Response to the comments of Anonymous Referee #3 on "Inverse modeling of black carbon emissions over China using ensemble data assimilation" by P. Wang et al.

We thank the Referee for the constructive feedback. We respond to each specific comment below. The original comments by the Referee are shown in bold italics. Our reply is shown in blue.

In this study, the assimilation technique is used to investigate the possibility of optimally recovering the spatially resolved emissions bias of black carbon (BC). An inverse modeling system for emissions is established for an atmospheric chemistry aerosol model and two key problems related to ensemble data assimilation in the top-down emissions estimation are discussed. In general, I found the paper appropriate for ACP audience. However, it need to be major revised before accepted this paper for publication in ACP with addressing those comments listed below:

Thanks for the comments!

- Major Comments and reply：

1.My primary criticism is that the authors spend too little time discussing the uncertainty in the results.

reply: Thank you! Yes, it' very important to evaluated the uncertainty of the result. Therefore we conducted the Monte Carlo simulation to quantify the uncertainty of the total bottom-up emission and the inversed emission inventory in China . The lognormal distribution was assumed, and the standard deviation was calculated by combining the root-mean-square error between observation and simulation with standard deviation of the inventory. Monte Carlo simulations with randomly selected values within the PDFs were repeatedly implemented for 10000 times. The uncertainty in Chinese BC bottom-up emission and inversed emission inventory at the 95% were obtained, as shown in Fig. 10. The mean value, 2.5th percentile value, and 97.5th percentile value were 1570, 321, and 5138 Gg (bottom-up) and 2650, 1114, 5471 Gg (inversed emission), respectively. Therefore, the uncertainty of these two emission inventory were about [-80%, 227%], and [-58, 102%], correspondingly. Using the ensemble inversion modeling, the uncertainty of BC emission inventory decreased by 50%. We also compare our estimation with results from previous study. Streets et al (2003) estimated the 1.05Tg BC emission in China for the year 2000 with ±360% uncertainty measured as 95% confidence intervals. Zhang et al (2009) estimation of the China BC emission is 1.61Tg. Qin and Xie (2012) estimated the 1.57Tg BC emission in China for the year 2005 with [-51%, 148] uncertainty. Our estimation are nearly 40% higher than these bottom-up inventories. One reason is there was very little emissions for the northwest China and northeast China in all of these emission inventory. They are so similar low in these regions probably because these bottom-up inventories are based on the same statics data source. Based on top-down regression method, Fu et al (2012) estimated the annual BC emission is 3.05±0.78Tg which is higher than our estimation. One possible reason is that their estimation may be biased high in central China which had been pointed out in their paper.

2. Author need to conduct at least more than 12 months simulation for comparison and validation. One month is not reliable to simulate the black carbon emissions over China.

reply: Thank you for the advice, we have conducted one year simulation for comparison. Fig.5e compares the seasonal variation of observed and simulated surface BC. The simulated concentrations were significantly lower than the observed throughout the year. This indicates a region wide underestimate in monthly and annual bottom-up emission inventory. BC observations were higher in Winter than summer, suggesting strong emission associated heating. Fig. 5a and 5b compared the spatial distribution of monthly mean observed and simulated BC concentrations for January and July. The model showed higher BC concentrations in January than in July, which is similar to the observation. The simulated BC concentration had much higher values in east than west with highest concentration over northern China, corresponding to the strong emission there. However, the model simulations not only underestimated BC concentration at urban sites but also significantly underestimated at rural sites such as TaZhong, Hami, Dunhong, Gaolanshan in northwest China where the bottom-up emission have very little emissions both for January and July. Fig.7 shows the daily BC concentrations variation in January at background, rural and urban sites. At background site AkeDala and WLG, the model still produces the relatively low concentration indicating underestimation of BC emission in northwest China. Little bias presented in simulation at background site ZhuZhang showed that emission in Yunnan province is relatively accurate. The simulated BC concentration at background site SD was a little higher than the observation. That is because SD located in northwest China where is the densely populated and industrialized area, and the emission rate around the SD were relatively high. Besides these four background sites, the model performance of daily BC concentration based on bottom-up inventory at rural and urban was very poor largely because the underestimation in emission. This suggests that the bottom-up emission was very low and misrepresented in space and time.

3. The purpose of this manuscript is unclear. When first quickly look through the title, I think this paper will discuss about the black carbon emissions over China, which is compared with the previous studies. After I read it very carefully, the theme just discuss the comparison of the two simulation methods with the observations.

reply: The purpose of this manuscript is to evaluate the current bottom-up emission inventory and use ensemble data assimilation for inversion to reduce large bias in the inventory. After we introduced the methodology of inversion using ensemble data assimilation and model and measurements employed in this study, we discussed two problems related to ensemble data assimilation in the top-down emission estimation, then evaluated the bottom-up emission inventory by comparing model simulations with observation. Then used monthly mean observation to do the inversion, and evaluated the top-down estimation and its uncertainty. Comparison with previous estimation of BC inventory was also presented. The main conclusion from our study is that the bottom-up BC inventory has large bias and can be reduced by the ensemble data assimilation.

4. What I concerned is about the author illustrated that the inversed emission over China in January is over 1.8 times of bottom-up emission inventory. But how much for the other months and seasons. Is it possible that the bottom-up emission inventory is larger than inverse model ?

reply: We employed an ensemble optimal interpolation to inverse 12 months bottom-up emission inventory. Fig. 11 presented the bottom-up and inversed BC emission inventory . The emissions in every month had been enhanced after the inversion.

Minor comments:

1. Line 2 in page: Author need to provide the reference for why " BC aerosols have been shown to act as cloud condensation nuclei when they become hydrophilic, affecting cloud micro- physical properties and rainfall processes."

reply: We added two references in the manuscript for this matter.

Lary D. J., Lee A. M., Toumi R., Newchurch M. J., Pirre M., and Renard J. B.: Carbon aerosols and atmospheric photochemistry, J. Geophys. Res., 102, 3671-3682, 1997

Bond T. C.,S. J. Doherty, D. W. Fahey,P. M. Forster,T. Berntsen,B. J. DeAngelo, M. G. Flanner,S. Ghan, B. Kärcher,D. Koch,S. Kinne, Y. Kondo,P. K. Quinn,M. C. Sarofim, M. G. Schultz,M. Schulz,C. Venkataraman,H. Zhang,S. Zhang,N. Bellouin,S. K. Guttikunda,P. K. Hopke,M. Z. Jacobson,J. W. Kaiser,Z. Klimont,U. Lohmann,J. P. Schwarz,D. Shindell, T. Storelvmo,S. G. Warren,and C. S. Zender : Bounding the role of black carbon in the climate system: A scientific assessment, J. Geophys. Res., 118, 5380–5552,2013

2. Line 26 in page 11: Authors need to provide some discussions about model fails to capture the spatiotemporal variability in the BC observations and underestimates the BC concentrations at almost all assimilated sites except site SD.

reply: Please see the reply for Major comments 2.

3. Line 6 in page 13: Author should give the reason why the Most of them feature a positive bias.

reply: It 's typo, should be low bias or negative bias, thank you.

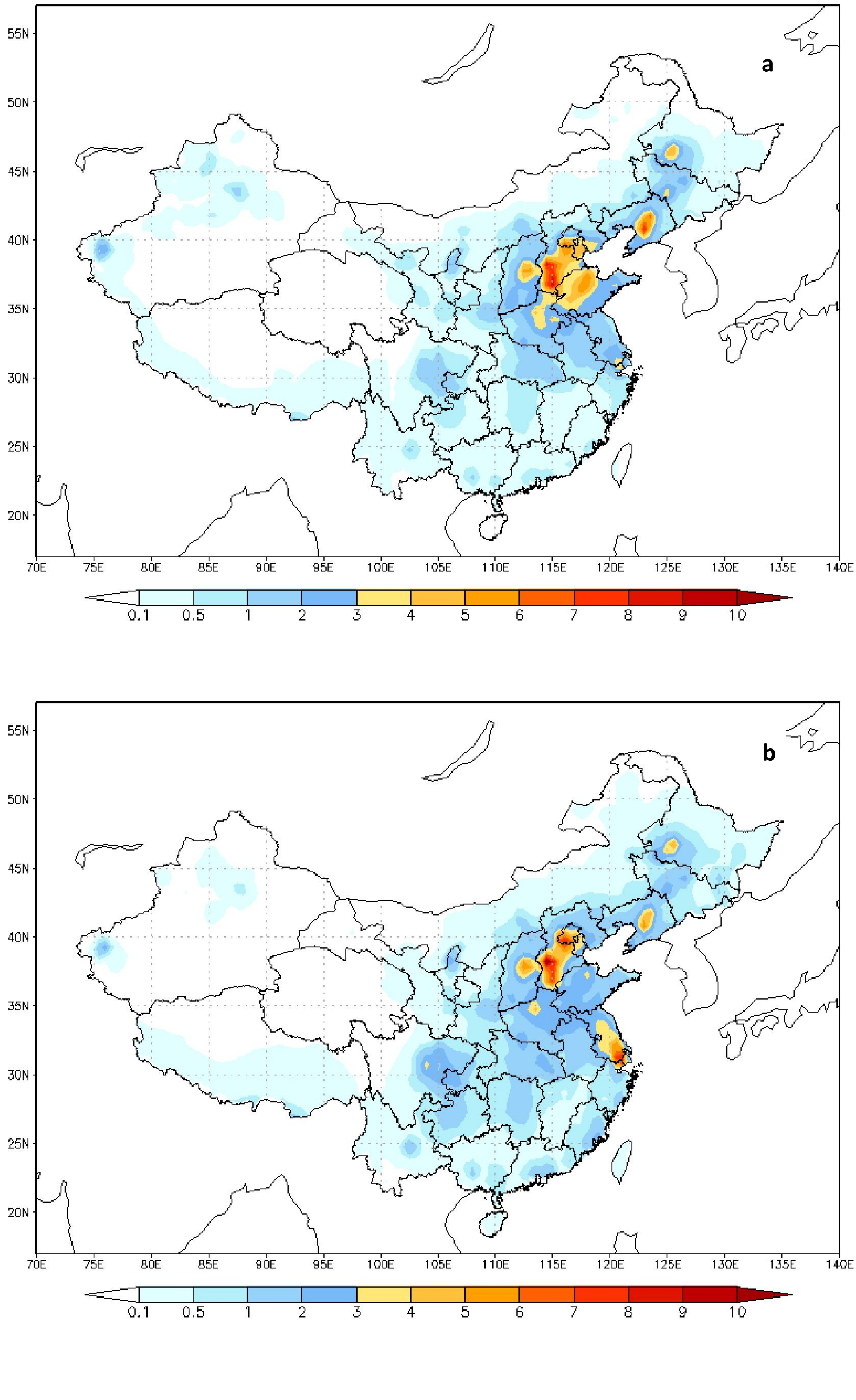
4. Line 10 in page 13: Figure7 is too small to see.

reply: Modified and enlarged as suggested.

5. Line 14 in page 13: Authors need to provide some discussions about why "Even though we only employ the monthly mean BC measurements to inverse the emissions, the accuracy of the daily model simulation is also improved."

reply: Thank you, we added some discussion about this.

Because there were large region wide underestimation in the bottom-up emission, not only in the densely populated and industrialized areas such as northern China, the Yangtze River Delta and the Sichuan basin, but also in northwest China which have lower population densities and lower economic level, the model performance of daily BC concentration was very poor. The simulation at rural and urban sites were significantly lower than the observations. With inversion by EnOI, the emission low bias had been corrected, the simulated concentration were increased and improved. The average RMSE reduced from 5.08 to 3.47. However, there were still large difference between the daily observations and simulation, because there are some other source of uncertainties such as meteorology and other factors of model error. We had used monthly mean data in the inversion process to reduce these effects, but when come to hourly and daily simulation, these effects should be considered reasonably which is the future work we plan to work on.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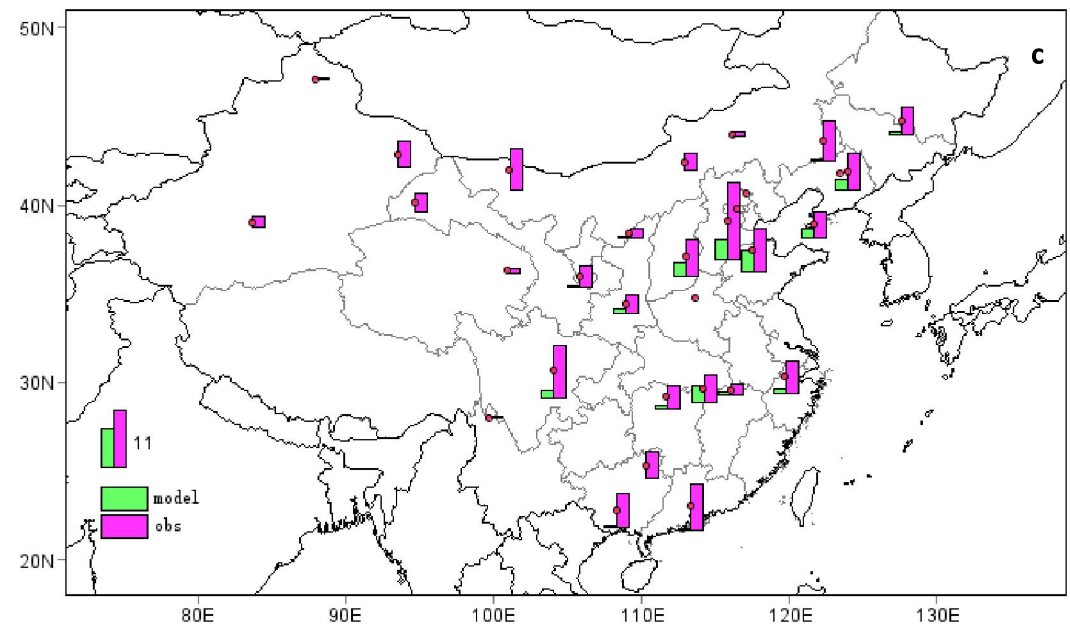
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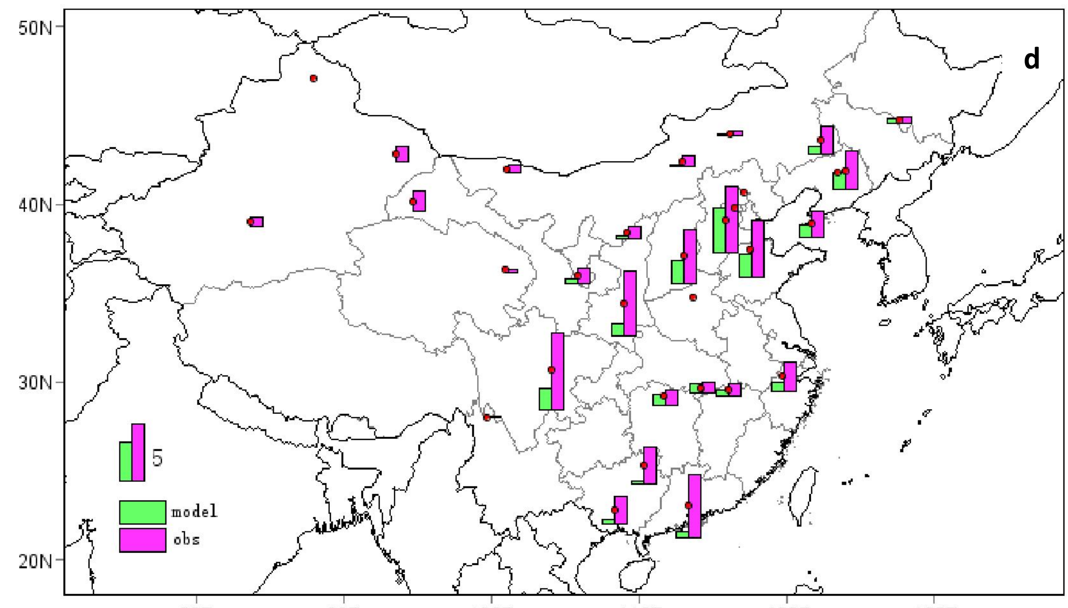
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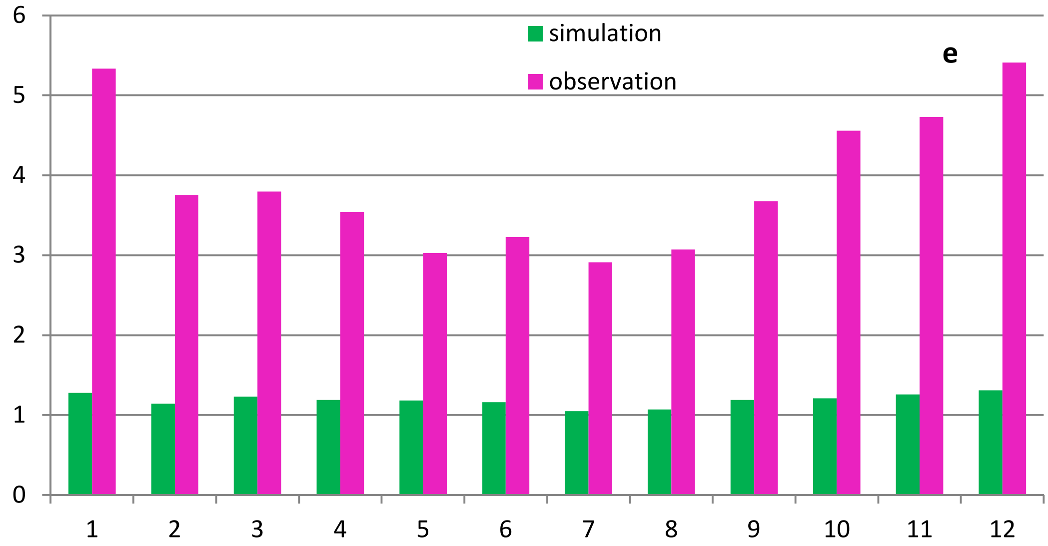

Fig. 5 BC monthly mean concentrations (units: μg/m^3). (a) model simulation with bottom-up emission in January ; (b) model simulation with bottom-up emission in July; (c) comparison between model simulation and observation in January; (d) comparison between model simulation and observation in July, Green bars show the model-simulated BC using the bottom-up emissions inventory; pink bars show the observed surface BC concentrations; (e) Seasonality of BC concentration

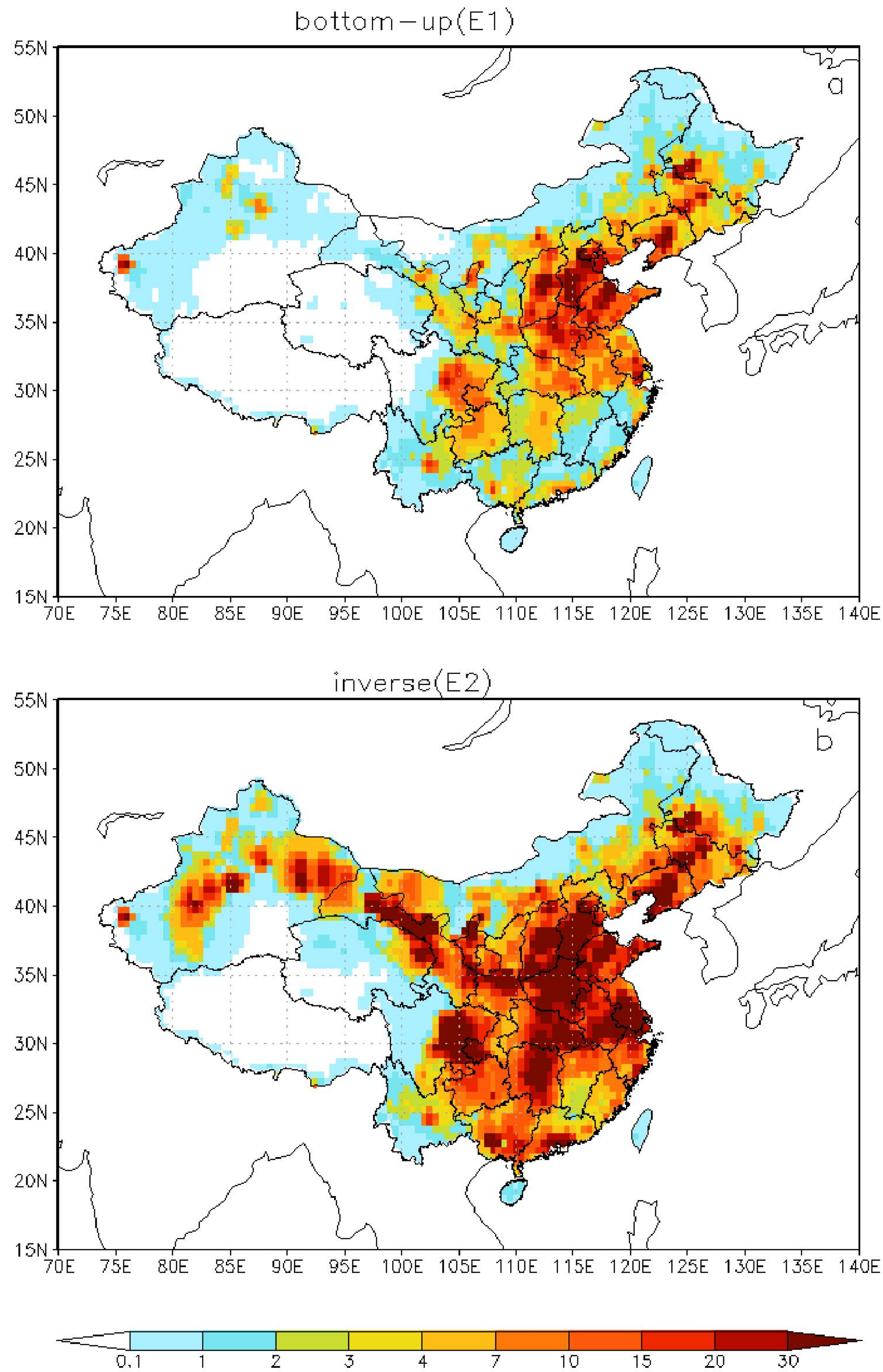

Fig. 6 BC emissions from China inversed by 27 observation sites (units: $\mu g/s\cdot m^2$): (a) bottom-up emissions, E1; (b) inversed emissions, E2.


AK
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XG
SD
E1
E2
obs
TZ
HMi
DH
GLS
E1
E2
obs
TL
GC
ZZ
XA
E1
E2
obs

Fig. 7 BC daily concentrations in January 2008 (units: μg/m^3). The red dotted line shows the observation; the blue line is the model simulation driving by prior emissions (E1); and the green line is the model simulation driven by inversed emissions (E2).

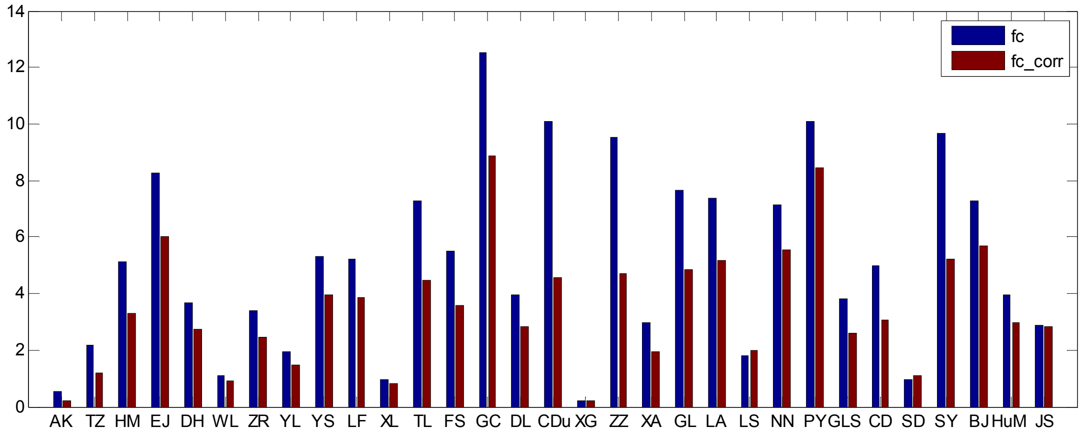

Fig. 8 The root-mean-square error (RMSE) between the daily model simulation and observation. The blue bars show the RMSE between the daily model simulation driven by prior emissions (E1) and observations, and the red bars show the RMSE between the daily model simulation driven by inversed emissions (E2) and observations. (units: μg/m^3)

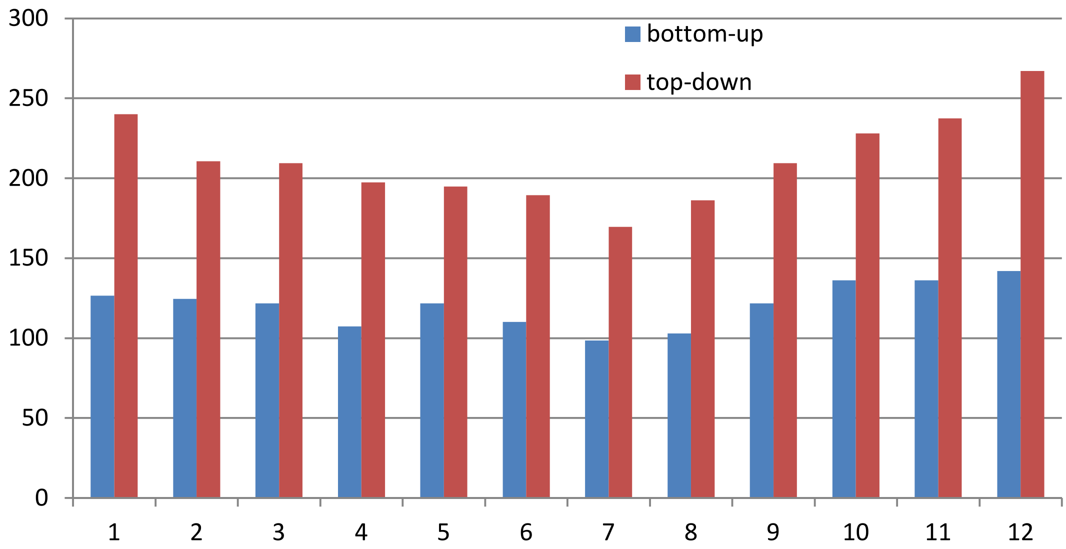

Fig. 9 Seasonality of BC emission in China

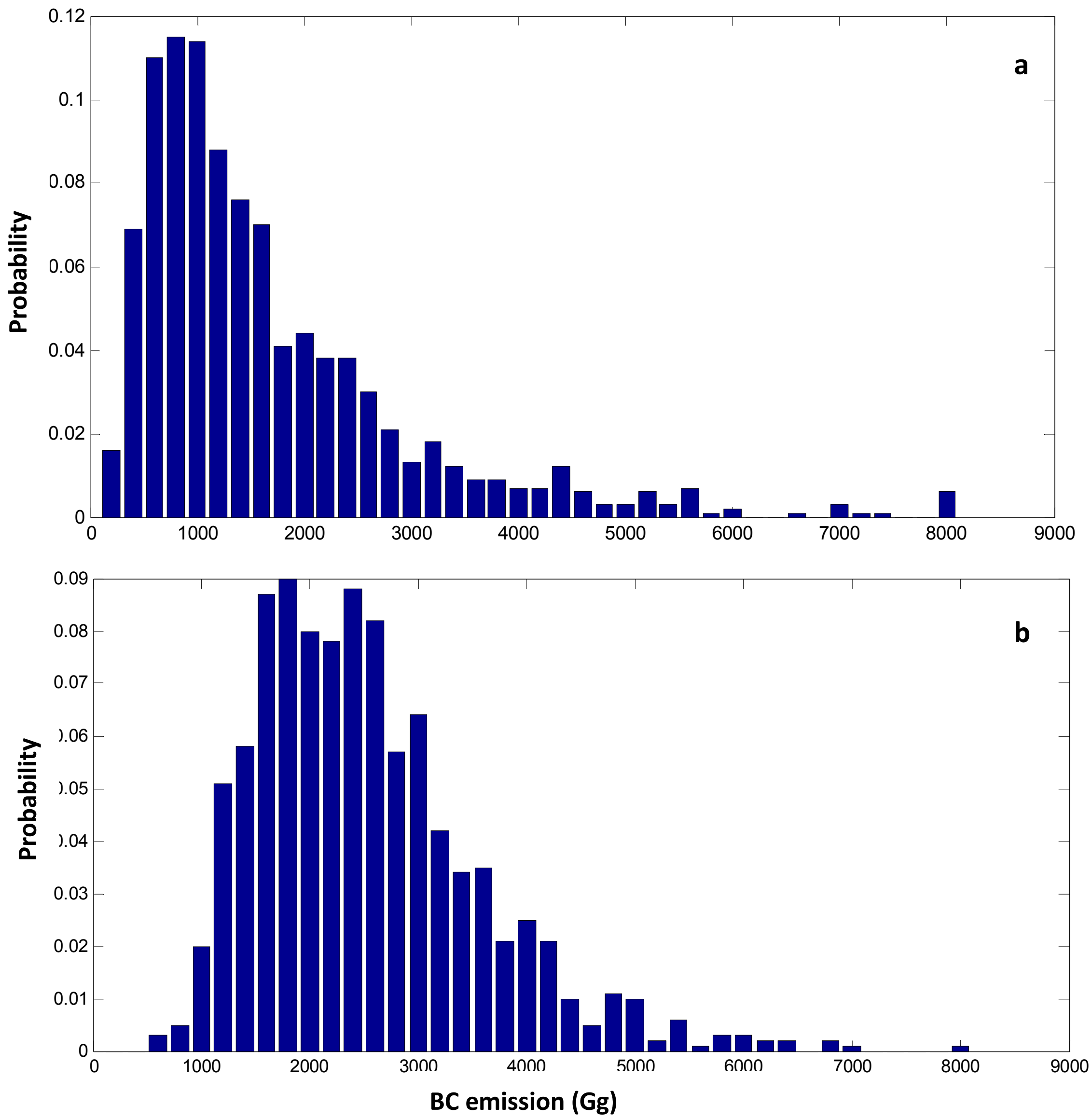

Fig. 10 Uncertainty analysis for annual Chinese BC bottom-up and inversed emission inventory

Table 1 Observation site information

num	observation sites	LON	LAT	ALT	description
1	AKeDaLa (AK)	87.97	47.12	562	background site
2	TaZhong (TZ)	83.67	39	1099.3	rural site
3	HaMi (HM)	93.52	42.82	737.2	rural site
4	EJiNaQi (EJ)	101.07	41.95	940.5	urban site
5	DunHuang (DH)	94.68	40.15	1140	rural site
6	WaLiGan (WL)	100.92	36.28	3816	background site
7	ZhuRiHe (ZR)	112.9	42.4	1151.9	rural site
8	YuLin (YL)	109.2	38.43	1105	urban site
9	YuShe (YS)	112.98	37.07	1041.4	urban site
10	LongFengShan (LF)	127.6	44.73	330.5	rural site
11	XiLinHaoTe (XL)	116.12	43.95	1003	rural site
12	TongLiao (TL)	122.27	43.6	178.7	urban site
13	FuShun (FS)	123.95	41.88	163	urban site
14	GuCheng (GC)	115.8	39.13	11	urban site
15	DaLian (DL)	121.63	38.9	91.5	urban site
16	ChengDu (CDu)	104.04	30.65	553	urban site
17	ZhuZhang (XG)	99.73	28.02	3580	background site
18	ZhengZhou (ZZ)	113.68	34.78	110	urban site
19	XiAn (XA)	108.97	34.43	410	urban site
20	GuiLin (GL)	110.3	25.32	164.4	rural site
21	LinAN (LA)	119.73	30.3	138.6	rural site
22	LuShan (LS)	115.99	29.57	1165	rural site
23	NanNing (NN)	108.35	22.82	172	urban site
24	PanYu (PY)	113.35	23	131	urban site
25	GaoLanShan (GLs)	105.85	36	2161.5	rural site
26	ChangDe (CD)	111.71	29.17	565	rural site
27	ShangDianZi (SD)	117.12	40.65	293.3	background site
28	ShenYang(SY)	123.41	41.76	110	urban site
29	Beijing (BJ)	116.47	39.8	31.3	urban site
30	HuiMin (HM)	117.53	37.48	11.7	urban site
31	JinSha (JS)	114.2	29.63	330.5	rural site

Table 3 Model simulations and surface observations of monthly mean BC concentrations at assimilation sites and verification sites (units: μg/m^3) and the relative error percentage (=(|model − obs| / obs) × 100%).

site	Model (E1)	Model (E2)	observation	observation std	Relative error percentage (E1)	Relative error percentage (E2)
AK	0.07	0.44	0.51	0.34	86.9	13.0
TZ	0.04	1.08	2.20	0.79	98.4	51.1
HMi	0.06	2.21	4.90	3.15	98.9	54.9
EJ	0.05	2.67	7.84	4.03	99.4	66.0
DH	0.06	1.02	3.55	1.93	98.4	71.4
WL	0.13	1.03	0.94	0.61	85.7	9.3
ZR	0.14	1.00	3.37	1.29	95.7	70.2
YL	0.31	0.89	1.88	1.60	83.6	52.9
YS	2.70	5.56	6.94	3.61	61.1	19.9
LF	0.58	2.23	5.16	3.81	88.8	56.8
XL	0.14	0.37	0.93	0.76	84.7	59.8
TL	0.47	2.97	7.42	3.05	93.6	59.9
FS	2.00	4.82	7.06	4.09	71.7	31.6
GC	3.79	7.60	14.24	8.06	73.4	46.7
DL	1.74	4.13	4.85	2.21	64.1	14.8
CDu	1.45	7.14	9.71	5.21	85.0	26.5
XG	0.20	0.21	0.30	0.18	34.8	29.2
ZZ	3.28	10.68	10.89	4.41	69.9	2.0
XA	1.02	3.57	3.66	1.73	72.1	2.5
GLs	0.26	1.92	4.99	2.75	94.8	61.5
LA	1.00	4.19	6.19	2.88	83.8	32.3
LS	0.73	2.69	2.08	1.12	65.0	29.4
NN	0.55	2.26	6.36	3.32	91.3	64.5
PY	0.55	2.15	8.69	4.64	93.6	75.2
GL	0.46	1.82	4.11	2.13	88.8	55.8
CD	0.71	3.02	4.38	1.86	83.7	31.0
SD	1.39	1.38	0.81	0.73	71.8	70.5
BJ	3.45	8.68	11.96	5.57	71.2	27.4
HM	4.27	6.41	8.06	4.96	47.0	20.4
JS	3.20	4.47	5.35	2.61	40.1	16.4
SY	0.89	3.14	3.05	1.69	70.7	2.8
Assi_sites mean	0.88	2.93	4.96	2.60	82.2	42.9
Veri_sites mean	2.95	5.67	7.10	3.71	57.2	16.8
All_sites mean	1.15	3.28	5.24	2.75	79.0	39.5