

Supplemental Information

Predicting the effects of nano-scale cerium additives in diesel fuel on regional-scale air quality

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Section S1

References for the Full Set of Emissions Studies Compiled for this Work

Table S1-1. Studies on nCe fuel additive effects on diesel exhaust emissions.

- Arul Mozhi Selvan, V.; Anand, R. B.; Udayakumar, M. (2009) Effects of Cerium Oxide Nanoparticle Addition in Diesel and Diesel-Biodiesel-Ethanol Blends on the Performance and Emission Characteristics of a CI Engine. *Journal of Engineering and Applied Sciences* 7, 1-6.
- Burtscher, H.; Matter, U.; Skillas, G. (1999) *Journal of Aerosol Science* 1999, 30, S851-S852.
- Cerulean International Ltd. (2005) Envirox™ Tier 1, 211b Submission, EPA/OTAQ Fuel and Fuel Additives Registration Documents.
- Clean Diesel Technologies, Inc. (1999) Platinum Plus® DFX Tier 1, 211b Submittal, EPA/OTAQ Fuel and Fuel Additives Registration Documents.
- Czerwinski, J.; Napoli, S.; Compte, P.; Matter, U.; Mosimann, T. (2000) Investigations with the Corning-Diesel Particulate Filter and Cerium-Additive Eolys DPX 9 on the Liebherr D914T Engine, VERT Report, Technik Thermische Maschinen, Niederrohrdorf, Switzerland.
- Czerwinski, J.; Napoli, S.; Matter, U.; Kasper, M.; Mosimann, T. (1999) Investigations with the IBIDEN SiC-Diesel Particulate Filter on the Liebherr D914T Engine with Detailed Analysis of the Particulate Emissions, VERT Report, Technik Thermische Maschinen, Niederrohrdorf, Switzerland.
- Czerwinski, J.; Mosimann, T.; Matter, U. (1997) Comparison of the Engines Liebherr I (86) and Liebherr II (96) with Particulate Trap, with Fuel Additives and with Detailed Analysis of the Particulate Emissions, VERT Report, Technik Thermische Maschinen, Niederrohrdorf, Switzerland.
- Dwyer, H.; Ayala, A.; Zhang, S.; Collins, J.; Huai, T.; Herner, J.; Chau, W. (2010) Emissions from a diesel car during regeneration of an active diesel particulate filter. *Journal of Aerosol Science* 41, 541-552.
- Farfaletti, A.; Astorga, C.; Martini, G.; Manfredi, U.; Mueller, A.; Rey, M.; De Santi, G.; Krasenbrink, A.; Larsen, B. R. (2005) Effect of Water/Fuel Emulsions and a Cerium-Based Combustion Improver Additive on HD and LD Diesel Exhaust Emissions. *Environmental Science & Technology* 39, 6792-6799.
- Gangal, M.; Leprince, T.; Muter, J.; Rubeli, B. (1999) The Effects of a Diesel Particulate Filter and Fuel Borne Catalyst on Engine Emissions, *Proceedings of the 8th U.S. Mine Ventilation Symposium*, Rolla, Missouri, 265-271.
- Heeb N. V. (1998a) Influence of Particulate Trap Systems on the Composition of Diesel Engine Exhaust Gas Emissions, Eidgenössische Materialprüfungs Und Forschungsanstalt (EMPA) Research Report 167985.
-

Table S1-1. Studies on nCe fuel additive effects on diesel exhaust emissions.

Jackson, M. D.; Remillard, R. L. (2001) Demonstration of a Diesel Fuel-Borne Catalyst System and Low NO_x Control Technology for Reducing Particulate and NO_x Emissions, Final Report (FR-00-104) Prepared for: South Coast Air Quality Management District, California Air Resources Board and California Environmental Protection Agency.

Japan Automobile Research Institute (JARI). (1995) Investigation on the Influence of Cerium Additive (Eolys) on Diesel Emissions, Tsukuba, Japan.

Jung, H.; Kittelson, D. B.; Zachariah, M. R. (2005) The influence of a cerium additive on ultrafine diesel particle emissions and kinetics of oxidation, *Combustion and Flame* 142, 276-288.

Khair, M.; Lemaire, J.; Fischer, S. (2000) Achieving Heavy-Duty Diesel NO_x/PM Levels Below the EPA 2002 Standards—An Integrated Solution. SAE Technical Paper Series 2000-01-0187.

Ladegaard, N.; Sorenson, S. C.; Schramm, J.; Gratz, L.; Stobbe, P. (1997) Fuel Additive Effects on Particulate Emissions from a Diesel Engine, SAE Technical Paper Series 970181.

Lemaire, J.; Khair, M. (1994) Effect of Cerium Fuel Additive on the Emissions Characteristics of a Heavy-Duty Diesel Engine, SAE Technical Paper Series 942067.

Lepperhoff, G.; Lüders, H.; Barthe, P.; Lemaire, J. (1995) Quasi-Continuous Particle Trap Regeneration by Cerium-Additives, SAE Technical Paper Series 950369.

Nikolaou, K. (2000) Five Years Studies on Vehicle Emission Reduction Using a CeO₂ Catalytic System, *Journal of Environmental Protection and Ecology* 1, 468-472.

Okuda, T.; Schauer, J. J.; Olson, M. R.; Shafer, M. M.; Rutter, A. P.; Walz, K. A.; Morschauer, P. A. (2009) Effects of a Platinum-Cerium Bimetallic Fuel Additive on the Chemical Composition of Diesel Engine Exhaust Particles, *Energy & Fuel* 23, 4974-4980.

Oxonica Ltd. (2006) Envirox™ Tier 1, 211b Supplemental Submission, EPA/OTAQ Fuel and Fuel Additives Registration Documents.

Pattas, K.; Samaras, Z.; Sherwood, D.; Umehara, K.; Cantiani, C.; Chariol, O. A.; Barthe, Ph.; Lemaire, J. (1992) Cordierite Filter Durability with Cerium Fuel Additive: 100,000 km of Revenue Service in Athens, SAE Technical Paper Series 920363.

Rhône-Poulenc Inc. (1997) Eolys™: DPX1-2, 4-9 Tier 1, 211b Submission, EPA/OTAQ Fuel and Fuel Additives Registration Documents.

Samaras, Z. (1994) Cerium Based Fuel Additive for the Effective Reduction of Diesel Particulate Emissions: Assessment of its Environmental Effects, Final Report Prepared for Rhône-Poulenc Project "Diesel Propre."

Shafer, M. M.; Schauer, J. J.; Copan, W.; Hoblyn, J. P.; Sprague, B.; Valentine, J. (2006) Investigation of Platinum and Cerium from Use of a Fuel Based Catalyst (FBC). Society of Automotive Engineers (SAE) Paper 2006-01-1517.

Table S1-1. Studies on nCe fuel additive effects on diesel exhaust emissions.

Skillas, G.; Qian, Z.; Baltensperger, U.; Matter, U.; Burtscher, H. (2000) The Influence of Additives on the Size Distribution and Composition of Particles Produced by Diesel Engines, *Combustion Science and Technology* 154, 259-273.

Southwest Research Institute (SwRI). (1997) Study of Long Term Effect of Cerium Additive on Heavy Duty Diesel Engine Emissions and Durability, Final Report SwRI 5870.

Summers, J. C.; Van Houtte, S.; Psaras, D. (1996) Simultaneous control of particulate and NO_x emissions from diesel engines, *Applied Catalysis B: Environmental* 10, 139-156.

Wakefield, G.; Gardener, M. (2007) 8nm cerium oxide as a fuel additive for fuel consumption and emissions reduction, *Technical Proceedings of the NSTI Nanotech: The Nanotechnology Conference and Trade Show* 4, 644-647.

Zhang, J.; Nazarenko, Y.; Zhang, L.; Calderon, L.; Lee, K-B.; Garfunkel, E.; Schwander, S.; Tetley, T.D.; Chung, K.F.; Porter, A.E.; Ryan, M.; Kipen, H.; Liroy, P.J.; Mainelis, G. (2013) Impacts of a nanosized ceria additive on diesel engine emissions of particulate and gaseous pollutants. *Environ. Sci. Technol.* 47, 13077-13085.

Section S2

Impacts on Emissions from Engines Not Equipped with DPFs

Table S2-1. The subset of nCe fuel additive studies used in this work.^a

Reference	Additive	Additive Dosing Level (ppm Ce in fuel)	Engine Tested	Engine Test Protocol
Skillas et al., 2000	Eolys	5, 10, 20, 35	Liebherr 914-T	Four operating points from ECE R49 13-point steady state test
Clean Diesel Technologies, 1999	Platinum Plus DFX	7.5, 15	Cummins N14-370E+	EPA transient cycle
Cerulean, 2005	Envirox	9.5	Cummins ISB 305	EPA transient cycle
Oxonica, 2006	Envirox	9.3	Cummins ISB 305	EPA transient cycle
Summers, 1996	Eolys	10	Detroit Diesel DDECII 6V92TA	CTA bus cycle
Czerwinski et al., 2000	Eolys DPX-9	25	Liebherr 914-T	Four operating points from ISO 8178 C1 and D2 cycles
Czerwinski et al., 1999	Eolys DPX-9	50	Liebherr 914-T	Four operating points from ISO 8178 C1 and D2 cycles
Czerwinski et al., 1997	Eolys DPX-9	50, 100	Liebherr 914-T	Four operating points from ISO 8178 C1 and D2 cycles
Heeb, 1998a	Eolys DPX-9	64	Liebherr 924-TI	ISO 8178/4 C1 cycle
SwRI, 1997	Eolys DPX-4	100	Cummins C series	EPA transient cycle

^a Fuel sulfur content in these studies was 500 ppm, except for Skillas et al. who did not report a value; All studies listed include emission measurements with and without additive for heavy-duty engines without use of a DPF.

Table S2-2. Emissions measured in diesel engine test studies without (base) and with (additive) the use of nCe fuel additives. The percent changes (%Δ) calculated from these data are plotted in Figure 1. The median %Δ values tabulated in the bottom row are used for subsequent air quality modeling calculations.

Reference	Additive Concentration (ppm Ce in fuel)	Emissions ^a											
		PM			CO			NO _x			THC		
		base	additive	%Δ	base	additive	%Δ	base	additive	%Δ	base	additive	%Δ
Skillas, 2000	5	19.9	19.9	0	-	-	-	-	-	-	-	-	-
	5	26.4	22.8	-14	-	-	-	-	-	-	-	-	-
	5	19.8	13.9	-30	-	-	-	-	-	-	-	-	-
	5	12.1	6.5	-46	-	-	-	-	-	-	-	-	-
CDTI, 1999 ^b	7.5	0.091	0.090	-1.5	0.544	0.500	-8.0	3.86	3.6	-6.8	0.184	0.20	8.9
Oxonica, 2006	9.3	0.08	0.10	25	1.3	1.4	7.7	3.7	3.5	-5.4	0.06	0.11	83
Cerulean, 2005	9.5	0.08	0.08	0	1.3	1.5	15.4	3.7	4.1	11	0.06	0.09	50
Cerulean, 2005	9.5	0.08	-	-	1.3	1.4	7.7	3.7	3.8	2.7	0.06	0.08	33
Skillas, 2000	10	19.9	16.5	-17	-	-	-	-	-	-	-	-	-
	10	26.4	22.0	-17	-	-	-	-	-	-	-	-	-
	10	19.8	11.6	-41	-	-	-	-	-	-	-	-	-
	10	12.1	6.8	-44	-	-	-	-	-	-	-	-	-
CDTI, 1999	15	0.091	0.099	8.8	0.544	0.562	3.4	3.86	3.76	-2.6	0.184	0.17	-8.5
Skillas, 2000	20	19.9	16.9	-15	-	-	-	-	-	-	-	-	-
	20	26.4	16.8	-36	-	-	-	-	-	-	-	-	-
	20	19.8	11.3	-43	-	-	-	-	-	-	-	-	-
	20	12.1	6.8	-44	-	-	-	-	-	-	-	-	-
Czerwinski, 2000	25	0.078	0.056	-28	0.22	0.27	23	17.3	15.9	-8.0	0.20	0.23	15
	25	0.042	0.036	-14	0	0.4	-	14.9	14.0	-6.3	0.20	0.23	15
	25	0.103	0.087	-16	0.22	0.3	36	11.2	10.7	-4.7	0.31	0.34	9.7
	25	0.086	0.078	-9.3	0.67	0.88	31	14.1	14.4	2.3	0.27	0.28	3.7
	25	0.075	0.051	-32	0.17	0.22	29	17.4	16.2	-6.7	0.18	0.19	5.6
Median %Δ				-17	15				-5				12
Mean %Δ				-20	16				-2.5				22
Standard error of mean				4.3	4.9				1.9				8.6

^a Emissions data from Skillas are in units of mg/m³; from CDTI, Oxonica and Cerulean, units are in g/bhp-hr; from Czerwinski, units are in g/kW-h.

^b CDTI = Clean Diesel Technologies, Inc.

Table S2-3. Speciated HC emission data from three studies without (base) and with (additive) use of nCe fuel additives.

Organic compound	Emissions (mg/bhp-hr)						Species Mapping ^b
	CDTI, 1999 ^a		Cerulean, 2005		Oxonica, 2006		
	base	additive	base	additive	base	additive	
1,2,3,5-tetramethylbenzene	0.2	0.1					PAR, XYL
1,2,3-trimethylbenzene			0.04	0	0.04	0	PAR, XYL
1,2,4-trimethylbenzene	0.4	0.1	0.19	0	0.19	0.17	PAR, XYL
1,3,5-trimethylbenzene	0.5	0.1					PAR
1,3-butadiene	1.1	1.1	0.15	0.03		0.98	PAR, OLE
1,3-dimethyl-4-ethylbenzene			0.02	0	0.02	0	PAR, XYL
1-butene	0.2	0.2	0.6	0.76	0.6	0.4	PAR, ALDX
1-decene			0.04	0	0.04	0	PAR, OLE
1-hexene	0.5	0.5	0.3	0.25	0.3	0.37	PAR, OLE
1-methyl-2-isopropylbenzene	0.1	0.0	0.41	0	0.41	0.76	PAR, XYL
1-methyl-2n-propylbenzene			0.03	0	0.03	0.44	PAR, XYL
1-methyl-3-isopropylbenzene			0.09	0	0.09	0.57	PAR, XYL
1-methyl-4n-propylbenzene			0.01	0	0.01	0	PAR, XYL
1-nonene	0.5	0.0					PAR, OLE
1-pentene	0.2	0.6	1.05	0.13	1.05	1.04	PAR, OLE
1-propyne	0.2	0.2	0.08	0.08	0.08	0.08	PAR, ALDX
1-undecene	1.1	1.1	0.55	0	0.55	0.24	PAR, OLE
2,2,3-trimethylpentane	0.1	0.2	0.06	0	0.06	0.05	PAR, UNR
2,2,4-trimethylpentane	0.6	1.5	0.09	0.07	0.09	0.17	PAR, UNR
2,2,5-trimethylhexane	0.1	0.3					PAR, UNR
2,2-dimethylbutane	0.1	0.1					PAR, UNR
2,2-dimethyloctane			0.06	0	0.06	0.06	PAR, UNR
2,2-dimethylpentane			0.03	0	0.03	0	PAR, UNR
2,2-dimethylpropane			0.12	0.61	0.12	0.06	PAR, UNR
2,3,3-trimethylpentane			0.04	0	0.04	0.04	PAR, UNR
2,3,4-trimethylpentane	0.2	0.3					PAR

Table S2-3. Speciated HC emission data from three studies without (base) and with (additive) use of nCe fuel additives.

Species	Emissions (mg/bhp-hr)						Species Mapping
	CDTI, 1999		Cerulean, 2005		Oxonica, 2006		
	base	additive	base	additive	base	additive	
2,3-dimethylbutane	0.1	0.1	0.07	0.31	0.07	0.11	PAR
2,3-dimethylhexane	0.1	0.3	0.09	0	0.09	0.06	PAR
2,3-dimethylpentane	0.1	0.3	0.06	0.03	0.06	0.07	PAR
2,4,4-trimethyl-2-pentene			0.21	0	0.21	0	PAR, ALDX, UNR
2,4-dimethyl hexane			0.09	0	0.09	0	PAR
2,4-dimethyloctane	0.3	0.2					PAR
2,4-dimethylpentane	0.3	0.2					PAR
2,5-dimethylheptane	0.1	0.1					PAR
2,6-dimethylheptane	0.5	0.4					PAR
2-methyl-1-butene			1.61	0.15	1.61	0.26	PAR, FORM
2-methyl-1-pentene	0.5	0.5	0.3	0.25	0.3	0.37	PAR, FORM
2-methyl-2-butene	0.3	0.1	0.02	0.07	0.02	0	PAR, ALD2
2-methyl-butane	0.3	0.4					PAR
2-methyloctane	0.3	0.2					PAR
2-methylpropane (isobutane)			0.02	0.01	0.02	0.1	PAR
2-methylpropene (isobutene)	0.7	0.7	0.48	0.49	0.48	0.37	PAR, FORM
3,3-dimethylpentane	0.1	0.0					PAR, UNR
3,4-dimethyl-1-pentene			0.02	0	0.02	0	PAR, OLE
3-ethylpentane	0.3	0.0					PAR
3-methylpentane	0.1	0.2	0.36	0.9	0.36	0.55	PAR
4-methyl-1-pentene	0.2	0.1	0.1	0.02	0.1	0.05	PAR, XYL
4-methylheptane	0.1	0.1					PAR
acetaldehyde	5.6	4.3	5.2	4.5	5.2	5.2	ALD2
acetone	1.4	1.0	0.95	1.91	0.95	2.4	PAR

Table S2-3. Speciated HC emission data from three studies without (base) and with (additive) use of nCe fuel additives.

Species	Emissions (mg/bhp-hr)						Species Mapping
	CDTI, 1999		Cerulean, 2005		Oxonica, 2006		
	base	additive	base	additive	base	additive	
acetylene	1.6	1.5	2.64	2.72	2.64	2.43	PAR, UNR
acrolein	1.9	1.0	2.2	1	2.2	0.3	OLE, ALDX
benzaldehyde	0.6	0.5	0.34	2.41	0.34	0.25	TOL
benzene	0.8	1.1	1.4	1.1	1.4	0.5	PAR, UNR
cis-1,3-dimethylcyclopentane			0.08	0.01	0.08	0.04	PAR
cis-1-methyl-3-ethylcyclopentane	0.3	0.1	0.14	0.06	0.14	0.15	PAR
cis-2-pentene	0.5	0.0	0.06	0	0.06	0	PAR, IOLE
cis-3-hexene			0.09	0	0.09	0	PAR, IOLE
crotonaldehyde	1.8	1.8	2.34	1.32	2.34	2.04	OLE, ALDX
cyclopentane			0.17	0	0.17	0.18	PAR
cyclopentene			0.02	0	0.02	0	PAR, IOLE
dimethylbenzaldehyde	0.1	0.2	0.07	0.23	0.07	0.14	PAR, XYL
ethane	0.2	0.1	0.11	0.06	0.11	0.05	ETHA
ethene	8.8	9.1	11.74	9.3	11.74	11.87	ETH
ethylbenzene	0.8	0.6	0.25	0	0.25	0.16	PAR, TOL
formaldehyde	13.8	11.3	15.7	13.4	15.7	13.7	FORM
hexanal (hexanaladehyde)	0.1	0.2		0.29		0.12	PAR, ALDX
isobutyraldehyde	0.4	0.3	0.16	0.25	0.16	0.2	PAR, ALDX
isovaleraldehyde (3-methylbutanal)	0.5	0.4	0.18	0.18	0.18	0.35	PAR, ALDX
m&p-tolualdehyde	1.8	0.5	1.96	1.68	1.96	1.86	TOL, UNR
m&p-xylene	1.2	0.8	0.53	0.33	0.53	0	XYL
methyl ethyl ketone	0.4	0.3	0.16	0.2	0.16	0.25	PAR
methylcyclohexane	0.2	0.1	0.05	0.15	0.05	0.02	PAR
methylcyclopentane			0.03	0	0.03	0	PAR
naphthalene			0.02	0.28	0.02	0	PAR, XYL

Table S2-3. Speciated HC emission data from three studies without (base) and with (additive) use of nCe fuel additives.

Species	Emissions (mg/bhp-hr)						Species Mapping
	CDTI, 1999		Cerulean, 2005		Oxonica, 2006		
	base	additive	base	additive	base	additive	
n-butane			0.06	0.12	0.06	0.36	PAR
n-decane	0.4	0.3	0.05	0	0.05	0.19	PAR
n-dodecane	0.1	0.7					PAR
n-hexane	0.1	0.2					PAR
n-nonane	0.8	0.7	0.11	0	0.11	0.05	PAR
n-octane	0.4	0.4	0.03	0.17	0.03	0.09	PAR
n-pentane	0.2	0.4	0.2	0.24	0.2	0.37	PAR
n-propylbenzene	0.2	0.1	0.07	0	0.07	0.04	PAR,TOL
o-Tolualdehyde	0.0	0.1	0.43	0.45	0.43	0.24	TOL, UNR
o-xylene	0.6	0.4	0.34	0.17	0.34	0	XYL
propionaldehyde	1.8	2.1	0.9	0.11	0.9	2.8	PAR, ALDX
propylene	3.0	2.9	3.09	2.94	3.09	2.43	PAR, OLE
toluene	1.0	2.5	1.66	0.87	1.66	0.38	TOL
trans-1,3-dimethylcyclopentane			0.02	0	0.02	0	PAR
trans-2-hexene			0.06	0	0.06	0	PAR, IOLE
trans-2-pentene			0.04	0.01	0.04	0.02	PAR, IOLE
valeraldehyde	0.3	0.6	0.29	0.31	0.29	0.16	PAR, ALDX

^a CDTI = Clean Diesel Technologies, Inc.

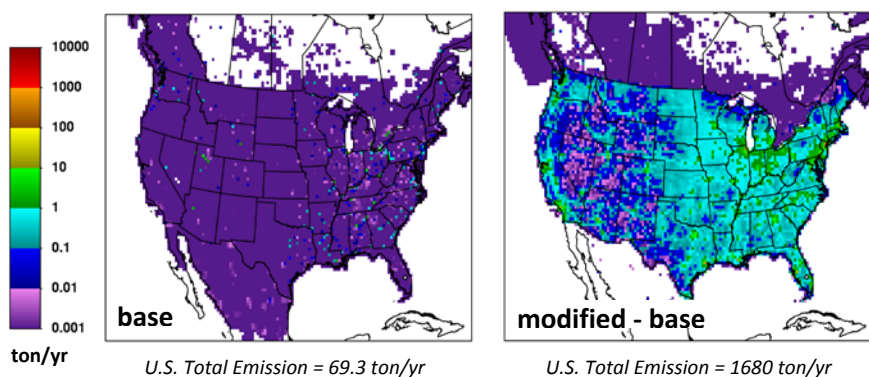
^b Model compounds are as follows: ALD2 = acetaldehyde; ALDX = propionaldehyde and higher aldehydes; ETH = ethene; ETHA = ethane; FORM = formaldehyde; IOLE = olefins with internal carbon double bond(s); OLE = other olefins; PAR = paraffins; TOL = toluene and other monoalkyl aromatics; UNR = unreactive species; XYL = xylene and other polyalkyl aromatics.

Table S2-4. Model compound mass fractions computed for emissions from three studies using the Speciation Tool procedure.

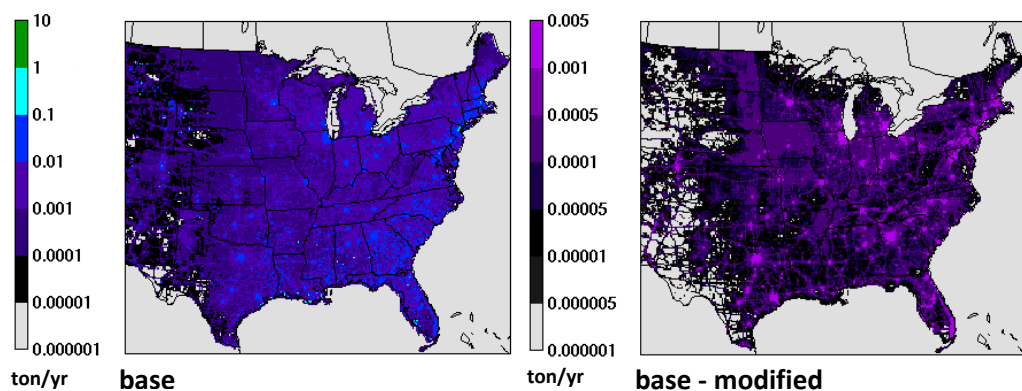
Species	Emission mass fractions						Median %Δ
	CDTI, 1999		Cerulean, 2005		Oxonica, 2006		
	base	additive	base	additive	base	additive	
ALD2	0.0029	0.0010	0.0002	0.0009	0.0002	0.0000	-65
ALDX	0.0989	0.0949	0.1002	0.0705	0.1006	0.0987	-4
ETH	0.2095	0.2264	0.3008	0.2913	0.3020	0.3181	6
ETHA	0.0048	0.0025	0.0028	0.0019	0.0028	0.0013	-48
FORM	0.0062	0.0064	0.0126	0.0061	0.0127	0.0055	-52
IOLE	0.0095	0.0000	0.0050	0.0003	0.0050	0.0004	-95
OLE	0.1105	0.1076	0.1197	0.0972	0.1182	0.1022	-14
PAR	0.3709	0.3908	0.2600	0.2869	0.2590	0.3155	10
TOL	0.0960	0.1027	0.1118	0.1611	0.1123	0.0707	7
UNR	0.0275	0.0271	0.0436	0.0547	0.0438	0.0409	-2
XYL	0.0634	0.0407	0.0433	0.0296	0.0434	0.0465	-31

Figure S2-1. Average emissions without use of nCe fuel additives (base) and difference between base case and additive case emissions (modified – base or base – modified, depending on sign of difference) of cerium and VOC species for which diesel sources contribute $\geq 5\%$ of total emissions. Emissions are shown for (a) cerium, (b) acrolein and (c) formaldehyde.

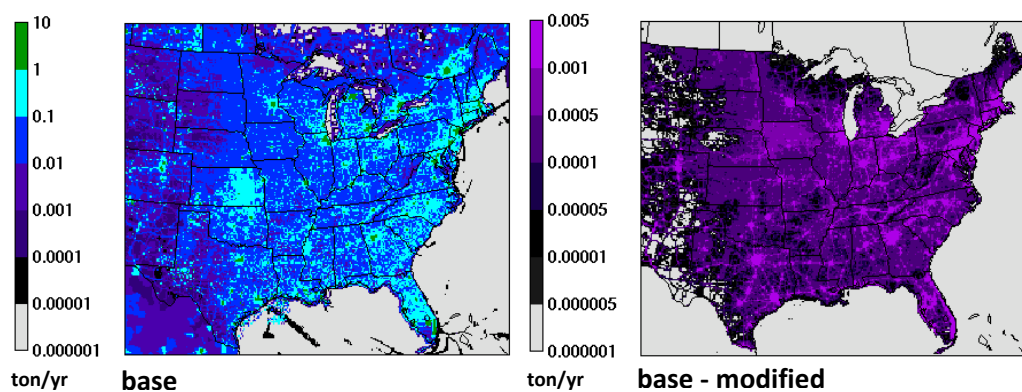
(a) Cerium



(b) Acrolein



(c) Formaldehyde



Section S3

Model-Predicted Impacts on Air Quality

Table S3-1. Domainwide ambient concentrations for Eastern U.S. in 2005 in base and nCe additive case CMAQ simulations (winter and summer 14-day periods averages) and relative changes in concentration.^a

Species ^b	Winter			Summer		
	base case	nCe case	% Change	base case	nCe case	% Change
PM _{2.5}	6.33 (12.9)	6.35 (12.8)	0.3 (-0.5)	4.45 (9.68)	4.42 (9.59)	-0.7 (-1.0)
Diesel PM _{2.5}	0.103 (0.399)	0.091 (0.347)	-11.6 (-13)	0.090 (0.371)	0.078 (0.321)	-13 (-13)
O ₃	34.3 (25.4)	34.2 (25.5)	-0.3 (0.4)	40.5 (41.7)	40.4 (41.6)	-0.2 (-0.2)
CO	146 (258)	147 (258)	0.8 (0.06)	102 (181)	102 (181)	-0.05 (-0.01)
NO _x	3.08 (13.1)	3.06 (13.0)	-0.8 (-1.2)	1.39 (7.37)	1.38 (7.26)	-0.9 (-1.3)
NO	0.324 (2.72)	0.318 (2.66)	-1.6 (-2.2)	0.102 (0.878)	0.101 (0.863)	-1.0 (-1.7)
NO ₂	2.76 (10.4)	2.74 (10.3)	-0.7 (-1.0)	1.29 (6.49)	1.28 (6.40)	-0.9 (-1.3)
VOC	24.6 (60.9)	24.7 (60.9)	0.7 (0.1)	34.4 (71.2)	34.3 (71.0)	-0.08 (-0.3)
SOA	0.089 (0.129)	0.091 (0.128)	2.7 (-0.4)	0.412 (0.767)	0.401 (0.741)	-2.8 (-3.4)
PM _{2.5} species						
EC	0.268 (0.735)	0.265 (0.709)	-1.4 (-3.5)	0.139 (0.472)	0.133 (0.447)	-4.4 (-5.2)
OC	0.980 (2.21)	0.981 (2.18)	0.1 (-1.3)	0.499 (1.08)	0.485 (1.04)	-2.7 (-3.9)
SO ₄	1.24 (1.64)	1.26 (1.65)	2.1 (0.1)	2.13 (4.11)	2.12 (4.10)	-0.1 (-0.1)
NO ₃	1.31 (2.83)	1.30 (2.83)	-0.7 (-0.3)	0.130 (0.403)	0.128 (0.397)	-1.2 (-1.3)
VOC species ^c						
Naphthalene	0.00099 (0.0046)	0.0011 (0.0049)	7.3 (6.2)	0.00030 (0.0025)	0.00033 (0.0027)	11 (8.2)
m-Xylene	0.029 (0.132)	0.029 (0.132)	0.2 (-0.2)	0.012 (0.077)	0.012 (0.076)	-0.2 (-0.2)
o-Xylene	0.012 (0.050)	0.012 (0.049)	0.3 (-0.2)	0.0053 (0.030)	0.0053 (0.030)	-0.3 (-0.3)
Toluene	0.129 (0.435)	0.129 (0.435)	0.04 (-0.1)	0.041 (0.224)	0.041 (0.223)	-0.3 (-0.2)
Benzene	0.090 (0.278)	0.090 (0.278)	0.8 (-0.1)	0.044 (0.179)	0.043 (0.178)	-0.2 (-0.2)
Acrolein	0.0052 (0.013)	0.0052 (0.013)	0.2 (-2.5)	0.0012 (0.0054)	0.0011 (0.0051)	-4.0 (-5.3)
1,3-Butadiene	0.0057 (0.025)	0.0057 (0.025)	-1.0 (-1.4)	0.0013 (0.0099)	0.0013 (0.0097)	-1.9 (-2.0)
Acetaldehyde	0.024 (0.096)	0.024 (0.096)	0.03 (-0.1)	0.0067 (0.034)	0.0067 (0.034)	-0.3 (-0.3)
Formaldehyde	0.067 (0.236)	0.066 (0.235)	-1.2 (-0.3)	0.023 (0.106)	0.022 (0.105)	-0.5 (-0.6)

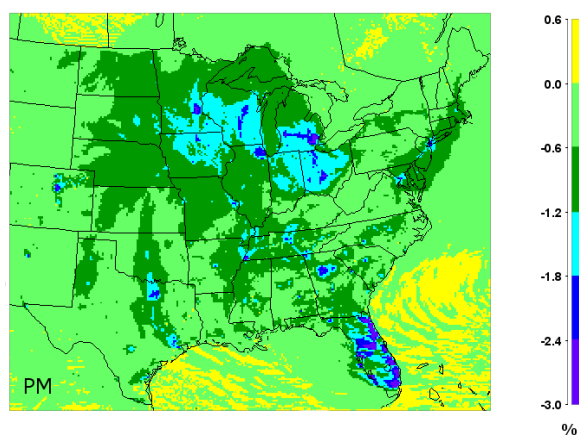
^a Values given in parentheses correspond to grid cells in the domain for which the land use is $\geq 5\%$ urban.

^b Concentrations of PM and secondary organic aerosol (SOA) concentrations in units of $\mu\text{g}/\text{m}^3$; all other species in units of ppbV.

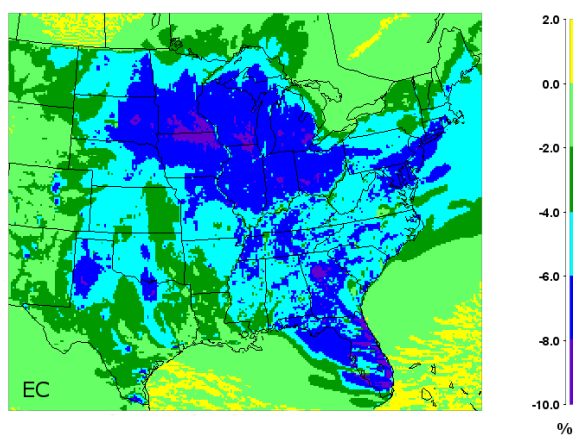
^c VOC species are listed in order of decreasing molecular weight.

Figure S3-1. Absolute and relative changes in $PM_{2.5}$ and $PM_{2.5}$ -EC average concentrations in summer and winter. Panels (a) and (b) are relative changes for one summer day (07/20/2005), (c)-(f) are changes in summer (7/17-7/30/2005) 14-day average concentrations. Panels (g)-(j) are changes in winter (1/25/2005) 24-hour average concentrations. Panels (k)-(n) are changes in winter (1/17-1/30/2005) 14-day average concentrations.

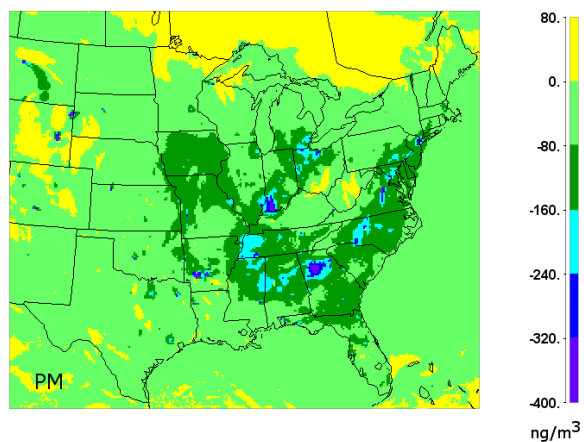
(a) PM on 07/20/2005



(b) PM-EC on 07/20/2005



(c) summer 14-day average PM



(d) summer 14-day average PM-EC

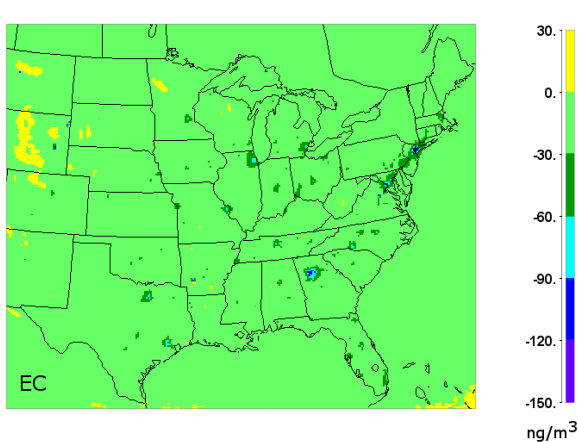
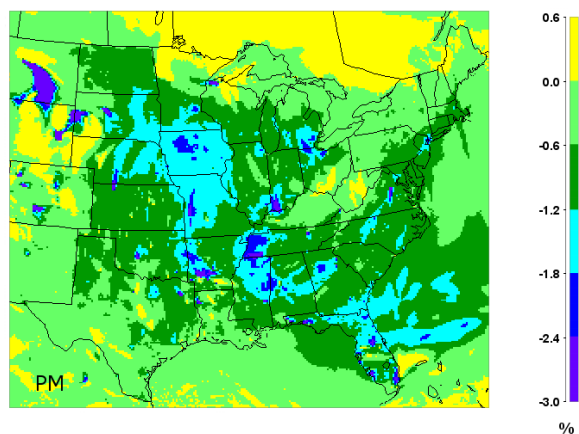
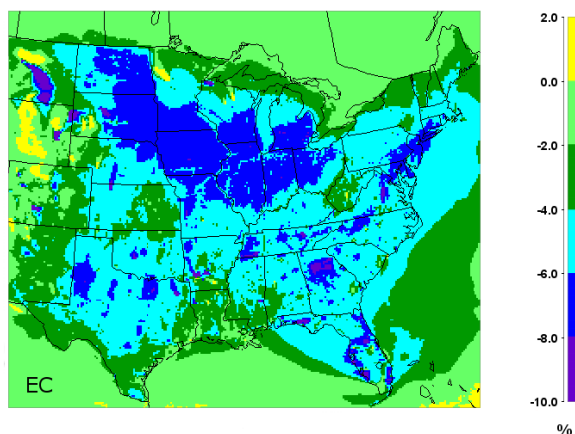


Figure S3-1. Absolute and relative changes in $\text{PM}_{2.5}$ and $\text{PM}_{2.5}\text{-EC}$ average concentrations in summer and winter. Panels (a) and (b) are relative changes for one summer day (07/20/2005), (c)-(f) are changes in summer (7/17-7/30/2005) 14-day average concentrations. Panels (g)-(j) are changes in winter (1/25/2005) 24-hour average concentrations. Panels (k)-(n) are changes in winter (1/17-1/30/2005) 14-day average concentrations.

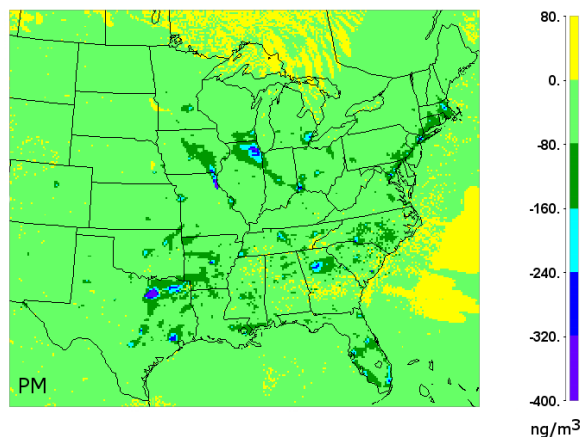
(e) summer 14-day average PM



(f) summer 14-day average PM-EC



(g) 1/20/2005 24-hour average PM



(h) 1/20/2005 24-hour average PM-EC

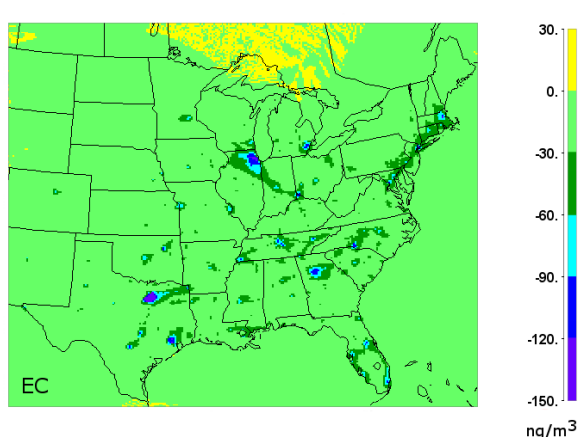
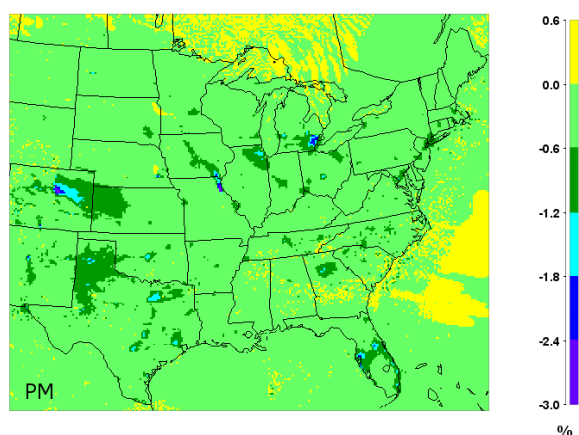
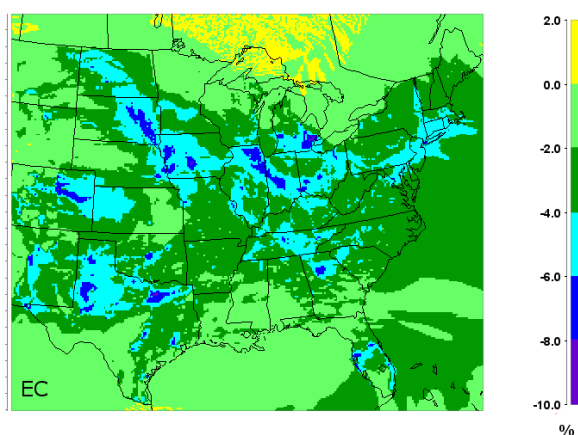


Figure S3-1. Absolute and relative changes in $PM_{2.5}$ and $PM_{2.5}$ -EC average concentrations in summer and winter. Panels (a) and (b) are relative changes for one summer day (07/20/2005), (c)-(f) are changes in summer (7/17-7/30/2005) 14-day average concentrations. Panels (g)-(j) are changes in winter (1/25/2005) 24-hour average concentrations. Panels (k)-(n) are changes in winter (1/17-1/30/2005) 14-day average concentrations.

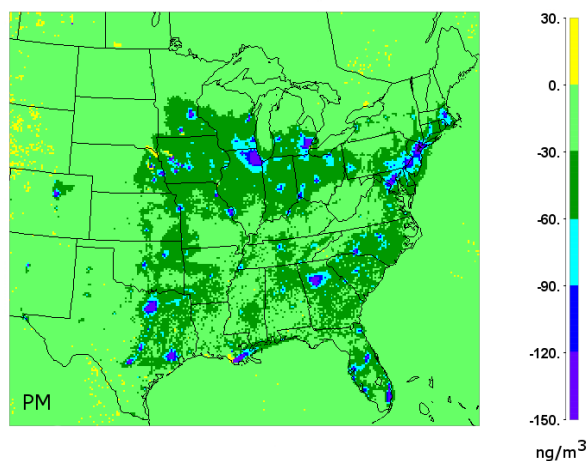
(i) 1/20/2005 24-hour average PM



(j) 1/20/2005 24-hour average PM-EC



(k) winter 14-day average PM



(l) winter 14-day average PM-EC

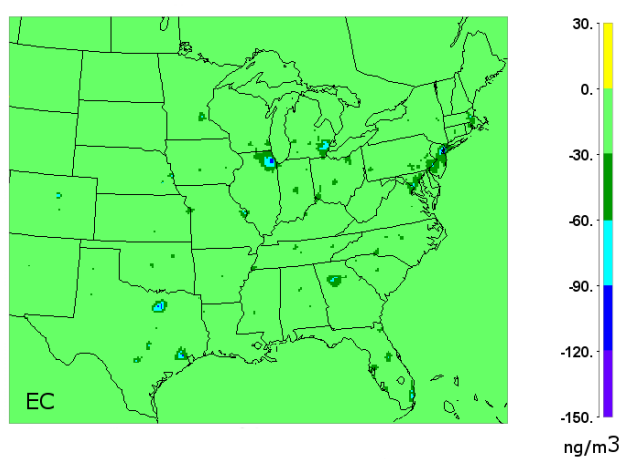
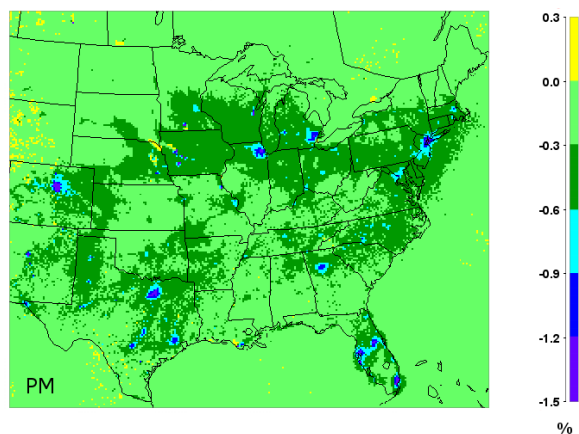


Figure S3-1. Absolute and relative changes in $PM_{2.5}$ and $PM_{2.5}$ -EC average concentrations in summer and winter. Panels (a) and (b) are relative changes for one summer day (07/20/2005), (c)-(f) are changes in summer (7/17-7/30/2005) 14-day average concentrations. Panels (g)-(j) are changes in winter (1/25/2005) 24-hour average concentrations. Panels (k)-(n) are changes in winter (1/17-1/30/2005) 14-day average concentrations.

(m) winter 14-day average PM



(n) winter 14-day average PM-EC

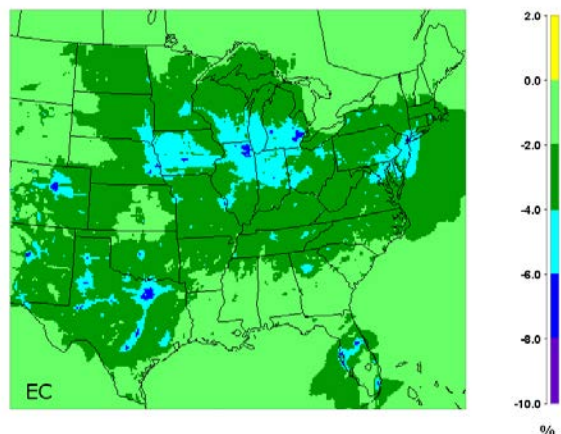
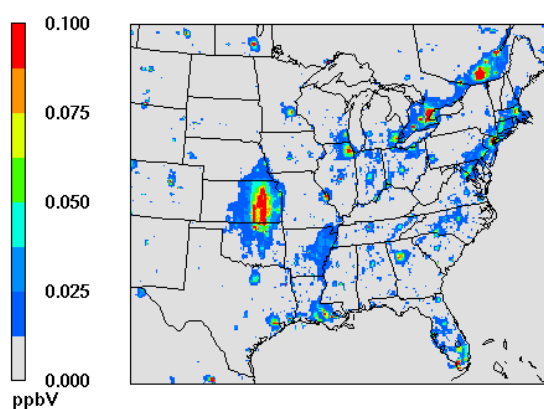
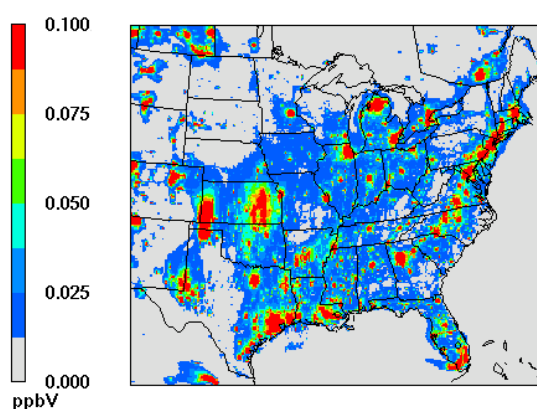


Figure S3-2. Summer and winter 14-day average select HAP base case (no cerium additive) concentrations and relative changes in concentrations. Panels (a)-(d) are summer base case concentrations, panels (e) and (f) are relative changes in summer concentrations of two of the HAPs, and (g)-(j) are winter base case concentrations, and panels (k)-(n) are relative changes in winter concentrations. The four species considered in these figures were selected based on a combination of their roles as key atmospheric pollutants, the size of diesel source contributions to their emissions, and the extent to which nCe additives modify their emissions. Extremely low base case concentrations occur in many areas of the model domain, particularly the central states, and result in extraordinarily large relative changes. Therefore, only relative changes computed for base concentrations > 0.001 ppbV are shown in these figures.

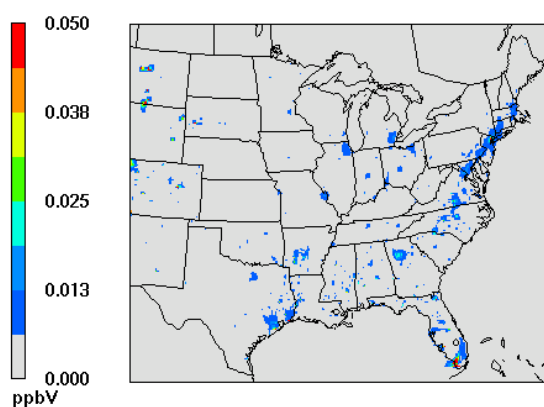
(a) base summer acetaldehyde



(b) base summer formaldehyde



(c) base summer acrolein



(d) base summer naphthalene

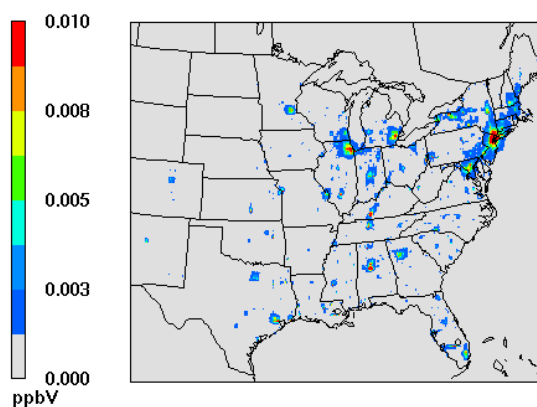
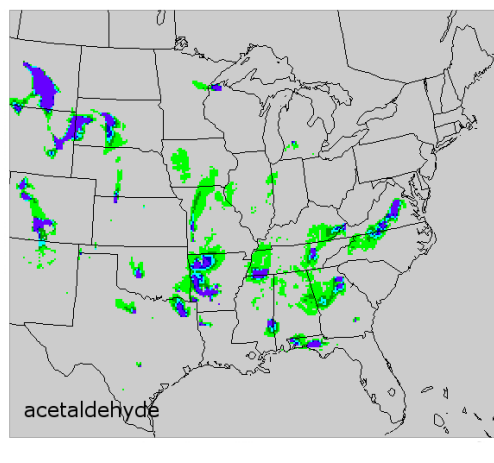
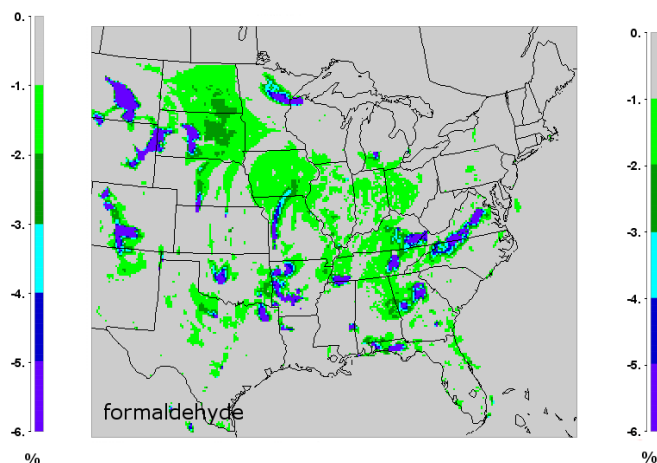


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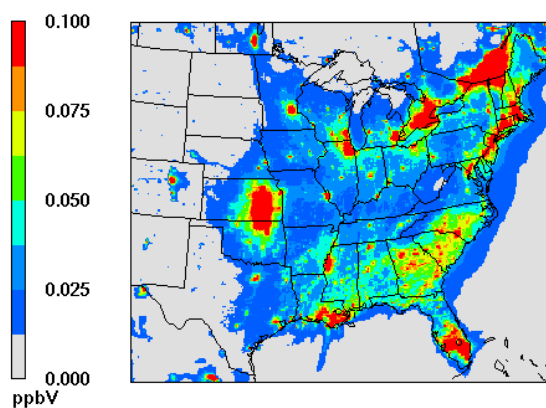
(e) change in summer acetaldehyde



(f) change in summer formaldehyde



(g) base winter acetaldehyde



(h) base winter formaldehyde

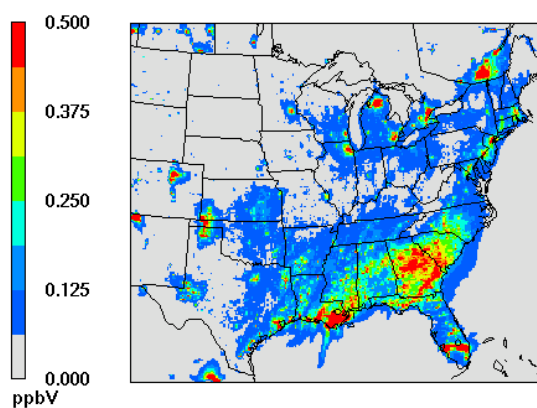
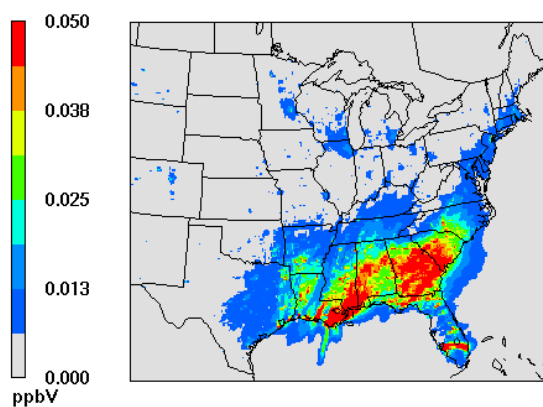
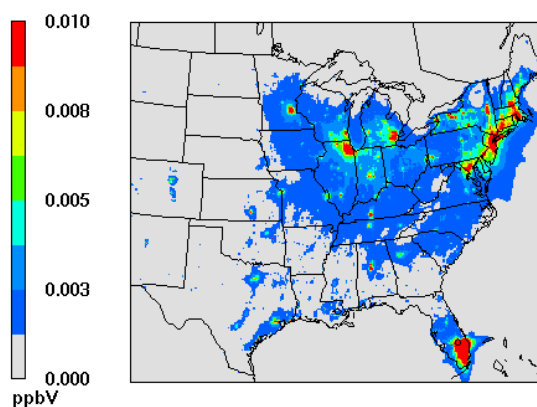


Figure S3-2. Summer and winter 14-day average select HAP base case (no cerium additive) concentrations and relative changes in concentrations. Panels (a)-(d) are summer base case concentrations, panels (e) and (f) are relative changes in summer concentrations of two of the HAPs, and (g)-(j) are winter base case concentrations, and panels (k)-(n) are relative changes in winter concentrations.

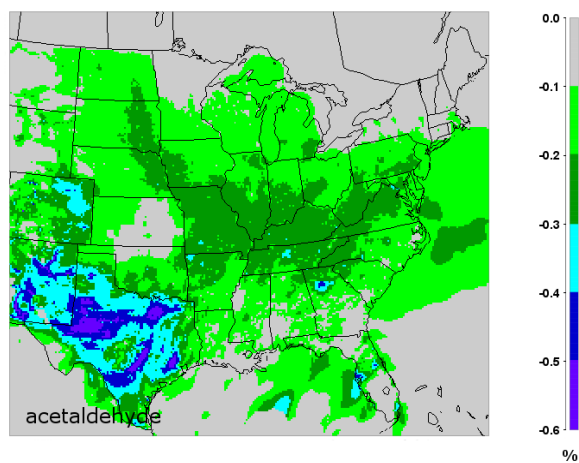
(i) base winter acrolein



(j) base winter naphthalene



(k) change in winter acetaldehyde



(l) change in winter formaldehyde

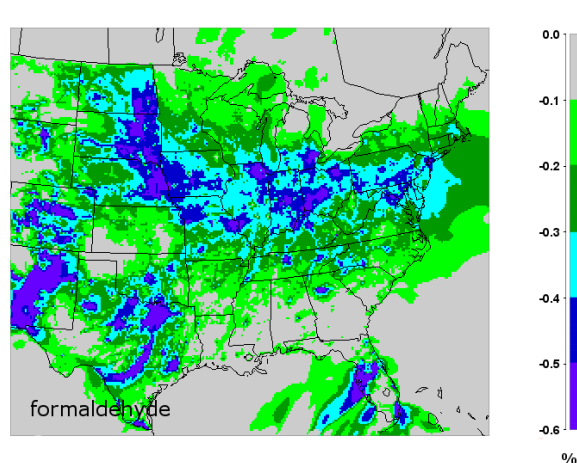
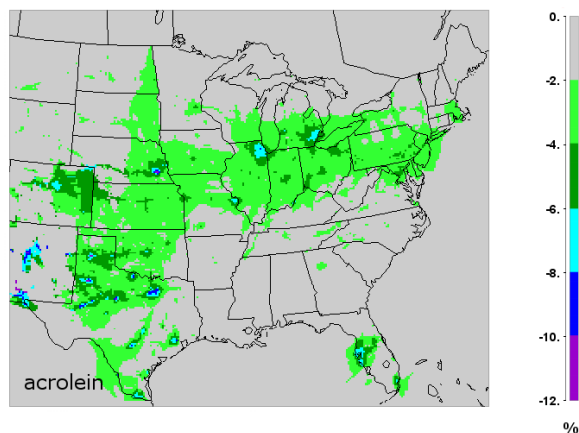
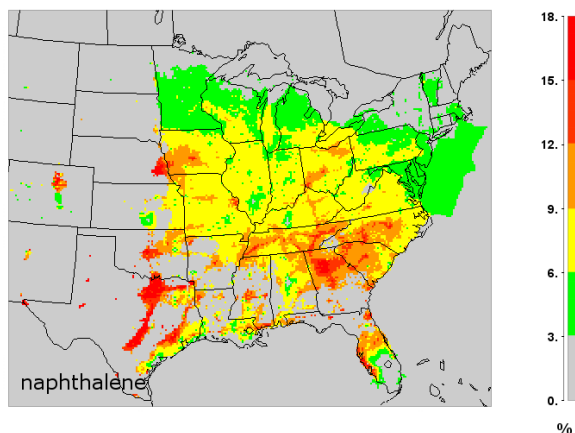


Figure S3-2. Summer and winter 14-day average select HAP base case (no cerium additive) concentrations and relative changes in concentrations. Panels (a)-(d) are summer base case concentrations, panels (e) and (f) are relative changes in summer concentrations of two of the HAPs, and (g)-(j) are winter base case concentrations, and panels (k)-(n) are relative changes in winter concentrations.

(m) change in winter acrolein



(n) change in winter naphthalene



Section S4

Impacts on Emissions from Engines Equipped with DPFs

Table S4-1. Emissions measured in diesel engine test studies using a diesel particulate filter (DPF), without (base) and with (additive) the use of nCe fuel additive. The percent changes (%Δ) calculated from these data are plotted in Figure S4-1.

Reference	Additive Concentration (ppm Ce in fuel)	Emissions ^a											
		PM			CO			NO _x			THC		
		base	additive	%Δ	base	additive	%Δ	base	additive	%Δ	base	additive	%Δ
Summers, 1996	10	14.6	14.9	2.3	-	-	-	-	-	-	-	-	-
Czerwinski, 2000	25	0.009	0.007	-22	0.37	0.57	54	16.2	15.9	-2.1	0.18	0.19	5.6
	25	0.010	0.009	-10	0	0	0	13.0	13.6	4.8	0.20	0.16	-20
	25	0.018	0.014	-22	0.19	0.20	5.3	11.2	11.7	4.3	0.31	0.29	-6.5
	25	0.011	0.010	-9.1	0.74	1.20	62	13.4	13.9	4.3	0.23	0.19	-17
	25	0.007	0.008	14	0.24	0.62	158	16.3	16.0	-2.2	0.18	0.17	-5.6
	25	0.007	0.007	0	0.56	0.57	1.8	15.2	15.9	4.5	0.12	0.19	58
	25	0.012	0.009	-25	0.03	0	-	13.9	13.6	-2.4	0.14	0.16	14
	25	0.015	0.014	-6.7	0.32	0.20	-38	10.8	11.7	8.2	0.27	0.29	7.4
	25	0.011	0.010	-9.1	0.92	1.20	30	13.6	13.9	2.1	0.18	0.19	5.6
	25	0.009	0.008	-11	0.44	0.62	41	15.8	16.0	1.0	0.10	0.17	70
Median %Δ				-9.1			18			3.2			5.6
Mean %Δ				-9.0			22			2.3			11
Standard error of mean				3.5			21			1.1			9.5

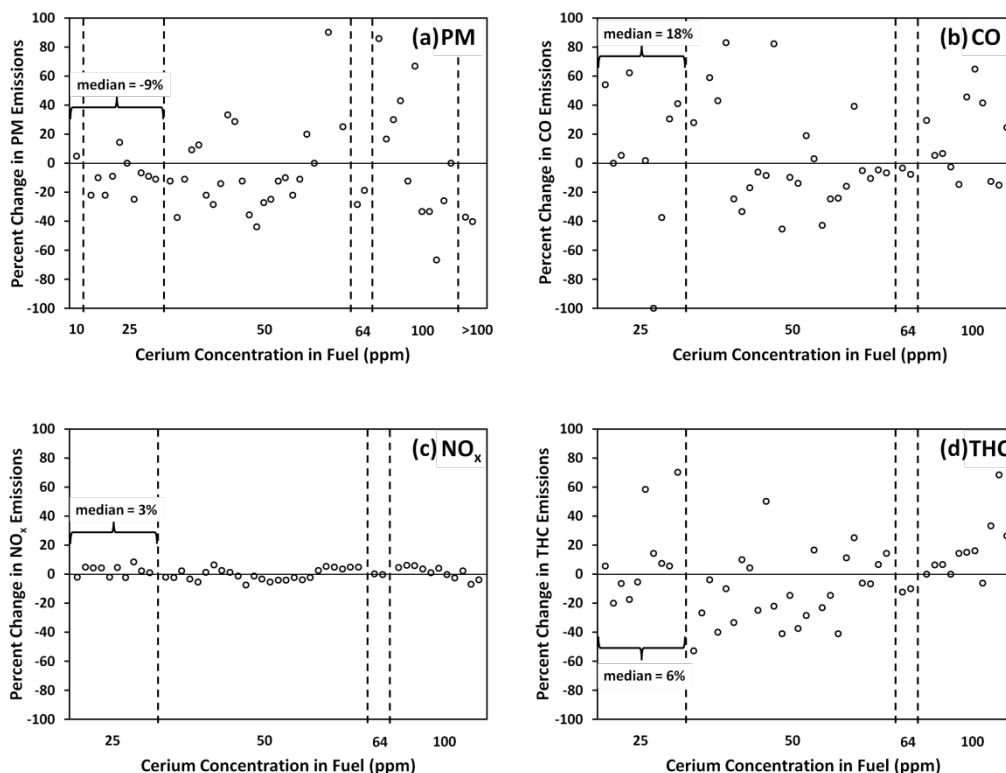
^a Emissions data from Summers are in units of g/10 engine testing cycles; from Czerwinski, units are in g/kW-h.

Table S4-2. Median percent change in diesel emissions due to nCe additives when using a diesel particulate filter (DPF)

Species	%Δ
PM	-9.1
CO	+18
NO _x	+3.2
NO	+3.2
NO ₂	+3.2
VOC	+5.6
PM species	
EC	-6.6
OC	-19.3
SO ₄	-10.1
NO ₃	-10.1
Other	-10.1
Cerium	a

^a The median mass fraction of cerium in DPM with a diesel particulate filter, computed from the available test data is 0.02.

Figure S4-1. Relative changes in emissions of (a) PM, (b) CO, (c) NO_x, and (d) THC measured in the six studies using diesel particulate filters listed in Table S4-1. Individual changes from each pair of data (emissions without additive and with additive) are plotted as open circles. All studies were performed using the Eolys™ additive. Median percent changes across all test pairs that used ≤ 25 ppm Ce are shown in each panel. (See Table S4-1 for raw data, mean values, and standard errors).



References

Cerulean International Limited. 2005. *Envirox™ Tier 1, 211b Summary Report*, U.S. Environmental Protection Agency, Office of Transportation and Air Quality Fuel and Fuel Additives Registration Submittal.

Clean Diesel Technologies, Inc. 1999. *Platinum Plus® DFX Tier 1, 211b Summary Report*, U.S. Environmental Protection Agency, Office of Transportation and Air Quality Fuel and Fuel Additives Registration Submittal.

Czerwinski, J., T. Mosimann, and U. Matter. 1997. *Comparison of the Engines Liebherr I (86) and Liebherr II (96) with Particulate Trap, with Fuel Additives and with Detailed Analysis*

of the Particulate Emissions. VERT Report, Technik Thermische Maschinen, Niederrohrdorf, Switzerland.

Czerwinski, J., S. Napoli, U. Matter, M. Kasper, and T. Mosimann. 1999. *Investigations with the IBIDEN SiC-Diesel Particulate Filter on the Liebherr D914T Engine with Detailed Analysis of the Particulate Emissions*. VERT Report, Technik Thermische Maschinen, Niederrohrdorf, Switzerland.

Czerwinski, J., S. Napoli, P. Compte, U. Matter, and T. Mosimann. 2000. *Investigations with the Corning-Diesel Particulate Filter and Cerium-Additive Eolys DPX 9 on the Liebherr D914T Engine*. VERT Report, Technik Thermische Maschinen, Niederrohrdorf, Switzerland.

Heeb N. V. 1998a. *Influence of Particulate Trap Systems on the Composition of Diesel Engine Exhaust Gas Emissions*, Eidgenössische Materialprüfungs Und Forschungsanstalt (EMPA) Research Report 167985.

Oxonica Energy Limited. 2006. *Envirox™ Tier 1, 211b Summary Report*. U.S. Environmental Protection Agency, Office of Transportation and Air Quality Fuel and Fuel Additives Registration Supplemental Submittal.

Skillas, G., Z. Qian, U. Baltensperger, U. Matter, and H. Burtscher. 2000. The Influence of Additives on the Size Distribution and Composition of Particles Produced by Diesel Engines, *Combust. Sci. and Technol.* 154:259-273.

Southwest Research Institute (SwRI). 1997. *Study of Long Term Effect of Cerium Additive on Heavy Duty Diesel Engine Emissions and Durability*. Final Report SwRI 5870.

Summers, J. C., S. Van Houtte, and D. Psaras. 1996. Simultaneous control of particulate and NO_x emissions from diesel engines. *Appl. Catal. B: Environmental*. 10:139-156.