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Quantification and evaluation of atmospheric pollutant emissions from open biomass burning with multiple methods: a case study for the Yangtze River Delta region, China

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1. Observation of PM_{10} concentrations

The PM_{10} mass concentrations were obtained with Air Pollution Index (API) from China National Environmental Monitoring Center (http://www.cnemc.cn/). The API of PM_{10}, SO_2 and NO_2 of a city were calculated in every day of 2010 and 2012, and the highest of the three values were published. The API for PM_{10} was usually highest during the OBB event, so the PM_{10} concentrations were calculated from API were reasonable in this study. If API scores were between I_i and I_{i+1}, the PM_{10} concentration was calculated with the following equation:

$$C = \frac{(I - I_i) \times (C_{i+1} - C_i)}{(I_{i+1} - I_i)} + C_i \qquad (1)$$

where I is API score; C is the concentration of PM_{10}, and i represents the rank. The relationship between the API score and PM_{10} concentration in different rank was shown in Table S15.

Tables

Table S1. Emission factors for OBB used in traditional bottom-up method.

	Emission factor（g/kg dry crop residue）										
	PM_{10}	$PM_{2.5}$	EC	OC	CH_4	$NMVOC_S$	CO	CO_2	NO_X	SO_2	NH_3
Rice straw	15.7[a]	13.77[b,c]	0.499[d,b]	6.16[d,b]	3.89[e]	8.94[e]	65.2[d,f,b]	1215.3[d,f,b]	2.67[d,f]	0.147[d]	0.525[e]
Wheat straw	25.4[a]	22.25[g,c]	0.505[d,g]	3.26[d,g]	3.36[g]	7.48[g]	88.8[d,g,f]	1502.5[d,g,f]	2.34[d,g,f]	0.449[d,g]	0.37[g]
Maize straw	19.7[a]	17.24[g,c]	0.565[d,g]	3.08[d,g]	4.41[g]	10.4[g]	79.3[d,g,f]	1605.2[d,g,f]	2.98[d,g,f]	0.233[d,g]	0.68[g]
Other	18.0[e]	15.78[e]	0.519[e]	5.38[e]	3.89[e]	8.94[e]	75.5[e]	1358.6[e]	2.75[e]	0.351[e]	0.525[e]

a Ratio of PM_{10} to $PM_{2.5}$ is from Akagi et al. (2011).

b Zhang et al., 2013.

c Zhu et al., 2005.

d Cao et al., 2008.

e Values are the average of the known straw species.

f Zhang et al., 2008.

g Li et al., 2007.

Table S2. The ratios of straw to grain and combustion efficiencies by crop type.

	Ratio of straw to grain [a]	Combustion efficiency [b]
Rice	0.95	92.5%
Wheat	1.3	91.7%
Maize	1.1	91.7%
Other corn	1.1	92.0%
Potato	0.526[c]	92.0%
Peanut	1.5	92.0%
Rapeseed	1.97	92.0%
Cotton	5	92.0%
Bean	1.6	92.0%

a Bi, 2010.
b Zhang et al., 2008.
c Wang et al., 2013.

Table S3. The percentages of CRBF of Shanghai, Jiangsu, Zhejiang and Anhui from 2005 to 2012 used in traditional bottom-up method.

	Shanghai [a, b]	Jiangsu [a, c, d]	Zhejiang [a, e]	Anhui [a, f]
2005	12.50%	20.5%	15%	23.50%
2006	12.50%	20.5%	15%	23.50%
2007	12.50%	20.5%	15%	23.50%
2008	12.50%	20.5%	15%	23.50%
2009	10.40%	17.4%	15%	23.50%
2010	8.60%	14.7%	15%	23.50%
2011	7.20%	12.5%	15%	23.50%
2012	6.00%	9.5%	11%	21.30%

a NDRC, 2014.
b SMDRC and SMAC, 2009.
c JPDRC and SMAC, 2009.
d APDRC, 2012.
e Qian, 2012.
f Xu and Wu, 2012.

Table S4. Emission factors used in FRP-based method (g/Kg dry crop residue).

	PM_{10} [a]	$PM_{2.5}$ [a]	EC [a]	OC [a]	CH_4 [a]	$NMVOC_S$ [a]	CO [a]	CO_2 [a]	NO_X [a]	SO_2 [b]	NH_3 [a]
EF	7.2	6.3	0.8	2.3	5.8	51.4	102.2	1584.9	3.1	0.4	2.2

a Akagi et al., 2011.
b Andreae and Merlet, 2001.

Table S5. Inter-annual Terra/Aqua (T/A) FRP ratios, estimated per-pixel FRE and total crop burnt from 2005 to 2015 in YRD.

	T/A FRP ratio	FRE ($\times 10^6$ MJ)	Total crop burnt（Tg）
2005	0.94	1.95	5.74
2006	0.88	1.78	5.55
2007	0.94	1.70	6.95
2008	1.02	1.64	5.36
2009	1.02	1.49	5.70
2010	0.97	1.59	8.02
2011	0.96	1.53	6.33
2012	0.92	1.80	12.60
2013	0.94	1.61	8.51
2014	1.04	2.49	10.66
2015	0.72	1.52	4.23

Table S6. Model performance statistics for meteorological parameters in D2 at 9km horizontal resolution for June of 2010, 2012 and 2014.

Variables	Parameter	June 2010	June 2012	June 2014	Benchmark
Wind speed	Mean OBS(m/s)	2.29	2.39	2.55	
	Mean SIM(m/s)	2.28	2.45	3.22	
	Bias(m/s)	-0.01	0.06	0.67	
	RMSE(m/s)	0.39	0.38	0.90	⩽2.0[a]
	IOA	0.91	0.89	0.72	⩾0.6[a]
Wind direction	Mean OBS(°)	155.32	138.79	152.69	
	Mean SIM(°)	145.48	132.00	134.47	
	Bias(°)	-9.84	-7.00	-18.22	
	RMSE(°)	26.76	21.02	39.58	⩽44.7[b]
	IOA	0.93	0.89	0.75	
Temperature	Mean OBS（℃）	24.08	25.45	24.23	
	Mean SIM（℃）	23.11	24.81	23.80	
	Bias（℃）	-0.97	-0.64	-0.43	⩽0.5[a]
	RMSE（℃）	1.46	1.76	1.35	
	IOA（%）	0.96	0.91	0.94	⩾0.8[a]
Relative Humidity	Mean OBS（%）	75.16	69.62	77.53	
	Mean SIM（%）	78.15	72.69	77.60	
	Bias（%）	2.99	3.08	0.07	
	RMSE（%）	4.79	7.02	4.59	
	IOA（%）	0.96	0.89	0.97	⩾0.6[a]

Note: [a] from Emery et al. (2001); [b] from Jiménez et al. (2006). OBS and SIM indicate the results from observation and simulation, respectively. The Bias, IOA and RMSE were calculated using following equations (P and O indicates the results from modeling prediction and observation, respectively):

$$Bias = \frac{1}{n}\sum_{i=1}^{n}\left(P_i - O_i\right);\quad IOA = 1 - \frac{\sum_{i=1}^{n}\left(P_i - O_i\right)^2}{\sum_{i=1}^{n}\left(\left|P_i - \bar{O}\right| + \left|O_i - \bar{O}\right|\right)^2};\quad RMSE = \sqrt{\frac{1}{n}\sum_{i=1}^{n}\left(P_i - O_i\right)^2}.$$

Table S7. Model performance statistics for PM_{10} concentrations from observation and CMAQ simulation during non-OBB event in June 2010 and 2012.

Province	City	June 2010		June 2012	
		NMB	NME	NMB	NME
Anhui	Fuyang	-17.9%	42.7%	-7.5%	24.6%
	Bozhou	-43.9%	44.2%	-6.7%	24.4%
	Bengbu	-21.8%	27.5%	-20.4%	25.2%
	Huainan	-30.1%	41.5%	-18.0%	20.2%
	Hefei	2.2%	41.9%	-22.6%	23.0%
	Chuzhou	-31.5%	38.2%	-1.9%	42.6%
Jiangsu	Xuzhou			-50.8%	50.8%
	Lianyungang	-42.2%	40.6%	-52.2%	52.2%
	Nanjing	-34.6%	57.8%	-20.3%	20.3%
	Yangzhou	-26.4%	37.4%	-28.8%	32.3%
	Zhenjiang	-7.4%	36.3%	1.3%	26.9%
	Taizhou	-	-	-31.6%	32.9%
	Nantong	-43.2%	53.2%	-35.8%	35.8%
	Changzhou	-	-	-15.9%	21.9%
	Wuxi	-	-	4.9%	26.2%
	Suzhou	14.0%	36.9%	21.8%	32.4%
Zhejiang	Huzhou	-27.0%	32.3%	-52.2%	52.6%
	Jiaxing	-	-	-27.2%	42.0%
	Hangzhou	-13.1%	18.7%	-27.2%	38.5%
	Shaoxing	-24.6%	31.4%	-49.6%	49.6%
	Ningbo	-38.1%	28.5%	-27.5%	27.3%
Shanghai	Shanghai	43.4%	52.8%	8.1%	44.3%
Average		-19.2%	38.9%	-20.9%	33.9%

Note: NMB and NME are calculated using following equations (P and O indicate the results from modeling prediction and observation, respectively):

$$NMB = \frac{\sum_{i=1}^{n}(P_i - O_i)}{\sum_{i=1}^{n} O_i} \times 100\%; \quad NME = \frac{\sum_{i=1}^{n}|P_i - O_i|}{\sum_{i=1}^{n} O_i} \times 100\%.$$

Table S8. Model performance statistics for $PM_{2.5}$ and PM_{10} concentrations from observation and CMAQ simulation during non-OBB event in June 2014.

Province	City	$PM_{2.5}$				PM_{10}			
		Hourly		Daily		Hourly		Daily	
		NMB	NME	NMB	NME	NMB	NME	NMB	NME
Anhui	Hefei	7.7%	34.9%	7.5%	22.0%	-3.2%	31.9%	-3.2%	15.9%
	Maanshan	-10.1%	45.0%	-10.1%	36.6%	-19.7%	46.3%	-19.6%	37.0%
	Wuhu	-6.5%	53.9%	-6.5%	42.1%	-26.6%	48.4%	-26.8%	40.9%
Jiangsu	Xuzhou	-25.9%	46.6%	-25.3%	32.3%	-43.3%	58.0%	-42.9%	44.8%
	Liangyungang	-43.7%	43.3%	-42.8%	42.8%	-65.3%	68.3%	-65.0%	65.0%
	Huaian	-50.2%	57.9%	-50.1%	53.1%	-51.9%	60.8%	-51.9%	51.9%
	Suqian	-53.2%	53.5%	-53.1%	53.1%	-60.6%	60.9%	-60.8%	60.8%
	Yancheng	-54.4%	58.6%	-53.9%	53.9%	-61.1%	64.3%	-60.5%	60.5%
	Nanjing	-29.5%	42.4%	-29.6%	32.5%	-37.9%	47.8%	-37.8%	40.0%
	Yangzhou	-34.8%	44.3%	-34.6%	37.2%	-50.6%	56.1%	-50.4%	50.4%
	Taizhou	-44.0%	47.2%	-43.3%	43.3%	-51.8%	56.5%	-51.0%	51.0%
	Nantong	-55.7%	56.0%	-54.8%	54.8%	-58.8%	60.4%	-58.2%	58.2%
	Suzhou	-31.6%	45.2%	-31.8%	35.0%	-40.2%	50.1%	-40.3%	41.9%
	Wuxi	-4.5%	56.7%	-2.7%	42.4%	-19.0%	61.7%	-17.5%	47.6%
	Zhenjiang	-29.1%	37.2%	-29.4%	32.1%	-41.2%	51.0%	-41.1%	41.6%
	Changzhou	-17.1%	37.9%	-17.2%	25.8%	-28.7%	45.5%	-28.6%	31.7%
Zhejiang	Hangzhou	-15.5%	44.8%	-16.0%	34.6%	-30.9%	47.8%	-31.0%	40.9%
	Huzhou	-20.2%	49.2%	-19.3%	41.1%	-28.6%	51.1%	-29.4%	46.6%
	Jiaxing	-31.5%	52.6%	-31.0%	39.1%	-38.3%	56.1%	-38.0%	40.3%
	Ningbo	-41.5%	59.9%	-41.7%	44.1%	-50.4%	58.5%	-50.4%	50.4%
	Shaoxing	-46.3%	61.1%	-45.5%	48.4%	-40.3%	54.1%	-41.0%	52.3%
	Shanghai	-20.6%	69.2%	-19.7%	56.2%	-27.0%	69.1%	-30.1%	58.7%
Average		-29.9%	49.8%	-29.6%	41.0%	-39.8%	54.7%	-39.8%	46.7%
Benchmark [a]		-33.0%	43.0%			-45.0%	49.0%		

Note: [a] from Zhang et al. (2006). NMB and NME are calculated using following equations (P and O indicate the results from modeling prediction and observation, respectively):

$$NMB = \frac{\sum_{i=1}^{n}\left(P_i - O_i\right)}{\sum_{i=1}^{n} O_i} \times 100\% ; \quad NME = \frac{\sum_{i=1}^{n}\left|P_i - O_i\right|}{\sum_{i=1}^{n} O_i} \times 100\% .$$

Table S9. Model performance statistics for CO concentrations from observation and CMAQ simulation during non-OBB event in June 2014.

Province	City	Hourly		Daily	
		NMB	NME	NMB	NME
Anhui	Hefei	-31.3%	32.5%	-31.2%	33.9%
	Maanshan	-62.6%	62.6%	-62.6%	62.0%
	Wuhu	-37.3%	41.6%	-35.8%	44.2%
Jiangsu	Xuzhou	-61.1%	61.3%	-61.0%	61.0%
	Liangyungang	-71.3%	71.3%	-71.0%	71.0%
	Huaian	-63.1%	63.3%	-63.1%	63.1%
	Suqian	-60.2%	57.7%	-60.1%	60.1%
	Yancheng	-61.2%	60.1%	-61.5%	61.5%
	Nanjing	-23.2%	36.5%	-23.5%	26.5%
	Yangzhou	-27.2%	36.7%	-27.7%	32.6%
	Taizhou	-58.2%	58.7%	-58.1%	58.1%
	Nantong	-40.1%	43.9%	-40.8%	40.8%
	Suzhou	-21.8%	35.4%	-21.7%	25.7%
	Wuxi	-21.6%	44.2%	-20.3%	33.5%
	Zhengjiang	-52.9%	52.6%	-53.0%	53.0%
	Changzhou	-38.3%	44.4%	-38.2%	38.2%
Zhejiang	Hangzhou	-12.3%	27.1%	-12.6%	14.8%
	Huzhou	-44.8%	46.7%	-44.9%	44.9%
	Jiaxing	-42.8%	46.0%	-43.0%	43.0%
	Ningbo	-64.7%	64.2%	-64.8%	64.8%
	Shaoxing	-10.3%	36.2%	-10.2%	23.1%
	Shanghai	-24.3%	38.7%	-24.1%	25.8%
Average		-42.3%	48.3%	-42.2%	44.6%

Note: NMB and NME are calculated using following equations (P and O indicate the results from modeling prediction and observation, respectively):

$$NMB = \frac{\sum_{i=1}^{n}\left(P_i - O_i\right)}{\sum_{i=1}^{n} O_i} \times 100\%; \quad NME = \frac{\sum_{i=1}^{n}\left|P_i - O_i\right|}{\sum_{i=1}^{n} O_i} \times 100\%.$$

Table S10. Annual OBB emissions in YRD based on traditional bottom-up method from 2005 to 2012 (Unit: Gg).

	PM_{10}	$PM_{2.5}$	EC	OC	CH_4	NMVOCs	CO	CO_2	NO_X	SO_2	NH_3
2005	327.4	286.5	8.6	84.2	64.0	146.2	1276.9	23000	44.4	4.9	8.3
2006	362.9	317.6	9.5	91.4	70.2	160.4	1409.7	25377	48.7	5.4	9.1
2007	352.9	308.8	9.1	86.4	67.0	152.8	1360.9	24426	46.4	5.2	8.6
2008	392.3	343.2	10.1	94.5	73.9	168.4	1509.2	27061	51.2	5.9	9.5
2009	380.4	332.9	9.8	91.1	71.7	163.5	1464.1	26283	49.6	5.7	9.2
2010	362.4	317.1	9.3	85.7	67.9	154.9	1391.8	24978	47.0	5.4	8.7
2011	348.0	304.5	8.9	82.1	65.3	148.9	1336.2	24000	45.2	5.2	8.4
2012	290.3	254.0	7.4	68.0	54.2	123.5	1113.3	19973	37.5	4.3	7.0

Table S11. Annual OBB emissions in YRD based on FRP-method from 2005 to 2015 (Unit: Gg).

	PM_{10}	$PM_{2.5}$	EC	OC	CH_4	NMVOCs	CO	CO_2	NO_X	SO_2	NH_3
2005	41.3	36.1	4.6	13.2	33.3	294.9	586.4	9093.3	17.8	2.3	12.6
2006	40.0	35.0	4.4	12.8	32.2	285.3	567.3	8797.2	17.2	2.2	12.2
2007	50.0	43.8	5.6	16.0	40.3	357.0	709.8	11007.9	21.5	2.8	15.3
2008	38.6	33.8	4.3	12.3	31.1	275.7	548.1	8500.1	16.6	2.1	11.8
2009	41.1	35.9	4.6	13.1	33.1	293.2	583.0	9040.5	17.7	2.3	12.5
2010	57.8	50.6	6.4	18.5	46.5	412.5	820.1	12718.0	24.9	3.2	17.7
2011	45.6	39.9	5.1	14.6	36.7	325.5	647.1	10035.2	19.6	2.5	13.9
2012	90.8	79.4	10.1	29.0	73.1	647.9	1288.2	19977.3	39.1	5.0	27.7
2013	61.3	53.6	6.8	19.6	49.4	437.5	869.8	13489.4	26.4	3.4	18.7
2014	76.8	67.2	8.5	24.5	61.9	548.2	1089.9	16902.2	33.1	4.3	23.5
2015	30.5	26.7	3.4	9.7	24.5	217.5	432.4	6706.0	13.1	1.7	9.3

Table S12. OBB emissions with traditional bottom-up method by city in YRD 2012 (Unit: Gg).

	PM_{10}	$PM_{2.5}$	EC	OC	CH_4	$NMVOC_S$	CO	CO_2	NO_X	SO_2	NH_3
Anqing	11.2	9.8	0.3	3.8	2.6	5.9	46	845	1.8	0.1	0.3
Bengbu	15.3	13.4	0.4	3.1	2.6	6	58	1021	1.8	0.2	0.3
Bozhou	26.1	22.9	0.6	3.9	4	9.1	95	1674	2.8	0.4	0.5
Chaohu	10.2	9	0.3	3.2	2.2	5.1	41	752	1.5	0.1	0.3
Chizhou	3.1	2.7	0.1	1.1	0.7	1.6	13	235	0.5	0	0.1
Chuzhou	19.2	16.8	0.5	4.7	3.6	8.2	74	1313	2.5	0.3	0.5
Fuyang	29.5	25.8	0.7	4.7	4.7	10.8	108	1923	3.3	0.5	0.6
Hefei	8.9	7.8	0.3	2.7	1.9	4.4	36	649	1.3	0.1	0.3
Huaibei	7.1	6.2	0.2	1.1	1.1	2.4	26	453	0.8	0.1	0.1
Huainan	6.2	5.4	0.1	1.4	1.1	2.4	23	407	0.7	0.1	0.1
Huangshan	1.5	1.3	0	0.5	0.3	0.8	6	112	0.2	0	0
Luan	19.1	16.7	0.5	5.2	3.8	8.7	74	1338	2.6	0.3	0.5
Maanshan	2.1	1.8	0.1	0.6	0.4	1	8	150	0.3	0	0.1
Suzhou	22.4	19.6	0.5	3.6	3.7	8.4	83	1484	2.5	0.4	0.5
Tongling	0.7	0.7	0	0.2	0.2	0.4	3	55	0.1	0	0
Wuhu	3.1	2.7	0.1	1	0.7	1.6	13	234	0.5	0	0.1
Xuancheng	5.8	5	0.2	1.7	1.2	2.8	23	418	0.8	0.1	0.2
Changzhou	2.1	1.9	0.1	0.6	0.4	0.9	8	148	0.3	0	0.1
Huaian	8.9	7.8	0.2	2.1	1.7	3.8	34	607	1.1	0.1	0.2
Lianyungang	7.3	6.4	0.2	1.7	1.3	3.1	28	495	0.9	0.1	0.2
Nanjing	2.3	2	0.1	0.7	0.5	1.1	9	164	0.3	0	0.1
Nantong	7.7	6.7	0.2	1.9	1.5	3.4	30	541	1	0.1	0.2
Suqian	8	7	0.2	1.7	1.4	3.2	30	533	1	0.1	0.2
Suzhou	2.2	1.9	0.1	0.6	0.4	1	9	151	0.3	0	0.1
Taizhou	6.5	5.7	0.2	1.6	1.2	2.8	25	442	0.8	0.1	0.2
Wuxi	1.5	1.4	0	0.4	0.3	0.7	6	105	0.2	0	0
Xuzhou	10.4	9.1	0.3	2	1.8	4.2	39	702	1.3	0.2	0.2
Yancheng	13.7	12	0.4	3.4	2.7	6.1	53	962	1.8	0.2	0.3
Yangzhou	6	5.3	0.2	1.5	1.1	2.6	23	409	0.8	0.1	0.1
Zhenjiang	2.5	2.2	0.1	0.7	0.5	1.1	10	172	0.3	0	0.1
Hangzhou	2.2	1.9	0.1	0.7	0.5	1.1	9	165	0.3	0	0.1
Huzhou	1.9	1.6	0.1	0.6	0.4	0.9	8	137	0.3	0	0.1
Jiaxing	2.8	2.5	0.1	0.9	0.6	1.4	11	207	0.4	0	0.1
Jinhua	1.8	1.5	0.1	0.6	0.4	0.9	7	135	0.3	0	0.1
Lishui	1	0.8	0	0.3	0.2	0.5	4	73	0.2	0	0
Ningbo	1.7	1.5	0.1	0.6	0.4	0.9	7	128	0.3	0	0.1
Quzhou	1.6	1.4	0	0.6	0.4	0.9	7	123	0.3	0	0.1
Shaoxing	2.3	2	0.1	0.8	0.5	1.2	9	170	0.4	0	0.1
Taizhou	1.6	1.4	0	0.5	0.4	0.8	7	121	0.3	0	0
Wenzhou	1.5	1.3	0	0.5	0.4	0.8	6	116	0.2	0	0
Zhoushan	0.1	0.1	0	0	0	0.1	1	9	0	0	0
Shanghai	1.3	1.1	0	0.4	0.3	0.6	5	93	0.2	0	0
Total	290.3	254	7.4	68	54.2	123.5	1113	19973	37.5	4.3	7

Table S13. OBB emissions with FRP-based method by city in YRD 2012 (Unit: Gg).

	PM_{10}	$PM_{2.5}$	EC	OC	CH_4	NMVOCs	CO	CO_2	NO_X	SO_2	NH_3
Anqing	0.7	0.6	0.1	0.2	0.6	5.2	10	160	0.3	0	0.2
Bengbu	4	3.5	0.4	1.3	3.2	28.7	57	885	1.7	0.2	1.2
Bozhou	7.8	6.8	0.9	2.5	6.3	55.9	111	1723	3.4	0.4	2.4
Chaohu	0.7	0.6	0.1	0.2	0.5	4.7	9	144	0.3	0	0.2
Chizhou	0.5	0.4	0.1	0.2	0.4	3.6	7	111	0.2	0	0.2
Chuzhou	2.8	2.4	0.3	0.9	2.2	19.9	40	613	1.2	0.2	0.8
Fuyang	6.5	5.7	0.7	2.1	5.3	46.6	93	1436	2.8	0.4	2
Hefei	1.2	1.1	0.1	0.4	1	8.9	18	275	0.5	0.1	0.4
Huaibei	5.2	4.6	0.6	1.7	4.2	37.2	74	1147	2.2	0.3	1.6
Huainan	0.7	0.6	0.1	0.2	0.6	4.9	10	151	0.3	0	0.2
Huangshan	0.4	0.3	0	0.1	0.3	2.7	5	84	0.2	0	0.1
Luan	1.6	1.4	0.2	0.5	1.3	11.7	23	361	0.7	0.1	0.5
Maanshan	0.5	0.4	0.1	0.2	0.4	3.6	7	110	0.2	0	0.2
Suzhou	16.5	14.4	1.8	5.3	13.3	117.6	234	3627	7.1	0.9	5
Tongling	0.2	0.2	0	0.1	0.1	1.3	3	40	0.1	0	0.1
Wuhu	0.9	0.8	0.1	0.3	0.7	6.6	13	204	0.4	0.1	0.3
Xuancheng	1.1	1	0.1	0.4	0.9	8	16	248	0.5	0.1	0.3
Changzhou	0.9	0.8	0.1	0.3	0.8	6.7	13	206	0.4	0.1	0.3
Huaian	2.5	2.2	0.3	0.8	2	18.1	36	560	1.1	0.1	0.8
Lianyungang	2.9	2.5	0.3	0.9	2.3	20.8	41	640	1.3	0.2	0.9
Nanjing	1.1	1	0.1	0.4	0.9	7.9	16	243	0.5	0.1	0.3
Nantong	0.4	0.4	0	0.1	0.4	3.1	6	97	0.2	0	0.1
Suqian	4.6	4	0.5	1.5	3.7	32.8	65	1011	2	0.3	1.4
Suzhou	1.4	1.2	0.2	0.4	1.1	10	20	310	0.6	0.1	0.4
Taizhou	1.1	0.9	0.1	0.3	0.9	7.6	15	236	0.5	0.1	0.3
Wuxi	1.2	1	0.1	0.4	1	8.5	17	261	0.5	0.1	0.4
Xuzhou	7.2	6.3	0.8	2.3	5.8	51.5	102	1589	3.1	0.4	2.2
Yancheng	2.2	1.9	0.2	0.7	1.8	15.9	32	489	1	0.1	0.7
Yangzhou	1.4	1.3	0.2	0.5	1.2	10.3	21	318	0.6	0.1	0.4
Zhenjiang	0.4	0.3	0	0.1	0.3	2.5	5	77	0.2	0	0.1
Hangzhou	1.8	1.5	0.2	0.6	1.4	12.5	25	386	0.8	0.1	0.5
Huzhou	1.1	1	0.1	0.4	0.9	8.1	16	250	0.5	0.1	0.3
Jiaxing	1.1	1	0.1	0.4	0.9	8	16	245	0.5	0.1	0.3
Jinhua	1	0.9	0.1	0.3	0.8	6.9	14	214	0.4	0.1	0.3
Lishui	0.6	0.5	0.1	0.2	0.5	4.1	8	125	0.2	0	0.2
Ningbo	1.6	1.4	0.2	0.5	1.3	11.5	23	356	0.7	0.1	0.5
Quzhou	0.9	0.8	0.1	0.3	0.7	6.3	13	195	0.4	0	0.3
Shaoxing	1	0.8	0.1	0.3	0.8	6.8	14	209	0.4	0.1	0.3
Taizhou	0.7	0.6	0.1	0.2	0.5	4.7	9	146	0.3	0	0.2
Wenzhou	0.7	0.6	0.1	0.2	0.5	4.8	10	147	0.3	0	0.2
Zhoushan	0.1	0.1	0	0	0.1	0.5	1	14	0	0	0
Shanghai	1.5	1.3	0.2	0.5	1.2	10.9	22	336	0.7	0.1	0.5
Total	90.8	79.4	10.1	29	73.1	648	1288	19977	39.1	5	27.7

Table S14. Constrained OBB emissions by city in YRD 2012 (Unit: Gg).

	PM_{10}	$PM_{2.5}$	EC	OC	CH_4	$NMVOC_S$	CO	CO_2	NO_X	SO_2	NH_3
Anqing	2.2	1.9	0.1	0.7	0.5	6.3	9	166	0.3	0	0.1
Bengbu	16.4	14.4	0.4	3.2	2.8	36.8	61	1090	2	0.2	0.3
Bozhou	37.9	33.2	0.9	5.7	5.8	75.9	138	2433	4.1	0.6	0.7
Chaohu	7	6.2	0.2	2.2	1.5	20.1	28	513	1	0.1	0.2
Chizhou	10.4	9.2	0.3	3.6	2.4	31.6	43	790	1.7	0.2	0.3
Chuzhou	11.1	9.7	0.3	2.8	2	27.0	43	758	1.4	0.2	0.2
Fuyang	31.5	27.6	0.7	5.1	5	66.1	116	2061	3.5	0.6	0.7
Hefei	4.2	3.6	0.1	1.3	0.9	12.1	17	300	0.6	0.1	0.1
Huaibei	26.2	23	0.6	3.9	3.9	51.7	95	1666	2.8	0.5	0.5
Huainan	2.8	2.4	0.1	0.6	0.5	6.3	10	182	0.3	0.1	0.1
Huangshan	1.2	1	0	0.4	0.3	3.4	5	95	0.2	0	0
Luan	6.4	5.6	0.2	1.7	1.3	16.7	25	450	0.9	0.1	0.2
Maanshan	1.7	1.5	0.1	0.6	0.4	5.2	7	126	0.2	0	0.1
Suzhou	75.6	66.1	1.8	12.1	12.4	162.1	280	5007	8.6	1.3	1.6
Tongling	0.6	0.5	0	0.2	0.2	1.7	2	39	0.1	0	0
Wuhu	2.6	2.3	0.1	0.9	0.6	7.5	11	197	0.4	0	0.1
Xuancheng	3.9	3.4	0.1	1.2	0.9	10.9	16	284	0.6	0.1	0.1
Changzhou	3.6	3.2	0.1	0.9	0.7	9.2	14	245	0.5	0.1	0.1
Huaian	10.8	9.5	0.2	2.6	2	26.4	41	735	1.3	0.2	0.2
Lianyungang	11.5	10.1	0.3	2.7	2.1	27.6	44	790	1.5	0.2	0.2
Nanjing	3.8	3.3	0.1	1.1	0.8	10.3	15	276	0.6	0.1	0.1
Nantong	1.5	1.3	0.1	0.4	0.3	4.0	6	111	0.2	0	0
Suqian	15.8	13.8	0.4	3.5	2.8	36.8	59	1058	2	0.2	0.4
Suzhou	5.1	4.4	0.2	1.3	0.9	12.6	20	348	0.6	0.1	0.2
Taizhou	4.1	3.6	0.1	1	0.8	9.8	16	276	0.6	0.1	0.1
Wuxi	4.5	3.9	0.1	1.1	0.9	10.9	17	308	0.6	0.1	0.1
Xuzhou	31.3	27.4	0.8	6.2	5.5	71.9	117	2109	3.8	0.5	0.7
Yancheng	8.5	7.4	0.2	2.1	1.7	21.8	33	600	1.2	0.2	0.2
Yangzhou	5.8	5.1	0.2	1.4	1.1	13.8	22	395	0.7	0.1	0.2
Zhenjiang	1.3	1.2	0	0.4	0.2	3.4	6	95	0.2	0	0
Hangzhou	6.1	5.4	0.2	1.9	1.3	17.8	25	458	1	0.1	0.2
Huzhou	3.9	3.4	0.1	1.3	0.9	11.5	16	284	0.6	0.1	0.1
Jiaxing	3.6	3.2	0.1	1.1	0.8	9.8	14	261	0.6	0.1	0.1
Jinhua	3.1	2.8	0.1	1	0.7	9.8	13	245	0.5	0.1	0.1
Lishui	1.9	1.7	0.1	0.6	0.5	5.7	8	142	0.3	0	0.1
Ningbo	4.7	4.1	0.2	1.6	1.1	13.8	20	355	0.7	0.1	0.2
Quzhou	2.7	2.4	0.1	0.9	0.6	8.6	11	205	0.5	0	0.1
Shaoxing	2.7	2.4	0.1	0.9	0.6	8.6	11	205	0.5	0	0.1
Taizhou	2.2	2	0.1	0.7	0.5	6.9	10	166	0.3	0	0.1
Wenzhou	3.1	2.7	0.1	1	0.7	9.8	13	237	0.5	0	0.1
Zhoushan	0.2	0.2	0	0.1	0.1	0.6	1	16	0	0	0
Shanghai	5.2	4.6	0.2	1.6	1.1	14.9	22	395	0.8	0.1	0.2
Total	389.0	340.4	9.6	83.6	70.2	919.4	1479	26474	48.6	6.0	9.0

Table S15. Model performance statistics for CO concentrations from observation and CMAQ simulation without OBB emissions (No_OBB) and with OBB emissions based on FRP-based (FRP_OBB) and constraining methods (Constrained_OBB) for the OBB events of June 2014.

	No_OBB		FRP_OBB		Constrained_OBB	
	NMB	NME	NMB	NME	NMB	NME
Hourly	-59.2%	59.9%	-46.7%	50.3%	-44.6%	49.7%
Daily	-59.5%	59.5%	-47.0%	47.0%	-44.9%	45.0%

Table S16. Uncertainties of emission factors and percentages of CRBF, expressed as the probability distribution functions (PDF).

Parameters		Distribution	Key characteristics for distribution functions		
			P10 [a]	P90 [a]	P50 [a]
Emission factors g/kg					
PM_{10}	Rice straw	Uniform	14.7	16.7	15.7
	Wheat straw	Uniform	7.8	38.3	23.0
	Maize straw	Uniform	13.5	24.5	19.0
	Other	Uniform	12.8	26.7	19.8
$PM_{2.5}$	Rice straw	Uniform	12.9	14.6	13.8
	Wheat straw	Uniform	6.7	33.6	20.1
	Maize straw	Uniform	11.9	21.6	16.8
	Other	Uniform	11.1	23.4	17.4
EC	Rice straw	Uniform	0.5	0.5	0.5
	Wheat straw	Uniform	0.4	0.6	0.5
	Maize straw	Uniform	0.3	0.7	0.5
	Other	Uniform	0.4	0.6	0.5
OC	Rice straw	Uniform	2.8	9.5	6.1
	Wheat straw	Uniform	1.9	3.6	2.8
	Maize straw	Uniform	2.4	3.8	3.1
	Other	Uniform	3.2	9.6	6.5
CH_4	Rice straw	Uniform	3.5	4.3	3.9
	Wheat straw	Normal	2.3	4.4	3.4
	Maize straw	Normal	3.2	5.7	4.4
	Other	Uniform	3.5	4.3	3.9
NMVOCs	Rice straw	Uniform	7.8	10.1	8.9
	Wheat straw	Normal	5.1	9.9	7.5
	Maize straw	Normal	4.2	17.3	10.5
	Other	Uniform	7.8	10.1	8.9
CO	Rice straw	Uniform	60.3	70.9	65.6
	Wheat straw	Uniform	67.6	132.8	100.1
	Maize straw	Uniform	55.5	108.1	82.0
	Other	Uniform	63.7	102.1	82.7
CO_2	Rice straw	Uniform	885.4	1659.9	1267.2
	Wheat straw	Uniform	1433.8	1544.1	1488.9
	Maize straw	Uniform	1356.7	2109.2	1727.4
	Other	Uniform	1156.9	1662.9	1409.4
NO_X	Rice straw	Uniform	2.0	3.3	2.7
	Wheat straw	Uniform	1.5	4.7	3.1
	Maize straw	Uniform	1.8	5.6	3.7
	Other	Uniform	1.6	3.6	2.6
SO_2	Rice straw	Normal	0.1	0.6	0.3
	Wheat straw	Uniform	0.1	0.8	0.5
	Maize straw	Uniform	0.1	0.4	0.2
	Other	Uniform	0.1	0.6	0.4
NH_3	Rice straw	Uniform	0.4	0.7	0.5
	Wheat straw	Normal	0.2	0.6	0.4
	Maize straw	Normal	0.2	1.4	0.8
	Other	Uniform	0.4	0.7	0.5

Parameters	Distribution	Key characteristics for distribution functions		
		P10[a]	P90[a]	P50[a]
Percentages of CRBF				
Anhui	Normal	12.3%	30.4%	21.3%
Jiangsu	Normal	5.4%	13.6%	9.5%
Zhejiang	Normal	6.5%	15.6%	11.0%
Jiangsu	Normal	5.4%	13.6%	9.5%

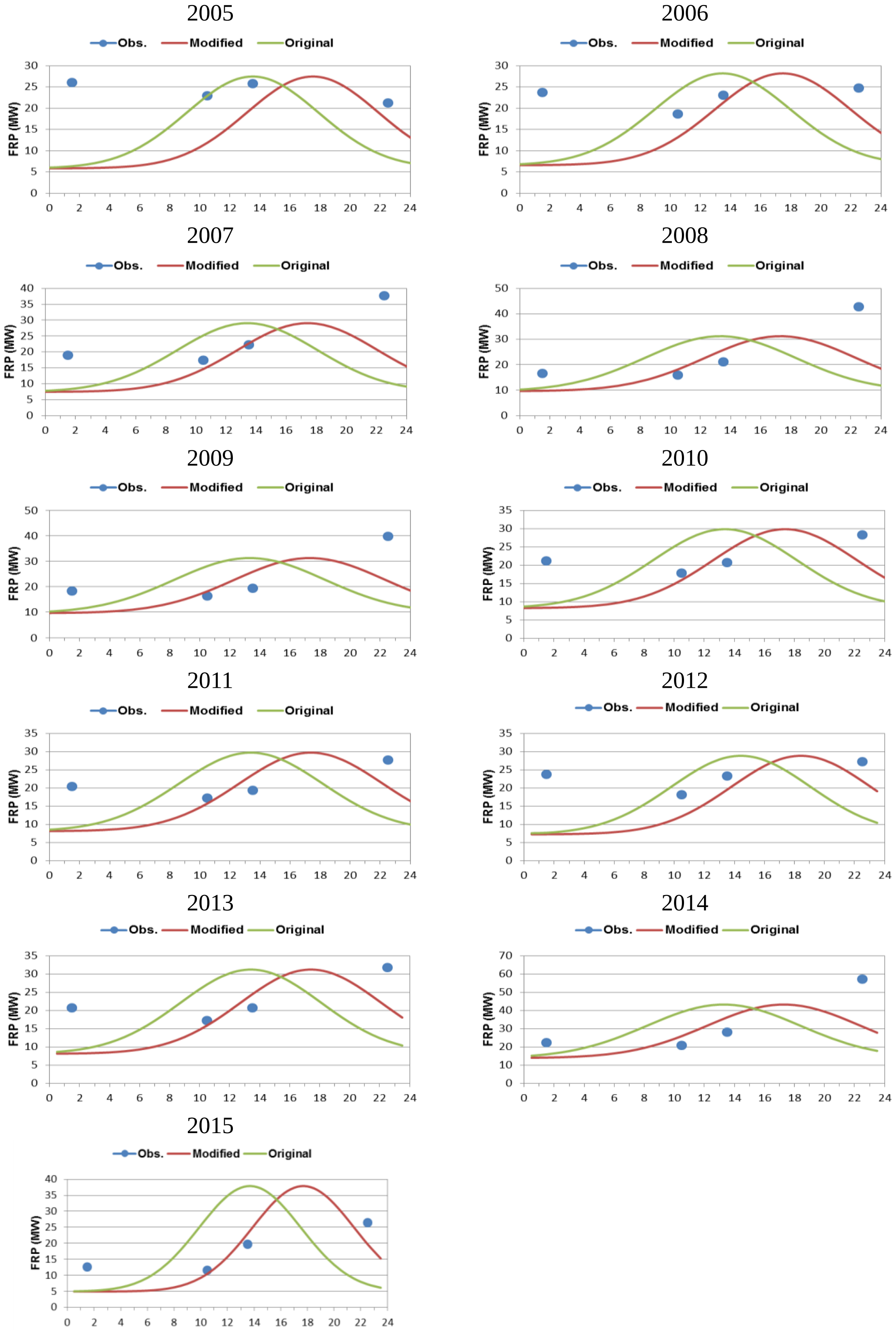
[a] P10 values mean that there is a probability of 10% that the actual result would be equal to or below the P10 values; P50 mean that there is a probability of 50% that the actual result would be equal to or below the P50 values; and P90 mean that there is a probability of 90% that the actual result would be equal to or below the P90 values.

Table S17. The relationship between the API score and PM_{10} concentration in different rank.

Rank	1	2	3	4	5	6
API	50	100	200	300	400	500
PM_{10} (μg/m^3)	50	150	350	420	500	600

Figures

Figure S1. Comparison of original (green lines) and modified (red lines) FRP diurnal curves for 2005-2015. Blue scatters represent the observed mean FRP values at each overpass time.



179 Figure S2. Observed $PM_{2.5}$ and PM_{10} concentrations at Caochangmen Station in
180 Nanjing in June 2012.

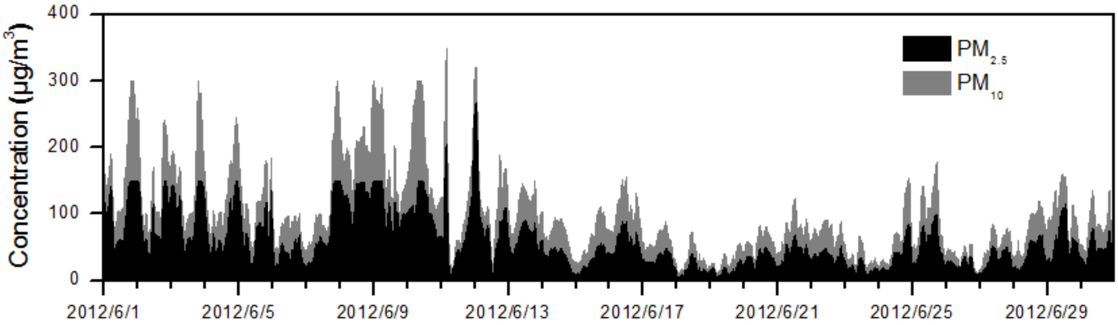


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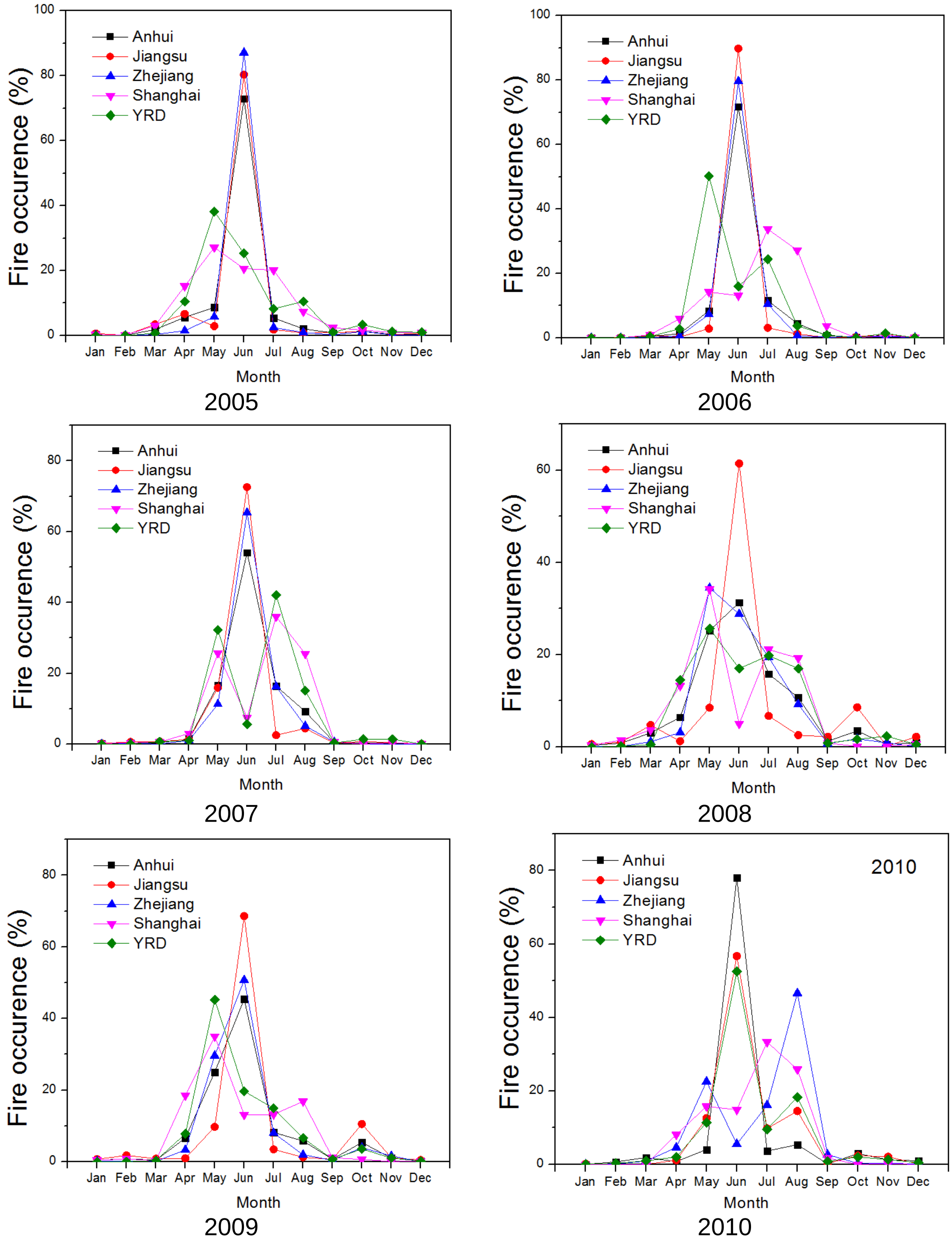
Figure S3. Monthly variations of fire occurrences from 2005-2015.



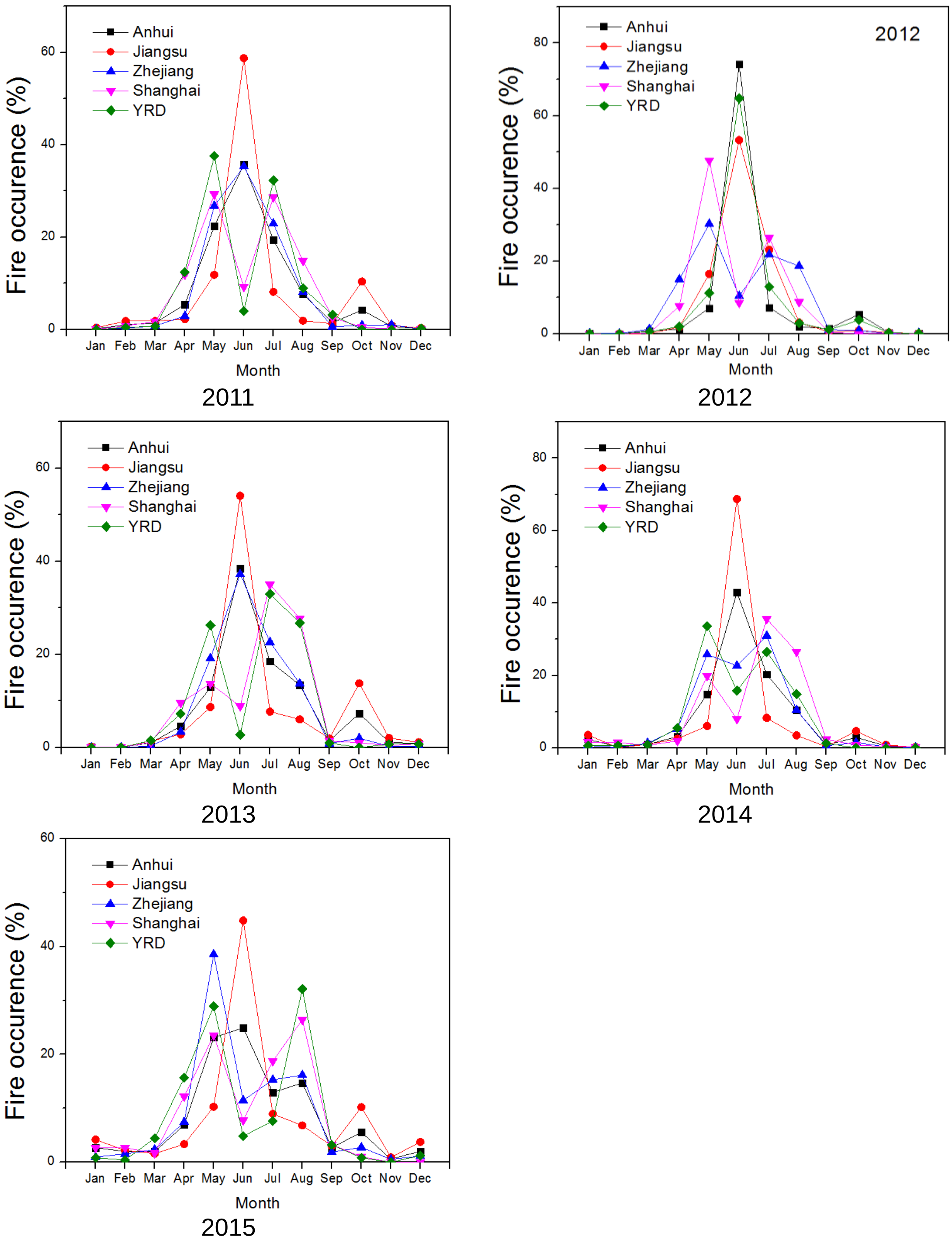
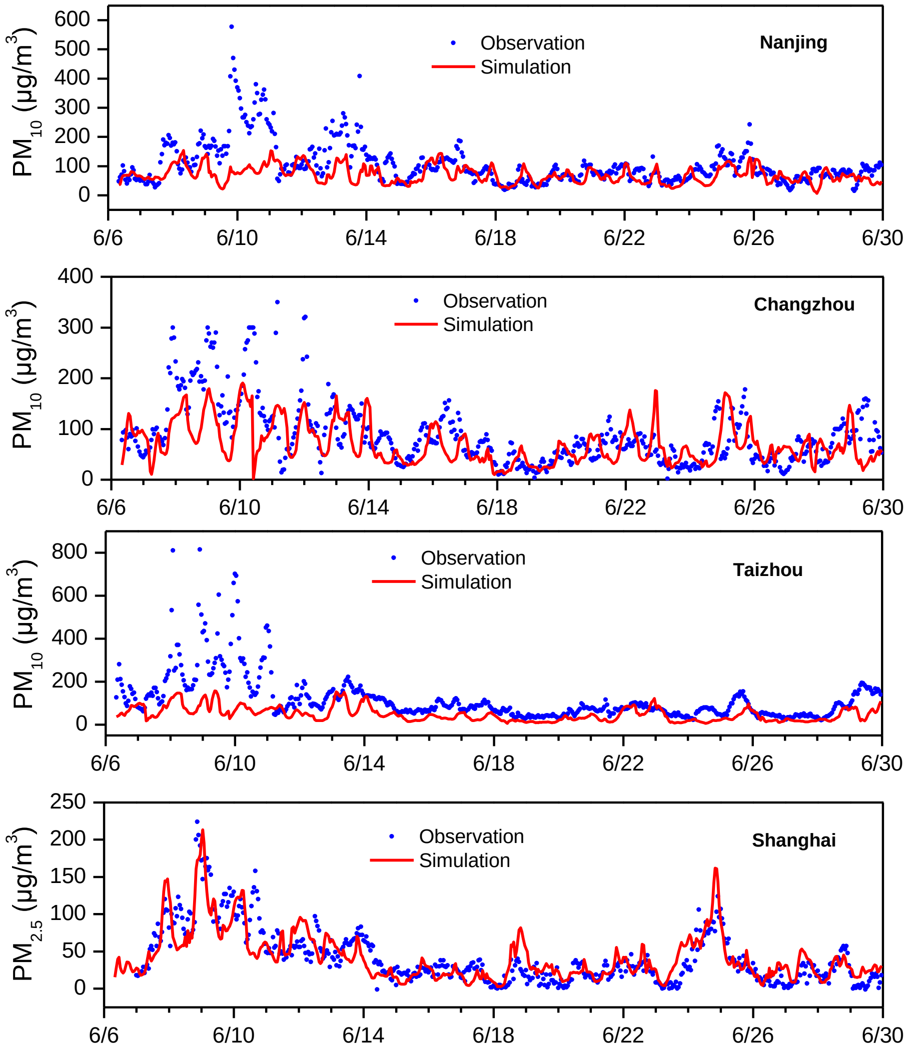

Anhui
Jiangsu
Zhejiang
Shanghai
YRD
Fire occurence (%)
Month
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec
2011
2012
2013
2014
2015

Figure S4. The hourly PM_{10} or $PM_{2.5}$ ground concentrations from observation and CMAQ simulation using emission inventories without OBB at the four air quality monitoring sites in YRD for June 2012.



Note: The hourly PM_{10} concentrations of Nanjing, Changzhou and Taizhou were obtained from Jiangsu province environmental monitoring center (http://www.jsem.net.cn/); the hourly $PM_{2.5}$ concentrations of Shanghai were obtained from U.S. Embassy & Consulates in China (https://china.usembassy-china.org.cn/embassy-consulates/shanghai/air-quality-monitor-stateair).

Figure S5. The inter-annual trends for original FRE, modified FRE, improved FRE and constrained mass of CRBF from 2005 to 2012. All the data are normalized to 2010 level.

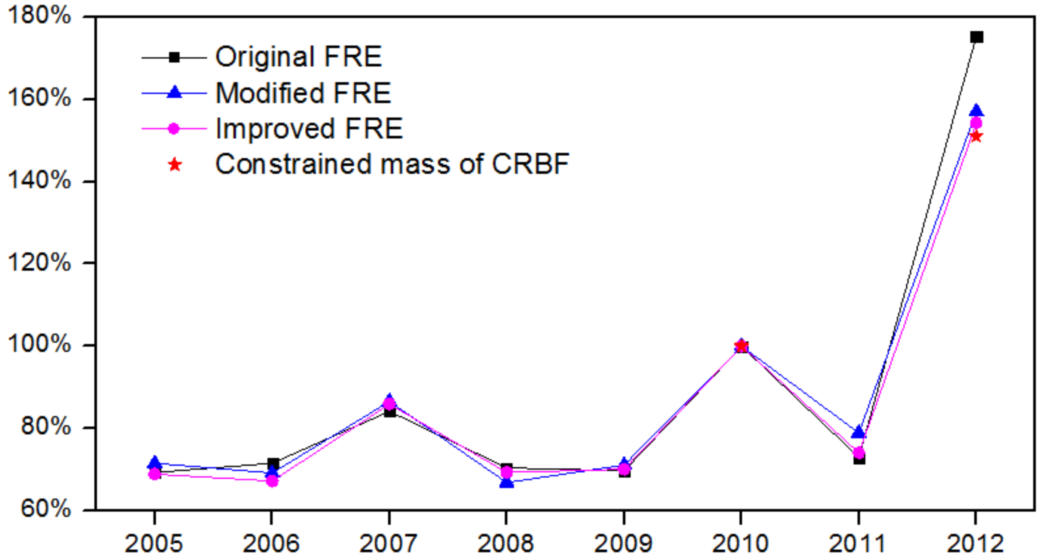


Figure S6. FRP diurnal curves based on curve fitting for 2005-2015. Blue scatters represent the observed total FRP values at each overpass time.

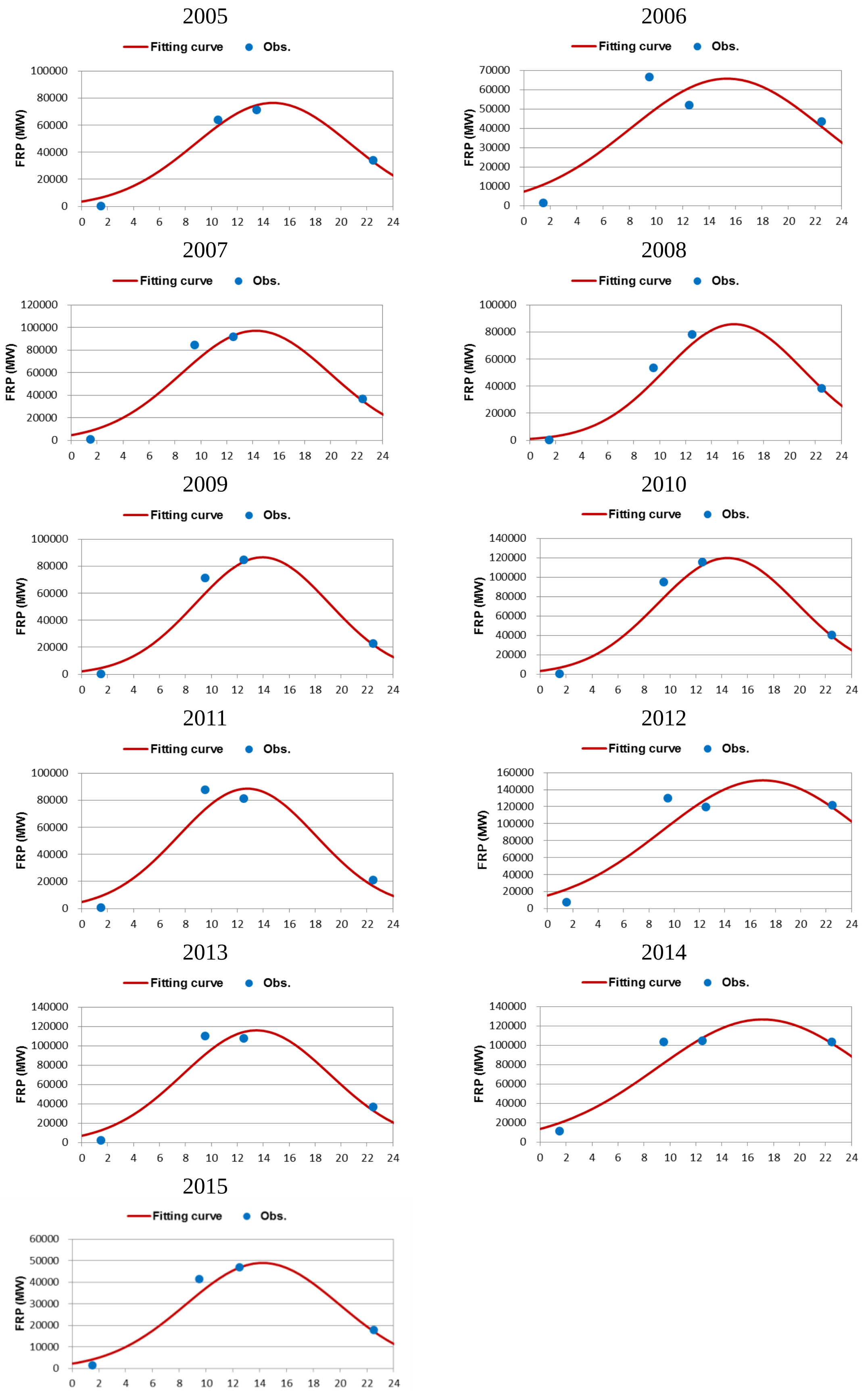


Figure S7. The mass of CRBF estimated in bottom-up method for 2005-2012, and that derived from FRP-based and constraining methods for 2005-2015.

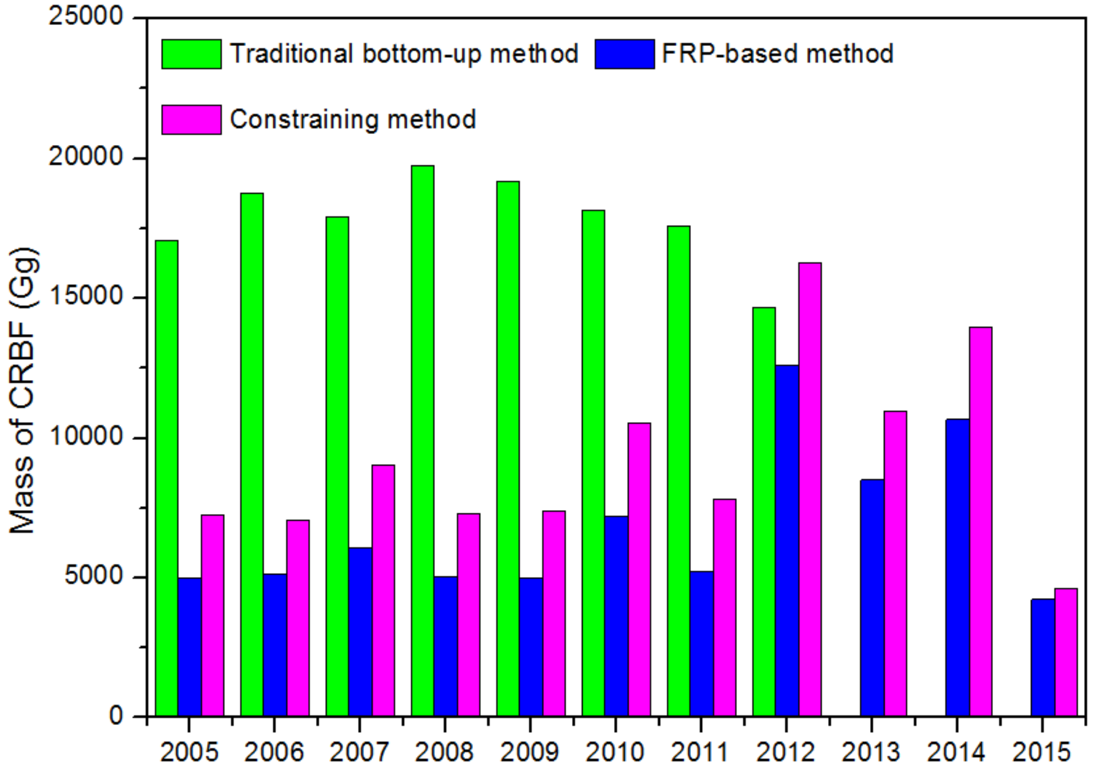


Figure S8. The constrained percentages of CRBF for different regions in YRD from 2005 to 2012.

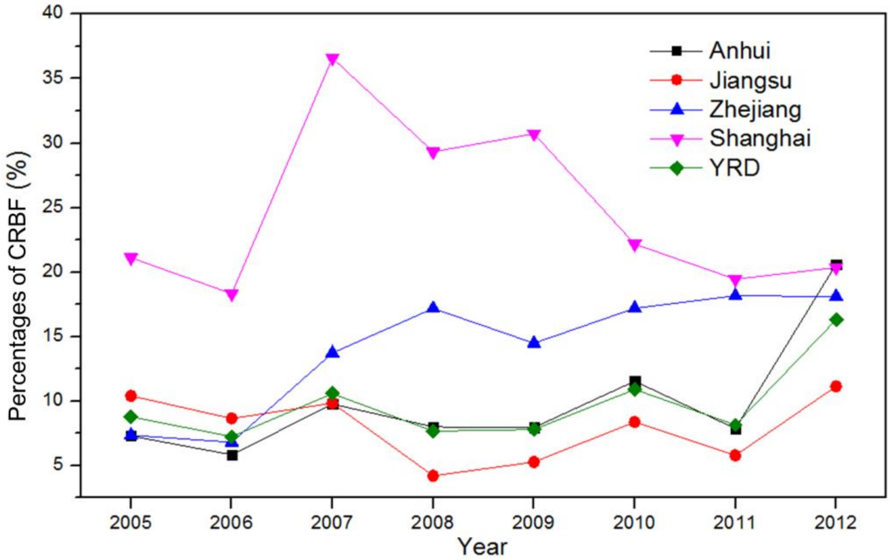
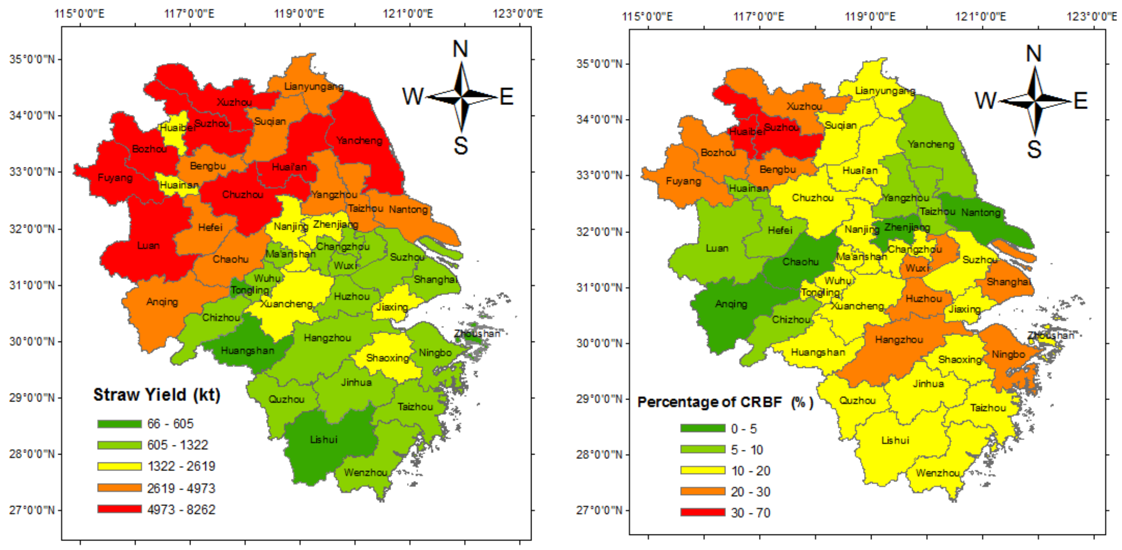
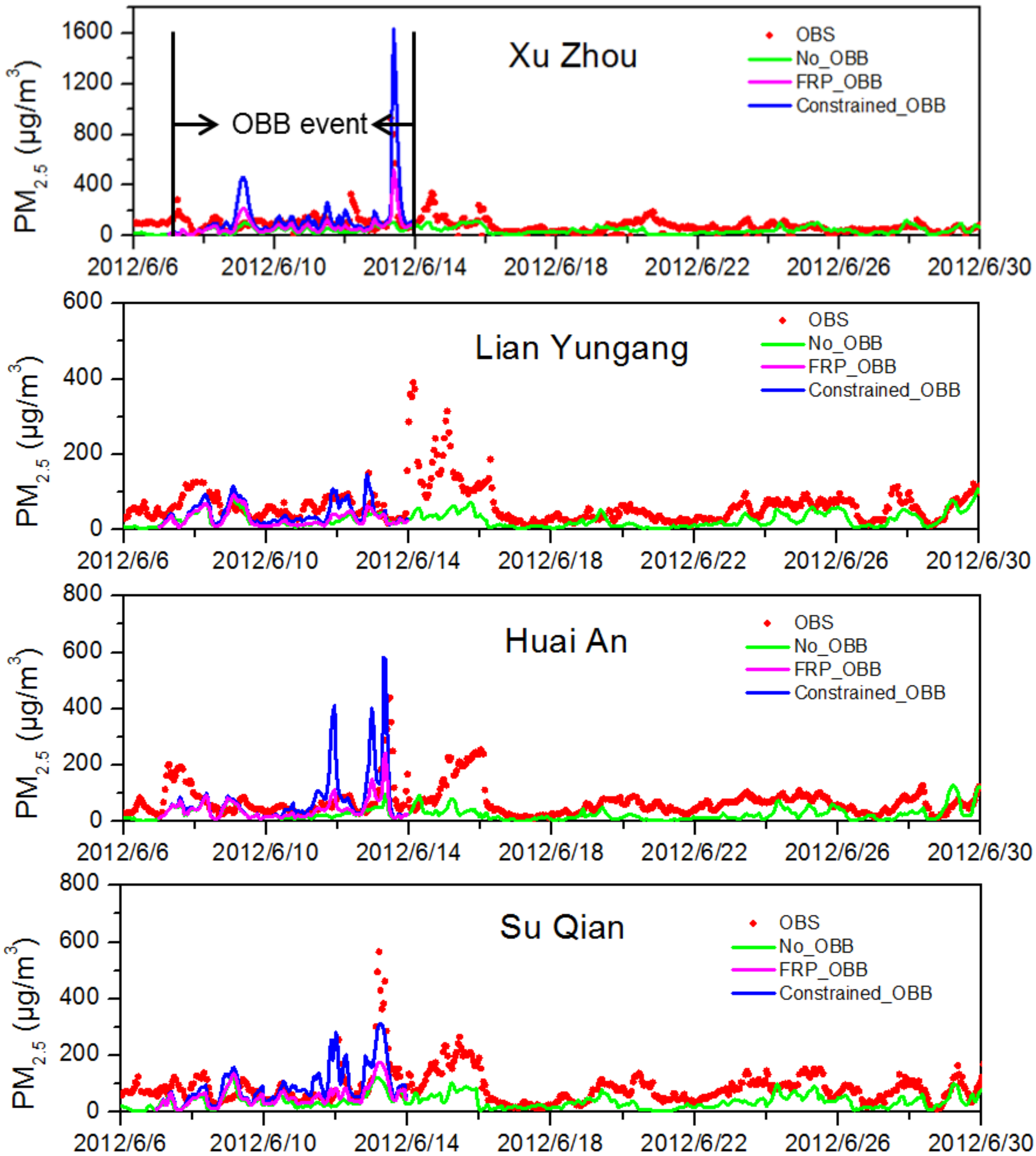


Figure S9. The spatial distribution of straw yield (a) and constrained percentage of CRBF (b) for YRD in 2012.

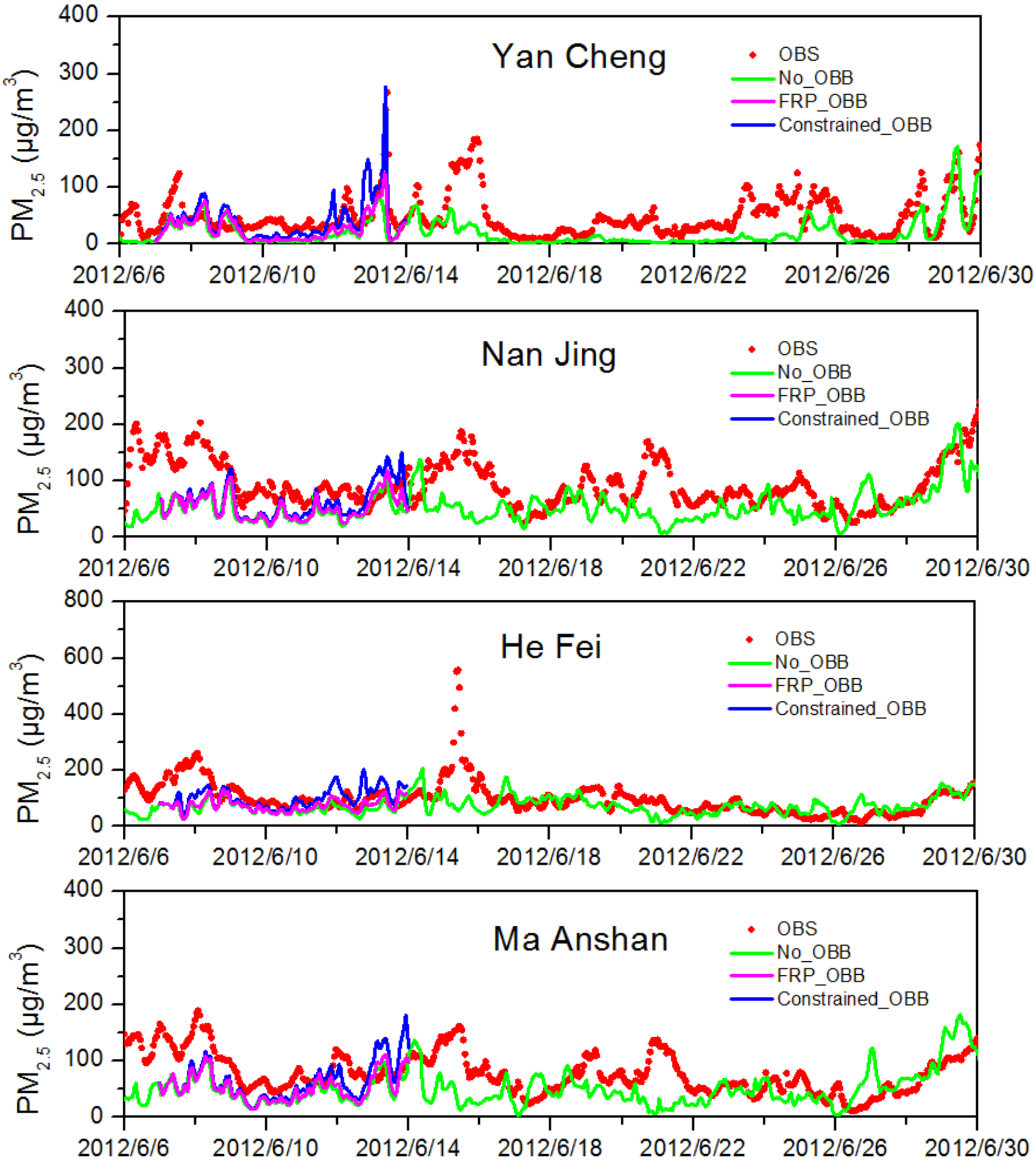


(a) (b)

Figure S10. Observed and simulated hourly $PM_{2.5}$ concentrations without OBB emissions (No_OBB) during June 2014 and with OBB emissions with FRP-based (FRP_OBB) and constraining (Constrained_OBB) methods in Xuzhou, Lianyungang, Huaian, Suqian, Yancheng, Nanjing, Hefei and Maanshan during June 7-13, 2014.



225

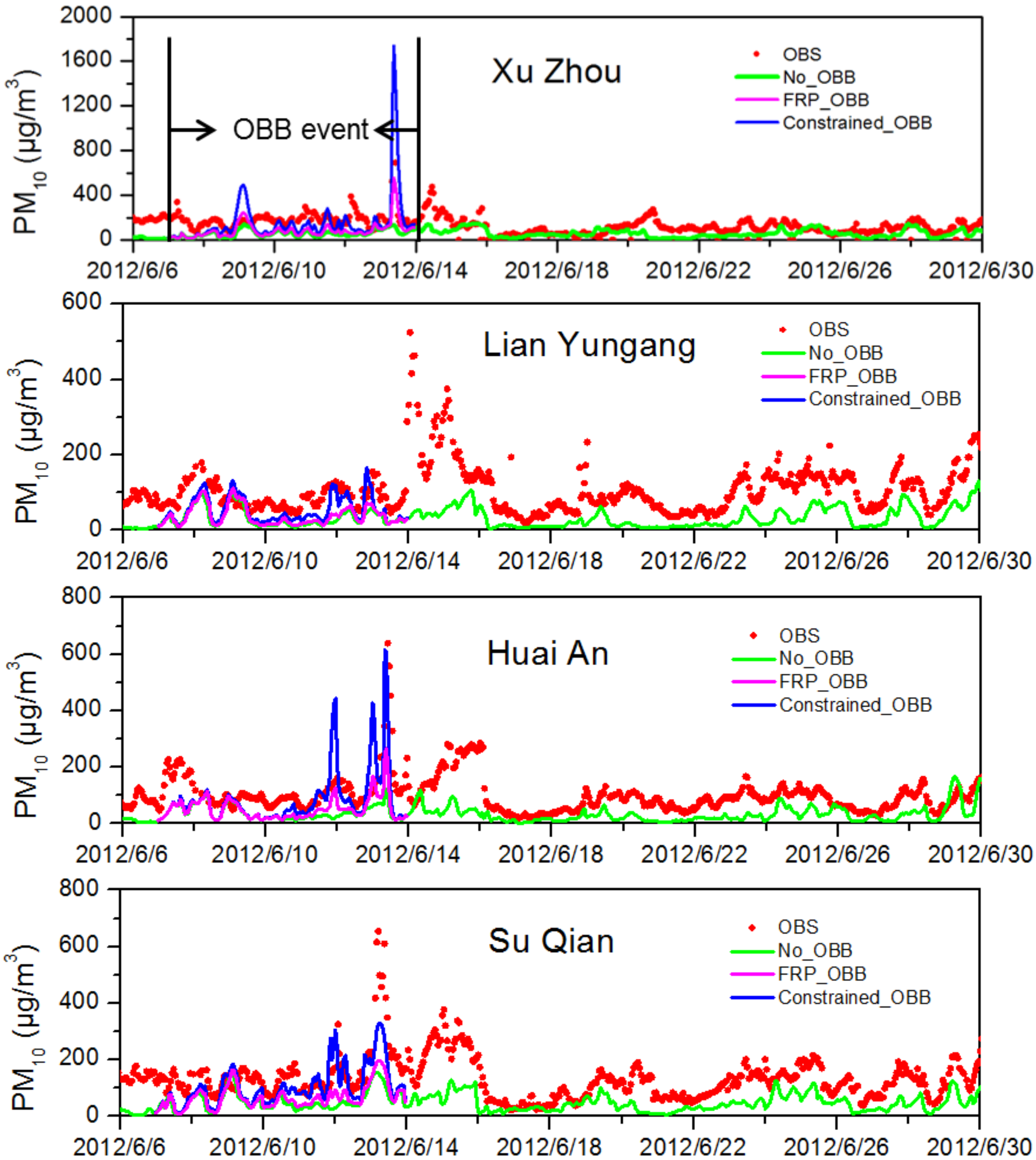


Yan Cheng
Nan Jing
He Fei
Ma Anshan
OBS
No_OBB
FRP_OBB
Constrained_OBB
PM2.5 (μg/m3)
2012/6/6
2012/6/10
2012/6/14
2012/6/18
2012/6/22
2012/6/26
2012/6/30

226

227

Figure S11. Observed and simulated hourly PM_{10} concentrations without OBB emissions (No_OBB) during June 2014 and with OBB emissions with FRP-based (FRP_OBB) and constraining (Constrained_OBB) methods in Xuzhou, Lianyungang, Huaian, Suqian, Yancheng, Nanjing, Hefei and Maanshan during June 7-13, 2014.



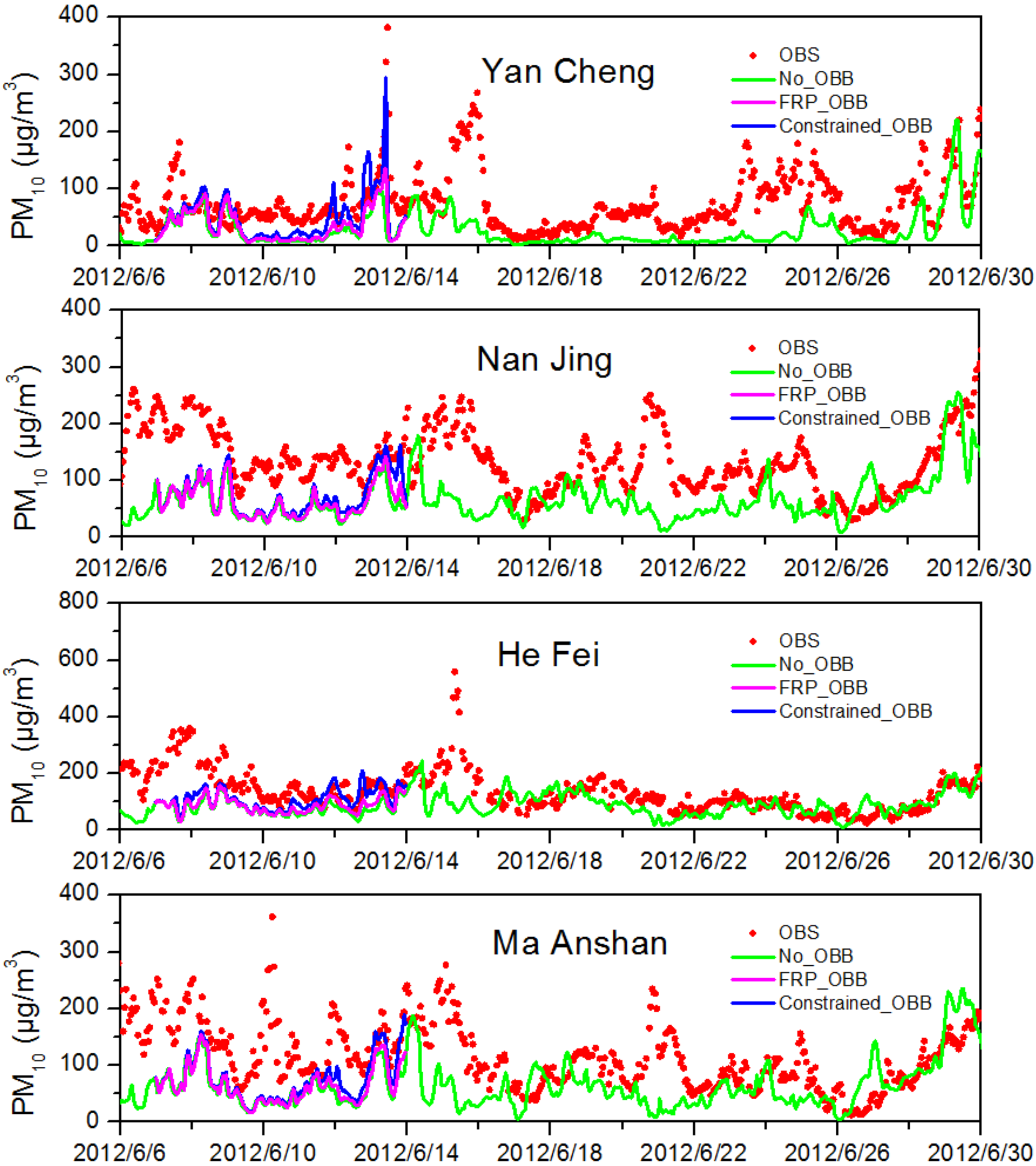
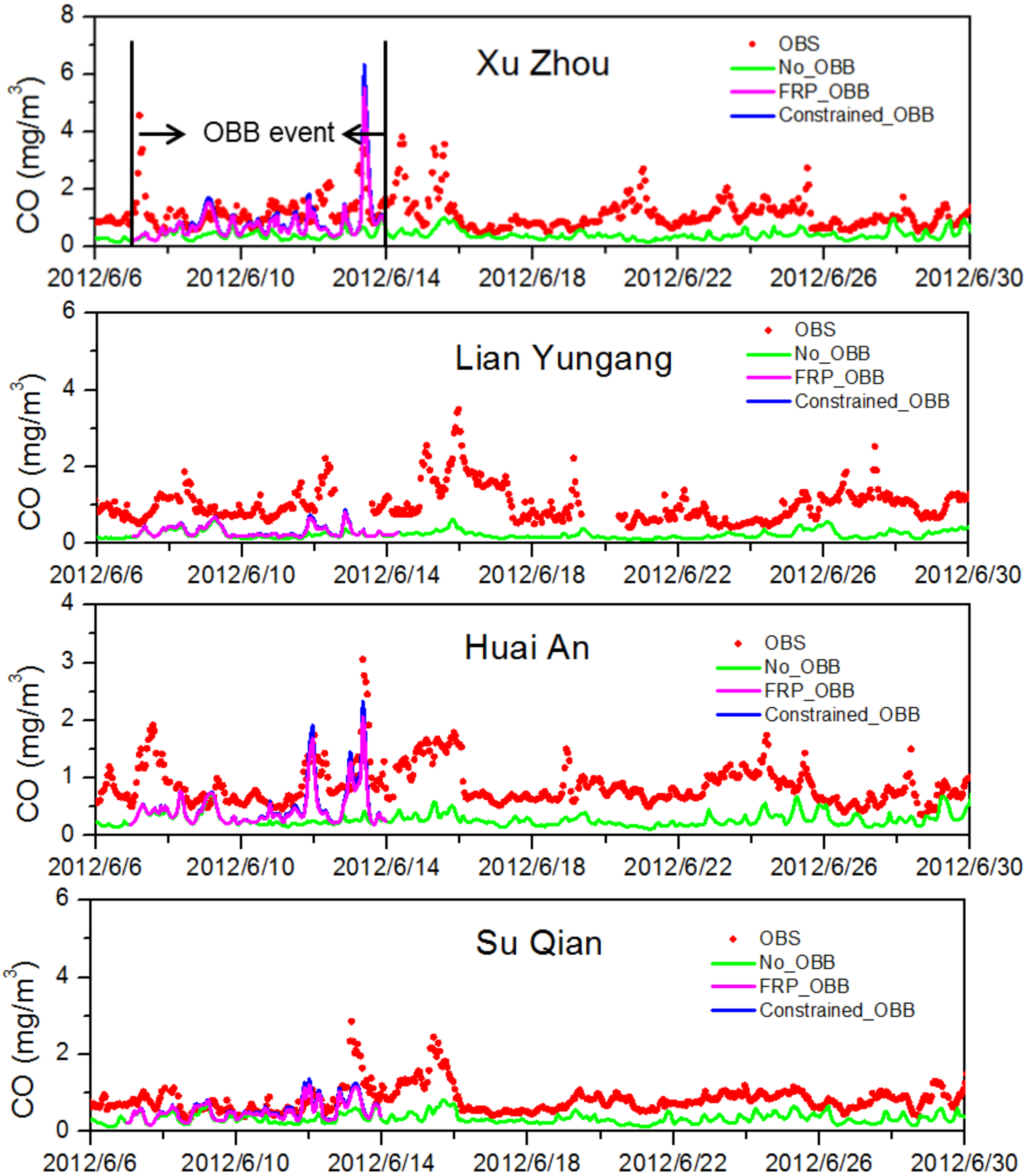
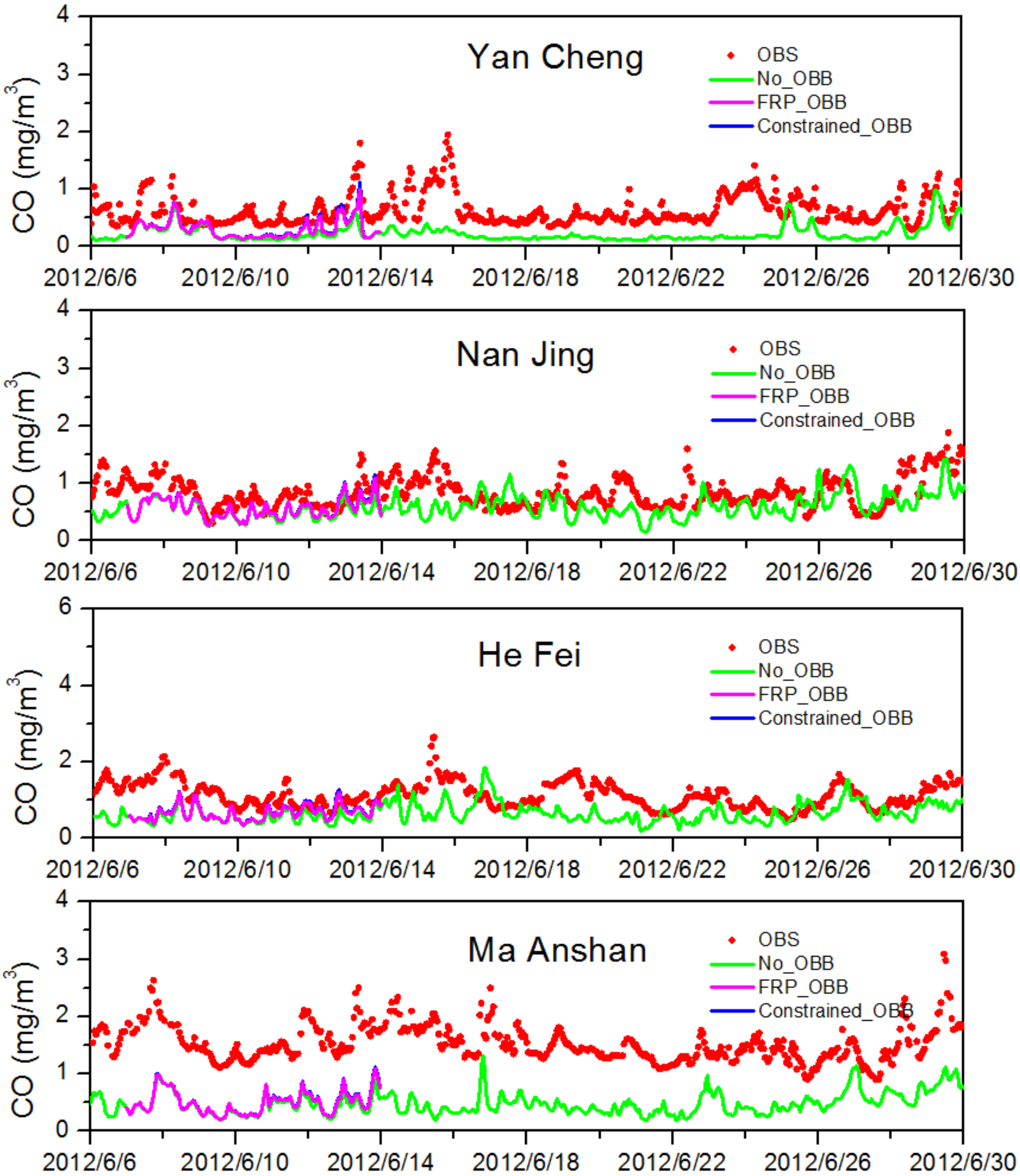

Yan Cheng
Nan Jing
He Fei
Ma Anshan
OBS
No_OBB
FRP_OBB
Constrained_OBB
PM10 (μg/m3)
2012/6/6
2012/6/10
2012/6/14
2012/6/18
2012/6/22
2012/6/26
2012/6/30

Figure S12. Observed and simulated hourly CO concentrations without OBB emissions (No_OBB) during June 2014 and with OBB emissions based on FRP-based (FRP_OBB) and constraining (Constrained_OBB) methods in Xuzhou, Lianyungang, Huaian, Suqian, Yancheng, Nanjing, Hefei and Maanshan during June 7-13, 2014.




Yan Cheng
Nan Jing
He Fei
Ma Anshan
OBS
No_OBB
FRP_OBB
Constrained_OBB
CO (mg/m3)
2012/6/6
2012/6/10
2012/6/14
2012/6/18
2012/6/22
2012/6/26
2012/6/30

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