

Response to Reviewers

No.: ACP-2025-10

Title: Anthropogenic and Natural Causes for the Interannual Variation of PM_{2.5} in East Asia During Summer Monsoon Periods From 2008 to 2018

Anonymous referee #3:

The manuscript “Anthropogenic and Natural Causes for the Interannual Variation of PM_{2.5} in East Asia During Summer Monsoon Periods From 2008 to 2018” by Ma et al. used a regional climate chemistry-ecosystem coupled model to investigate interannual variations in PM_{2.5} across East Asia from 2008 to 2018 and investigates the drivers. This has been an important topic in the past years, and this work improves over previous studies by exploring the impact of CO₂. I feel this point is of interest to the community and falls within the scope of ACP. The manuscript is also well written and easy to follow. I recommend publication after addressing the following points.

Response: We thank referee #3 for careful reading and valuable comments. We have responded to each specific comment in blue below. Please note that the line numbers given below refer to the clean version of the manuscript.

1. My major concern is the boundary between CO₂ change and meteorology change in the work. As mentioned in the text, CO₂ could influence PM via changing radiation, temperature, and precipitation. Aren't these already counted in the meteorology change? This needs to be explained clearer.

Response: Thanks. As shown in Table 1, the difference between SIM_{Base} and SIM_{MET=2008} (SIM_{Base} - SIM_{MET=2008}) quantifies the impact of meteorological variability on PM_{2.5} concentrations. Here, “meteorological variability” refers to the year-to-year changes in weather relative to the fixed 2008 baseline. In contrast, the difference between SIM_{Base} and SIM_{CO2=2008} (SIM_{Base} - SIM_{CO2=2008}) isolates the effect of CO₂ emission changes on PM_{2.5}. As a principal greenhouse gas, CO₂ modifies meteorological parameters—such as radiation, temperature, and precipitation—which in turn influence PM_{2.5} levels. In this comparison, all meteorological changes derive solely from variations in CO₂ concentration, a mechanism fundamentally different from the meteorological influences identified in Experiments SIM_{Base} and SIM_{MET=2008}.

We have added some discussions on this aspect.

Table 1. The Numerical experimental in this study.

Experiment	Time	Meteorological fields	CO ₂ emissions	Anthropogenic pollutant emissions
SIM ₂₀₀₈	2008	2008	2008	2008
SIM _{Base}	2009-2018	2009-2018	2009-2018	2009-2018

SIM _{MET=2008}	2009-2018	2008	2009-2018	2009-2018
SIM _{CO2=2008}	2009-2018	2009-2018	2008	2009-2018

Changes in manuscript:

2.1 Model description

(L141–148): “In the RegCM-Chem-YIBs model, changes in CO₂ concentrations affect PM_{2.5} primarily via two mechanisms: first, CO₂-induced radiative forcing alters the atmospheric radiation balance, leading to shifts in temperature, precipitation, and boundary-layer structure that modulate PM_{2.5} formation, transport, and removal(Li and Mölders, 2008; Matthews, 2007); And second, through the YIBs module, changes in CO₂ concentration modulate photosynthetic activity and stomatal behavior, altering BVOCs emissions that undergo atmospheric photochemical oxidation to form secondary organic aerosols, a significant fraction of PM_{2.5} (Kergoat et al., 2002; Kellomaki and Wang, 1998).”

2.3 Experiment settings

(L189–193): “It is noteworthy that, as a principal greenhouse gas, CO₂ modifies meteorological parameters—such as radiation, temperature, and precipitation—which in turn influence PM_{2.5} levels. In this comparison, all meteorological changes derive solely from variations in CO₂ emissions, a mechanism fundamentally different from the meteorological influences identified in experiments SIM_{Base} and SIM_{MET=2008}.”

References

- Kellomaki, S. and Wang, K. Y.: Growth, respiration and nitrogen content in needles of Scots pine exposed to elevated ozone and carbon dioxide in the field, *Environmental pollution* (Barking, Essex : 1987), 101, 263-274, 10.1016/s0269-7491(98)00036-0, 1998.
- Kergoat, L., Lafont, S., Douville, H., Berthelot, B., Dedieu, G., Planton, S., and Royer, J. F.: Impact of doubled CO₂ on global-scale leaf area index and evapotranspiration:: Conflicting stomatal conductance and LAI responses -: art. no. 4808, *Journal of Geophysical Research-Atmospheres*, 107, 10.1029/2001jd001245, 2002.
- Li, Z. and Mölders, N.: Interaction of impacts of doubling CO₂ and changing regional land-cover on evaporation, precipitation, and runoff at global and regional scales, *International Journal of Climatology*, 28, 1653-1679, 10.1002/joc.1666, 2008.
- Matthews, H. D.: Implications of CO₂ fertilization for future climate change in a coupled climate-carbon model, *Global Change Biology*, 13, 1068-1078, 10.1111/j.1365-2486.2007.01343.x, 2007.

2. Another important concern is for Section 3.2 and 3.3: I would suggest present some statistics other than just make the conclusions by spatial distribution plots. e.g., when you say a reduction of PM_{2.5} is associated with an increase of a certain factor, did you find a correlation? We cannot simply say a decrease of A is due to an increase of B and a decrease of C, they may just not relate to each other with small correlation.

Response: Thanks. We have provided the relevant statistical data in the supplementary information and have incorporated Tables S1–S4 into the main manuscript (Tables 3–6) to facilitate easier access for readers.

We attribute changes in PM_{2.5} concentrations to three primary factors: meteorological variability, CO₂ emission changes, and anthropogenic pollutant emissions changes. In Section 3.2, we assessed the combined effects of meteorological factors—including temperature, precipitation, wind speed, and planetary boundary layer height—on PM_{2.5} concentrations, without isolating the individual contributions of each factor. As illustrated in Figure 4 and Table 4, PM_{2.5} concentrations exhibit a negative correlation with precipitation and a positive correlation with temperature, elucidating the mechanisms by which meteorological conditions influence PM_{2.5} levels.

Similarly, in Section 3.3, we quantified the integrated impact of CO₂ on atmospheric PM_{2.5} concentrations through its modulation of biogenic volatile organic compound (BVOC) emissions and alteration of meteorological conditions.

Your insightful suggestion has provided us with a new perspective. In our forthcoming research, we plan to conduct sensitivity experiments by individually fixing specific meteorological variables. This approach will enable us to independently assess the impact of temperature, precipitation, wind speed, and other factors on PM_{2.5} concentrations. Additionally, we aim to distinguish the respective impacts of CO₂-induced meteorological changes and CO₂-driven alterations in BVOC emissions on PM_{2.5} levels. This line of inquiry represents a deeper exploration of the subject and promises to yield valuable insights.

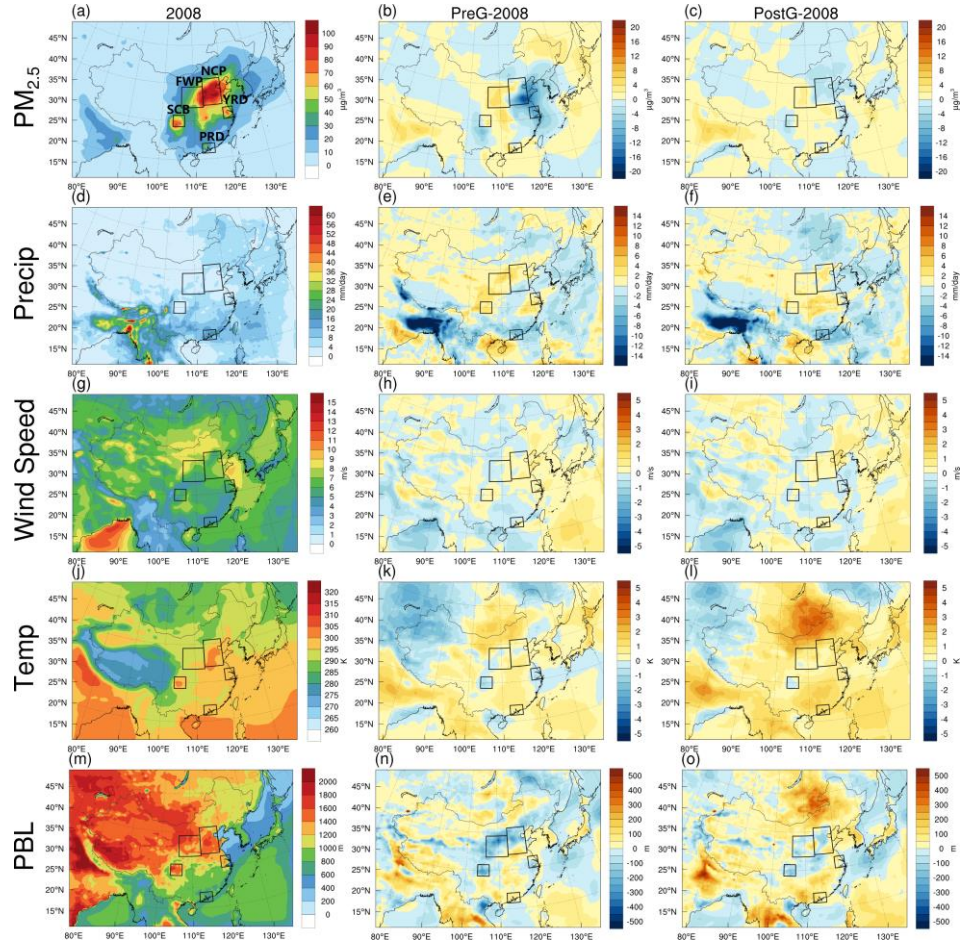


Figure 4. The PM_{2.5} (a–c, $\mu\text{g}/\text{m}^3$), precipitation (d–f, mm/day), wind speed (g–i, m/s), temperature (j–l, K), and Planetary Boundary Layer (PBL) height (m–o, m) during the EASM period in 2008 (left), and their mean changes due to meteorological variations in PreG (2009–2013, center) and PostG (2014–2018, right) phase relative to 2008 ($\text{SIM}_{\text{Base}} - \text{SIM}_{\text{MET}=2008}$).

Table 4. Impact of meteorological condition changes on PM_{2.5} ($\mu\text{g}/\text{m}^3$), precipitation (mm/day), wind speed (m/s), near-surface temperature (K), and Planetary Boundary Layer (PBL) height (m) during the EASM period in PreG (2009–2013) and PostG (2014–2018) phase relative to 2008 ($\text{SIM}_{\text{Base}} - \text{SIM}_{\text{MET}=2008}$).

Region	Period	PM _{2.5} ($\mu\text{g}/\text{m}^3$)	Precipitation (mm/day)	Wind Speed (m/s)	Near-Surface Temperature (K)	PBL (m)
NCP	PreG	-4.01	0.58	0.17	0.32	-46.8
	PostG	-1.6	0.6	0.26	0.6	-14.5
FWP	PreG	2.32	1.68	-0.06	0.1	-108.5
	PostG	1	0.81	0.05	0.46	-15.3
YRD	PreG	-6.31	1.02	0.18	-0.29	-33.9
	PostG	-0.43	0.48	-0.08	0.45	21.9
PRD	PreG	1.49	-2.39	-0.02	0.36	29.6
	PostG	0.11	-3.24	0.18	1.00	52.2

SCB	PreG	0.29	1.81	0.13	-0.58	-136.5
	PostG	-1.14	0.37	-0.03	-0.14	-76

Changes in manuscript:

Table 3. Changes in near-surface PM_{2.5} concentrations ($\mu\text{g}/\text{m}^3$) during the EASM period from 2009 to 2018 relative to 2008 in the North China Plain (NCP), Fen-Wei Plain (FWP), Yangtze River Delta (YRD), Pearl River Delta (PRD), and Sichuan Basin (SCB) (SIM_{Base} - SIM₂₀₀₈).

Year	NCP	FWP	YRD	PRD	SCB
2009	-11.24	-1.29	-11.37	1.41	-3.16
2010	-3.87	1.9	-15.2	-3.57	-4.79
2011	-6.27	0.22	-14.76	0.13	-8.65
2012	-7.42	1.69	-17.61	2.35	-15.99
2013	-14.67	-15.49	-14.9	-6.34	-20.37
2014	-24.26	-15.36	-19.95	-6.72	-22.87
2015	-31.41	-16.9	-27.76	-9.91	-31.75
2016	-38.5	-25.23	-32.43	-8.18	-35.58
2017	-40.69	-25.49	-26.21	-5.82	-37.43
2018	-48.96	-27.83	-33.08	-9.53	-42.19
PreG	-8.69	-2.59	-14.77	-1.20	-10.59
PostG	-36.76	-22.16	-27.89	-8.03	-33.96

Table 4. Impact of meteorological condition changes on PM_{2.5} ($\mu\text{g}/\text{m}^3$), precipitation (mm/day), wind speed (m/s), near-surface temperature (K), and Planetary Boundary Layer (PBL) height (m) during the EASM period in PreG (2009–2013) and PostG (2014–2018) phase relative to 2008 (SIM_{Base} - SIM_{MET=2008}).

Region	Period	PM _{2.5} ($\mu\text{g}/\text{m}^3$)	Precipitation (mm/day)	Wind Speed (m/s)	Near-Surface Temperature (K)	PBL (m)
NCP	PreG	-4.01	0.58	0.17	0.32	-46.8
	PostG	-1.6	0.6	0.26	0.6	-14.5
FWP	PreG	2.32	1.68	-0.06	0.1	-108.5
	PostG	1	0.81	0.05	0.46	-15.3
YRD	PreG	-6.31	1.02	0.18	-0.29	-33.9
	PostG	-0.43	0.48	-0.08	0.45	21.9
PRD	PreG	1.49	-2.39	-0.02	0.36	29.6
	PostG	0.11	-3.24	0.18	1.00	52.2
SCB	PreG	0.29	1.81	0.13	-0.58	-136.5
	PostG	-1.14	0.37	-0.03	-0.14	-76

Table 5. Impact of CO₂ emission changes on PM_{2.5} ($\mu\text{g}/\text{m}^3$), CO₂ (ppm), precipitation (mm/day), and isoprene ($\mu\text{g}/\text{m}^3$) during the EASM period in PreG (2009–2013) and PostG (2014–2018) phase relative to 2008 (SIM_{Base} - SIM_{CO2=2008}).

Region	Period	PM _{2.5} ($\mu\text{g}/\text{m}^3$)	CO ₂ (ppm)	Precipitation (mm/day)	Isoprene ($\mu\text{g}/\text{m}^3$)
NCP	PreG	0.6	3.19	0.27	-0.1
	PostG	-1.3	4.24	0.13	0.26
FWP	PreG	0.84	1.70	0.21	-0.16
	PostG	-0.98	2.05	0.06	0.33
YRD	PreG	-0.02	4.1	0.13	-0.32
	PostG	-0.05	6.2	0.09	-0.58
PRD	PreG	1.13	1.97	-1.02	0.31
	PostG	0.31	3.20	-0.33	0.92
SCB	PreG	-0.49	2.80	0.64	-0.78
	PostG	-0.73	2.78	0.21	0.69

Table 6. Changes in total PM_{2.5} concentrations (ALL, SIM_{Base} - SIM₂₀₀₈) and the impacts of anthropogenic pollutant emissions (Emis, All-Met-CO₂), meteorological conditions (Met, SIM_{Base} - SIM_{MET=2008}), and CO₂ emission (CO₂, SIM_{Base} - SIM_{CO2=2008}) variations on PM_{2.5} concentrations ($\mu\text{g}/\text{m}^3$) during the EASM period in PreG (2009–2013) and PostG (2014–2018) phase relative to 2008.

Region	Period	ALL	Emis	Met	CO ₂
NCP	PreG	-8.69	-5.28	-4.01	0.6
	PostG	-36.76	-33.86	-1.6	-1.3
FWP	PreG	-2.59	-5.75	2.32	0.84
	PostG	-22.16	-22.18	1	-0.98
YRD	PreG	-14.77	-8.44	-6.31	-0.02
	PostG	-27.89	-27.41	-0.43	-0.05
PRD	PreG	-1.2	-3.82	1.49	1.13
	PostG	-8.03	-8.45	0.11	0.31
SCB	PreG	-10.59	-10.39	0.29	-0.49
	PostG	-33.96	-32.09	-1.14	-0.73

Other comments:

3. Section 2.3 and Table 1: Can you introduce a bit more detail of what processes CO₂ will influence in your model? Are the meteorological fields used in SIM_{Base} and SIM_{CO2=2008} the same or SIM_{Base} also reflect the meteorology change due to CO₂? In Fig 1, it seems that meteorology responses to YIBs that changes with CO₂, then how do you apply the fixed meteorological field for SIM_{MET=2008}? ignoring the response of CO₂ variation?

Response: Thanks. We added some descriptions of what processes CO₂ will influence in our model.

To clarify the experimental design, we have revised Table 1 and its description in the revised manuscript. The SIM₂₀₀₈ experiment represents the baseline conditions for the year 2008. In the SIM_{Base} experiment, interannual variations in meteorological

fields, CO₂ emissions, and anthropogenic pollutant emissions (excluding CO₂ emissions) were considered for simulations spanning 2009–2018, representing the baseline conditions for 2009–2018. Additionally, the SIM_{MET=2008} and SIM_{CO₂=2008} experiments were designed, where meteorological fields and CO₂ emissions were fixed at their 2008 levels, respectively, while simulations were conducted for 2009–2018.

As shown in Table 1, SIM₂₀₀₈ and SIM_{Base} serve as baseline experiments that collectively capture the evolution of PM_{2.5} concentrations under the combined influences of meteorological variability, CO₂ emission changes, and anthropogenic pollutant emissions changes (SIM_{Base} - SIM₂₀₀₈).

The SIM_{Base} and SIM_{CO₂=2008} experiments share identical meteorological conditions, differing only in their CO₂ emission datasets; by comparing SIM_{Base} and SIM_{CO₂=2008} (SIM_{Base} - SIM_{CO₂=2008}), we isolate the impact of CO₂ emission changes on PM_{2.5}. Likewise, since SIM_{Base} and SIM_{MET=2008} use the same CO₂ emission inputs and differ only in meteorological fields, their comparison (SIM_{Base} - SIM_{MET=2008}) quantifies the effect of meteorological variability on PM_{2.5}.

Table 1. The Numerical experimental in this study.

Experiment	Time	Meteorological fields	CO ₂ emissions	Anthropogenic pollutant emissions
SIM ₂₀₀₈	2008	2008	2008	2008
SIM _{Base}	2009-2018	2009-2018	2009-2018	2009-2018
SIM _{MET=2008}	2009-2018	2008	2009-2018	2009-2018
SIM _{CO₂=2008}	2009-2018	2009-2018	2008	2009-2018

Changes in manuscript:

2.1 Model description

(L141–148): “In the RegCM-Chem-YIBs model, changes in CO₂ concentrations affect PM_{2.5} primarily via two mechanisms: first, CO₂-induced radiative forcing alters the atmospheric radiation balance, leading to shifts in temperature, precipitation, and boundary-layer structure that modulate PM_{2.5} formation, transport, and removal(Li and Mölders, 2008; Matthews, 2007); And second, through the YIBs module, changes in CO₂ concentration modulate photosynthetic activity and stomatal behavior, altering BVOCs emissions that undergo atmospheric photochemical oxidation to form secondary organic aerosols, a significant fraction of PM_{2.5} (Kergoat et al., 2002; Kellomaki and Wang, 1998).”

2.3 Experiment settings:

(L169–175): “The numerical experiments are presented in Table 1. The SIM₂₀₀₈ experiment represents the baseline conditions for the year 2008. In the SIM_{Base} experiment, interannual variations in meteorological fields, CO₂ emissions, and anthropogenic pollutant emissions (excluding CO₂ emissions) were considered for

simulations spanning 2009–2018, representing the baseline conditions for 2009–2018. Additionally, the $SIM_{MET=2008}$ and $SIM_{CO_2=2008}$ experiments were designed, where meteorological fields and CO_2 emissions were fixed at their 2008 levels, respectively, while simulations were conducted for 2009–2018.”

(L179–188): “By comparing the simulation results from different years in the SIM_{Base} experiment to SIM_{2008} ($SIM_{Base} - SIM_{2008}$), we quantified changes in $PM_{2.5}$ concentrations relative to 2008 for the period 2009–2018. To evaluate the impact of meteorological conditions on $PM_{2.5}$ concentrations, we compared the results of the SIM_{Base} experiment with those of the $SIM_{MET=2008}$ experiment for the same year ($SIM_{Base} - SIM_{MET=2008}$). Similarly, the contribution of CO_2 emission changes to $PM_{2.5}$ variations was assessed by comparing the SIM_{Base} experiment with the $SIM_{CO_2=2008}$ experiment ($SIM_{Base} - SIM_{CO_2=2008}$) in the same year. The contribution of anthropogenic pollutant emissions was then determined by subtracting the effects of meteorological and CO_2 emission changes from the total $PM_{2.5}$ variation.”

References

- Kellomaki, S. and Wang, K. Y.: Growth, respiration and nitrogen content in needles of Scots pine exposed to elevated ozone and carbon dioxide in the field, *Environmental pollution* (Barking, Essex : 1987), 101, 263-274, 10.1016/s0269-7491(98)00036-0, 1998.
- Kergoat, L., Lafont, S., Douville, H., Berthelot, B., Dedieu, G., Planton, S., and Royer, J. F.: Impact of doubled CO_2 on global-scale leaf area index and evapotranspiration:: Conflicting stomatal conductance and LAI responses -: art. no. 4808, *Journal of Geophysical Research-Atmospheres*, 107, 10.1029/2001jd001245, 2002.
- Li, Z. and Mölders, N.: Interaction of impacts of doubling CO_2 and changing regional land-cover on evaporation, precipitation, and runoff at global and regional scales, *International Journal of Climatology*, 28, 1653-1679, 10.1002/joc.1666, 2008.
- Matthews, H. D.: Implications of CO_2 fertilization for future climate change in a coupled climate-carbon model, *Global Change Biology*, 13, 1068-1078, 10.1111/j.1365-2486.2007.01343.x, 2007.

4. line 148: as the impact of meteorological conditions is calculated by $SIM_{base} - SIM_{met=2008}$, it is likely to also include influences of CO_2 .

Response: Thanks. SIM_{Base} and $SIM_{MET=2008}$ use the same CO_2 emission inputs and differ only in meteorological fields, their comparison ($SIM_{Base} - SIM_{MET=2008}$) quantifies the effect of meteorological variability on $PM_{2.5}$. Please refer to comment 3 for a detailed response.

5. line 154-158: it would be better to bring some information of model evaluation to the text instead of letting the audience check all the information in other references. e.g., you might also show numbers for measured $PM_{2.5}$ trend when discussing the simulation results in Section 3.1, or include figures to compare with observations in

supplements.

Response: Thank you for your invaluable suggestions. We have incorporated the model evaluation results into the manuscript and expanded the corresponding descriptions.

Changes in manuscript:

2.4 Model evaluations

(L198–207): “Observed PM_{2.5} data were obtained from the China National Environmental Monitoring Center (CNEMC). This study used hourly PM_{2.5} concentrations during the summer monsoon period (May 1 to August 31) from 2015 to 2018. A total of 366 monitoring stations across Chinese cities, selected based on data completeness and representativeness, were used for model validation. The locations of these stations are shown in Fig. S5. CO₂ observations were sourced from the World Data Centre for Greenhouse Gases (WDCGG), including all seven sites in East Asia: Waliguan, Korea Tae-ahn Peninsula, Ulaanbaatar in Mongolia, Lulin, Yonagunijima, Cape D'Aguilar (Hong Kong), and King's Park. Detailed station locations are shown in Fig. S6. Reanalysis data for temperature, wind fields, and relative humidity were obtained from the ERA-Interim dataset.”

(L208–21): “As shown in Table 2 and Figures S1–S6, the SIM_{Base} experiments reproduce 2015–2018 PM_{2.5} and CO₂ concentrations with high correlations and low biases relative to observations, while their simulated meteorological fields closely match reanalysis data. Overall, the RegCM-Chem-YIBs model effectively captures the fundamental characteristics and temporal trends of meteorological factors, PM_{2.5}, and CO₂ concentrations in East Asia.”

Table 2. Evaluations of the near-surface CO₂ and PM_{2.5} in East Asia.

Species	Year	Observation	Simulation	Bias	RMSE	R
CO ₂ (ppm)	2015	402.82	406.98	4.16	9.37	0.44
	2016	407.12	410.44	3.32	8.22	0.69
	2017	408.35	413.62	5.27	11	0.39
	2018	409.61	416.68	7.07	11.32	0.41
PM _{2.5} (ug/m ³)	2015	36.6	25.57	-11.03	12.99	0.71
	2016	31.03	22.91	-8.12	10.31	0.64
	2017	29.61	24.02	-5.59	10.57	0.71
	2018	27.18	19.04	-8.14	11.62	0.61

RMSE: root mean square error; R: correlation coefficient.

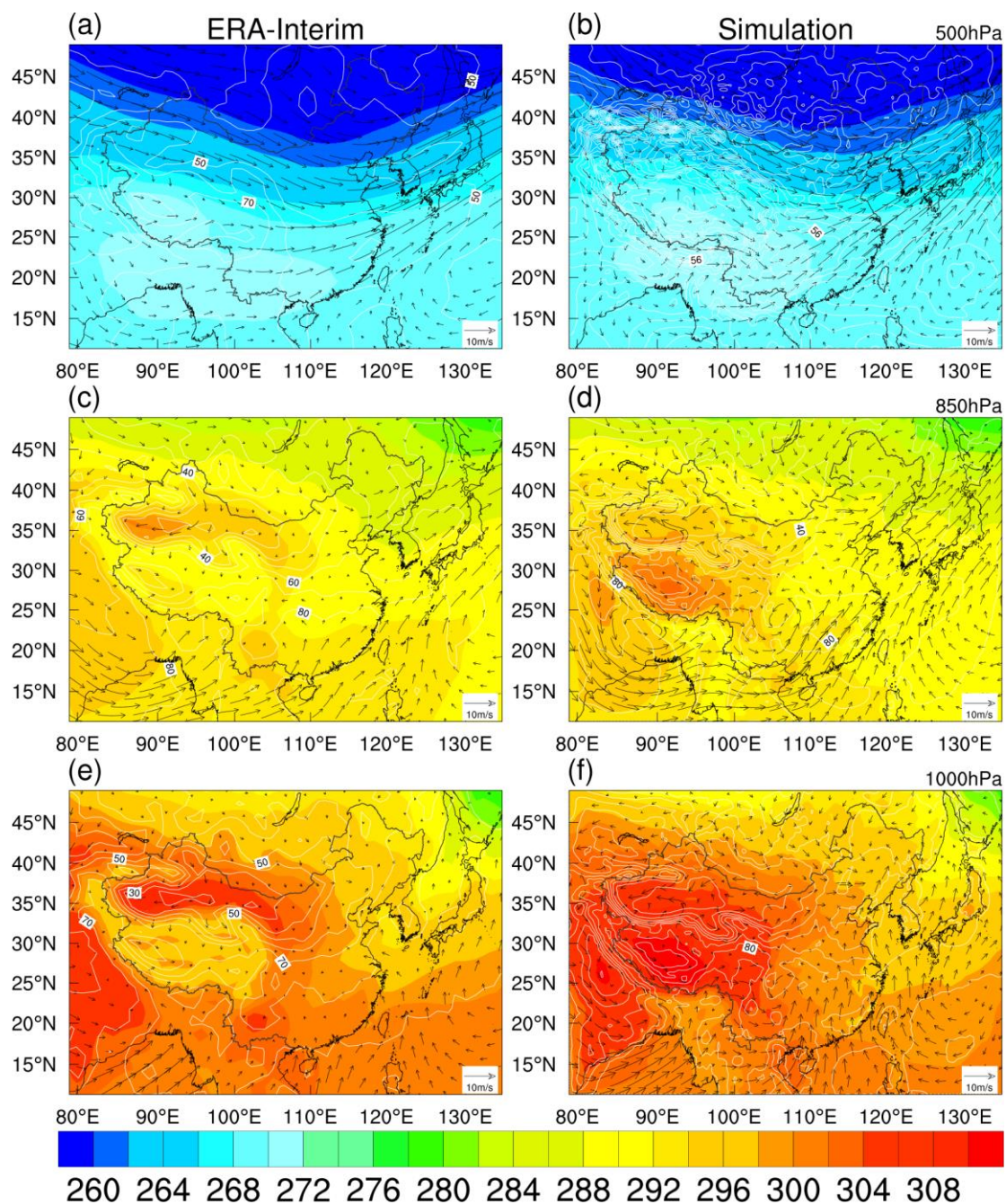


Figure S1. Comparisons between the simulated (right) and reanalysis (left) mean temperature (shading, units: K), wind (vectors, units: m/s), and relative humidity (contours, units: %) at 500 hPa (a, b), 850 hPa (c, d) and 1000 hPa (e, f) during the EASM period in 2015.

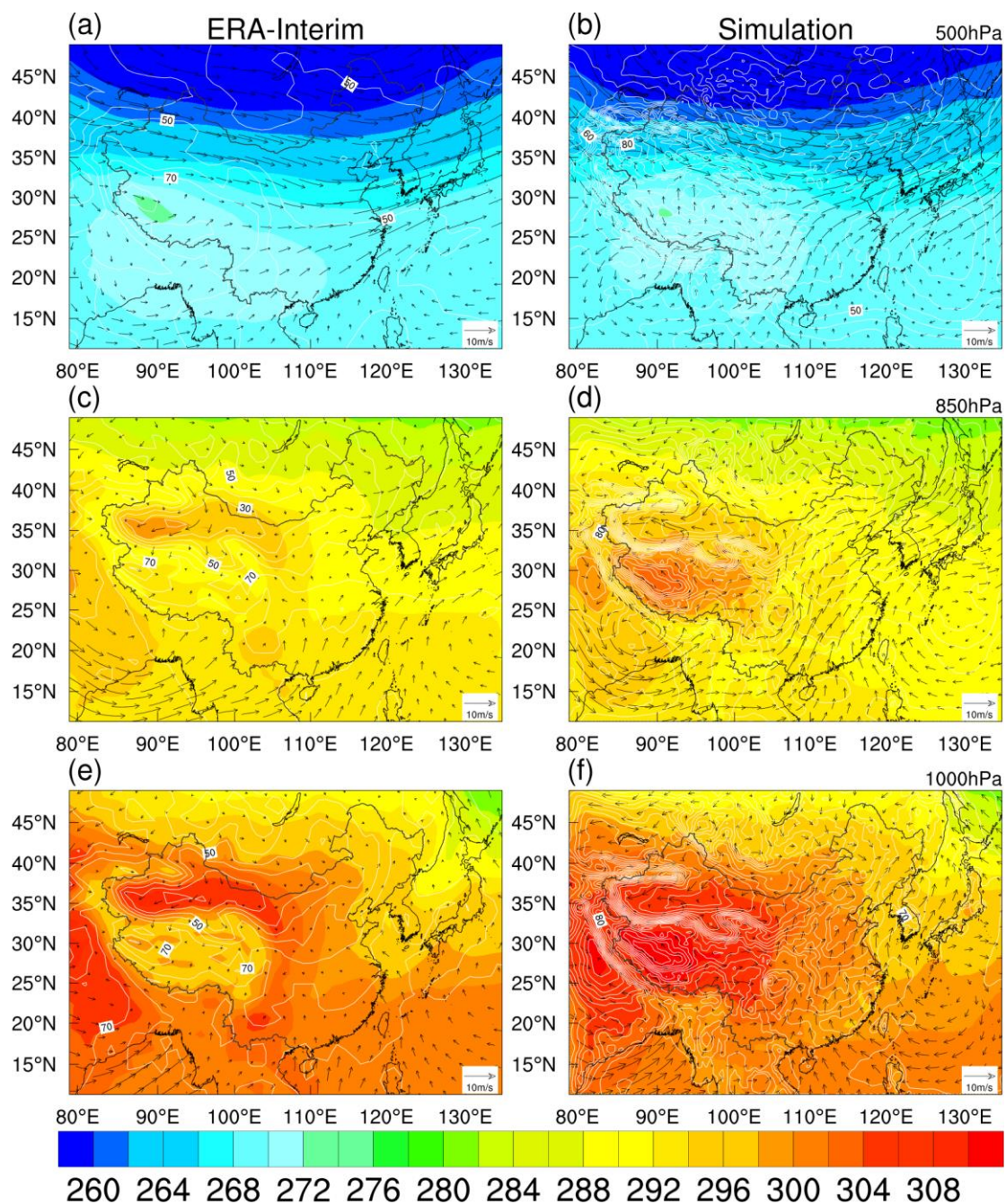


Figure S2. Comparisons between the simulated (right) and reanalysis (left) mean temperature (shading, units: K), wind (vectors, units: m/s), and relative humidity (contours, units: %) at 500 hPa (a, b), 850 hPa (c, d) and 1000 hPa (e, f) during the EASM period in 2016.

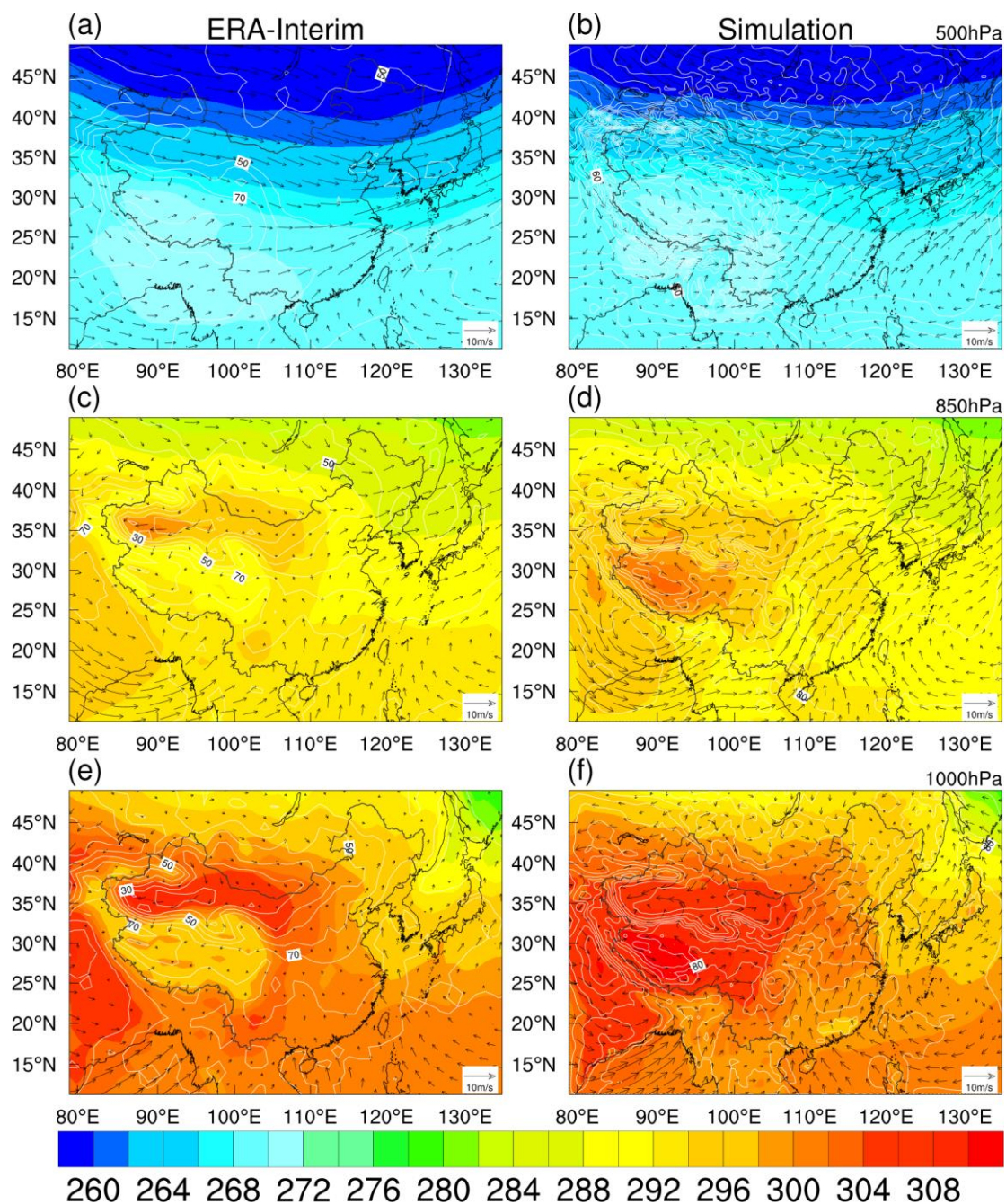


Figure S3. Comparisons between the simulated (right) and reanalysis (left) mean temperature (shading, units: K), wind (vectors, units: m/s), and relative humidity (contours, units: %) at 500 hPa (a, b), 850 hPa (c, d) and 1000 hPa (e, f) during the EASM period in 2017.

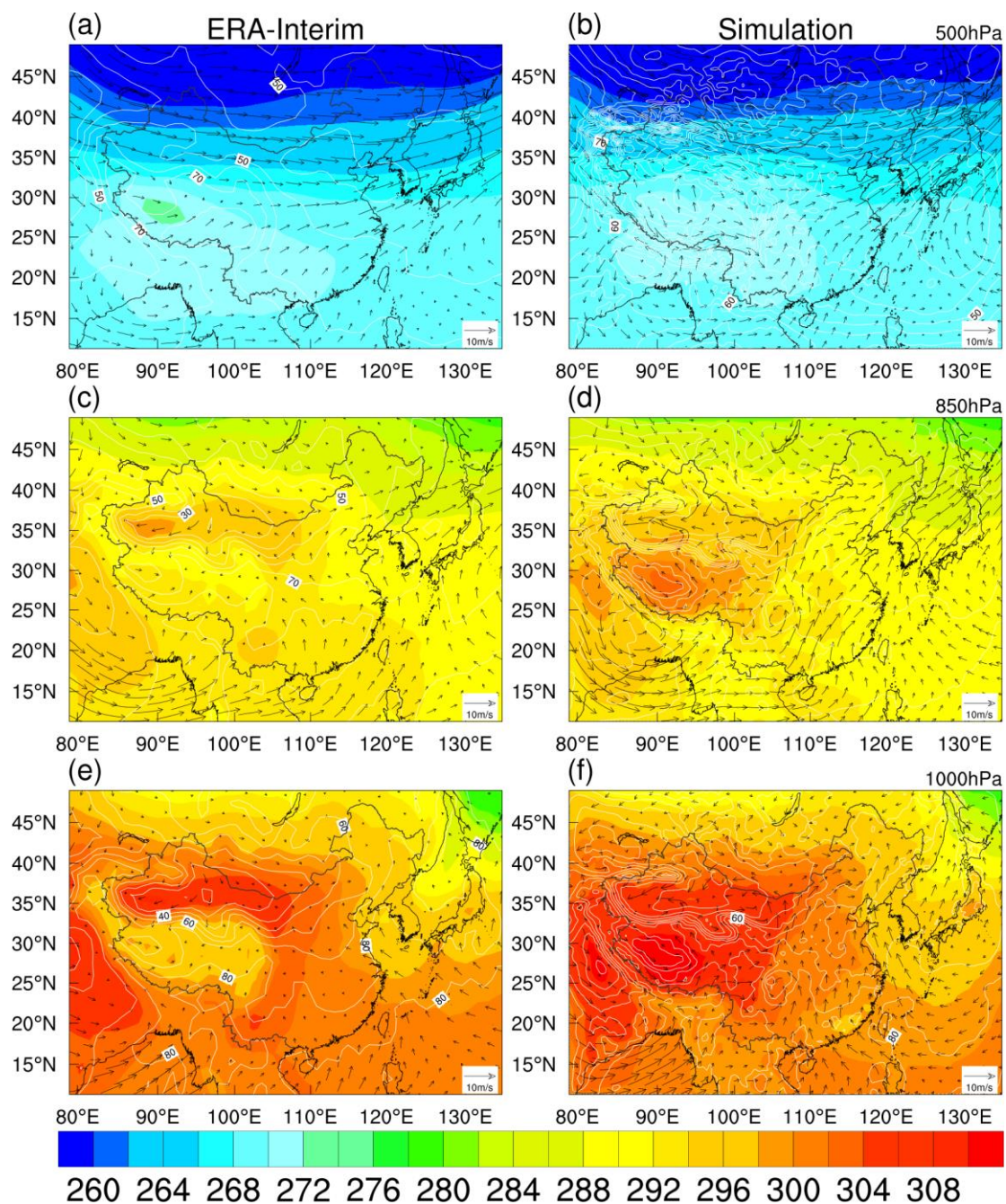


Figure S4. Comparisons between the simulated (right) and reanalysis (left) mean temperature (shading, units: K), wind (vectors, units: m/s), and relative humidity (contours, units: %) at 500 hPa (a, b), 850 hPa (c, d) and 1000 hPa (e, f) during the EASM period in 2018.

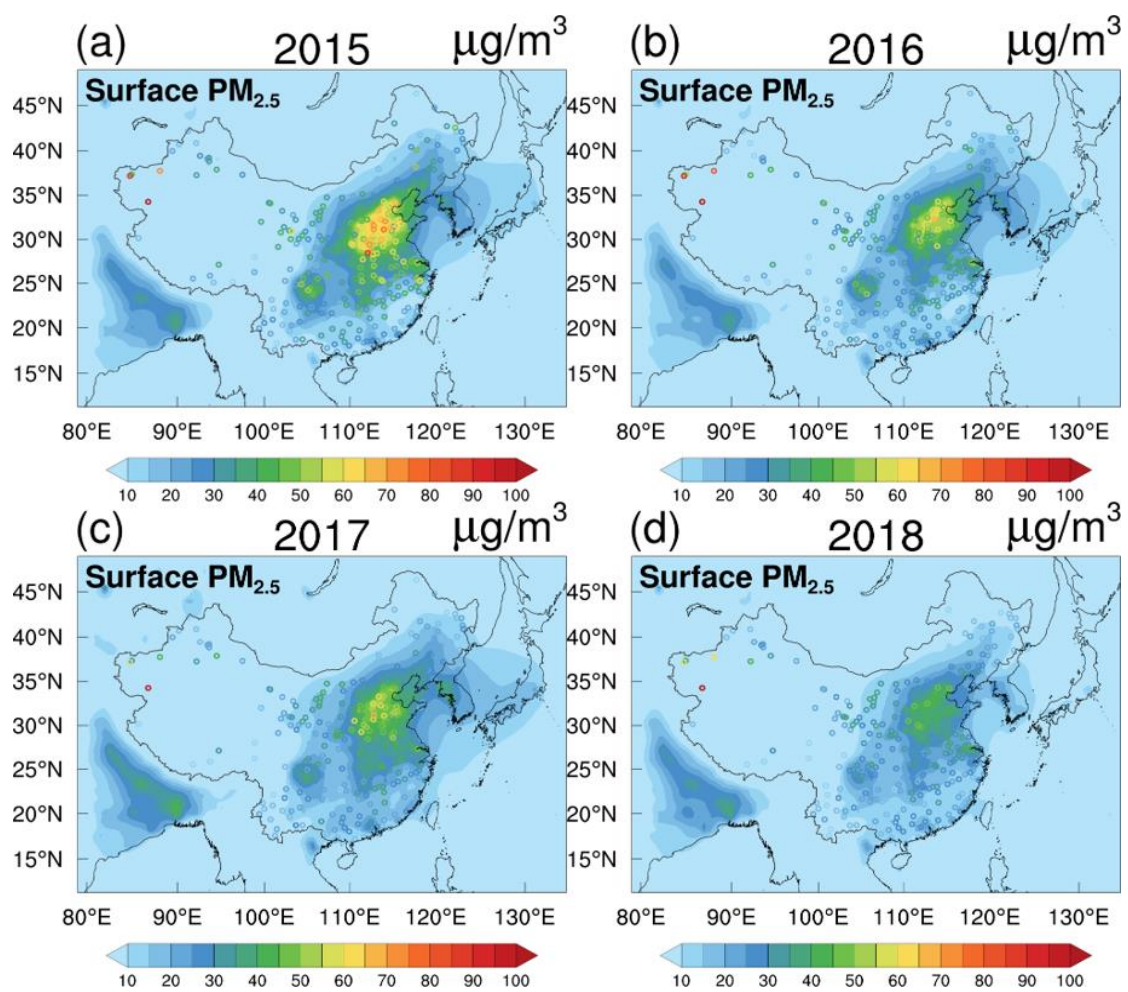


Figure S5. Comparisons between the simulated and observed near-surface PM_{2.5} concentrations (units: $\mu\text{g}/\text{m}^3$) during the EASM period in (a)2015, (b)2016, (c)2017, (d)2018. Colored circles represent the observations.

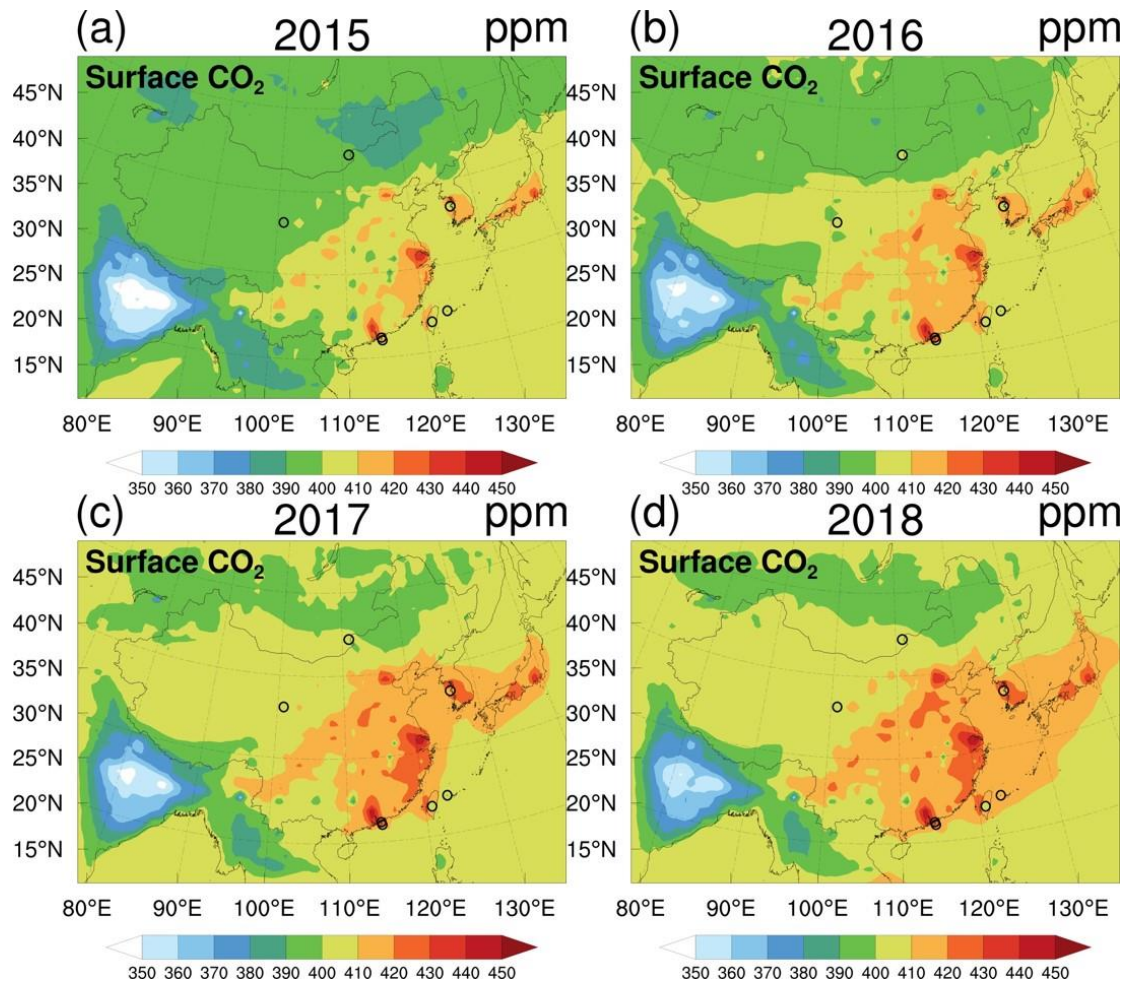


Figure S6. Comparisons between the simulated and observed near-surface CO₂ concentrations (units: ppm) during the EASM period in (a)2015, (b)2016, (c)2017, (d)2018. Colored circles represent the observations.

6. The wording of “anthropogenic emissions” driver in many places of the manuscript might need to be clearer. One key question is whether it is also the anthropogenic emissions contributing to the CO₂ changes. If so, the “anthropogenic emissions” in the text should means non-CO₂ emissions.

Response: Thank you for highlighting this critical issue. In response, we have revised the manuscript to replace all instances of “anthropogenic emissions” with “anthropogenic pollutant emissions”. Additionally, in Section 2.3 Experiment settings, we have clarified that “anthropogenic pollutant emissions” exclude CO₂ emissions.

2.3 Experiment settings:

(L170–173): “In the SIM_{Base} experiment, interannual variations in meteorological fields, CO₂ emissions, and anthropogenic pollutant emissions (excluding CO₂ emissions) were considered for simulations spanning 2009–2018, representing the baseline conditions for 2009–2018.”

7. Section 3.3: Will CO₂ also change temperature and cloud due to its effects on

radiation balance? Are those negligible factors comparing to those shown in Fig 5?

Response: Thanks. CO₂ alters atmospheric radiative properties, thereby influencing meteorological factors such as temperature, cloud cover, and precipitation. Our analysis of the relationships between temperature, cloud cover, and PM_{2.5} concentrations indicates that their direct effects are insignificant. Specifically, cloud cover affects PM_{2.5} primarily through indirect mechanisms, including modulation of solar radiation and changes in planetary boundary layer height. Temperature influences PM_{2.5} via multiple complex pathways, such as regulating secondary organic aerosol formation, vertical convection, and boundary layer dynamics. In contrast, precipitation directly removes PM_{2.5} through wet deposition processes. Therefore, the primary pathways through which CO₂ impacts PM_{2.5} concentrations are its modulation of precipitation patterns and its influence on biogenic volatile organic compound (BVOC) emissions from vegetation.

Response to Reviewers

No.: ACP-2025-10

Title: Anthropogenic and Natural Causes for the Interannual Variation of PM_{2.5} in East Asia During Summer Monsoon Periods From 2008 to 2018

Anonymous referee #1:

This study presents the results of simulations performed with the RegCM-Chem-YIBs Model over the period 2008-2018. The study focuses on PM_{2.5} concentrations over China and explores the drivers of change in simulated concentrations before and after the implementation of a Clean Air Action Plan in 2013. This is an interesting study and certainly within the scope of ACP.

Response: We thank Referee #1 for his/her valuable comments, which have greatly improved our manuscript. We have attempted to make a revision addressing each of the points mentioned in his/her review. Please note that the line numbers given below refer to the clean version of the manuscript.

1. Whilst the paper presents an interesting set of experiments, it would be strengthened further if the authors could comment on the extent to which this modelling framework captures the temporal and spatial variability of observed PM_{2.5} across China during the specific time period concerned. There are many studies that have analysed and reported measured PM_{2.5} concentrations, several of which are already cited in your Introduction, and could be used to offer a comparison. Others include:

Silver et al., 2018, Environ. Res. Lett. 13 114012

Ma et al, 2019, Atmos. Chem. Phys., 19, 6861–6877

Kong et al., 2021, Earth Syst. Sci. Data, 13, 529–570

Silver et al., 2025, Environment International, 197, 109318

Response: Thank you for your valuable suggestions. We have added a comparative analysis with existing studies on PM_{2.5} concentration changes in China in Section 3.1 “PM_{2.5} variation”. The results show that the simulated PM_{2.5} trends from 2008 to 2018 in this study are highly consistent with most previous findings, further validating the reliability of our simulation results.

Changes in manuscript:

3.1 PM_{2.5} variation

(L259–270): “Table S1 shows that the mean PM_{2.5} trend over China during the PreG (2009-2013) and PostG (2014-2018) periods was $-1.84 \mu\text{g}/\text{m}^3/\text{yr}$ and $-2.90 \mu\text{g}/\text{m}^3/\text{yr}$, respectively. These values are consistent with the findings of Silver et al. (2025), who reported a PM_{2.5} trend of $-2.47 \mu\text{g}/\text{m}^3/\text{yr}$ for 2014–2017 in China based on ground-based observations. Similarly, Lin et al. (2018) reported PM_{2.5} trends of -0.65 and $-2.30 \mu\text{g}/\text{m}^3/\text{yr}$ for 2006–2010 and 2011–2015 in China, respectively. Using satellite remote sensing data, Ma et al. (2019) found declines of 1.03 and 4.27

$\mu\text{g}/\text{m}^3/\text{yr}$ for 2010-2013 and 2013-2017 in China, respectively. The high-resolution Chinese air quality reanalysis (CAQRA), developed by Kong et al. (2021) using data assimilation techniques, indicated a more pronounced decline of $-5.80 \mu\text{g}/\text{m}^3/\text{yr}$ for $\text{PM}_{2.5}$ from 2013 to 2018 in China. In addition, Silver et al. (2018), based on multi-source data, reported a trend of $-3.40 \mu\text{g}/\text{m}^3/\text{yr}$ for 2015–2017 in China. Therefore, our simulation accurately captures the observed $\text{PM}_{2.5}$ trends over China from 2008 to 2018, providing a robust foundation for subsequent attribution analyses.”

Table S1. Interannual trends of near-surface $\text{PM}_{2.5}$ concentrations ($\mu\text{g}/\text{m}^3/\text{year}$) during the PreG period (2009–2013) and PostG period (2014–2018) relative to 2008 over the NCP, FWP, YRD, PRD, and SCB regions.

Year	NCP	FWP	YRD	PRD	SCB	Average
PreG	-0.69	-2.84	-0.71	-1.55	-3.44	-1.84
PostG	-4.94	-2.49	-2.63	-0.56	-3.86	-2.90

References

Kong, L., Tang, X., Zhu, J., Wang, Z., Li, J., Wu, H., Wu, Q., Chen, H., Zhu, L., Wang, W., Liu, B., Wang, Q., Chen, D., Pan, Y., Song, T., Li, F., Zheng, H., Jia, G., Lu, M., Wu, L., and Carmichael, G. R.: A 6-year-long (2013-2018) high-resolution air quality reanalysis dataset in China based on the assimilation of surface observations from CNEMC, *Earth System Science Data*, 13, 529-570, 10.5194/essd-13-529-2021, 2021.

Lin, C. Q., Liu, G., Lau, A. K. H., Li, Y., Li, C. C., Fung, J. C. H., and Lao, X. Q.: High-resolution satellite remote sensing of provincial $\text{PM}_{2.5}$ trends in China from 2001 to 2015, *Atmos Environ*, 180, 110-116, 10.1016/j.atmosenv.2018.02.045, 2018.

Ma, Z., Liu, R., Liu, Y., and Bi, J.: Effects of air pollution control policies on $\text{PM}_{2.5}$ pollution improvement in China from 2005 to 2017: a satellite-based perspective, *Atmospheric Chemistry and Physics*, 19, 6861-6877, 10.5194/acp-19-6861-2019, 2019.

Silver, B., Reddington, C. L., Arnold, S. R., and Spracklen, D. V.: Substantial changes in air pollution across China during 2015-2017, *Environmental Research Letters*, 13, 10.1088/1748-9326/aae718, 2018.

Silver, B., Reddington, C. L., Chen, Y., and Arnold, S. R.: A decade of China's air quality monitoring data suggests health impacts are no longer declining, *Environment International*, 197, 10.1016/j.envint.2025.109318, 2025.

Specific comments:

2. Line 11: This sentence doesn’t quite make sense - remove word “changes”.

Response: Thanks. We have removed the word “changes” in the revised version.

Changes in manuscript:

Abstract

(L11–12): “There was a significant difference in near-surface PM_{2.5} across China after the implementation of the Clean Air Action Plan in 2013.”

3. Line 16: You haven’t defined PreG or PostG yet so it’s a bit confusing to mention these here. You could either define them or describe the time periods without referring to them by these names.

Response: Thanks. We have revised the expressions and clearly defined PreG and PostG in the updated manuscript.

Changes in manuscript:

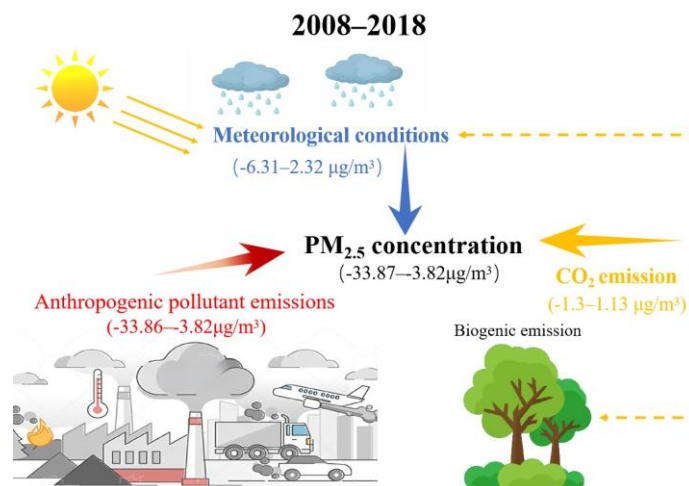
Abstract

(L15–18): “Compared to 2008, PM_{2.5} showed little variation during the PreG phase (2009–2013). However, during the PostG phase (2014–2018), a substantial decline in PM_{2.5} was simulated, particularly in the North China Plain ($-36.76 \mu\text{g}/\text{m}^3$) and the Sichuan Basin ($-33.96 \mu\text{g}/\text{m}^3$).”

4. Graphical abstract: At the moment the diagram is slightly confusing and it ideally needs to be entirely self-explanatory since the graphical abstracts do not come with a caption. It’s also not clear what “~” is being used to represent here: are these ranges?

Response: Thanks for pointing that out. We have replaced the tilde (~) with a hyphen (–) to indicate ranges. Additionally, the Graphical abstract has been optimized to enhance clarity and make it more self-explanatory.

Changes in manuscript:



5. Lines 31–38: You refer to PM_{2.5} as if it is a single entity, whereas in reality it’s an aggregation of part of the aerosol size distribution, and could be comprised of many different components (this is most relevant to your description of the impact of PM_{2.5} on climate) - I suggest rewording this paragraph to reflect this.

Response: Thanks for pointing out this issue. We have added a more detailed

description of PM_{2.5} in the revised manuscript.

Changes in manuscript:

Introduction

(L32–36): “PM_{2.5} refers to fine particulate matter with an aerodynamic diameter less than or equal to 2.5 micrometers (Chen et al., 2018). Its sources include industrial emissions, vehicular exhaust, biomass burning, and secondary formation from atmospheric gases (Wu et al., 2020). Major chemical components of PM_{2.5} include sulfates, nitrates, ammonium salts, organic carbon, elemental carbon, and heavy metals (Van Donkelaar et al., 2019; Li et al., 2017a).”

References

- Chen, G. B., Li, S. S., Knibbs, L. D., Hamm, N. A. S., Cao, W., Li, T. T., Guo, J. P., Ren, H. Y., Abramson, M. J., and Guo, Y. M.: A machine learning method to estimate PM_{2.5} concentrations across China with remote sensing, meteorological and land use information, *Sci Total Environ*, 636, 52-60, 10.1016/j.scitotenv.2018.04.251, 2018.
- Li, G. H., Bei, N. F., Cao, J. J., Huang, R. J., Wu, J. R., Feng, T., Wang, Y. C., Liu, S. X., Zhang, Q., Tie, X. X., and Molina, L. T.: A possible pathway for rapid growth of sulfate during haze days in China, *Atmospheric Chemistry and Physics*, 17, 3301-3316, 10.5194/acp-17-3301-2017, 2017a.
- van Donkelaar, A., Martin, R. V., Li, C., and Burnett, R. T.: Regional Estimates of Chemical Composition of Fine Particulate Matter Using a Combined Geoscience-Statistical Method with Information from Satellites, Models, and Monitors, *Environmental Science & Technology*, 53, 2595-2611, 10.1021/acs.est.8b06392, 2019.
- Wu, K., Yang, X. Y., Chen, D., Gu, S., Lu, Y. Q., Jiang, Q., Wang, K., Ou, Y. H., Qian, Y., Shao, P., and Lu, S. H.: Estimation of biogenic VOC emissions and their corresponding impact on ozone and secondary organic aerosol formation in China, *Atmospheric Research*, 231, 10.1016/j.atmosres.2019.104656, 2020.

6. Lines 42-46: Reword this slightly to clarify whether these are all annual average values, as written it sounds as though some of them could be the maximum value recorded.

Response: Thank you for pointing out this issue. We have slightly revised the wording in the manuscript to clarify that these values represent averages.

Changes in manuscript:

Introduction

(L48–52): “From 2000 to 2008, the national average PM_{2.5} concentration in China was $49.4 \pm 14.2 \mu\text{g}/\text{m}^3$. In eastern China, the average concentration was $55.4 \pm 16.1 \mu\text{g}/\text{m}^3$, while the Beijing-Tianjin-Hebei region experienced average levels as high as $62.1 \pm 22.5 \mu\text{g}/\text{m}^3$. The Yangtze River Delta saw an average concentration of $63.0 \pm 11.1 \mu\text{g}/\text{m}^3$, the Pearl River Delta recorded an average of $52.4 \pm 5.8 \mu\text{g}/\text{m}^3$, and

the Sichuan Basin averaged $61.6 \pm 13.4 \mu\text{g}/\text{m}^3$.

7. Lines 74-81: clarify here that as well as affecting photosynthesis, elevated CO₂ concentrations can directly inhibit the emission of isoprene. Temperature is mentioned in the previous section in terms of its impact on chemical reactions, but changes in temperature will also drive changes to BVOC emissions (and the partitioning of BVOC oxidation products from the gas to particle phase) so this could be mentioned in the Introduction too.

Response: Thanks. We have added some discussions on this aspect.

Changes in manuscript:

Introduction

(L68–74): “In addition, moderate increases in temperature can significantly enhance the emissions of biogenic volatile organic compounds (BVOCs) by stimulating the activity of the synthase enzyme. However, when temperatures exceed the physiological tolerance threshold of plants, decreased enzyme activity or metabolic disruption may suppress emissions(Lindwall et al., 2016; Kleist et al., 2012). Therefore, temperature changes can influence atmospheric PM_{2.5} concentrations by modulating the emissions of BVOCs.”

(L89–94): “It is worth noting that elevated CO₂ concentrations may also directly inhibit BVOCs emissions by reducing the activity of BVOCs synthase enzymes(Heald et al., 2009; Pegoraro et al., 2004). Therefore, the impact of increased CO₂ on vegetation BVOCs emissions can be either positive or negative, depending primarily on the relative strength of the inhibitory effect from enzyme suppression versus the stimulatory effect from enhanced photosynthesis(Sun et al., 2012).”

References

- Heald, C. L., Wilkinson, M. J., Monson, R. K., Alo, C. A., Wang, G. L., and Guenther, A.: Response of isoprene emission to ambient CO₂ changes and implications for global budgets, *Global Change Biology*, 15, 1127-1140, 10.1111/j.1365-2486.2008.01802.x, 2009.
- Kleist, E., Mentel, T. F., Andres, S., Bohne, A., Folkers, A., Kiendler-Scharr, A., Rudich, Y., Springer, M., Tillmann, R., and Wildt, J.: Irreversible impacts of heat on the emissions of monoterpenes, sesquiterpenes, phenolic BVOC and green leaf volatiles from several tree species, *Biogeosciences*, 9, 5111-5123, 10.5194/bg-9-5111-2012, 2012
- Lindwall, F., Schollert, M., Michelsen, A., Blok, D., and Rinnan, R.: Fourfold higher tundra volatile emissions due to arctic summer warming, *Journal of Geophysical Research-Biogeosciences*, 121, 895-902, 10.1002/2015jg003295, 2016.
- Pegoraro, E., Rey, A., Bobich, E. G., Barron-Gafford, G., Grieve, K. A., Malhi, Y., and Murthy, R.: Effect of elevated CO₂ concentration and vapour pressure deficit on isoprene emission from leaves of *Populus deltoides* during drought, *Functional Plant Biology*, 31, 1137-1147, 10.1071/fp04142, 2004.

Sun, Z. H., Niinemets, Ü., Hüve, K., Noe, S. M., Rasulov, B., Copolovici, L., and Vislap, V.: Enhanced isoprene emission capacity and altered light responsiveness in aspen grown under elevated atmospheric CO₂ concentration, *Global Change Biology*, 18, 3423-3440, 10.1111/j.1365-2486.2012.02789.x, 2012.

8. Lines 82-85: specify here that you are referring to China as the same may not be true for other regions.

Response: Thanks. We have now specified that the region in question is China.

Changes in manuscript:

Introduction

(L97–99): “Numerous studies have used statistical models and numerical simulations to investigate the impacts of meteorological conditions and anthropogenic pollution emissions on PM_{2.5} concentration changes in China.”

9. Line 168: can you be more specific than “more favourable meteorological conditions”? What are the main differences in meteorology in this region that lead to lower PM_{2.5} concentrations?

Response: Thanks. We have included an explanation of “more favourable meteorological conditions” in the revised manuscript.

Changes in manuscript:

3.1 PM_{2.5} variation

(L226–230): “In contrast, regions in western China (Yunnan, Gansu, Xinjiang) exhibit lower PM_{2.5} levels due to limited industrial activity, lower population density, and more favorable meteorological conditions (Low water vapor content, lower temperatures, and weak solar radiation are unfavorable for the formation of secondary aerosols such as sulfates, nitrates, and organic aerosols) (Wei et al., 2021; Xue et al., 2020).

References

- Wei, J., Li, Z. Q., Lyapustin, A., Sun, L., Peng, Y. R., Xue, W. H., Su, T. N., and Cribb, M.: Reconstructing 1-km-resolution high-quality PM_{2.5} data records from 2000 to 2018 in China: spatiotemporal variations and policy implications, *Remote Sens Environ*, 252, ARTN 11213610.1016/j.rse.2020.112136, 2021.
- Xue, W. H., Zhang, J., Zhong, C., Ji, D. Y., and Huang, W.: Satellite-derived spatiotemporal PM_{2.5} concentrations and variations from 2006 to 2017 in China, *Sci Total Environ*, 712, 10.1016/j.scitotenv.2019.134577, 2020

10. Line 105: Model Description - it would be useful to include some details around how the model calculates BVOC emissions (since this process is important to your results) - specifically, how are these emissions affected by CO₂ concentration; I don't think this is covered in your previous paper (Ma et al 2023a).

Response: Thanks. We have added some discussions on this aspect.

Changes in manuscript:

2.1 Model description

(L129–135): “The YIBs model employs a leaf-level BVOC emission scheme based on vegetation photosynthesis. Unlike the traditional MEGAN (Model of Emissions of Gases and Aerosols from Nature) model, this approach incorporates the influence of plant photosynthesis on BVOC emissions, making it more representative of actual plant physiological processes. In this scheme, leaf-level BVOC emission rates depend on the photosynthetic rate, leaf surface temperature, and intracellular CO₂ concentration (Yue and Unger, 2015; Lei et al., 2020; Yue et al., 2015).

(L141–148): “In the RegCM-Chem-YIBs model, changes in CO₂ concentrations affect PM_{2.5} primarily via two mechanisms: first, CO₂-induced radiative forcing alters the atmospheric radiation balance, leading to shifts in temperature, precipitation, and boundary-layer structure that modulate PM_{2.5} formation, transport, and removal (Li and Mölders, 2008; Matthews, 2007); And second, through the YIBs module, changes in CO₂ concentration modulate photosynthetic activity and stomatal behavior, altering BVOCs emissions that undergo atmospheric photochemical oxidation to form secondary organic aerosols, a significant fraction of PM_{2.5} (Kergoat et al., 2002; Kellomaki and Wang, 1998).”

References

- Kellomaki, S. and Wang, K. Y.: Growth, respiration and nitrogen content in needles of Scots pine exposed to elevated ozone and carbon dioxide in the field, *Environmental pollution* (Barking, Essex : 1987), 101, 263-274, 10.1016/s0269-7491(98)00036-0, 1998.
- Kergoat, L., Lafont, S., Douville, H., Berthelot, B., Dedieu, G., Planton, S., and Royer, J. F.: Impact of doubled CO₂ on global-scale leaf area index and evapotranspiration:: Conflicting stomatal conductance and LAI responses -: art. no. 4808, *Journal of Geophysical Research-Atmospheres*, 107, 10.1029/2001jd001245, 2002.
- Lei, Y. D., Yue, X., Liao, H., Gong, C., and Zhang, L.: Implementation of Yale Interactive terrestrial Biosphere model v1.0 into GEOS-Chem v12.0.0: a tool for biosphere-chemistry interactions, *Geoscientific Model Development*, 13, 1137-1153, 10.5194/gmd-13-1137-2020, 2020.
- Li, Z. and Mölders, N.: Interaction of impacts of doubling CO₂ and changing regional land-cover on evaporation, precipitation, and runoff at global and regional scales, *International Journal of Climatology*, 28, 1653-1679, 10.1002/joc.1666, 2008.
- Matthews, H. D.: Implications of CO₂ fertilization for future climate change in a coupled climate-carbon model, *Global Change Biology*, 13, 1068-1078, 10.1111/j.1365-2486.2007.01343.x, 2007.
- Yue, X. and Unger, N.: The Yale Interactive terrestrial Biosphere model version 1.0: description, evaluation and implementation into NASA GISS ModelE2, *Geoscientific Model Development*, 8, 2399-2417, 10.5194/gmd-8-2399-2015,

2015.

Yue, X., Unger, N., and Zheng, Y.: Distinguishing the drivers of trends in land carbon fluxes and plant volatile emissions over the past 3 decades, *Atmospheric Chemistry and Physics*, 15, 11931-11948, 10.5194/acp-15-11931-2015, 2015.

11. Figure 1: it would be better if the words in each box weren't split across lines, as some of them currently are. This could be solved by making some of the boxes slightly larger.

Response: Thank you for your valuable suggestion. We have enlarged the boxes to prevent word wrapping.

Changes in manuscript:

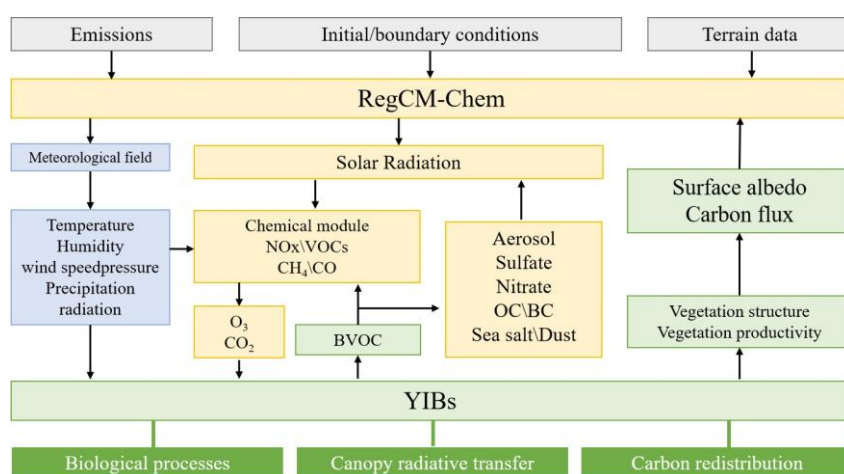


Figure 1. Framework of the RegCM-Chem-YIBs Model.

12. Figure 3: Correct the units on the legend within the figure.

Response: Thanks. We have corrected the units on the legend within the figure.

Changes in manuscript:

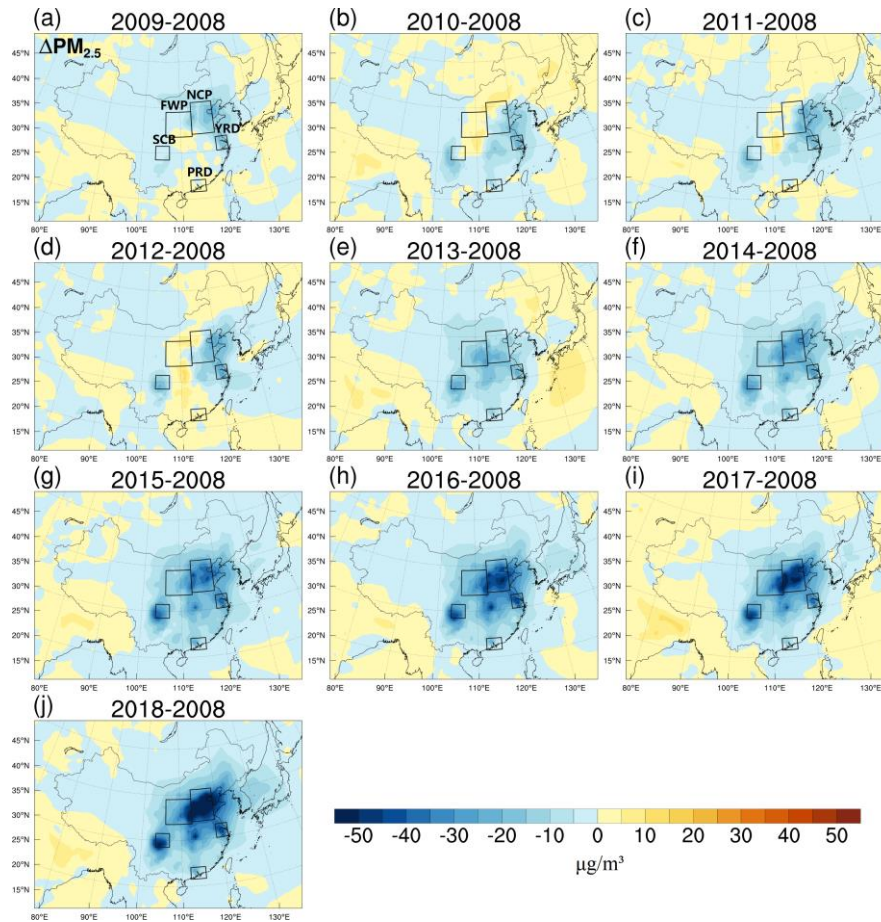


Figure 3. Changes in near-surface PM_{2.5} concentrations ($\mu\text{g}/\text{m}^3$) during the EASM period from 2009 (a) to 2018 (j) relative to 2008 in East Asia ($\text{SIM}_{\text{Base}} - \text{SIM}_{2008}$).

13. Line 207: In Section 3.2 it's not completely clear which time periods the values you report are referring to, i.e., for the PreG period, are these the changes between 2008 and 2013? Or is it the average of 2008-2013 minus 2008. I think this is confused by Figure 4 where it's not clear which time period the central and right-hand columns refer to. Some Figure captions specify May to August but add this to the others that don't.

Response: Thanks. Sorry for the mistake. "PreG-2008" represents the average of 2008-2013 minus 2008. We have revised the corresponding figure titles accordingly and replaced "May–August" with "EASM" throughout.

Changes in manuscript:

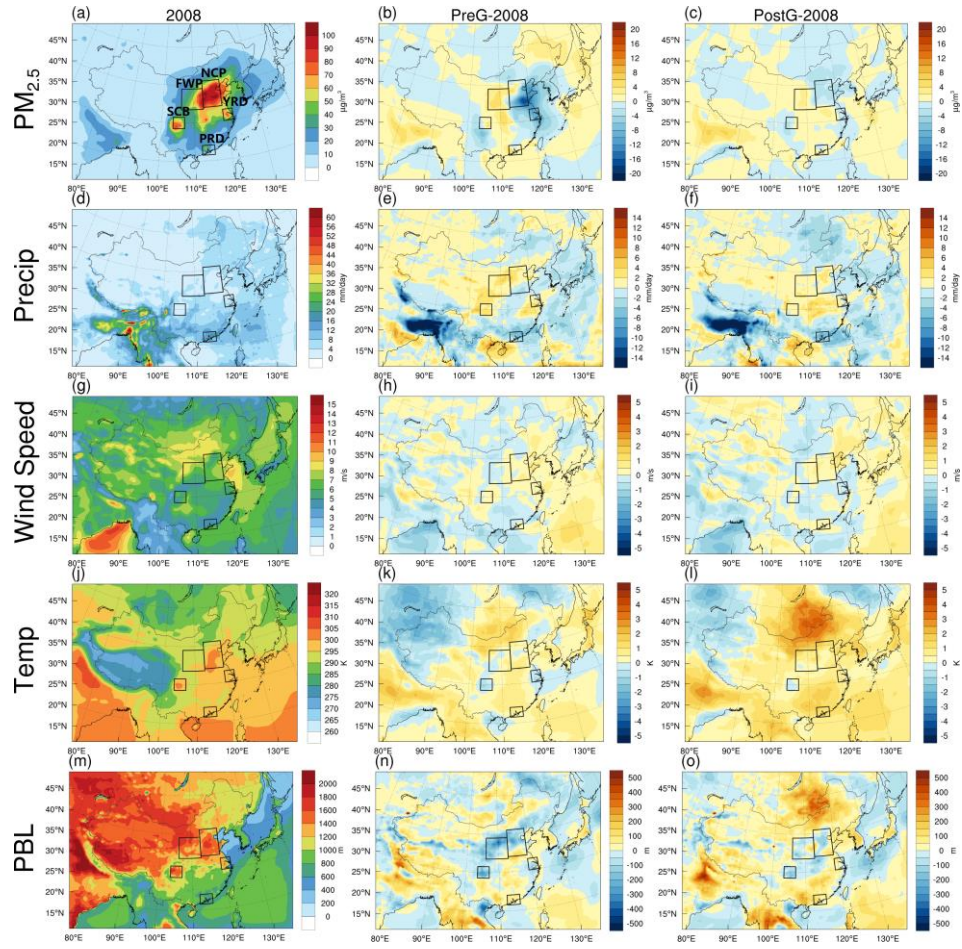


Figure 4. The PM_{2.5} (a–c, $\mu\text{g}/\text{m}^3$), precipitation (d–f, mm/day), wind speed (g–i, m/s), temperature (j–l, K), and Planetary Boundary Layer (PBL) height (m–o, m) during the EASM period in 2008 (left), and their mean changes due to meteorological variations in PreG (2009–2013, center) and PostG (2014–2018, right) phase relative to 2008 (SIM_{Base} - SIM_{MET=2008}).

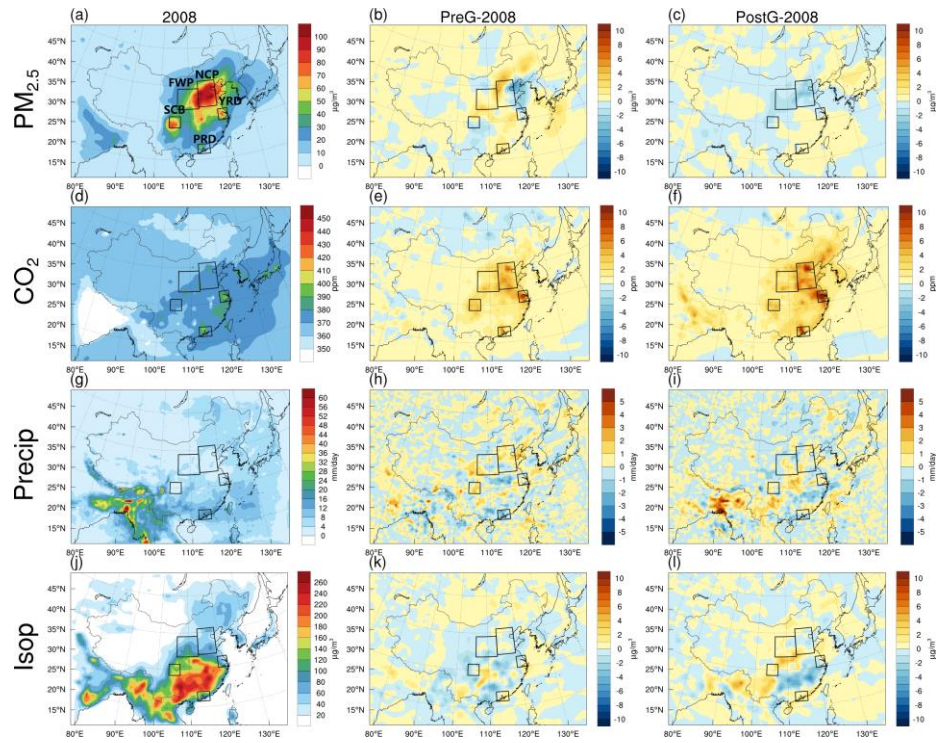


Figure 5. The $\text{PM}_{2.5}$ (a–c, $\mu\text{g}/\text{m}^3$), CO_2 (d–f, ppm), precipitation (g–i, mm/day), and isoprene (j–l, $\mu\text{g}/\text{m}^3$) during the EASM period in 2008 (left), and their mean changes due to CO_2 emission variations in PreG (2009–2013, center) and PostG (2014–2018, right) phase relative to 2008 ($\text{SIM}_{\text{Base}} - \text{SIM}_{\text{CO}_2=2008}$).

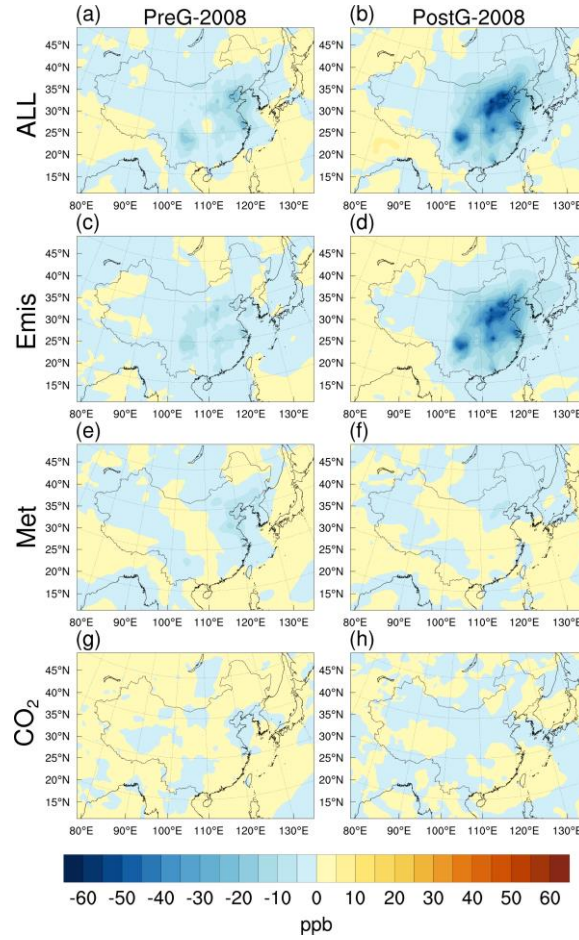


Figure 6. The total changes in PM_{2.5} concentrations (All, SIM_{Base} - SIM₂₀₀₈), and the changes in PM_{2.5} attributed to variations of anthropogenic pollutant emissions (Emis, All-Met-CO₂), meteorological conditions (Met, SIM_{Base} - SIM_{MET=2008}), and CO₂ emissions (CO₂, SIM_{Base} - SIM_{CO2=2008}) during the EASM period in PreG (2009–2013, left) and PostG (2014–2018, right) phase relative to 2008.

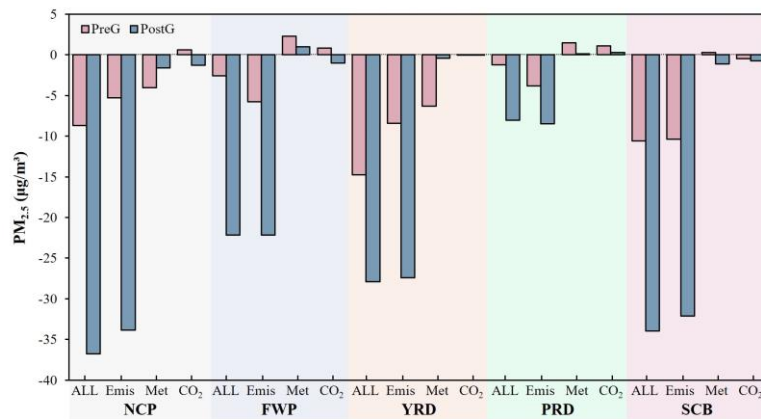


Figure 7. The total changes in PM_{2.5} concentrations (All, SIM_{Base} - SIM₂₀₀₈) for the North China Plain (NCP), Fenwei Plain (FWP), Yangtze River Delta (YRD), Pearl River Delta (PRD), and Sichuan Basin (SCB) during the EASM period in PreG (2009–2013) and PostG (2014–2018) phase relative to 2008, along with the variations

in PM_{2.5} due to anthropogenic pollutant emissions (Emis, All-Met-CO₂), meteorological conditions (Met, SIM_{Base} – SIM_{MET=2008}), and CO₂ emission (CO₂, SIM_{Base} - SIM_{CO2=2008}) changes.

14. Figure 4: Correct this caption (currently refers to O₃).

Response: Thanks. Sorry for the mistake. We replaced O₃ with PM_{2.5}.

15. Line 237: In Section 3.3 (and same for Section 3.2), it would be useful to reiterate which simulations have been used to generate these results, and include this in the captions for Figure.

Response: Thanks for your valuable suggestion. We have reiterated in the text which simulations were used to generate these results and included this information in the figure captions. Revisions to the figure captions are detailed in response to question 13.

Changes in manuscript:

3.1 PM_{2.5} variation

(L220–222): “Changes in PM_{2.5} concentrations from 2009 to 2018 relative to 2008 were quantified by comparing simulation results from each year in the SIM_{Base} experiment with SIM₂₀₀₈ (SIM_{Base} - SIM₂₀₀₈).”

3.2 Contribution of meteorological conditions

(L287–288): “The impact of meteorological conditions variations on PM_{2.5} concentrations were assessed by comparing SIM_{Base} results with those from SIM_{MET=2008} for the same year (SIM_{Base} - SIM_{MET=2008}).”

3.3 Contribution of CO₂

(L326–328): “The contribution of CO₂ emission changes to PM_{2.5} variability was quantified by comparing the SIM_{Base} experiment with the SIM_{CO2=2008} experiment (SIM_{Base} - SIM_{CO2=2008}) within the same year.”

3.4 Contribution of anthropogenic pollutant emissions

(L363–363): “The contribution of changed anthropogenic pollutant emissions to PM_{2.5} variation was determined by removing the effects of meteorological and CO₂ emission changes from the total variation.”

16. Line 267: Do you mean the average over the entire region?

Response: Thanks. This refers to the mean values, and we have revised the ambiguous wording accordingly.

Changes in manuscript:

(L364–365): “During the PreG period, PM_{2.5} levels decreased by an average of 5 to 10 µg/m³ over East Asia.”

17. Line 283: Correct the units on the legend in this Figure (should be µg/m³).

Response: Thanks. Sorry for the mistake. We have corrected the units on the legend in this Figure

Changes in manuscript:

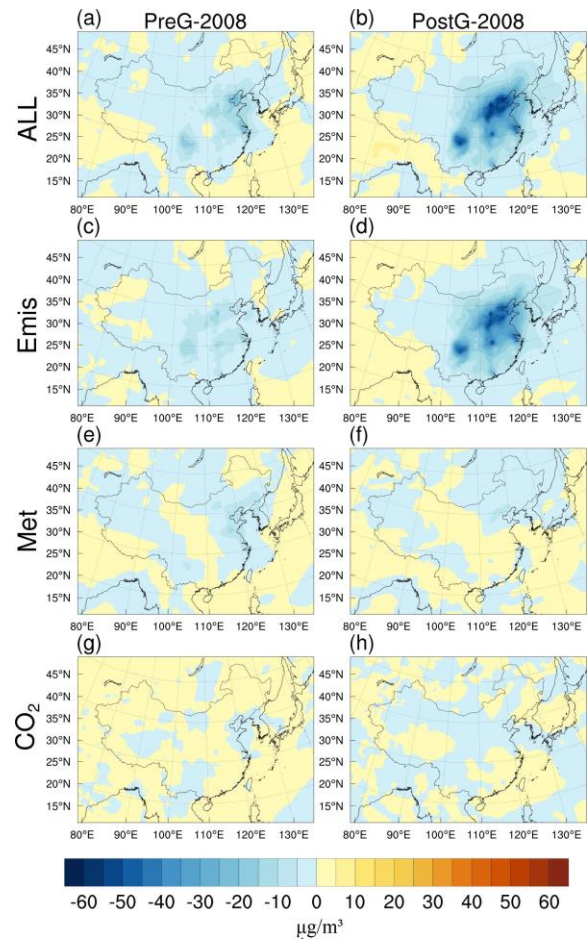


Figure 6. The total changes in PM_{2.5} concentrations (All, SIM_{Base} - SIM₂₀₀₈), and the changes in PM_{2.5} attributed to variations of anthropogenic pollutant emissions (Emis, All-Met-CO₂), meteorological conditions (Met, SIM_{Base} - SIM_{MET=2008}), and CO₂ emissions (CO₂, SIM_{Base} - SIM_{CO₂=2008}) during the EASM period in PreG (2009–2013, left) and PostG (2014–2018, right) phase relative to 2008.

18. In the simulations where meteorology varies with the year and you see an increase in temperature, would the model also have simulated an increase in BVOC emissions? If so that needs to be discussed.

Response: Thanks. As shown in Table 1, the only difference between the SIM_{Base} and SIM_{MET=2008} experiments lies in the meteorological conditions. Therefore, the difference SIM_{Base} - SIM_{MET=2008} represents the impact of meteorological changes on PM_{2.5}. Although elevated temperatures may lead to changes in BVOC emissions, Figure R1 indicates that BVOC changes are not significant and show little spatial correlation with PM_{2.5} trends (Figure 4 a-c). As a result, this aspect is not discussed further in the manuscript.

Table 1. The Numerical experimental in this study

Experiment	Time	Meteorological	CO ₂	Anthropogenic
------------	------	----------------	-----------------	---------------

		fields	emissions	pollutant emissions
	SIM ₂₀₀₈	2008	2008	2008
	SIM _{Base}	2009-2018	2009-2018	2009-2018
	SIM _{MET=2008}	2009-2018	2009-2018	2009-2018
	SIM _{CO2=2008}	2009-2018	2008	2009-2018

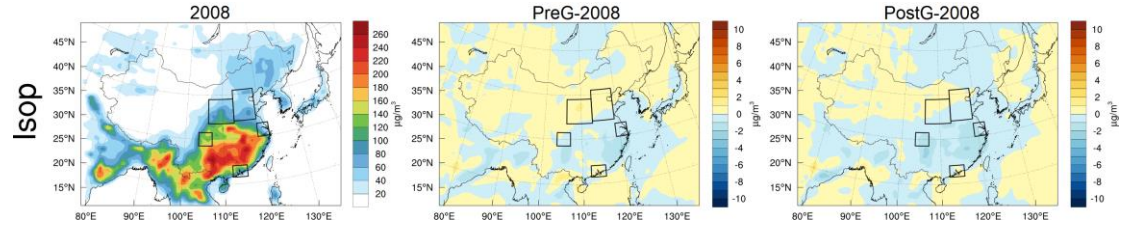


Figure R1. The isoprene ($\mu\text{g}/\text{m}^3$) during the EASM period in 2008 (left), and the changes due to meteorological variations in PreG (2009–2013, center) and PostG (2014–2018, right) phase relative to 2008 (SIM_{Base} - SIM_{MET=2008}).

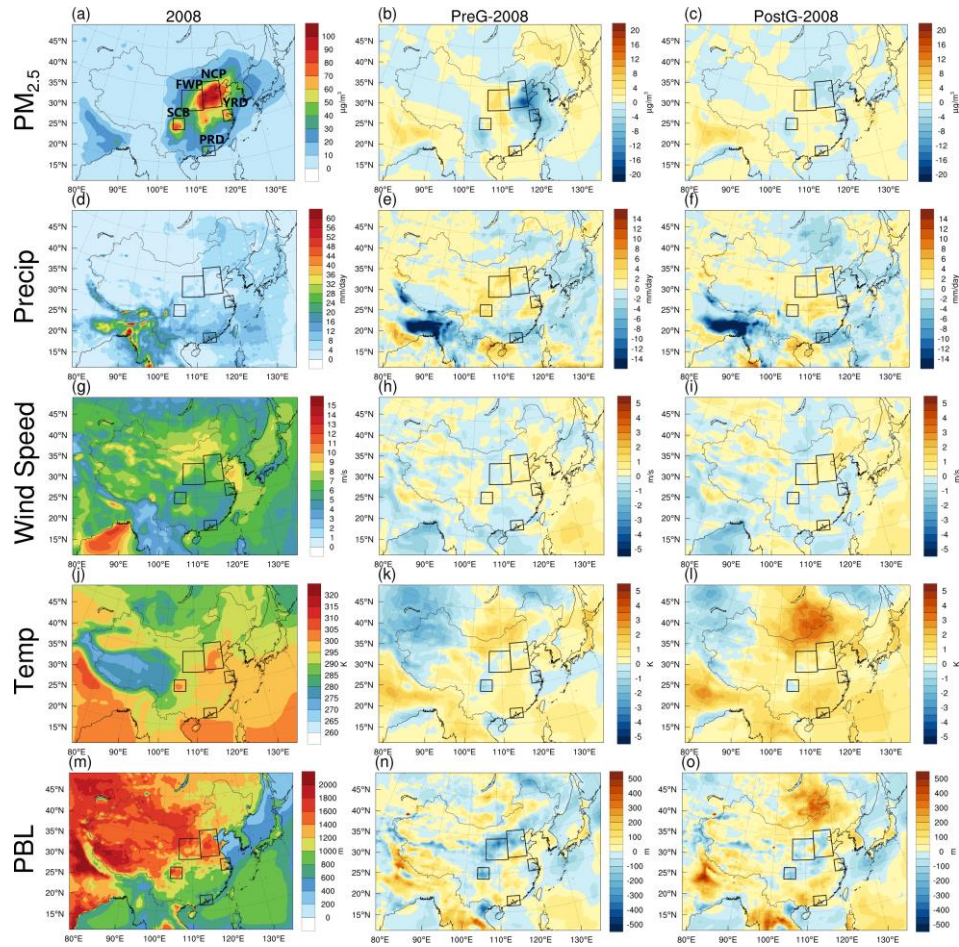


Figure 4. The PM_{2.5} (a–c, $\mu\text{g}/\text{m}^3$), precipitation (d–f, mm/day), wind speed (g–i, m/s), temperature (j–l, K), and Planetary Boundary Layer (PBL) height (m–o, m) during the EASM period in 2008 (left), and their mean changes due to meteorological variations

in PreG (2009–2013, center) and PostG (2014–2018, right) phase relative to 2008 ($\text{SIM}_{\text{Base}} - \text{SIM}_{\text{MET}=2008}$).