Response to reviewers' comments and main revisions

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Title: Long-term Variability in Black Carbon Emissions Constrained by Gap-filled Absorption Aerosol Optical Depth and Associated Premature Mortality in China

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We thank very much for the valuable comments and suggestions from the reviewers, which help us improve our manuscript. The comments have been carefully considered and revisions have been made in response to suggestions. Following are our point-by-point responses to the comments and corresponding revisions. **Please note that the line/table/figure numbers mentioned following refer to the clean version of the revised manuscript, unless specifically noted.**

Comments from Reviewer #1

General comment: Using a machine learning technique and a "top-down" inversion approach, with remote sensing observations and meteorological reanalysis data as input, the authors analyzed the evolution of black carbon emissions in China from 2000 to 2020. Moreover, using an attributional model, the authors related premature mortality and black carbon exposure, and investigated the mortality due to black carbon exposure and its drivers. In addition, the authors discussed the uncertainties in the calculation of black carbon AAOD and health impact estimation. The manuscript is well structured and written, the methodology is well established, the results are well presented and discussed. Due to the issues listed below, I suggest a major revision before it is suitable for publication.

Response and main revisions:

We appreciate the reviewer's positive comments on our paper, and have made point-by-point response and revisions as summarized below.

Q1. Major issues: In the introduction, there is no review of studies on BC-associated premature mortality, especially over China. The motivation behind the authors' investigation into BC-associated premature motility and the research status of this premature motility is not clear. As the associated premature mortality is listed in the title and it is supposed to be one of the important parts of the paper.

Response and main revisions:

We appreciate and agree with the reviewer's valuable comment. The review of studies on BC-associated premature mortality in China and the motivation of this study are summarized below.

Based on the "bottom-up" emission estimates with great uncertainty and CTMs, previous studies have evaluated the BC-associated premature mortality in China for limited years (2000, 2013, and 2016, Cui et al., 2022; Qin et al., 2019; Saikawa et al., 2009; Wang et al., 2021). Large discrepancy exists in the magnitude (50,100-1,436,960 cases) and few analyses are available on the long-term spatiotemporal variations and driving forces of BC-associated health effects. The influence of human activities on quickly changing BC emissions and their associated health impact is inadequately or inaccurately understood, weakening science-based decision making for air pollution control.

We have added and reorganized the information in **lines 95-100 in the revised manuscript**.

Q2. Major issues: In section 3.3, the results of premature mortality associated with BC exposure are only based on the estimation, without any validations, which makes the analysis less convincing. Is it possible to collect some data,

either released in government reports or published in papers, to support your estimations? It does not need to be very precise, the magnitudes of the same order are sufficient.

Response and main revisions:

We appreciate the reviewer's valuable comment. We have collected the all-cause premature deaths attributed to BC in China from limited published papers to validate our estimates. The all-cause premature deaths attributed to BC in China were reported as 50,100 cases in 2000 (Qin et al., 2019), 1,436,960 cases in 2013 (Wang et al., 2021) and 538, 400 cases in 2017 (Cui et al., 2022). All-cause premature deaths attributed to BC in China in this work were estimated as 733,910-937,990 in 2000-2020, within the wide range of 50,100-1,436,960 cases by previous studies.

The health impact estimation could be biased by rare domestic β_{BC} values in China. Previous studies commonly adopted the same β_{BC} with $PM_{2.5}$ (Qin et al., 2019) or β_{BC} obtained from experiments conducted in Europe or the United States (Wang et al., 2021), resulting in large uncertainty. In this study, we relied on a unique cohort study in China and calculated the all-cause premature deaths attributed to BC at 733,910-937,980/yr. The β_{BC} values obtained from national-scale studies in the US and Europe indicate a 10-fold difference (220,980-2,386,060/yr, Supplementary Table S14), similar to the estimation conducted in the US (Li et al., 2016). More domestic epidemiological studies focusing on BC emissions are expected to further reduce the uncertainty.

We have added the validations and uncertainty discussions in **lines 533-536 and lines 610-618 in the revised manuscript**.

Q3. Specific points: L137: The Chinese Academy of Sciences is a huge organization. A detailed name where the data is obtained is needed. The website of the dataset rather than the website of the data center should be provided. If the dataset is associated with a published paper, then the paper should be cited.

Response and main revisions:

We appreciate the reviewer's valuable comment. The land-use data were obtained from the Institute of Geographic Sciences and Natural Resources Research, Chinese Academy of Sciences at a horizontal resolution of 1 × 1 km (https://www.resdc.cn/DOI/DOI.aspx?DOIID=129; last accessed on 25 June 2022). The elevation data were obtained from the Shuttle Radar Topography Mission at a horizontal resolution of 1 × 1 km (https://www.resdc.cn/data.aspx?DATAID=123; last accessed on 25 June 2022). We have corrected the data source information in **lines 155-162 in the revised manuscript**.

Q4. Specific points: L148: References for CARSNET are needed.

Response and main revisions:

We appreciate the reviewer's valuable comment. We have added the reference for CARSNET (Che et al., 2015) in **line 172 in the revised manuscript**.

Q5. Specific points: L149: References for CARE-China and SONET are needed.

Response and main revisions:

We appreciate the reviewer's valuable comment. We have added the reference for CARE-China (Xin et al., 2015) and SONET (Li et al., 2018) in **lines 173-174 in the revised manuscript**.

Q6. Specific points: L163: Have you tested uncertainties brought by using four months to represent four seasons?

Response and main revisions:

We appreciate the reviewer's valuable comment. In this study, January, April, July, and October were selected as representative months of different seasons to avoid abundant calculations. This method has been widely applied in emission inversion researches (Zhang et al., 2015; Zhao et al., 2019). The relative difference between the average AAOD of four representative months and annual value were estimated within

-4%~-1% during 2000-2020. We do not quantify this uncertainty due to computational costs in this work. We have added relative information in **lines 193-194 in the revised manuscript**.

Q7. Specific points: L166: Add references for CMAQ.

Response and main revisions:

We appreciate the reviewer's valuable comment. We have added the reference for CMAQ (USEPA, 2017) in **lines 196-197 in the revised manuscript**.

Q8. Specific points: L221: References are needed for the log-linear model.

Response and main revisions:

We appreciate the reviewer's valuable comment. We have added the reference for the log-linear model (Wang et al., 2021) in **line 256 in the revised manuscript**.

Q9. Specific points: L221: L231: References are needed for Equation 7.

Response and main revisions:

We appreciate the reviewer's valuable comment. We have added the reference for the Equation 10 (Eq. 7 in the previous edition) (Wang et al., 2021) in **lines 266-267 in the revised manuscript**.

Q10. Specific points: L257: Where are the RMSE and NMB values from? From which plots or tables?

Response and main revisions:

We appreciate the reviewer's valuable comment. We have added the data sources in **line 295 in the revised manuscript** and corresponding **Supplementary Table S8 in the revised supplement**.

Table S8 in the revised supplement: Evaluation of the monthly XGBoost-predicted AAOD and MERRA-2 AAOD performance against ground measurements.

	BIAS	RMSE	NMB	NME
XGBoost-predicted AAOD (this study)	0.0023	0.017	5%	32%
MERRA-2 AAOD	-0.0079	0.021	-19%	37%

Q11. Specific points: L284: Double brackets at the right side.

Response and main revisions:

We appreciate the reviewer's reminder and the redundant bracket has been deleted in the revised manuscript.

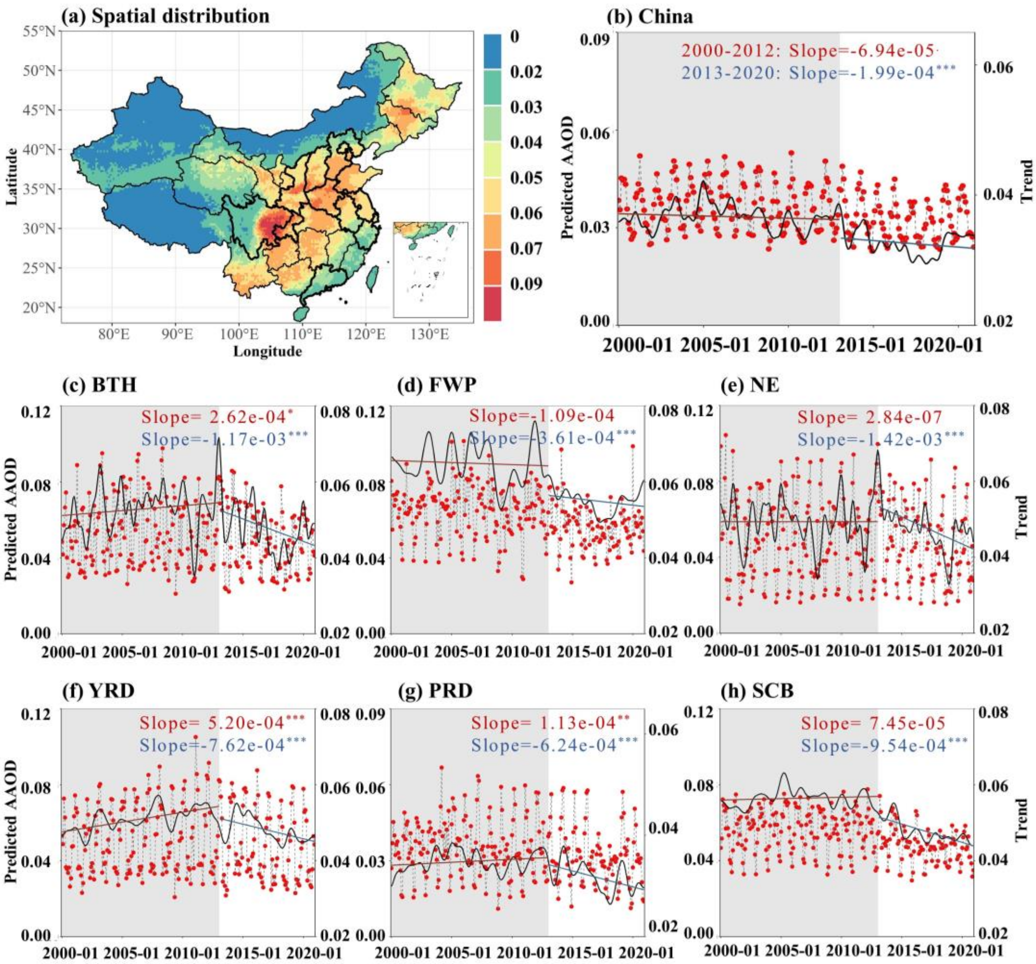
Q12. Specific points: L314: I do not agree that the increase in AAOD from 2018 to 2020 is due to the increasing surface wind speed. In general, the near-surface wind speed has decreased significantly since 1980 and has become flat or increased slightly since about 2010~2013. Why did the AAOD still decrease from 2013 to 2020? Thus, near-surface wind speed only cannot explain the changing trend in AAOD. Moreover, from 2018 to 2020, only 3 years, the time period is too short for changing trend analysis.

Response and main revisions:

We appreciate the reviewer's valuable comment. Continuous air pollution controls during 2013-2020 have resulted in the reduction of light-absorption BC emissions, thereby the decline of AAOD of China. However, recent observations show that the frequency of sandstorms in northern China has increased after 2015 due to increasing surface wind speed (Yang et al., 2021). This resulted in greater emissions of light-absorption dust aerosols, thereby AAOD, partly offset the AAOD decline owing to black carbon emission mitigation in northern China. We agreed with the reviewer that the time period from 2018 to 2020 is too short for changing trend analysis, so we removed the trend analysis results for 2018-2020 from Figure 3 and the main text.

We have included the discussion in **lines 352-355** and modified **Figure 3 in the revised manuscript**.

Figure 3 in the revised manuscript: (a) Spatial distribution of multiyear average AAOD during 2000-2020 and (b-h) interannual variations of AAOD for China and six key regions in 2000-2020. The grey and white present Phase 1 (2000-2012) and Phase 2 (2013-2020), respectively. The red dots and dashed line represent time series of monthly AAOD (left vertical axis). The black solid lines represent the interannual variability after removing the seasonal change through time-series decomposition (right vertical axis). The straight red and blue lines present the linear trends of AAOD for different phases (right vertical axis). The annual variation rates (1/yr) during different phases with significance levels (*p < 0.05, **p < 0.01, ***p < 0.001) are presented.



Q13. Specific points: L327-329: Could you explain why a larger underestimation appears in 2000 and 2020?

Response and main revisions:

We appreciate the reviewer's valuable comment. The larger underestimation of simulated prior BC concentration in 2000 and 2020 may be caused by larger underestimation of BC emissions in these years.

For 2000, the under-reporting of activity levels and lack of local measurements for specific BC emission factors (EFs, emissions per unit of activity level) in very early year may lead to larger uncertainties in BC emission estimation (Fu et al., 2012; Guan et al., 2012). The increased uncertainty in prior BC emissions in 2020 may have resulted partly from an underestimation of increased fuel use owing to residential heating and cooking during the COVID-19 lockdown and quarantine (Zheng et al., 2020).

We have discussed the possible reasons in **lines 366-367 and 407-418 in the revised manuscript**.

Q14. Specific points: L345: Simulation -> simulation.

Response and main revisions:

We appreciate the reviewer's reminder. We are sorry for the mistake and have corrected it in **line 384 in the revised manuscript**.

Q15. Specific points: L361: What do you mean by the factor here?

Response and main revisions:

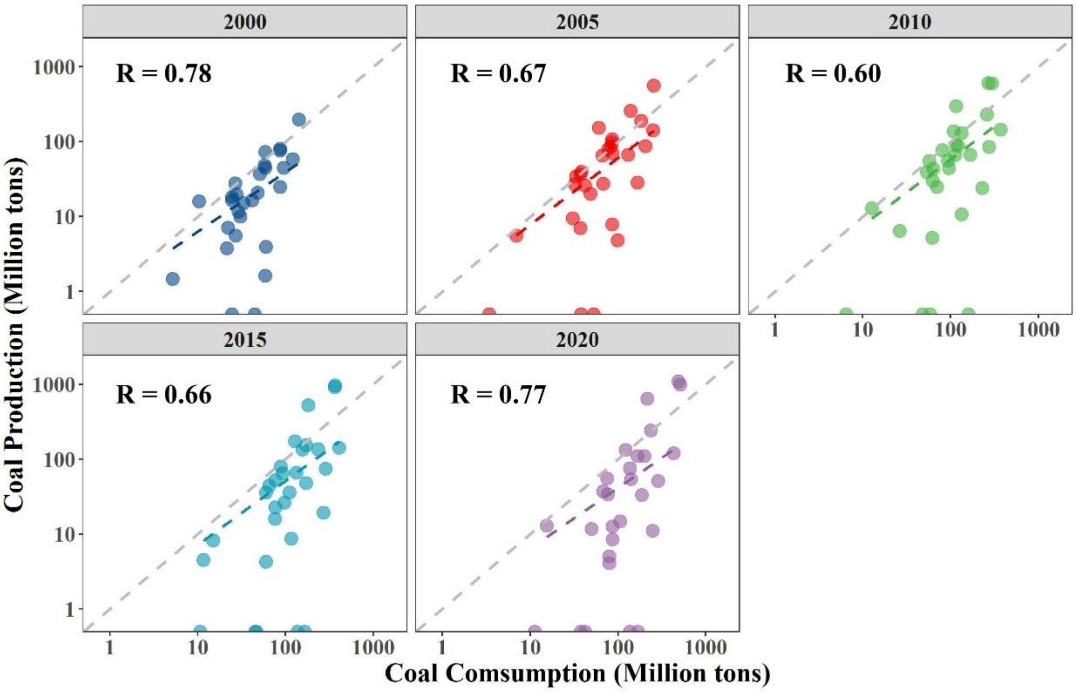
We appreciate the reviewer's valuable comment. The "factor" here represents the ratio of the difference between posterior and prior to the prior BC emissions, i.e., (a posterior – a prior) / a prior. We have modified the expression in **lines 400-401 in the revised manuscript**.

Q16. Specific points: L426: Why coal consumption or fossil fuel combustion are missing, which are also very important to BC emissions.

Response and main revisions:

We appreciate the reviewer's important comment. As shown in Figure R1 below, there is a great correlation between provincial coal production and consumption. In Section 3.2.3, we used coal production as an indicator to distinguish the main coal production provinces and to further highlight the unique BC emission patterns for those provinces (Shanxi, Inner Mongolia, Henan and Shaanxi Province). Those patterns cannot be clearly revealed if coal consumption is used as the indicator.

Figure R1 Correlation between provincial coal consumption and production by year (2000-2020, with a five-year interval).



Q17. Specific points: L490-493: How did you get those values? Calculated from equation 7?

Response and main revisions:

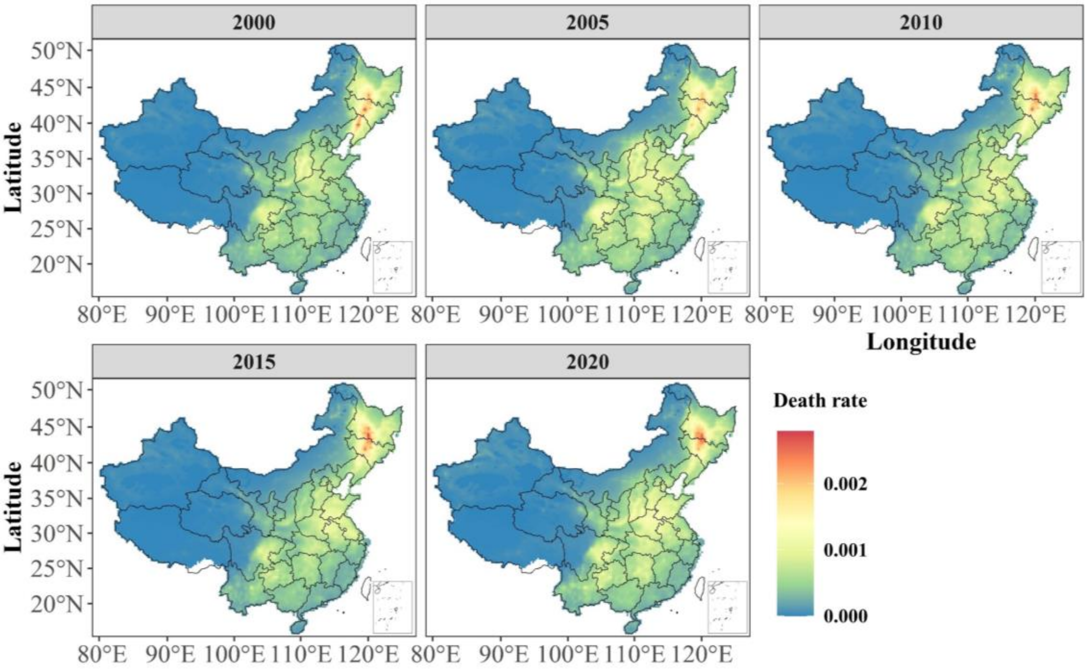
We appreciate the reviewer's valuable comment. The all-cause premature deaths attributed to BC here were calculated from equation 10 (equation 7 in the previous edition) in **lines 266-278 in the revised manuscript**.

Q18. Specific points: L496-498: Can you calculate the relative premature deaths that divide the premature death cases by the total population? Then, one can get rid of the impacts of population density when comparing premature deaths in different regions of China.

Response and main revisions:

We appreciate the reviewer's valuable comment. We calculated the premature death rate that divide the premature death cases by the total population and the results are shown in a **new Supplementary Figure S9 in the revised supplement**. Higher premature death rate in eastern China were attributed mainly to the relatively high BC exposure from developed industrial and commercial activities. We have added the corresponding statement in **lines 536-539 in the revised manuscript**.

Figure S9 in the revised supplement: Spatial distribution of premature mortality rate attributable to the posterior BC exposure during 2000-2020 (with a five-year interval).



Q19. Specific points: L499: What does cases/grid mean?

Response and main revisions:

We appreciate the reviewer's valuable comment. The "cases/grid" means the all-cause premature deaths attributed to BC in a model grid cell (27 × 27 km). Combined with the Q21 of reviewer #2, we have modified the unit "cases/grid" to "cases/1000 km^2" and re-calculated the corresponding values in **Table 3 in the revised manuscript** to make the expression easier to understand. The corresponding sentences were modified to "The highest multiyear average of premature mortality was 1482 cases/1000 km^2 (the all-cause premature deaths attributed to BC per area of 1000 km^2) in Shanghai, followed by 793, 761, 520, 450, and 442 cases/1000 km^2 in Beijing, Tianjin, Jiangsu, Henan, and Shandong, respectively (Table 3). These values were much higher than the national average of 86 cases/1000 km^2" in **lines 539-543 in the revised manuscript**.

Table 3 in the revised manuscript: The annual all-cause premature mortality associated with BC exposure by province in mainland China during 2000-2020 with a five-year interval (Unit: cases/1000 km^2). Locations of provinces are shown in Figure S1a.

	2000	2005	2010	2015	2020	Average
Shanghai	1050	1258	1661	2038	1403	1482
Beijing	410	686	781	930	1156	793
Tianjin	442	601	760	907	1094	761
Jiangsu	341	481	575	669	535	520
Henan	386	484	421	404	555	450
Shandong	299	433	455	494	529	442
Anhui	218	320	338	341	322	308
Liaoning	346	282	271	234	226	272
Hebei	194	250	238	249	303	247
Chongqing	253	285	249	204	239	246
Hubei	173	250	245	192	210	214
Jilin	207	200	206	212	188	203
Zhejiang	140	180	210	234	224	198
Hunan	163	227	206	171	216	197
Guangdong	149	177	213	165	192	179
Shanxi	192	213	134	130	217	177
Sichuan	124	148	130	105	113	124
Guizhou	109	128	111	120	132	120
Jiangxi	90	127	129	108	136	118
Shannxi	97	120	95	80	116	101
Heilongjiang	87	88	104	113	109	100
Fujian	81	87	99	79	84	86
Guangxi	70	96	99	72	81	84
Hainan	46	59	64	43	59	54
Yunnan	42	51	49	50	59	50
Ningxia	34	49	33	34	49	40
Gansu	23	30	19	17	22	22
Inner Mongolia	6	8	6	5	7	7
Qinghai	2	3	2	2	2	2
Xinjiang	1	1	1	1	1	1
Tibet	0	0	0	0	0	0

Q20. Specific points: Table 1: Units are needed for the values of emission.

Response and main revisions:

We appreciate the reviewer's valuable comment. Table 1 shows the multiyear average relative differences between the posterior and various "bottom-up" estimates of BC emissions. We have added "unitless" for the relative difference **in the caption of Table 1**.

Table 1 in the revised manuscript: The multiyear average relative differences between the posterior and various "bottom-up" estimates of BC emissions by region (unitless). Note: OBB emissions from GFED are added to each anthropogenic emission estimate as the total "bottom-up" estimate, except for PKU-Fuel which includes its own OBB emission estimate.

Region	CEDS + GFED	PKU-Fuel	EDGAR + GFED	REAS + GFED	MEIC + GFED	Average
BTH	1.19	0.31	2.35	0.69	0.99	1.11
FWP	2.18	0.56	3.63	1.50	2.70	2.11
YRD	0.80	1.44	1.23	1.49	1.56	1.30
PRD	0.75	1.15	2.01	2.05	2.65	1.72
SCB	3.23	2.23	6.38	3.01	3.17	3.60
NE	4.90	4.66	6.37	5.36	6.81	5.62
Other	3.35	1.98	4.96	3.68	3.62	3.52
China	2.68	1.72	4.13	2.82	3.26	2.92

Q21. Specific points: Figure 1: The quality of the figure needs to be improved. The words are too small and unreadable.

Response and main revisions:

We appreciate the reviewer's valuable comment. We have improved the quality of **Figure 1 in the revised manuscript**.

Q22. Specific points: Figure 2: I highly recommend using five different colors for the data from five different years. The two dashed lines have to be introduced in the caption. The interval of bins is also needed to be introduced. The equations of NMB, NME, and RMSE should go to section 2.1.

Response and main revisions:

We appreciate the reviewer's valuable comment and modified the Figure 2. Firstly, we use 16 different colors to represent the data from 16 years (2005-2020) in Figure 2. Secondly, we added the introduction of the dashed lines and the interval of bins in the caption of Figure 2, i.e., "The red dashed line indicates the 1:1 line. The blue dashed line indicates the regression line. The interval of bins of the marginal histograms is 0.02". Finally, the equations of NMB, NME and RMSE were moved to section 2.1 in **lines 181-188 in the revised manuscript**.

Q23. Specific points: Figure 3: The quality needs to be improved. Words and legends are not clear enough.

Response and main revisions:

We appreciate the reviewer's valuable comment and improve the quality of Figure 3 to make the words and legends clear enough.

Q24. Specific points: Figure 7: What do colors in Figures 7c and 7d stand for?

Response and main revisions:

We appreciate the reviewer's valuable comment. The colors in Figures 7c and 7d stand for changes in provincial BC emission intensity (annual BC emissions per km^2) in posterior BC estimates, which are the same as those in Figures 7a and 7b. We have added the information **in the caption of Figure 7**.

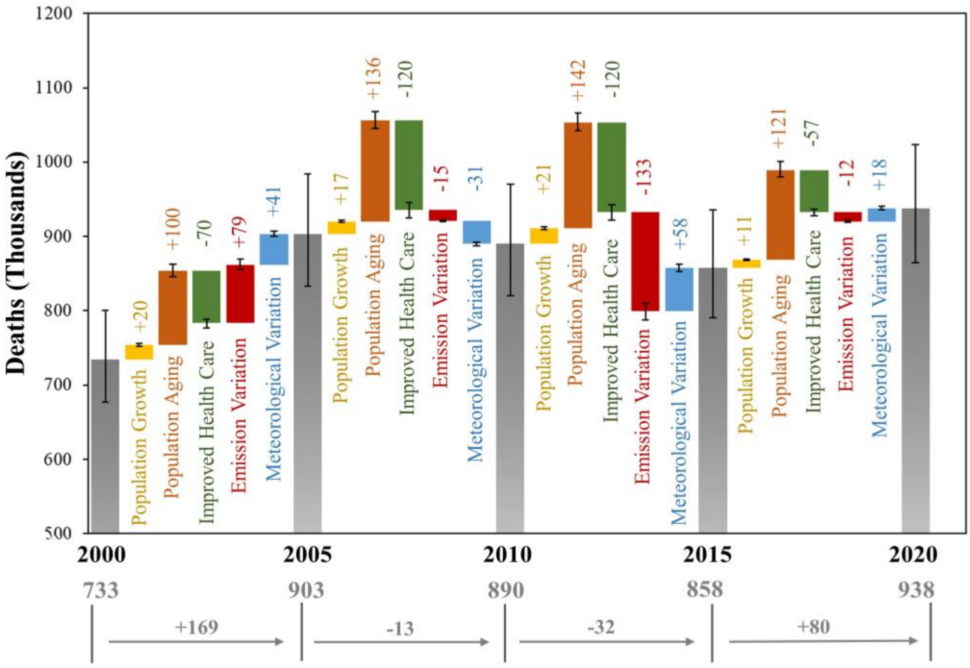
Q25. Specific points: Figure 9: The quality of the figure needs to be improved. Words are not readable. Important information should be included in the

caption, for example, what the gray bars are and why the numbers over the gray bars are missing.

Response and main revisions:

We appreciate the reviewer's valuable comment and improve the quality of Figure 9 to make the words and legends clear enough. The grey bars stand for the total all-cause premature deaths attributed to BC exposure in China during 2000-2020 (with a 5-year interval), and the corresponding numbers are at the bottom of the Figure. The colored bars and numbers above the bars show the contributions of major factors to the national changing mortality. We have added the information **in the caption of Figure 9 in the revised manuscript**.

Figure 9 in the revised manuscript: Total all-cause premature deaths attributed to BC exposure in China (grey bars) and drivers of changing premature mortality (colored bars) during 2000-2020 (with a 5-year interval). Error bars show the 95% CI of estimates in this study. The numbers of total all-cause premature deaths are at the bottom of the Figure. The contributions of major factors to the national changing mortality are above the colored bars.



Reference

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Comments from Reviewer #2

General comment: The authors analyzed BC emission changes and the associated premature mortality in China during 2000-2020. Overall, the methodology is robust and the findings are valuable. The paper was well written and I enjoyed reading it. The following comments need to be addressed before publishing.

Response and main revisions:

We appreciate the reviewer's positive comments on our paper, and have made point-by-point response and revisions as summarized below.

Q1. Line 31: Explicitly mention OMI and Extreme Gradient Boosting algorithm in the abstract.

Response and main revisions:

We appreciate the reviewer's valuable comment and have added the specific information. The sentence has been modified as "Here, we present the spatiotemporal evolution of BC emissions and the associated premature mortality in China during 2000-2020, based on an integrated framework combining satellite observations from Ozone Monitoring Instrument (OMI), an Extreme Gradient Boosting (XGBoost) algorithm, a "top-down" inversion approach, and an exposure-response model" in **lines 29-33 in the revised manuscript**.

Q2. Line 47: "BC poses greater health risks than total PM2.5 due to its absorption"? Why would absorption be related to health?

Response and main revisions:

We appreciate the reviewer's valuable comment. Here the "absorption" means the ability of BC to absorb the harmful matters. Epidemiological studies have indicated that BC absorbs polycyclic aromatic hydrocarbons (PAHs) and volatile organic compounds (VOCs) due to its fine particle size and porous structure, and readily penetrates human lung tissue (Pani et al., 2020). Thus BC exposure may cause cardiovascular diseases (CVDs) and respiratory diseases (RDs). We have modified the unclear expression and added the corresponding reference in **lines 48-51 in the revised manuscript**.

Q3. Line 64-65: "9%-22% (2005-2020) and 8%-12% (2006-2013)". Reference?

Response and main revisions:

We appreciate the reviewer's valuable comment. Here the data coverage ratio was calculated by the author based on the satellite AAOD datasets. We have added the data sources of OMI (https://disc.gsfc.nasa.gov/datasets/OMAEROe_003/summary; last accessed on 10 March 2022) and POLDER (https://www.grasp-open.com; last accessed on 4 May 2022) in **lines 65-69 in the revised manuscript**.

Q4. Line 72 "Existing bottom-up estimates". Please explicitly list the name of these inventories mentioned here.

Response and main revisions:

We appreciate the reviewer's valuable comment. We have listed the name of the inventories mentioned here in **lines 79-84 in the revised manuscript**, i.e., the Multiresolution Emission Inventory for China (MEIC; Tsinghua University, 2023), the Emissions Database for Global Atmospheric Research (EDGAR; European Commission, 2022), Community Emissions Data System (CEDS; Mcduffie et al., 2020), the Peking University Fuel Inventory (PKU-Fuel; Wang et al., 2014), Regional Emission inventory in ASia (REAS; Kurokawa and Ohara, 2020), and others (Lu et al., 2011; Lei et al., 2011; Klimont et al., 2009; Qin and Xie, 2012).

Q5. Section 2.1: Move the description of XGBoost model from Supplement to the main text. And explicitly explain how it differs from a Random Forest Model.

Response and main revisions:

We appreciate the reviewer's valuable comment. We have moved the description of XGBoost model from Supplement to **lines 120-127 in the revised manuscript**. XGBoost has been widely used in predicting air pollution and shown to outperform various statistical and machine learning models (Liang et al., 2020; Liu et al., 2022; Wang et al., 2023; Xiao et al., 2018). The XGBoost algorithm is an additive model based on hundreds of decision tree models. It first builds multiple Classification and Regression Trees, and then integrates these trees as a new tree model using an additive function (Liu et al., 2021). The model continues to iteratively improve, and the new tree model generated in each iteration will fit the residual of the previous tree. The complexity of the ensemble model will gradually increase until the training achieves the best results. Different from the boosting approach of XGBoost, the Random Forest model fits a set of decision trees, and then a majority vote method is taken for final prediction (Lyu et al., 2019). Generally, XGBoost model requires less training and prediction time and presents better performance than the Random Forest model. We have added the comparison of two models in **lines 128-131 in the revised manuscript.**

Q6. Line 98: "extreme gradient boosting" ==> "Extreme Gradient Boosting"

Response and main revisions:

We appreciate the reviewer's valuable comment. We have modified the "extreme gradient boosting" to "Extreme Gradient Boosting" in the main text (**lines 31-32 and lines 108-109**).

Q7. Line 110: "XGBoost has been widely used…" In this case, please provide more reference than Xiao et al., 2018.

Response and main revisions:

We appreciate the reviewer's valuable comment. We have added more relative references (Liang et al., 2020; Liu et al., 2022; Wang et al., 2023) in **lines 121-122 in the revised manuscript.**

Q8. In the Text S1 XGBoost model description, can you explain how you "integrates these trees as a new tree model"?

Response and main revisions:

We appreciate the reviewer's valuable comment. The XGBoost algorithm is an additive model. For a given dataset, the XGBoost algorithm continuously perform feature splitting to grow a tree. Each time a tree is added, a new function is actually learned to fit the residuals of the last tree prediction. Eventually the model uses an additive function to "integrates these trees as a new tree model", thus obtaining the final prediction. We have added the explanation in **lines 123-125 in the revised manuscript**.

Q9. Line 113: "random forest" ==> "Random Forest".

Response and main revisions:

We appreciate the reviewer's valuable comment. We have modified the "random forest" to "Random Forest" in the main text (**line 128 and line 131**).

Q10. Line 139: To regrid from 1 km to 0.25 degree, you should use average rather than Bilinear interpolation.

Response and main revisions:

We appreciate the reviewer's valuable comment. We are sorry for the mistake and have modified it to "These parameters were resampled to the 0.25° × 0.25° grid system by averaging the 1-km resolution data." in **lines 162-163 in the revised manuscript**.

Q11. OMI overpass time is ~13:30. Therefore OMI only measures AAOD at ~13:30 each day. Therefore when you use other MERRA-2 variables for model training, it's better to use hourly values near 13:30 rather than daily average.

You don't need to redo the training. However please add a few sentences to discuss this.

Response and main revisions:

We appreciate the reviewer's valuable comment. We used the daily MERRA-2 data because the daily average MERRA-2 data was proved more reliable than the hourly data compared to the observation (Xu et al., 2020). However, as the reviewer said, application of the daily data may result in a mismatch with OMI-measured AAOD at ~13:30, thus leading to uncertainties in AAOD prediction. The complicated nonlinear response relationship of machine learning (i.e., XGBoost model) can partially compensate this mismatch, and our validation results also proved the robustness of the model. Evaluated by 10-fold CV and individual ground measurements, the predicted AAOD shows good agreements with observations, with RMSE of 0.013 and 0.017, respectively (Figure 2 and Supplementary Table S8). We have added the explanation in **lines 153-155 in the revised manuscript**.

Q12. Your developed monthly AAOD data represent the value at ~13:30 if you use OMI AAOD as the truth for training. Please discuss the impact of diurnal variation on your emission estimates. GEMS will provide more diurnal information in the future and can be used to further understand this issue.

Response and main revisions:

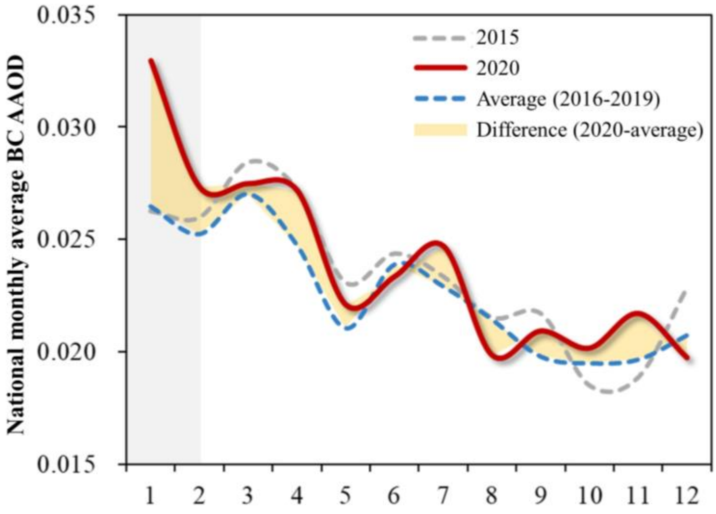
We appreciate the reviewer's valuable comment and acknowledged the limitation in emission inversion of this work. As XGBoost predicted monthly BC AAOD at ~13:30, we could not capture the diurnal distribution of BC emissions. We simply applied the ratio of the posterior to prior emissions at 13:30 to correct emissions in other hours, causing uncertainties in the diurnal distribution of emissions. As mentioned by the reviewer, the Geostationary Environment Monitoring Spectrometer (GEMS) was launched on board the Geostationary KOrea Multi-Purpose SATellite 2B (GEO-KOMPSAT-2B) satellite in 2020 and provided hourly daytime observations of aerosols (Kim et al., 2020; Park et al., 2023). This can potentially be helpful for improving the temporal accuracy of BC emission inversion in the future. We have added the discussion of the uncertainty in **lines 603-609 in the revised manuscript**.

Q13. Line 163: While it makes sense to have five-year intervals, using 2015 and 2020 to interpolate the years in between will introduce biases due to COVID interruption (i.e., 2020 emission is an anomaly).

Response and main revisions:

We appreciate the reviewer's valuable comment. We acknowledged that 2020 is an anomaly considering the influence of COVID-19. We compared the national monthly BC AAOD in 2015, 2020 and 2016-2019 (Figure R2). Annual average BC AAOD in 2020 was 0.0235, slightly lower than that in 2015 (0.0239), while both of them were higher than the multi-year average BC AAOD in 2016-2019 (0.0227). The COVID-19 lockdown and quarantine may cause the increase of fuel use owing to residential heating and cooking (Zheng et al., 2020), thereby the increase of BC emissions. We have briefly discussed the influence of COVID-19 in **lines 415-418 in the revised manuscript**. To describe an emission trend of entire two decades, we kept using the five-year interval in the emission inversion analysis.

Figure R2 Comparison of the national average monthly BC AAOD in different years, i.e., 2015, 2020 and the multi-year average for 2016-2019.



Q14. Line 181: To be more clear, I suggest to replace $AAOD\_simi$,, with $AAOD$_BC_$simi,m,n$ here and after.

Response and main revisions:

We appreciate the reviewer’s valuable comment. We have modified the “$AAOD\_sim_{i,m,n}$” to “$AAOD\_BC\_sim_{i,m,n}$” in the main text (**line 211, 212, 231, 233, 238 and 239**) and **Table S2 in the revised supplement**.

Q15. Section 2.2.2: I appreciate this part that the authors included four sensitivity tests to recalculate posterior BC emissions and explore the uncertainty in the inversion.

Response and main revisions:

We appreciate the reviewer’s positive comment on this work.

Q16. Line 227: “we applied the 1.25th percentile of BC concentrations as the threshold.” Why use 1.25? Is it from a previous study?

Response and main revisions:

We appreciate the reviewer’s valuable comment. The health impact threshold of BC (i.e., 1.25th percentile of BC concentrations) was suggested by previous study (Pani et al., 2020; Wang et al., 2021). We have added the reference in **lines 261-262 in the revised manuscript.**

Q17. Figure 4: Have you described these BC concentration observations in the text? How were they measured?

Response and main revisions:

We appreciate the reviewer’s valuable comment. The BC concentration observations here were collected from 64 published researches as comprehensive as possible, covering various sampling regions in China and study period from 2000 to 2020, which are listed in Supplementary Table S3. Most studies analyzed BC using well-acknowledged reliable and widely used analyzers (Tao et al., 2017), for example, a DRI carbon analyzer or Sunset carbon analyzer. We have added the description of BC concentration observations in **lines 244-248 in the revised manuscript**.

Q18. Did you add anthro and fire BC emis together and updated them as a whole? Please be explicit.

If that's the case, why would you compare your BC emission estimates (anthro+BB) with other anthropogenic emission inventories in Figure 5? If that's not the case, did you separately update anthro and biomass burning BC emissions? Or you only updated anthro emissions? Biomass burning emissions are a large source for BC (sometimes larger than anthropogenic sources) and have to be considered.

Response and main revisions:

We appreciate the reviewer's valuable comment and careful reminding. In this study, we added anthropogenic and fire BC emissions together and updated them as a whole. We acknowledged that comparing the posterior BC emissions (anthro+BB) with other "bottom-up" anthropogenic emission inventories (i.e., EDGAR, CEDS, REAS, et al.) was not rigorous enough. Here we added GFED open biomass burning BC emissions to various anthropogenic emission inventories and re-compared the posterior emissions with the new "bottom-up" emission estimates. It is worth noting that the PKU-Fuel, Lu et al. (2011) and Qin and Xie (2012) already includes emissions from wildfires. The posterior BC emissions presented an enhancement compared to various "bottom–up" estimates of China's BC emissions (sum of anthropogenic and OBB emissions), with the lowest relative difference of 1.7 for the PKU-Fuel (http://inventory.pku.edu.cn/; last accessed on 1 May 2023) and highest value of 4.1 for EDGAR+GFED (https://edgar.jrc.ec.europa.eu/dataset_ap61; last accessed on 1 May 2023) (Figure 5d and Table 1). The posterior emissions presented a smaller interannual variability compared to the prior and other "bottom-up" estimates, with a net growth of 8% during 2000-2010 (the analogous numbers are

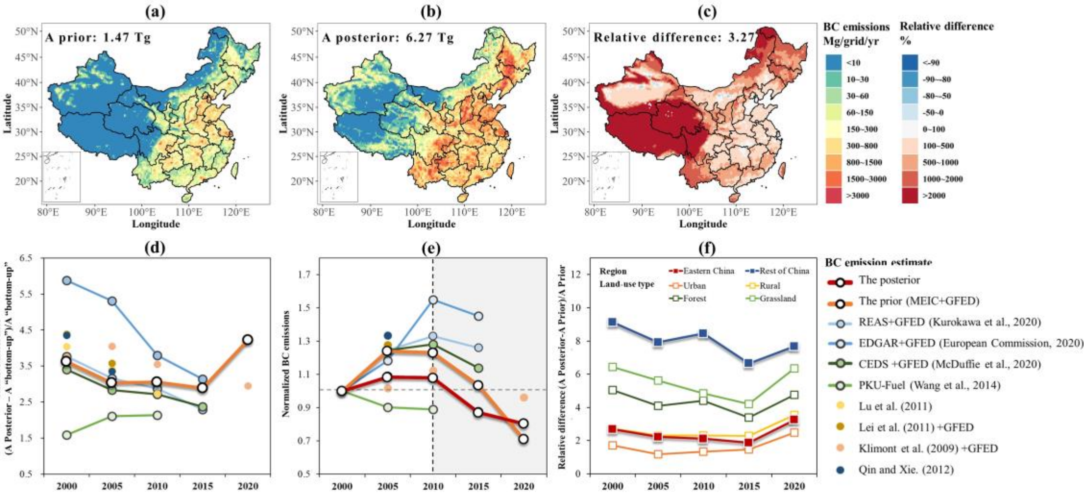
12%-55% for various “bottom-up” estimates including 24% for the prior used in this work, MEIC+GFED) and a decline of 26% during 2010-2020 (41% for MEIC+GFED, Figure 5e). Besides, the relative differences between posterior and other “bottom-up” BC emission estimates were smaller (1.1-2.1) in more economically developed regions (BTH, FWP, YRD, and PRD), but larger (3.5-5.6) in SCB, NE, and other regions (Table 1).

We have modified the **Figure 5 and Table 1** and corresponding descriptions in **lines 402-407, lines 419-423 and lines 429-432 in the revised manuscript**.

Table 1 in the revised manuscript: The multiyear average relative differences between the posterior and various “bottom-up” estimates of BC emissions by region (unitless). Note: OBB emissions from GFED are added to each anthropogenic emission estimate as the total “bottom-up” estimate, except for PKU-Fuel which includes its own OBB emission estimate.

Region	CEDS + GFED	PKU-Fuel	EDGAR + GFED	REAS + GFED	MEIC + GFED	Average
BTH	1.19	0.31	2.35	0.69	0.99	1.11
FWP	2.18	0.56	3.63	1.50	2.70	2.11
YRD	0.80	1.44	1.23	1.49	1.56	1.30
PRD	0.75	1.15	2.01	2.05	2.65	1.72
SCB	3.23	2.23	6.38	3.01	3.17	3.60
NE	4.90	4.66	6.37	5.36	6.81	5.62
Other	3.35	1.98	4.96	3.68	3.62	3.52
China	2.68	1.72	4.13	2.82	3.26	2.92

Figure 5 in the revised manuscript: Comparisons between posterior and various "bottom-up" BC emission estimates in China during 2000-2020. (a) Multiyear average spatial distribution of prior BC emissions (MEIC+GFED), (b) posterior BC emissions, and (c) their relative differences. (d) Long-term variability in the relative differences between posterior and various "bottom-up" BC emission estimates, with five-year intervals. Note: OBB emissions from GFED are added to each anthropogenic emission estimate as the total "bottom-up" estimate, except for PKU-Fuel, Lu et al. (2011) and Qin and Xie (2012) which include their own OBB emission estimate. (e) Long-term variability in normalized posterior and various "bottom-up" BC emission estimates (relative to 2000). The grey area indicates the period with declining national BC emissions. (f) Long-term variability in the relative differences between prior (MEIC+GFED) and posterior BC emissions by region and land use type.



Q19. Figure 5: The relative difference is larger in forest and grassland rather than rural or urban. Does that mean the uncertainties mainly come from biomass burning emissions rather than anthro?

Response and main revisions:

We appreciate the reviewer's valuable comment. Yes, it means that the uncertainties of BC emissions in remote regions, including those from open biomass burning should be larger than those from more intensive human activities. The "bottom-up" approach could capture information about energy consumption and pollution controls more easily and accurately in regions with more intensive human activities. However the omission of small fires from satellite observations and application of global EFs led to an underestimation of biomass burning emissions. We have included the discussions in **lines 411-413 and lines 442-446 in the revised manuscript**.

Q20. Line 485: Just out of curiosity, is transportation a significant source sector for BC in China?

Response and main revisions:

We appreciate the reviewer's valuable comment. The transportation sector is an important source of BC emissions in China. According to MEIC, the contributions of transportation sector to total anthropogenic BC emissions in China during 2000-2020 (i.e., our study period) varied between 17%-24%.

Q21. Line 501: It's more intuitive to use unit cases/km^2 than cases/grid.

Response and main revisions:

We appreciate the reviewer's valuable comment. We have modified the unit "cases/grid" to "cases/1000 km^2" and re-calculated the corresponding values in **Table 3 in the revised manuscript**. The corresponding sentences were modified to "The highest multiyear average of premature mortality was 1482 cases/1000 km^2 (the all-cause premature deaths attributed to BC per area of 1000 km^2) in Shanghai, followed by 793, 761, 520, 450, and 442 cases/1000 km^2 in Beijing, Tianjin, Jiangsu, Henan, and Shandong, respectively (Table 3). These values were much higher than the national average of 86 cases/1000 km^2" in **lines 539-543 in the revised manuscript**.

Table 3 in the revised manuscript: The annual all-cause premature mortality associated with BC exposure by province in mainland China during 2000-2020 with a five-year interval (Unit: cases/1000 km^2). Locations of provinces are shown in Figure S1a.

	2000	2005	2010	2015	2020	Average
Shanghai	1050	1258	1661	2038	1403	1482
Beijing	410	686	781	930	1156	793
Tianjin	442	601	760	907	1094	761
Jiangsu	341	481	575	669	535	520
Henan	386	484	421	404	555	450
Shandong	299	433	455	494	529	442
Anhui	218	320	338	341	322	308
Liaoning	346	282	271	234	226	272
Hebei	194	250	238	249	303	247
Chongqing	253	285	249	204	239	246
Hubei	173	250	245	192	210	214
Jilin	207	200	206	212	188	203
Zhejiang	140	180	210	234	224	198
Hunan	163	227	206	171	216	197
Guangdong	149	177	213	165	192	179
Shanxi	192	213	134	130	217	177
Sichuan	124	148	130	105	113	124
Guizhou	109	128	111	120	132	120
Jiangxi	90	127	129	108	136	118
Shannxi	97	120	95	80	116	101
Heilongjiang	87	88	104	113	109	100
Fujian	81	87	99	79	84	86
Guangxi	70	96	99	72	81	84
Hainan	46	59	64	43	59	54
Yunnan	42	51	49	50	59	50
Ningxia	34	49	33	34	49	40
Gansu	23	30	19	17	22	22
Inner Mongolia	6	8	6	5	7	7
Qinghai	2	3	2	2	2	2
Xinjiang	1	1	1	1	1	1
Tibet	0	0	0	0	0	0

Q22. I like the Section 3.4 (detailed discussion on the uncertainties). It makes this manuscript more convincing.

Response and main revisions:

We appreciate the reviewer's positive comment on this work.

Q23. It's not convenient for the reviewers when you separate figure captions from figures. I had to go back and forth to understand a figure. I suggest in your future manuscript submissions, put figure and its corresponding caption in the same place.

Response and main revisions:

We appreciate the reviewer's valuable comment. We have put figure and its corresponding caption in the same place.

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