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The effect of emission source chemical profiles on simulated $PM_{2.5}$ components: sensitivity analysis with the Community Multiscale Air Quality (CMAQ) modeling system version 5.0.2

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This supplement material contains 28 tables and 10 figures.

Table S1 The simulation error of chemical transport models (CTMs) on the components of $PM_{2.5}$ in different studies

$PM_{2.5}$ components	Model	NMB	R	Study area	Period	Reference
SO_4^{2-}	CMAQv4.7.1	-45%	0.73	Eastern China	2010	(Cheng et al., 2015)
NO_3^-		29%	0.82			
SO_4^{2-}	CMAQv4.7.1	-4.5%	0.87	Qing Dao	Jan. 2016	(Zhang et al., 2017)
NO_3^-		10%	0.87			
NH_4^+		-6%	0.9			
SO_4^{2-}	CMAQv5.0.1	-54%	0.6	Northern China	2013	(Zheng et al., 2015)
NO_3^-		-40%	0.8			
NH_4^+		-58%	0.7			
OC		-25%	0.8			
EC		196%	0.6			
SO_4^{2-}	Revised CMAQ	6%	0.7			
NO_3^-		6%	0.8			
NH_4^+		-4%	0.8			
OC		-28%	0.7			
EC		183%	0.6			
SO_4^{2-}	WRF-Chem3.6.1	-84%	0.31	Nanjing	Jan. 2017	(Sha et al., 2019)
		-71%	0.26		Apr. 2017	
NO_3^-		45%	0.51		Jan. 2017	
		67%	0.32		Apr. 2017	
NH_4^+		-34%	0.27		Jan. 2017	
		-13%	0.31		Apr. 2017	
SO_4^{2-}	CMAQv5.0.2	-41%	0.82	Qing Dao	Dec. 2015 ~ Jan. 2016	(Gao et al., 2020)

NO_3^-		41%	0.83			
NH_4^+		-5%	0.83			
SO_4^{2-}	RAQMS	-4%	0.83	Beijing	Feb. to Mar. 2014	(Li et al., 2020)
NO_3^-		-4%	0.77			
NH_4^+		4%	0.81			
OC		-39%	0.92			
EC		-9%	0.81			
SO_4^{2-}	CMAQv5.0.1	-56%~-29%	-	China	2013	(Shi et al., 2017)
NO_3^-		-47%~19%				
NH_4^+		-44%~1				
SO_4^{2-}	CMAQv4.7	-16% and -6%	-	USA	Jan. 2006	(Foley et al., 2010)
		-19%~-0.2%			Aug. 2006	
NO_3^-		-5% and 1%			Jan. 2006	
NH_4^+		13% and 14%			Jan. 2006	
		15% and -6%			Aug. 2006	
OC		-20%			Jan. 2006	
		-49%			Aug. 2006	
EC		-25%			Jan. 2006	
		-32%			Aug. 2006	
SO_4^{2-}	CMAQv4.5.1	-34%~7%	-	USA	Jan. 2002	(Liu et al., 2010)
		-18%~-37%			Jul. 2002	
NO_3^-		16%~118%			Jan. 2002	
		-69%~88%			Jul. 2002	
NH_4^+		-0.5%~61%			Jan. 2002	
		-43%~53%			Jul. 2002	

OC		-4%~13%			Jan. 2002	
		-71%~-64%			Jul. 2002	
EC		-16%~18%			Jan. 2002	
		-39%~38%			Jul. 2002	
SO_4^{2-}	CMAQv4.5.1	5%	0.7	South Eastern USA	Jan. 2002	(Zhang et al., 2013)
	CAMx-4.4.2	33%	0.6			
	CMAQv4.5.1	-39%	0.5		Jul. 2002	
	CAMx-4.4.2	-9%	0.6			
NO_3^-	CMAQv4.5.1	46%	0.8		Jan. 2002	
	CAMx-4.4.2	-21%	0.8			
	CMAQv4.5.1	-62%	0.2		Jul. 2002	
	CAMx-4.4.2	-80%	0.2			
NH_4^+	CMAQv4.5.1	-7%	0.8		Jan. 2002	
	CAMx-4.4.2	-8%	0.7			
	CMAQv4.5.1	-52%	0.7		Jul. 2002	
	CAMx-4.4.2	-45%	0.7			
OC	CMAQv4.5.1	-15%	0.8		Jan. 2002	
	CAMx-4.4.2	-18%	0.8			
	CMAQv4.5.1	-73%	0.7		Jul. 2002	
	CAMx-4.4.2	-47%	0.7			
EC	CMAQv4.5.1	-9%	0.7		Jan. 2002	
	CAMx-4.4.2	5%	0.7			
	CMAQv4.5.1	-47%	0.4		Jul. 2002	
	CAMx-4.4.2	-33%	0.4			
SO_4^{2-}	CMAQv5.0	0.7% and -31%	0.85	USA	1990-2010	(Xing et al., 2015)

		-2%	0.61	Europe		
NO_3^-		56%~59%	0.66	USA		
		-6%	0.70	Europe		
NH_4^+		-13%	0.52	USA		
		34%	0.62	Europe		
SO_4^{2-}	CMAQv4.5	-16%	0.82	USA	2002~2008	(Friberg et al., 2016)
NO_3^-		72%	0.64			
NH_4^+		13%	0.68			
OC		-30%	0.39			
EC		-22%	0.5			
SO_4^{2-}	CMAQv5.0.2	-50%~29%	-	California	2013	(Chen et al., 2020)
NO_3^-		-27%~48%				
NH_4^+		-32%~130%				
OC		-35%~13%				
EC		0~43%				

Table S2 The main model configurations in this work

Model	Description
WRF	Three nested domains covering the inland China with grid resolutions of 36 km × 36 km, 12 km × 12 km and 4 km × 4 km were set for the simulation domains. The initial and boundary conditions for WRF were based on the North American Regional Reanalysis data archived at National Center for Atmospheric Research (NCAR, https://doi.org/10.5065/D6M043C6). In addition, surface (https://doi.org/10.5065/4F4P-E398) and upper (https://doi.org/10.5065/39C5-Z211) air observations obtained from NCAR were used to further refine the analysis data through data assimilation.
CMAQ	The CMAQ model version 5.0.2 was used to simulate air quality over the modeling period. The modeling was conducted from Oct. 1 to Oct.30 in 2018. Gas phase chemistry was based on CB05 (Carbon Bond 05) mechanism, the aerosol dynamics/chemistry was based on aero6 (Sixth-generation modal CMAQ aerosol model) module (cb05tucl_ae6_aq) (Yarwood et al., 2005; Whitten et al., 2010), ISORROPIA II inorganic chemical mechanism (Fountoukis and Nenes, 2007) was adopted. The CMAQ were two nested modeling domains which took Dom2 and Dom3 from WRF, but both indented 6 grids each side to reduce the impact of the boundary effect, and we excluded the first 7 days CMAQ simulation results to minimize the impact of initial conditions. PM size representation was based on three log-normal modes (nuclei, accumulation, coarse) (Chapel Hill, 2012).
MEIC	The Multi-resolution Emission Inventory for China (MEIC) model has been developed and maintained by Tsinghua University since 2010, aiming to build a high-resolution inventory of anthropogenic air pollutants and carbon dioxide emissions in China. Anthropogenic emission data from the monthly MEIC which include five types of emission categories (power plant, industry, residential, vehicle and agriculture) at the year 2017 were used. The inventory contains the emissions of ten major species (including sulfur dioxide (SO_2), nitrogen oxides (NO_x), carbon monoxide (CO), non-methane volatile organic compounds, ammonia (NH_3), fine particulate matter ($PM_{2.5}$), coarse particulate matter (PM_{10}), black carbon (BC), organic carbon (OC) and carbon dioxide (CO_2) (http://meicmodel.org/) (Liu et al., 2017a; Zheng et al., 2018). In addition, the Inventory Spatial Allocate Tool (ISAT) was used in this work (Wang et al., 2019).

Table S3 Power plant source profiles from published literatures in China, SPAP and SPECIATE database

Year	SO_4^{2-}	NO_3^-	Cl^-	NH_4^+	Na	K	Mg	Ca	OC	EC	Fe	Mn	Al	Si	Ti	Other	City/Region	Data source
2005~2006	2.9	0.6	3.4	0.1	1	0.3	0.5	2.5	34	4	2	0.05	1.7		0.1	46.9	Southern China	(Liu, 2007)
2006	23		0.7	4			0.7	5.5	2	0.3	2		4.2			57.6	Shang Hai	.(Zheng et al., 2013)
2009~2013	0.8	0.2	0.1		0.3	0.8		1.7	0.3	1.8	2		15.1	20.3	0.6	56	Shijiazhuang	(Qi et al., 2015)
2012	5.8	1.5	1.8	0.6	1	2.6	2	3	13	0.4	0.9	0.03	2.3	12.8	0.1	52.2	Beijing	(Ma et al., 2015)
2013	2.4	0.2		0.03	2.2	0.2	0.9	8.8	4	0.8	0.8	0.04	5.7	7.5		66.4	Changzhou	(Teng et al., 2015)
2015~2016	8.7	0.9	16.5	4.9	2.1	3.9	1.1	9.6	4.2	0.4	2.3	0.1	1.3	2.4	0.1	41.7	Tianjin	(Bi et al., 2019)
2017	7.8		9.3		0.2	0.1	0.2	3.6	1.4	0.1	1.6		2.2	15.2		58.3	Yantai	(Wen et al., 2019)
	3.2		9.2	3.0	0.5	0.2	1.1	19.3	6.7	0.7	2.6	0.1	3.0	3.0	0.1	47		SPAP, Bi et al. 2019
	10.1	0.1	32.8	14.0	0.2	0.1	0.8	0.9	1.9	0.3	0.2	0.01	0.2	2.1	0.01	36		SPAP, Bi et al. 2019
	0.03	0.3	0.1	0.5	0.03	0.1	0.04	1.1	0.1	0.05	0.2	0.003	0.8	0.6	0.03	96		SPAP, Bi et al. 2019
	9.4	2.1	3.9	1.5	0.9	0.7	3.2	33.4	0.1	0.1	2.7	0.1	4.2	0.8	0.04	37		SPAP, Bi et al. 2019
	30.7	3.8	1.1	15.4	0.8	0.4	1.6	3.4	27.2	4.3	3.0	0.1	9.7	3.8	0.3			SPAP, Bi et al. 2019
	26.4	3.7	1.0	13.6	1.4	0.6	3.1	6.4	23.2	3.5	3.6	0.1	15.7	12.1	0.4			SPAP, Bi et al. 2019
	0.5	0.1	0.04	2.4		0.3	0.3	1.7	1.1	1.4	2.9	0.04	4.8	4.8	0.3	79		SPAP, Bi et al. 2019
	0.3	0.02	0.1	0.05		0.6	0.5	3.2	1.9	4.0	3.5	0.03	8.0	11.3	0.5	66		SPAP, Bi et al. 2019
	0.8	0.1	0.4	0.03		0.5	0.4	4.8	2.1	3.8	3.8	0.1	6.9	6.2	0.4	70		SPAP, Bi et al. 2019
	1.7	0.1	1.5	0.1	0.1	0.9	0.8	6.8	1.9	3.4	5.8	0.2	13.8	11.9	0.7	50		SPAP, Bi et al. 2019
	0.9	0.04	0.3	0.03	0.2	1.1	1.0	8.0	2.6	3.3	7.6	0.2	14.9	10.1	0.7	49		SPAP, Bi et al. 2019
			0.03	0.2	0.4	0.6	0.5	5.3	7.9	5.1	2.2	0.03	7.1	20.3	0.3	50		SPAP, Bi et al. 2019
	2.7	0.3	0.4	0.7	0.6	0.6	0.6	5.0	6.1	10.5	2.9	0.03	5.1	14.2	0.3	50		SPAP, Bi et al. 2019
			0.03	0.1	0.7	0.6	0.9	12.3	5.5	8.5	2.6	0.04	6.5	13.6	0.3	48		SPAP, Bi et al. 2019

	1.8	0.3	0.2	0.2	0.3	0.4	0.4	3.4	2.0	5.8	2.6	0.04	7.3	20.0	0.4	55		SPAP, Bi et al. 2019
	0.1		0.0	0.2	0.4	0.4	0.4	3.6	2.6	3.6	2.2	0.03	6.3	20.0	0.3	60		SPAP, Bi et al. 2019
1987	10.2		0.1	0.3		0.5		3.5		4.3	2.9	0.03	6	9.0	0.4	62.9	Colorado	3190
1987	2.1		0.1	0.3		0.5		2.6	4.4	6.7	2.7	0.03	6.4	9.1	0.4	64.7	Colorado	3191
1987	18.2	0.1	0.1	0.4		0.4		4.3	1.9	1.9	3.1	0.02	5.5	8.9	0.5	54.6	Colorado	3192
1987	2.4		0.1	0.3		0.8		7.2	2.9	1.2	4.7	0.06	9	12.0	0.5	58.9	Colorado	3194
1995	27.3	0.2	0.2	2.0	2.1	0.8	2.4	3.8	3.2	2.2	3.3	0.12	4.2	7.8	0.2	40.1	Colorado	3687
1995	15.4	0.4	1.7	0.2	0.3	0.1	0.3	10.0	1.9	2.5	0.7	0.01	1.3	2.3	0.01	62.9	Colorado	3691
1995	7.7	0.2	1.6	6.6	0.1	0.4	0.5	2.3	11.7	1.7	1.9	0.01	5.4	9.0	0.4	50.5	Colorado	3700
1997	1.5		0.3		0.6	2.0	1.9	4.0	8.7	0.4	1.9	0.03	19.7	23.9		35.2	South Africa	3987
1999	10.2	1.6	3.8	0.3	3.8	0.6	1.2	4.3	70.3	0.01	0.8	0.03	1.2	1.9	0.1	0.03	Texas	4290
1999	71.1		0.2	5.5	0.1	0.3		5.4	1.0	0.2	2.3	0.08	2.8	8.5	0.5	2.2	Texas	4307
1999	41.5	0.1		0.8	0.2	0.6		24.8	3.6	0.9	4.4	0.3	2.1	12.5	1.4	6.7	Texas	4310
1999	4.3	1.3		0.1	0.2	0.1	1.7	21.8	0.7		3.7	0.1	7.4	7.6	1.0	50.0	Texas	4315
2002	5.7	1.0	0.3	0.5	0.4	0.2	1.3	16.1	55.7	2.4	2.9	0.03	5.6	6.1	0.6	1.2	Texas	4368
2002	46.2	0.1	1.1	5.1	0.1	0.5	0.1	11.1	10.3	0.1	3.7	0.2	6.5	13.9	0.8	0.2	Texas	4371
2002	6.4	0.7	0.0	0.1	0.2	0.3	1.5	18.8	1.5	1.4	3.5	0.1	6.8	9.1	1.0	48.6	Texas	4317
2006	12.7	0.2	0.07	0.4	0.1	0.4	0.5	3.7	2.6	1.9	2.7	0.02	5.4	8.9	0.4	60.0		91041
2010	0.4				3.9	0.3	0.0	8.5			2.0		9.5	11.2	0.6	63.7	Canada	95518

Note:

1. The values under different components are the weight percentage in $PM_{2.5}$, %; the number under data source represents the code number in speciate_5.0_0;

2. SPAP: Database of Source Profiles of Air Pollution (SPAP, http://www.nkspap.com:9091/), measured by State Environmental Protection Key Laboratory of Urban Ambient Air Particulate Matter Pollution Prevention and Control & Tianjin Key Laboratory of Urban, Nankai University. SPAP data used in this table were deposited

to the Mendeley data repository and can be freely downloaded from https://doi.org/10.17632/x8dfshjt9j.2, Bi et al., 2019.

Table S4 Industrial process (Sintering) source profiles from published literatures in China, SPAP and SPECIATE database

Year	SO_4^{2-}	NO_3^-	Cl^-	NH_4^+	Na	K	Mg	Ca	OC	EC	Fe	Mn	Al	Si	Ti	Other	City/Region	Data source
2006	23		20.0	1.7	2.2	17.2	0.2	13	13		1.2	0.0				9.0	Shanghai	(Zheng et al., 2013)
2007	22	0.1	8.4	0.6	3.1	22.6	1.1	7	11	2.6	4.8	0.1	3.8	6.8	0.3	5.6		(Ma, 2009)
2007	6	0.1	7.1	0.1	4.3	3.2	3	15	9	3.3	30.1	0.7	5.9	9.2	0.4	2.4		
2012~2013	3	0.0	3.1	0.2	1.6	25.8	0.2	2	13	5.2	6.2	0.1	0.6	6.8		32.6		(Zhao, 2014)
2012~2013	7	0.2	6.6	0.1	0.4	7	0.5	5	10	3.4	25.3	0.2	2.9	9.2		22.7		
2012~2014					1.9	1.2	1.3	4	2		37.2	0.1	7.1	20.3	0.8	24.7	Guiyang	(Wang et al., 2016)
2015	13	0.5	23.2	8.9	2	13.9	0.1	17	6	0.3	2.7		0.1	0.1		12.7	Jing-Jin-Ji	(Guo et al., 2017)
2017	2	9	4	2	1	1	3	33	4	0.3	3	0.1	4	1	0.04	34		SPAP
2017	0.03	6	7	6	1	5	0.2	1			1	0.02	0.01	1	0.01	71		SPAP
2017	1	0.4	9	2	0.3	3	0.5	14	0.3	3	7	0.04	0.2	2	0.004	58		SPAP
2018	2	0.1	1	0.3	0.0004	1	0.4	13			35	1	1	5		41		SPAP
1988			0.3			0.3		13			27.5	0.7	2.6	6.4	0.3	49.2		283042.5
1989	20		17.0			20.0		1			13.0	0.6				28.8		283012.5
2009	10		17.0		13.0	21.0	0.2	1	3	0.2	6.9	0.3	0.1	1.2		26.8		91139

Note:

1. The values under different components are the weight percentage in $PM_{2.5}$, %; the number under data source represents the code number in speciate_5.0_0

2. SPAP: Database of Source Profiles of Air Pollution (SPAP, http://www.nkspap.com:9091/), measured by State Environmental Protection Key Laboratory of Urban Ambient Air Particulate Matter Pollution Prevention and Control & Tianjin Key Laboratory of Urban, Nankai University.

Table S5 Industrial process (Iron-making) source profiles from published literatures in China, SPAP and SPECIATE database

Year	SO_4^{2-}	NO_3^-	Cl^-	NH_4^+	Na	K	Mg	Ca	OC	EC	Fe	Mn	Al	Si	Ti	Other	City/Region	Data source

2007	12	2.6	6.6	1.1	4	4.4	2.9	6.5	12	0.8	14	0.4	10	17	0.7	5.5		(Ma, 2009)
2012~2013	29	1.3	1.5		2.8	12.2	0.8	4	5	1.3	32	0.3	1.2	2		7.6		(Zhao, 2014)
2014~2015	11	4.8		5.5	1.3	1.7	3.6	7.7	16	7	9	0.1	3.5	4	1.4	23.6		(Liu et al., 2017b)
2015	7	0.5	1.6	2.3	0.9	3.2	0.2	2.5	4	4.2	63	0.8	0.3	1	0.2	8.3	Jing-Jin-Ji	(Guo et al., 2017)
2018	2		0.4	1.7				8.7	6	0.8	25			7		49.5	Wuhan	(Wen et al., 2018)
	6.1	1.6	5.2	2.7	1.7	3.3	1.3	7.7	8.2	7.8	12.8	0.3	2.0	2.7	0.2	37		SPAP
2017	2	0.02	0.2	1	0.2	1	0.1	6	6	3	17	0.1	1	1	0.02	62		SPAP
1989			2.5		2.0	3.2		2			6	1.70	1.3	2	0.5	79.5		282012.5
1989			0.9		1.3	3.0		1.0			15	4.50	1.1	24	0.1	49.1		282022.5
1989			1.7		1.7	3.1		1.3			10	3.1	1.2	13	0.3	64.2		900102.5
2006	2	0.2			1.3	1.9	3.1	6.2	3	0.4	32	3.60	0.7	3	0.2	42.0		91011
2009	6	0.5	0.8		1.2	2.7		0.9	6	0.9	14	4.10	1.0	22	0.1	39.1		91157

Note:

1. The values under different components are the weight percentage in $PM_{2.5}$, %; the number under data source represents the code number in speciate_5.0_0

2. SPAP: Database of Source Profiles of Air Pollution (SPAP, http://www.nkspap.com:9091/), measured by State Environmental Protection Key Laboratory of Urban Ambient Air Particulate Matter Pollution Prevention and Control & Tianjin Key Laboratory of Urban, Nankai University.

Table S6 Industrial process (Steelmaking) source profiles from published literatures in China, SPAP and SPECIATE database

Year	SO_4^{2-}	NO_3^-	Cl^-	NH_4^+	Na	K	Mg	Ca	OC	EC	Fe	Mn	Al	Si	Ti	Other	City/Region	Data source
2010	0.9		1.1		1	1.3	3.5	8.7	4.2	1.6	16.6	0.1	5.3	8.9	0.3	46.5	Jincheng	(Cui, 2011)
2012~2013	14.1	0.5	2.2	0	2.8	11.1	1.6	2.2	2.3	1.5	6.4	3.5	0.6	2.5		48.7		(Zhao, 2014)
2012~2013	5.6	0.2	3.6	0	0.4	3.6	0.3	0.9	4.1	0.1	57.9	0.9	0.4	2.9		19.1		
2015	0.8		2.3		1.1	3	0.6	7.0	0.3		72.7	2.5	0.3	1.2	0.2	8	Jing-Jin-Ji	(Guo et al., 2017)
2018	1.4		0.3	2.0				20.3	8.8	0.7	8.2			11.0		47.3	Wuhan	(Wen et al., 2018)
2014	4	1	2	0.2					12	41		0.1	0.2	3	0.08	36		SPAP

2015	1	0.1	1	0.01		1	1	16	5	6	4	0.2	0.5	1	0.08	63		SPAP
	12	1	30	14	2	14	0.4	1	5	0.4	1	0.1	1	2	0.19	16		SPAP
1989	40.0	1				5.0		0.6			11.0	0.60		9.9		32.3		283032.5
1989	2.5		1.9		1.3	0.9	6.5	6			32.0	8.70	0.7	5.0	0.2	34.1		283052.5
1989			0.5			2.5		25			21.0	0.30	0.9	1.6	0.2	48		283062.5
2004	0.7		30.0		13.0	22.0	0.2	0.6	2.7	0.2	0.9	0.0	0.1	1.2		28.39	South Africa	3991
2004			0.5			0.3		22.0			12.0	0.60	0.9	3.0	0.2	60.5	Ohio	3547
2009	8.0		0.5			2.5		25.0			21.0	0.30	0.9	1.6	0.2	40		91179

Note:

1. The values under different components are the weight percentage in $PM_{2.5}$, %; the number under data source represents the code number in speciate_5.0_0

2. SPAP: Database of Source Profiles of Air Pollution (SPAP, http://www.nkspap.com:9091/), measured by State Environmental Protection Key Laboratory of Urban Ambient Air Particulate Matter Pollution Prevention and Control & Tianjin Key Laboratory of Urban, Nankai University.

Table S7 Industrial process (Cement) source profiles from published literatures in China, SPAP and SPECIATE database

Year	SO_4^{2-}	NO_3^-	Cl^-	NH_4^+	Na	K	Mg	Ca	OC	EC	Fe	Mn	Al	Si	Ti	Other	City/Region	Data source
2001	14				1.7	1.4	0.4	36	2	0.3	1.7	0.0	1.2	4.8	0.1	36.4	Hongkong	(Ho et al., 2003)
2006	3	0.3	0.2	0.3	0.2	2.7	0.5	24	3	0.7	3.7	0.1	5	6.2	0.4	49.7	Hangzhou	(Bao et al., 2010)
2014~2015	10	1.8		2.7	0.5	0.8	1.7	11	14	2.1	5.5	0.0	4.6	10.1	0.3	34.9		(Liu et al., 2017b)
2014~2015	16	0.7		0.6	0.7	0.5	1.8	12	5	6.2	4.7	0.1	3.1	9.4	0.2	39.2		
		0.4	0.2		0.1	2.3	0.8	59	20	1.9	5.3	0.3	2	5.5	0.4	1.4	Jing-Jin-Ji	(Ye et al., 2017)
			0.2		0.1	1.1	0.6	64	23	0.6	3.9	0.0	1.3	3.8	0.2	1.2	Jing-Jin-Ji	
	0.2	0.5	0.4		0.1	2.1	0.9	52	29	2.2	3.5	0.0	2.2	5.9	0.2	1.2	Jing-Jin-Ji	
		1.2	0.2		0.1	1.8	1.5	68	9		4.3	0.1	3.2	8.2	0.3	1.6	Jing-Jin-Ji	
2017	21	0.7	2.2	4.5	1.2	0.8	1.4	3	2	0.5	3.2	0.1	5.2	11.4		42.8	Wuhan	(Gong and Luo, 2018)
	4.9	0.4	0.5	1.6	1.4	1.1	1.5	19.1	4.9	1.9	2.3	0.1	3.5	7.0	0.1	50		SPAP
2016	4	0.3	0.1		0.3	1	1	31	1	5	2	0.03	2	9	0.2	42		SPAP

2016	1	2	1	2		0.4	0.3	15	4	0.2	2	0.04	3	0.3	0.03	69		SPAP
2017	1	2	1	9	9	1	2	18			3	0.05	3	6	0.1	44		SPAP
2017	0.5	0.02	0.2	1	0.3	0.5	1	17	10	1	1	0.03	1	2	0.02	66		SPAP
1989	18	0.2	7.8		2.3	5.4	0.2	10	5	0.2	0.9	0.0	4.3	8.4	0.3	36.6		272032.5
1997	0	0.1	0.1	0.0	0.0	0.3	0.1	30	14	0.0	1.7	0.0	0.7	2.8	0.0	49.0	Mexico	4087
1999	38	4.6	3.9	1.2	2.4	21.8	0.0	10	12	1.0	1.1	0.1	0.8	3.1	0.1	0.2	Texas	4333
2002	31	8.9	7.1	2.4	2.3	11.6	0.1	18	13	3.0	1.3	0.1	1.1	4.3	0.1		Texas	4378
2006	18	4.7	3.2	2.4	2.3	7.0	0.1	17	13	3.0	0.7	0.1	1.1	4.3	0.3	23.5		91004
2009	18	4.7	3.1	2.3	2.3	6.9	0.1	17	13	2.9	0.7	0.1	1.1	4.2	0.3	24.4		91127

Note:

1. The values under different components are the weight percentage in $PM_{2.5}$, %; the number under data source represents the code number in speciate_5.0_0

2. SPAP: Database of Source Profiles of Air Pollution (SPAP, http://www.nkspap.com:9091/), measured by State Environmental Protection Key Laboratory of Urban Ambient Air Particulate Matter Pollution Prevention and Control & Tianjin Key Laboratory of Urban, Nankai University.

Table S8 Transportation sector (Heavy duty gasoline) source profiles from published literatures in China, SPAP and SPECIATE database

Year	SO_4^{2-}	NO_3^-	Cl^-	NH_4^+	Na	K	Mg	Ca	OC	EC	Fe	Mn	Al	Si	Ti	Other	City/Region	Data source
2005~2006	1				0.8	0.3	0.3	2.4	60	24	0.4		0.2	1	0.1	9.7	Tianjin	(Zhang, 2007)
2009	2	0.2		0.6	0.35			0.71	40	22			1	3.1		30.0	Dongying	(Kong, 2012)
2012	6	1.4	1.1	1.8	2	0.9	0.9	1.5	52	24	1		0.2	0.3		7.6	Pearl River Delta	(Feng, 2013)
2012	1	0.7	0.5	0.1	0.7	0.3	0.1	0.9	29	61	0.6					4.7	Hubei	(Zhang et al., 2015)
2012	1	0.6	0.6	0.1	0.6	0.3	0.1	0.6	32	59	0.7					4.0	Hubei	
2014~2015	9	1.9	0.3	2.9	0.6	0.5	0.2	0.8	19	43	0.3		0.1			21.3	Hengshui	(Wang et al., 2015)
2014~2015	9	2.7	0.6	2.2	0.7	0.6	0.2	0.6	16	40	0.4		0.3			26.9	Hengshui	
2014~2015	8	1.9	0.4	4	0.5	0.5	0.3	0.8	20	39	0.3		0.2			24.6	Hengshui	
2015		1.0	0.2					0.2	31.4	19.8	0.1	0.01	0.3	0.8	0.002	46.2		SPAP

2015		1.0	1.0					0.2	41.6	24.4	0.1	0.01	0.4		0.001	31.3		SPAP
1989	5				0.8	0.2	0.9	0.7	21	55	0.6	0.0	1.0	1.6	0.0	13.3	-	322022.5
1989			0.0			0.0		0.1	36	52			0.0	0.0	0.0	11.9	-	322032.5
1990			0.0					0.1	36	52			0.0	0.0	0.0	11.9	-	322072.5
1999	0	0.3	0.3	0.1	0.2	0.1		0.2	33	41	0.1	0.0	0.1	0.6	0.0	23.9	Los Angeles	322082.5
2000	30	0.8	1.4	5.1		0.0		3	16	33	0.1	0.01		0.6	0.2	10.0	Ottawa	4750
2000	1	0.1	0.1	0.1				0	44	46		0.06	0.1	0.5		8.0	Ottawa	4749
2001	1		0.0	0.2	0.1	0.0	0.0	0	25	63	0.1		0.0	0.5		10.4	California	4860
2005	3	0.2	0.0	1.0		0.0	0.0	0	15	70	0.1	0.00		0.2		10.1	Los Angeles	4972
2005	2	0.3	0.1	0.4	0.0	0.0	0.0	1.2	62	30	0.3	0.0	0.0	0.0		3.3	Los Angeles	4978
2008	40				0.9	0.0	0.1	0.4	49	7	0.4	0.01	0.1	0.1	0.0	2.4		5679
2012	1	15.9		1.2	0.1	0.0		0.4	52	14	0.2	0.01	0.1	0.1	0.0	15.2		95334

Note:

1. The values under different components are the weight percentage in $PM_{2.5}$, %; the number under data source represents the code number in speciate_5.0_0

2. SPAP: Database of Source Profiles of Air Pollution (SPAP, http://www.nkspap.com:9091/), measured by State Environmental Protection Key Laboratory of Urban Ambient Air Particulate Matter Pollution Prevention and Control & Tianjin Key Laboratory of Urban, Nankai University.

Table S9 Transportation sector (Light diesel) source profiles from published literatures in China, SPAP and SPECIATE database

Year	SO_4^{2-}	NO_3^-	Cl^-	NH_4^+	Na	K	Mg	Ca	OC	EC	Fe	Mn	Al	Si	Ti	Other	City/Region	Data source
2002	5.9	1.0	0.2	0.5	0.0	0.0	0.0	0.0	10	21	0.2	0.0	0.0	0.0	0.0		Yantai	(Cui et al., 2017)
2009	15	1.2		1.8	0.9				38	6			1.0	2.5			Dongying	(Kong, 2012)
2009~2015	0.1	1.9	0.3	0.2	0.3	0.0	0.0	0.2	36	24	0.1	0.0	0.2	0.6	0.0		Fen-Wei plains	(Hao et al., 2019)
2013~2014	1.1	0.1	0.3	0.0		0.1		0.1	16	24	1.2	0.0	0.7	0.6	0.2		-	(Liu et al., 2018)
2015	0.3	3.3	0.2		0.0	0.00	0.1	0.4	32	19	0.3		0.2	0.7	0.4	44		SPAP
	3.0	2.3	0.8	1.5	0.4	0.1	0.4	0.5	34.6	19.2	0.2	0.04	0.3	0.8	0.1	36		SPAP

1987	3.2		0.1	0.6	0.1			0.1	49	43	0.0			0.3	0.0	California	3463
1988	1.4	0.1	0.1	0.3		0.0		0.2	18	78	0.6			0.4		Denver	3219
1989	2.4	0.3	1.6	0.9		0.0		0.2	40	33	0.2	0.0	0.2	0.5		Phoenix	3518
1996	0.4	0.2	0.0		0.0	0.0	0.0	0.1	42	48	0.1		0.0	0.4		Colorado	3960
1997	0.4	0.2	0.1		0.1		0.1	0.1	19	75	0.0		0.0	0.5		Colorado	3878
1998	3.2	2.8	0.6	1.4	0.4	0.8		0.4	52	37	0.3	0.0	0.5	0.8	0.0	Mexico	4014
2001	1.9		0.2	0.5	0.0	0.0	0.2	0.3	37	43	0.2		0.1			California	4842
2007	5.3	1.3	0.4	1.7	0.3	0.3	0.1	0.6	35	46	0.3		0.1	0.3	0.0		8994

Note:

1. The values under different components are the weight percentage in $PM_{2.5}$, %; the number under data source represents the code number in speciate_5.0_0

2. SPAP: Database of Source Profiles of Air Pollution (SPAP, http://www.nkspap.com:9091/), measured by State Environmental Protection Key Laboratory of Urban Ambient Air Particulate Matter Pollution Prevention and Control & Tianjin Key Laboratory of Urban, Nankai University.

Table S10 Transportation sector (Light duty gasoline) source profiles from published literatures in China, SPAP and SPECIATE database

Year	SO_4^{2-}	NO_3^-	Cl^-	NH_4^+	Na	K	Mg	Ca	OC	EC	Fe	Mn	Al	Si	Ti	Other	City/Region	Data source
2009~2015	0.1	2.0	0.3		0.3	0.0	0.1	0.4	48	6	0.5	0.5	0.4	0.4		41.0	Fen-Wei plains	(Hao et al., 2019)
2010	3.9	1.8	0.8		1.1	0.2	0.7	7.8	39	29	2.9	0.1	4.3	4.2	0.1	4.2	Xining	(Kong, 2012)
2010	9.7	1.7	2.4		2.3	1.2	2.8	6.6	29	18	1.6	0.4	8.6	4.9	0.2	10.8	Xining	
2013		1.5	5.4		3.3	0.1	0.4	2.5	54	21	0.6	0.1	0.5			10.7	Xiamen	(Zhang et al., 2016)
2017	5	6	4	2	1	1	0.2	1	33	34	0.4	1	1		0.02	14		SPAP
2017	2	5	5	5	0.5	0.3	0.4	0.4	25	38	0.5	0.5	1		0.02	16		SPAP
	1	3	1	4	0.2	0.1	0.2	1	54	12	0.5	0.5	0.5	0.4	0.01	23		SPAP
2015	0.03	2	0.2		0.0002	0.002	0.01	0.3	69	1	0.1	0.2	0.2	0.2	0.002	26		SPAP
2015	0.1	2	0.2		0.02	0.004	0.01	0.2	77	2	0.1	0.2	0.2	0.2	0.003	18		SPAP
1989	17		1.8			0.01		0.1	24	6	0.1	0.0	0.1	0.2		50.8	-	312302.5

1990			0.3			0.05		0.1	31	15	0.1	1.0	0.6	0.8	0.0	51.1	-	311062.5
1999	3.6	1.8	2.5		1.5				50	23	0.2		0.1	0.0	0.0	17.3	-	311072.5
1999	0.5	0.6	0.9		0.5	0.1			66	8	0.8	0.9	0.7	0.4	0.0	20.7	-	311082.5
2001	9.9	1.1	1.3	5.1	0.1	0.0	0.2	0.1	48	14	0.3	0.0	0.1	3.1	0.0	16.8	California	4895
2004	1.1	0.0			0.8	0.0	0.1	2.1	73	18	0.1	0.0	0.0	0.6	0.0	4.1	Kansas	5570
2005	7.4	0.1			0.1	0.0	0.0	0.2	66	12		0.0	0.1	0.3	0.0	13.8	Kansas	5592
2010	7.2	0.3	0.1	2.8		0.1	0.1	1.4	56	14	1.8	0.0	0.3	0.3	0.0	15.6		8993

Note:

1. The values under different components are the weight percentage in $PM_{2.5}$, %; the number under data source represents the code number in speciate_5.0_0

2. SPAP: Database of Source Profiles of Air Pollution (SPAP, http://www.nkspap.com:9091/), measured by State Environmental Protection Key Laboratory of Urban Ambient Air Particulate Matter Pollution Prevention and Control & Tianjin Key Laboratory of Urban, Nankai University.

Table S11 Residential coal combustion source profiles from published literatures in China, SPAP and SPECIATE database

Year	SO_4^{2-}	NO_3^-	Cl^-	NH_4^+	Na	K	Mg	Ca	OC	EC	Fe	Mn	Al	Si	Ti	Other	City/Region	Data source
2004			10.5		1.3	17.4		0.1	3		0.1		0.1	0.3		67.2	Yangquan	(Ge et al., 2004)
2004						1.2	0.3	0.6	12	4	0.8		1.5	3.4	0.1	76.4	Yangquan	
2009	31	0.8	5.6	0.5	2	10.4	0.1	0.3	36		0.4		0.2			12.4	Dongying	(Kong, 2012)
2009	8	0.6	0.7		1.8	1.3	0.9	2.4	69	6	2.1	0.1	1.9			5.6	Dongying	
2012~2014					0.8	1.5	1.1	1.0	18	8	7.8		11.3	25.4	2.2	23.3	Guiyang	(Wang et al., 2016)
2016~2017	17	0.4	1.9	2.1	0.8	0.4			49							28.0	Xian	(Dai et al., 2019)
2016~2017	29	1.1	1.1	9.9	1.8	0.1	0.2		32	5				0.1		19.5	Xian	
2017~2018	40	0.3	0.9	18	1.6	0.8	0.02	0.6	3	0.4	0.1		0.2	0.4		33.7		SPAP
2017	28	1.2	2	16	4.1	0.5	0.2	2.2	10	0.9	0.3		1.7	4.8		28.1		SPAP
2017	31	0.4	20.1	20	1.5	1	0.1	0.8	3	0.3	0.2		0.1	0.4		21.1		SPAP
	21.7	0.5	2.1	6.0	2.2	1.8	0.2	0.9	23.3	5.8	0.8	0.02	1.5	2.1	0.2	30.9		SPAP

1995	7	0.8	0.3	3.1		1.7	0.1	0.8	45	33	0.2		0.2	0.2		8.4	Colorado	3758
1995	2	0.2	0.1	1		0.1			76	21				0.1			Colorado	3759
1995	3	0.3	0.2	1		0.5		0.2	69	26	0.1		0.1	0.1			Colorado	3761
1997	1	0.2	0.1						56	19				0.1		23.3	South Africa	4007
2009	3	0.3	0.1	1.4		0.5	0.3	1.2	45	24	0.9		0.7	0.7		23.0	Colorado	91155

Note:

1. The values under different components are the weight percentage in $PM_{2.5}$, %; the number under data source represents the code number in speciate_5.0_0

2. SPAP: Database of Source Profiles of Air Pollution (SPAP, http://www.nkspap.com:9091/), measured by State Environmental Protection Key Laboratory of Urban Ambient Air Particulate Matter Pollution Prevention and Control & Tianjin Key Laboratory of Urban, Nankai University.

Table S12 Monitoring sites information

site	District	Lon	Lat	Meridional grid	Zonal grid
1	Ji Zhou	117.444	40.042	43	14
2	Bao Di	117.343	39.526	29	14
3	Wu Qing	117.044	39.373	24	8
4	Ning He	117.817	39.328	25	24
5	Bei Chen	117.185	39.226	20	12
6	Dong Li	117.375	39.151	19	16
7	Xi Qing	117.090	39.078	16	10
8	**Jin Nan**	**117.350**	**39.001**	**15**	**16**
9	Jing Hai	116.915	38.943	12	7
10	Bin Hai New Area	117.707	39.034	17	23

Table S13 The variations of simulated $PM_{2.5}$ and its components in CMAQ_SPE relative to that in CMAQ_SPA for other monitoring sites (%)

Components	ΔP_1	ΔP_2	ΔP_3	ΔP_4	ΔP_5	ΔP_6	ΔP_7	ΔP_8	ΔP_9	ΔP_{10}
SO_4^{2-}	-4	-7	-9	-7	-8	-8	-10	-9	-6	-3
NO_3^-	-3	-4	-5	-5	-7	-7	-11	-8	-6	-5
Cl	-43	-51	-48	-47	-28	-33	-26	-31	-31	-43
NH_4^+	-2	-4	-5	-3	-2	-3	-2	-3	-2	-2
Na	-68	-72	-70	-68	-54	-57	-50	-55	-56	-60
K	-45	-49	-47	-44	-28	-32	-23	-29	-30	-38
Mg	-44	-39	-39	-44	-54	-52	-59	-53	-54	-54
Ca	-47	-43	-44	-46	-52	-51	-53	-51	-51	-52
OC	53	57	58	53	44	45	34	45	44	39
EC	200	211	208	198	166	171	150	167	169	168
Fe	-13	-12	-13	-13	-16	-15	-17	-16	-15	-14
Mn	100	91	108	94	125	114	76	126	110	60
Al	-34	-38	-39	-37	-33	-34	-34	-33	-33	-26
Si	-27	-33	-33	-30	-22	-24	-21	-22	-22	-16
Ti	-59	-62	-63	-61	-59	-59	-59	-58	-58	-50
$PM_{2.5}$	0.2	0.3	0.4	0.4	0.7	0.6	0.8	0.6	0.4	0.2

ΔP_i represent the variations of simulated $PM_{2.5}$ and its components in CMAQ_SPE relative to that in CMAQ_SPA at monitoring site i.

Table S14 The sensitivity coefficients (δ) of simulated components in case DBL at different monitoring sites

Components	δ_1	δ_2	δ_3	δ_4	δ_5	δ_6	δ_7	δ_9	δ_{10}
Al	0.20	0.32	0.39	0.33	0.45	0.44	0.54	0.35	0.21
Ca	0.18	0.29	0.36	0.30	0.42	0.41	0.51	0.32	0.18
Cl	0.12	0.28	0.33	0.28	0.38	0.36	0.47	0.26	0.10
EC	0.18	0.29	0.36	0.30	0.41	0.41	0.51	0.32	0.18
Fe	0.20	0.32	0.39	0.33	0.45	0.44	0.54	0.35	0.21
K	0.18	0.29	0.36	0.30	0.42	0.41	0.51	0.32	0.18
Mg	0.18	0.29	0.36	0.30	0.42	0.41	0.51	0.32	0.18
Mn	0.20	0.32	0.39	0.33	0.45	0.44	0.54	0.35	0.21
Na	0.18	0.29	0.36	0.30	0.42	0.41	0.51	0.32	0.18
OC	0.18	0.30	0.36	0.30	0.42	0.41	0.51	0.32	0.18
Si	0.20	0.32	0.39	0.33	0.45	0.44	0.54	0.35	0.21
Ti	0.20	0.32	0.39	0.33	0.45	0.44	0.54	0.35	0.21
NH_4^+	-12.28	-16.94	-19.80	-16.35	-20.68	-20.35	-23.34	-17.89	-8.93
NO_3^-	1.23	1.63	1.54	1.36	1.35	1.50	1.14	1.66	1.96
SO_4^{2-}	0.21	0.32	0.40	0.33	0.48	0.48	0.62	0.39	0.19
Other	0.18	0.29	0.36	0.30	0.42	0.41	0.51	0.33	0.18

δ_i represent the sensitivity coefficients (δ) of simulated components in case DBL at monitoring site i (site index is listed in Table S12).

Table S15 The sensitivity coefficients (δ) of simulated components in case DBP at different monitoring sites

Components	δ_1	δ_2	δ_3	δ_4	δ_5	δ_6	δ_7	δ_9	δ_{10}
Al	0.20	0.33	0.40	0.34	0.46	0.45	0.54	0.35	0.21
Ca	0.19	0.30	0.36	0.30	0.42	0.41	0.51	0.33	0.18
Cl	0.12	0.28	0.34	0.29	0.39	0.37	0.47	0.26	0.11
EC	0.19	0.30	0.36	0.30	0.42	0.41	0.51	0.33	0.18
Fe	0.20	0.33	0.40	0.34	0.46	0.45	0.54	0.35	0.21
K	0.19	0.30	0.36	0.30	0.42	0.41	0.51	0.33	0.18
Mg	0.19	0.30	0.36	0.30	0.42	0.41	0.51	0.33	0.18
Mn	0.20	0.33	0.40	0.34	0.46	0.45	0.54	0.35	0.21
Na	0.19	0.30	0.36	0.30	0.42	0.41	0.51	0.33	0.18
OC	0.19	0.30	0.37	0.30	0.42	0.41	0.51	0.33	0.19
Si	0.20	0.33	0.40	0.34	0.46	0.45	0.54	0.35	0.21
Ti	0.20	0.33	0.40	0.34	0.46	0.45	0.54	0.35	0.21
Other	0.18	0.29	0.36	0.30	0.42	0.41	0.51	0.33	0.18

δ_i represent the sensitivity coefficients (δ) of simulated components in case DBP at monitoring site i (site index is listed in Table S12)

Table S16 The sensitivity coefficients (δ) of simulated components in case DBS at different monitoring sites

Components	δ_1	δ_2	δ_3	δ_4	δ_5	δ_6	δ_7	δ_9	δ_{10}
Al	-	-	-	-	-	-	-	-	-
Ca	-	-	-	-	-	-	-	-	-
Cl	-	-	-	-	-	-	-	-	-
EC	-	-	-	-	-	-	-	-	-
Fe	-	-	-	-	-	-	-	-	-
K	-	-	-	-	-	-	-	-	-
Mg	-	-	-	-	-	-	-	-	-
Mn	-	-	-	-	-	-	-	-	-
Na	-	-	-	-	-	-	-	-	-
OC	-	-	-	-	-	-	-	-	-
Si	-	-	-	-	-	-	-	-	-
Ti	-	-	-	-	-	-	-	-	-
NH_4^+	0.05	0.09	0.11	0.09	0.12	0.11	0.15	0.09	0.04
NO_3^-	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01	-0.02
SO_4^{2-}	0.15	0.24	0.30	0.24	0.35	0.35	0.43	0.27	0.16
Other	-0.19	-0.31	-0.38	-0.31	-0.44	-0.43	-0.54	-0.34	-0.19

δ_i represent the sensitivity coefficients (δ) of simulated components in case DBS at monitoring site i (site index is listed in Table S12)

Table S17 The sensitivity coefficients (δ) of simulated components in case TPS at different monitoring sites

Components	δ_1	δ_2	δ_3	δ_4	δ_5	δ_6	δ_7	δ_9	δ_{10}
Al	-	-	-	-	-	-	-	-	-
Ca	-	-	-	-	-	-	-	-	-
Cl	-	-	-	-	-	-	-	-	-
EC	-	-	-	-	-	-	-	-	-
Fe	-	-	-	-	-	-	-	-	-
K	-	-	-	-	-	-	-	-	-
Mg	-	-	-	-	-	-	-	-	-
Mn	-	-	-	-	-	-	-	-	-
Na	-	-	-	-	-	-	-	-	-
OC	-	-	-	-	-	-	-	-	-
Si	-	-	-	-	-	-	-	-	-
Ti	-	-	-	-	-	-	-	-	-
NH_4^+	0.05	0.09	0.11	0.09	0.12	0.11	0.15	0.09	0.04
NO_3^-	-0.01	-0.01	-0.01	-0.01	-0.01	-0.02	-0.01	-0.01	-0.02
SO_4^{2-}	0.15	0.24	0.30	0.24	0.35	0.34	0.42	0.27	0.16
Other	-0.19	-0.31	-0.38	-0.31	-0.44	-0.43	-0.54	-0.34	-0.19

δ_i represent the sensitivity coefficients (δ) of simulated components in case TPS at monitoring site i (site index is listed in Table S12)

Table S18 The sensitivity coefficients (δ) of simulated components in case TWN at different monitoring sites

Components	δ_1	δ_2	δ_3	δ_4	δ_5	δ_6	δ_7	δ_9	δ_{10}
Al	-	-	-	-	-	-	-	-	-
Ca	-	-	-	-	-	-	-	-	-
Cl	-	-	-	-	-	-	-	-	-
EC	-	-	-	-	-	-	-	-	-
Fe	-	-	-	-	-	-	-	-	-
K	-	-	-	-	-	-	-	-	-
Mg	-	-	-	-	-	-	-	-	-
Mn	-	-	-	-	-	-	-	-	-
Na	-	-	-	-	-	-	-	-	-
OC	-	-	-	-	-	-	-	-	-
Si	-	-	-	-	-	-	-	-	-
Ti	-	-	-	-	-	-	-	-	-
NH_4^+	0.04	0.07	0.09	0.07	0.10	0.10	0.12	0.08	0.04
NO_3^-	0.16	0.29	0.35	0.29	0.40	0.38	0.50	0.29	0.12
SO_4^{2-}	-	-0.03	-0.04	-0.03	-0.04	-0.03	-0.06	-0.02	0.03
Other	-0.19	-0.32	-0.39	-0.32	-0.45	-0.44	-0.55	-0.34	-0.19

δ_i represent the sensitivity coefficients (δ) of simulated components in case TWN at monitoring site i (site index is listed in Table S12)

Table S19 The sensitivity coefficients (δ) of simulated components in case FON at different monitoring sites

Components	δ_1	δ_2	δ_3	δ_4	δ_5	δ_6	δ_7	δ_9	δ_{10}
Al	-	-	-	-	-	-	-	-	-
Ca	-	-	-	-	-	-	-	-	-
Cl	-	-	-	-	-	-	-	-	-
EC	-	-	-	-	-	-	-	-	-
Fe	-	-	-	-	-	-	-	-	-
K	-	-	-	-	-	-	-	-	-
Mg	-	-	-	-	-	-	-	-	-
Mn	-	-	-	-	-	-	-	-	-
Na	-	-	-	-	-	-	-	-	-
OC	-	-	-	-	-	-	-	-	-
Si	-	-	-	-	-	-	-	-	-
Ti	-	-	-	-	-	-	-	-	-
NH_4^+	0.04	0.07	0.09	0.07	0.10	0.10	0.12	0.07	0.04
NO_3^-	0.15	0.28	0.35	0.29	0.40	0.38	0.49	0.29	0.12
SO_4^{2-}	-	-0.03	-0.04	-0.03	-0.04	-0.03	-0.06	-0.02	0.03
Other	-0.19	-0.31	-0.38	-0.32	-0.44	-0.43	-0.54	-0.34	-0.19

δ_i represent the sensitivity coefficients (δ) of simulated components in case FON at monitoring site i (site index is listed in Table S12)

Table S20 The sensitivity coefficients (δ) of simulated components in case OHA at different monitoring sites

Components	δ_1	δ_2	δ_3	δ_4	δ_5	δ_6	δ_7	δ_9	δ_{10}
Al	-	-	-	-	0.01	-	0.01	-	-
Ca	-	-	-	-	-	-	-	-	-
Cl	-	-	-	-	-	0.01	0.01	-	-
EC	-	-	-	-	0.01	-	0.01	-	-
Fe	-	-	-	-	-	-	-	-	-
K	-	-	-	-	-	-	-	-	-
Mg	-	-	-	-	-	-	-	-	-
Mn	-	-	-	-	-	-	-	-	-
Na	-	-	-	-	-	-	-	-	-
OC	-	0.01	0.01	0.01	0.01	0.01	0.01	-	-
Si	-	0.01	0.01	0.01	0.01	0.01	0.01	0.01	-
Ti	-	-	-	-	-	-	-	-	-
NH_4^+	0.04	0.05	0.07	0.06	0.08	0.09	0.10	0.08	0.07
NO_3^-	0.03	0.03	0.03	0.03	0.04	0.05	0.03	0.04	0.05
SO_4^{2-}	0.08	0.14	0.16	0.15	0.19	0.18	0.24	0.15	0.06
Other	-0.17	-0.25	-0.31	-0.26	-0.36	-0.36	-0.44	-0.30	-0.18

δ_i represent the sensitivity coefficients (δ) of simulated components in case OHA at monitoring site i (site index is listed in Table S12)

Table S21 The sensitivity coefficients (δ) of simulated components in case THA at different monitoring sites

Components	δ_1	δ_2	δ_3	δ_4	δ_5	δ_6	δ_7	δ_9	δ_{10}
Al	-	-	-	-	0.01	-	0.01	-	-
Ca	-	-	-	-	-	-	-	-	-
Cl	-	-	-	-	-	0.01	0.01	-	-
EC	-	-	-	-	0.01	-	0.01	-	-
Fe	-	-	-	-	-	-	-	-	-
K	-	-	-	-	-	-	-	-	-
Mg	-	-	-	-	-	-	-	-	-
Mn	-	-	-	-	-	-	-	-	-
Na	-	-	-	-	-	-	-	-	-
OC	-	0.01	0.01	0.01	0.01	0.01	0.01	-	-
Si	-	0.01	0.01	0.01	0.01	0.01	0.01	0.01	-
Ti	-	-	-	-	-	-	-	-	-
NH_4^+	0.04	0.05	0.07	0.06	0.08	0.09	0.10	0.08	0.07
NO_3^-	0.03	0.03	0.03	0.03	0.04	0.05	0.03	0.04	0.05
SO_4^{2-}	0.09	0.14	0.17	0.15	0.19	0.18	0.24	0.15	0.06
Other	-0.17	-0.26	-0.31	-0.26	-0.36	-0.36	-0.44	-0.30	-0.18

δ_i represent the sensitivity coefficients (δ) of simulated components in case THA at monitoring site i (site index is listed in Table S12)

Table S22 Equilibrium relations and *K* used in ISORROPIA II

Number	Reaction	K^0 （298.15K）
I1	$Ca(NO_3)_{2(s)} \leftrightarrow Ca^{2+}_{(aq)} + 2NO^{-}_{3(aq)}$	6.067×10^{5}
I2	$Ca(Cl)_{2(s)} \leftrightarrow Ca^{2+}_{(aq)} + 2Cl^{-}_{(aq)}$	7.974×10^{11}
I3	$CaSO_4 \cdot 2H_2O_{(s)} \leftrightarrow Ca^{2+}_{(aq)} + SO^{2-}_{4\ (aq)} + 2H_2O$	4.319×10^{-5}
I4	$K_2SO_{4(s)} \leftrightarrow 2K^{+}_{(aq)} + SO^{2-}_{4\ (aq)}$	1.569×10^{-2}
I5	$KHSO_{4(s)} \leftrightarrow K^{+}_{(aq)} + HSO^{-}_{4(aq)}$	24.016
I6	$KNO_{3(s)} \leftrightarrow K^{+}_{(aq)} + NO^{-}_{3(aq)}$	0.872
I7	$KCl_{(s)} \leftrightarrow K^{+}_{(aq)} + Cl^{-}_{(aq)}$	8.680
I8	$MgSO_{4(s)} \leftrightarrow Mg^{2+}_{(aq)} + SO^{2-}_{4\ (aq)}$	1.079×10^{5}
I9	$Mg(NO_3)_{2(s)} \leftrightarrow Mg^{2+}_{(aq)} + 2NO^{-}_{3(aq)}$	2.507×10^{15}
I10	$Mg(Cl)_{2(s)} \leftrightarrow Mg^{2+}_{(aq)} + 2Cl^{-}_{(aq)}$	9.557×10^{21}
I11	$HSO^{-}_{4(aq)} \leftrightarrow H^{+}_{(aq)} + SO^{2-}_{4\ (aq)}$	1.015×10^{-2}
I12	$NH_{3(g)} \leftrightarrow NH_{3(aq)}$	57.64
I13	$NH_{3(aq)} + H_2O_{(aq)} \leftrightarrow NH^{+}_{4(aq)} + OH^{-}_{(aq)}$	1.805×10^{-5}
I14	$HNO_{3(g)} \leftrightarrow H^{+}_{(aq)} + NO^{-}_{3(aq)}$	2.511×10^{6}
I15	$HNO_{3(g)} \leftrightarrow HNO_{3(aq)}$	2.1×10^{5}
I16	$HCl_{(g)} \leftrightarrow H^{+}_{(aq)} + Cl^{-}_{(aq)}$	1.971×10^{6}
I17	$HCl_{(g)} \leftrightarrow HCl_{(aq)}$	2.5×10^{3}
I18	$H_2O_{(aq)} \leftrightarrow H^{+}_{(aq)} + OH^{-}_{(aq)}$	1.010×10^{-14}
I19	$Na_2SO_{4(s)} \leftrightarrow 2Na^{+}_{(aq)} + SO^{2-}_{4\ (aq)}$	0.4799
I20	$(NH_4)_2SO_{4(s)} \leftrightarrow 2NH^{+}_{4(aq)} + SO^{2-}_{4\ (aq)}$	1.817

I21	$NH_4Cl_{(s)} \leftrightarrow NH_{3(g)} + HCl_{(g)}$	1.086×10^{-16}
I22	$NaNO_{3(s)} \leftrightarrow Na^+_{(aq)} + NO^-_{3(aq)}$	11.97
I23	$NaCl_{(s)} \leftrightarrow Na^+_{(aq)} + Cl^-_{(aq)}$	37.66
I24	$NaHSO_{4(s)} \leftrightarrow Na^+_{(aq)} + HSO^-_{4(aq)}$	2.413×10^{4}
I25	$NH_4NO_{3(s)} \leftrightarrow NH_{3(g)} + HNO_{3(g)}$	4.199×10^{-17}
I26	$NH_4HSO_{4(s)} \leftrightarrow NH^+_{4(aq)} + HSO^-_{4(aq)}$	1.383
I27	$(NH_4)_3H(SO_4)_{2(s)} \leftrightarrow 3NH^+_{4(aq)} + HSO^-_{4(aq)} + SO^{2-}_{4\ (aq)}$	29.72

Source: (Fountoukis and Nenes, 2007)

Table S23 Main reactions related to sulfate and nitrate production in CMAQ

	Gas-phase chemistry (G)
G1	$SO_2+OH+H_2O+O_2 \rightarrow H_2SO_4+HO_2$
G2	$NO_2+OH \rightarrow HNO_3$
G3	$N_2O_5+H_2O \rightarrow 2HNO_3$
G4	$NO_3+HO_2 \rightarrow HNO_3+O_2$
G5	$NTR+OH \rightarrow HNO_3$
G6	$NO_3+VOCs \rightarrow HNO_3$
	Aqueous-phase chemistry (Aq)
Aq1	$HSO_3^-+H_2O_2 \rightarrow SO_4^{2-}+H^+$
Aq2	$SO_2+O_3 \rightarrow SO_4^{2-}+2H^+$
Aq3	$HSO_3^-+O_3 \rightarrow SO_4^{2-}+H^+$
Aq4	$SO_3^{2-}+O_3 \rightarrow SO_4^{2-}$
Aq5	$HSO_3^-+MHP \rightarrow SO_4^{2-}+H^+$
Aq6	$HSO_3^-+PAA \rightarrow SO_4^{2-}+H^+$
Aq7	$S(IV)+Fe(III)/Mn(II) \rightarrow SO_4^{2-}$
	Heterogeneous chemistry (H)
H1	$N_2O_5+H_2O \rightarrow 2HNO_3$
H2	$2NO_2+H_2O \rightarrow HONO+HNO_3$

NTR: Organic nitrate; MHP: methyl hydroperoxide; PAA: peroxyacetic acid.

Source: (Zheng et al., 2015; Chapel Hill, 2012)

Table S24 Calculation method for sensitivity coefficient of gaseous pollutants

Equation	$\delta_i = \dfrac{\dfrac{C_{i\_case} - C_{i\_base}}{C_{i\_base}} \times 100\%}{P_{j\_case} - P_{j\_base}}$
Description	C_{i_case} is the simulation result of gas pollutant i in different sensitivity experiment cases, μg/m^3; C_{i_base} is the simulation result of gas pollutant *i* in base case, μg/m^3; P_{case} is the sum of the percentage of perturbed components which cause the variation of gas pollutant in sensitivity experiment cases, %; P_{base} is the percentage of P_{case} in base case, %.

Table S25 The sensitivity coefficients (δ) of gas pollutants from different cases at different monitoring sites

Cases and sites	δ_{SO_2}	δ_{NH_3}	δ_{NO_x}
1DBL	-0.08	0.05	-0.006
1DBP	-0.15	0.20	-0.008
1DBS	0.02	-0.18	-0.002
1TPS	0.02	-0.18	-0.002
1TWN	0.02	-0.12	0.002
1FON	0.02	-0.12	0.002
1OHA	-0.07	0.46	0.001
1THA	-0.07	0.46	0.001
2DBL	-0.03	0.04	-0.005
2DBP	-0.02	0.16	-0.006
2DBS	0.03	-0.17	-0.002
2TPS	0.01	-0.17	-0.002
2TWN	0.02	-0.13	0.002
2FON	0.02	-0.13	0.001
2OHA	-0.07	0.44	0.001
2THA	-0.03	0.44	0.001
3DBL	-0.03	0.06	-0.004
3DBP	-0.05	0.27	-0.005
3DBS	0.04	-0.28	-0.002
3TPS	0.04	-0.28	-0.001
3TWN	0.03	-0.21	0.001
3FON	0.03	-0.21	0.001
3OHA	-0.04	0.73	0.001
3THA	-0.04	0.73	0.001
4DBL	-0.05	0.04	-0.004
4DBP	-0.07	0.17	-0.005
4DBS	0.04	-0.18	-0.002
4TPS	0.04	-0.18	-0.002
4TWN	0.03	-0.14	0.001
4FON	0.03	-0.14	0.001

4OHA	-0.11	0.47	0.001
4THA	-0.07	0.47	0.001
5DBL	0.00	0.10	-0.002
5DBP	-0.02	0.45	-0.002
5DBS	0.04	-0.48	-0.001
5TPS	0.04	-0.48	-0.001
5TWN	0.03	-0.37	0.000
5FON	0.03	-0.37	0.000
5OHA	-0.11	1.30	0.000
5THA	-0.11	1.30	0.000
6DBL	-0.05	0.15	-0.002
6DBP	-0.11	0.64	-0.003
6DBS	0.05	-0.64	-0.001
6TPS	0.05	-0.65	-0.001
6TWN	0.04	-0.50	0.000
6FON	0.04	-0.50	0.000
6OHA	-0.14	1.76	0.001
6THA	-0.14	1.77	0.001
7DBL	-0.05	0.14	-0.001
7DBP	-0.11	0.59	-0.001
7DBS	0.05	-0.69	0.000
7TPS	0.05	-0.70	0.000
7TWN	0.04	-0.55	0.000
7FON	0.04	-0.55	0.000
7OHA	-0.15	1.92	0.000
7THA	-0.15	1.93	0.000
9DBL	-0.07	0.09	-0.003
9DBP	-0.13	0.38	-0.004
9DBS	0.05	-0.39	-0.001
9TPS	0.05	-0.39	-0.001
9TWN	0.04	-0.28	0.001
9FON	0.04	-0.28	0.001
9OHA	-0.14	1.02	0.001
9THA	-0.14	1.02	0.001
10DBL	-0.09	0.13	-0.004
10DBP	-0.16	0.53	-0.006
10DBS	0.04	-0.54	-0.002
10TPS	0.04	-0.53	-0.002
10TWN	0.03	-0.36	0.001
10FON	0.03	-0.36	0.001
10OHA	-0.12	1.39	0.001
10THA	-0.12	1.40	0.001

1DBL represent the simulation results of case DBL in site 1 (which is listed in Table S25 ‘cases and sites’ column), and the others named by the same rule.

Table S26 The selected information of source profile in SPECIATE and SAPPC database

Code	Profile Name	Controls	Profile Date	Profile Notes	Keywords	Match MEIC source
91041[a]	Draft Sub-Bituminous Combustion - Composite	Mixture of Baghouse, None, Electrostatic Precipitator, Wet Scrubber, Mechanical Collectors, Dry Lime Scrubber, Ammonia Injection	2006-5-24	Replaced by Profile 91110. Median of Profiles 3191, 3192, 3690, 3694, and 3700	Sub-Bituminous Coal Combustion; PM Composite	PP
900162.5[b]	Industrial Manufacturing - Average	Not Applicable	1989-1-5	Average profile developed from original profiles representing the source category group 3xxxxxxx	INDUSTRIAL	IN
91155[c]	Residential Coal Combustion - Composite	Uncontrolled	2009-7-12	Median of Profiles 3761, 432012.5	Residential Coal Combustion; Inventory speciation	RE
91022[a]	Draft On-road Gasoline Exhaust - Composite	Mixture of Catalytic converter and Not available	2006-5-24	Replaced by Profile 91122. Median of Profiles 311072.5, 3517, 3884, 3892, 3904, 3947, 3951, 3955, 3959, and 4558	On-road Gasoline Exhaust; PM Composite	TR
91162[c]	LDDV Exhaust - Composite	Mixture of Catalytic converter and Not available	2009-7-12	Median of Profiles 321042.5, 3912, 3963, 4675	LDDV Exhaust; Inventory speciation	

Local[d]	Coal combustion by power plants	Mixture of Baghouse, None, Electrostatic Precipitator, Wet Scrubber, Mechanical Collectors, Dry Lime Scrubber	Average of profiles power and heating power plant	PP
Local[d]	Industrial processes	Wet Scrubber, Dry Lime Scrubber	Average of profiles steel, metallurgy, cement, glass, industrial boiler	IN
Local[d]	Transportation sector	Mixture of Catalytic converter	Average of profiles gasoline, diesel, gasoline-diesel exhaust	TR
Local[d]	Residential emission		Average of profiles civil boiler	RE

a, Hsu, Ying, Randy Strait, Stephen Roe, David Holoman. 2006. 'SPECIATE 4.0 Speciation database development document - Final Report', Prepared for US EPA, RTP, NC, EPA Contract Nos. EP-D-06-001, Work Assignment Numbers 0-03 and 68-D-02-063, WA 4-04 and WA 5-05, by E.H. Pechan & Associates, Incorporation, Durham, NC. https://www.epa.gov/sites/production/files/2015-10/documents/speciatedoc_1206.pdf.

b, Shareef, G. S. Engineering Judgement, Radian Corporation. August 1987.

c, Reff, Adam, Prakash V Bhave, Heather Simon, Thompson G Pace, George A Pouliot, J David Mobley, and Marc Houyoux. 2009. 'Emissions Inventory of $PM_{2.5}$ Trace Elements across the United States', Environmental Science & Technology, 43, no. 15: 5790-96. DOI: 10.1021/es802930x.

d, Database of Source Profiles of Air Pollution (SPAP), measured by State Environmental Protection Key Laboratory of Urban Ambient Air Particulate Matter Pollution Prevention and Control & Tianjin Key Laboratory of Urban, Nankai University. Coal combustion by power plants (PP), industrial processes (IN), residential emission (RE) and transportation sector (TR).

Table S27 Potential species for the five aerosol types

R_1	R_2	R_3	Aerosol type	Solid phase
$R_1<1$	any value	any value	Sulfate Rich (free acid)	$NaHSO_4$, NH_4HSO_4, $KHSO_4$, $CaSO_4$
$1\leq R_1\leq 2$	any value	any value	Sulfate Rich	$NaHSO_4$, NH_4HSO_4, Na_2SO_4, $(NH_4)_2SO_4$, $(NH_4)_3H(SO_4)_2$, $CaSO_4$, $KHSO_4$, K_2SO_4, $MgSO_4$
$R_1\geq 2$	$R_2<2$	any value	Sulfate Poor, Crustal & Sodium Poor	Na_2SO_4, $(NH_4)_2SO_4$, NH_4NO_3, NH_4Cl, $CaSO_4$, K_2SO_4, $MgSO_4$
$R_1\geq 2$	$R_2\geq 2$	$R_3<2$	Sulfate Poor, Crustal & Sodium Rich, Crustal Poor	Na_2SO_4, $NaNO_3$, $NaCl$, NH_4NO_3, NH_4Cl, $CaSO_4$, K_2SO_4, $MgSO_4$
$R_1\geq 2$	$R_2\geq 2$	$R_3>2$	Sulfate Poor, Crustal & Sodium Rich, Crustal Rich	$NaNO_3$, $NaCl$, NH_4NO_3, NH_4Cl, $CaSO_4$, K_2SO_4, $MgSO_4$, $Ca(NO_3)_2$, $CaCl_2$, $Mg(NO_3)_2$, $MgCl_2$, KNO_3, KCl

Table S28 The sensitivity coefficients (δ) of simulated components for different meteorological pattern

Pattern I

Components	DBL	DBP	DBS	TPS	TWN	FON	OHA	THA
EC	0.35	0.35	0.00	0.00	0.00	0.00	0.00	0.00
OC	0.35	0.36	0.00	0.00	0.00	0.00	0.00	0.00
Fe	0.39	0.39	0.00	0.00	0.00	0.00	0.00	0.00
Mn	0.39	0.39	0.00	0.00	0.00	0.00	0.00	0.00
Ti	0.39	0.39	0.00	0.00	0.00	0.00	0.00	0.00
Al	0.39	0.39	0.00	0.00	0.00	0.00	0.00	0.00
Si	0.39	0.39	0.00	0.00	0.00	0.00	0.00	0.00
Ca	0.35	0.35	0.00	0.00	0.00	0.00	0.00	0.00
Mg	0.35	0.35	0.00	0.00	0.00	0.00	0.00	0.00
K	0.35	0.35	0.00	0.00	0.00	0.00	0.00	0.00
Na	0.35	0.35	0.00	0.00	0.00	0.00	0.00	0.00
Cl	0.26	0.27	0.00	0.00	0.00	0.00	0.01	0.01
NH_4^+	-19.04		0.09	0.08	0.07	0.07	0.13	0.12
NO_3^-	1.01		-0.03	-0.03	0.27	0.27	0.08	0.07
SO_4^{2-}	0.40		0.32	0.32	0.01	0.01	0.11	0.12
Other	0.36	0.36	-0.37	-0.37	-0.36	-0.36	-0.34	-0.34

Pattern II

Components	DBL	DBP	DBS	TPS	TWN	FON	OHA	THA
EC	0.43	0.44	0.00	0.00	0.00	0.00	0.01	0.01
OC	0.43	0.44	0.00	0.00	0.00	0.00	0.01	0.01
Fe	0.46	0.47	0.00	0.00	0.00	0.00	0.00	0.00
Mn	0.46	0.47	0.00	0.00	0.00	0.00	0.00	0.00
Ti	0.46	0.47	0.00	0.00	0.00	0.00	0.00	0.00

Al	0.46	0.47	0.00	0.00	0.00	0.00	0.01	0.01
Si	0.46	0.47	0.00	0.00	0.00	0.00	0.01	0.01
Ca	0.43	0.44	0.00	0.00	0.00	0.00	0.00	0.00
Mg	0.43	0.44	0.00	0.00	0.00	0.00	0.00	0.00
K	0.43	0.44	0.00	0.00	0.00	0.00	0.00	0.00
Na	0.43	0.44	0.00	0.00	0.00	0.00	0.00	0.00
Cl	0.37	0.38	0.00	0.00	0.00	0.00	0.01	0.01
NH_4^+	-23.29		0.12	0.12	0.10	0.10	0.11	0.10
NO_3^-	1.54		-0.02	-0.02	0.37	0.37	0.07	0.07
SO_4^{2-}	0.46		0.39	0.39	-0.01	-0.01	0.13	0.13
Other	0.44	0.44	-0.46	-0.46	-0.45	-0.45	-0.37	-0.37
				Pattern III				
Components	DBL	DBP	DBS	TPS	TWN	FON	OHA	THA
EC	0.40	0.41	0.00	0.00	0.00	0.00	0.00	0.00
OC	0.40	0.41	0.00	0.00	0.00	0.00	0.01	0.01
Fe	0.44	0.44	0.00	0.00	0.00	0.00	0.00	0.00
Mn	0.44	0.44	0.00	0.00	0.00	0.00	0.00	0.00
Ti	0.44	0.44	0.00	0.00	0.00	0.00	0.00	0.00
Al	0.44	0.44	0.00	0.00	0.00	0.00	0.01	0.01
Si	0.44	0.44	0.00	0.00	0.00	0.00	0.01	0.01
Ca	0.40	0.41	0.00	0.00	0.00	0.00	0.00	0.00
Mg	0.40	0.41	0.00	0.00	0.00	0.00	0.00	0.00
K	0.40	0.41	0.00	0.00	0.00	0.00	0.00	0.00
Na	0.40	0.41	0.00	0.00	0.00	0.00	0.00	0.00
Cl	0.33	0.34	0.00	0.00	0.00	0.00	0.01	0.01
NH_4^+	-23.56		0.11	0.11	0.09	0.09	0.11	0.11
NO_3^-	1.35		-0.02	-0.02	0.34	0.34	0.06	0.06
SO_4^{2-}	0.43		0.36	0.36	-0.01	-0.01	0.13	0.13
Other	0.41	0.41	-0.43	-0.43	-0.42	-0.42	-0.35	-0.35
				Pattern VI				
Components	DBL	DBP	DBS	TPS	TWN	FON	OHA	THA
EC	0.40	0.41	0.00	0.00	0.00	0.00	0.01	0.01
OC	0.41	0.41	0.00	0.00	0.00	0.00	0.01	0.01
Fe	0.44	0.45	0.00	0.00	0.00	0.00	0.00	0.00
Mn	0.44	0.45	0.00	0.00	0.00	0.00	0.00	0.00
Ti	0.44	0.45	0.00	0.00	0.00	0.00	0.00	0.00
Al	0.44	0.45	0.00	0.00	0.00	0.00	0.01	0.01
Si	0.44	0.45	0.00	0.00	0.00	0.00	0.01	0.01
Ca	0.40	0.41	0.00	0.00	0.00	0.00	0.00	0.00
Mg	0.40	0.41	0.00	0.00	0.00	0.00	0.00	0.00
K	0.40	0.41	0.00	0.00	0.00	0.00	0.00	0.00
Na	0.40	0.41	0.00	0.00	0.00	0.00	0.00	0.00
Cl	0.35	0.36	0.00	0.00	0.00	0.00	0.01	0.01
NH_4^+	-24.00		0.11	0.11	0.09	0.09	0.10	0.10

NO_3^-	0.96		-0.02	-0.02	0.36	0.36	0.05	0.05
SO_4^{2-}	0.43		0.35	0.35	-0.02	-0.02	0.15	0.15
Other	0.41	0.41	-0.43	-0.43	-0.42	-0.42	-0.35	-0.35

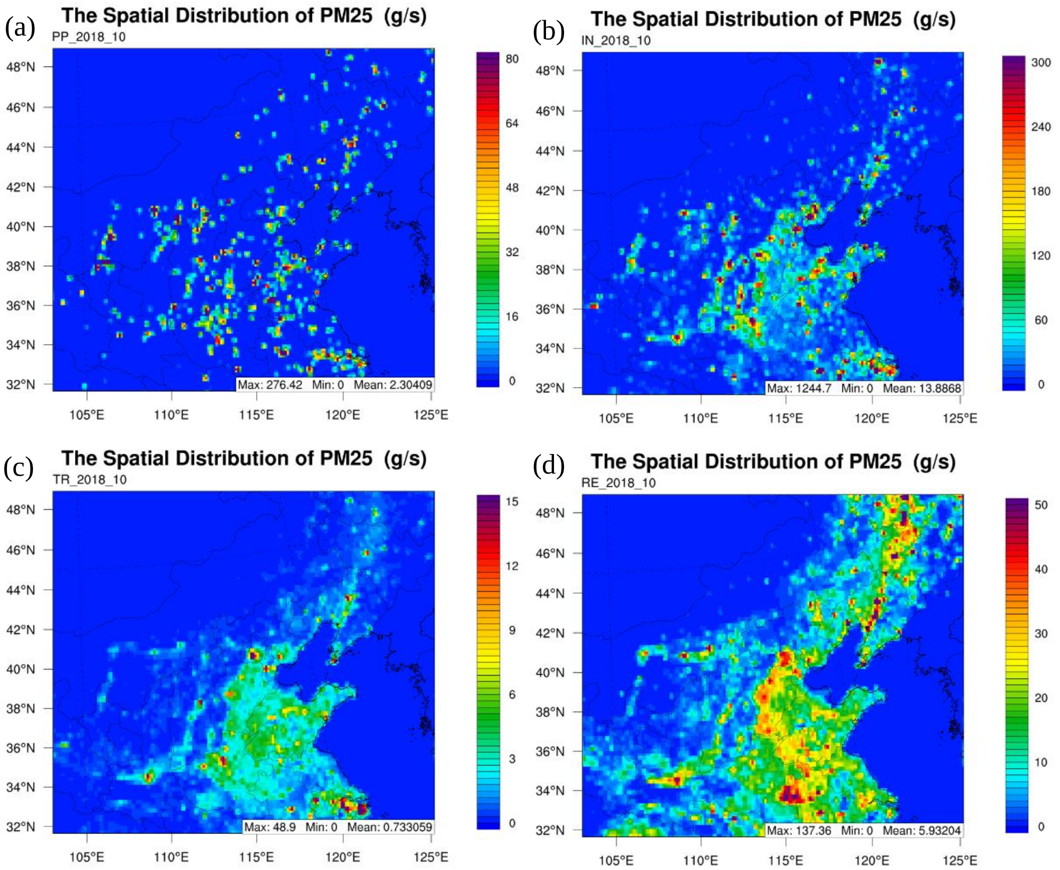


Figure S1 The regional distribution of $PM_{2.5}$ emission sources. (a) coal-fired power plant; (b) industry process; (c) transportation sector; (d) residential coal combustion.

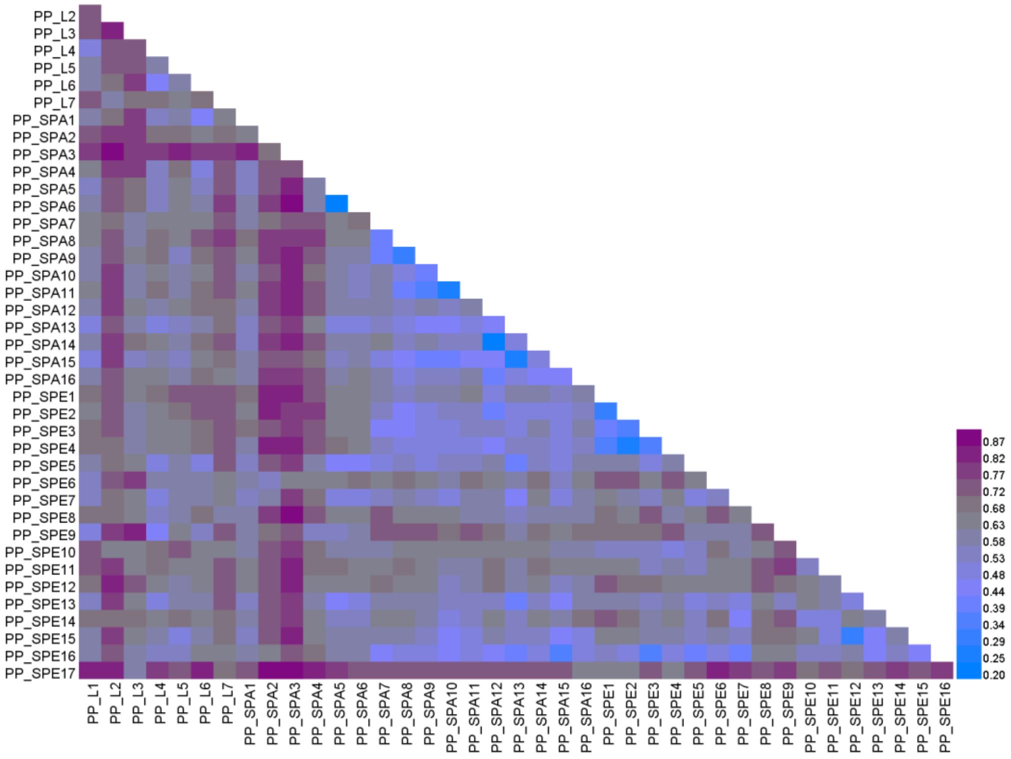


Figure S2 The coefficient divergence values for PP source profile

Note: Power plant source profiles from published literatures in China (PP_L), SPAP (PP_SPA) and SPECIATE database (PP_SPE). Numbers represent source profile sequence number.

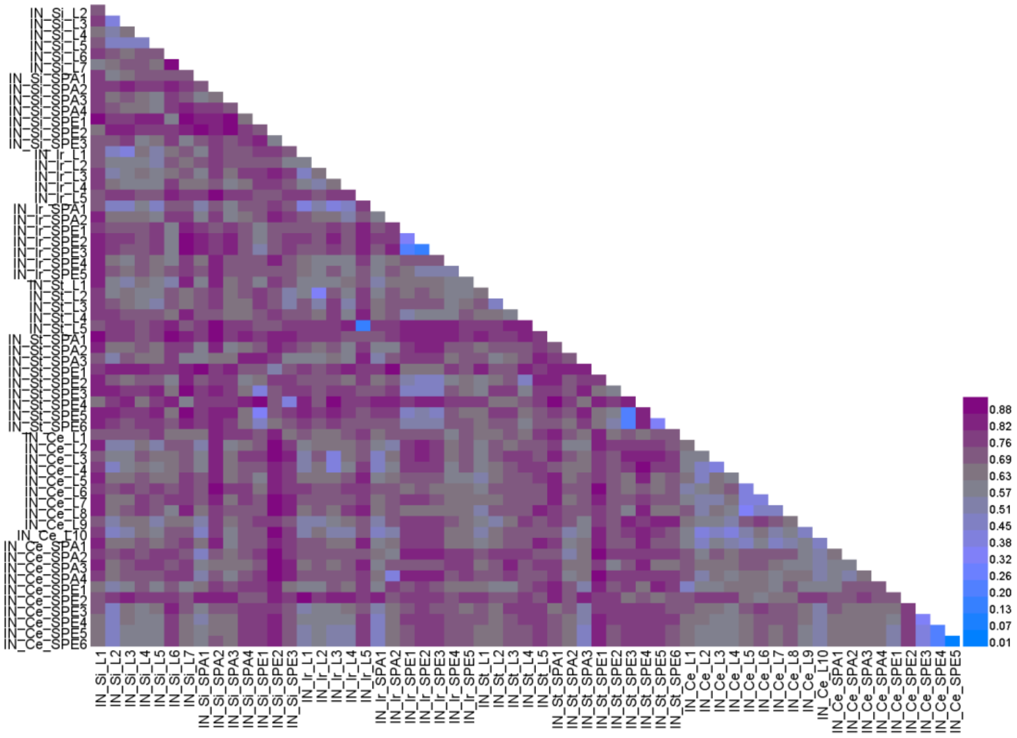


Figure S3 The coefficient divergence values for IN source profile

Note: Industrial process (Sintering) source profiles from published literatures (IN_Si_L) in China, SPAP (IN_Si_SPA) and SPECIATE database (IN_Si_SPE); Industrial process (Iron-making) source profiles from published literatures in China (IN_Ir_L), SPAP (IN_Ir_SPA) and SPECIATE database(IN_Ir_SPE); Industrial process (Steelmaking) source profiles from published literatures in China (IN_St_L), SPAP (IN_St_SPA) and SPECIATE (IN_St_SPE) database; Industrial process (Cement) source profiles from published literatures in China (IN_Ce_L), SPAP (IN_Ce_SPA) and SPECIATE database (IN_Ce_SPE). Numbers represent source profile sequence number.

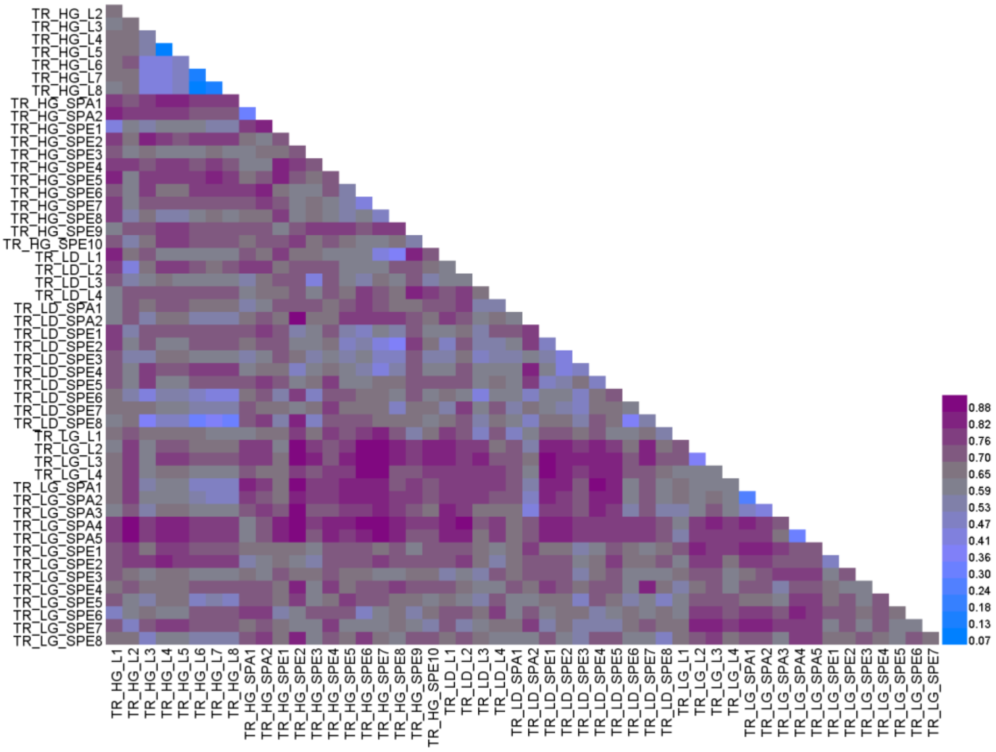


Figure S4 The coefficient divergence values for TR source profile

Note: Transportation sector (Heavy duty gasoline) source profiles from published literatures in China, SPAP and SPECIATE database. Transportation sector (Light diesel) source profiles from published literatures in China, SPAP and SPECIATE database. Transportation sector (Light duty gasoline) source profiles from published literatures in China, SPAP and SPECIATE database. Numbers represent source profile sequence number.

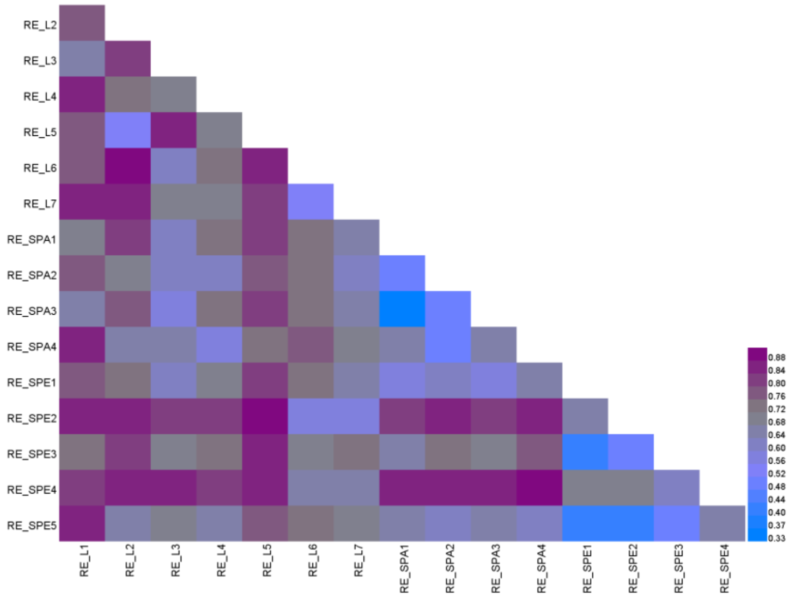


Figure S5 The coefficient divergence values for RE source profile

Note: Residential coal combustion source profiles from published literatures in China, SPAP and SPECIATE database. Numbers represent source profile sequence number.

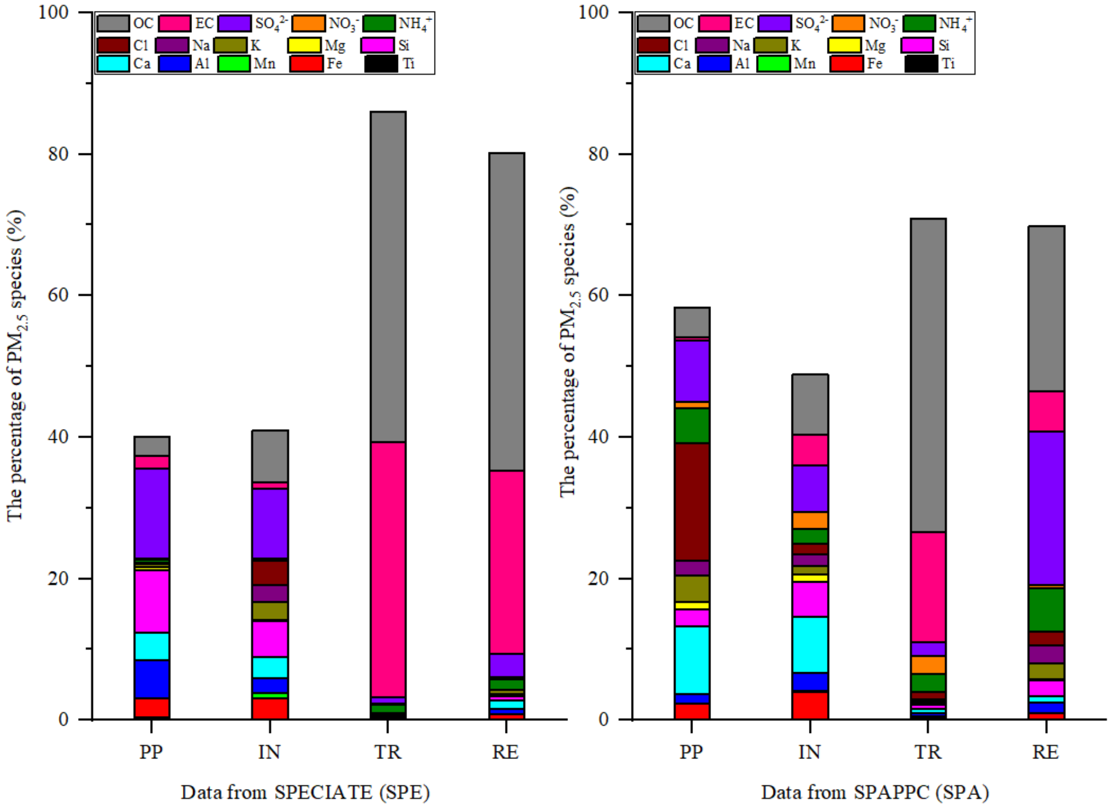
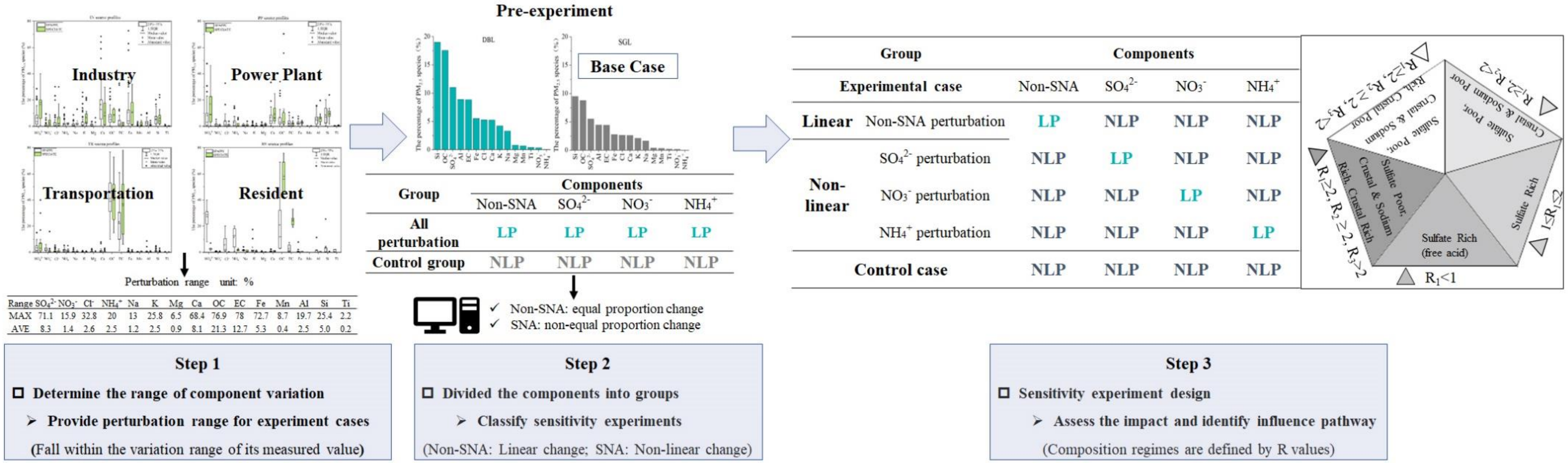


Figure S6 The selected speciation profile of PM2.5 for case CMAQ_SPE and CMAQ_SPA

In SPE, the selected source profiles were average profile developed from original profiles of the source category group in SPECIATE database, the power plant (PP) source profile code was 91041, industrial process (IN) was 900162.5, Residential coal combustion (RE) was 91155, Transportation sector (TR) was 91022 and 91162. In SPA, the selected source profiles were from SPAPPC database which were measured from local emission sources.



Range	SO_4^{2-}	NO_3^-	Cl^-	NH_4^+	Na	K	Mg	Ca	OC	EC	Fe	Mn	Al	Si	Ti
MAX	71.1	15.9	32.8	20	13	25.8	6.5	68.4	76.9	78	72.7	8.7	19.7	25.4	2.2
AVE	8.3	1.4	2.6	2.5	1.2	2.5	0.9	8.1	21.3	12.7	5.3	0.4	2.5	5.0	0.2

Group	**Components**			
	Non-SNA	SO_4^{2-}	NO_3^-	NH_4^+
All perturbation	**LP**	**LP**	**LP**	**LP**
Control group	**NLP**	**NLP**	**NLP**	**NLP**

- ✓ Non-SNA: equal proportion change
- ✓ SNA: non-equal proportion change

Group		**Components**			
Experimental case		Non-SNA	SO_4^{2-}	NO_3^-	NH_4^+
Linear	Non-SNA perturbation	**LP**	**NLP**	**NLP**	**NLP**
Non-linear	SO_4^{2-} perturbation	**NLP**	**LP**	**NLP**	**NLP**
	NO_3^- perturbation	**NLP**	**NLP**	**LP**	**NLP**
	NH_4^+ perturbation	**NLP**	**NLP**	**NLP**	**LP**
Control case		**NLP**	**NLP**	**NLP**	**NLP**

Step 1

☐ **Determine the range of component variation**

➢ **Provide perturbation range for experiment cases**

(Fall within the variation range of its measured value)

Step 2

☐ **Divided the components into groups**

➢ **Classify sensitivity experiments**

(Non-SNA: Linear change; SNA: Non-linear change)

Step 3

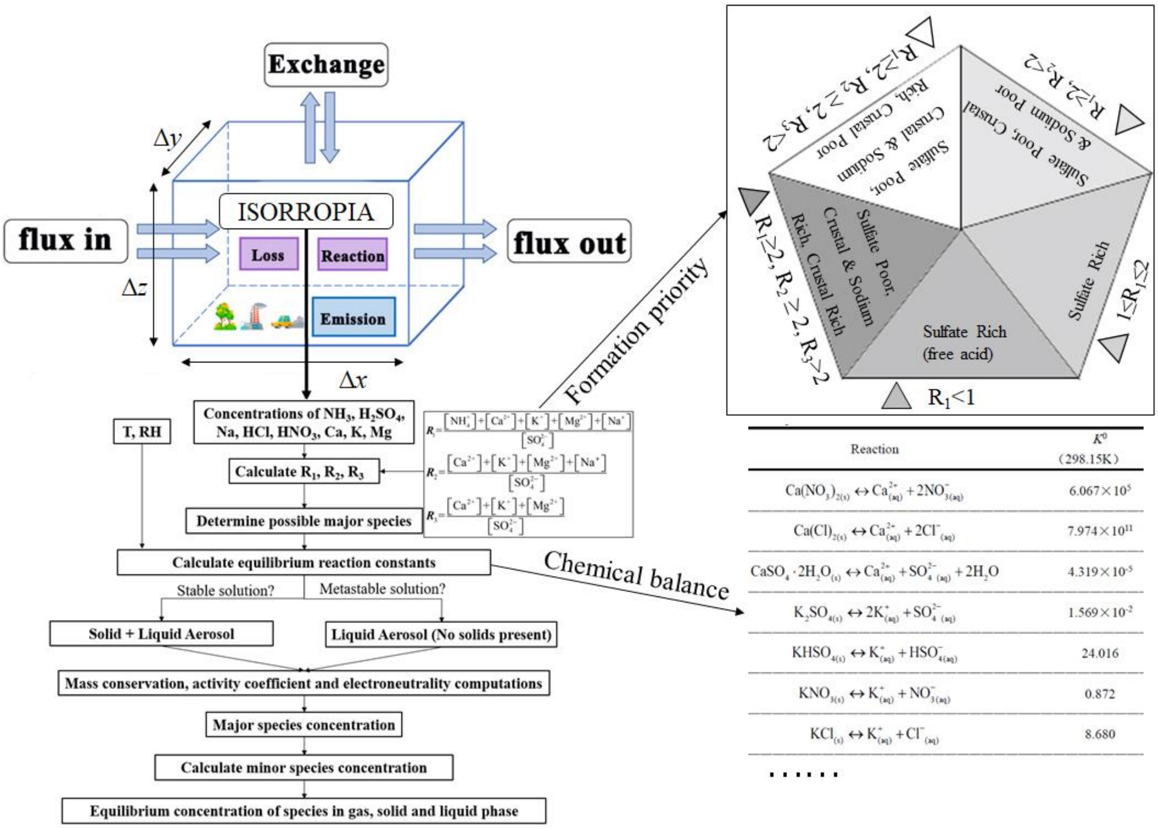
☐ **Sensitivity experiment design**

➢ **Assess the impact and identify influence pathway**

(Composition regimes are defined by R values)

Note: SNA represent SO_4^{2-}, NO_3^-, and NH_4^+, Non-SNA represent other components in $PM_{2.5}$; LP: Load perturbation; NLP: Not load perturbation

Figure S7 The sketch of sensitivity experiment design idea



$$R_1=\frac{[NH_4^+]+[Ca^{2+}]+[K^+]+[Mg^{2+}]+[Na^+]}{[SO_4^{2-}]}$$

$$R_2=\frac{[Ca^{2+}]+[K^+]+[Mg^{2+}]+[Na^+]}{[SO_4^{2-}]}$$

$$R_3=\frac{[Ca^{2+}]+[K^+]+[Mg^{2+}]}{[SO_4^{2-}]}$$

Reaction	K^0 (298.15K)
$Ca(NO_3)_{2(s)} \leftrightarrow Ca^{2+}_{(aq)} + 2NO^-_{3(aq)}$	6.067×10^5
$Ca(Cl)_{2(s)} \leftrightarrow Ca^{2+}_{(aq)} + 2Cl^-_{(aq)}$	7.974×10^{11}
$CaSO_4\cdot2H_2O_{(s)} \leftrightarrow Ca^{2+}_{(aq)} + SO^{2-}_{4\,(aq)} + 2H_2O$	4.319×10^{-5}
$K_2SO_{4(s)} \leftrightarrow 2K^+_{(aq)} + SO^{2-}_{4\,(aq)}$	1.569×10^{-2}
$KHSO_{4(s)} \leftrightarrow K^+_{(aq)} + HSO^-_{4(aq)}$	24.016
$KNO_{3(s)} \leftrightarrow K^+_{(aq)} + NO^-_{3(aq)}$	0.872
$KCl_{(s)} \leftrightarrow K^+_{(aq)} + Cl^-_{(aq)}$	8.680

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Figure S8 The general solution path of ISORROPIA for each grid at time *t*

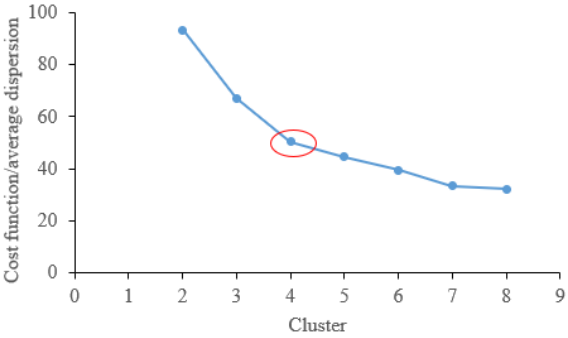

Figure S9 The elbow plot of K-mean clustering

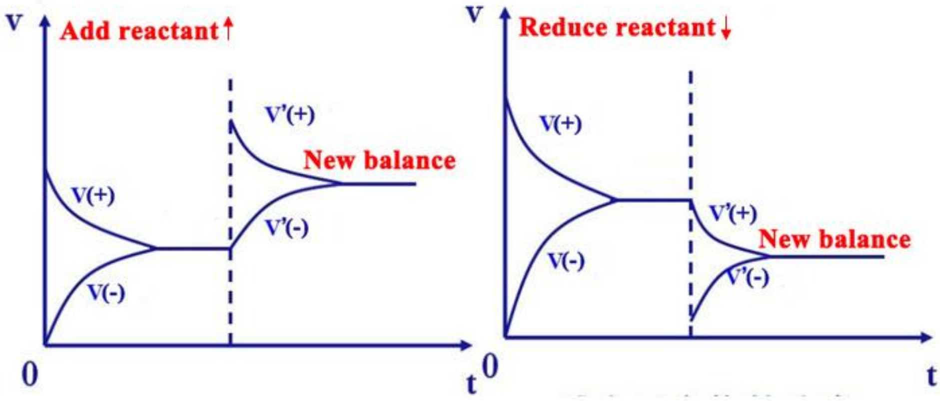

Figure S10 The shift direction of chemical reaction equilibrium

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