Response to the comments of Anonymous Referee #2 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.

This study employs an ensemble data assimilation technique to reduce the bias in the black carbon (BC) emission inventory over China. From the point view of methodology, using the ensemble optimal interpolation (EnOI) for emission inversion with low computational cost is an interesting attempt. Significant reduction of bias in BC modeling are obtained after assimilating the surface BC observations, which would be valuable for better knowledge of the bottom-up BC emission inventory over China. Nevertheless, there are several issues need to be addressed before publishing in ACP.

Thanks for the comments!

Major Comments and reply

1. Because the background error covariance in chemical transport modeling can vary very quickly. In this study, static background error covariance is used in ensemble optical interpolation (EnOI) to do inverse estimation. How to deal with the rapid variation of background error covariance using the current method?

We used monthly mean meteorological field to run the model with perturbed emissions and got background error covariance.

2. Meteorological simulations and its uncertainty are key impact factor for inverse modeling. This paper seems not pay much attention to this issue. Is the simulated meteorological well consistent with the observations?

We employed NCEP 1×1° Reanalysis data for the model's initial and 6h meteorological lateral boundary input fields and the forecast time was every 24h throughout the month. This ensure the monthly mean meteorological factor were consistent with

NCEP reanalysis data. Besides, we had compared the model's daily averaged wind speeds with the NCEP analysis for 1–31 July and found the modeled daily wind speed agrees well with the NCEP wind speed, showing the model's fair ability to simulation of meteorological field (Wang 2015). Moreover, in this study, the bottom-up inventory had been found to have large bias and we only use monthly mean BC concentration observation to inversed the inventory. Therefore, we didn't pay much attention to meteorological simulations. However, it is a very important factor when we focus in daily and hourly simulation which will be studied in our future work. Thank you.

Wang H., Xue M., Zhang X. Y., Liu H. L., Zhou C. H., Tan S.C., Che H. Z., Chen B., and Li T.: Mesoscale modeling study of the interactions between aerosols and PBL meteorology during a haze episode in Jing–Jin–Ji (China) and its nearby surrounding region – Part 1: Aerosol distributions and meteorological features, Atmos. Chem. Phys., 15, 3257–3275, doi:10.5194/acp-15-3257-2015, 2015

3. BC measurements could contain errors and affect the results of inverse estimation. This issue should be considered in their experiments.

The error of BC measurements had been considered by their standard deviation in the inversion process as observation error in equation (2) and added in Table 3.

4. The uncertainty related to the inverse estimation should be investigated and well discussed.

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 Figure of uncertainty. 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.

Minor Comments and reply:

1. Figure 7 is not clear and too many graphs are included.

Modified and enlarged as suggested.

2. The methodology description is too simple and not well constructed.

We added some description of ensemble strategy and reconstructed the manuscript. Thank you!

3.There are some grammatical errors in the manuscript. Help from an English editor is recommended.

The manuscript has been edited by a native English speaker following the suggestion.


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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).

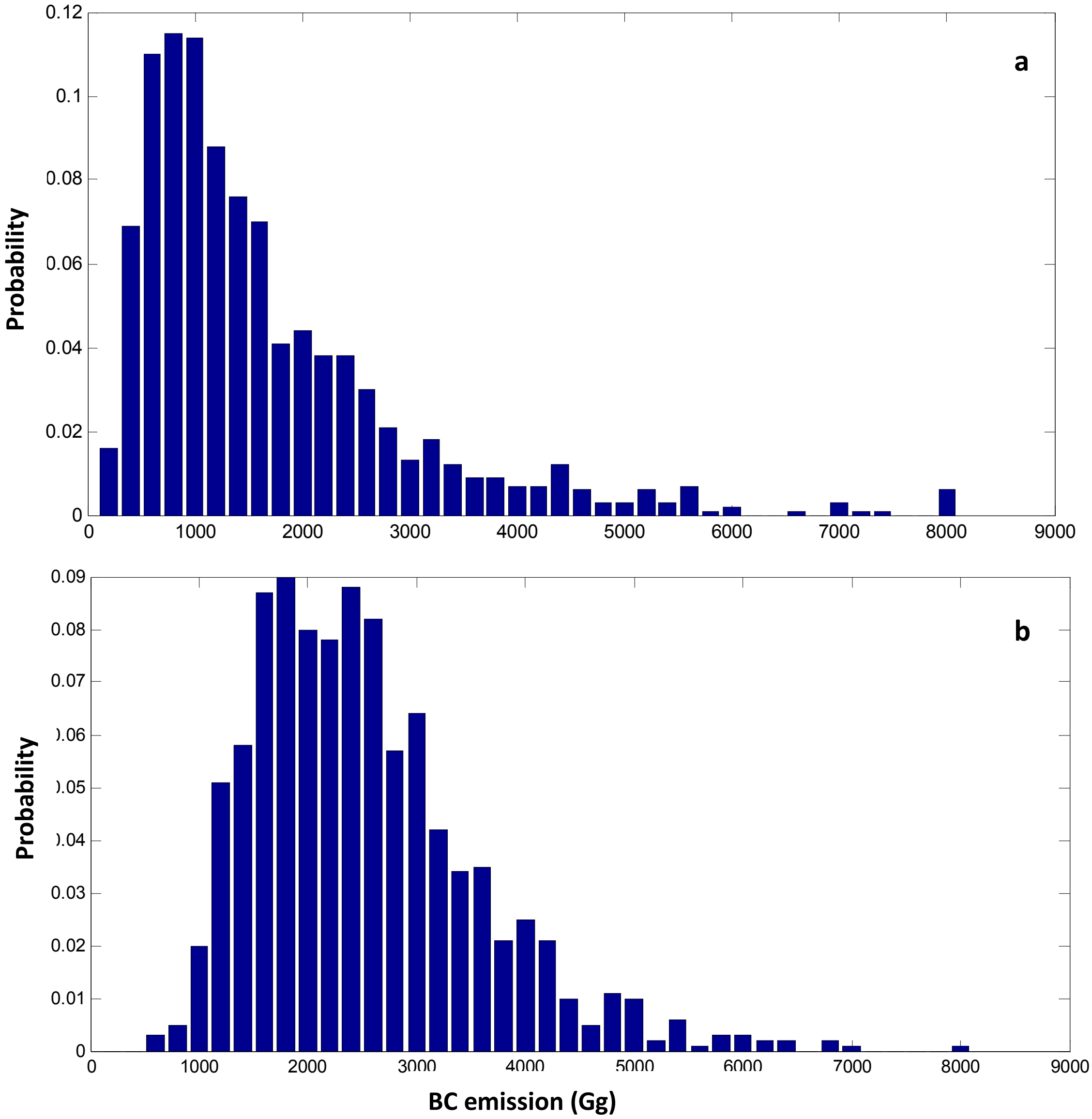

Fig. 10 Uncertainty analysis for annual Chinese BC bottom-up and inversed emission inventory

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