

## Supplementary Material

### Changes in PM<sub>2.5</sub> concentrations and their sources in the US from 1990 to 2010

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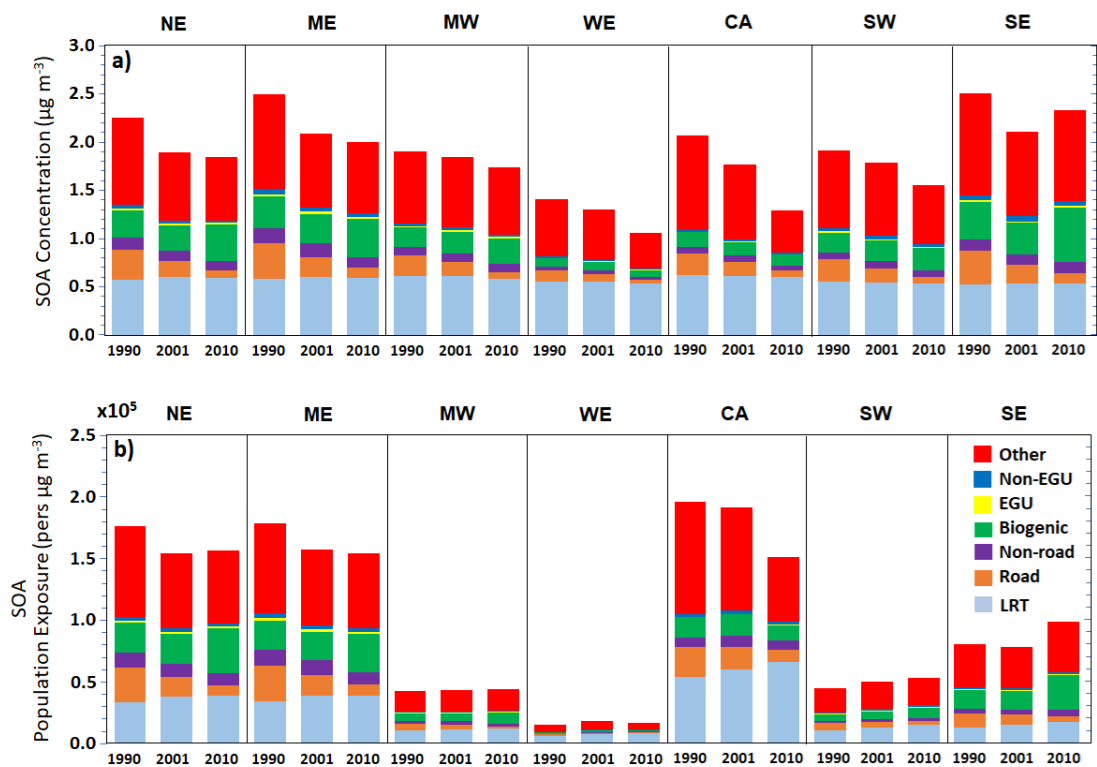
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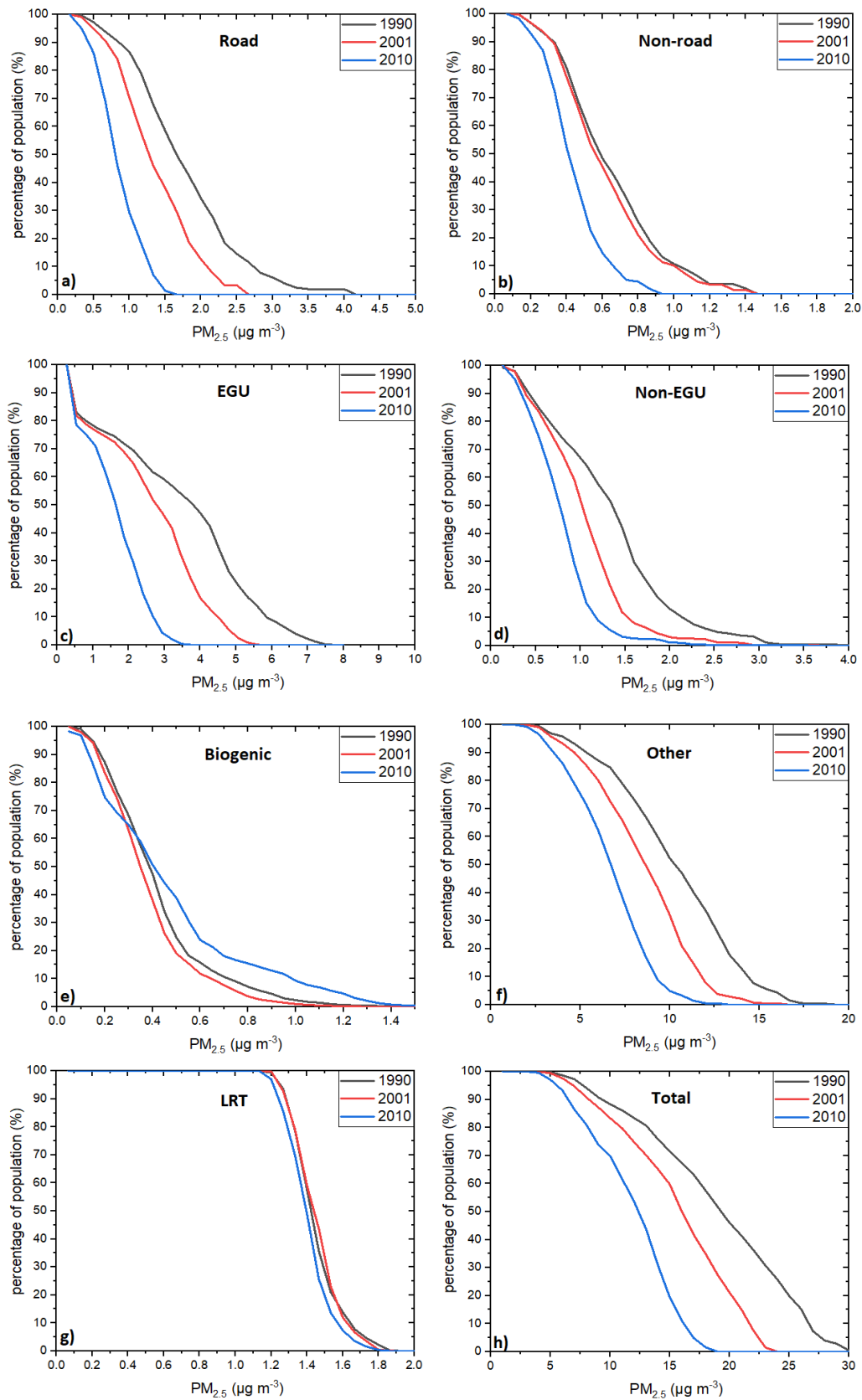
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**Table S1:** Evaluation metrics for annual average concentrations of PM<sub>2.5</sub> for each region and for each examined year (the analysis for 1990 is not included due to the very few stations in some regions).

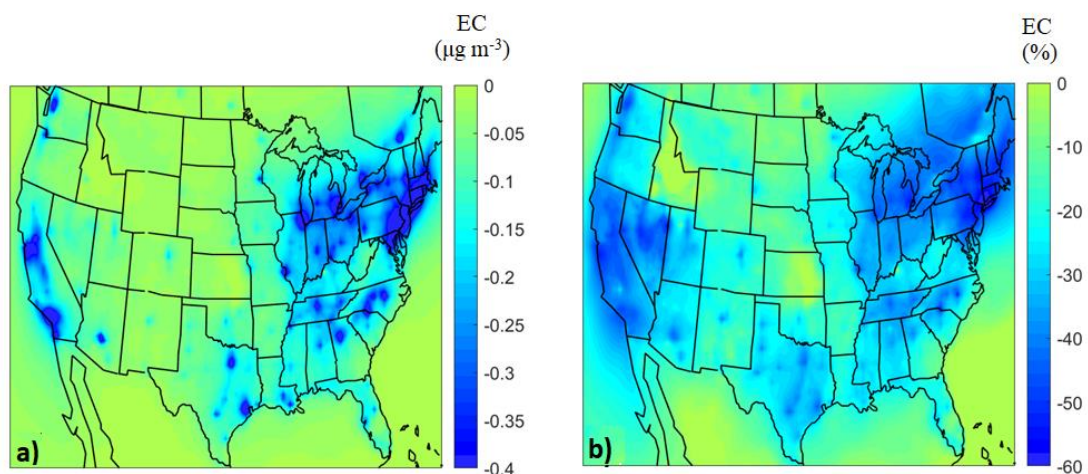
	<b>MB</b> ( $\mu\text{g m}^{-3}$ )	<b>MAGE</b> ( $\mu\text{g m}^{-3}$ )	<b>NMB</b>	<b>NME</b>	<b>FBIAS</b>	<b>FERROR</b>	<b>Stations</b>
<b>NE</b>							
<b>2001</b>	2.53	2.95	0.18	0.21	0.17	0.20	208
<b>2010</b>	2.21	2.41	0.22	0.24	0.21	0.23	235
<b>ME</b>							
<b>2001</b>	2.93	3.26	0.19	0.22	0.18	0.20	152
<b>2010</b>	2.25	2.42	0.19	0.21	0.18	0.19	156
<b>MW</b>							
<b>2001</b>	2.83	3.06	0.26	0.29	0.21	0.24	125
<b>2010</b>	1.41	1.79	0.15	0.18	0.13	0.18	131
<b>WE</b>							
<b>2001</b>	-1.69	2.63	-0.22	0.35	-0.18	0.37	154
<b>2010</b>	-1.82	2.43	-0.29	0.39	-0.25	0.43	141
<b>CA</b>							
<b>2001</b>	-5.06	5.27	-0.39	0.41	-0.38	0.43	79
<b>2010</b>	-2.21	2.80	-0.25	0.32	-0.22	0.36	120
<b>SW</b>							
<b>2001</b>	2.01	3.33	0.21	0.35	0.13	0.30	110
<b>2010</b>	0.23	1.74	0.03	0.24	0.03	0.23	82
<b>SE</b>							
<b>2001</b>	0.61	1.60	0.05	0.12	0.04	0.12	202
<b>2010</b>	1.13	1.49	0.11	0.14	0.10	0.13	193



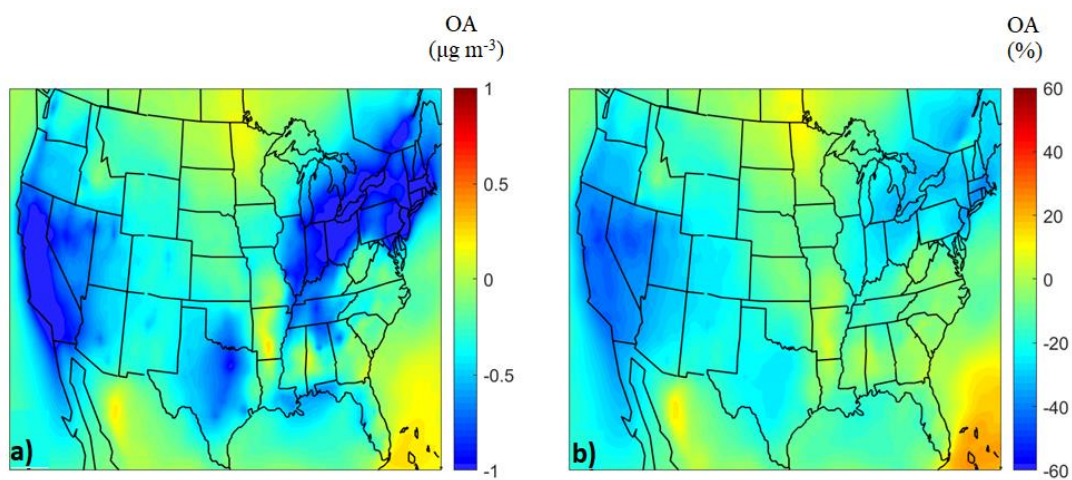
**Figure S1:** Sources of annual average SOA (aSOA+bSOA) for the different regions during 1990, 2001, and 2010 for: a) concentrations ( $\mu\text{g m}^{-3}$ ) and b) population exposure (persons  $\mu\text{g m}^{-3}$ ).



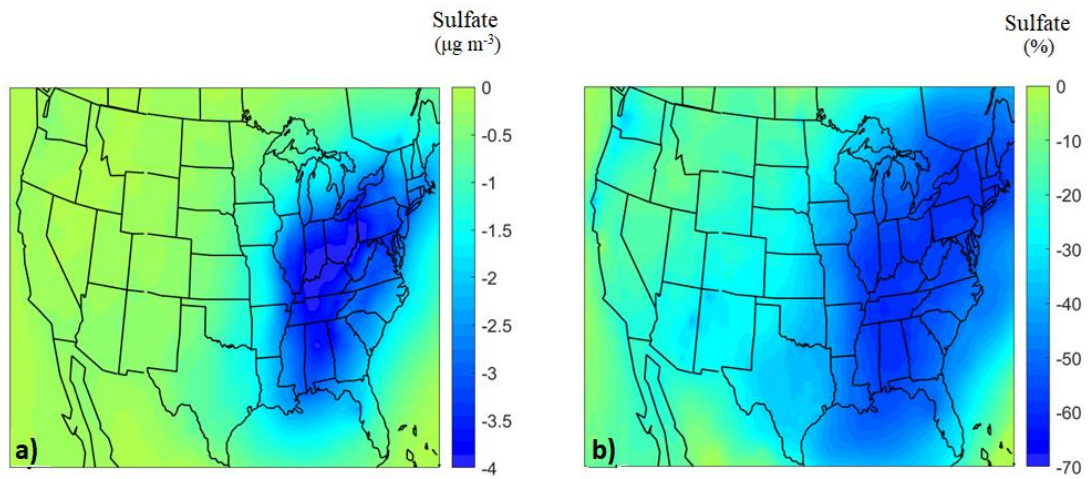
**Figure S2:** Cumulative distributions of annual average  $PM_{2.5}$  during 1990 (grey), 2001 (red), 2010 (blue); and for each source of  $PM_{2.5}$ : a) road transport, b) non-road transport, c) EGU, d) non-EGU, e) biogenic, f) other, g) LRT, and h) total  $PM_{2.5}$ .



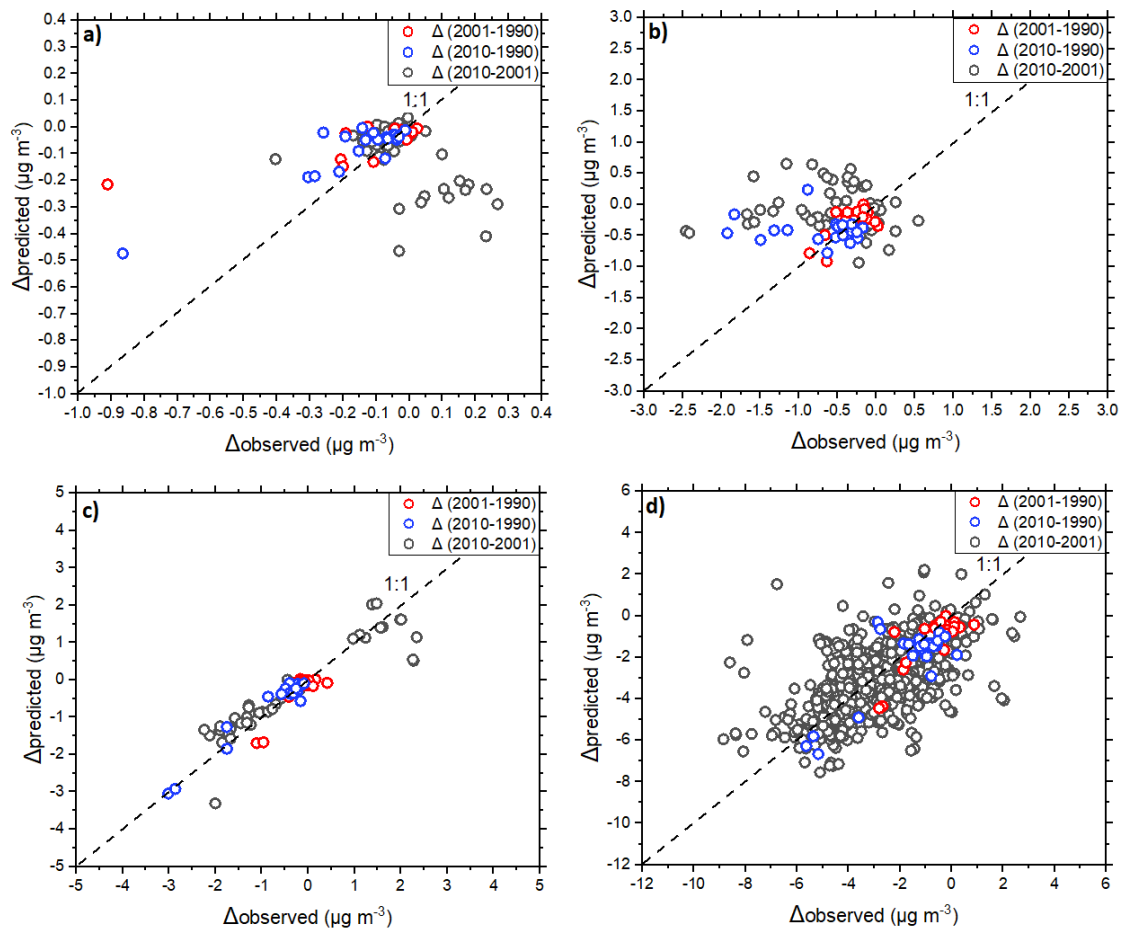
**Figure S3:** Changes in PM<sub>2.5</sub> EC concentrations, cool colors indicate reduction from 1990 to 2010; a) changes in  $\mu\text{g m}^{-3}$  and b) percent changes (%).



**Figure S4:** Changes in PM<sub>2.5</sub> OA concentrations, cool colors indicate reduction from 1990 to 2010; a) changes in  $\mu\text{g m}^{-3}$  and b) percent changes (%).



**Figure S5:** Changes in PM<sub>2.5</sub> sulfate concentrations, cool colors indicate reduction from 1990 to 2010; a) changes in  $\mu\text{g m}^{-3}$  and b) percent changes (%).



**Figure S6:** Predicted changes compared to observed, shown together for the 3 different cases, from 1990 to 2001, from 1990 to 2010, and from 2001 to 2010; the different figures stand for different  $\text{PM}_{2.5}$  components: a) elemental carbon, b) OA, c) sulfate, and d) total  $\text{PM}_{2.5}$ .