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Chemical composition of ambient PM_{2.5} over China and relationship to precursor emissions during 2005–2012

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Collection of ground-based PM_{2.5} and chemical compositions measurements from literatures

In this study, we collected PM_{2.5} and chemical composition measurements for 2005–2012 from literatures to evaluate our estimations. The sources of the data, site locations, sampling period, and other information are all listed in Table S1.

GEOS-Chem model

In this work, we used the nested-grid GEOS-Chem model v9-01-02 over Southeast Asia to simulate the conversion factors between PM_{2.5} species and AOD, as well as tropospheric SO₂ and NO₂ column densities. The nested-grid model, which covers China and most of its surrounding countries (11°S–55N, 70–150E), has a spatial resolution of 0.5° lat × 0.667° lon with 47 vertical layers (Chen et al., 2009). The lateral boundary conditions for the nested model were provided by the associated global simulations with horizontal resolution of 2° lat × 2.5° lon. Both global and nested simulations were driven by the assimilated GEOS-5 meteorology from the Goddard Earth Observing System (GEOS) at the NASA Global Modeling and Assimilation Office (GMAO; <http://gmao.gsfc.nasa.gov/>). A correction to the GEOS-5 predicted nocturnal mixed layer depth was applied, as described in Heald et al. (2012) and Walker et al. (2012).

The model was run with the full HO_x-NO_x-VOC-ozone-aerosol chemistry (Bey et al., 2001; Park et al., 2004), which includes the sulfate-nitrate-ammonium system (Park et al., 2006), primary (Park et al., 2003) and secondary (Liao et al., 2007) carbonaceous aerosols, mineral dust (Fairlie et al., 2007) and sea salt (Alexander et al., 2005). The ISORROPIA II thermodynamic scheme (Fountoukis et al., 2007) was used for partitioning of gases and aerosols (Pye et al., 2009). The nitric acid was reduced to 75% of its value for each time step to correct for an overestimation in HNO₃ suggested by Heald et al. (2012). AOD at 550 nm were calculated using the RH-dependent aerosol

optical extinctions (Martin et al., 2003) with an updated growth factor for organic matter, and updates to the dust (Ridley et al., 2012) and ammonium sulfate optics.

Anthropogenic emissions over China used in the GEOS-Chem model were year-by-year emissions during 2005–2012 taken from the MEIC inventory, including SO₂, NO_x, NH₃, CO, OC, BC and NMVOCs. Anthropogenic emissions over Southeast Asia outside China were taken from the INTEX-B inventory (Zhang et al., 2009). Other non-anthropogenic emissions include biomass burning (Mu et al., 2011), soil NO_x (Yienger et al., 1995) and lightning NO_x (Martin et al., 2007; Murray et al., 2012). A total of eight years simulations were conducted (2005–2012) with initialization on January 1, 2005 after a one year spin-up simulation. The study time period could not be expanded due to the restrictions by GEOS-5 meteorology data.

Satellite observations of SO₂ and NO₂ column densities

Satellite observation of SO₂ and NO₂ columns retrieved from the OMI instrument were used to better understand the emissions changes of SO₂ and NO_x in this work. OMI is a nadir-viewing UV-visible spectrometer, which observes the atmosphere in ground pixels varying from $13 \times 24 \text{ km}^2$ at nadir to about $28 \times 150 \text{ km}^2$ at edges.

The SO₂ products used in this study were taken from Wang et al. (2015), which were improved based on the standard NASA products to reduce the uncertainties. The latitude-dependent offsets in the SO₂ slant columns were removed using the reference sector method on a daily basis (Lee et al., 2009). Local AMFs were recalculated using the LIDORT radiative transfer model (Spurr et al., 2001) weighted by the relative vertical SO₂ profile taken from the global GEOS-Chem model. In addition, the NASA Cloud Pressure and Fraction products using O₂-O₂ absorption (OMCLDO2 v003) were used to replace the original Cloud Pressure and Fraction retrievals using Raman scattering (OMCLDRR v003) used in the operational NASA SO₂ products to diminish their effects on the inter-annual

variations of the AMFs for OMI retrievals, which would have impacts on the trend of SO₂ columns (Wang et al., 2015).

The NO₂ products used in this work were the OMI standard product, OMNO2 (version 2.1) (Bucsela et al., 2013), which is publicly available from the NASA Goddard Earth Sciences Data Active Archive Center (GES DISC, <http://disc.sci.gsfc.nasa.gov>). This version is significantly improved compared to previous versions (Bucsela et al., 2006; Celarier et al., 2008). The uncertainties in the final products were suggested to be 20% for clear-sky conditions (Lamsal et al., 2014).

For both SO₂ and NO₂ retrievals, we only used pixels with cloud radiance fraction ≤ 0.3 , solar zenith angle (SZA) $\leq 70^\circ$ and surface albedo ≤ 3 . Those five pixels at each edges were also excluded. Since June 2007, an anomaly happened to the CCD detectors, which can affect the quality of the radiance data at all wavelengths (i.e., row anomaly, <http://disc.sci.gsfc.nasa.gov/Aura/data-holdings/OMI/index.shtml#info>). The row anomaly has been expanded and changed over time ever since. In this work, we restricted all pixels affected by the row anomaly as suggested by NASA.

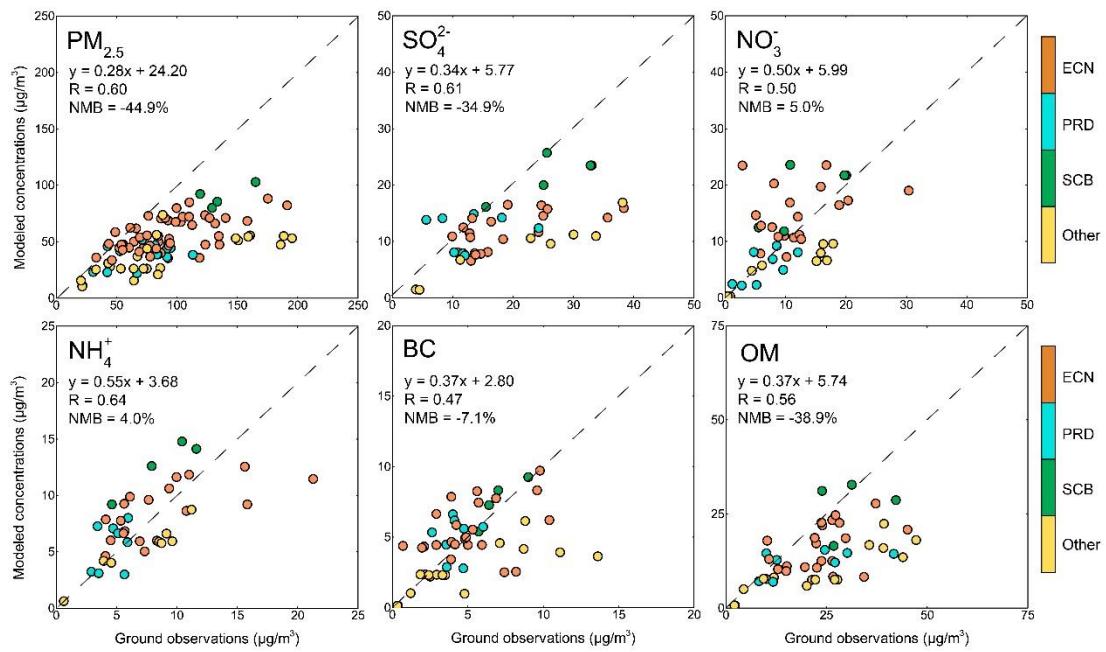


Figure S1. Evaluation of the GEOS-Chem modeled PM_{2.5} and chemical compositions concentrations using in situ data collected from publications. The dashed lines show the 1:1 line.

Table S1. Ground measurements collected from publications.

City	Lat	Lon	(µg/m³)						Sample Period	Source
			PM _{2.5}	SO ₄ ²⁻	NO ₃ ⁻	NH ₄ ⁺	OM	BC		
Baotou	40.65	109.85	66.6	10.3	3.2	2.7	18.1	2.7	Sep 2011 - Jun 2012	(Zhang and Zhang, 2014)
Beijing	40.32	116.32	118.5	15.8	10.1	7.3	24.5	8.2	Mar 2005 - Feb 2006	(Yang et al., 2011)
Beijing	39.99	116.30	55.4						2010	(Yu et al., 2013)
Beijing	39.99	116.30	135.0	13.6	11.3	6.9	16.9	5.0	Apr 2009, Jul 2009, Oct 2009, Jan 2010	(Zhang et al., 2013)
Beijing	39.98	116.34	131.5	18.4	10.7	10.8	32.2	9.6	2009 summer and winter	(Song et al., 2012)
Beijing	39.97	115.43	33.0						2009 - 2011	(Xin et al., 2014)
Beijing	39.95	116.30	92.6	14.6	9.7	8.3	18.8	5.9	Jun 2009, Sep 2009, Dec 2009, Mar 2010	(Liu et al., 2014)
Beijing	39.93	116.28	64.2					4.8	2007	(Yu et al., 2011)
Beijing	39.93	116.28	84.5						2007	(Zhao et al., 2009)
Beijing	39.90	116.30	123.4	19.1	20.5	6.4	18.2	6.3	Apr 2009, Jul 2009, Oct 2009, Jan 2010	(Zhao et al., 2013)
Beijing	39.98	116.35	112.4	24.2	20.3	15.8	17.1	5.6		(Wang et al., 2015)
Benxi	41.19	123.47	78.2						2007	(Guo et al., 2009)
Changsha	28.16	112.95	92.3						Jun - Oct 2009	(Yang et al., 2010)
Chengde	40.97	117.93	92.4	13.0	5.8	4.1	19.0	7.4	Apr 2009, Jul 2009, Oct 2009, Jan 2010	(Zhao et al., 2013)
Chengdu	30.66	104.00	165.0	33.0	20.0	10.4	22.3	9.0	Apr 2009, Jul 2009, Oct 2009, Jan 2010	(Tao et al., 2013)
Chengdu	30.61	104.04	133.2	15.5	9.7	4.6	19.1	5.7	Apr - May 2009	(Yang et al., 2012)
Chengdu	30.66	104.00		32.8	19.7	10.4			Apr 2009, Jul 2009, Oct 2009, Jan 2010	(Li et al., 2014)
Chengdu	30.65	104.03	119.0	25.0	10.7	11.6	17.0	7.0	Jan 2011, Apr 2011, Jul 2011, Oct 2011	(Tao et al., 2014)
Chonging	29.57	106.53	129.0	25.6	5.5	7.9	30.1	6.4	Mar 2005 - Feb 2006	(Yang et al., 2011)
Duolun	42.20	116.52	64.1						Mar - Apr 2007	(Deng et al., 2011)
Fuzhou	26.10	119.31	44.3	10.8	4.4	3.9	8.5	2.2	Apr 2007 - Jan 2008	(Xu et al., 2012)
Guangzhou	23.09	113.30	42.4				7.1	4.0	Aug 2006 - Jul 2007	(Huang et al., 2010)

Guangzhou	23.25	113.60	81.7	5.6	12.0	4.7	17.5	4.1	Dec 2008 - Feb 2009	(Yang et al., 2011)
Guangzhou	23.14	113.36	94.7	8.3	4.7	3.4	21.5	4.7	Jan 2008	(Tan et al., 2009)
Guangzhou	23.12	113.35	76.8	18.1	7.8	5.1	9.0	6.0	Apr 2009, Jul 2009, Oct 2009, Jan 2010	(Tao et al., 2014)
Guangzhou	23.10	113.35	91.4						Apr 2009, Jul 2009, Oct 2009, Jan 2010	(Lin et al., 2013)
Guangzhou	22.71	113.55	70.8	13.5	8.5	5.9	29.7	2.6	Nov - Dec 2010	(Wang et al., 2012)
Guangzhou	22.71	113.55	113.0	24.2	9.5	5.9	19.3	3.6	Oct - Nov 2007	(Ding et al., 2011)
Guangzhou	22.70	113.53	91.6						Aug - Dec 2008	(Ding et al., 2012)
Gucheng	39.13	115.80	86.6						2007	(Guo et al., 2009)
Guilin	25.32	110.30	66.9						2007	(Guo et al., 2009)
Hangzhou	30.27	120.15	84.3						Sep 2010 - Jul 2011	(Sun et al., 2013)
Hangzhou	30.27	120.15	69.0						2006 - 2008	(Hong et al., 2013)
Hangzhou	30.24	120.12	77.5	12.8	5.9	5.3				(Bao et al., 2010)
Hong kong	22.33	114.10	30.5	10.2	1.1	2.9	5.8	3.6	Aug - Sep 2009, Jan - Feb 2010	(Yau et al., 2013)
Huai'an	33.61	119.01	75.0						2008	(Cai et al., 2009)
Jinan	36.67	117.03	123.2	24.7	8.0	11.0	21.2	4.2	Mar 2006 - Feb 2007	(Yang et al., 2012)
Jinan	36.74	117.07		38.3	15.8	21.3			Dec 2007, Apr 2008, Jun 2008, Sep 2008	(Gao et al., 2011)
Jinan	36.67	117.12	158.1				16.0	5.3	2010	(Gu et al., 2014)
Jinsha	29.63	114.20	48.7	13.2	5.0	5.6	7.3	0.7	Mar 2012 - Mar 2013	(Zhang et al., 2014)
Lin'an	30.30	119.73	93.3						2007	(Guo et al., 2009)
Lushan	29.57	115.99	43.4						2007	(Guo et al., 2009)
Nanjing	32.12	118.95	90.0						Nov 2011 - Mar 2012	(Herrmann et al., 2013)
Nanjing	32.05	118.76	103.0						Jun 2007 - May 2008	(Yang et al., 2010)
Nanjing	32.05	118.76	98.8				15.7	10.4	Jun 2007 - May 2008	(Chen et al., 2010)
Nanjing	32.06	118.74	104.7	16.3	2.8	9.9			2010	(Zhang et al., 2013)
Nanjing	32.05	118.78	76.1						Jan 2011 - Apr 2011	(Zhuang et al., 2014)
Nanning	22.82	108.35	42.8						2007	(Guo et al., 2009)

Ningbo	29.68	121.60	46.0			7.1	2.5	Jul 2009 - Mar 2010	(Liu et al., 2013)
Panyu	23.00	113.35	52.6					2007	(Guo et al., 2009)
Qingdao	36.06	120.34	86.6					Jun 2007 - May 2008	(Li et al., 2012)
Qinghai Lake	36.98	99.90	21.5	3.9	0.8	0.6	1.6	0.4	Jun - Aug 2010
Qinghai Lake	36.98	99.90	21.3	4.5	0.4		1.5	0.3	Jun - Sep 2010
Qingyuan	23.70	113.06	84.1					Aug 2009 - Jan 2010	(Wei et al., 2011)
Qingyuan	23.60	113.08	83.2					Aug 2009 - Jan 2010	(Wei et al., 2011)
Sanya	18.30	109.52	20.4			3.1	1.2	Jan - Feb 2012	(Zhou et al., 2012)
Shangdianzi	40.70	117.10	71.8	13.8	12.2	4.5	10.8	3.9	Apr 2009, Jul 2009, Oct 2009, Jan 2010
Shangdianzi	40.65	117.12	60.2					2007	(Guo et al., 2009)
Shangdianzi	40.65	117.12	51.9					2007	(Zhao et al., 2009)
Shanghai	31.33	121.35	92.9			16.1	2.9	Oct 2005, Jan 2006, Apr 2006, Jul 2006	(Feng et al., 2009)
Shanghai	31.30	121.40	77.7					Nov 2011 - Dec 2011	(Jahn et al., 2013)
Shanghai	31.30	121.40	82.7					Jul 2009 - Sep 2010	(Wang et al., 2013)
Shanghai	31.23	121.48	54.8					2007	(Waheed et al., 2010)
Shanghai	31.22	121.55	53.9			3.9		Apr 2007 - Dec 2008	(Geng et al., 2013)
Shanghai	31.17	121.43	54.9	9.9	8.7	5.7	9.2	2.1	2011 - 2012
Shanghai	31.15	121.43	94.0	11.7	7.7	4.1	14.0	4.1	Apr 2009 - Feb 2010
Shanghai	31.15	121.43	68.4	12.9	12.6	5.6	10.7	2.0	Dec 2011 - Nov 2012
Shenyang	41.73	123.41	75.0					2006 - 2008	(Ma et al., 2011)
Shenzhen	22.59	113.97	42.2	11.7	2.7	3.5	8.3	4.7	2009
Shenzhen	22.58	113.97	66.9	12.0	5.1	5.6			Jul - Aug 2009 - 2010, Nov - Dec 2009 - 2010
Shijiazhuang	38.02	114.52	99.4					Jun - Oct 2008	(Du et al., 2010)
Shijiazhuang	38.00	114.40	191.2	35.6	30.4	9.3	26.5	9.8	Apr 2009, Jul 2009, Oct 2009, Jan 2010
Shijiazhuang	38.02	114.54	66.0					Jun - Jul 2012	(Ning et al., 2012)
Taishan	36.27	117.10	61.2					Mar - Jul 2007	(Zhou et al., 2012)

Taiyuan	37.80	112.58	334.5	35.1	12.1					Dec 2011 - Jan 2012	(Jia et al., 2013)
Tianjin	39.10	117.20	140.0	25.0	18.8	7.6	18.8	6.9		Apr 2009, Jul 2009, Oct 2009, Jan 2010	(Zhao et al., 2013)
Tianjin	39.10	117.20	109.8				16.9	5.7		Apr 2008, Jul 2008, Oct 2008, Jan 2009	(Gu et al., 2010)
Tianjin	39.10	117.15	134.5				7.6	4.9		Jun - Aug 2011	(Wei et al., 2012)
Tianjin	39.01	117.17		19.1	12.0	6.1				Jan 2008, Apr 2008, Jul 2008, Oct 2008	(Gu et al., 2013)
Urumqi	43.83	87.60		33.1	13.2	13.3	25.2	5.5		Mar 2013 - Feb 2014	(Zhao et al., 2015)
Wuhan	30.50	114.35	127.0				19.4	2.9		Jul 2011 - Feb 2012	(Cheng et al., 2012)
Wuhan	30.50	114.35	110.7	18.0	13.9	9.6	27.0	2.3		Aug 2012 - Jul 2013	(Zhang et al., 2015)
Xiamen	24.61	118.06	32.7							2012	(Niu et al., 2013)
Xiamen	24.58	118.09	86.2	11.2	6.0	4.5	15.2	2.9		Jun 2009 - May 2010	(Zhang et al., 2012)
Xiamen	24.58	118.09	63.9				15.8	2.4		Apr 2009, Jul 2009, Oct 2009, Jan 2010	(Zhang et al., 2011)
Xiamen	24.48	118.04	74.8				19.7	3.5		Apr 2009, Jul 2009, Oct 2009, Jan 2010	(Zhang et al., 2011)
Xiamen	24.48	118.11	72.1				19.3	3.3		Apr 2009, Jul 2009, Oct 2009, Jan 2010	(Zhang et al., 2011)
Xiamen	24.43	118.08	53.4				6.7	1.9		2008 - 2011	(Chen et al., 2012)
Xi'an	34.31	108.95					28.0	8.8		2006 - 2008	(Huang et al., 2011)
Xi'an	34.31	108.95		38.1	16.2	11.2				2006	(Huang et al., 2011)
Xi'an	34.30	108.93	88.1							2007	(Guo et al., 2009)
Xi'an	34.23	108.89	185.9	33.7	16.8	8.5	31.4	13.6		2005	(Cao, 2014)
Xi'an	34.23	108.89	195.2							2006	(Cao, 2014)
Xi'an	34.23	108.89	188.4	30.0	15.1	9.6	27.9	11.1		2007	(Cao, 2014)
Xi'an	34.23	108.89	150.4							2008	(Cao, 2014)
Xi'an	34.23	108.89	160.4	26.2	15.8	8.7	25.4	8.7		2009	(Cao, 2014)
Xi'an	34.23	108.89	148.4							2010	(Cao, 2014)
Xi'an	34.23	108.89	159.0	22.9	17.8	9.1	33.7	7.1		2011	(Cao, 2014)
Yong'an	25.97	117.36	84.1				14.3	4.8		Apr 2007 - Jan 2008	(Yin et al., 2012)
Yulin	38.29	109.74	83.0							Mar - Apr 2007	(Deng et al., 2011)

Zhengzhou	34.80	113.52	175.0	25.7	16.7	15.6	20.1	3.9	2010	(Geng et al., 2013)
Zhengzhou	34.78	113.68	110.2						2007	(Guo et al., 2009)

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