Response to the comments of Anonymous Referee #1 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.

General comments:This paper uses an ensemble optimal interpolation (EnOI) data assimilation technique to reduce emission bias of black carbon (BC) in China. The bottom-up emission inventories in China are associated with large uncertainties. The authors demonstrated that using the EnOI approach can considerably bring the model prediction closer to observed BC concentration. The manuscript is well organized and the results are clearly presented. The findings presented here provide a reliable alternative to predict BC variations in China in the absence of accurate emission inventories. Hence, I recommend publishing this manuscript after the following comments being satisfactorily addressed.

Thanks for the comments!

Specific comments:

1. While inverse modeling can provide a simplified solution, the processes contributing to the model bias go beyond emission. Therefore, emission inversing is likely to lump up uncertainties from other processes into emission.

reply: Yes, this is quite right, so we added this comment and uncertainty analysis of inverse estimation in our manuscript. 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 compared 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. There are a number of formulas given but not all variables are explicitly denoted. Suggest a throughout checking of the manuscript on this matter. For instance, Eq. 10 and 12.

reply: We have done a throughout checking of the manuscript to make sure that all variables in the formulas are explicitly denoted. Thank you very much.

3. P20858, L22-24, how was the subset of assimilation or verification sites chosen from CAWNET?

reply: We chose the verification sites because 1) they had less missing value, 2) at least one more site not far away from them so there would be a inversion increment.

4. P20858, L26: "city's average elevation" is confusing. May choose another term,

such as above ground level.

reply: Thank you, modified as suggested.

5. P20859, L3: [at] a 5min time [interval].

reply: Fixed as suggested.

6.L5: [the] optical absorption.

reply: Fixed as suggested.

7. L13-14: How representative is the monthly mean to the actual daily and hourly variability? It may be useful to provide some measures, such as standard deviation, from at least observations to gauge the robustness of this choice.

reply: Yes, we have calculated the standard deviation of daily observations and used them to represent the error of the measurements in the inversion process. We also presented them in the Table 3.

8. P20860, L12: [than] Scheme A. I wonder how the RMSE from Scheme B compared to that from A.

reply: the RMSE of Scheme A is 2.10 and Scheme B is 2.58 . This also indicated that Scheme B is better than Scheme A because good ensemble should not only include the observation but also have large spread to contain large possibility.

9. P20861-P20863: The observed BC concentrations are five-fold of the model prediction (5.2 vs 1.1 ug/m3). An increase of emission by 1.8 times will reduce the model bias by 50%. Why is that?

reply: The increase of emission by over 1.8 times refers to increment at the nationwide scale. Yet, the model bias reduced by 50% is calculate by the BC concentration of observation site shown in table 3 where emission rate were significantly corrected by EnOI. However, there was a typo in Table 3, observation and model E2 were labeled opposite, and had been corrected. Thank you!

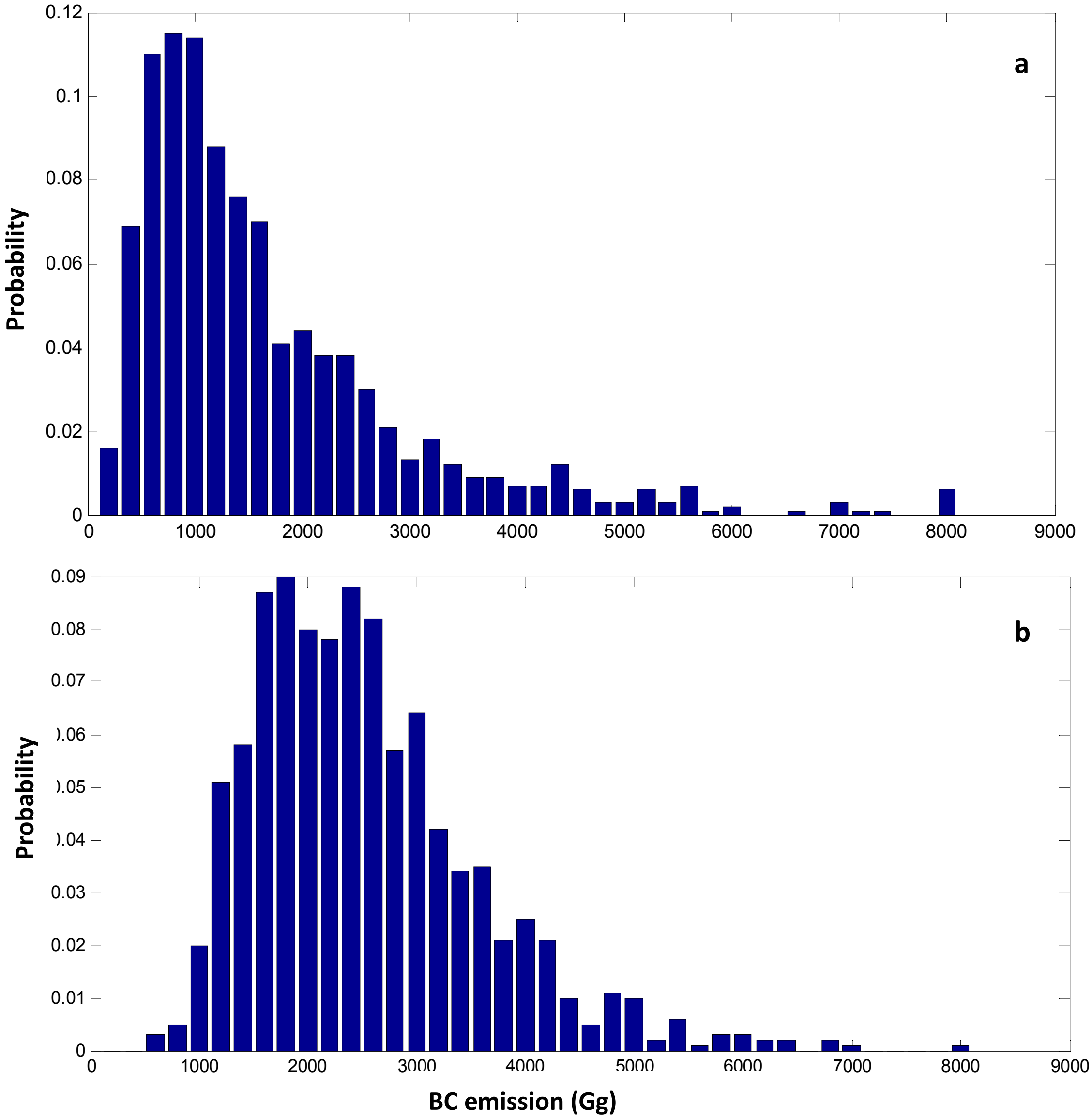

Fig. 10 Uncertainty analysis for annual Chinese BC bottom-up and inversed emission inventory

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