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Supplement of

Effectiveness of emission control in reducing PM$_{2.5}$ pollution in central China during winter haze episodes under various potential synoptic controls

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S1. Emissions in GEOS-Chem global model and nested model over China

Table S1 and Table S2 show a list of emission inventories in the global model and nested model over China, respectively. Global anthropogenic emissions of CO, NO_x and SO_2 in 2013-2014 are from EDGAR (Emission Database for Global Atmospheric Research) v4.2 with a resolution of 0.1° × 0.1°. Monthly global anthropogenic emission of non-methane volatile organic compounds (NMVOCs) are taken from RETRO (REanalysis of the TROpospheric chemical composition) in the year of 2000 with the grid resolution of 0.5° × 0.5°. The monthly emission data of biomass burning comes from GFED4 (Global Fire Emissions Database version 4). Other natural source emissions, such as NO_x from lightning and soil and NMVOCs from biogenic emissions, are calculated on-line by parameterization based on meteorological conditions in the simulations. The parameterization of soil NO_x emission follows Hudman et al. (2012). The NMVOCs biogenic emissions are calculated by MEGAN (Model of Emissions of Gases and Aerosols from Nature) v2.1 according to the monthly averaged MODIS (MODerate resolution Imaging Spectroradiometer) leaf area index.

S2. Optimation in GEOS-Chem model

(1) Optimize PM$_{2.5}$ emissions

The primary particulate matter emitted by human activities is an important source of PM$_{2.5}$ (Pui et al., 2014). Only natural emission sources of PM$_{2.5}$, such as dust and sea salt, are considered in the standard version of GEOS-Chem v11-01 (Yan et al., 2019). According to the emission inventory of MEIC in 2013-2014 and SEEA, this study adds the PM$_{2.5}$ primary anthropogenic emissions in the model.

(2) Increase the proportion of sulfate primary emission

Human activities would lead to the primary emission of sulfate (Fu et al., 2013). At present, there is no global/regional sulfate emission inventory, so the model can not grasp the primary emission of sulfate very well. According to previous observations,

the primary emission concentration of sulfate is about 3% of SO_2 emission concentration (Fu et al., 2013). Thus the primary emission of sulfate in the model is simply parameterized to 3% of SO_2 emission. However, the latest observation shows that the primary emission of sulfate in China is relatively high (Yan et al., 2020). In addition, the primary emission of SO_3 and its conversion to sulfate are not considered in the model, so we increase the primary emission proportion of sulfate to 4.5% of SO_2 emission.

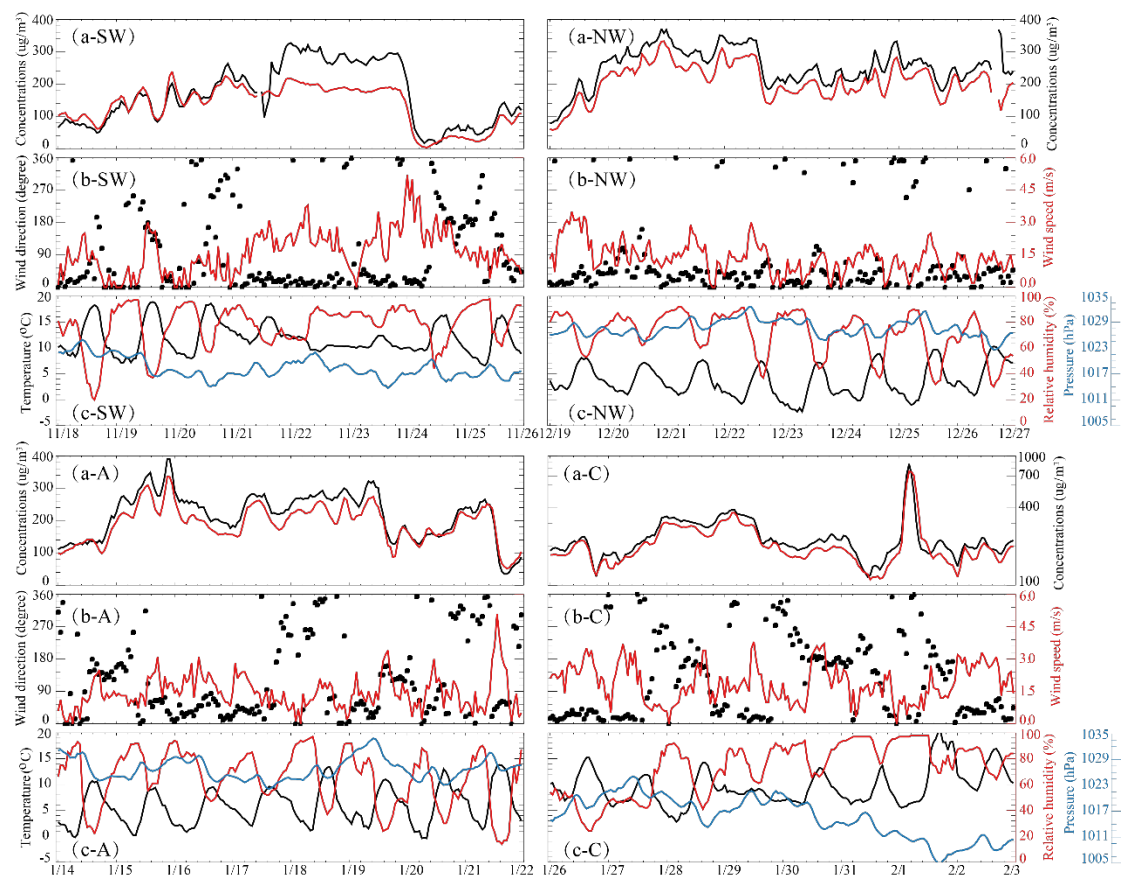Figure S1 (a) Modeled (red lines) and observed (black lines) hourly PM$_{2.5}$ concentration (μg/m^3) at Jingzhou during the four typical heavy pollution processes, forced by SW-type, NW-type, A-type and C-type circulation, respectively. (b) Observed wind speed (red line) and wind direction (black dots). (c) Obseved temperature (black line), relative humidity (red line) and sea level pressure (blue line).
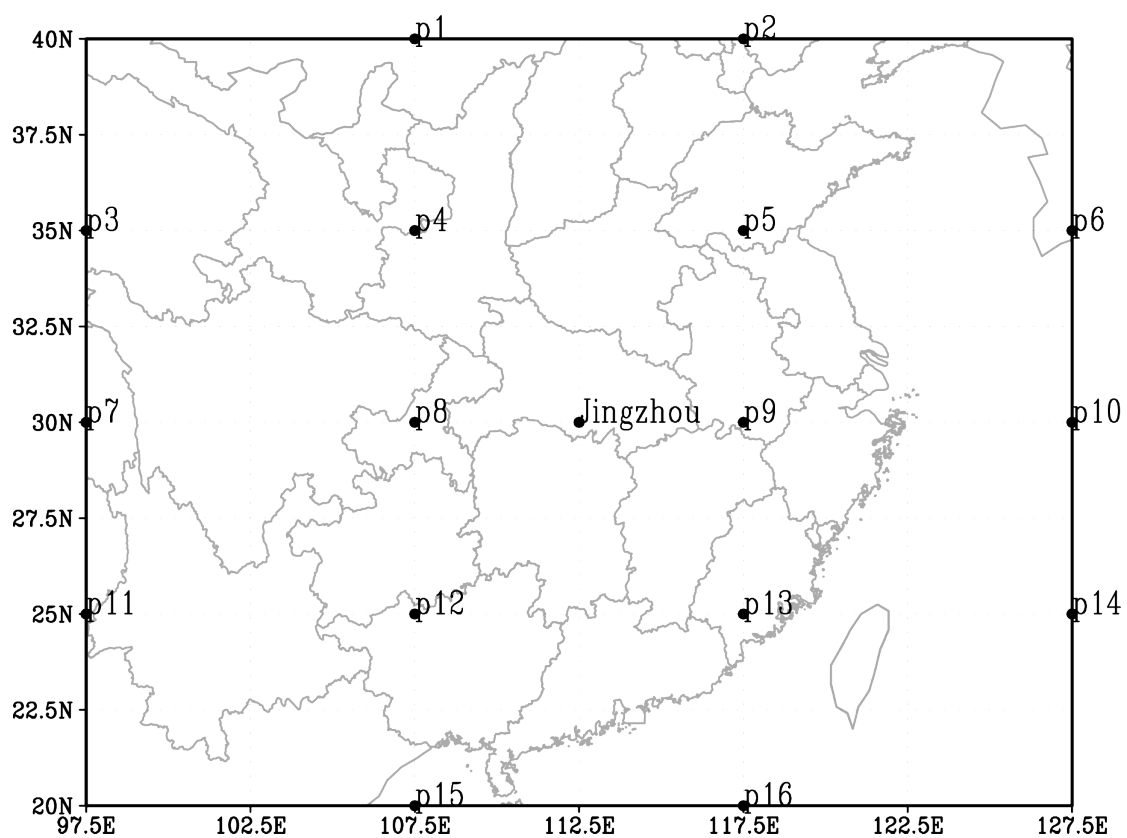
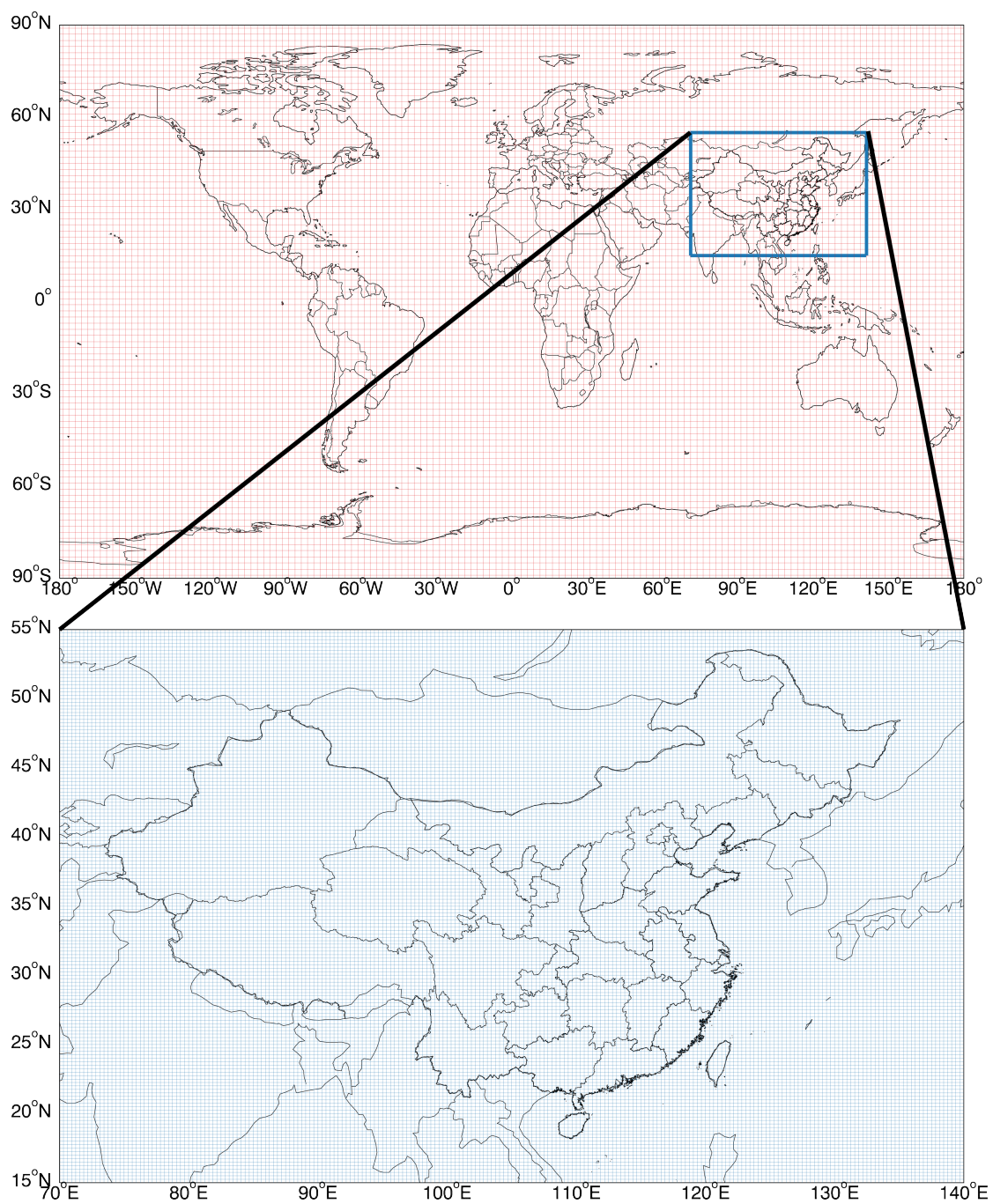Figure S2 Location of the grid points used in circulation classification calculation

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°).
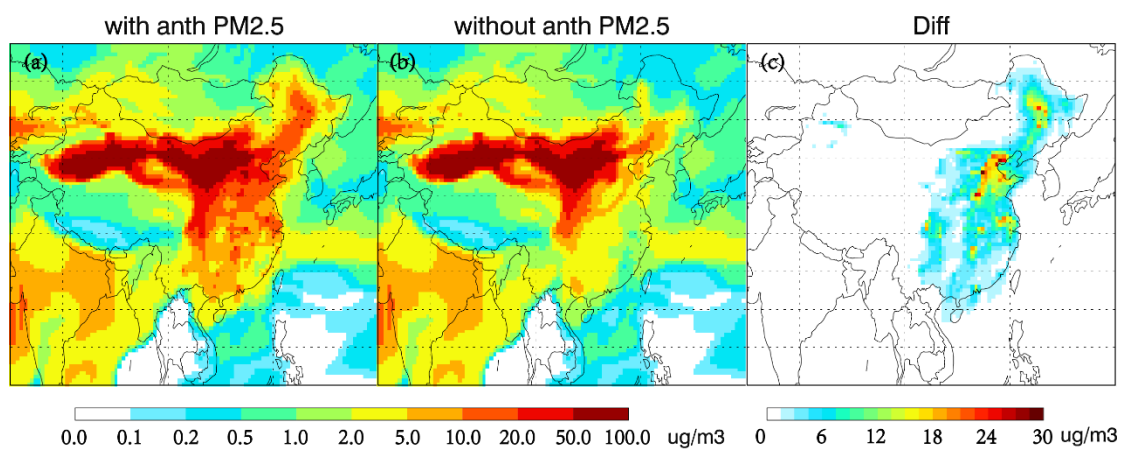
Figure S4 Modeled PM$_{2.5}$ concentrations with and without PM$_{2.5}$ primary anthropogenic emissions. Also shown is the difference between the two simulations.
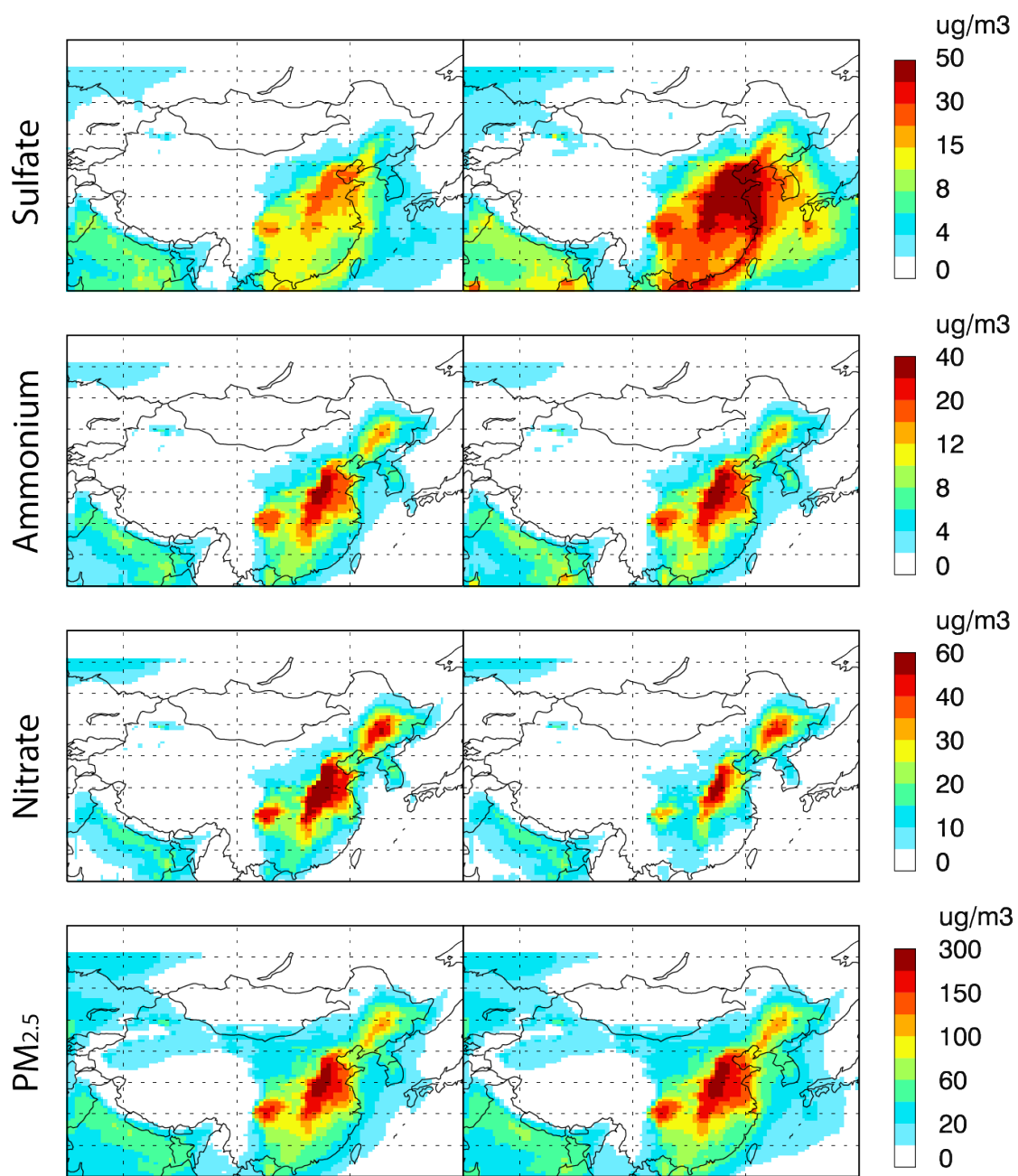
Figure S5 Modeled PM$_{2.5}$, sulfate, nitrate and ammonium concentrations in standard GEOS-Chem v11-01 (left column) and in optimized model version (right column).

Figure S6 Spatial distribution of 500 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.
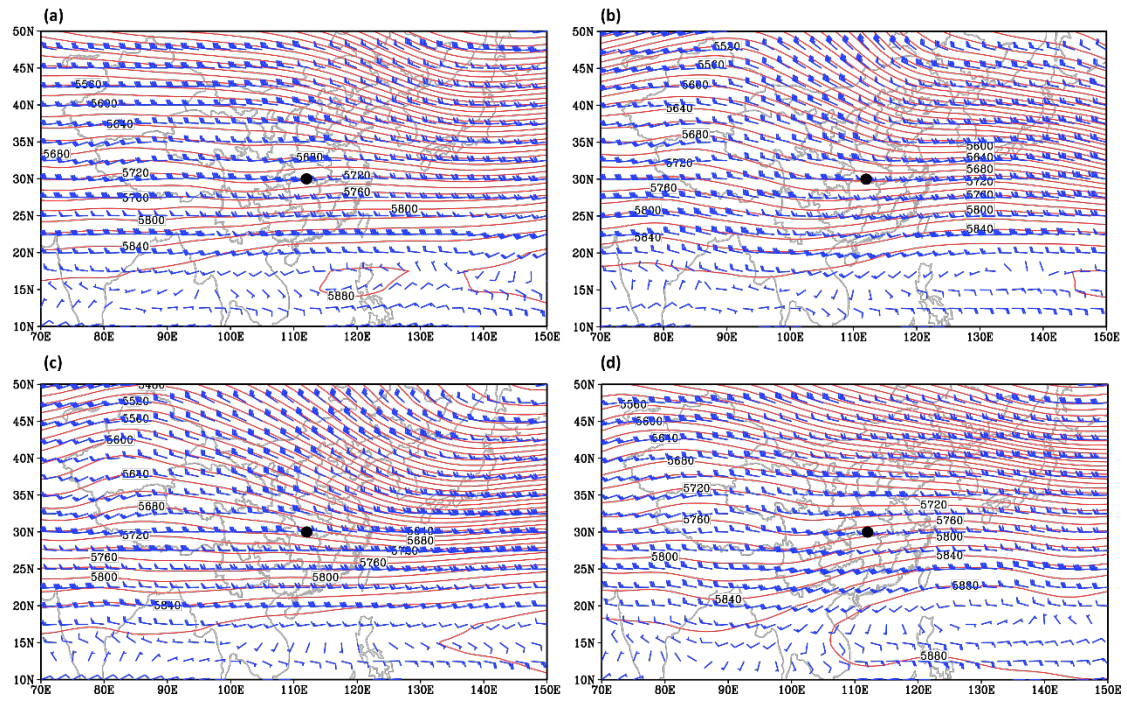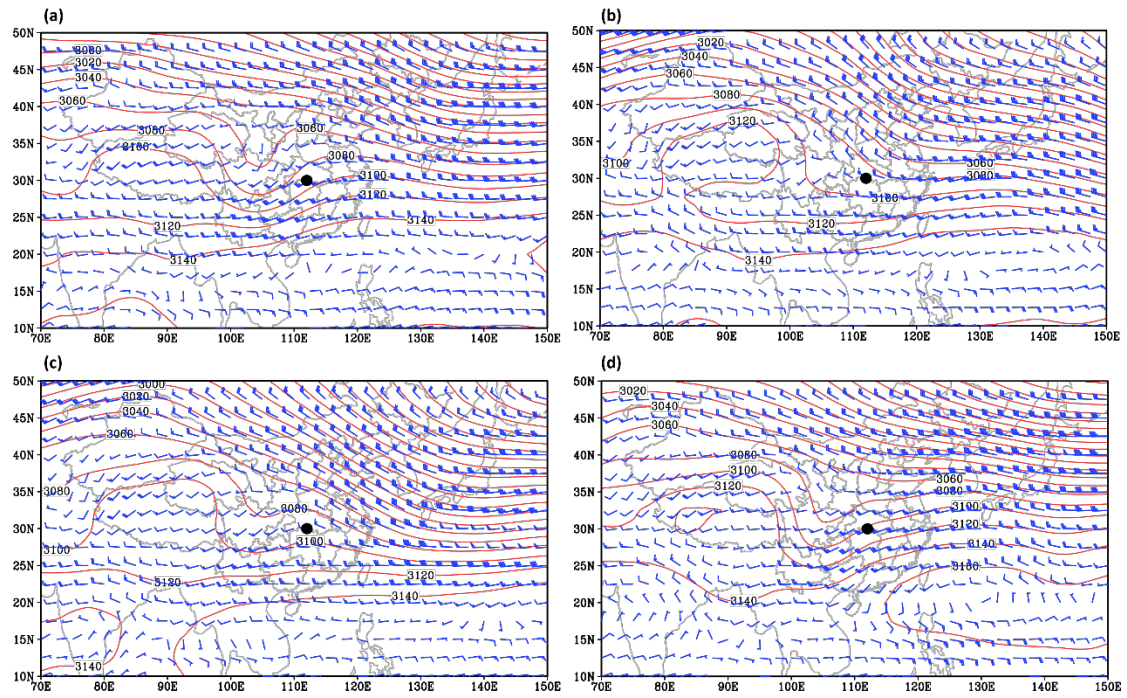
Figure S7 Spatial distribution of 700 hPa geopotential height and wind vector for SW-type (a), NW-type (b), A-type (c) and C-type (d) synoptic control averaged over 2013-2018.

Figure S8 Spatial distribution of PM$_{2.5}$, sulfate, nitrate and ammonium concentrations averaged in four typical heavy pollution processes over Jingzhou simulated by GEOS-Chem control simulation (μg/m^3).
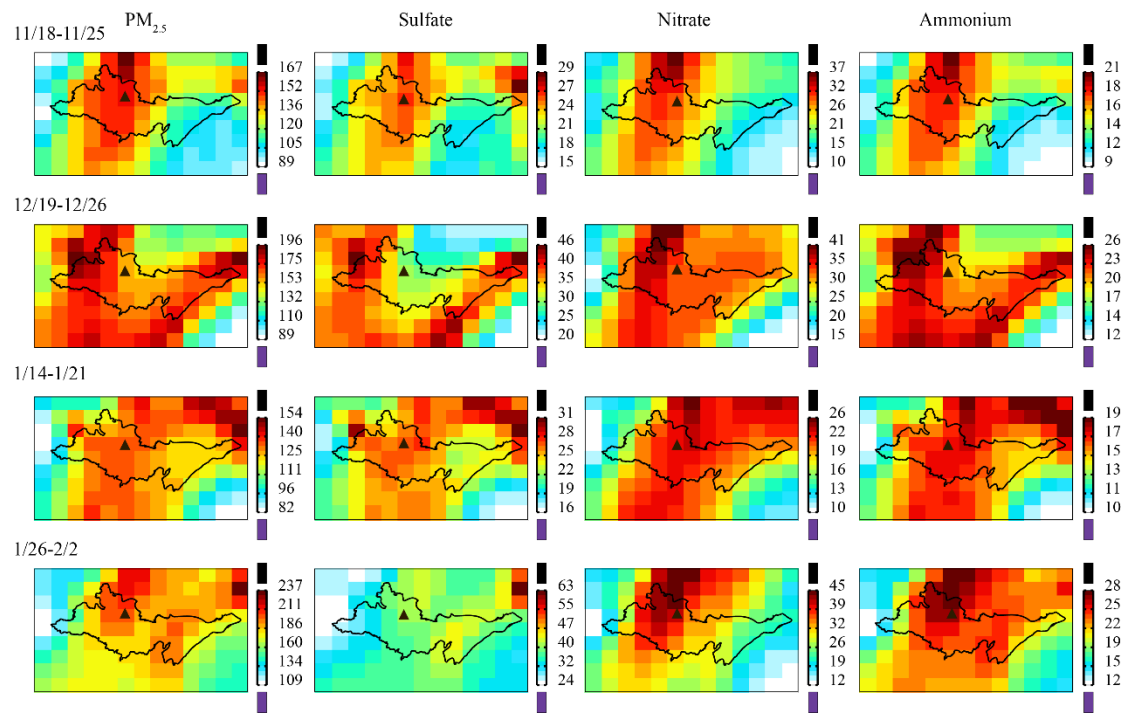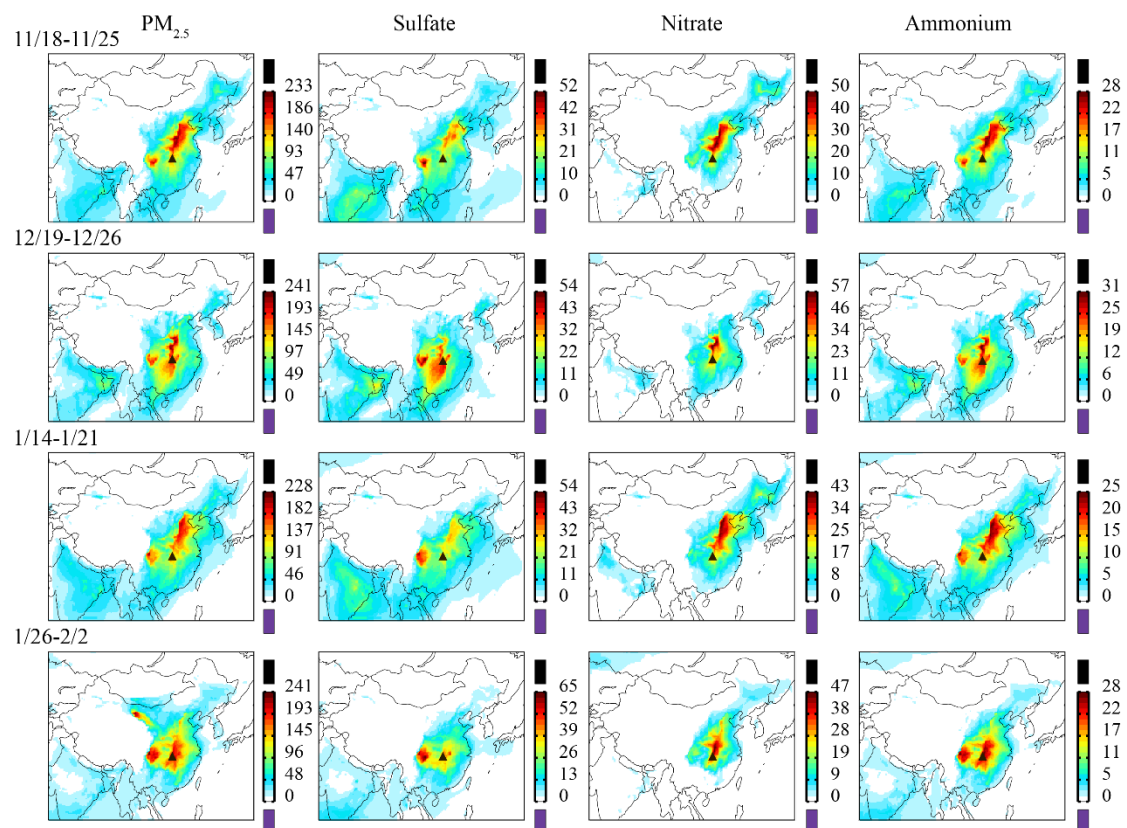
Figure S9 Spatial distribution of PM$_{2.5}$, sulfate, nitrate and ammonium concentrations averaged in four typical heavy pollution processes over China simulated by GEOS-Chem control simulation (μg/m^3).

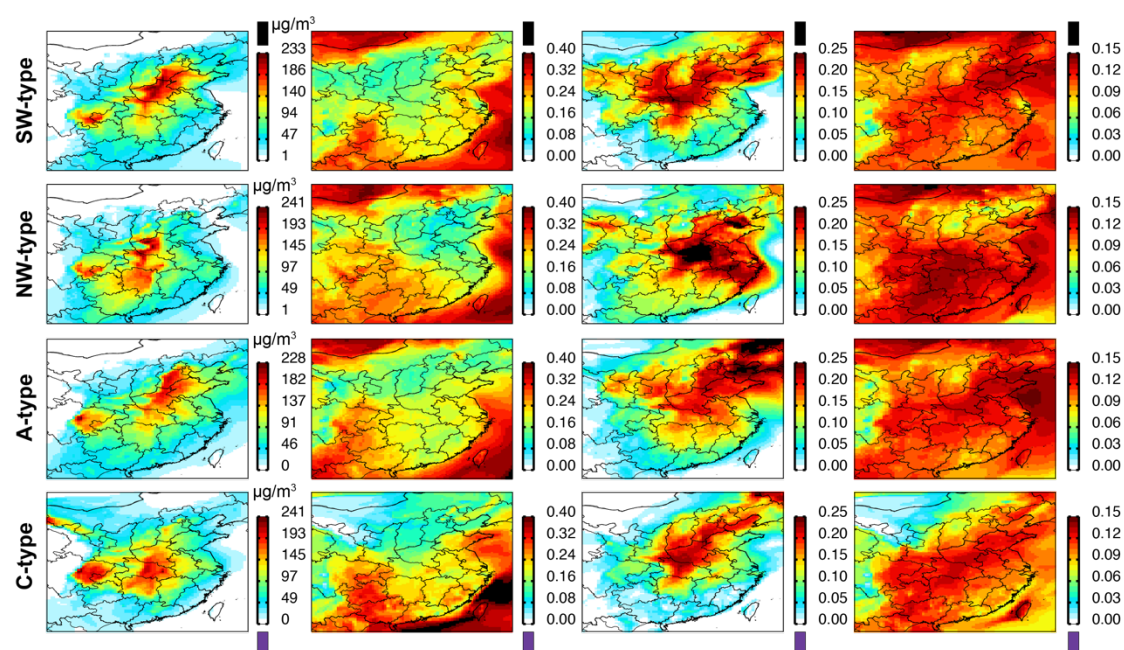Figure S10 Spatial distribution of PM$_{2.5}$ concentrations and the fraction of each inorganic salt (sulfate: second column; nitrate: third column; ammonium: forth column) to PM$_{2.5}$ for these four typical heavy pollution processes simulated by GEOS-Chem control simulation.

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 e 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	NO_x, 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 e 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).

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

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