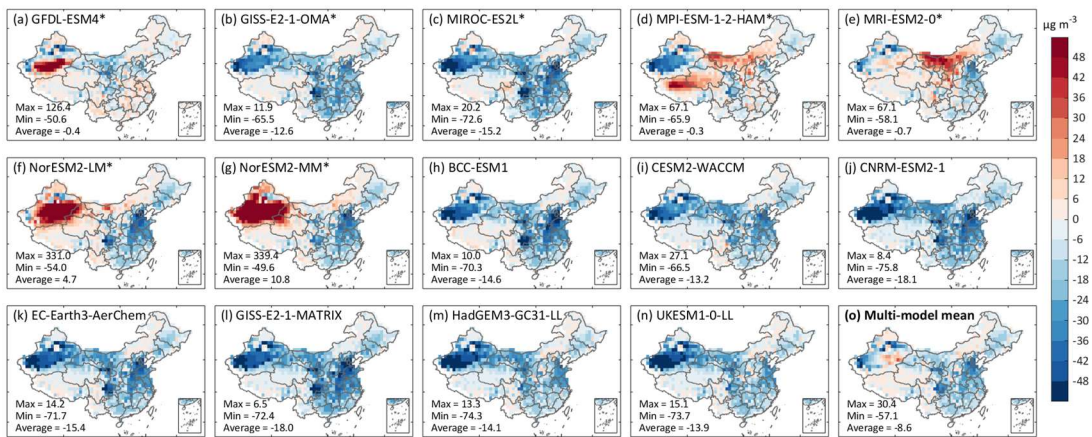


1

2 **Figure S1.** Provincial observed records over China during 2000–2014. The color and labeling of provinces are

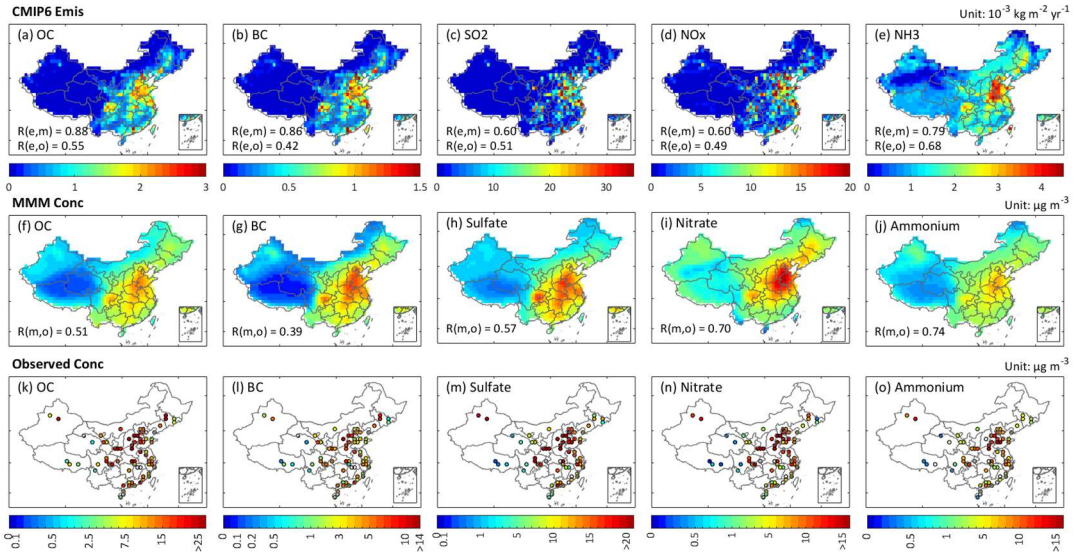
3 consistent with Figure 1.



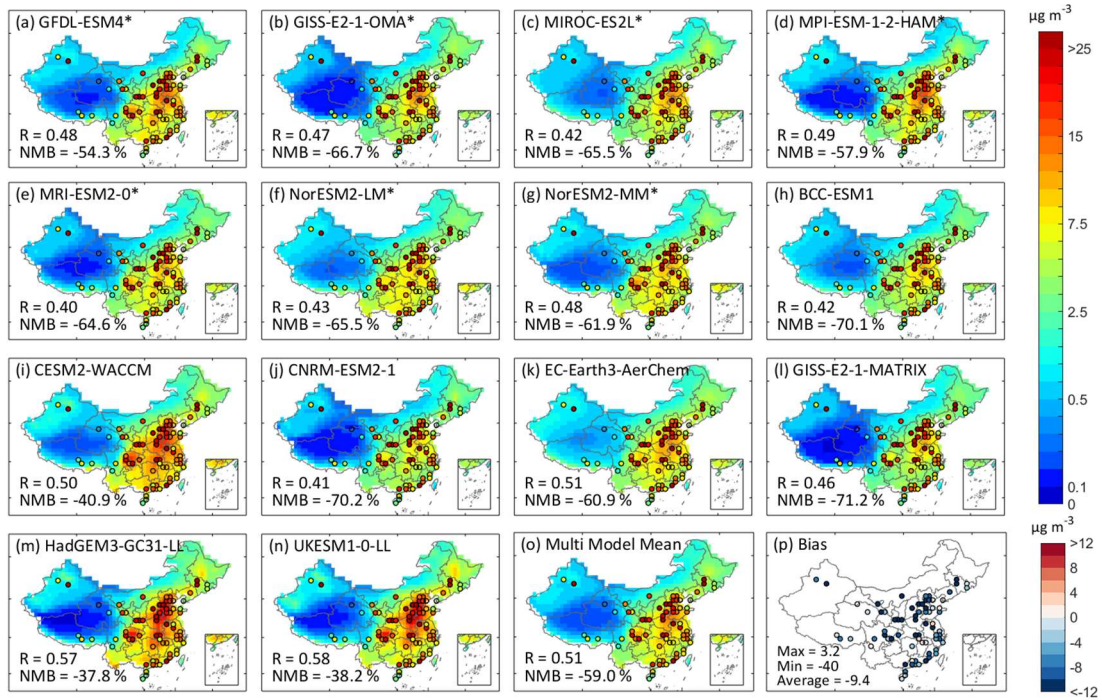
4

5 **Figure S2.** Spatial distribution of bias in the multi-year average of simulate-based $PM_{2.5}$ concentrations during 2000–

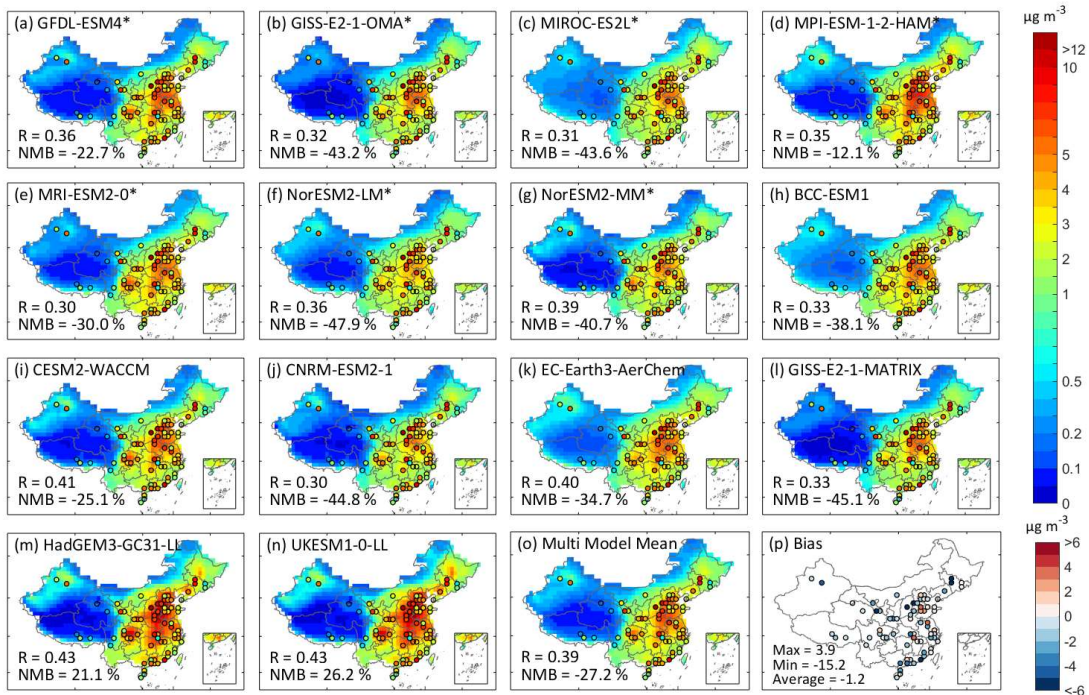
6 2014 for each model.



7
8 **Figure S3.** Multi-year average of CMIP6 emissions (a-e), multi-model mean concentrations (f-j), and observed
9 concentrations (k-o) of air pollutants over 2000–2014. $R(e, m)$, $R(e, o)$, and $R(m, o)$ denote the spatial correlation
10 coefficients between CMIP6 emissions and multi-model mean concentrations, between CMIP6 emissions and
11 observed concentrations, and between multi-model mean concentrations and observed concentrations.



12
13 **Figure S4.** Multi-year mean annual average near-surface OC concentrations over China during 2000–2014. (a-n)
14 OC in individual models overlaid with ground-based observations. (o) Multi-model mean overlaid with ground-
15 based observations. (p) Bias in multi-model mean.

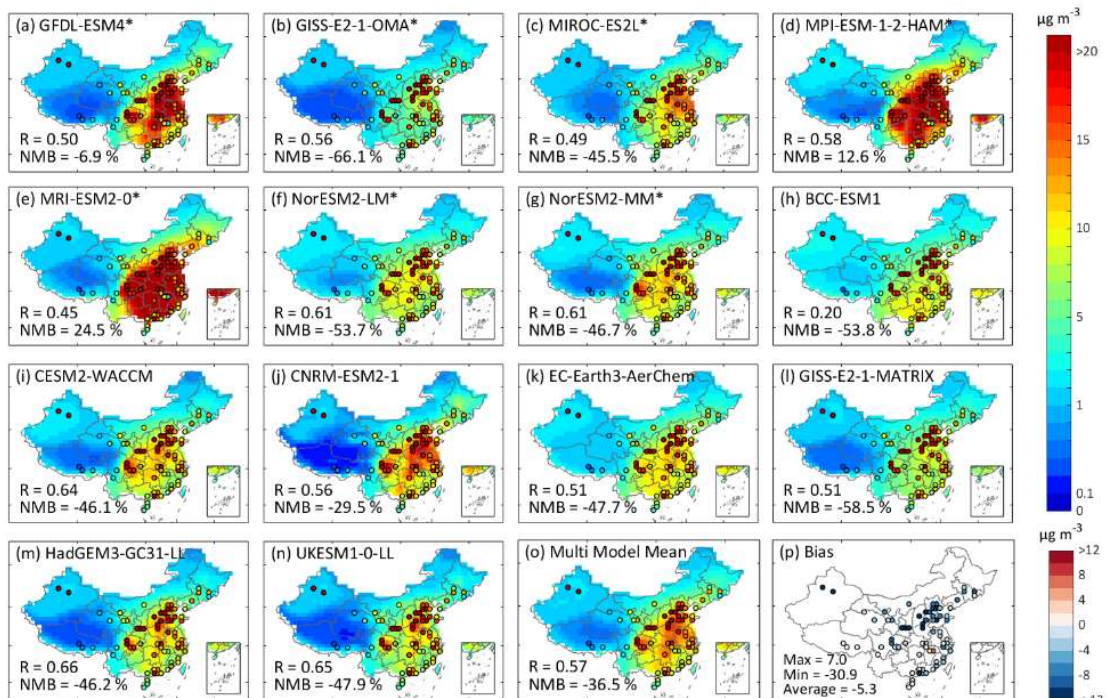


16

17 **Figure S5.** Multi-year mean annual average near-surface BC concentrations over China during 2000–2014. (a-n)

18 BC in individual models overlaid with ground-based observations. (o) Multi-model mean overlaid with ground-

19 based observations. (p) Bias in multi-model mean.

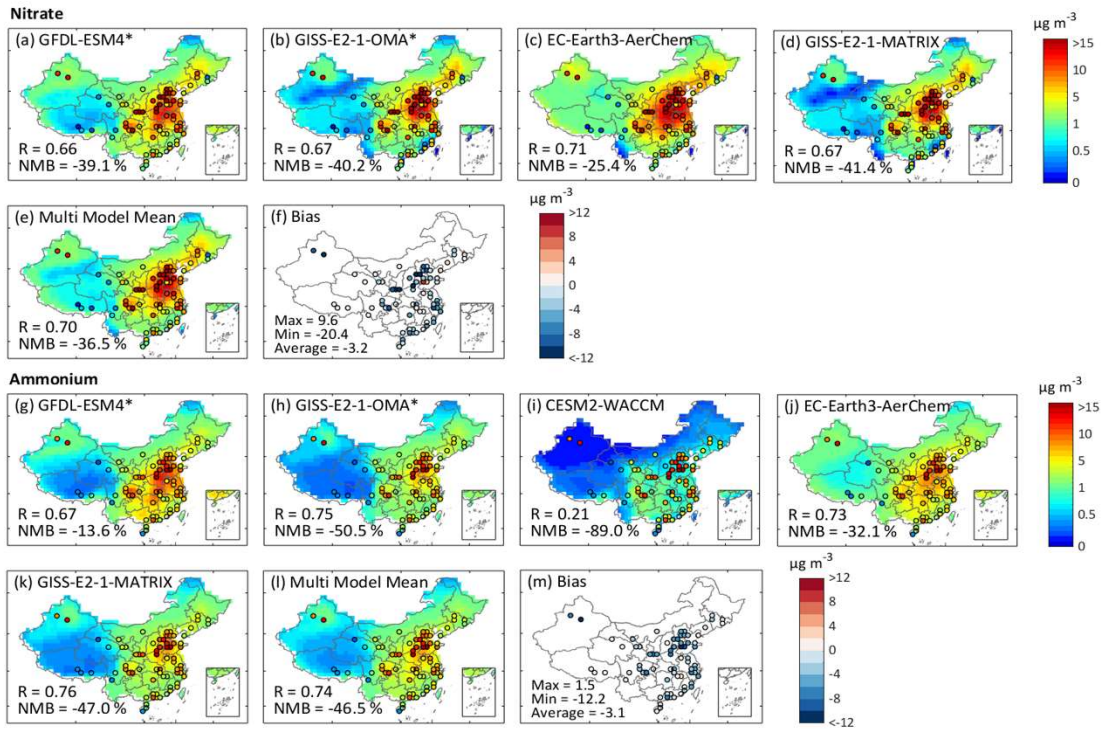


20

21 **Figure S6.** Multi-year mean annual average near-surface sulfate concentrations over China during 2000–2014. (a-n)

22 Sulfate in individual models overlaid with ground-based observations. (o) Multi-model mean overlaid with ground-

23 based observations. (p) Bias in multi-model mean.

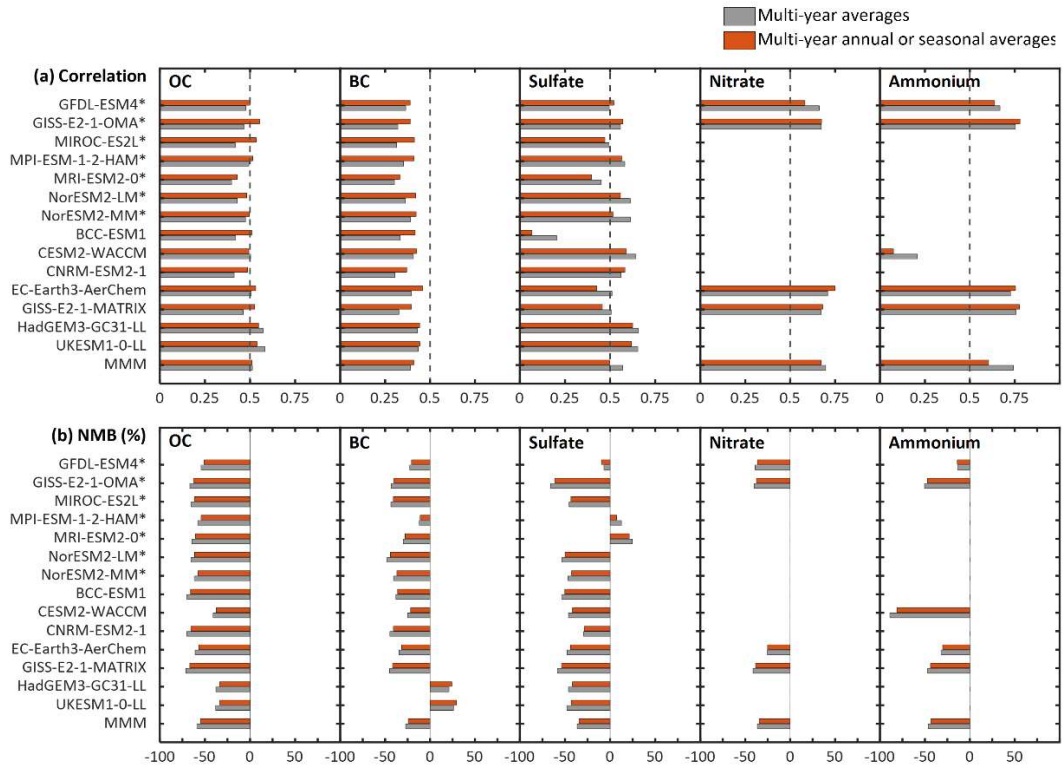


24

25 **Figure S7.** Multi-year mean annual average near-surface nitrate and ammonium concentrations over China during

26 2000–2014. (a-d) Nitrate and (g-k) ammonium in individual models overlaid with ground-based observations. (e, l)

27 Multi-model mean of overlaid with ground-based observations. (f, m) Bias in multi-model mean.



28

29 **Figure S8.** Spatial correlation and bias of multi-year averages (gray bar) and multi-year annual or seasonal averages

30 (red bar) of PM_{2.5} components during 2000–2014 for individual models.

31 **Table S1.** CMIP6 models and PM_{2.5} outputs.

Model	Resolution (Lat × Lon)	Number of members	OA, BC, SO ₄ ²⁻ , SSLT, DST	NO ₃ ⁻ , NH ₄ ⁺	PM _{2.5}	Model references and data citation
BCC-ESM1	2.813° × 2.813°	3	Y			(Wu et al., 2019; Wu et al., 2020; Zhang et al., 2018)
CESM2- WACCM	0.9° × 1.25°	3	Y	Y (No NO ₃ ⁻)		(Danabasoglu, 2019; Gettelman et al., 2019; Tilmes et al., 2019; Emmons et al., 2020)
CNRM- ESM2-1	1.4° × 1.4°	3	Y			(Séférian, 2018; Séférian et al., 2019; Michou et al., 2020)
EC-Earth3- AerChem	2° × 3°	3	Y	Y		(Van Noije et al., 2021; Döscher et al., 2022; EC-Earth Ec-Earth- Consortium, 2020)
GFDL- ESM4	1° × 1.25°	1	Y	Y	Y	(Horowitz et al., 2020; Dunne et al., 2020; Krasting et al., 2018)
GISS-E2-1- OMA	2° × 2.5°	15	Y	Y	Y	(Bauer et al., 2020; Miller et al., 2021; Nasa Goddard Institute for Space Studies, 2018)
GISS-E2-1- MATRIX	2° × 2.5°	12	Y	Y		(Bauer et al., 2020; Miller et al., 2021; Nasa Goddard Institute for Space Studies, 2018)
HadGEM3- GC31-LL	1.25° × 1.875°	4	Y			(Ridley et al., 2018; Kuhlbrodt et al., 2018)
MIROC- ES2L	2.813° × 2.813°	10	Y		Y	(Hajima et al., 2020; Hajima et al., 2019)
MPI-ESM- 1-2-HAM	1.875° × 1.875°	3	Y		Y	(Tegen et al., 2019; Neubauer et al., 2019)
MRI-ESM2- 0	1.875° × 1.875°	5	Y		Y	(Yukimoto et al., 2019a; Yukimoto et al., 2019b; Oshima et al., 2020)

NorESM2-LM	1.9° × 2.5°	3	Y	Y	(Karset et al., 2018; Seland et al., 2019; Seland et al., 2020; Kirkevåg et al., 2018)
NorESM2-MM	0.9° × 1.25°	3	Y	Y	(Karset et al., 2018; Bentsen et al., 2019; Seland et al., 2020; Kirkevåg et al., 2018)
UKESM1-0-LL	1.25° × 1.875°	4	Y		(Tang et al., 2019; Sellar et al., 2019)

32 **Table S2.** The specific values of a_1 and a_2 from Eq. 1. The average, trend, and spatial correlation coefficients of
33 PM_{2.5} concentrations over the eastern region and western region during 2000–2014.

	Model	a_1	a_2	Eastern region			Western region		
				Average ($\mu\text{g m}^{-3}$)	Trend ($\mu\text{g m}^{-3} \text{ yr}^{-1}$) ^a	Spatial Corr. ^b	Average ($\mu\text{g m}^{-3}$)	Trend ($\mu\text{g m}^{-3} \text{ yr}^{-1}$)	Spatial Corr.
Satellite-based				39.0	0.72	1	22.7	0.06*	1
Total PM_{2.5} from Direct ESM output	GFDL-ESM4			37.7	1.14	0.92	22.1	0.28	0.66
	GISS-E2-1-OMA			24.4	0.69	0.91	10.9	0.13	0.79
	MIROC-ES2L			20.3	0.49	0.90	8.9	0.13	0.59
	MPI-ESM1-2-HAM			36.6	0.93	0.91	22.5	0.20*	0.36
	MRI-ESM2-0			30.4	0.57	0.83	24.5	0.24	0.71
	NorESM2-LM			22.1	0.32	0.87	35.5	0.03*	0.49
	NorESM2-MM			23.6	0.40	0.90	43.1	-0.10*	0.53
Total PM_{2.5} from Eq. 1	BCC-ESM1			19.5	0.40	0.87	10.2	0.15	0.62
	CESM2-WACCM	0.25	0.1	24.0	0.73	0.92	10.1	0.22	0.67
	CNRM-ESM2-1	0.02	0.25	18.9	0.42	0.90	5.5	0.11	0.51
	EC-Earth3-AerChem	0.25	0.1	21.4	0.56	0.91	8.3	0.18	0.53

GISS-E2-1-MATRIX	0.25	0.1	17.0	0.43	0.92	6.4	0.10	0.67
HadGEM3-GC31-LL	0.27	0.35	26.5	0.80	0.89	7.9	0.18	0.50
UKESM1-0-LL	0.27	0.35	26.5	0.71	0.89	8.1	0.18	0.52

34 ^a Trends are estimated using the Theil-Sen Median method (Theil, 1950; Sen, 1968). Significant changes are
35 identified using the non-parametric Mann-Kendall test (Kendall, 1938). * represents non-significant monotonous
36 change at $p = 0.05$. ^b Spatial correlation coefficients between simulations and satellite-based data over the eastern
37 and western regions are calculated. The spatial correlation coefficients of 14 models are at the 0.05 significance level.

38 **Table S3.** Multi-year averages of PM_{2.5} concentrations including five aerosol species (Eq. 1) and all fine aerosol
39 species from 4 models providing nitrate and ammonium simulations.

Model	PM _{2.5} according to Eq. 1 ($\mu\text{g m}^{-3}$)	PM _{2.5} including all fine aerosol species ($\mu\text{g m}^{-3}$) ^a	Nitrate proportion ^b	Ammonium proportion
EC-Earth3-AerChem	12.1	18.7	20.6%	14.3%
GFDL-ESM4	13.0	18.5	15.1%	14.6%
GISS-E2-1-OMA	10.0	14.1	17.6%	11.4%
GISS-E2-1-MATRIX	9.5	13.8	17.5%	13.2%

40 ^a represents that $\text{PM}_{2.5} = \text{OA} + \text{BC} + \text{SO}_4^{2-} + 0.25\text{SSLT} + 0.1\text{DST} + \text{NO}_3^- + \text{NH}_4^+$. ^b represents that the proportion
41 of nitrate to PM_{2.5} including all fine aerosol species.

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