. The contribution of local emission sources is about 82% (Fig. 8).

Figure 11

Under the C-type synoptic pattern, the southwest low pressure center is formed at 850 hPa, and Jingzhou is located in the low pressure system of the SLP field (Fig. 11). In combination with the large relative humidity (78% on average; Fig. 5; because that the occurrence season of C-type is the late winter and early spring), it can promote the haze pollution due to its impact on hydrophilic aerosols (Twohy et al., 2009; Zheng et al., 2015). Together with the small wind speed (less than 4 m/s; Fig. 5), it is easy to cause the accumulation of pollutants. The average concentration of $PM_{2.5}$ over Central China is as high as 203.7 μg/m^3. Air pollution controlled by this weather condition is the most serious of the four typical synoptic controls. However, in this weather situation, pollutants in North China are easy to diffuse (Miao et al., 2017; Li et al., 2019), and the concentration of $PM_{2.5}$ is significantly lower than that in the former three weather situations (Fig. 11 and Fig. S9). The contribution of pollution transport from non-Central China region simulated by GEOS-Chem is less than 8%, and the contribution of local emission sources at Jingzhou is more than 85% (Fig. 7).

3.5 Effectiveness of emission reduction under the four main PSCs

In order to estimate the effectiveness of emission reduction in severe pollution events forced by the four potential synoptic controls, we conduct sensitivity simulations by applying seven emission scenarios (Table 2). All emission scenarios use the reduction ratio of 20% which is close to the average of the target emission reduction of

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all provinces in the 13th Five-year plan (The State Council of the People's Republic of China, 2016). Although the base year of emission reduction is 2015 for the 13th Five-year plan, it does not affect to use the simulation results of emission scenarios (with the reduction ratio of 20% applied to the simulated year 2013/2014) to explore the emission reduction effect of specific haze pollution events. The differences in model results between CON (control simulation) and JSN/JSNN/JALL (emissions of SO_2+NO_x/SO_2+NO_x+NH_3/all pollution sources at Jingzhou are reduced by 20%) represent the environmental benefits caused by different local emission reduction scenarios. The potential $PM_{2.5}$ mitigations by joint prevention and control in different regions are calculated by sensitivity experiments of CCALL (emissions of all pollution sources over Central China are reduced by 20%), CNALL (over Central China and NCP region), CPALL (over Central China and PRD region) and TALL (over Central China, NCP, YRD, PRD and SCB regions).

In the JSN emission reduction scenario, the sulfate and ammonium concentrations over Jingzhou are significantly reduced by 3.2-5.8 μg/m^3 (12.7-14.5%) and 0.6-1.9 μg/m^3 (3.2-5.9%) in these four pollution events, respectively. However, the concentration of nitrate increases (1.3-1.7%). This is because there is a competition mechanism between nitrate and sulfate. Ammonium ions always react with sulfate ions first to generate ammonium sulfate, which will continue to react with nitrate ions to generate ammonium nitrate when ammonium ions are rich (Mao et al., 2010). Thus the reduction of SO_2 emission increases the concentration of nitrate, which offset the contribution of sulfate particle reduction to the environment to some extent. Therefore, the application of JSN emission reduction scheme only reduces the $PM_{2.5}$ concentrations by 3.1-7.2 μg/m^3 (2.0-3.5%, Fig. 12). This inefficient emission reduction scheme is most widely used in heavy pollution areas over China in the past decade, ignoring the synergistic effect of various precursors.

Figure 12

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By applying the JSNN and JALL emission reduction scenarios, we aim to evaluate the synergistic effect of multiple precursors on emission reduction. These two scenarios reduce the average sulfate concentration in Jingzhou by 2.8-6.7 μg/m^3 (11.3-17.3%) and 2.9-7.2 μg/m^3 (11.7-17.9%), and the ammonium concentration by 2.0-4.8 μg/m^3 (12.1-16.5%) and 2.2-4.7 μg/m^3 (13.2-17.3%), respectively. Unlike the increments of nitrate in JSN emission reduction scenario, the nitrate decreases (JSNN: 0.3-1.2 μg/m^3; JALL: 0.4-1.5 μg/m^3). Therefore, through the application of JSNN and JALL emission reduction schemes, $PM_{2.5}$ concentrations decrease by 4.9-8.3% and 9.0-15.9%, respectively (Fig. 12), much higher than the improvement by JSN scenario. Zheng et al. (2019b) has also evaluated the sensitiveness of NH_3 control to $PM_{2.5}$ reduction based on observations. However, these results indicate that it is unrealistic to substantially reduce local emissions to achieve the national air quality standard in the long term.

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Additionally, the sensitivity simulations by excluding emission sources over upwind regions are conducted to estimate the potential $PM_{2.5}$ mitigations of inter-regional and intra-regional joint control. Our results show that after applying TALL emission reduction scenario, $PM_{2.5}$ concentrations have been significantly improved, with the improvement rates increased from 9.0-15.9% (by JALL scenario) to 17.4-18.8% (Fig. 12). Especially, the NW-type synoptic controlled air pollution episode shows the best effect of joint prevention, followed by SW-type. For NW-type, by reducing emissions over Central China and Northern China (CNALL scheme), $PM_{2.5}$ concentrations are reduced by 26.5 μg/m^3 (16.9%), much more effective than JALL emission reduction scheme (14.1 μg/m^3, 9.0%). In SW-type controlled pollution episode, it should be otherwise to decrease the emissions over Southern China in addition to Central China.

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4. Conclusion

The $PM_{2.5}$ pollution in autumn and winter haze periods is now the key obstacle for further improving air quality in China. The extremely severe and persistent $PM_{2.5}$ pollution episodes are attributed to adverse synoptic conditions in addition to high precursor emissions. For the $PM_{2.5}$ mitigations during winter haze episodes in specific region forced by various potential synoptic controls, how to effectively reduce emissions has become an urgent scientific question to be answered. Our results over Central China could provide reference for regional air quality policy-making.

Through Lamb-Jenkension circulation classification, the top four potential synoptic controls of heavy $PM_{2.5}$ pollution days (totally 109 days) over Central China from 2013 to 2018 are decomposed to be SW-type, NW-type A-type and C-type, accounting for 29%, 27%, 19% and 12% of the total pollution days, respectively. In these four PSCs, three inorganic salt aerosols (sulfate: ~20%; nitrate: ~18%; ammonium: ~13%) totally accounted for ~51% of $PM_{2.5}$ concentrations simulated by optimized GEOS-Chem modelling.

In the SW-type/NW-type synoptic situation, affected by the southerly/northerly airflow, pollutants over southern/northern China could be transmitted to Central China, with the transport contribution of 7.6%/37%. In the situation A-type/C-type weather, affected by stable weather condition/high relative humidity, the pollution processes are less affected by the emission sources from non-local regions. And the local emission sources dominate the contribution (82%/85%) to $PM_{2.5}$.

By only reducing SO_2 and NO_x emission and not controlling NH_3, due to the competition mechanism between nitrate and sulfate, the concentrations of sulfate and ammonium decrease, but the concentration of nitrate increases instead. The enhanced nitrate counteracts the effect of sulfate reduction on $PM_{2.5}$ mitigations, with less than 4% decrease in $PM_{2.5}$. Even if the NH_3 emission is also reduced, the $PM_{2.5}$ concentration reduction is less than 9%. By applying the TALL emission reduction scenario, $PM_{2.5}$ concentrations would decrease significantly, with the improvement rate increased from 9.0-15.9% (by JALL scenario) to 17.4-18.8%.

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These results provide an opportunity to effectively mitigate haze pollution by local emission control actions in coordination with regional collaborative actions according to different synoptic patterns. Especially, the NW-type synoptic controlled air pollution episode shows the best effect of joint prevention, followed by SW-type. It is noted that in this study, the division of transmission areas is relatively rough, and more accurate source area identification and refined assessment of emission reduction effect of multiple pollutants from source groups are needed in the follow-up.

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Author contributions

Yingying Yan and Shaofei Kong conceived and designed the research. Yingying Yan performed the data processing, model simulations, and analyses. Yue Zhou assisted in the circulation classification. Jian Wu provided the emission data over Central China. Shaofei Kong, Tianliang Zhao and Dantong Liu contributed the funding acquisition. Yingying Yan wrote the paper with input from all authors.

Data availability

Observational data are obtained from individual sources (see links in the text). Model results are available upon request. Model codes are available on a collaborative basis.

Competing interests

The authors declare that they have no conflict of interest.

References

The People's Government of Beijing Municipality (PGBM): Emergency plan for severe air pollution in Beijing, available at: http://www.beijing.gov.cn/zhengce/zhengcefagui/201905/t20190522_61613.html (last access: 14 July 2018), 2018 (in Chinese).

The People's Government of Guangdong Province (PGGP): Emergency plan for severe air pollution in Pearl River Delta, available at: http://www.gd.gov.cn/gkmlpt/content/0/142/post_142657.html#7 (last access: 14 July 2018), 2014 (in Chinese).

The People's Government of Shanghai Municipality (PGSM): Special emergency plan for heavy air pollution in Shanghai, available at: http://www.shanghai.gov.cn/nw2/nw2314/nw2319/nw31973/nw32019/nw32022/nw32023/u21aw1316153.html (last access: 14 July 2018), 2018 (in Chinese).

The State Council of the People's Republic of China (SCPPC): The Thirteenth Five-Year Plan for Energy Saving and Emission Reduction, available at: http://www.gov.cn/gongbao/content/2017/content_5163448.htm (last access: 14 July 2018), 2016 (in Chinese).

The State Council of the People's Republic of China (SCPPC): Air Pollution Prevention and Control Action Plan, available at: http://www.gov.cn/zhengce/content/2013-09/13/content_4561.htm (last access: 14 July 2018), 2013 (in Chinese).

Jenkinson A. F., Collison F. P.: An initial climatology of gales over the North Sea. Synoptic Climatology Branch Memorandum, 62. Bracknell: Meteorological Office, 1-18, 1977.

Lamb H H. Types and spells of weather around the year in the British Isles. Quarterly Journal Royal Meteorological Society, 76, 393-438, 1950.

Agarwal, N. K., Sharma, P., and Agarwal, S. K.: Particulate matter air pollution and cardiovascular disease, Medical Science, 21, 270-279, 2017.

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An, Z., Huang, R.J., Zhang, R., Tie, X., Li, G., Cao, J., Zhou, W., Shi, Z., Han, Y., Gu, Z., and Ji, Y.: Severe haze in northern China: A synergy of anthropogenic emissions and atmospheric processes, Proceedings of the National Academy of Sciences of the United States of America, 116, 8657-8666, 10.1073/pnas.1900125116, 2019.

Bei, N., Zhao, L., Xiao, B., Meng, N., and Feng, T.: Impacts of local circulations on the wintertime air pollution in the Guanzhong Basin, China, Science of the Total Environment, 592, 373-390, 10.1016/j.scitotenv.2017.02.151, 2017.

Bi, X., Dai, Q., Wu, J., Zhang, Q., Zhang, W., Luo, R., Cheng, Y., Zhang, J., Wang, L., Yu, Z., Zhang, Y., Tian, Y., and Feng, Y.: Characteristics of the main primary source profiles of particulate matter across China from 1987 to 2017, Atmospheric Chemistry and Physics, 19, 3223-3243, 10.5194/acp-19-3223-2019, 2019.

Cheng, J., Su, J., Cui, T., Li, X., Dong, X., Sun, F., Yang, Y., Tong, D., Zheng, Y., Li, Y., Li, J., Zhang, Q., and He, K.: Dominant role of emission reduction in $PM_{2.5}$ air quality improvement in Beijing during 2013-2017: a model-based decomposition analysis, Atmospheric Chemistry and Physics, 19, 6125-6146, 10.5194/acp-19-6125-2019, 2019.

Cheng, Z., Luo, L., Wang, S., Wang, Y., Sharma, S., Shimadera, H., Wang, X., Bressi, M., de Miranda, R. M., Jiang, J., Zhou, W., Fajardo, O., Yan, N., and Hao, J.: Status and characteristics of ambient $PM_{2.5}$ pollution in global megacities, Environment International, 89-90, 212-221, 10.1016/j.envint.2016.02.003, 2016.

Chuang, M.T., Chiang, P.C., Chan, C.C., Wang, C.F., Chang, E. E., and Lee, C.T.: The effects of synoptical weather pattern and complex terrain on the formation of aerosol events in the Greater Taipei area, Science of the Total Environment, 399, 128-146, 10.1016/j.scitotenv.2008.01.051, 2008.

Geng, G., Xiao, Q., Zheng, Y., Tong, D., Zhang, Y., Zhang, X., Zhang, Q., He, K., and Liu, Y.: Impact of China's Air Pollution Prevention and Control Action Plan on $PM_{2.5}$ chemical composition over eastern China, Science China-Earth Sciences, 62, 1872-1884, 10.1007/s11430-018-9353-x, 2019.

Gong, W., Zhang, T., Zhu, Z., Ma, Y., Ma, X., and Wang, W.: Characteristics of $PM_{1.0}$, $PM_{2.5}$, and PM_{10}, and Their Relation to Black Carbon in Wuhan, Central China, Atmosphere, 6, 1377-1387, 10.3390/atmos6091377, 2015.

Guo, L., Guo, X., Fang, C., and Zhu, S.: Observation analysis on characteristics of formation, evolution and transition of a long-lasting severe fog and haze episode in North China, Science China-Earth Sciences, 58, 329-344, 10.1007/s11430-014-4924-2, 2015.

He, J., Gong, S., Zhou, C., Lu, S., Wu, L., Chen, Y., Yu, Y., Zhao, S., Yu, L., and Yin, C.: Analyses of winter circulation types and their impacts on haze pollution in

Beijing, Atmospheric Environment, 192, 94-103, 10.1016/j.atmosenv.2018.08.060, 2018.

Huang, R.J., Zhang, Y., Bozzetti, C., Ho, K.F., Cao, J.J., Han, Y., Daellenbach, K. R., Slowik, J. G., Platt, S. M., Canonaco, F., Zotter, P., Wolf, R., Pieber, S. M., Bruns, E. A., Crippa, M., Ciarelli, G., Piazzalunga, A., Schwikowski, M., Abbaszade, G., Schnelle-Kreis, J., Zimmermann, R., An, Z., Szidat, S., Baltensperger, U., El Haddad, I., and Prevot, A. S. H.: High secondary aerosol contribution to particulate pollution during haze events in China, Nature, 514, 218-222, 10.1038/nature13774, 2014.

Huang, X., Ding, A., Gao, J., Zheng, B., Zhou, D., Qi, X., Tang, R., Wang, J., Ren, C., Nie, W., Chi, X., Xu, Z., Chen, L., Li, Y., Che, F., Pang, N., Wang, H., Tong, D., Qin, W., Cheng, W., Liu, W., Fu, Q., Liu, B., Chai, F., Davis, S. J., Zhang, Q., and He, K.: Enhanced secondary pollution offset reduction of primary emissions during COVID-19 lockdown in China, National Science Review, 10.1093/nsr/nwaa137, 2020.

Kurita, H., Sasaki, K., Muroga, H., Ueda, H., and Wakamatsu, S.: Long-range transport of air pollution under light gradient wind conditions, Journal of Climate and Applied Meteorology, 24, 425-434, 10.1175/1520-0450(1985)024<0425:lrtoap>2.0.co;2, 1985.

Leung, D. M., Tai, A. P. K., Mickley, L. J., Moch, J. M., van Donkelaar, A., Shen, L., and Martin, R. V.: Synoptic meteorological modes of variability for fine particulate matter ($PM_{2.5}$) air quality in major metropolitan regions of China, Atmospheric Chemistry and Physics, 18, 6733-6748, 10.5194/acp-18-6733-2018, 2018.

Li, J., Liao, H., Hu, J., and Li, N.: Severe particulate pollution days in China during 2013-2018 and the associated typical weather patterns in Beijing-Tianjin-Hebei and the Yangtze River Delta regions, Environmental Pollution, 248, 74-81, 10.1016/j.envpol.2019.01.124, 2019.

Li, M., Zhang, Q., Kurokawa, J.-i., Woo, J.-H., He, K., Lu, Z., Ohara, T., Song, Y., Streets, D. G., Carmichael, G. R., Cheng, Y., Hong, C., Huo, H., Jiang, X., Kang, S., Liu, F., Su, H., and Zheng, B.: MIX: a mosaic Asian anthropogenic emission inventory under the international collaboration framework of the MICS-Asia and HTAP, Atmospheric Chemistry and Physics, 17, 935-963, 10.5194/acp-17-935-2017, 2017a.

Li, X., Zhang, Q., Zhang, Y., Zhang, L., Wang, Y., Zhang, Q., Li, M., Zheng, Y., Geng, G., Wallington, T. J., Han, W., Shen, W., and He, K.: Attribution of $PM_{2.5}$ exposure in Beijing-Tianjin-Hebei region to emissions: implication to control strategies, Science Bulletin, 62, 957-964, 10.1016/j.scib.2017.06.005, 2017b.

Li, Z., Guo, J., Ding, A., Liao, H., Liu, J., Sun, Y., Wang, T., Xue, H., Zhang, H., and Zhu, B.: Aerosol and boundary-layer interactions and impact on air quality, National Science Review, 4, 810-833, 10.1093/nsr/nwx117, 2017c.

Liao, Z., Gao, M., Sun, J., and Fan, S.: The impact of synoptic circulation on air quality and pollution-related human health in the Yangtze River Delta region, Science of the Total Environment, 607, 838-846, 10.1016/j.scitotenv.2017.07.031, 2017.

Liao, Z., Xie, J., Fang, X., Wang, Y., Zhang, Y., Xu, X., and Fan, S.: Modulation of synoptic circulation to dry season $PM_{2.5}$ pollution over the Pearl River Delta region: An investigation based on self-organizing maps, Atmospheric Environment, 230, 10.1016/j.atmosenv.2020.117482, 2020.

Lin, Y., Zou, J., Yang, W., and Li, C.Q.: A Review of Recent Advances in Research on $PM_{2.5}$ in China, International Journal of Environmental Research and Public Health, 15, 10.3390/ijerph15030438, 2018.

Liu, M., Huang, X., Song, Y., Tang, J., Cao, J., Zhang, X., Zhang, Q., Wang, S., Xu, T., Kang, L., Cai, X., Zhang, H., Yang, F., Wang, H., Yu, J. Z., Lau, A. K. H., He, L., Huang, X., Duan, L., Ding, A., Xue, L., Gao, J., Liu, B., and Zhu, T.: Ammonia emission control in China would mitigate haze pollution and nitrogen deposition, but worsen acid rain, Proceedings of the National Academy of Sciences of the United States of America, 116, 7760-7765, 10.1073/pnas.1814880116, 2019.

Mao, J., Jacob, D. J., Evans, M. J., Olson, J. R., Ren, X., Brune, W. H., Clair, J. M. S., Crounse, J. D., Spencer, K. M., Beaver, M. R., Wennberg, P. O., Cubison, M. J., Jimenez, J. L., Fried, A., Weibring, P., Walega, J. G., Hall, S. R., Weinheimer, A. J., Cohen, R. C., Chen, G., Crawford, J. H., McNaughton, C., Clarke, A. D., Jaeglé, L., Fisher, J. A., Yantosca, R. M., Le Sager, P., and Carouge, C.: Chemistry of hydrogen oxide radicals (HOx) in the Arctic troposphere in spring, Atmospheric Chemistry and Physics, 10, 5823-5838, 10.5194/acp-10-5823-2010, 2010.

Miao, Y., Guo, J., Liu, S., Liu, H., Li, Z., Zhang, W., and Zhai, P.: Classification of summertime synoptic patterns in Beijing and their associations with boundary layer structure affecting aerosol pollution, Atmospheric Chemistry and Physics, 17, 3097-3110, 10.5194/acp-17-3097-2017, 2017.

Ning, G., Wang, S., Yim, S. H. L., Li, J., Hu, Y., Shang, Z., Wang, J., and Wang, J.: Impact of low-pressure systems on winter heavy air pollution in the northwest Sichuan Basin, China, Atmospheric Chemistry and Physics, 18, 13601-13615, 10.5194/acp-18-13601-2018, 2018.

Pope, R. J., Savage, N. H., Chipperfield, M. P., Ordonez, C., and Neal, L. S.: The influence of synoptic weather regimes on UK air quality: regional model studies of

Deleted: Lin, J., Tong, D., Davis, S., Ni, R., Tan, X., Pan, D., Zhao, H., Lu, Z., Streets, D., Feng, T., Zhang, Q., Yan, Y., Hu, Y., Li, J., Liu, Z., Jiang, X., Geng, G., He, K., Huang, Y., and Guan, D.: Global climate forcing of aerosols embodied in international trade, Nature Geoscience, 9, 790-+, 10.1038/ngeo2798, 2016.

Deleted: Liu, M., Lin, J., Wang, Y., Sun, Y., Zheng, B., Shao, J., Chen, L., Zheng, Y., Chen, J., Fu, T.-M., Yan, Y., Zhang, Q., and Wu, Z.: Spatiotemporal variability of NO_2 and $PM_{2.5}$ over Eastern China: observational and model analyses with a novel statistical method, Atmospheric Chemistry and Physics, 18, 12933-12952, 10.5194/acp-18-12933-2018, 2018.

tropospheric column NO_2, Atmospheric Chemistry and Physics, 15, 11201-11215, 10.5194/acp-15-11201-2015, 2015.

Russo, A., Trigo, R. M., Martins, H., and Mendes, M. T.: NO_2, PM_{10} and O_3 urban concentrations and its association with circulation weather types in Portugal, Atmospheric Environment, 89, 768-785, 10.1016/j.atmosenv.2014.02.010, 2014.

Shu, L., Xie, M., Gao, D., Wang, T., Fang, D., Liu, Q., Huang, A., and Peng, L.: Regional severe particle pollution and its association with synoptic weather patterns in the Yangtze River Delta region, China, Atmospheric Chemistry and Physics, 17, 12871-12891, 10.5194/acp-17-12871-2017, 2017.

Sun, Y., Niu, T., He, J., Ma, Z., Liu, P., Xiao, D., Hu, J., Yang, J., and Yan, X.: Classification of circulation patterns during the formation and dissipation of continuous pollution weather over the Sichuan Basin, China, Atmospheric Environment, 223, 10.1016/j.atmosenv.2019.117244, 2020.

Sun, Y. L., Chen, C., Zhang, Y. J., Xu, W. Q., Zhou, L. B., Cheng, X. L., Zheng, H. T., Ji, D. S., Li, J., Tang, X., Fu, P. Q., and Wang, Z. F.: Rapid formation and evolution of an extreme haze episode in Northern China during winter 2015, Scientific Reports, 6, 10.1038/srep27151, 2016.

Tian, S. L., Pan, Y. P., and Wang, Y. S.: Size-resolved source apportionment of particulate matter in urban Beijing during haze and non-haze episodes, Atmospheric Chemistry and Physics, 16, 1-19, 10.5194/acp-16-1-2016, 2016.

Trigo, R. M., and DaCamara, C. C.: Circulation weather types and their influence on the precipitation regime in Portugal, International Journal of Climatology, 20, 1559-1581, 10.1002/1097-0088(20001115)20:13<1559::aid-joc555>3.0.co;2-5, 2000.

Twohy, C. H., Coakley, J. A., Jr., and Tahnk, W. R.: Effect of changes in relative humidity on aerosol scattering near clouds, Journal of Geophysical Research-Atmospheres, 114, 10.1029/2008jd010991, 2009.

Wang, X., Wei, W., Cheng, S., Li, J., Zhang, H., and Lv, Z.: Characteristics and classification of $PM_{2.5}$ pollution episodes in Beijing from 2013 to 2015, Science of the Total Environment, 612, 170-179, 10.1016/j.scitotenv.2017.08.206, 2018.

Wang, Y., Chen, Y., Wu, Z., Shang, D., Bian, Y., Du, Z., Schmitt, S. H., Su, R., Gkatzelis, G. I., Schlag, P., Hohaus, T., Voliotis, A., Lu, K., Zen, L., Zhao, C., Alfarra, M. R., McFiggans, G., Wiedensohler, A., Kiendler-Scharr, A., Zhang, Y., and Hu, M.: Mutual promotion between aerosol particle liquid water and particulate nitrate enhancement leads to severe nitrate-dominated particulate matter pollution and low visibility, Atmospheric Chemistry and Physics, 20, 2161-2175, 10.5194/acp-20-2161-2020, 2020.

Wu, J., Kong, S., Wu, F., Cheng, Y., Zheng, S., Yan, Q., Zheng, H., Yang, G., Zheng, M., Liu, D., Zhao, D., and Qi, S.: Estimating the open biomass burning emissions in central and eastern China from 2003 to 2015 based on satellite observation, Atmospheric Chemistry and Physics, 18, 11623-11646, 10.5194/acp-18-11623-2018, 2018.

Xu, G., Jiao, L., Zhang, B., Zhao, S., Yuan, M., Gu, Y., Liu, J., and Tang, X.: Spatial and temporal variability of the $PM_{2.5}/PM_{10}$ ratio in Wuhan, Central China, Aerosol and Air Quality Research, 17, 741-751, 10.4209/aaqr.2016.09.0406, 2017.

Yan, Q., Kong, S., Yan, Y., Liu, H., Wang, W., Chen, K., Yin, Y., Zheng, H., Wu, J., Yao, L., Zeng, X., Cheng, Y., Zheng, S., Wu, F., Niu, Z., Zhang, Y., Zheng, M., Zhao, D., Liu, D., and Qi, S.: Emission and simulation of primary fine and submicron particles and water-soluble ions from domestic coal combustion in China, Atmospheric Environment, 224, 10.1016/j.atmosenv.2020.117308, 2020.

Yan, Y., Cabrera-Perez, D., Lin, J., Pozzer, A., Hu, L., Millet, D. B., Porter, W. C., and Lelieveld, J.: Global tropospheric effects of aromatic chemistry with the SAPRC-11 mechanism implemented in GEOS-Chem version 9-02, Geoscientific Model Development, 12, 111-130, 10.5194/gmd-12-111-2019, 2019.

Yan, Y. Y., Lin, J. T., Kuang, Y., Yang, D., and Zhang, L.: Tropospheric carbon monoxide over the Pacific during HIPPO: two-way coupled simulation of GEOS-Chem and its multiple nested models, Atmospheric Chemistry and Physics, 14, 12649-12663, 10.5194/acp-14-12649-2014, 2014.

Yang, Y., Zheng, X., Gao, Z., Wang, H., Wang, T., Li, Y., Lau, G. N. C., and Yim, S. H. L.: Long-term trends of persistent synoptic circulation events in planetary boundary layer and their relationships with haze pollution in winter half year over Eastern China, Journal of Geophysical Research-Atmospheres, 123, 10991-11007, 10.1029/2018jd028982, 2018.

Yu, C., Zhao, T., Bai, Y., Zhang, L., Kong, S., Yu, X., He, J., Cui, C., Yang, J., You, Y., Ma, G., Wu, M., and Chang, J.: Heavy air pollution with a unique "non-stagnant" atmospheric boundary layer in the Yangtze River middle basin aggravated by regional transport of $PM_{2.5}$ over China, Atmospheric Chemistry and Physics, 20, 7217-7230, 10.5194/acp-20-7217-2020, 2020.

Zhang, J. P., Zhu, T., Zhang, Q. H., Li, C. C., Shu, H. L., Ying, Y., Dai, Z. P., Wang, X., Liu, X. Y., Liang, A. M., Shen, H. X., and Yi, B. Q.: The impact of circulation patterns on regional transport pathways and air quality over Beijing and its surroundings, Atmospheric Chemistry and Physics, 12, 5031-5053, 10.5194/acp-12-5031-2012, 2012.

Zhang, Q., Jiang, X., Tong, D., Davis, S. J., Zhao, H., Geng, G., Feng, T., Zheng, B., Lu, Z., Streets, D. G., Ni, R., Brauer, M., van Donkelaar, A., Martin, R. V., Huo, H., Liu, Z., Pan, D., Kan, H., Yan, Y., Lin, J., He, K., and Guan, D.: Transboundary health impacts of transported global air pollution and international trade, Nature, 543, 10.1038/nature21712, 2017.

Zhang, Q., Zheng, Y., Tong, D., Shao, M., Wang, S., Zhang, Y., Xu, X., Wang, J., He, H., Liu, W., Ding, Y., Lei, Y., Li, J., Wang, Z., Zhang, X., Wang, Y., Cheng, J., Liu, Y., Shi, Q., Yan, L., Geng, G., Hong, C., Li, M., Liu, F., Zheng, B., Cao, J., Ding, A., Gao, J., Fu, Q., Huo, J., Liu, B., Liu, Z., Yang, F., He, K., and Hao, J.: Drivers of improved PM2.5 air quality in China from 2013 to 2017, Proceedings of the National Academy of Sciences of the United States of America, 116, 24463-24469, 10.1073/pnas.1907956116, 2019.

Zheng, B., Tong, D., Li, M., Liu, F., Hong, C., Geng, G., Li, H., Li, X., Peng, L., Qi, J., Yan, L., Zhang, Y., Zhao, H., Zheng, Y., He, K., and Zhang, Q.: Trends in China's anthropogenic emissions since 2010 as the consequence of clean air actions, Atmospheric Chemistry and Physics, 18, 14095-14111, 10.5194/acp-18-14095-2018, 2018.

Zheng, H., Kong, S., Wu, F., Cheng, Y., Niu, Z., Zheng, S., Yang, G., Yao, L., Yan, Q., Wu, J., Zheng, M., Chen, N., Xu, K., Yan, Y., Liu, D., Zhao, D., Zhao, T., Bai, Y., Li, S., and Qi, S.: Intra-regional transport of black carbon between the south edge of the North China Plain and central China during winter haze episodes, Atmospheric Chemistry and Physics, 19, 4499-4516, 10.5194/acp-19-4499-2019, 2019a.

Zheng, M., Wang, Y., Bao, J., Yuan, L., Zheng, H., Yan, Y., Liu, D., Xie, M., and Kong, S.: Initial Cost Barrier of Ammonia Control in Central China, Geophysical Research Letters, 46, 14175-14184, 10.1029/2019gl084351, 2019b.

Zheng, X. Y., Fu, Y. F., Yang, Y. J., and Liu, G. S.: Impact of atmospheric circulations on aerosol distributions in autumn over eastern China: observational evidence, Atmospheric Chemistry and Physics, 15, 12115-12138, 10.5194/acp-15-12115-2015, 2015.

Zhong, J., Zhang, X., Dong, Y., Wang, Y., Liu, C., Wang, J., Zhang, Y., and Che, H.: Feedback effects of boundary-layer meteorological factors on cumulative explosive growth of $PM_{2.5}$ during winter heavy pollution episodes in Beijing from 2013 to 2016, Atmospheric Chemistry and Physics, 18, 247-258, 10.5194/acp-18-247-2018, 2018.

Bai, Z., Winiwarter, W., Klimont, Z., Velthof, G., Misselbrook, T., Zhao, Z., Jin, X., Oenema, O., Hu, C., and Ma, L.: Further Improvement of Air Quality in China Needs

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Clear Ammonia Mitigation Target, Environmental Science & Technology, 53, 10542-10544, 10.1021/acs.est.9b04725, 2019.

Xu, Z., Liu, M., Zhang, M., Song, Y., Wang, S., Zhang, L., Xu, T., Wang, T., Yan, C., Zhou, T., Sun, Y., Pan, Y., Hu, M., Zheng, M., and Zhu, T.: High efficiency of livestock ammonia emission controls in alleviating particulate nitrate during a severe winter haze episode in northern China, Atmospheric Chemistry and Physics, 19, 5605-5613, 10.5194/acp-19-5605-2019, 2019.

Ye, Z., Guo, X., Cheng, L., Cheng, S., Chen, D., Wang, W., and Liu, B.: Reducing PM2.5 and secondary inorganic aerosols by agricultural ammonia emission mitigation within the Beijing-Tianjin-Hebei region, China, Atmospheric Environment, 219, 10.1016/j.atmosenv.2019.116989, 2019.

Cao, J.-J., Shen, Z.-X., Chow, J. C., Watson, J. G., Lee, S.-C., Tie, X.-X., Ho, K.-F., Wang, G.-H., and Han, Y.-M.: Winter and Summer PM2.5 Chemical Compositions in Fourteen Chinese Cities, Journal of the Air & Waste Management Association, 62, 1214-1226, 10.1080/10962247.2012.701193, 2012.

Huang, W., Cao, J., Tao, Y., Dai, L., Lu, S.-E., Hou, B., Wang, Z., and Zhu, T.: Seasonal Variation of Chemical Species Associated With Short-Term Mortality Effects of PM2.5 in Xi'an, a Central City in China, American Journal of Epidemiology, 175, 556-566, 10.1093/aje/kwr342, 2012.

Liu, Z., Gao, W., Yu, Y., Hu, B., Xin, J., Sun, Y., Wang, L., Wang, G., Bi, X., Zhang, G., Xu, H., Cong, Z., He, J., Xu, J., and Wang, Y.: Characteristics of PM2.5 mass concentrations and chemical species in urban and background areas of China: emerging results from the CARE-China network, Atmospheric Chemistry and Physics, 18, 8849-8871, 10.5194/acp-18-8849-2018, 2018.

Luo, Y., Zhou, X., Zhang, J., Xiao, Y., Wang, Z., Zhou, Y., and Wang, W.: PM2.5 pollution in a petrochemical industry city of northern China: Seasonal variation and source apportionment, Atmospheric Research, 212, 285-295, 10.1016/j.atmosres.2018.05.029, 2018.

Wang, H. L., Qiao, L. P., Lou, S. R., Zhou, M., Ding, A. J., Huang, H. Y., Chen, J. M., Wang, Q., Tao, S., Chen, C. H., Li, L., and Huang, C.: Chemical composition of PM2.5 and meteorological impact among three years in urban Shanghai, China, Journal of Cleaner Production, 112, 1302-1311, 10.1016/j.jclepro.2015.04.099, 2016a.

Wang, J., Li, X., Zhang, W., Jiang, N., Zhang, R., and Tang, X.: Secondary PM2.5 in Zhengzhou, China: Chemical Species Based on Three Years of Observations, Aerosol and Air Quality Research, 16, 91-104, 10.4209/aaqr.2015.01.0007, 2016b.

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Wang, Q., Fang, J., Shi, W., and Dong, X.: Distribution characteristics and policy-related improvements of PM2.5 and its components in six Chinese cities, Environmental Pollution, 266, 10.1016/j.envpol.2020.115299, 2020.

Xu, Q., Wang, S., Jiang, J., Bhattarai, N., Li, X., Chang, X., Qiu, X., Zheng, M., Hua, Y., and Hao, J.: Nitrate dominates the chemical composition of PM2.5 during haze event in Beijing, China, Science of the Total Environment, 689, 1293-1303, 10.1016/j.scitotenv.2019.06.294, 2019.

Zhang, T., Cao, J. J., Tie, X. X., Shen, Z. X., Liu, S. X., Ding, H., Han, Y. M., Wang, G. H., Ho, K. F., Qiang, J., and Li, W. T.: Water-soluble ions in atmospheric aerosols measured in Xi'an, China: Seasonal variations and sources, Atmospheric Research, 102, 110-119, 10.1016/j.atmosres.2011.06.014, 2011.

Zheng, J., Hu, M., Peng, J., Wu, Z., Kumar, P., Li, M., Wang, Y., and Guo, S.: Spatial distributions and chemical properties of PM2.5 based on 21 field campaigns at 17 sites in China, Chemosphere, 159, 480-487, 10.1016/j.chemosphere.2016.06.032, 2016.

Chang, W., and Zhan, J.: The association of weather patterns with haze episodes: Recognition by PM2.5 oriented circulation classification applied in Xiamen, Southeastern China, Atmospheric Research, 197, 425-436, 10.1016/j.atmosres.2017.07.024, 2017.

Dai, H., Zhu, J., Liao, H., Li, J., Liang, M., Yang, Y., and Yue, X.: Co-occurrence of ozone and PM2.5 pollution in the Yangtze River Delta over 2013-2019: Spatiotemporal distribution and meteorological conditions, Atmospheric Research, 249, 10.1016/j.atmosres.2020.105363, 2021.

Ding, A., Huang, X., Nie, W., Chi, X., Xu, Z., Zheng, L., Xu, Z., Xie, Y., Qi, X., Shen, Y., Sun, P., Wang, J., Wang, L., Sun, J., Yang, X.-Q., Qin, W., Zhang, X., Cheng, W., Liu, W., Pan, L., and Fu, C.: Significant reduction of PM2.5 in eastern China due to regional-scale emission control: evidence from SORPES in 2011-2018, Atmospheric Chemistry and Physics, 19, 11791-11801, 10.5194/acp-19-11791-2019, 2019.

Fu, X., Wang, S., Xing, J., Zhang, X., Wang, T., and Hao, J.: Increasing Ammonia Concentrations Reduce the Effectiveness of Particle Pollution Control Achieved via SO2 and NOX Emissions Reduction in East China, Environmental Science & Technology Letters, 4, 221-227, 10.1021/acs.estlett.7b00143, 2017.

Xing, J., Ding, D., Wang, S., Zhao, B., Jang, C., Wu, W., Zhang, F., Zhu, Y., and Hao, J.: Quantification of the enhanced effectiveness of NOx control from simultaneous reductions of VOC and NH3 for reducing air pollution in the Beijing-Tianjin-Hebei region, China, Atmospheric Chemistry and Physics, 18, 7799-7814, 10.5194/acp-18-7799-2018, 2018.

Xing, J., Ding, D., Wang, S., Dong, Z., Kelly, J. T., Jang, C., Zhu, Y., and Hao, J.: Development and application of observable response indicators for design of an effective ozone and fine-particle pollution control strategy in China, Atmospheric Chemistry and Physics, 19, 13627-13646, 10.5194/acp-19-13627-2019, 2019.

Philipp, A., Beck, C., Huth, R., and Jacobeit, J.: Development and comparison of circulation type classifications using the COST 733 dataset and software, International Journal of Climatology, 36, 2673-2691, 10.1002/joc.3920, 2016.

Pope, R. J., Savage, N. H., Chipperfield, M. P., Arnold, S. R., and Osborn, T. J.: The influence of synoptic weather regimes on UK air quality: analysis of satellite column NO2, Atmospheric Science Letters, 15, 211-217, 10.1002/asl2.492, 2014.

Russo, A., Trigo, R. M., Martins, H., and Mendes, M. T.: NO2, PM10 and O3 urban concentrations and its association with circulation weather types in Portugal, Atmospheric Environment, 89, 768-785, 10.1016/j.atmosenv.2014.02.010, 2014.

Santurtun, A., Carlos Gonzalez-Hidalgo, J., Sanchez-Lorenzo, A., and Teresa Zarrabeitia, M.: Surface ozone concentration trends and its relationship with weather types in Spain (2001-2010), Atmospheric Environment, 101, 10-22, 10.1016/j.atmosenv.2014.11.005, 2015.

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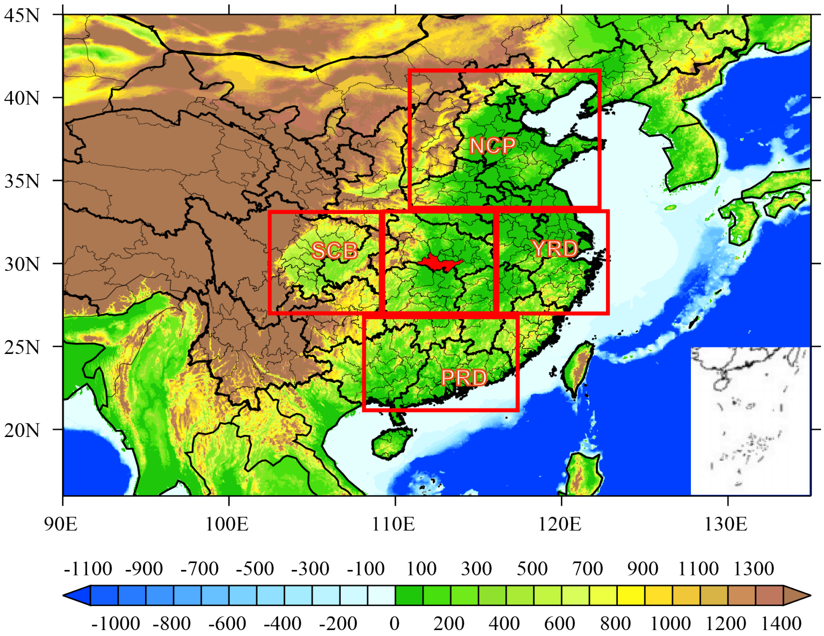

Figue 1 The location of Jingzhou (red area) and the major haze pollution regions of NCP, YRD, PRD, and SCB. The areas framed in red are used to investigate the inter-regional imapcts by GEOS-Chem sensitivity simulations. The overlaid map shows the surface elevation (m) from a 2 min Gridded Global Relief Data (ETOPO2v2) available at NGDC Marine Trackline Geophysical database (http://www.ngdc.noaa.gov/mgg/global/etopo2.html).

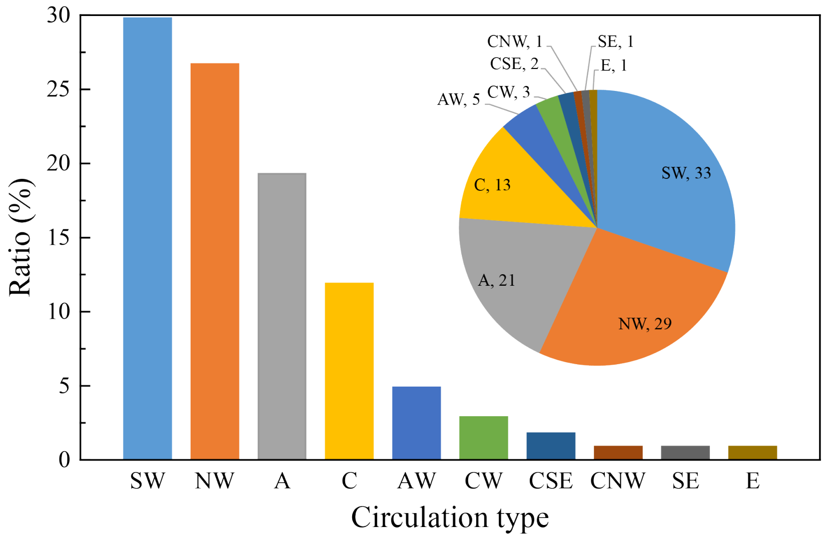

Figure 2 Frequency distributions of ten circulation types for the heavy pollution days of 2013-2018 over Jingzhou. The occurrence numbers of each type are shown. The ten circulation types include Southwest (SW), Northwest (NW), Anticyclone (A), Cyclone (C), Anticyclone-West (AW), Cyclone-West (CW), Cyclone-Southeast (CSE), Cyclone-Northwest (CNW), Southeast (SE) and East (E), respectively.

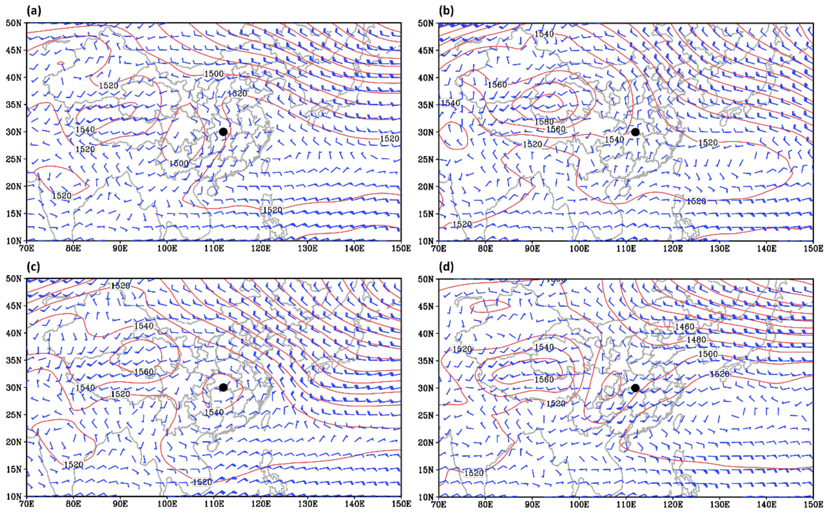

Figure 3 Spatial distribution of 850 hPa geopotential height and wind vector for SW-type (a), NW-type (b), A-type (c) and C-type (d) synoptic control averaged over 2013-2018. The black dot indicates the location of Jingzhou.

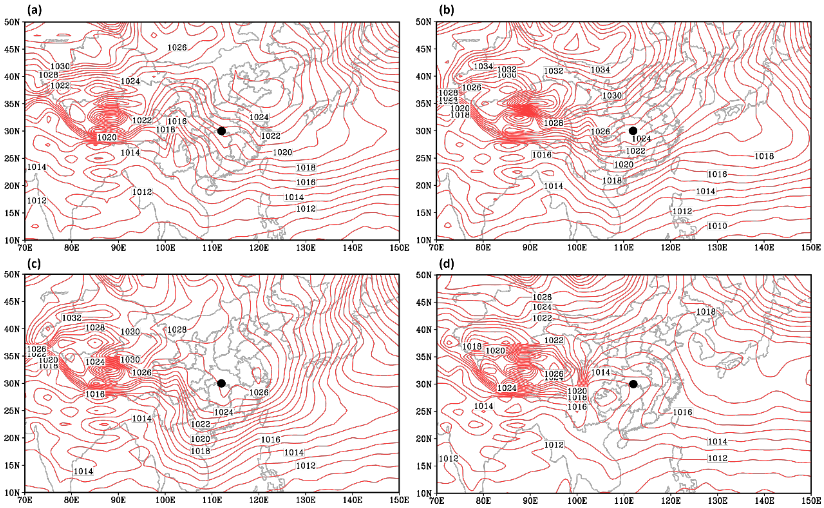

Figure 4 Spatial distribution of sea level pressure for SW-type (a), NW-type (b), A-type (c) and C-type (d) synoptic control averaged over 2013-2018. The black dot indicates the location of Jingzhou.

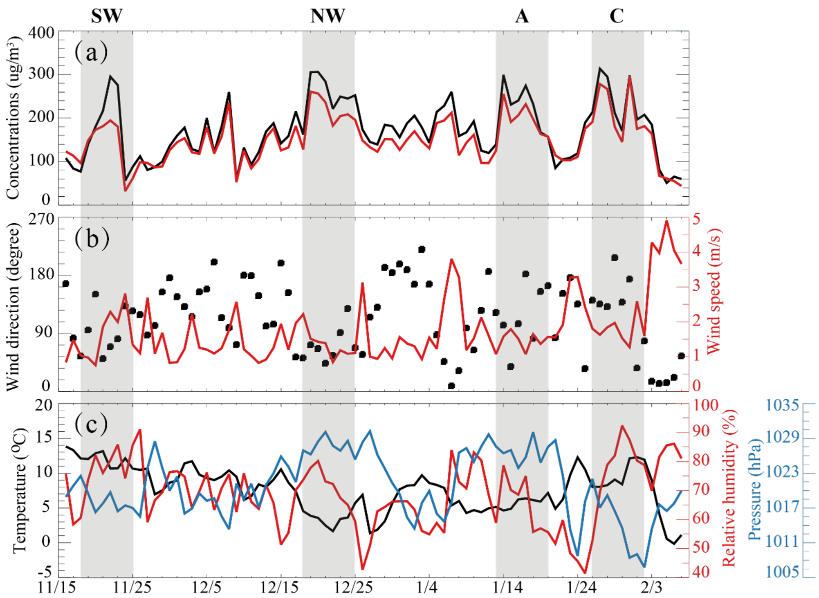

Figure 5 (a) Daily mean values of modeled (red line) and observed (black line) $PM_{2.5}$ concentration ($\mu g/m^3$) at Jingzhou and four severe pollution events (grey area) from November, 2013 to February, 2014. (b) Observed daily mean wind speed (red line) and wind direction (black dots). (c) Obseved temperature (black line), relative humidity (red line) and sea level pressure (blue line).

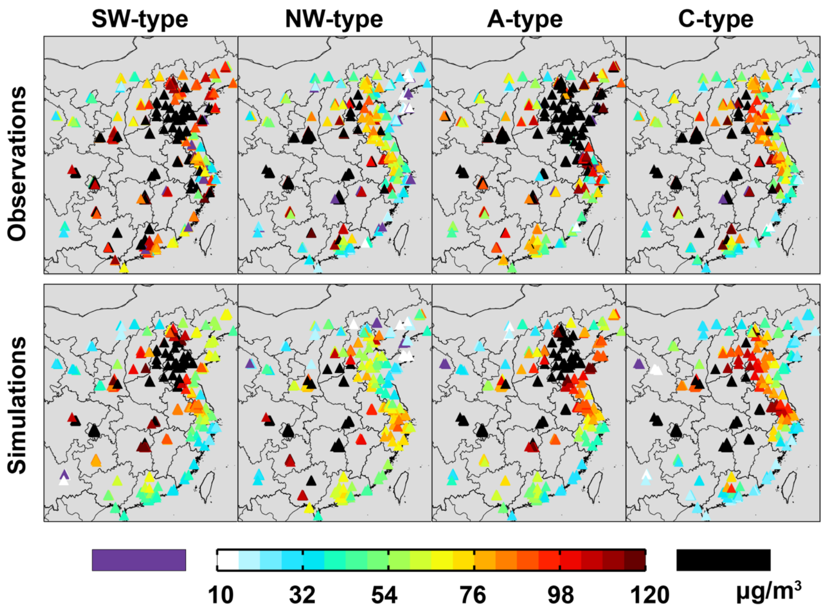

Figure 6 Spatial distribution of observed (top row) and modeled (bottom row, by CON case) $PM_{2.5}$ concentrations ($\mu g/m^3$) averaged over four severe pollution episodes controlled by SW-type (first column), NW-type (second column), A-type (third column) and C-type (forth column) synoptic pattern, respectively.

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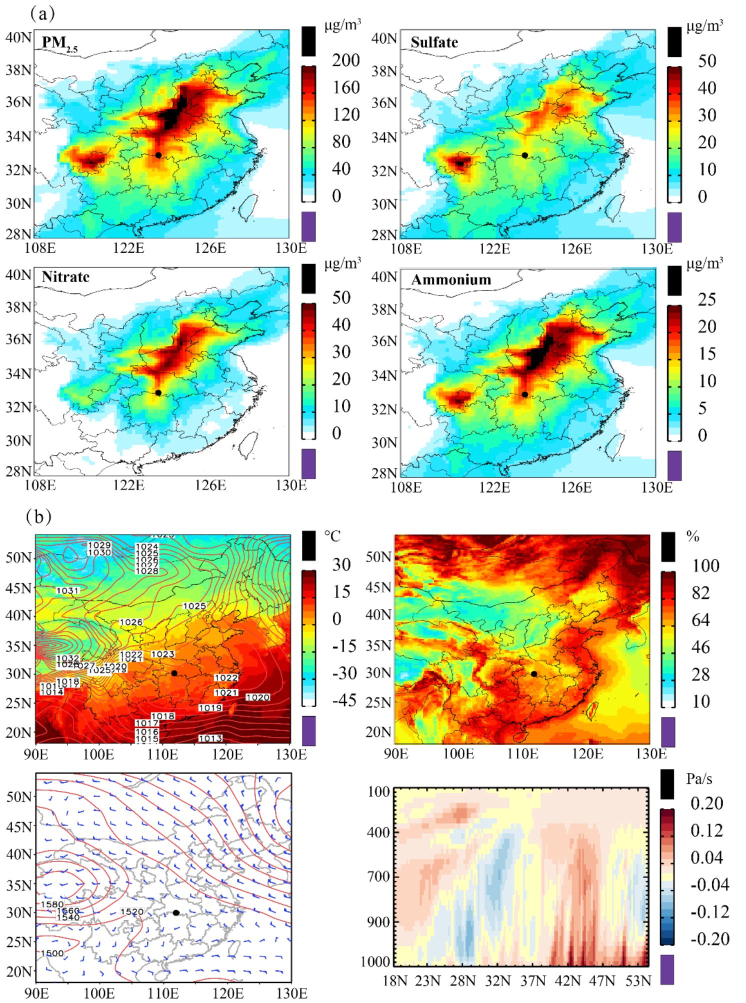

Figure 7 (a) Spatial distribution of $PM_{2.5}$, sulfate, nitrate and ammonium concentrations averaged over SW-type synoptic controls (18-25 November, 2013) simulated by GEOS-Chem control simulation (μg/m^3). (b) Meteorological conditions of SW-type: sea level pressure (red line) and temperature (colour shades), surface relative humidity (%) fields, 850 hPa wind and geopotential height (red line) and height–latitude cross-sections of vertical velocity (Pa/s).

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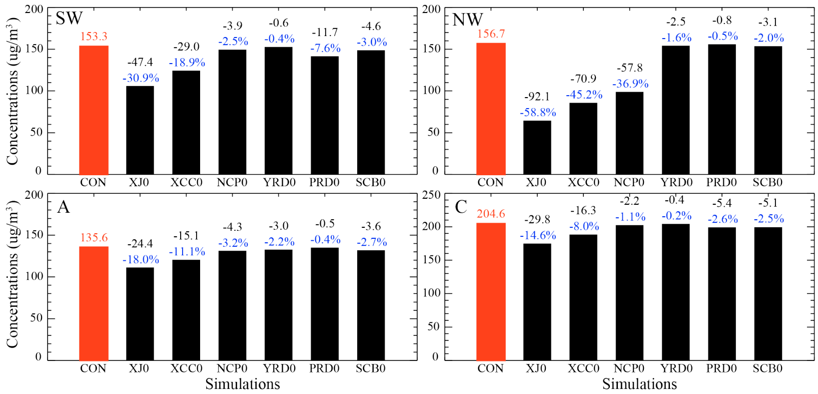

Figure 8 Modeled concentrations (μg/m^3) of $PM_{2.5}$ at Jingzhou in the GEOS-Chem control (red bar) and sensitivity (black bar) simulations in view of the regional transportation, and the differences (black characters for mass concentrations and blue characters for mass percentages) between the sensitivity and the control simulations. The abbreviations of each simulation referred to Table 2.

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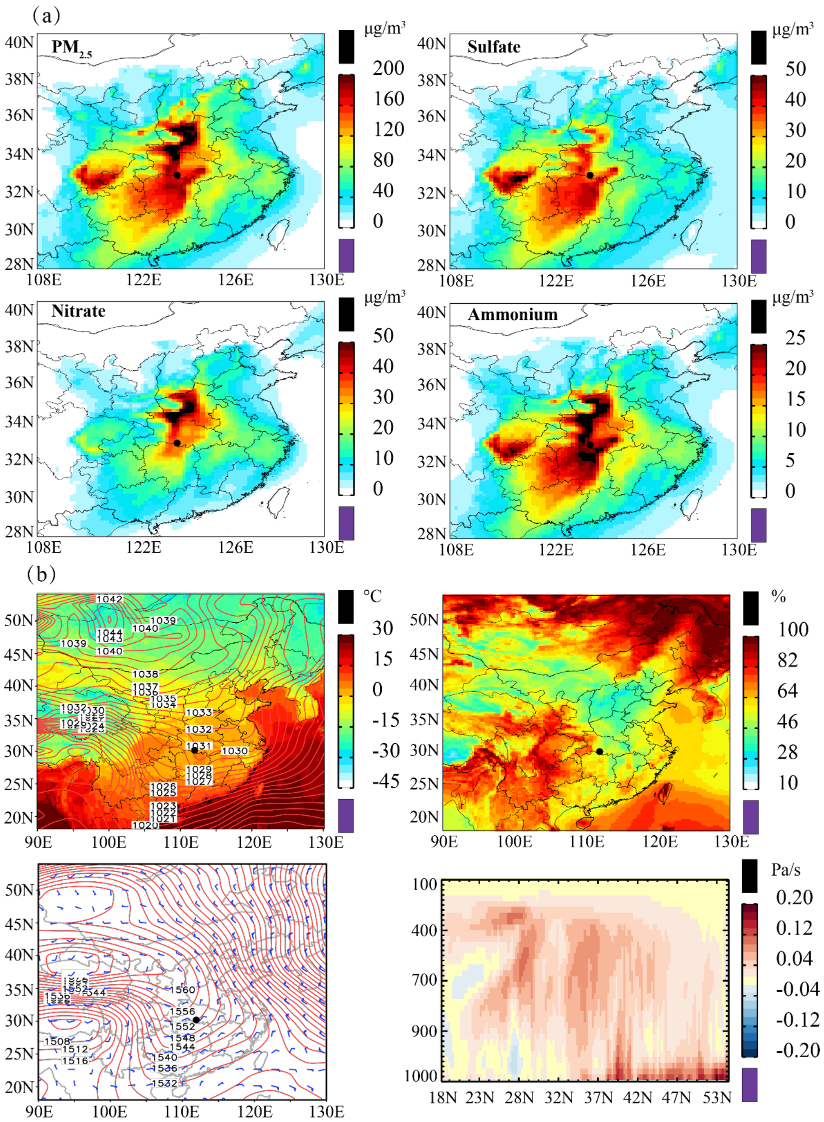

Figure 9 As in Fig. 6 but for NW-type synoptic control (19-26 December, 2013).

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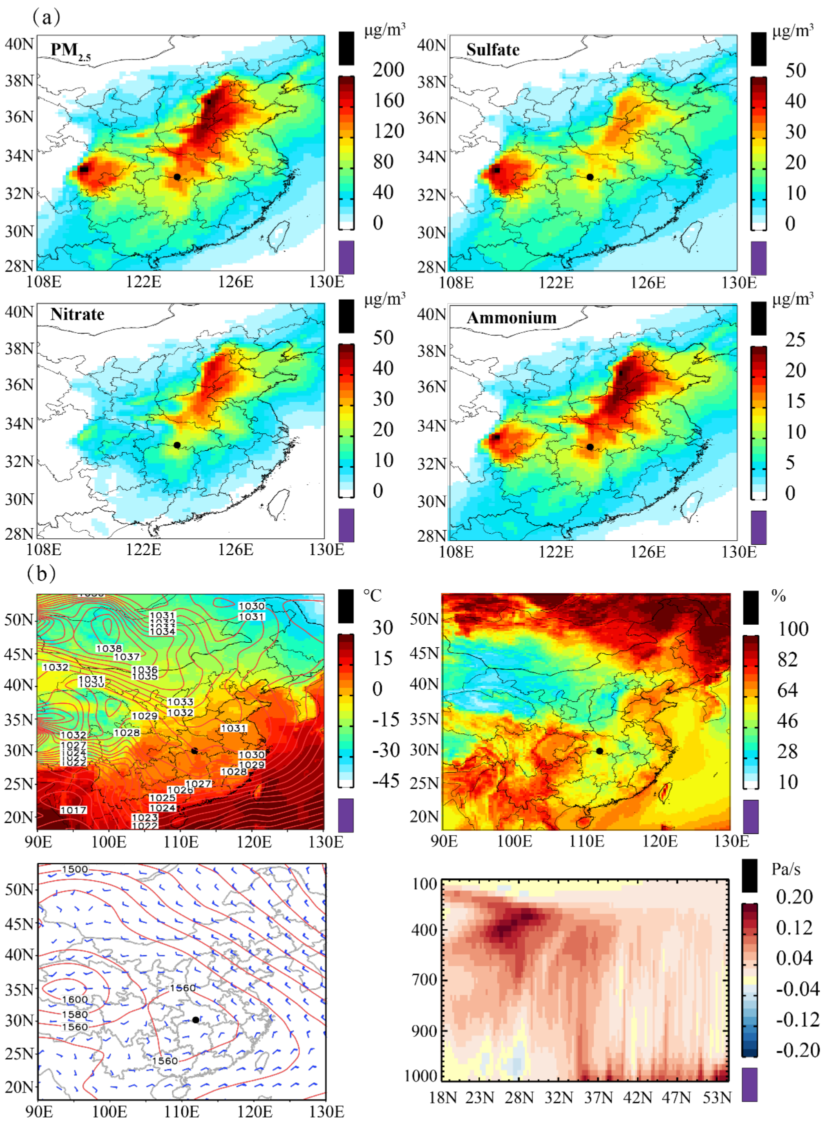

Figure 10 As in Fig. 6 but for A-type synoptic control (14-21 January, 2014).

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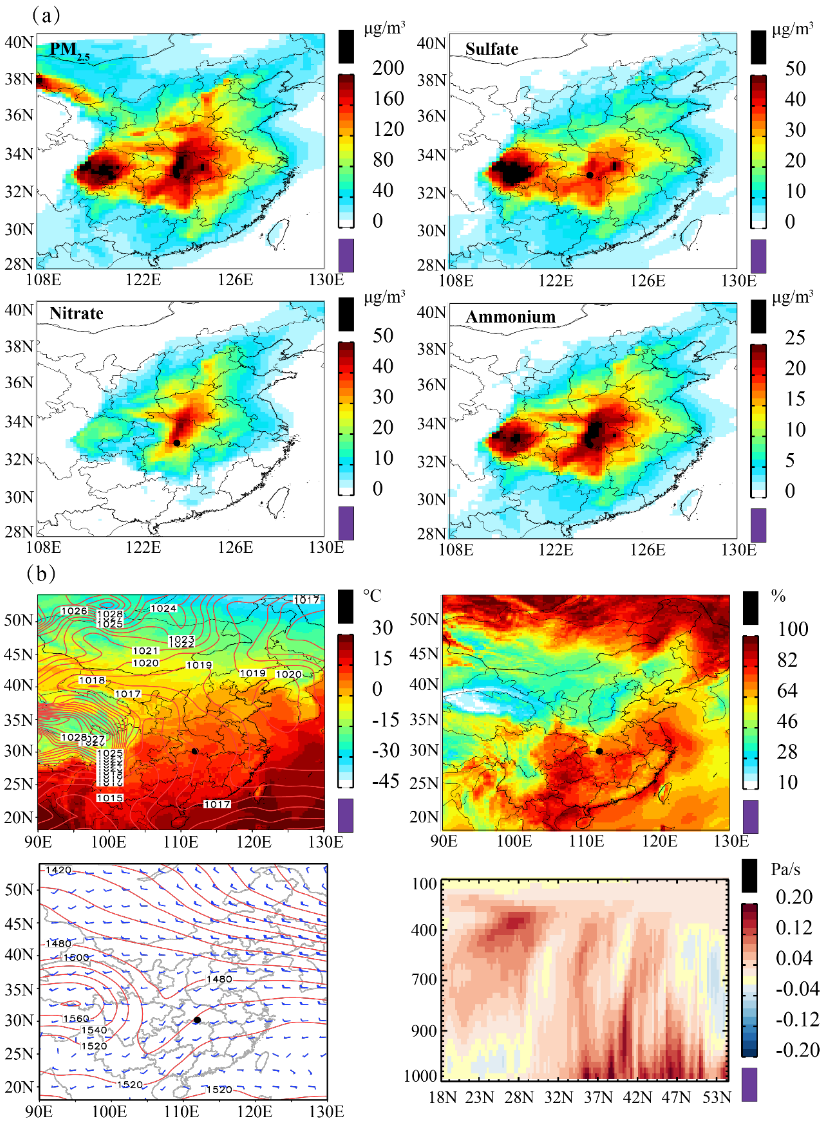

Figure 11 As in Fig. 6 but for C-type synoptic control (26 January - 2 February, 2014).

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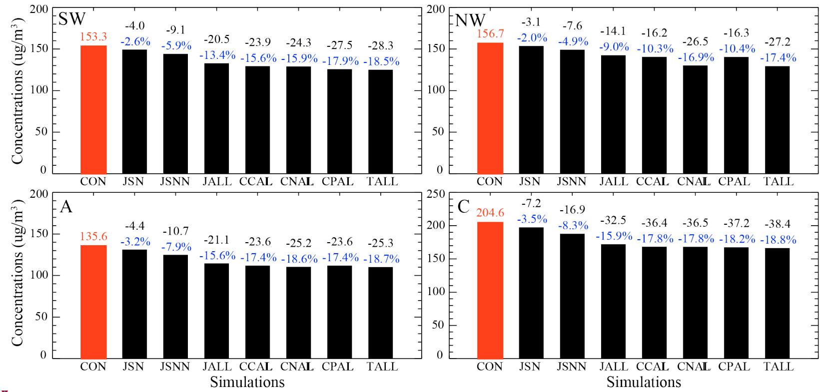

Figure 12 Modeled concentrations (μg/m^3) of $PM_{2.5}$ at Jingzhou in the GEOS-Chem control (red bar) and sensitivity (black bar) simulations for emission reduction, and the differences (black characters for mass concentrations and blue characters for mass percentages) between the sensitivity and the control simulations. The abbreviations of each simulation referred to Table 2.

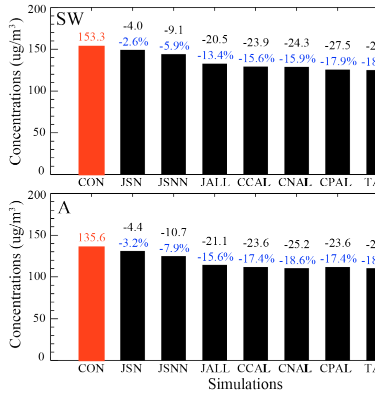

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Table 1 Lamb-Jenkinson circulation types

$\lvert\xi\rvert \leq V$ (Flat airflow type)	$\lvert\xi\rvert \geq 2V$ (Rotating airflow type)	$V < \lvert\xi\rvert < 2V$ (Mixed type)
East (E), Southeast (SE), Southwest (SW), Northwest (NW)	Anticyclone (A), Cyclone (C)	Cyclone-Southeast (CSE), Cyclone-West (CW), Cyclone-Northwest (CNW), Anticyclone-West (AW)

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Table 2 Description of sensitivity simulations by GEOS-Chem model. The NCP, YRD, PRD and SCB are the areas framed in red showed by Fig. 1.

Simulations	Description
CON	Applying the original emission situation in Table S1 and Table S2
XJ0	Emissions of all pollution sources[1] outside Jingzhou are set to be zero
XCC0	Emissions of all pollution sources outside Central China are set to be zero
NCP0	Emissions of all pollution sources over NCP region are set to be zero
YRD0	Emissions of all pollution sources over YRD region are set to be zero
PRD0	Emissions of all pollution sources over PRD region are set to be zero
SCB0	Emissions of all pollution sources over SCB region are set to be zero
JSN	Emissions of SO_2 and NO_x at Jingzhou are reduced by 20%
JSNN	Emissions of SO_2, NO_x and NH_3 at Jingzhou are reduced by 20%
JALL	Emissions of all pollution sources at Jingzhou are reduced by 20%
CCALL	Emissions of all pollution sources over Central China are reduced by 20%
CNALL	Emissions of all pollution sources over Central China and NCP region are reduced by 20%
CPALL	Emissions of all pollution sources over Central China and PRD region are reduced by 20%
TALL	Emissions of all pollution sources over Central China, NCP, YRD, PRD and SCB region are reduced by 20%

1. All pollution sources include emissions of SO_2, NO_x, NH_3, CO, BC, OC and NMVOCs.

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Table 3 Simulated $PM_{2.5}$ concentrations and associated chemical components averaged for the four typical heavy pollution episodes at Jingzhou. Also shown in brackets are the percentages of each component in $PM_{2.5}$.

$PM_{2.5}$ components	Typical heavy pollution episodes			
μg/m^3	11/18-11/25（SW-type）	12/19-12/26（NW-type）	1/14-1/21（A-type）	1/26-2/2（C-type）
Nitrate	30.6 (20.0%)	34.6 (22.1%)	23.4 (17.3%)	42.3 (20.7%)
Sulfate	26.5 (13.4%)	30.7 (19.6%)	27.7 (20.4%)	40.4 (19.7%)
Ammonium	18.8 (12.3%)	21.6 (13.8%)	17.1 (12.6%)	27.1 (13.2%)
Dust	24.4 (15.9%)	22.3 (14.2%)	19.8 (14.6%)	29.2 (14.3%)
BC	10.5 (6.8%)	9.6 (6.1%)	9.5 (7.0%)	13.8 (6.7%)
POA	21.6 (14.1%)	18.9 (12.1%)	18.9 (13.9%)	27.7 (13.5%)
SOA	20.9 (13.6%)	19.0 (12.1%)	19.2 (14.2%)	24.1 (11.8%)
$PM_{2.5}$	153.3	156.7	135.6	204.6

Table 4 The reported concentrations of $PM_{2.5}$ and the three inorganic salts (sulfate, nitrate and ammonium, μg/m^3) in other cities.

References	Site	Time	$PM_{2.5}$	Sulfate	Nitrate	Ammonium
Cao et al., 2012	Beijing	01/03	115.6±46.6	20.0±4.2 (17.3%)	13.1±4.5 (11.3%)	9.4±4.1 (8.1%)
Cao et al., 2012	Qingdao	01/03	134.8±43.0	21.1±7.7 (15.7%)	19.3±9.2 (14.3%)	15.3±5.2 (11.4%)
Cao et al., 2012	Tianjin	01/03	203.1±76.2	32.5±15.1 (16.0%)	25.2±10.3 (12.4%)	22.2±9.8 (10.9%)
Cao et al., 2012	Xi'an	01/03	356.3±118.4	53.8±25.6 (15.1%)	29.0±10.0 (8.1%)	29.8±11.5 (8.4%)
Cao et al., 2012	Chongqing	01/03	316.6±101.2	60.9±19.6 (19.2%)	18.1±6.4 (5.7%)	28.8±8.9 (9.1%)
Cao et al., 2012	Hangzhou	01/03	177.3±59.5	33.4±16.7 (18.8%)	25.7±14.8 (14.5%)	19.1±10.7 (10.8%)
Cao et al., 2012	Shanghai	01/03	139.4±50.6	21.6±12.3 (15.5%)	17.5±8.7 (12.6%)	14.5±5.9 (10.4%)
Cao et al., 2012	Wuhan	01/03	172.3±67.0	31.4±15.6 (18.2%)	22.2±10.7 (12.9%)	18.4±10.2 (10.7%)
Zhang et al., 2011	Xi'an	03/06-03/07	194.1	35.6 (18.3%)	16.4 (8.4%)	11.4 (5.9%)
Huang et al., 2012	Xi'an	01/06-02/06	235.8±125.1	44.8±31.3 (19.0%)	20.5±14.2 (8.7%)	14.5±10.8 (6.1%)
Wang et al., 2020	Jinan	10/17	104±54	14.4±9.2 (13.8%)	33.4±23.2 (32.1%)	13.0±8.3 (12.5%)
Wang et al., 2020	Shijiazhuang	10/17	152±109	19.3±19.6 (12.7%)	42.8±41.1 (28.2%)	18.2±17.1 (12.0%)
Wang et al., 2020	Wuhan	12/17	117±33	13.6±3.2	26.6±11.1	13.1±3.8

				(11.6%)	(22.7%)	(11.2%)
Wang et al., 2016a	Zhengzhou	01/11-02/11	297±160	48±36 (16.2%)	31±19 (10.4%)	21±16 (7.1%)
Wang et al., 2016a	Zhengzhou	01/12-02/12	234±125	23±10 (9.8%)	22±9 (9.4%)	16±5 (6.8%)
Wang et al., 2016a	Zhengzhou	01/13-02/13	337±168	56±39 (16.6%)	39±20 (11.6%)	31±18 (9.2%)
Luo et al., 2018	Zibo	12/06-02/07	224.9±85.4	40.1±19.2 (17.9%)	18.1±9.0 (8.1%)	21.7±10.2 (9.7%)
Wang et al., 2016b	Shanghai	12/11, 12/12, 12/13	73.9±57.5	12.2±9.2 (16.5%)	14.6±12.2 (19.8%)	8.2±6.7 (11.1%)
Xu et al., 2019	Beijing	02/17-03/17	180.5	20.1 (11.1%)	45.6 (25.3%)	22.5 (12.5%)
Xu et al., 2019	Beijing	05/17-09/17	186.7	20.2 (10.8%)	32.4 (17.4%)	17.1 (9.2%)
Xu et al., 2019	Beijing	10/17-11/17	167.5	17.9 (10.7%)	44.5 (26.6%)	20.9 (12.5%)
Zheng et al., 2016	Beijing	03/10-05/10	65.2±65.1	11.1±10.1 (17.0%)	11.1±11.0 (17.0%)	6.8±6.7 (10.4%)
Zheng et al., 2016	Beijing	07/09-08/09	88.9±39.1	23.0±13.9 (25.9%)	16.2±11.8 (18.2%)	11.8±6.8 (13.3%)
Zheng et al., 2016	Beijing	12/09-02/10	84.0±66.6	8.1±8.3 (9.1%)	8.0±9.6 (9.0%)	5.9±7.1 (6.6%)
Zheng et al., 2016	Guangzhou	11/10	73.3±16.5	16.6±4.0 (22.6%)	5.7±3.8 (7.8%)	6.2±2.0 (8.5%)
Zheng et al., 2016	Shenzhen	12/09	64.6±24.7	20.6±3.5 (31.9%)	4.9±3.5 (7.6%)	4.6±1.0 (7.1%)
Zheng et al., 2016	Wuxi	04/10-05/10	82.1±27.0	12.8±3.8	9.9±6.3	7.0±2.0

				(15.6%)	(12.1%)	(8.5%)
Zheng et al., 2016	Jinhua	10/11-11/11	81.9±26.2	18.3±6.7 (22.3%)	12.6±7.0 (15.4%)	10.4±4.1 (12.7%)
Liu et al., 2018	Chongqing	2012-2013	73.5±30.5	19.7±9.6 (26.8%)	6.5±6.2 (8.8%)	6.1±2.7 (8.3%)
Liu et al., 2018	Shanghai	2012-2013	68.4±20.3	13.6±6.4 (19.9%)	11.9±5.0 (17.4%)	5.8±2.1 (8.5%)
Liu et al., 2018	Beijing	2012-2013	71.7±36.0	11.9±8.2 (16.6%)	9.3±7.5 (13.0%)	5.3±2.7 (7.4%)

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