

Supporting Information for

Historical gaseous and primary aerosol emissions in
the United States from 1990-2010

Table S1 Sectors grouped by NEI SCC in this study

(a) Power plants

Fuel	SCC	Explanation	Controlled species
Coal	101001XX	External Combustion Boilers	Anthracite Coal
	101002XX		Bituminous/ Subbituminous Coal
	101003XX		Lignite
	2101002XXX	Electric Utility	Bituminous/ Subbituminous Coal
Residual oil	101004XX	External Combustion Boilers	Residual Oil
	2101005XXX	Electric Utility	
Distillate oil	101005XX	External Combustion Boilers;	Distillate Oil
	201001XX	Internal Combustion Engines	
	2101004XXX	Electric Utility	
Natural gas	101006XX	External Combustion Boilers	Natural Gas
	201002XXX	Internal Combustion Engines	

(b) Industry combustion

Fuel	SCC	Explanation	Controlled species
Coal	102001XXX	External Combustion Boilers	Anthracite Coal
	102002XXX		Bituminous/ Subbituminous Coal
	102003XXX		Lignite
	2102001XXX	Stationary Source	Anthracite Coal
	2102002XXX		Bituminous/ Subbituminous Coal
Residual oil	102004XXX	External Combustion Boilers	Residual Oil
	2102005XXX	Stationary Source	
Distillate oil	102005XXX	External Combustion Boilers	Distillate Oil
	202001XXX	Internal Combustion Engines	
	2102004XXX	Stationary Source	
Natural gas	102006XXX	External Combustion Boilers	Natural Gas

202002XXX	Internal Combustion Engines
2102006XXX	Stationary Source

(c) Commercial combustion

Fuel	SCC	Explanation	Controlled species
Coal	103001XXX	External Combustion Boilers	Anthracite Coal
	103002XXX		Bituminous/ Subbituminous Coal
	2103001XXX	Stationary Source Fuel Combustion	Anthracite Coal
	2103002XXX		Bituminous/ Subbituminous Coal
Residual oil	103004XXX	External Combustion Boilers	Residual Oil
	2103005XXX	Stationary Source Fuel Combustion	
Distillate oil	103005XXX	External Combustion Boilers	Distillate Oil
	2103004XXX	Stationary Source Fuel Combustion	
Natural gas	103006XXX	External Combustion Boilers	Natural Gas
	2103006XXX	Stationary Source Fuel Combustion	
LPG	103010XXX	External Combustion Boilers	Liquified Petroleum Gas (LPG)
	2103007XXX	Stationary Source Fuel Combustion	
Kerosene	2103011XXX	Stationary Source Fuel Combustion	Kerosene

(d) Residential combustion

Fuel	SCC	Explanation	Controlled species
Coal	2104001XXX	Stationary Source Fuel Combustion	Anthracite Coal
	2104002XXX		Bituminous/ Subbituminous Coal
Distillate oil	2104004XXX	Stationary Source Fuel Combustion	Distillate Oil
Natural gas	2104006XXX		Natural Gas
LPG	2104007XXX		Liquified Petroleum Gas (LPG)

Wood	2104008XXX	Wood	PM
Kerosene	2104011XXX	Kerosene	PM

(e) On road transportation

Type	SCC	Explanation	Controlled species
Light Duty Vehicle	220100XXXX	Light Duty Gasoline Vehicles	NO _x , SO ₂ , CO, VOC, PM
	223000XXXX	Light Duty Diesel Vehicles	
Light Duty Truck	220102XXXX	Light Duty Gasoline Trucks 1 & 2	NO _x , SO ₂ , CO, VOC, PM
	220104XXXX	Light Duty Gasoline Trucks 3 & 4	
	223006XXXX	Light Duty Diesel Trucks 1 thru 4 (M6) (LDDT)	
Heavy duty	220107XXXX	Heavy Duty Gasoline Vehicles	NO _x , SO ₂ , CO, VOC, PM
	223007XXXX	Heavy Duty Diesel Vehicles (HDDV) Class 2B	
Motorcycle	220108XXXX	Motorcycles (MC)	NO _x , SO ₂ , CO, VOC, PM

(f) Off road transportation

Fuel	Type	SCC	Explanation	Controlled species	
Gasoline	Industry & Commercial	2260003XXXX	Industrial Equipment	SO ₂	
		2265003XXXX			
		2265010XXXX			
	Construction	2260006XXXX	Commercial Equipment		
		2265006XXXX			
	Agriculture	2260002XXXX	Construction and Mining Equipment		
		2265002XXXX			
	Marine, gasoline	2260005XXXX	Agricultural Equipment		
		2265005XXXX			
	Other	Marine, gasoline	2280004XXXX		Marine Vessels, Gasoline
			2260008XXXX		Airport Ground Support Equipment
		Other	2265008XXXX		Recreational Equipment
			2260001XXXX		
2265001XXXX					
Other		2260004XXXX	Lawn and Garden Equipment		
		2265004XXXX			
Other	2260007XXXX	Logging Equipment			
	2265007XXXX				
Distillate oil	Rail	2285XXXXXX	Class I Operations		
	Marine, diesel	2280002XXXX	Marine Vessels, Diesel		
	Other diesel	2270XXXXXX	Other diesel		

Residual oil	Marine	2280003XXXX	Marine Vessels, Residual oil	-
Natural gas	All Transport	2268XXXXXX	CNG	-
LPG	All Transport	2267XXXXXX	LPG	-
Jet fuel	Aviation	2275XXXXXX	Aircraft	-

(g) Industry processes

Category	SCC	Explanation	
Chemical Manufacturing	301XXXXX	Chemical Manufacturing	Point
	2301XXXXXX		Area
Metals processing	303XXXXX	Primary Metal Production	Point
	304XXXXX	Secondary Metal Production	
	309XXXXX	Fabricated Metal Products	Area
	2303XXXXXX	Primary Metal Production	
	2304XXXXXX	Secondary Metal Production	
	2309XXXXXX	Fabricated Metal Products	
Petroleum & related industries	306XXXXX	Petroleum Refining	Point
	310XXXXX	Oil and Gas Production	Area
	2306XXXXXX	Petroleum Refining	
	2310XXXXXX	Oil and Gas Production	
Other industry processes	302XXXXX	Food and Agriculture	Point
	305XXXXX	Mineral Products	
	307XXXXX	Pulp and Paper and Wood Products	
	308XXXXX	Rubber and Miscellaneous Plastics	
	311XXXXX	Building Construction	
	312XXXXX	Machinery	
	313XXXXX	Electrical Equipment	
	314XXXXX	Transportation Equipment	
	315XXXXX	Photo Equip/Health Care/Labs/Air Condit/SwimPools	
	32XXXXXX	Leather and Leather Products	
	33XXXXXX	Textile Products	
	36XXXXXX	Printing and Publishing	
	38XXXXXX	Cooling Tower	
	39XXXXXX	Miscellaneous	
	2302XXXXXX	Food and Kindred Products	
2305XXXXXX	Mineral Processes		
2307XXXXXX	Wood Products		
2308XXXXXX	Rubber/Plastics		
2312XXXXXX	Machinery		
2313XXXXXX	Electrical Equipment		
239XXXXXX	Miscellaneous		

(h) Others

Category	SCC	Explanation		Controlled species
Solvent Utilization	401XXXXXX	Organic Solvent Evaporation	Point	VOC
	402XXXXXX	Surface Coating Operations		
	405XXXXXX	Printing/Publishing		
	410XXXXXX	Dry Cleaning		
	425XXXXXX	Loss		
	49XXXXXXX	Organic Solvent Evaporation		
	24XXXXXXXXX	Solvent Utilization		
Storage & Transport	403XXXXXX	Petroleum Product Storage at Refineries	Point	-
	404XXXXXX	Petroleum Liquids Storage (non-Refinery)		
	406XXXXXX	Transportation and Marketing of Petroleum Products		
	407XXXXXX	Organic Chemical Storage		
	408XXXXXX	Organic Chemical Transportation		
	25XXXXXXXXX	Storage and Transport		
Waste disposal & recycling	5XXXXXXXXX	Waste Disposal	Point	
	26XXXXXXXXX	Waste Disposal, Treatment, and Recovery	Area	-
	6XXXXXXXXX	MACT Source Categories	Point	-
Miscellaneous	2294XXXXXXXX	All Paved Roads	Mobile	PM
	2296XXXXXXXX	All Unpaved Roads		
	2311XXXXXXXX	Construction		
	2325XXXXXXXX	Mining and Quarrying		-
	2801XXXXXXXX	Agriculture Production - Crops		-
	2805XXXXXXXX	Agriculture Production - Livestock		NH ₃
	2810001XXX	Forest Wildfires		-
	2810015XXX	Prescribed Burning for Forest Management	Area	-
	2810XXXXXXXX	Other Combustion		-
	2820XXXXXXXX	Cooling Towers		-
	2830XXXXXXXX	Catastrophic/Accidental Releases		-
	284XXXXXXXXX	Repair Shops		-
	285XXXXXXXXX	Health Services		-

Table S2 Activity and emission trends during 1990-2010
(a) energy-related stationary sources (absolute values for Fig. 3)

Power plant		1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	
Coal	Activity	16.259	16.248	16.464	17.194	17.259	17.465	18.428	18.903	19.216	19.279	20.220	19.614	19.783	20.185	20.305	20.737	20.461	20.807	20.513	18.226	19.133	
	NOx	NEI	5.638					5.579	5.539			4.909		4.208	4.150			3.335					
		This study	5.837	5.694	5.677	5.843	5.753	5.629	5.550	5.637	5.553	4.953	4.667	4.325	4.156	3.886	3.604	3.335	3.158	2.981	2.811	1.878	1.943
	SO2	NEI	15.219					11.603	12.136			11.746		10.004	9.970			9.936					
		This study	15.565	15.396	14.963	14.853	14.511	11.982	12.274	12.696	12.873	11.839	10.941	10.341	10.134	10.029	9.926	9.935	9.153	8.328	7.442	5.528	4.970
Natural Gas	Activity	3.333	3.399	3.535	3.559	4.000	4.327	3.882	4.147	4.698	4.924	5.318	5.496	5.789	5.259	5.609	6.036	6.394	7.028	6.849	7.044	7.550	
	NOx	NEI	0.691					0.650	0.344			0.519		0.445	0.292			0.217					
		This study	0.414	0.405	0.417	0.417	0.467	0.487	0.354	0.362	0.420	0.425	0.409	0.375	0.276	0.210	0.219	0.208	0.224	0.249	0.241	0.247	0.271
	SO2	NEI	0.002					0.013	0.008			0.179		0.211	0.060			0.060					
		This study	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.002	0.001	0.001	0.002	0.002	0.002	0.002	0.002	0.002
Distillate fuel oil	Activity	0.097	0.084	0.074	0.086	0.120	0.108	0.109	0.111	0.136	0.140	0.175	0.171	0.127	0.161	0.111	0.115	0.074	0.089	0.073	0.070	0.080	
	NOx	NEI	0.084					0.033	0.029			0.054		0.052	0.084			0.078					
		This study	0.015	0.012	0.011	0.013	0.018	0.016	0.016	0.016	0.020	0.021	0.026	0.025	0.019	0.023	0.016	0.016	0.010	0.012	0.010	0.010	0.011
	SO2	NEI	0.057					0.056	0.056			0.042		0.046	0.029			0.030					
		This study	0.031	0.025	0.021	0.026	0.037	0.033	0.031	0.026	0.035	0.031	0.036	0.029	0.016	0.017	0.013	0.013	0.009	0.011	0.009	0.008	0.009
Residual fuel oil	Activity	1.163	1.085	0.872	0.959	0.869	0.566	0.628	0.715	1.047	0.959	0.871	1.003	0.659	0.869	0.879	0.876	0.361	0.397	0.240	0.181	0.154	
	NOx	NEI	0.192					0.094	0.094			0.185		0.156	0.129			0.108					
		This study	0.165	0.154	0.124	0.136	0.123	0.080	0.089	0.098	0.143	0.131	0.119	0.134	0.091	0.105	0.105	0.096	0.036	0.040	0.022	0.015	0.012
	SO2	NEI	0.603					0.408	0.410			0.559		0.492	0.345			0.369					
		This study	0.520	0.495	0.396	0.433	0.389	0.251	0.278	0.312	0.459	0.421	0.379	0.422	0.287	0.339	0.339	0.313	0.116	0.130	0.072	0.046	0.037

Industrial Combustion		1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	
Coal	Activity	2.754	2.600	2.512	2.500	2.507	2.500	2.438	2.396	2.254	2.188	2.259	2.194	2.020	2.044	2.046	1.954	1.914	1.864	1.792	1.394	1.666	
	NOx	NEI	0.511					0.560	0.485			0.407		0.419	0.343			0.316					
		This study	0.694	0.665	0.633	0.634	0.643	0.632	0.569	0.560	0.576	0.487	0.524	0.477	0.355	0.342	0.325	0.317	0.293	0.262	0.239	0.246	0.263
	SO2	NEI	0.082					0.091	0.103			0.109		0.113	0.126			0.119					
		This study	2.368	2.323	2.111	2.126	2.160	2.076	1.714	1.695	1.656	1.494	1.541	1.461	1.120	1.093	1.091	1.025	0.918	0.762	0.644	0.662	0.748
	Natural Gas	Activity	8.520	8.637	8.996	9.129	9.202	9.678	9.999	10.109	9.882	9.438	9.550	8.674	8.865	8.510	8.573	7.930	7.881	8.098	8.102	7.629	7.982
NOx		NEI	1.648					1.775	1.844			1.543		1.583	1.071			0.917					
		This study	1.023	1.023	1.065	1.085	1.091	1.136	1.144	1.149	1.119	1.062	1.064	0.959	0.886	0.817	0.819	0.731	0.727	0.748	0.748	0.704	0.738
SO2		NEI	0.031					0.035	0.046			0.060		0.061	0.035			0.034					
		This study	0.002	0.002	0.002	0.002	0.002	0.003	0.003	0.003	0.003	0.003	0.003	0.002	0.002	0.002	0.002	0.002	0.002	0.002	0.002	0.002	0.002
Distillate fuel oil		Activity	1.150	1.078	1.107	1.117	1.111	1.131	1.187	1.203	1.211	1.187	1.200	1.300	1.204	1.136	1.214	1.264	1.263	1.265	1.277	1.107	1.188
	NOx	NEI	0.080					0.086	0.077			0.099		0.111	0.113			0.107					
		This study	0.125	0.117	0.121	0.122	0.121	0.123	0.129	0.131	0.132	0.129	0.131	0.142	0.131	0.124	0.132	0.138	0.137	0.138	0.139	0.121	0.129
	SO2	NEI	0.181					0.177	0.153			0.148		0.163	0.135			0.131					
		This study	0.242	0.215	0.225	0.228	0.225	0.220	0.219	0.214	0.218	0.202	0.201	0.212	0.147	0.127	0.136	0.131	0.133	0.136	0.137	0.120	0.128
	Residual fuel oil	Activity	0.411	0.334	0.387	0.446	0.419	0.337	0.335	0.291	0.230	0.207	0.241	0.203	0.190	0.220	0.249	0.281	0.239	0.193	0.199	0.106	0.120
NOx		NEI	0.158					0.147	0.133			0.099		0.111	0.056			0.056					
		This study	0.068	0.055	0.064	0.074	0.069	0.056	0.055	0.048	0.038	0.035	0.040	0.034	0.032	0.037	0.041	0.047	0.040	0.032	0.033	0.018	0.019
SO2		NEI	0.651					0.683	0.623			0.360		0.396	0.185			0.174					
		This study	0.195	0.158	0.183	0.211	0.198	0.159	0.158	0.138	0.109	0.098	0.114	0.096	0.086	0.086	0.101	0.101	0.084	0.073	0.072	0.037	0.037

Commercial Combustion		1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	
Coal	Activity	0.124	0.115	0.117	0.117	0.117	0.116	0.120	0.129	0.101	0.102	0.086	0.088	0.088	0.083	0.103	0.096	0.064	0.070	0.072	0.065	0.062	
	NOx	NEI	0.035					0.038	0.035			0.036		0.038	0.034			0.032					
		This study	0.037	0.034	0.035	0.035	0.035	0.035	0.036	0.038	0.030	0.029	0.026	0.026	0.026	0.025	0.031	0.029	0.019	0.021	0.021	0.019	0.018
	SO2	NEI	0.211					0.199	0.183			0.145		0.147	0.129			0.126					
		This study	0.225	0.202	0.198	0.205	0.207	0.206	0.216	0.224	0.172	0.160	0.144	0.143	0.126	0.118	0.135	0.110	0.079	0.092	0.095	0.087	0.084
Natural Gas	Activity	2.698	2.807	2.883	2.944	2.978	3.117	3.251	3.306	3.098	3.132	3.261	3.109	3.223	3.271	3.211	3.083	2.908	3.095	3.235	3.199	3.172	
	NOx	NEI	0.123					0.162	0.162			0.170		0.180	0.163			0.162					
		This study	0.147	0.153	0.157	0.160	0.162	0.170	0.177	0.180	0.169	0.170	0.178	0.169	0.175	0.178	0.175	0.168	0.158	0.168	0.176	0.174	0.173
	SO2	NEI	0.006					0.007	0.008			0.010		0.013	0.002			0.002					
		This study	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001
Distillate fuel oil	Activity	0.536	0.517	0.507	0.493	0.501	0.479	0.483	0.444	0.429	0.438	0.491	0.508	0.444	0.481	0.470	0.447	0.401	0.384	0.372	0.413	0.410	
	NOx	NEI	0.050					0.049	0.049			0.042		0.044	0.050			0.045					
		This study	0.041	0.040	0.039	0.038	0.039	0.037	0.037	0.034	0.033	0.034	0.038	0.039	0.034	0.037	0.036	0.035	0.031	0.030	0.029	0.032	0.032
	SO2	NEI	0.238					0.222	0.196			0.154		0.161	0.128			0.121					
		This study	0.209	0.199	0.193	0.189	0.192	0.178	0.168	0.149	0.145	0.143	0.154	0.160	0.096	0.095	0.095	0.083	0.076	0.071	0.072	0.080	0.083
Residual fuel oil	Activity	0.230	0.212	0.189	0.173	0.172	0.141	0.137	0.111	0.085	0.073	0.092	0.070	0.080	0.111	0.122	0.116	0.075	0.075	0.073	0.076	0.077	
	NOx	NEI	0.035					0.041	0.026			0.014		0.015	0.021			0.022					
		This study	0.037	0.035	0.030	0.029	0.028	0.023	0.023	0.018	0.012	0.012	0.015	0.012	0.013	0.018	0.020	0.019	0.013	0.013	0.012	0.012	0.012
	SO2	NEI	0.186					0.174	0.142			0.098		0.102	0.093			0.093					
		This study	0.093	0.087	0.074	0.068	0.068	0.057	0.055	0.045	0.030	0.029	0.036	0.027	0.030	0.037	0.039	0.033	0.023	0.023	0.022	0.022	0.023

Residential Combustion		1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	
Coal	Activity	0.031	0.025	0.026	0.026	0.021	0.017	0.016	0.016	0.012	0.014	0.011	0.011	0.012	0.012	0.011	0.008	0.006	0.008	0.008	0.008	0.008	
	NOx	NEI	0.106					0.111	0.100			0.007		0.007	0.003			0.003					
		This study	0.009	0.008	0.008	0.008	0.006	0.005	0.005	0.005	0.004	0.004	0.003	0.003	0.004	0.004	0.003	0.002	0.002	0.002	0.002	0.002	0.002
	SO2	NEI	0.371					0.029	0.021			0.052		0.052	0.021			0.021					
		This study	0.070	0.057	0.058	0.056	0.045	0.038	0.035	0.035	0.027	0.030	0.022	0.022	0.016	0.018	0.015	0.010	0.009	0.011	0.009	0.009	0.009
	Natural Gas	Activity	4.519	4.684	4.820	5.098	4.981	4.984	5.391	5.125	4.671	4.857	5.104	4.902	5.006	5.224	4.993	4.958	4.483	4.849	5.018	4.899	4.893
NOx		NEI	0.326					0.519	0.481			0.219		0.221	0.240			0.241					
		This study	0.246	0.255	0.262	0.277	0.271	0.271	0.293	0.279	0.254	0.264	0.278	0.267	0.272	0.284	0.272	0.270	0.244	0.264	0.273	0.267	0.266
SO2		NEI	0.001					0.002	0.002			0.001		0.001	0.001			0.001					
		This study	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001
Distillate fuel oil		Activity	0.978	0.930	0.980	0.974	0.960	0.905	0.926	0.874	0.772	0.828	0.905	0.908	0.860	0.905	0.924	0.854	0.712	0.726	0.669	0.602	0.583
	NOx	NEI	0.191					0.210	0.194			0.053		0.053	0.062			0.062					
		This study	0.057	0.054	0.057	0.057	0.056	0.053	0.054	0.051	0.045	0.048	0.053	0.053	0.050	0.053	0.054	0.050	0.042	0.042	0.039	0.035	0.034
	SO2	NEI	0.962					0.144	0.108			0.125		0.126	0.135			0.135					
		This study	0.333	0.251	0.262	0.260	0.255	0.172	0.164	0.154	0.135	0.144	0.157	0.157	0.142	0.145	0.147	0.133	0.111	0.112	0.104	0.093	0.091

Activity – energy combustion, unit: 10⁶ MMBtu year⁻¹

Emission – unit: Tg year⁻¹

(b) on-road mobile sources (absolute values for Fig. 4)

Light Duty Vehicle		1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
Activity		1.404	1.360	1.367	1.375	1.407	1.438	1.466	1.505	1.556	1.563	1.603	1.624	1.658	1.669	1.705	1.707	1.692	1.742	1.776	1.765	1.731
NOx	NEI							3.001			2.421		2.512	2.295			1.986					
	This study	4.529	3.990	3.682	3.410	3.229	3.031	2.840	2.751	2.675	2.485	2.427	2.385	2.295	2.189	1.964	1.807	1.537	1.461	1.370	1.238	1.138
CO	NEI							28.573			43.816		39.590	36.482			21.699					
	This study	81.111	71.175	64.940	59.222	54.969	51.045	47.127	44.698	43.008	40.409	39.145	38.160	36.482	34.844	30.425	27.712	23.709	23.055	22.220	20.934	19.926
SO2	NEI							0.129			0.101		0.084	0.081			0.034					
	This study	0.173	0.160	0.149	0.137	0.125	0.114	0.102	0.102	0.101	0.101	0.087	0.083	0.081	0.070	0.059	0.048	0.036	0.025	0.014	0.013	0.012
VOC	NEI							2.851			3.126		2.377	2.783			1.861					
	This study	5.916	5.210	4.779	4.400	4.153	3.920	3.664	3.521	3.411	3.230	3.104	2.976	2.783	2.611	2.375	2.166	1.934	1.829	1.715	1.555	1.412
PM10	NEI							0.062			0.051		0.047	0.045			0.032					
	This study	0.088	0.084	0.080	0.076	0.072	0.069	0.065	0.062	0.058	0.054	0.052	0.048	0.045	0.044	0.044	0.043	0.041	0.038	0.036	0.031	0.026
PM2.5	NEI							0.038			0.029		0.026	0.024			0.010					
	This study	0.054	0.052	0.049	0.046	0.043	0.041	0.038	0.036	0.033	0.031	0.029	0.026	0.024	0.024	0.023	0.023	0.021	0.019	0.018	0.016	0.015

Light Duty Truck	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
Activity	0.578	0.644	0.698	0.731	0.756	0.776	0.802	0.834	0.860	0.889	0.910	0.929	0.955	0.970	1.012	1.022	1.055	1.089	1.083	1.082	1.081
NOx	NEI						1.962			1.478		1.903	1.925			2.549					
	This study	2.068	2.129	2.158	2.106	2.049	1.954	1.896	1.883	1.867	1.874	1.902	1.925	1.925	1.893	1.794	1.662	1.534	1.480	1.360	1.270
CO	NEI						19.282			30.659		32.404	31.732			29.613					
	This study	45.993	47.361	47.460	45.858	43.839	41.383	39.191	38.090	36.817	35.439	34.290	33.197	31.732	29.127	26.435	23.090	20.887	20.298	18.689	17.481
SO2	NEI						0.096			0.067		0.069	0.074			0.037					
	This study	0.153	0.143	0.133	0.122	0.113	0.103	0.092	0.092	0.091	0.091	0.078	0.075	0.074	0.064	0.054	0.044	0.033	0.023	0.013	0.012
VOC	NEI						2.065			1.989		1.920	2.104			2.195					
	This study	3.045	3.109	3.074	2.959	2.841	2.696	2.546	2.482	2.405	2.347	2.293	2.221	2.104	1.981	1.834	1.620	1.514	1.460	1.297	1.199
PM10	NEI						0.043			0.034		0.033	0.035			0.034					
	This study	0.067	0.064	0.061	0.058	0.055	0.052	0.050	0.047	0.044	0.041	0.039	0.036	0.035	0.034	0.034	0.033	0.031	0.029	0.028	0.024
PM2.5	NEI						0.028			0.020		0.019	0.020			0.011					
	This study	0.044	0.042	0.040	0.038	0.036	0.034	0.031	0.030	0.027	0.025	0.024	0.021	0.020	0.020	0.019	0.019	0.017	0.016	0.014	0.013

Heavy Duty		1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
Activity		0.152	0.155	0.159	0.166	0.177	0.185	0.190	0.198	0.203	0.210	0.213	0.216	0.221	0.225	0.228	0.229	0.247	0.258	0.266	0.269	0.268
NOx	NEI							2.873			4.585		4.295	4.149			4.980					
	This study	4.353	4.241	4.214	4.299	4.543	4.707	4.729	4.874	4.928	4.896	4.656	4.352	4.149	3.834	3.479	3.205	3.149	3.011	2.774	2.573	2.299
CO	NEI							5.219			6.056		4.552	3.941			5.291					
	This study	9.226	8.695	8.165	7.764	7.558	7.156	6.563	6.121	5.641	5.200	4.716	4.269	3.941	3.418	3.117	2.860	2.703	2.632	2.444	2.038	1.911
SO2	NEI							0.091			0.118		0.117	0.082			0.101					
	This study	0.172	0.161	0.150	0.137	0.128	0.117	0.104	0.104	0.102	0.102	0.088	0.083	0.082	0.072	0.061	0.050	0.038	0.026	0.014	0.013	0.012
VOC	NEI							0.538			0.575		0.471	0.420			0.549					
	This study	0.796	0.750	0.690	0.661	0.640	0.619	0.580	0.555	0.521	0.496	0.466	0.438	0.420	0.390	0.370	0.340	0.328	0.318	0.300	0.275	0.255
PM10	NEI							0.177			0.155		0.136	0.121			0.038					
	This study	0.236	0.226	0.216	0.205	0.198	0.188	0.176	0.167	0.156	0.146	0.138	0.127	0.121	0.120	0.118	0.115	0.109	0.103	0.096	0.084	0.071
PM2.5	NEI							0.155			0.135		0.118	0.104			0.020					
	This study	0.233	0.223	0.211	0.199	0.191	0.180	0.166	0.156	0.143	0.132	0.123	0.111	0.104	0.102	0.099	0.096	0.089	0.082	0.075	0.070	0.065

Motorcycle		1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
Activity		0.010	0.009	0.010	0.010	0.010	0.009	0.010	0.010	0.010	0.010	0.010	0.010	0.010	0.010	0.011	0.011	0.012	0.013	0.014	0.014	0.014
NOx	NEI							0.012			0.015		0.019	0.021			0.020					
	This study	0.022	0.021	0.022	0.023	0.023	0.021	0.022	0.022	0.023	0.023	0.023	0.021	0.021	0.022	0.023	0.023	0.026	0.029	0.030	0.030	0.032
CO	NEI							0.189			0.173		0.209	0.225			0.691					
	This study	0.255	0.234	0.249	0.251	0.256	0.235	0.245	0.236	0.245	0.239	0.240	0.221	0.225	0.226	0.246	0.248	0.282	0.308	0.318	0.317	0.327
SO2	NEI							0.000			0.000		0.000	0.000			0.000					
	This study	0.001	0.001	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
VOC	NEI							0.036			0.028		0.028	0.032			0.067					
	This study	0.042	0.037	0.039	0.038	0.038	0.035	0.036	0.034	0.036	0.035	0.034	0.032	0.032	0.033	0.035	0.035	0.040	0.044	0.046	0.046	0.047
PM10	NEI							0.000			0.000		0.000	0.000			0.000					
	This study	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
PM2.5	NEI							0.000			0.000		0.000	0.000			0.000					
	This study	0.001	0.001	0.001	0.001	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000

Activity – vehicle miles traveled, unit: 10^{12} miles year⁻¹

Emission – unit: Tg year⁻¹

(c) industrial process sources (absolute values for Fig. 6)

Chemical Manufacturing		1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
NOx	NEI	0.186					0.186	0.169			0.123		0.129	0.096			0.085					
	This study	0.233	0.226	0.218	0.210	0.203	0.195	0.187	0.180	0.172	0.164	0.157	0.149	0.141	0.134	0.126	0.118	0.111	0.103	0.095	0.088	0.080
CO	NEI	1.266					1.245	1.182			0.478		0.508	0.385			0.291					
	This study	1.655	1.585	1.515	1.444	1.374	1.303	1.233	1.162	1.092	1.021	0.951	0.881	0.810	0.740	0.669	0.599	0.528	0.458	0.388	0.317	0.247
SO2	NEI	0.312					0.300	0.301			0.291		0.304	0.265			0.261					
	This study	0.310	0.304	0.299	0.293	0.288	0.282	0.277	0.271	0.266	0.260	0.255	0.249	0.244	0.239	0.233	0.228	0.222	0.217	0.211	0.206	0.200
VOC	NEI	0.618					0.688	0.410			0.260		0.270	0.261			0.248					
	This study	0.669	0.644	0.618	0.593	0.567	0.542	0.516	0.491	0.465	0.440	0.414	0.389	0.363	0.338	0.312	0.287	0.261	0.236	0.210	0.185	0.159
PM10	NEI	0.078					0.069	0.067			0.057		0.060	0.043			0.039					
	This study	0.083	0.081	0.078	0.076	0.073	0.071	0.068	0.065	0.063	0.060	0.058	0.055	0.052	0.050	0.047	0.045	0.042	0.040	0.037	0.034	0.032
PM2.5	NEI	0.048					0.044	0.041			0.048		0.050	0.032			0.032					
	This study	0.051	0.050	0.049	0.048	0.047	0.045	0.044	0.043	0.042	0.041	0.039	0.038	0.037	0.036	0.035	0.033	0.032	0.031	0.030	0.029	0.027

Metals processing		1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
NOx	NEI	0.126					0.133	0.104			0.109		0.121	0.090			0.081					
	This study	0.119	0.118	0.116	0.114	0.113	0.111	0.110	0.108	0.107	0.105	0.104	0.102	0.101	0.099	0.097	0.096	0.094	0.093	0.091	0.090	0.088
CO	NEI	2.642					2.400	1.771			1.358		1.498	1.041			0.883					
	This study	2.812	2.714	2.616	2.518	2.420	2.322	2.225	2.127	2.029	1.931	1.833	1.736	1.638	1.540	1.442	1.344	1.246	1.149	1.051	0.953	0.855
SO2	NEI	0.729					0.535	0.447			0.323		0.320	0.229			0.189					
	This study	0.787	0.756	0.726	0.695	0.665	0.634	0.604	0.573	0.542	0.512	0.481	0.451	0.420	0.390	0.359	0.329	0.298	0.267	0.237	0.206	0.176
VOC	NEI	0.130					0.136	0.086			0.082		0.088	0.057			0.059					
	This study	0.146	0.141	0.137	0.132	0.127	0.122	0.118	0.113	0.108	0.104	0.099	0.094	0.089	0.085	0.080	0.075	0.070	0.066	0.061	0.056	0.051
PM10	NEI	0.231					0.247	0.207			0.154		0.167	0.090			0.091					
	This study	0.234	0.226	0.218	0.210	0.202	0.194	0.186	0.178	0.170	0.162	0.154	0.147	0.139	0.131	0.123	0.115	0.107	0.099	0.091	0.083	0.075
PM2.5	NEI	0.164					0.151	0.129			0.126		0.137	0.058			0.066					
	This study	0.172	0.167	0.161	0.156	0.150	0.145	0.140	0.134	0.129	0.123	0.118	0.112	0.107	0.101	0.096	0.091	0.085	0.080	0.074	0.069	0.063

Petroleum & related industries		1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
NOx	NEI	0.301					0.267	0.282			0.244		0.244	0.453			0.556					
	This study	0.564	0.564	0.564	0.564	0.564	0.564	0.564	0.564	0.564	0.564	0.564	0.564	0.564	0.564	0.564	0.564	0.564	0.564	0.564	0.564	0.564
CO	NEI	0.363					0.427	0.459			0.200		0.201	0.407			0.449					
	This study	0.457	0.457	0.457	0.457	0.457	0.457	0.457	0.457	0.457	0.457	0.457	0.457	0.457	0.457	0.457	0.457	0.457	0.457	0.457	0.457	0.457
SO2	NEI	0.643					0.538	0.520			0.470		0.484	0.375			0.281					
	This study	0.608	0.588	0.568	0.548	0.527	0.507	0.487	0.466	0.446	0.426	0.406	0.385	0.365	0.345	0.324	0.304	0.284	0.264	0.243	0.223	0.203
VOC	NEI	0.617					0.658	0.505			0.466		0.450	0.625			0.957					
	This study	1.023	1.023	1.023	1.023	1.023	1.023	1.023	1.023	1.023	1.023	1.023	1.023	1.023	1.023	1.023	1.023	1.023	1.023	1.023	1.023	1.023
PM10	NEI	0.030					0.033	0.028			0.038		0.036	0.027			0.034					
	This study	0.066	0.064	0.063	0.061	0.059	0.057	0.055	0.054	0.052	0.050	0.048	0.046	0.045	0.043	0.041	0.039	0.037	0.036	0.034	0.032	0.030
PM2.5	NEI	0.022					0.024	0.021			0.033		0.031	0.022			0.029					
	This study	0.036	0.036	0.036	0.035	0.035	0.035	0.034	0.034	0.033	0.033	0.033	0.032	0.032	0.032	0.031	0.031	0.030	0.030	0.030	0.029	0.029

Other industry processes		1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	
NOx	NEI	0.487					0.534	0.526			0.558		0.612	0.516			0.556						
	This study	0.495	0.495	0.495	0.495	0.495	0.495	0.495	0.495	0.495	0.495	0.495	0.495	0.495	0.495	0.495	0.495	0.495	0.495	0.495	0.495	0.495	0.495
CO	NEI	0.590					0.688	0.649			0.631		0.710	0.584			0.626						
	This study	0.575	0.575	0.575	0.575	0.575	0.575	0.575	0.575	0.575	0.575	0.575	0.575	0.575	0.575	0.575	0.575	0.575	0.575	0.575	0.575	0.575	0.575
SO2	NEI	0.552					0.578	0.480			0.453		0.496	0.378			0.400						
	This study	0.442	0.435	0.427	0.420	0.412	0.405	0.398	0.390	0.383	0.375	0.368	0.361	0.353	0.346	0.338	0.331	0.324	0.316	0.309	0.301	0.294	
VOC	NEI	0.408					0.463	0.445			0.445		0.434	0.451			0.467						
	This study	0.452	0.452	0.452	0.452	0.452	0.452	0.452	0.452	0.452	0.452	0.452	0.452	0.452	0.452	0.452	0.452	0.452	0.452	0.452	0.452	0.452	0.452
PM10	NEI	0.734					0.633	0.449			0.462		0.535	0.411			0.437						
	This study	0.253	0.249	0.246	0.242	0.239	0.235	0.231	0.228	0.224	0.221	0.217	0.214	0.210	0.206	0.203	0.199	0.196	0.192	0.188	0.185	0.181	
PM2.5	NEI	0.346					0.315	0.242			0.306		0.365	0.253			0.284						
	This study	0.209	0.207	0.205	0.203	0.201	0.198	0.196	0.194	0.192	0.190	0.188	0.186	0.184	0.182	0.180	0.178	0.175	0.173	0.171	0.169	0.167	

Emission – unit: Tg year⁻¹

(d) other non-energy related sources (absolute values for Fig. 7)

		1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
(a) Solvent utilization (NMVOC)	Activity	1.093	1.038	1.060	1.144	1.212	1.192	1.248	1.263	1.283	1.311	1.281	1.235	1.291	1.326	1.362	1.381	1.360	1.353	1.201	1.089	1.153
	NEI data	4.400					6.186	5.541			5.039		5.016	4.282			4.247					
	NEI trend	2.099	2.110	2.154	2.196	2.249	2.257	1.999	2.052	1.879	1.838	1.763	1.829	1.561	1.557	1.553	1.549	1.415	1.280	1.145	1.040	0.935
	This study	5.752	5.422	5.538	5.975	6.330	6.186	5.479	5.191	5.276	5.038	5.057	5.014	4.280	4.231	4.349	4.247	4.183	4.163	3.695	3.349	3.243
(b) Fertilizer Application (NH3)	Activity	12.671	12.953	13.561	12.286	13.494	12.814	13.979	13.672	13.845	13.516	14.156	13.440	13.589	13.784	13.610	14.113	13.080	14.760	13.784	13.673	14.716
	NEI data	0.420					0.219	0.656			0.726		0.727	1.164			1.164					
	This study	1.086	1.134	1.176	1.076	1.161	1.123	1.233	1.189	1.224	1.188	1.238	1.168	1.164	1.172	1.162	1.187	1.101	1.214	1.166	1.145	1.211
(c) Livestock (NH3)	Activity	0.199	0.202	0.210	0.210	0.215	0.217	0.215	0.215	0.228	0.231	0.228	0.228	0.233	0.231	0.234	0.235	0.238	0.243	0.253	0.253	0.246
	NEI data	3.160					1.784	3.451			3.536		2.419	2.112			2.112					
	This study	2.024	2.030	2.087	2.091	2.121	2.122	2.082	2.069	2.134	2.161	2.123	2.108	2.135	2.112	2.119	2.112	2.130	2.149	2.196	2.189	2.141
(d) Construction processes (PM10)	Activity	0.260	0.282	0.310	0.337	0.355	0.369	0.390	0.407	0.434	0.459	0.486	0.498	0.482	0.503	0.530	0.566	0.595	0.632	0.601	0.525	0.453
	NEI data	4.249					3.654	0.960			1.923		0.919	0.894			0.894					
	This study	1.151	1.000	1.064	1.184	1.219	1.069	1.053	0.998	1.011	0.994	0.971	0.961	0.935	0.881	0.900	0.894	0.963	1.028	1.033	0.985	0.920
(e) Mining & quarrying (PM10)	Activity	2.897	2.961	3.071	3.048	3.113	3.257	3.357	3.514	3.626	3.308	3.269	3.369	3.410	3.450	4.062	4.156	4.265	4.219	3.983	3.507	4.094
	NEI data	0.321					0.573	0.550			0.708		0.365	0.608			0.608					
	This study	0.462	0.472	0.489	0.486	0.496	0.507	0.519	0.531	0.557	0.550	0.542	0.560	0.550	0.562	0.593	0.608	0.630	0.604	0.560	0.498	0.560
(f) Paved/Unpaved road (PM10)	Activity	2.137	2.174	2.254	2.310	2.373	2.436	2.498	2.582	2.651	2.705	2.770	2.812	2.878	2.912	2.995	3.011	3.048	3.168	3.185	3.139	3.090
	NEI data	13.353					12.752	3.578			11.443		5.470	5.169			5.169					
	This study	13.353	12.919	12.486	12.052	11.618	11.185	10.751	10.317	9.884	9.450	9.016	8.583	8.149	7.715	7.282	6.848	6.414	5.981	5.547	5.113	4.680
(g) Agriculture tilling (PM10)	Activity	0.167	0.174	0.177	0.166	0.173	0.167	0.180	0.176	0.175	0.175	0.176	0.173	0.175	0.173	0.173	0.173	0.168	0.183	0.175	0.172	0.174
	NEI data	4.826					4.441	1.191			4.482		3.391	2.491			2.491					
	This study	2.418	2.521	2.529	2.375	2.480	2.403	2.625	2.557	2.548	2.544	2.576	2.533	2.551	2.513	2.514	2.491	2.416	2.653	2.541	2.485	2.513
(h) Forest wildfires (PM10)	Activity	1.664	1.063	0.745	0.647	1.466	0.663	2.184	1.028	0.876	1.988	1.189	1.645	2.686	2.875	2.462	2.306	2.721	3.148	1.942	2.531	2.318
	NEI data	0.447					0.487	0.530			0.310		0.287	0.171			0.171					
	This study	0.165	0.105	0.074	0.064	0.145	0.066	0.216	0.102	0.087	0.197	0.118	0.163	0.171	0.283	0.297	0.253	0.282	0.349	0.282	0.294	0.369

(i) Prescribed Forest Management (PM10)	Activity	4.602	2.941	2.061	1.790	4.057	1.833	6.041	2.845	1.324	5.603	7.363	3.556	7.186	3.962	8.098	8.686	9.871	9.302	5.254	5.922	3.233
	NEI data	0.202					0.145	0.000			0.683		1.196	0.800			0.800					
	This study	0.712	0.455	0.319	0.277	0.627	0.283	0.934	0.440	0.205	0.866	1.139	0.550	0.726	0.640	0.915	1.055	1.909	1.876	1.063	0.865	0.470

Activity –

(a) Estimated Quantity of Shipments of Paint and Allied Products, unit: 10^9 gallons year⁻¹

(b) Fertilizer (NITROGEN) Application, unit: 10^9 lb year⁻¹

(c) Weighted sum of livestock, unit: 10^9 Head year⁻¹

(d) Value of Construction Put in Place, unit: 10^{12} Head dollars year⁻¹

(e) Weighted sum of crude ore and coal handled on surface mine, unit: 10^9 Ton year⁻¹

(f) Vehicle miles traveled, unit: 10^{12} miles year⁻¹

(g) Weighted sum of number of acres harvested, unit: 10^9 acres year⁻¹

(h, i) Acres Burned, unit: 10^6 acres year⁻¹

Emission – unit: Tg year⁻¹

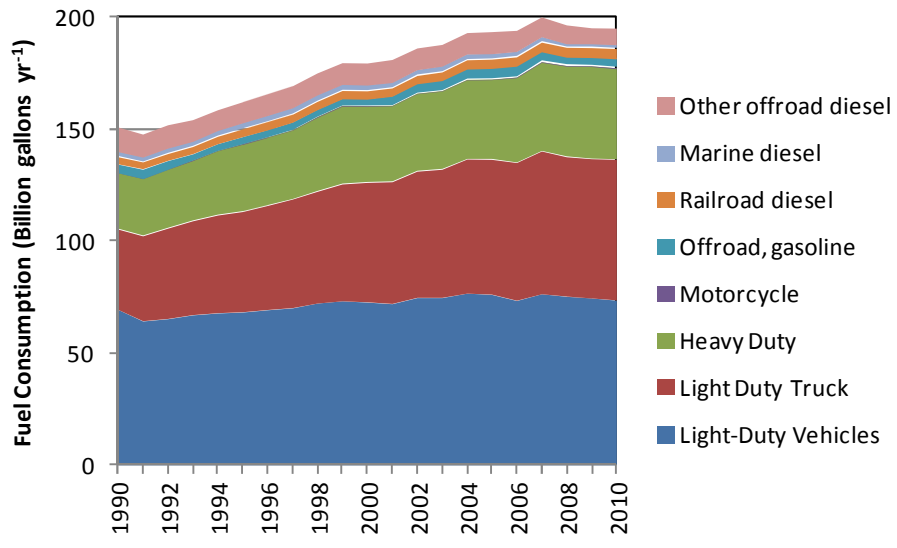


Fig. S1 Mobile source fuel consumption in the United States, 1990–2010

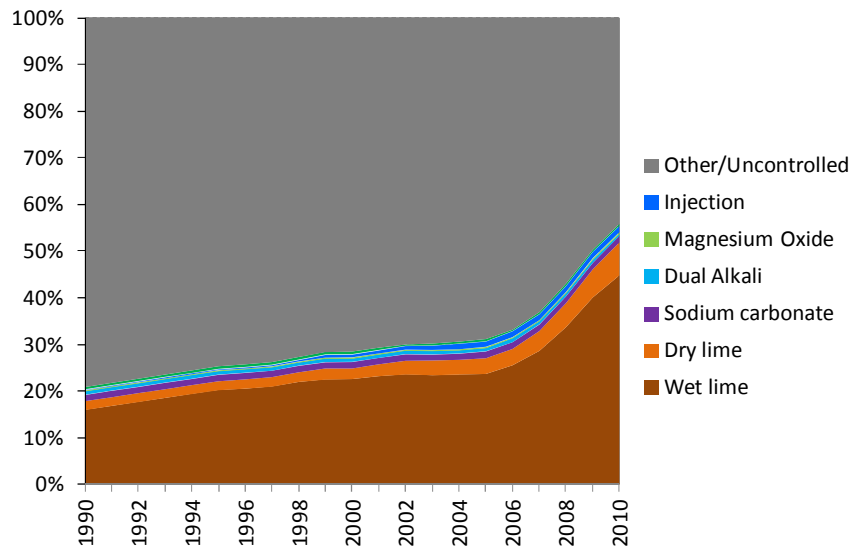


Fig. S2 Application of post-combustion SO₂ control technologies in coal-fired power plants from 1990-2010 (weighted by unit capacity, based on the Clean Air Markets data, <http://ampd.epa.gov/ampd/>)

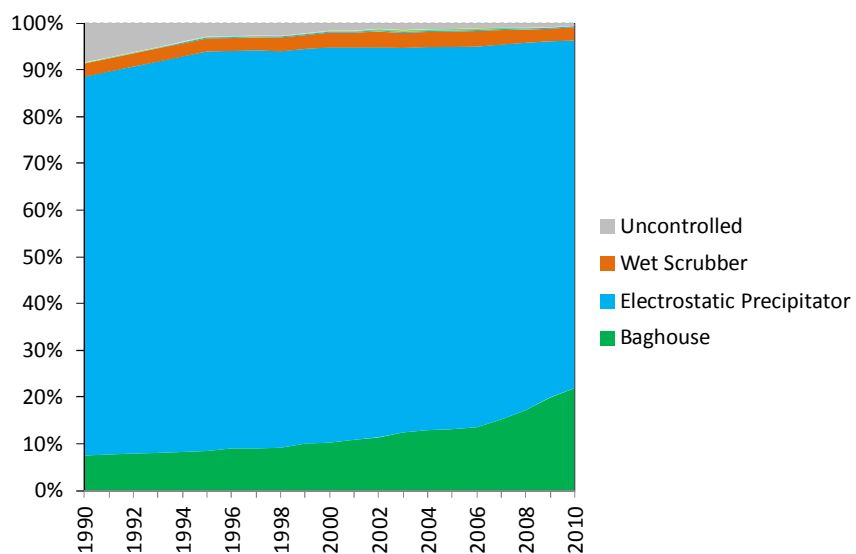


Fig. S3 Application of post-combustion PM control technologies in coal-fired power plants from 1990 to 2010 (weighted by unit capacity, based on the Clean Air Markets data, <http://ampd.epa.gov/ampd/>)

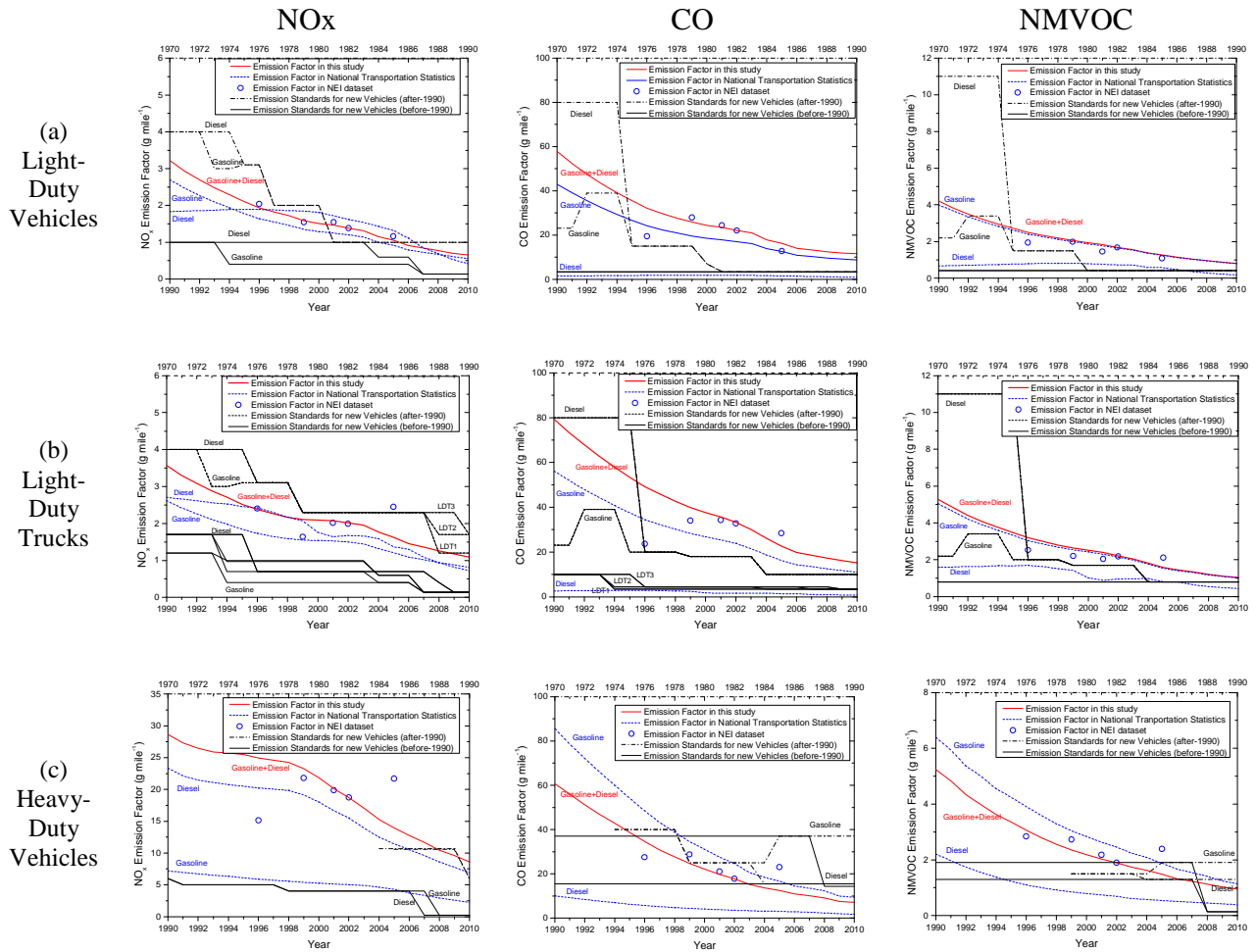


Fig. S4 Comparison of on-road NO_x, CO and NMVOC emission factors with the federal exhaust emissions certification standards for newly manufactured vehicles