



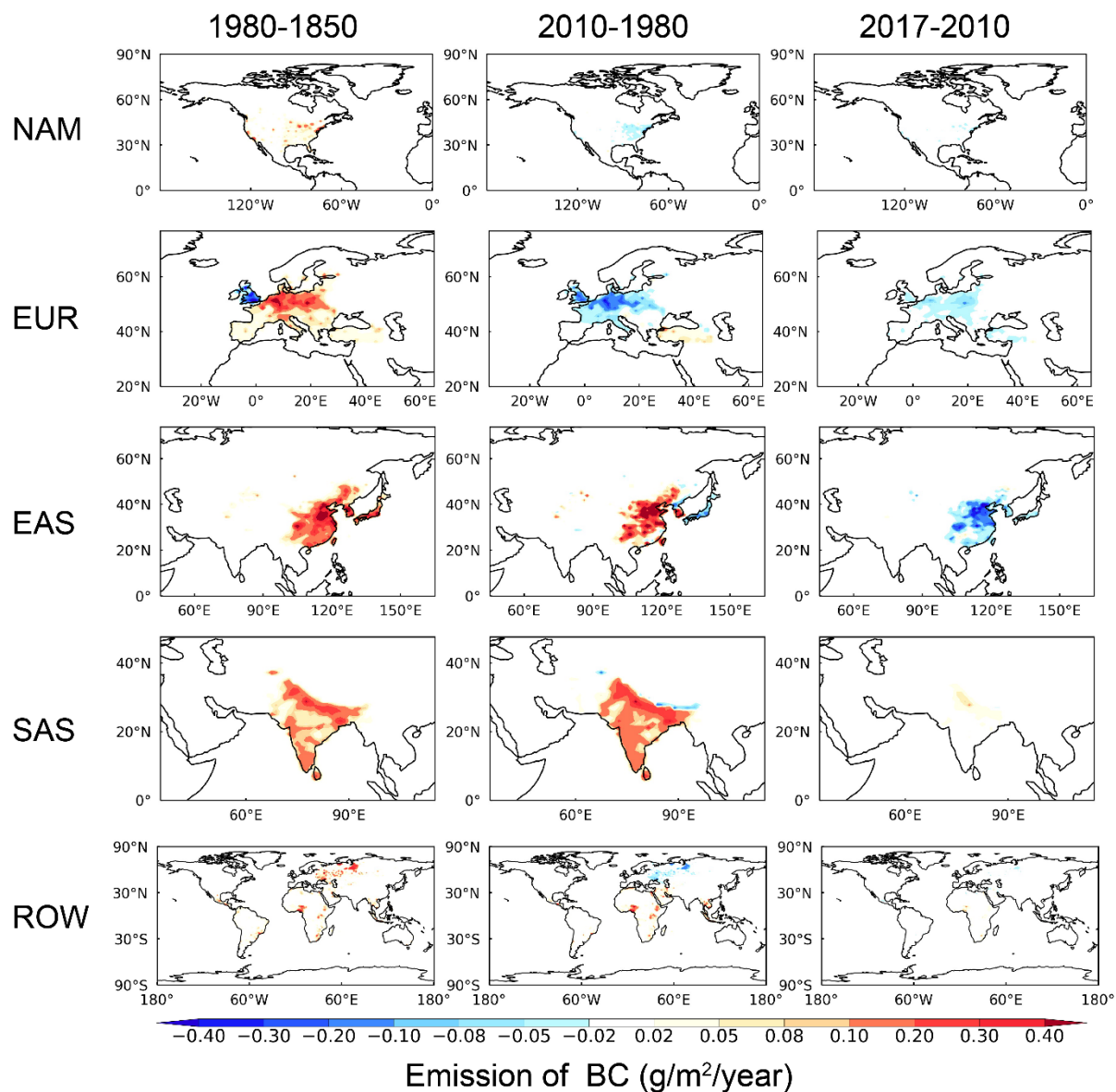
*Supplement of*

## **Global source apportionment of aerosols into major emission regions and sectors over 1850–2017**

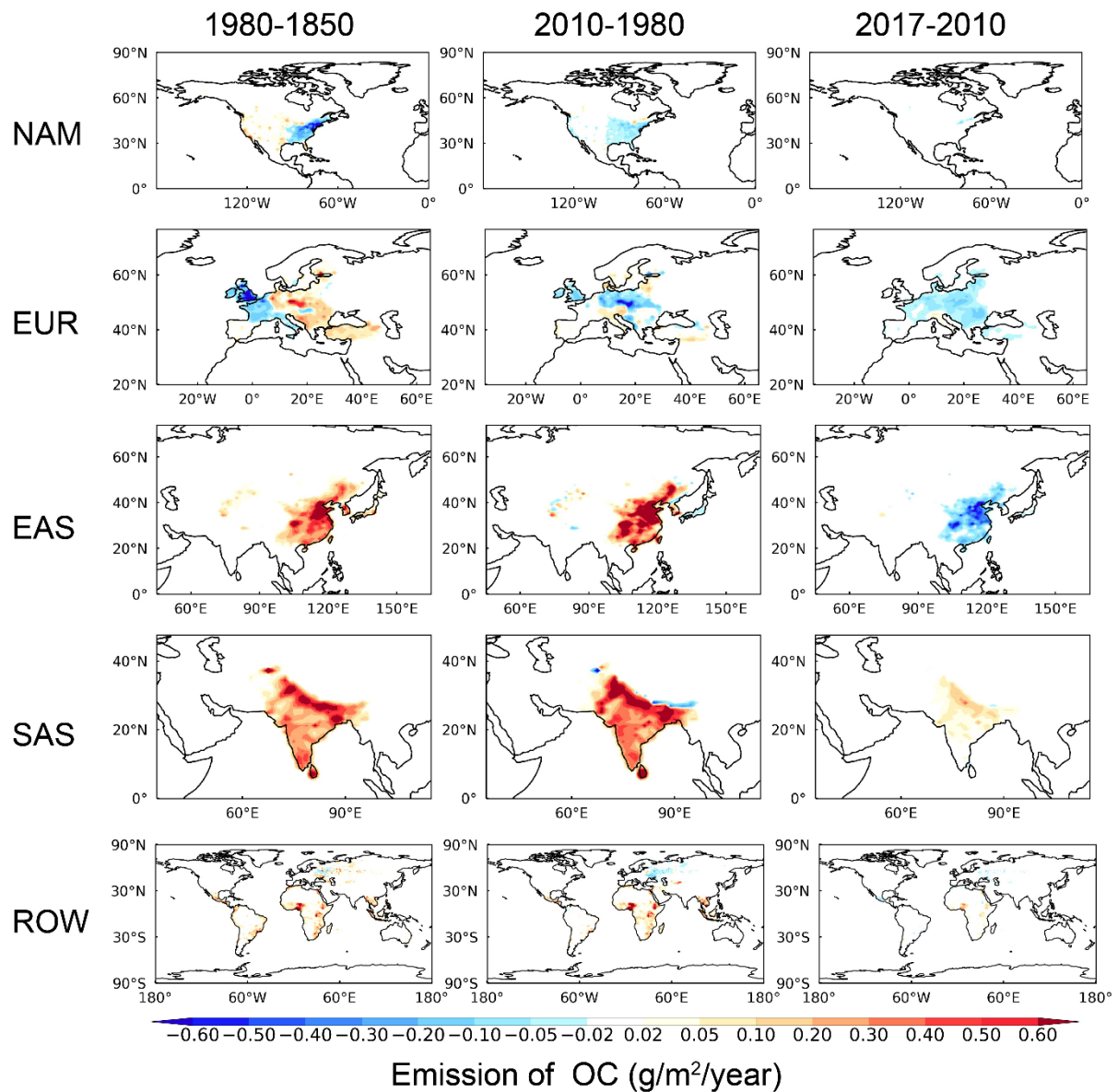
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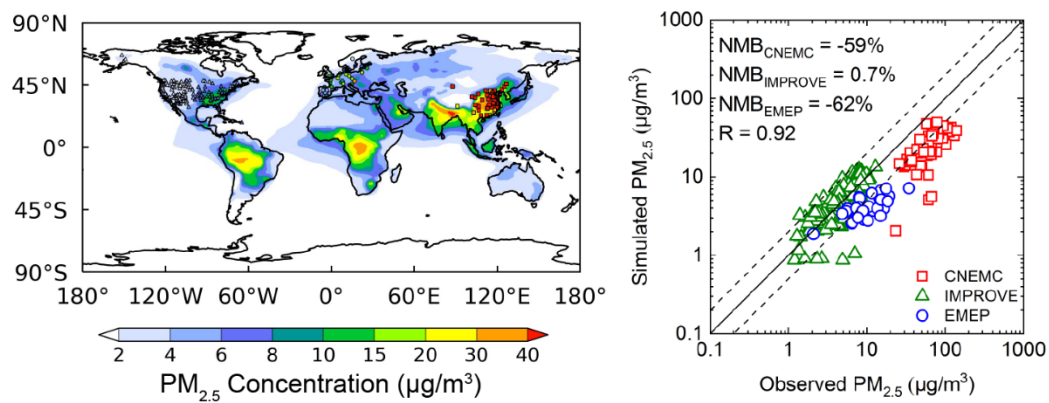
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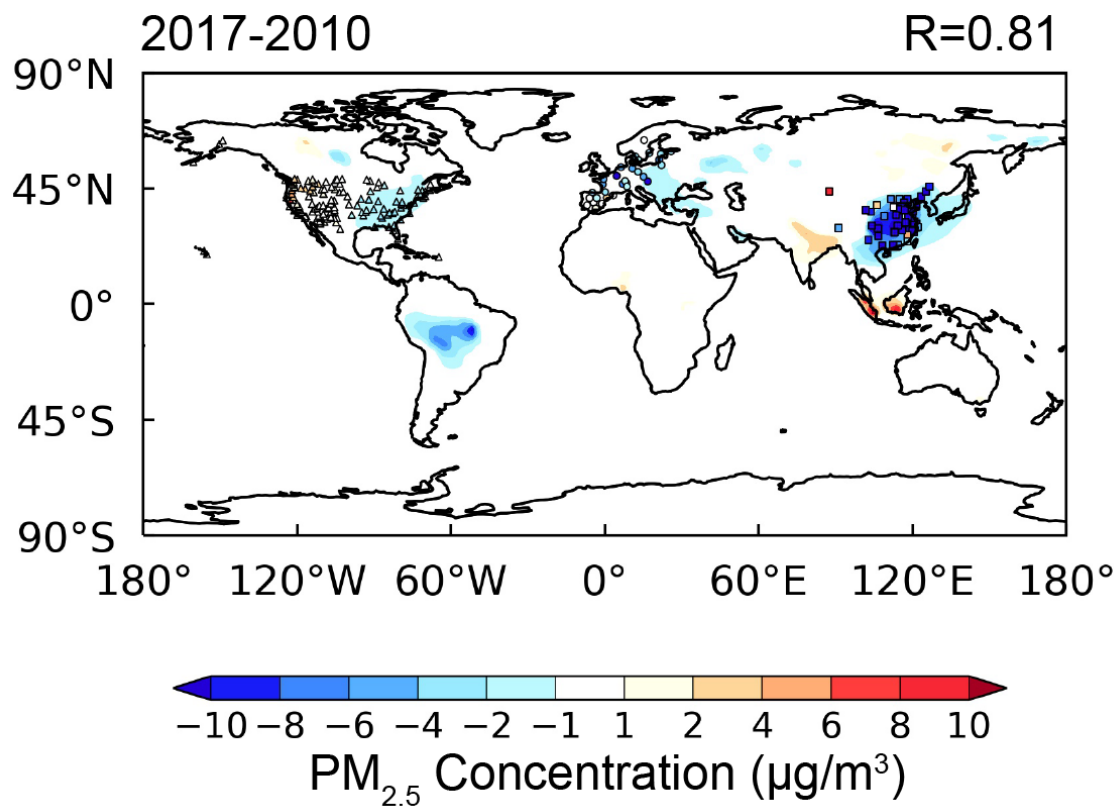
**Figure S1.** Changes in anthropogenic emissions of black carbon (BC) between 1850 and 1980 (left), between 1980 and 2010 (middle), and between 2010 and 2017 (right) in the 5 tagged source regions (NAM, EUR, EAS, SAS and ROW from top to bottom).



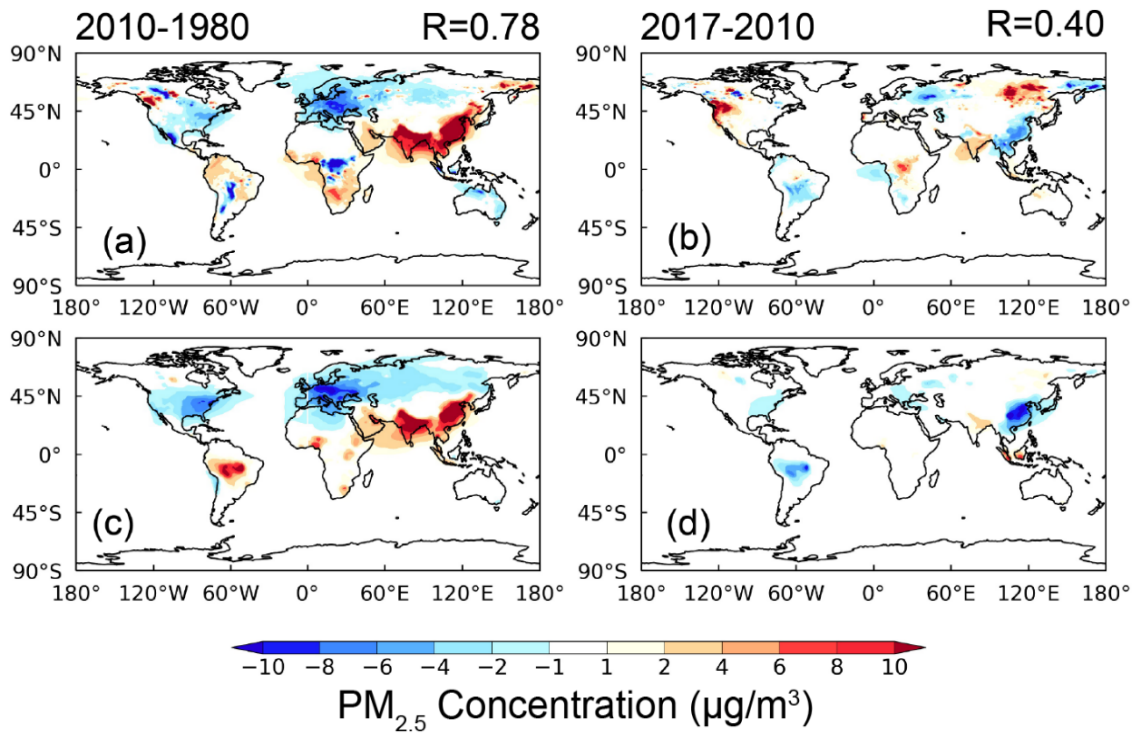
**Figure S2.** Changes in anthropogenic emissions of organic carbon (OC) between 1850 and 1980 (left), between 1980 and 2010 (middle), and between 2010 and 2017 (right) in the 5 tagged source regions (NAM, EUR, EAS, SAS and ROW from top to bottom).



**Figure S3.** Spatial distribution (left panel) and scatter plot (right) between the simulated and observed annual mean near-surface  $\text{PM}_{2.5}$  concentrations ( $\mu\text{g}/\text{m}^3$ ) in 2010. Observational data are from IMPROVE (triangle), EMEP (circle) and CNEMC (square, in 2013). The solid line marks the 1:1 ratio and dashed lines mark the 1:2 and 2:1 ratios. Normalized mean bias (NMB) and correlation coefficient (R) between observation and simulation are shown on the right panel.  $\text{NMB} = 100\% \times \sum(M_i - O_i) / \sum O_i$ , where  $M_i$  and  $O_i$  are the modeled and observed values at site  $i$ , respectively.



**Figure S4.** Spatial distribution of the change in simulated and observed annual mean near-surface PM<sub>2.5</sub> concentrations (µg/m<sup>3</sup>) between 2010 and 2017. Observational data are from IMPROVE (triangle), EMEP (circle) and CNEMC (square, between 2013 and 2017).



**Figure S5.** Spatial distribution of the changes in annual mean near-surface PM<sub>2.5</sub> concentrations ( $\mu\text{g}/\text{m}^3$ ) from (a, b) MERRA-2 and (c, d) model simulation between 1980 and 2010 (left) and between 2010 and 2017 (right).