Supplement of

Decadal changes in anthropogenic source contribution of $PM_{2.5}$ pollution and related health impacts in China, 1990–2015

Jun Liu et al.

Correspondence to: Qiang Zhang (qiangzhang@tsinghua.edu.cn)

Validation of WRF-CMAQ modelling system

The domain-wide meteorological evaluation was carried out with the observed hourly surface temperature, relative humidity, wind speed, wind direction from the National Climate Data Center (www.ncdc.noaa.gov/) dataset. The evaluation parameters include mean bias (MB), root mean square error (RMSE), mean error (ME), normalized mean bias (NMB), normalized mean error (NME), and correlation coefficient (R). More than 1500 meteorological stations were located in the simulation domain. Table S1 shows the performance statistics in details. In general, the WRF model performed consistently over the past twenty-five years, and model performances were comparable among different years. The 2-m temperature agreed well with observations with the MB and RMSE ranging at -0.3~-0.1K and 2.6~2.8K, respectively. For 2-m relative humidity, the MB and NMB were 3.3~3.9% and 4.7~5.7%, respectively, showing close agreements between the simulations and observations. The simulated 10-m wind speed and the 10-m wind direction agreed fairly close with observations. Overall, the WRF model performed reasonably well during the simulation years, and the performance statistics are comparable with previous WRF-CMAQ (Hu et al. 2016; Zheng et al. 2015) and WRF-Chem (Liu et al. 2016) simulations over China.

Since 2013, direct $PM_{2.5}$ measurements have been included in China National Monitoring Network. Therefore, we conducted a detailed CMAQ model evaluation for year 2015. Daily $PM_{2.5}$ observations at 495 stations in 74 major cities were derived from monitoring data published the China National Environmental Monitoring Center (CNEMC, http://106.37.208.233:20035). Simulated hourly concentrations in the surface grid cell at the measurement location were used to derive the daily average concentrations. The averages of observations and simulations of $PM_{2.5}$ for each city are used for model evaluation. For the entire year, the average observed and simulated $PM_{2.5}$ concentrations were 54.4 μg m^{-3} and 59.4 μg m^{-3}, with correlation coefficient, RMSE, NMB at 0.73, 34.2 μg m^{-3} and 9.1%, respectively, indicating good agreements between observations and simulations. Figure S2 shows the overlay of observed and simulated seasonal $PM_{2.5}$ concentrations. Spatially, high $PM_{2.5}$ concentrations were distributed in Eastern and Central China, with concentration peaks in the Beijing-Tianjin-Hebei and the surrounding regions. There were obvious seasonal variations in $PM_{2.5}$, with the highest concentration in winter, and lowest concentration in Summer. Compared with the observations, the modelling system well reproduced the spatial and seasonal pattern of $PM_{2.5}$ in most of the regions over China, but underestimated the $PM_{2.5}$ concentrations in the west, where the dust source region was located.

Besides, to provide a view for other years, we compared the CMAQ simulated anthropogenic $PM_{2.5}$ with the satellite-derived dust-free $PM_{2.5}$ from van Donkelaar et al. (2016) for all the overlapping years, including 2000, 2005, 2010 and 2015 (Figure S3). In general, the CMAQ simulated anthropogenic $PM_{2.5}$ has a good correlation with satellite-derived dust-free $PM_{2.5}$, with R^2 ranging from 0.76 to 0.86. Better agreements occurred in recent years, especially in 2015, when direct $PM_{2.5}$ measurements were available to evaluate the CMAQ model and calibrate the satellite-based estimates. Higher $PM_{2.5}$ estimates from CMAQ occurred in early years, such as 2000, indicating larger uncertainties in exposure estimates for these early years. Since the satellite-based $PM_{2.5}$ estimates have their own uncertainties, it’s unknown which estimate is more reliable. However, since we focus more on the relative contribution of different source sectors in this study, the uncertainties from exposure estimates could be partially offset by the division operation.

References

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van Donkelaar, A., Martin, R. V., Brauer, M., Hsu, N. C., Kahn, R. A., Levy, R. C., Lyapustin, A., Sayer, A. M. and Winker, D. M.: Global Estimates of Fine Particulate Matter using a Combined Geophysical-Statistical Method with Information from Satellites, Models, and Monitors, Environ. Sci. Technol., 50(7), 3762–3772, doi:10.1021/acs.est.5b05833, 2016.

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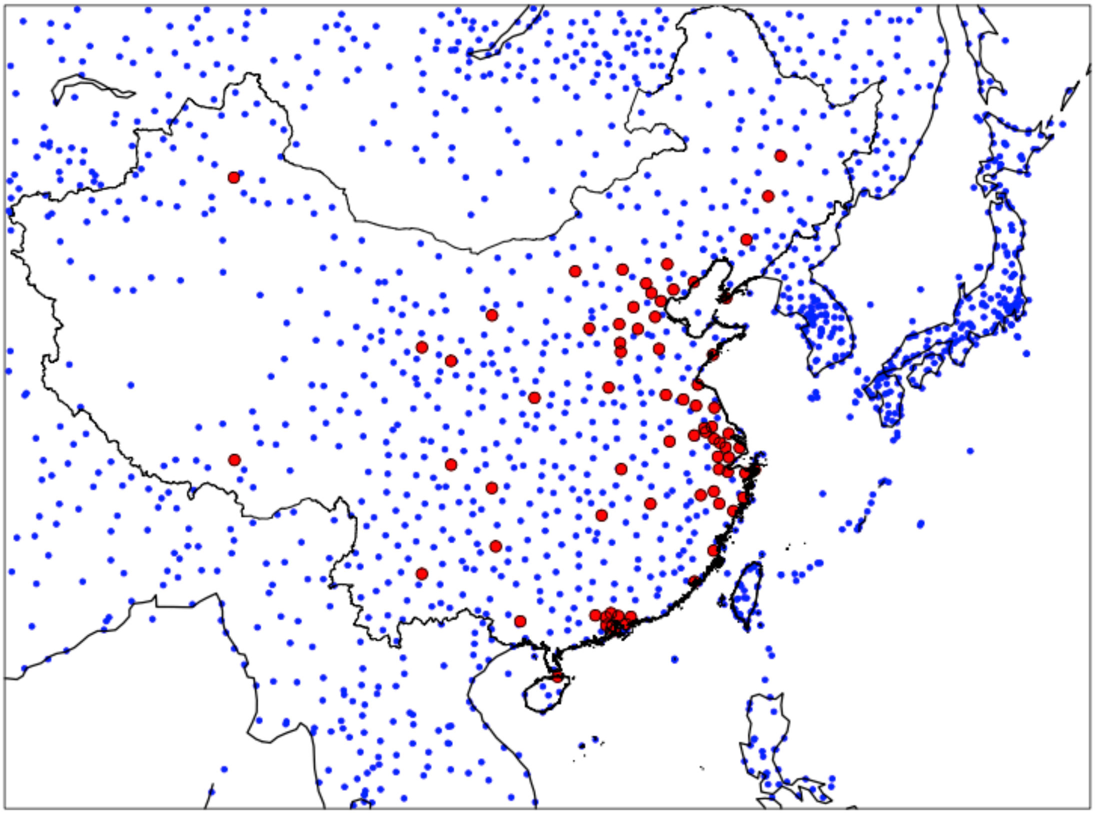

Figure S1: Domain used in WRF-CMAQ and location of monitoring stations for model evaluation. Blue dots are the meteorological sites included in the National Climate Data Center dataset, and red dots are the location of 74 major cities with monitoring stations included in the China National Monitoring Network for $PM_{2.5}$ concentrations.

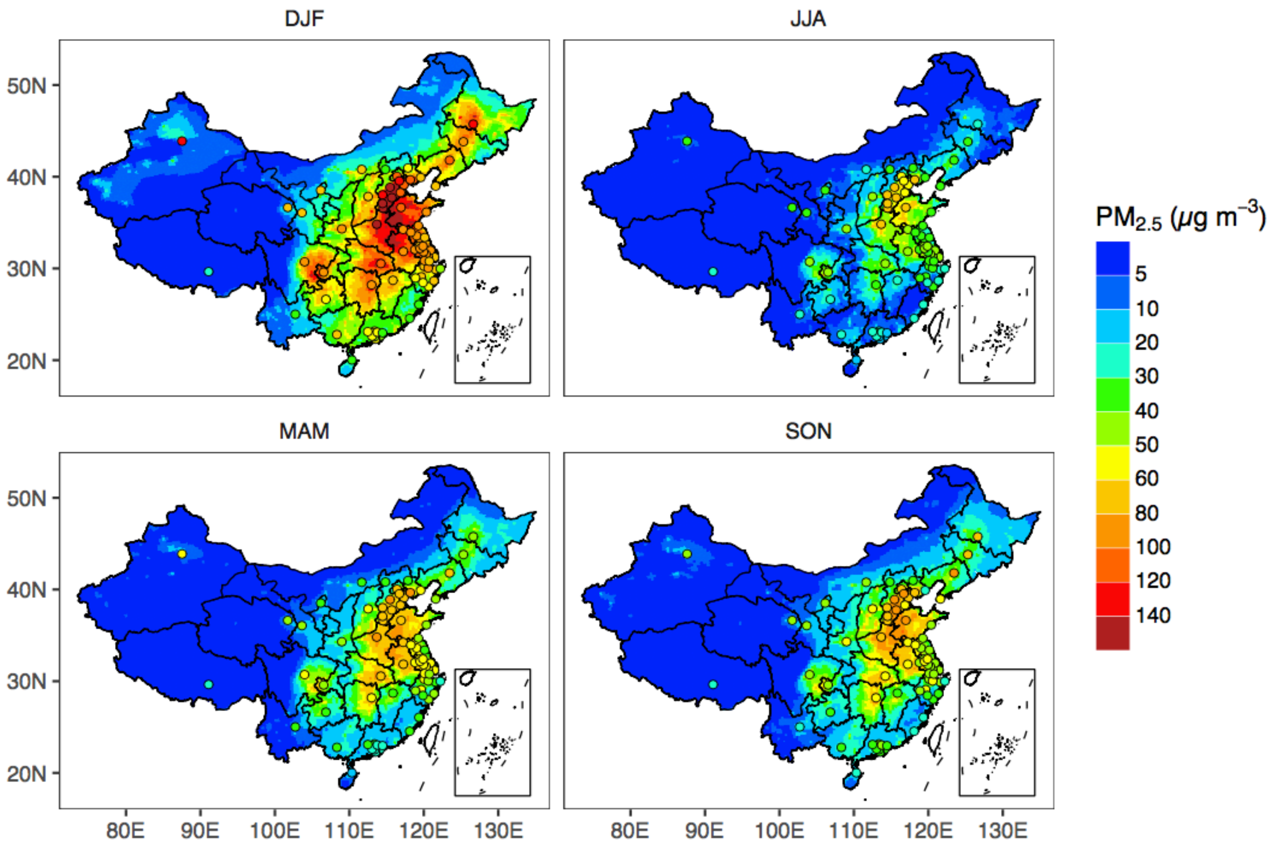

Figure S2: Simulated and observed seasonal average $PM_{2.5}$ concentration in 74 cities in China.

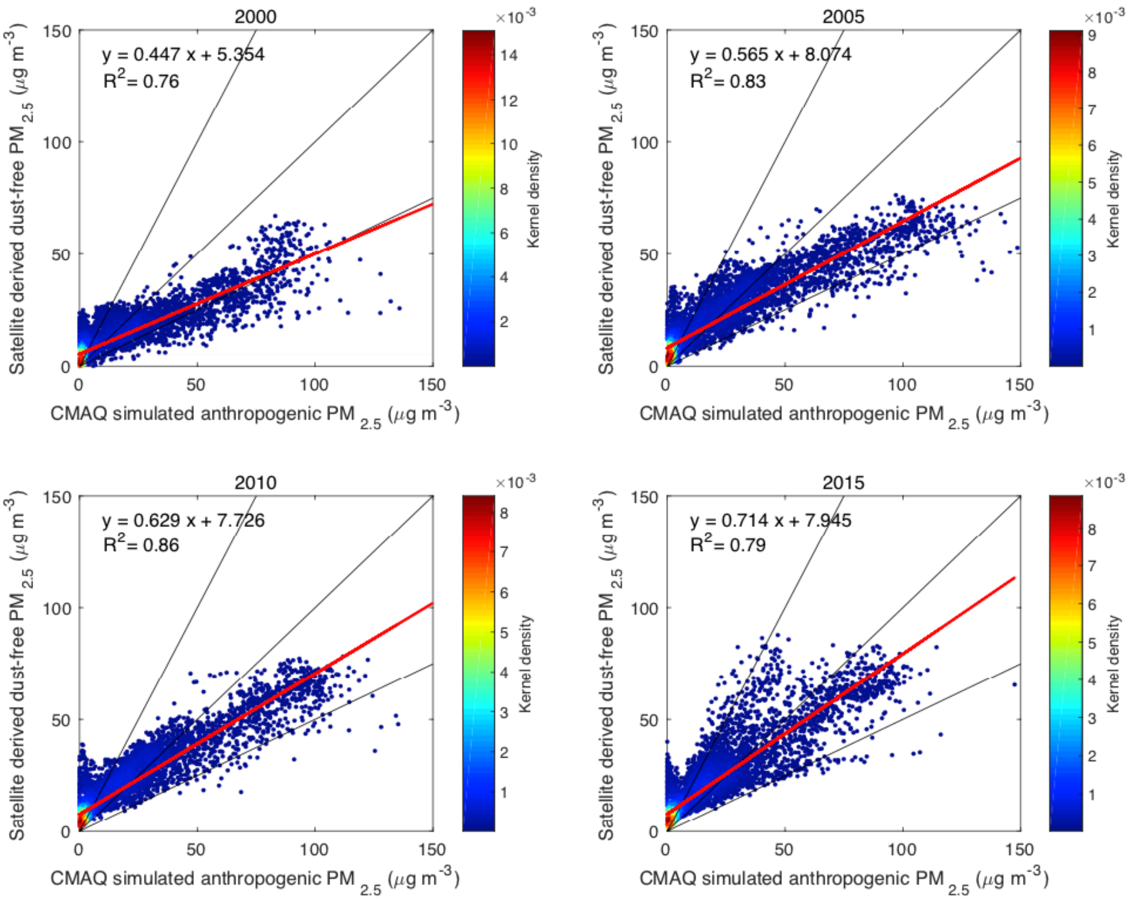

Figure S3: Comparison of CMAQ simulated anthropogenic $PM_{2.5}$ and satellite derived dust-free $PM_{2.5}$ in China (the dots are colored with the kernel density estimate)

<table>
<tr><th rowspan="2">Year</th><th rowspan="2">Power</th><th colspan="2">Industry</th><th colspan="2">Transportation</th></tr>
<tr><th>Cement</th><th>Iron and steel</th><th>Light-duty gasline</th><th>Heavy-duty diesel</th></tr>
<tr><td>1990</td><td>Pre-stage</td><td rowspan="7">GB4915-1985</td><td rowspan="7">GB9078-1988</td><td rowspan="10">Pre-stage</td><td rowspan="11">Pre-stage</td></tr>
<tr><td>1991</td><td rowspan="6">GB13223-1991</td></tr>
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<tr><td>2002</td></tr>
<tr><td>2003</td><td rowspan="4">Euro II</td></tr>
<tr><td>2004</td><td rowspan="8">GB13223-2003</td><td rowspan="3">Euro II</td></tr>
<tr><td>2005</td><td rowspan="9">GB4915-2004</td></tr>
<tr><td>2006</td></tr>
<tr><td>2007</td><td rowspan="7">Euro III</td><td rowspan="8">Euro III</td></tr>
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<tr><td>2011</td></tr>
<tr><td>2012</td><td rowspan="4">GB13223-2011</td><td rowspan="4">GB28662-2012
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GB28665-2012</td></tr>
<tr><td>2013</td></tr>
<tr><td>2014</td><td rowspan="2">GB4915-2013</td><td rowspan="2">Euro IV</td></tr>
<tr><td>2015</td><td>Euro IV</td></tr>
</table>

Figure S4: Emission standards implemented in major sectors during 1990-2015.

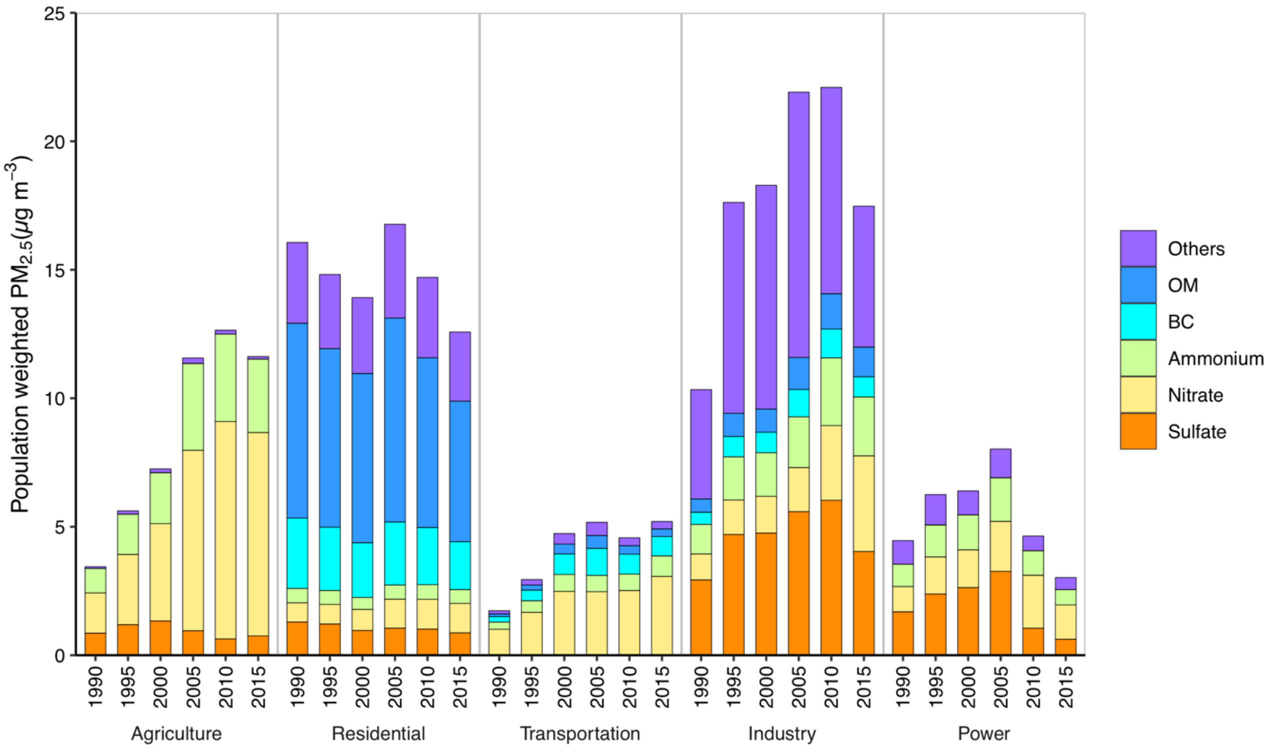

Figure S5: **Chemical composition of the population-weighted $PM_{2.5}$ concentrations by source sectors during 1990-2015.**

Table S1: Performance statistics for meteorological fields simulated by WRF

year	Meteorological field	Data Pairs	R	Observed mean	Simulated mean	MB	RMSE	ME	NMB (%)	NME (%)
2000	2-m temperature (K)	4192027	0.98	13.6	13.5	-0.1	2.8	2.0	-0.6	14.4
	2-m relative humidity (%)	4178440	0.79	70.5	73.8	3.3	12.8	9.4	4.7	13.4
	10-m wind speed (m/s)	4150033	0.57	2.8	2.8	0.0	2.1	1.5	0.7	55.2
	10-m wind direction (°)	3513773	0.41	193.9	190.9	1.9	64.2	46.6	1.0	24.0
2005	2-m temperature (K)	4747580	0.98	13.8	13.6	-0.2	2.7	1.9	-1.2	13.8
	2-m relative humidity (%)	4737573	0.81	68.8	72.8	3.9	12.6	9.4	5.7	13.7
	10-m wind speed (m/s)	4741462	0.60	2.8	2.9	0.1	2.0	1.5	3.7	53.5
	10-m wind direction (°)	4087836	0.42	194.8	196.8	1.8	64.6	46.7	0.9	24.0
2010	2-m temperature (K)	5099963	0.98	14.2	14.0	-0.2	2.6	1.8	-1.2	12.9
	2-m relative humidity (%)	5090225	0.82	69.5	73.2	3.7	12.1	9.1	5.3	13.0
	10-m wind speed (m/s)	5093539	0.53	2.7	3.1	0.4	2.2	1.6	14.4	60.6
	10-m wind direction (°)	4368053	0.38	191.0	197.5	0.8	68.8	50.6	0.4	26.5
2015	2-m temperature (K)	5153973	0.98	14.9	14.5	-0.3	2.7	1.9	-2.3	13.0
	2-m relative humidity (%)	5145148	0.82	69.9	73.7	3.8	12.6	9.5	5.4	13.5
	10-m wind speed (m/s)	4924354	0.61	2.8	2.8	0.1	1.9	1.4	2.8	50.8
	10-m wind direction (°)	4165012	0.40	193.6	188.5	2.0	65.7	47.6	1.0	24.6