

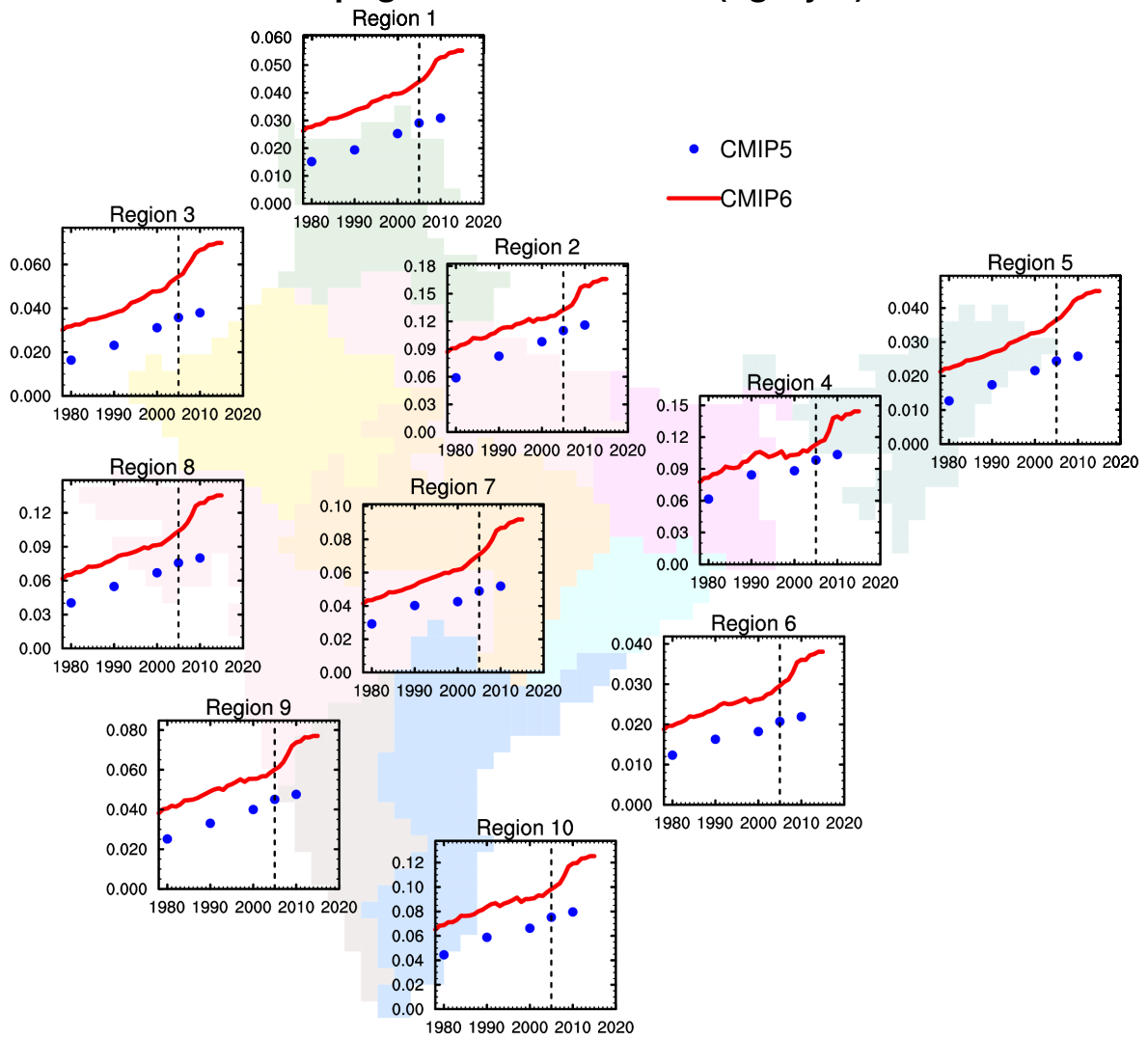
**Table S1.** Heterogeneous uptake coefficients.

Species	Gamma
N <sub>2</sub> O <sub>5</sub>	0.1
N <sub>2</sub> O <sub>5</sub> dust	0.0
NO <sub>3</sub>	0.1
NO <sub>3</sub> dust	0.0
NO <sub>2</sub>	0.0001
NH <sub>3</sub>	0.05
HO <sub>2</sub>	1.0

**Table S2.** Time series correlation of each station's observed total PM<sub>2.5</sub> with the individual components of modeled dry PM<sub>2.5</sub>, total dry PM<sub>2.5</sub>, and total wet PM<sub>2.5</sub> in the grid cell the station is located within. The last two columns show the normalized mean bias (%) of modeled dry and wet PM<sub>2.5</sub>. For each station, the component with the highest correlation is in bold.

Site	Correlation of modeled component with daily averaged observed PM <sub>2.5</sub>										Normalized Mean Bias	
	Dust	Salt	BC	OM	SOA	NH <sub>4</sub>	SO <sub>4</sub>	NO <sub>3</sub>	PM <sub>2.5</sub> (dry)	PM <sub>2.5</sub> (wet)	PM <sub>2.5</sub> (dry)	PM <sub>2.5</sub> (wet)
1	-0.36	-0.46	0.51	0.53	-0.18	0.53	-0.17	<b>0.67</b>	0.62	0.61	3.8	28.1
2	-0.36	-0.25	0.66	<b>0.66</b>	-0.33	0.50	-0.08	0.60	0.61	0.60	-32.7	-18.7
3	-0.40	-0.37	<b>0.60</b>	0.60	-0.32	0.44	-0.19	0.56	0.56	0.55	-23.9	-7.8
4	-0.20	-0.14	0.59	<b>0.60</b>	-0.13	0.43	-0.02	0.46	0.49	0.47	-57.7	-48.8
5	-0.24	-0.10	<b>0.41</b>	0.40	0.25	0.25	0.14	0.24	0.34	0.33	-26.0	-10.3
6	0.01	0.19	<b>0.47</b>	0.44	0.23	0.13	-0.11	0.09	0.24	0.21	-26.4	-10.3
7	-0.30	-0.21	0.63	<b>0.66</b>	-0.14	0.53	0.08	0.57	0.58	0.57	-34.2	-20.3
8	-0.34	-0.23	0.63	<b>0.65</b>	-0.14	0.52	0.05	0.55	0.56	0.55	-48.1	-37.2
9	-0.31	-0.21	0.51	<b>0.54</b>	-0.27	0.39	-0.08	0.44	0.45	0.44	-45.9	-34.6
10	-0.21	-0.19	0.25	0.23	-0.12	<b>0.25</b>	0.10	0.18	0.21	0.21	-31.2	-16.7
11	-0.29	-0.21	<b>0.32</b>	0.30	0.06	-0.14	-0.03	-0.24	-0.12	-0.13	-52.3	-45.6
12	-0.33	-0.24	0.66	<b>0.68</b>	-0.19	0.53	-0.03	0.55	0.57	0.56	-39.9	-27.2
13	0.27	0.07	0.23	0.21	<b>0.53</b>	0.21	0.27	0.22	0.28	0.27	-36.7	-23.9
14	-0.23	-0.16	<b>0.44</b>	0.43	-0.15	0.35	-0.11	0.36	0.35	0.34	13.4	38.8
15	-0.40	-0.30	0.45	<b>0.48</b>	0.10	0.38	0.09	0.41	0.37	0.36	-50.0	-39.0
16	-0.15	-0.22	0.29	<b>0.31</b>	0.05	0.18	-0.18	0.21	0.16	0.17	-47.5	-37.3
17	-0.38	-0.22	<b>0.66</b>	0.65	-0.26	0.54	-0.02	0.56	0.58	0.57	-30.5	-15.9
18	-0.35	-0.17	0.63	<b>0.64</b>	-0.18	0.50	-0.05	0.56	0.57	0.55	-21.0	-3.8
19	-0.25	-0.16	0.58	0.58	-0.25	0.53	0.06	<b>0.62</b>	0.59	0.58	-26.9	-11.0
20	-0.24	-0.20	<b>0.60</b>	0.60	-0.14	0.47	0.09	0.54	0.55	0.54	-24.7	-8.4
21	-0.36	-0.32	0.59	0.58	-0.10	0.61	0.21	0.64	<b>0.64</b>	0.64	-30.9	-15.8
22	-0.37	-0.20	0.52	<b>0.53</b>	-0.28	0.40	-0.15	0.49	0.47	0.46	-10.0	9.5

## Anthropogenic BC Emissions (TgC yr<sup>-1</sup>)



**Figure S1.** Time series (1980–2015) of anthropogenic black carbon emissions (TgC yr<sup>-1</sup>) over 10 regions of India for the CMIP5 inventory (blue dots) and CMIP6 inventory (red lines).

# Anthropogenic OM Emissions (Tg yr<sup>-1</sup>)

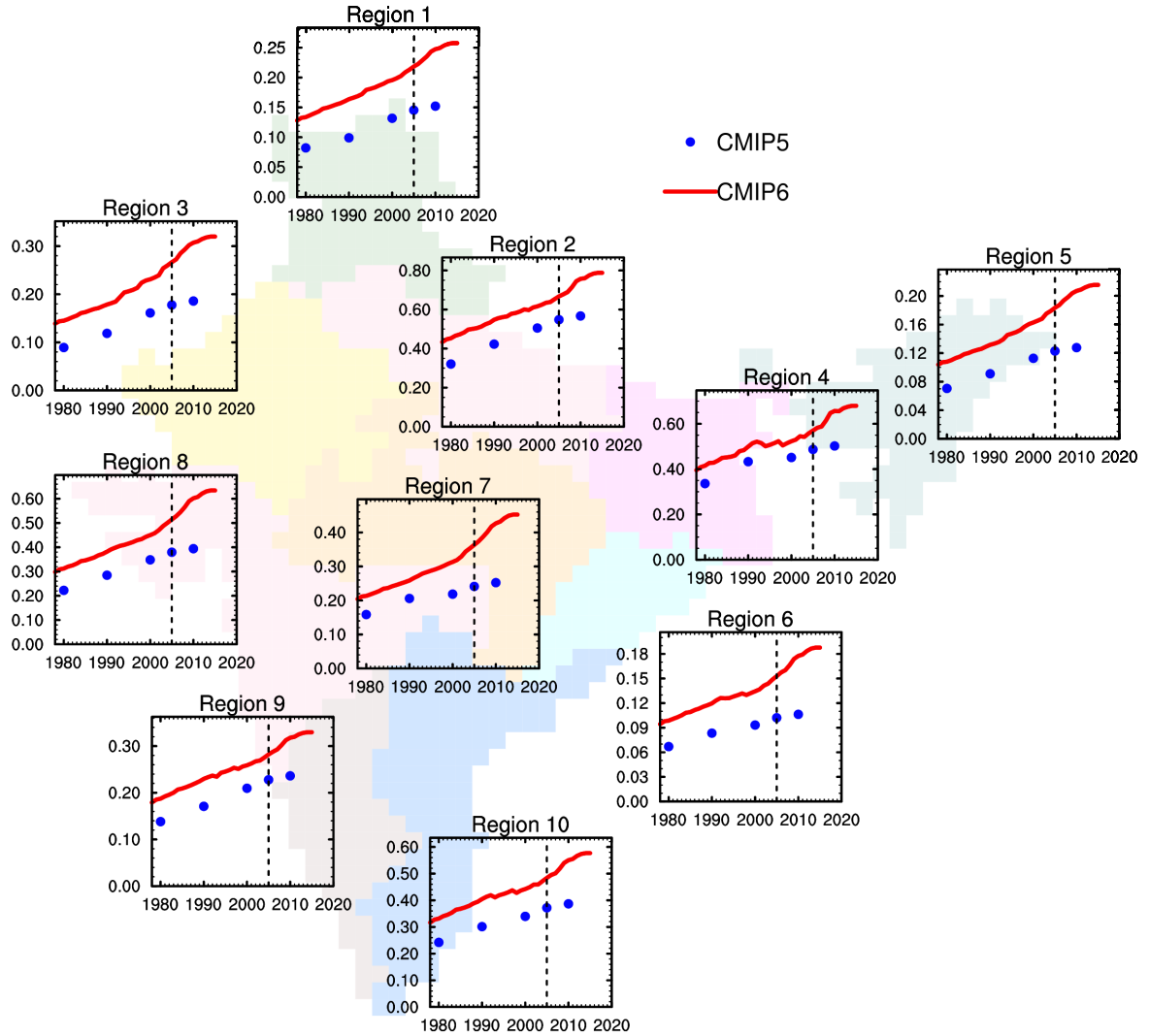


Figure S2. Same as Figure S1, but for organic matter (Tg yr<sup>-1</sup>)

# Anthropogenic NO Emissions (Tg yr<sup>-1</sup>)

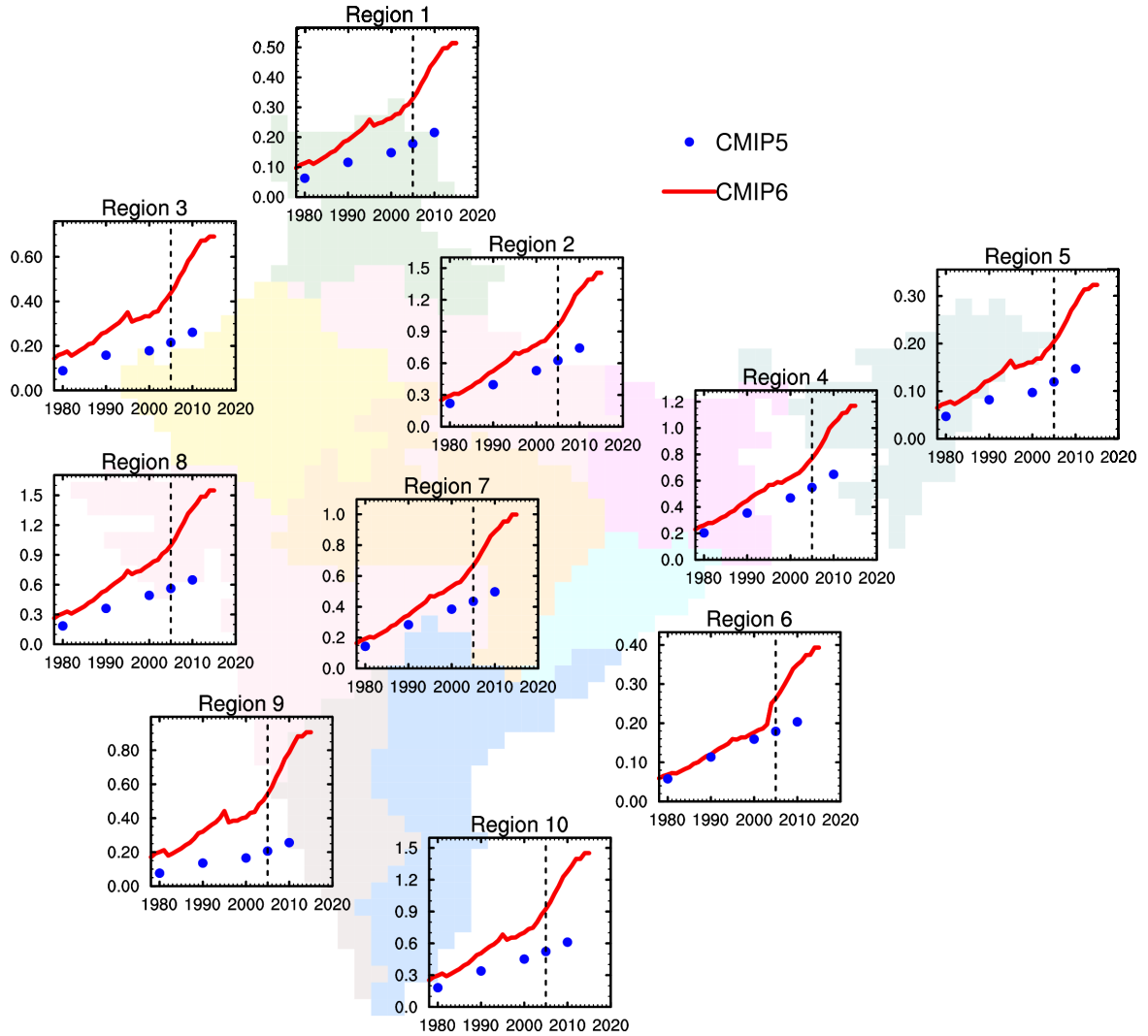


Figure S3. Same as Figure S1, but for NO (Tg yr<sup>-1</sup>)

# Anthropogenic SO<sub>2</sub> Emissions (Tg yr<sup>-1</sup>)

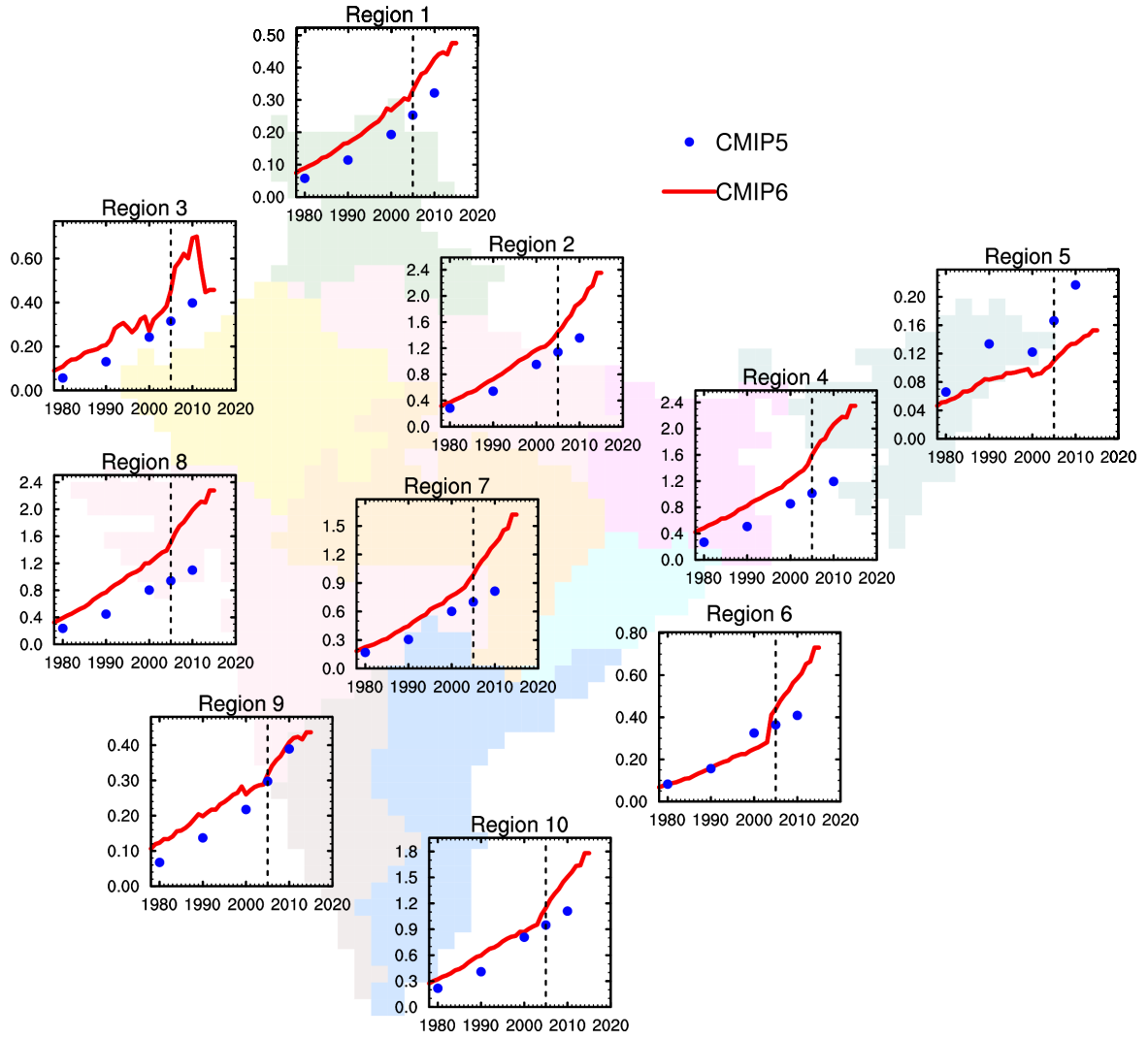
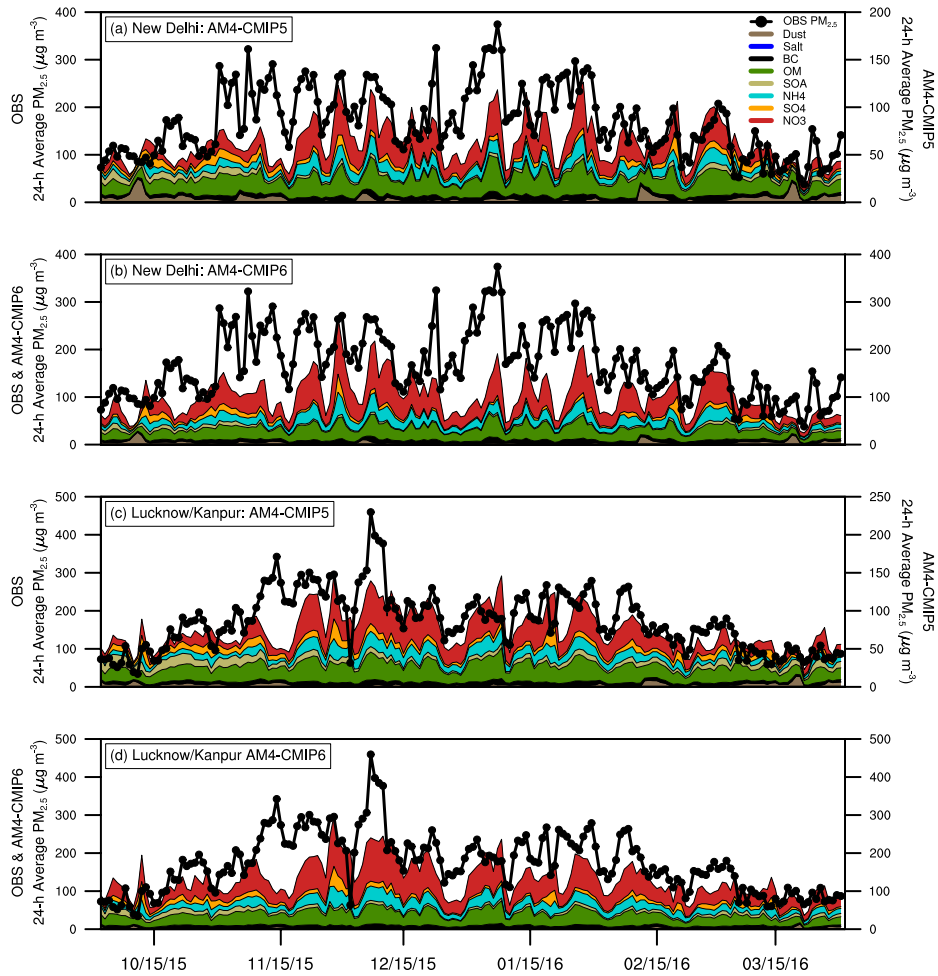
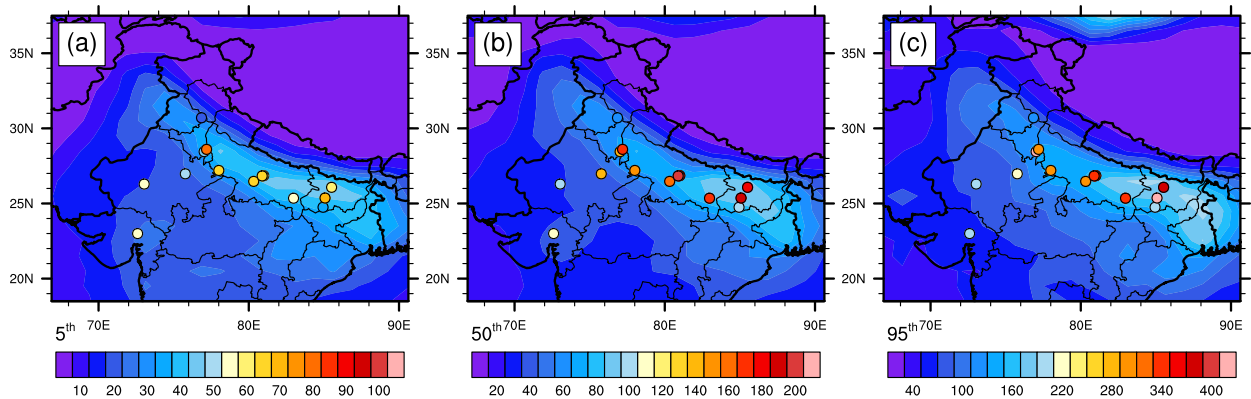


Figure S4. Same as Figure S1, but for SO<sub>2</sub> (Tg yr<sup>-1</sup>)

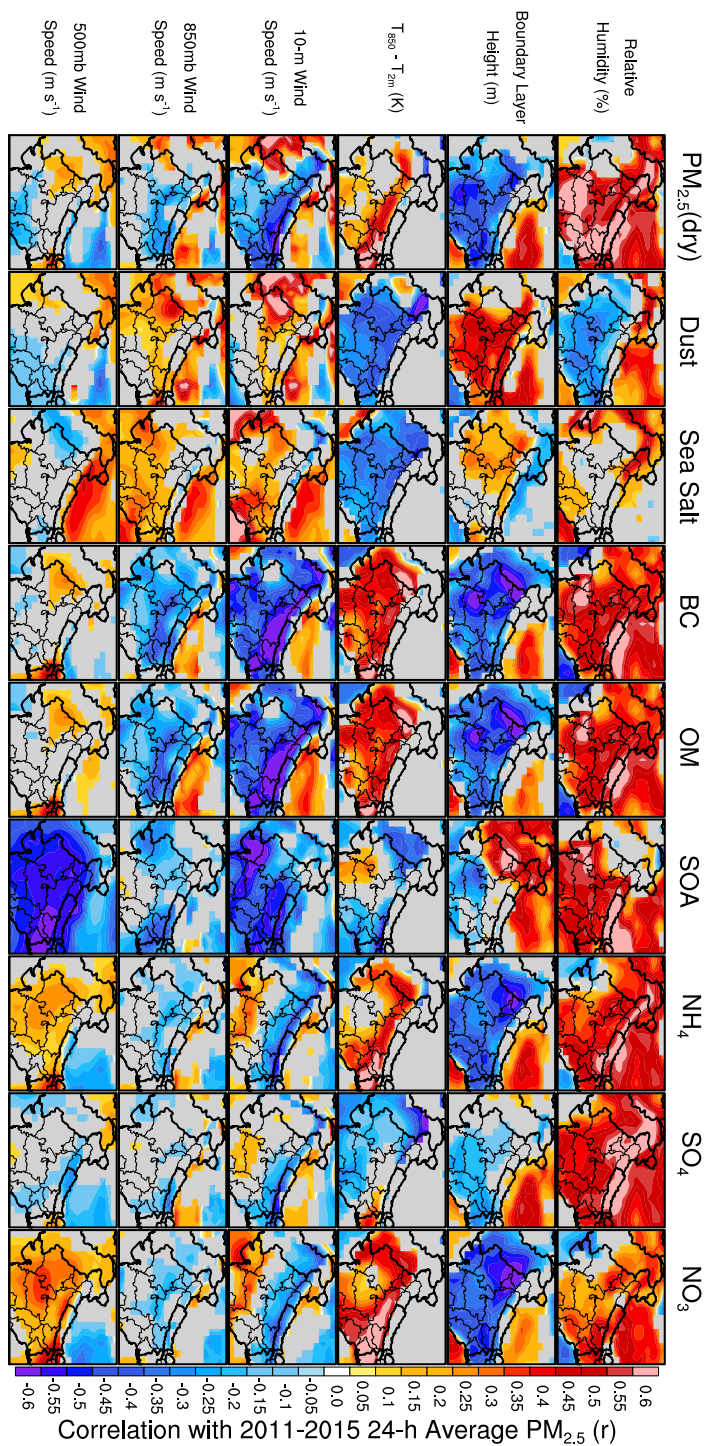




**Figure S6.** Six month time series of daily total observed  $PM_{2.5}$  (black) and modeled components (colors) over (a, b) New Delhi and (c, d) Lucknow/Kanpur, Uttar Pradesh for AM4-CMIP5 (a, c) and AM4-CMIP6 (b, d). Note the different y-axis scales in (a) and (c) for the AM4-CMIP5 versus the observations.

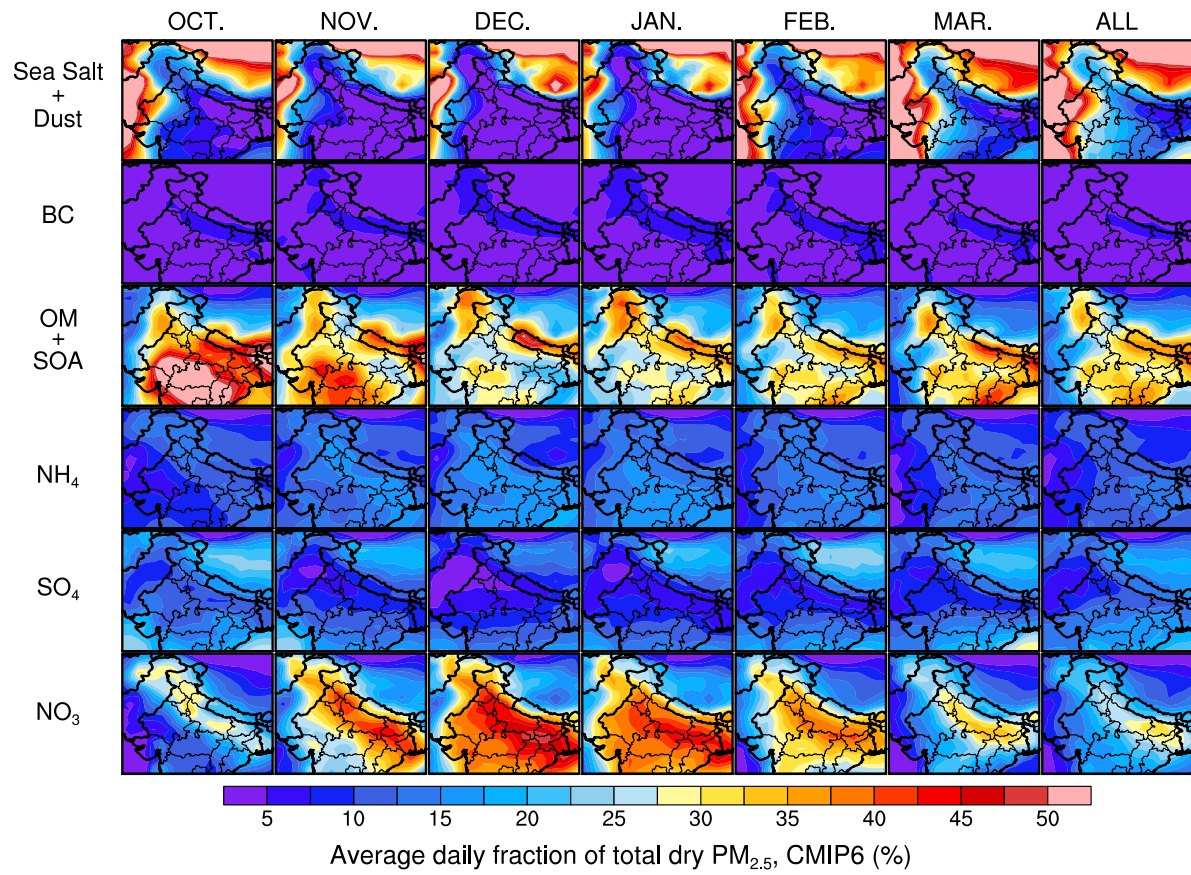


**Figure S7.** (a-b) 5th (c-d) 50th, and (e-f) 95th percentile of 24-h average  $PM_{2.5}$  ( $\mu g m^{-3}$ ) over 1 October 2015 – 31 March 2016 for the observations (circles), and the AM4-CMIP5-wet (grid).

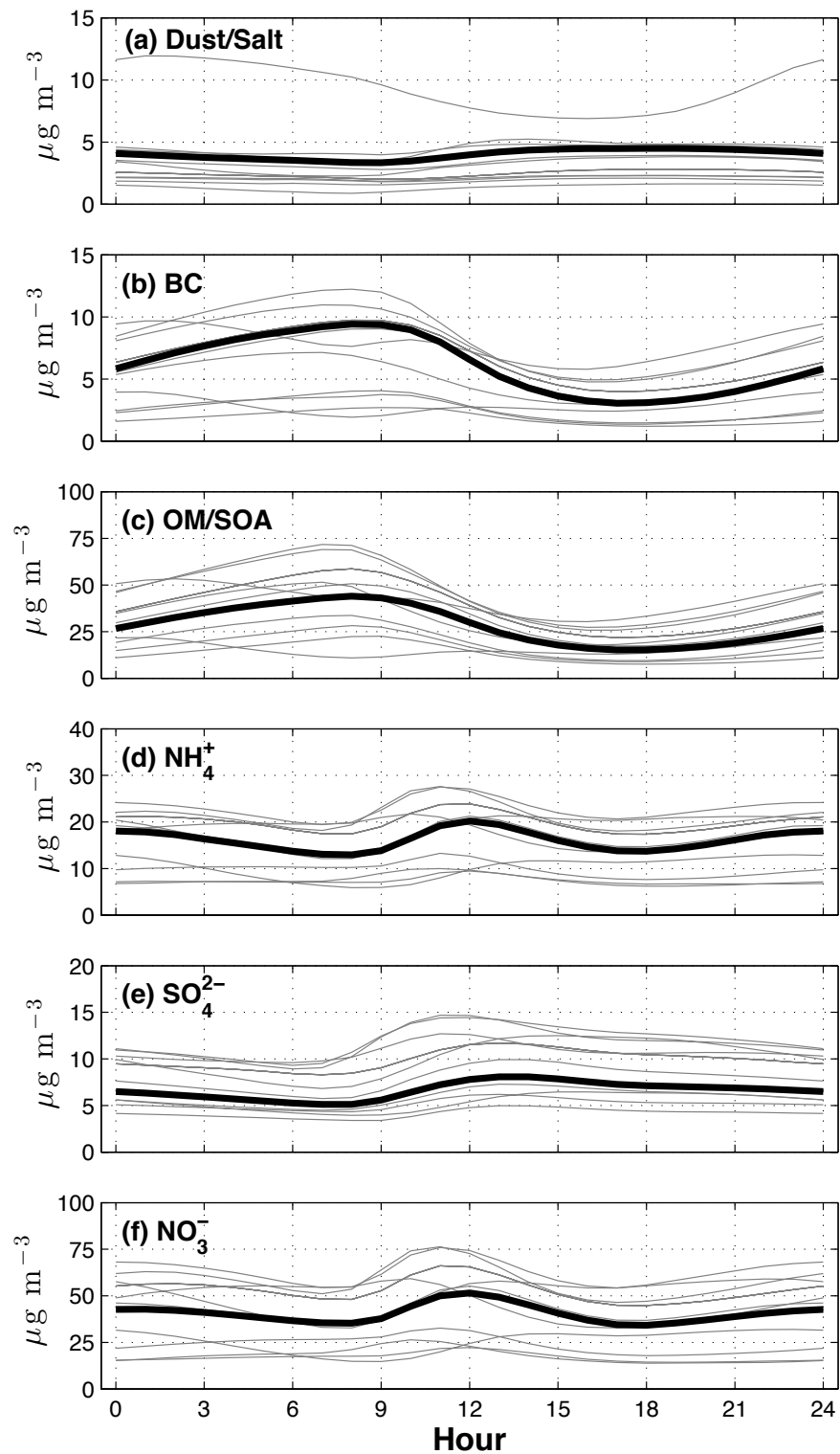


**Figure S8.** Correlation of daily average modeled dry  $PM_{2.5}$  and its components (columns) with the meteorological variables shown in Figure 4 (rows) over October–March, 2011–2015. Gray areas are where the correlation is not significant at the 95% confidence level.

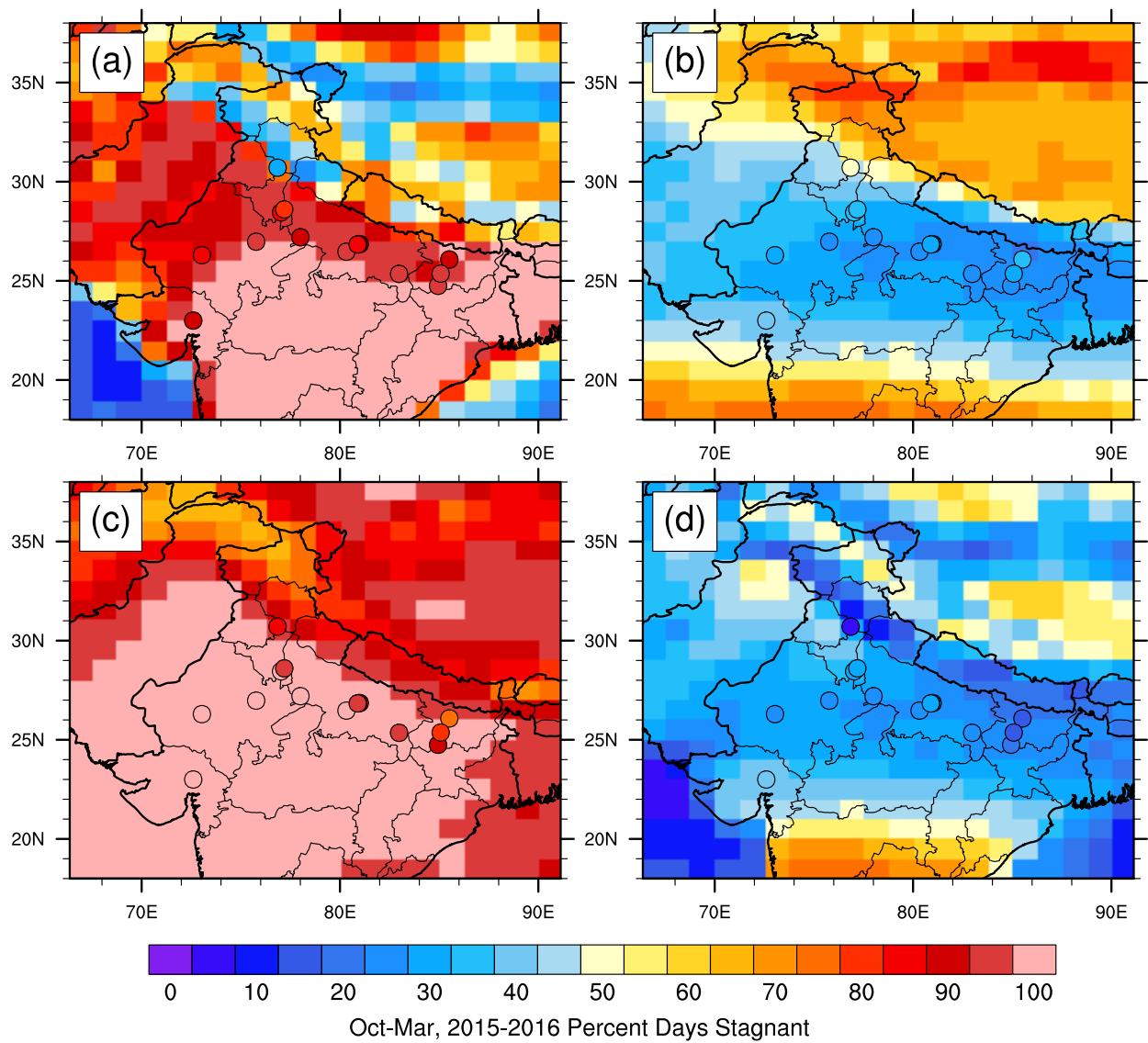




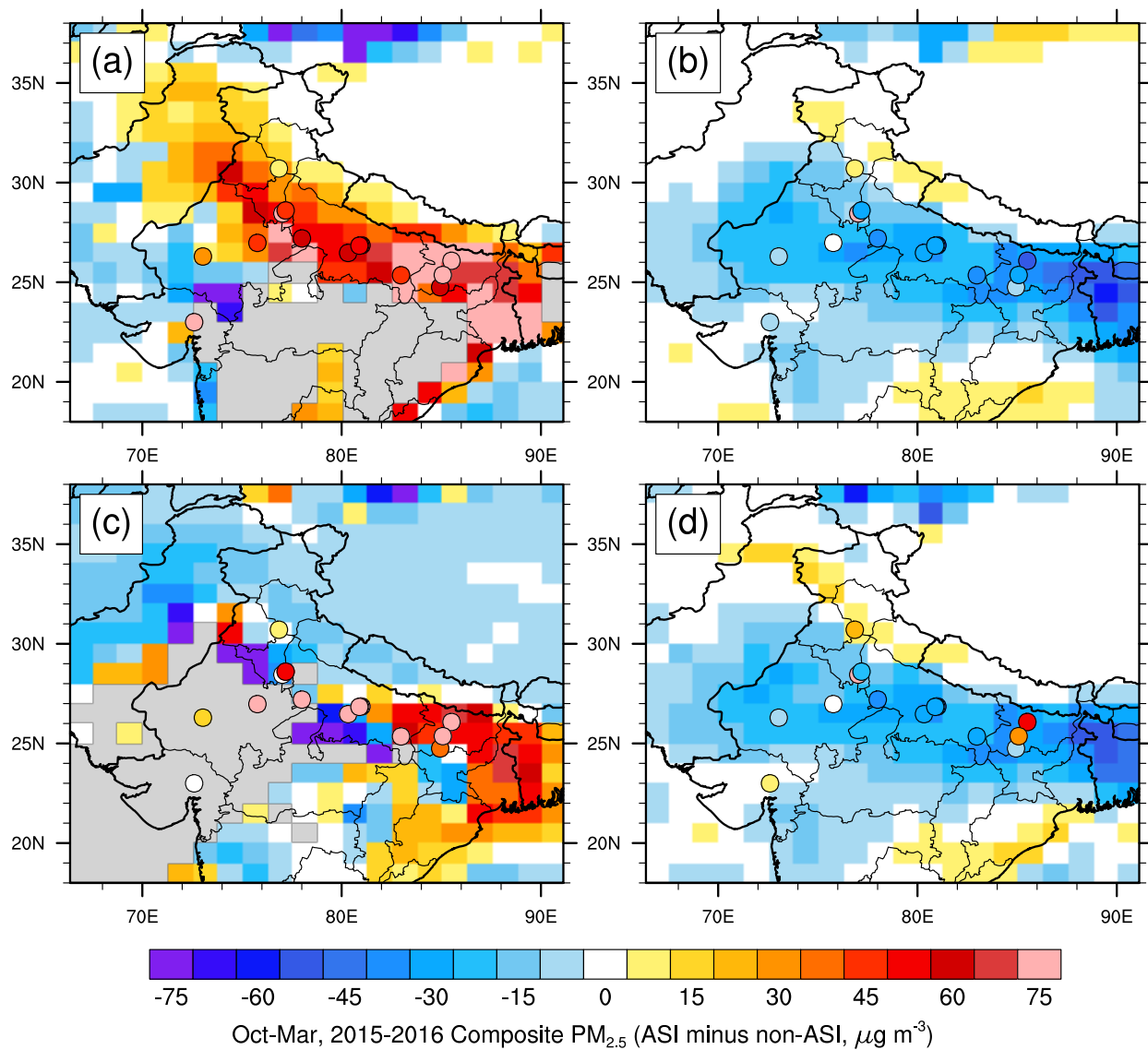
**Figure S9.** Average fraction of AM4-CMIP6 dry PM<sub>2.5</sub> for each of the major components (rows) for each month and the 6-month mean (columns) averaged over 2011–2015.



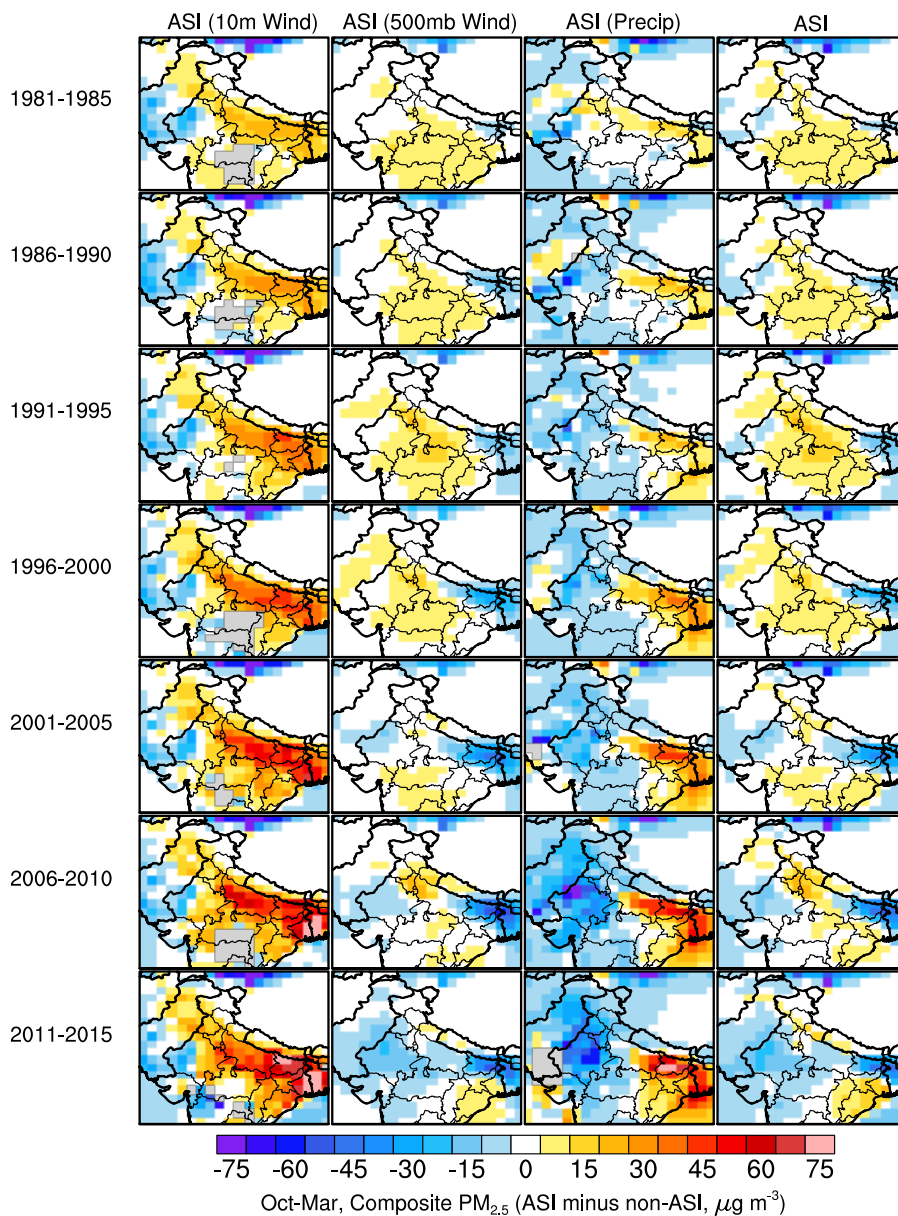
**Figure S10.** Diurnal cycles of the abundance ( $\mu\text{g m}^{-3}$ ) of (a) dust and sea salt, (b) black carbon, (c) organic matter and secondary organic aerosol, (d) ammonium, (e) sulfate, and (f) nitrate in AM4-CMIP6 for the individual grid cells where observations are located (gray) and their median (black) averaged over 1 October 2015 – 31 March 2016.



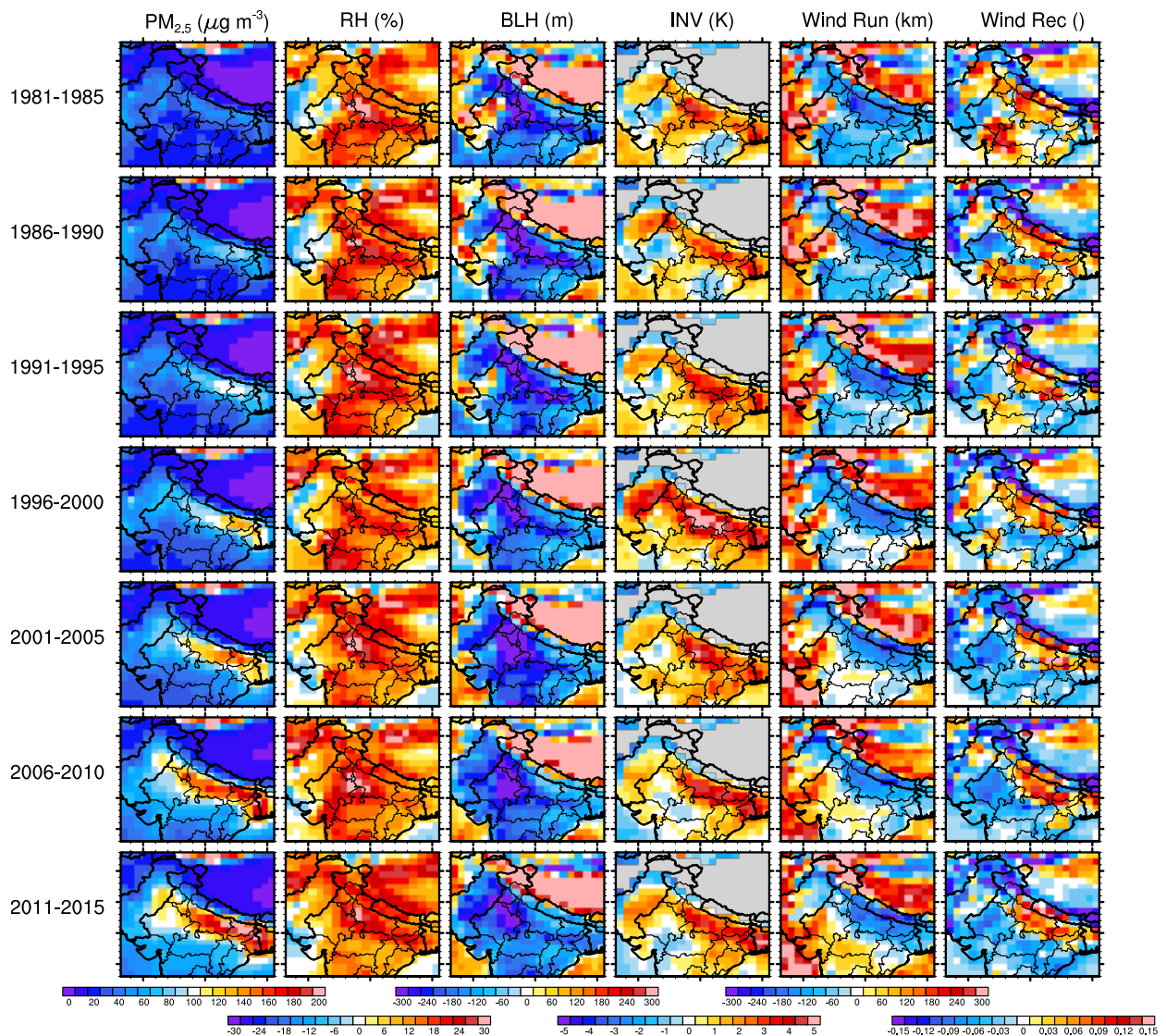
**Figure S11.** Percentage of October–March, 2015–2016 days that are considered stagnant with respect to (a) 10-m wind speed ( $< 3.2 \text{ m s}^{-1}$ ), (b) 500 mb wind speed ( $< 13.0 \text{ m s}^{-1}$ ), (c) precipitation ( $< 1.0 \text{ mm day}^{-1}$ ), and (d) all three; i.e. the ASI is met for the observations (circles) and the AM4-CMIP6 (background).



**Figure S12.** Composite of raw PM<sub>2.5</sub> on days when ASI components (a) 10-m wind speed, (b) 500 mb wind speed, (c) precipitation, and (d) total ASI are met minus days when they are not for October 2015 – March 2016. Gray areas are locations where a composite cannot be constructed since 100% of the days are considered stagnant.



**Figure S13.** Composite of AM4-CMIP6 wet PM<sub>2.5</sub> on days when ASI components (first column) 10-m wind speed, (second column) 500 mb wind speed, (third column) precipitation, and (last column) total ASI are met minus days when they are not for October–March over seven 5-year intervals (rows). Gray areas are locations where a composite cannot be constructed since 100% of the days are considered stagnant.



October - March, 50 Worst Days minus 50 Best Days

**Figure S14.** Composite of the 50 days with the highest PM<sub>2.5</sub> abundance minus the 50 days with the lowest for October – March over seven 5-year intervals (rows) for (first column) PM<sub>2.5</sub> (wet,  $\mu\text{g m}^{-3}$ ), (second column) relative humidity (%), (third column) boundary layer height (m), (fourth column) temperature inversion strength (K), (fifth column) wind run (km), and (sixth column) wind recirculation (unitless) for AM4-CMIP6-wet.