



Supplement of

Changes in PM_{2.5} concentrations and their sources in the US from 1990 to 2010

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Table S1: Evaluation metrics for annual average concentrations of PM_{2.5} for each region and for each examined year (the analysis for 1990 is not included due to the very few stations in some regions).

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

Table S2: Evaluation metrics for annual average concentrations of PM_{2.5} and for its major components for each examined year, except CA region.

	MB ($\mu\text{g m}^{-3}$)	MAGE ($\mu\text{g m}^{-3}$)	NMB	NME	FBIAS	FERROR	Stations
Without CA							
EC							
1990	-0.02	0.07	-0.07	0.27	0.04	0.28	27
2001	0.14	0.19	0.42	0.59	0.28	0.38	104
2010	-0.05	0.15	-0.10	0.34	0.05	0.37	268
OA							
1990	0.01	0.47	0.01	0.26	0.07	0.26	27
2001	-0.21	0.60	-0.09	0.25	-0.01	0.27	103
2010	0.20	0.50	0.09	0.23	0.09	0.23	269
Sulfate							
1990	0.13	0.23	0.09	0.15	0.20	0.23	27
2001	0.19	0.40	0.14	0.29	0.31	0.38	101
2010	0.07	0.31	0.04	0.17	0.16	0.25	287
Nitrate							
1990	0.01	0.18	0.03	0.65	-0.31	0.59	27
2001	-0.15	0.32	-0.14	0.30	-0.19	0.47	97
2010	-0.25	0.32	-0.26	0.34	-0.34	0.49	282
Ammonium							
1990	-0.04	0.14	-0.06	0.22	0.07	0.24	27
2001	0.02	0.20	0.02	0.21	0.10	0.26	96
2010	0.09	0.16	0.11	0.20	0.19	0.26	286
PM_{2.5}							
1990	1.20	1.63	0.23	0.31	0.19	0.26	27
2001	1.49	2.72	0.12	0.23	0.09	0.23	951
2010	1.10	2.08	0.12	0.22	0.09	0.23	938

Table S3: Evaluation metrics for daily average concentrations of PM_{2.5} and for its major components for each examined year, except CA region.

	MB ($\mu\text{g m}^{-3}$)	MAGE ($\mu\text{g m}^{-3}$)	NMB	NME	FBIAS	FERROR	Points	Comment
Without CA								
EC								
1990	-0.03	0.14	-0.11	0.48	0.11	0.55	2373	Average ^a
2001	0.12	0.26	0.31	0.72	0.37	0.61	16490	Average
2010	-0.04	0.22	-0.09	0.52	0.21	0.58	25869	Average
OA								
1990	-0.11	0.93	-0.06	0.49	0.05	0.49	2443	Good
2001	-0.35	1.29	-0.14	0.51	0.06	0.54	16435	Average
2010	0.11	1.16	0.05	0.56	0.18	0.53	25858	Average
Sulfate								
1990	0.13	0.65	0.09	0.46	0.28	0.50	2607	Good
2001	0.01	0.99	0.00	0.44	0.25	0.52	15813	Average
2010	0.18	0.79	0.12	0.51	0.32	0.57	29294	Average
Nitrate								
1990	0.01	0.30	0.05	1.13	-0.74	1.29	2428	Problematic
2001	-0.09	0.65	-0.12	0.81	-0.57	1.07	15771	Problematic
2010	-0.22	0.58	-0.25	0.64	-0.70	1.09	27118	Problematic
Ammonium								
1990	-0.04	0.29	-0.07	0.46	0.13	0.49	2425	Good
2001	0.02	0.47	0.02	0.51	0.26	0.55	15589	Average
2010	0.09	0.38	0.12	0.52	0.34	0.58	26445	Average
PM_{2.5}								
1990	0.98	2.64	0.19	0.51	0.18	0.46	2184	Good
2001	1.82	5.29	0.15	0.45	0.16	0.43	149851	Good
2010	0.50	4.24	0.05	0.44	0.07	0.45	185862	Good

^a Following Morris et al. (2005) criteria: Good: FBIAS $\leq \pm 0.30$, FERROR ≤ 0.50 ; Average: FBIAS $\leq \pm 0.60$, FERROR ≤ 0.75 ; Problematic: FBIAS $> \pm 0.60$, FERROR > 0.75

Table S4: Evaluation metrics for annual average concentrations of PM_{2.5} and its major components for each examined year (all sites are included).

	NMB	NME	r	Points
EC				
1990	-0.01	0.23	0.95	33
2001	0.39	0.56	0.82	122
2010	-0.11	0.35	0.76	304
OA				
1990	-0.02	0.21	0.97	33
2001	-0.12	0.28	0.88	121
2010	0.02	0.27	0.66	306
Sulfate				
1990	0.09	0.16	0.99	33
2001	0.13	0.30	0.94	118
2010	0.05	0.18	0.92	327
Nitrate				
1990	-0.30	0.65	0.39	33
2001	-0.24	0.37	0.97	114
2010	-0.35	0.41	0.75	321
Ammonium				
1990	-0.09	0.25	0.89	33
2001	0.00	0.23	0.96	113
2010	0.08	0.23	0.88	326
PM_{2.5}				
1990	0.16	0.26	0.96	33
2001	0.08	0.24	0.71	1040
2010	0.07	0.22	0.77	1067

Table S5: Evaluation metrics for daily average concentrations of PM_{2.5} and its major components for each examined year (all sites are included).

	NMB	NME	r	Points	criteria
EC					
1990	-0.11	0.53	0.68	2940	yes
2001	0.28	0.71	0.51	18763	yes
2010	-0.10	0.54	0.56	29423	yes
OA					
1990	-0.11	0.53	0.48	2940	yes
2001	-0.17	0.53	0.40	18706	yes
2010	-0.01	0.56	0.35	29412	yes
Sulfate					
1990	0.11	0.47	0.81	3228	yes
2001	-0.01	0.45	0.76	18077	yes
2010	0.12	0.52	0.64	33051	no
Nitrate					
1990	-0.26	0.99	0.30	2998	yes
2001	-0.31	0.83	0.31	18019	yes
2010	-0.34	0.68	0.58	30867	yes
Ammonium					
1990	-0.09	0.48	0.69	2996	yes
2001	-0.03	0.54	0.59	17828	no
2010	0.08	0.54	0.62	30162	no
PM_{2.5}					
1990	0.13	0.50	0.65	2706	yes
2001	0.11	0.46	0.54	161909	yes
2010	0.00	0.45	0.51	212899	yes

^a Following Emery et al. (2017), the model meets the acceptable criteria: for PM_{2.5}; sulfate and ammonium if NMB < ± 0.30, NME < 0.50, r > 0.4; for nitrate if NMB < ± 0.65, NME < 1.15; for OM if NMB < ± 0.50, NME < 0.65; for EC if NMB < ± 0.40, NME < 0.75

Table S6: Evaluation metrics for daily average concentrations of PM_{2.5} and its major components for each examined year, for two cases: except CA and only CA region.

	NMB		NME		r		Points		criteria	
	without CA	CA	without CA	CA	without CA	CA	without CA	CA	without CA	CA
EC										
1990	-0.11	-0.10	0.48	0.67	0.81	0.28	2373	567	yes ^a	yes ^a
2001	0.31	0.06	0.72	0.65	0.51	0.59	16490	2117	yes	yes
2010	-0.09	-0.17	0.52	0.67	0.58	0.49	25869	3257	yes	yes
OA										
1990	-0.06	-0.14	0.49	0.54	0.52	0.36	2443	589	yes	yes
2001	-0.14	-0.36	0.51	0.64	0.44	0.27	16435	2117	yes	yes
2010	0.05	-0.40	0.56	0.61	0.39	0.25	25858	3261	yes	yes
Sulfate										
1990	0.09	0.23	0.46	0.56	0.81	0.62	2607	621	yes	no
2001	0.00	-0.13	0.44	0.59	0.77	0.53	15813	2108	yes	no
2010	0.12	0.18	0.51	0.65	0.63	0.42	29294	3453	no	no
Nitrate										
1990	0.05	-0.61	1.13	0.83	0.42	0.34	2428	570	yes	yes
2001	-0.12	-0.83	0.81	0.91	0.46	0.29	15771	2096	yes	no
2010	-0.25	-0.79	0.64	0.88	0.69	0.23	27118	3437	yes	no
Ammonium										
1990	-0.07	-0.35	0.46	0.62	0.73	0.25	2425	570	yes	no
2001	0.02	-0.44	0.51	0.73	0.66	0.34	15589	2085	no	no
2010	0.12	-0.28	0.52	0.76	0.68	0.22	26445	3416	no	no
PM_{2.5}										
1990	0.19	-0.04	0.51	0.47	0.70	0.53	2184	522	yes	no
2001	0.15	-0.50	0.45	0.61	0.59	0.30	149851	8213	yes	no
2010	0.05	-0.46	0.44	0.82	0.56	-0.05	185862	17030	yes	no

^a Following Emery et al. (2017), the model meets the acceptable criteria: for PM_{2.5}; sulfate and ammonium if NMB < ± 0.30, NME < 0.50, r > 0.4; for nitrate if NMB < ± 0.65, NME < 1.15; for OM if NMB < ± 0.50, NME < 0.65; for EC if NMB < ± 0.40, NME < 0.75

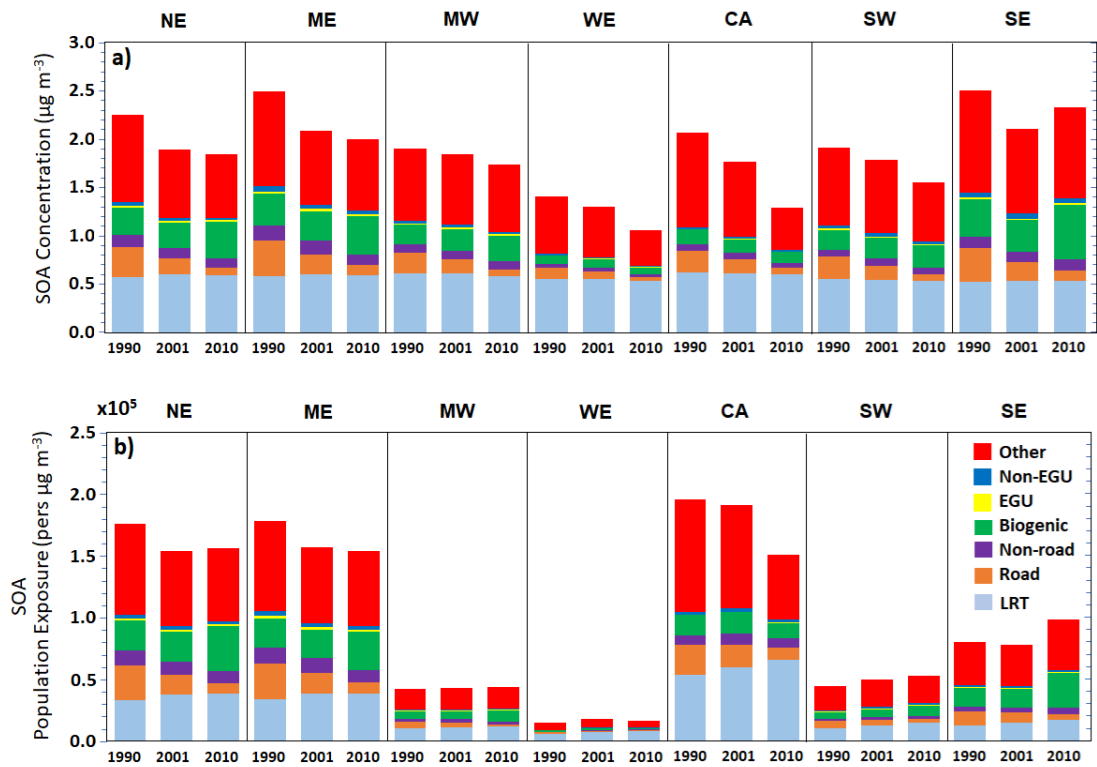


Figure S1: Sources of annual average SOA (aSOA+bSOA) for the different regions during 1990, 2001, and 2010 for: a) concentrations ($\mu\text{g m}^{-3}$) and b) population exposure (persons $\mu\text{g m}^{-3}$).

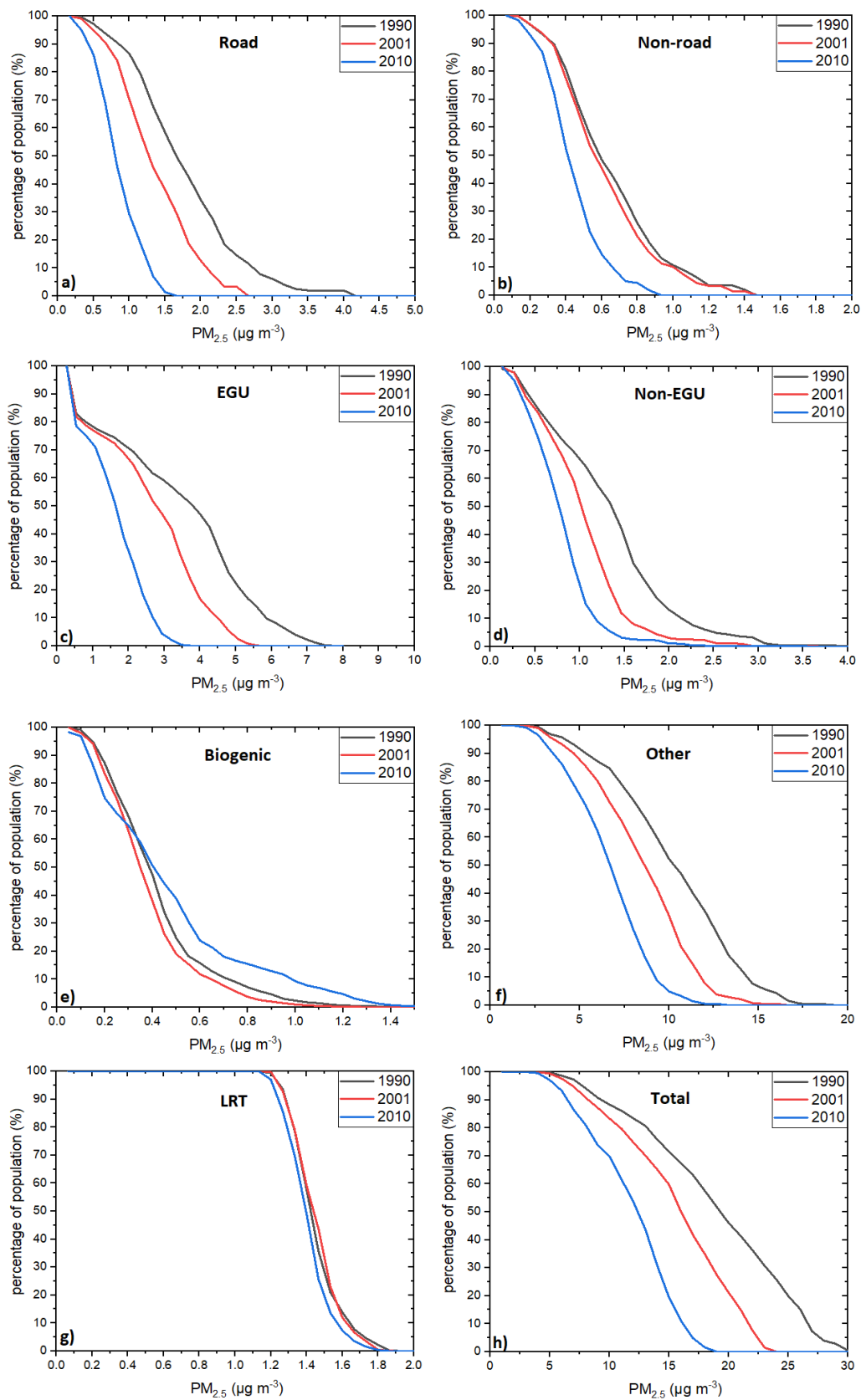


Figure S2: Cumulative distributions of annual average PM_{2.5} during 1990 (grey), 2001 (red), 2010 (blue); and for each source of PM_{2.5}: a) road transport, b) non-road transport, c) EGU, d) non-EGU, e) biogenic, f) other, g) LRT, and h) total PM_{2.5}.

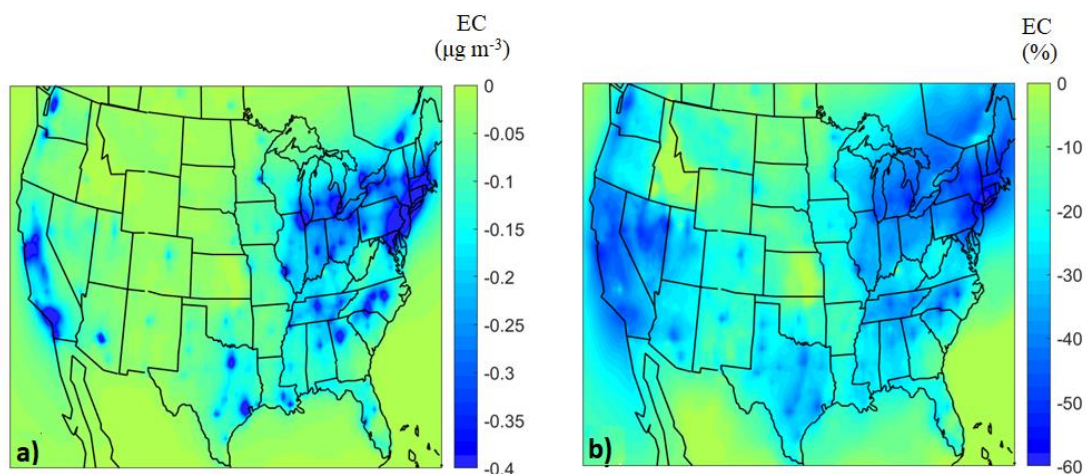


Figure S3: Changes in PM_{2.5} EC concentrations, cool colors indicate reduction from 1990 to 2010; a) changes in $\mu\text{g m}^{-3}$ and b) percent changes (%).

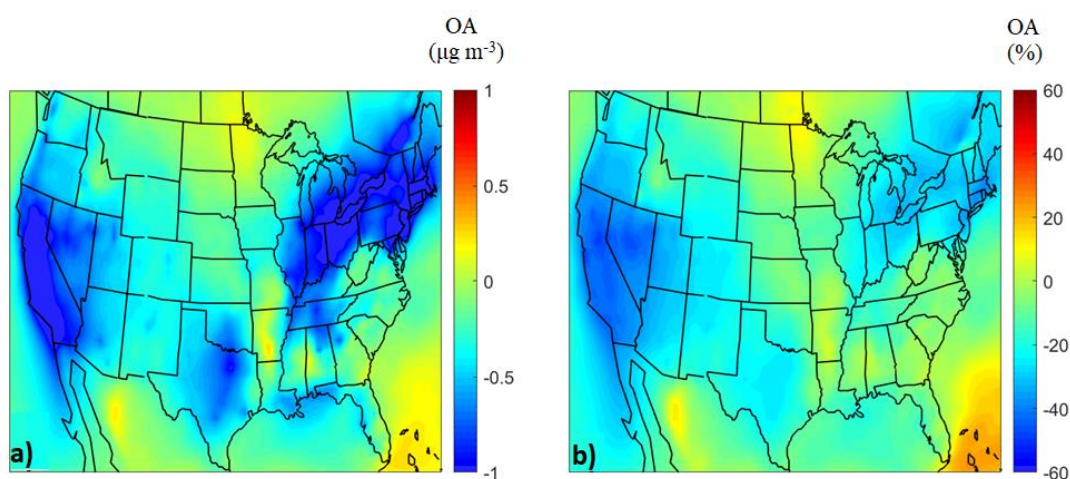


Figure S4: Changes in PM_{2.5} OA concentrations, cool colors indicate reduction from 1990 to 2010; a) changes in $\mu\text{g m}^{-3}$ and b) percent changes (%).

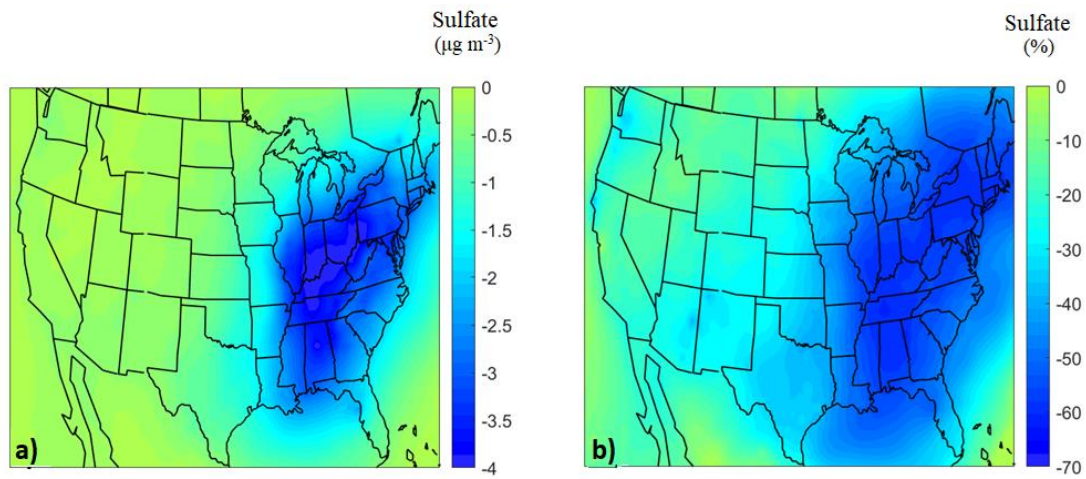


Figure S5: Changes in PM_{2.5} sulfate concentrations, cool colors indicate reduction from 1990 to 2010; a) changes in $\mu\text{g m}^{-3}$ and b) percent changes (%).

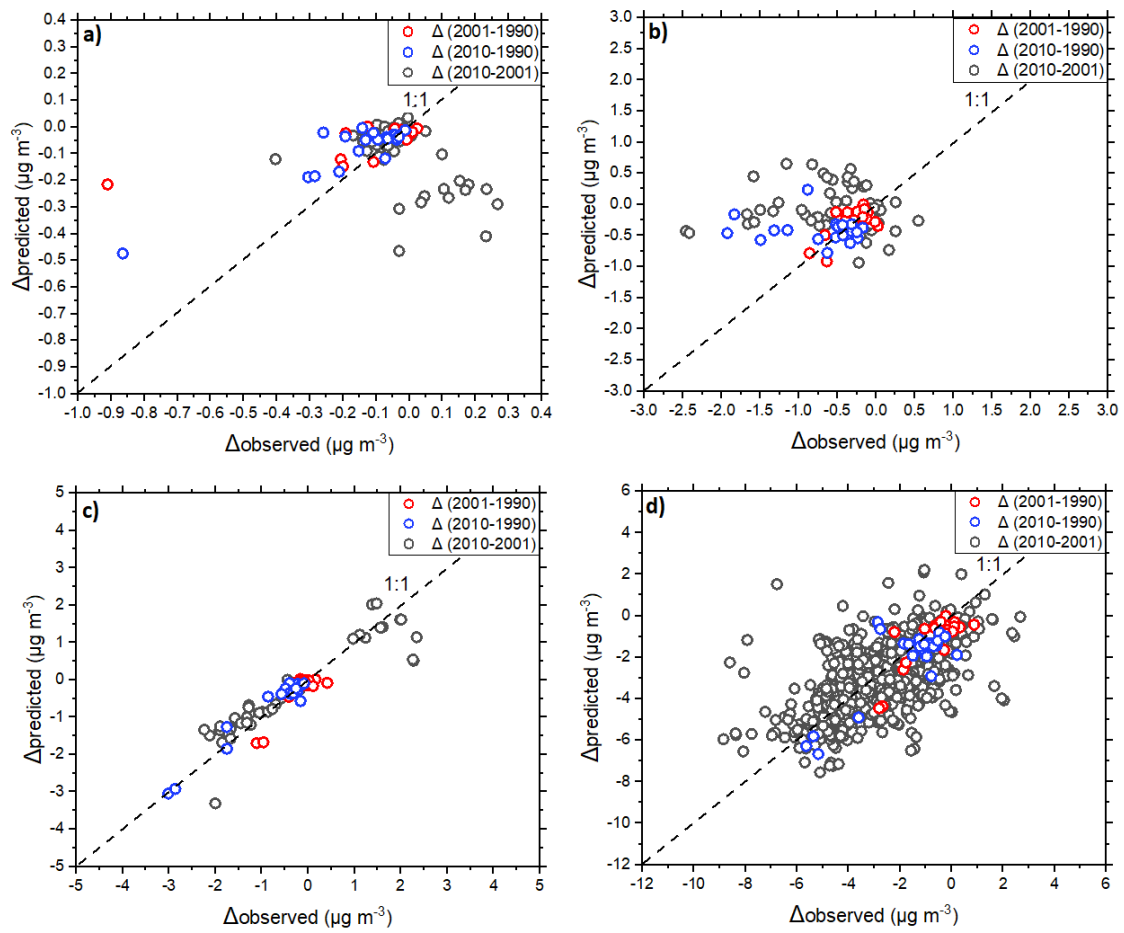


Figure S6: Predicted changes compared to observed, shown together for the 3 different cases, from 1990 to 2001, from 1990 to 2010, and from 2001 to 2010; the different figures stand for different $\text{PM}_{2.5}$ components: a) elemental carbon, b) OA, c) sulfate, and d) total $\text{PM}_{2.5}$.